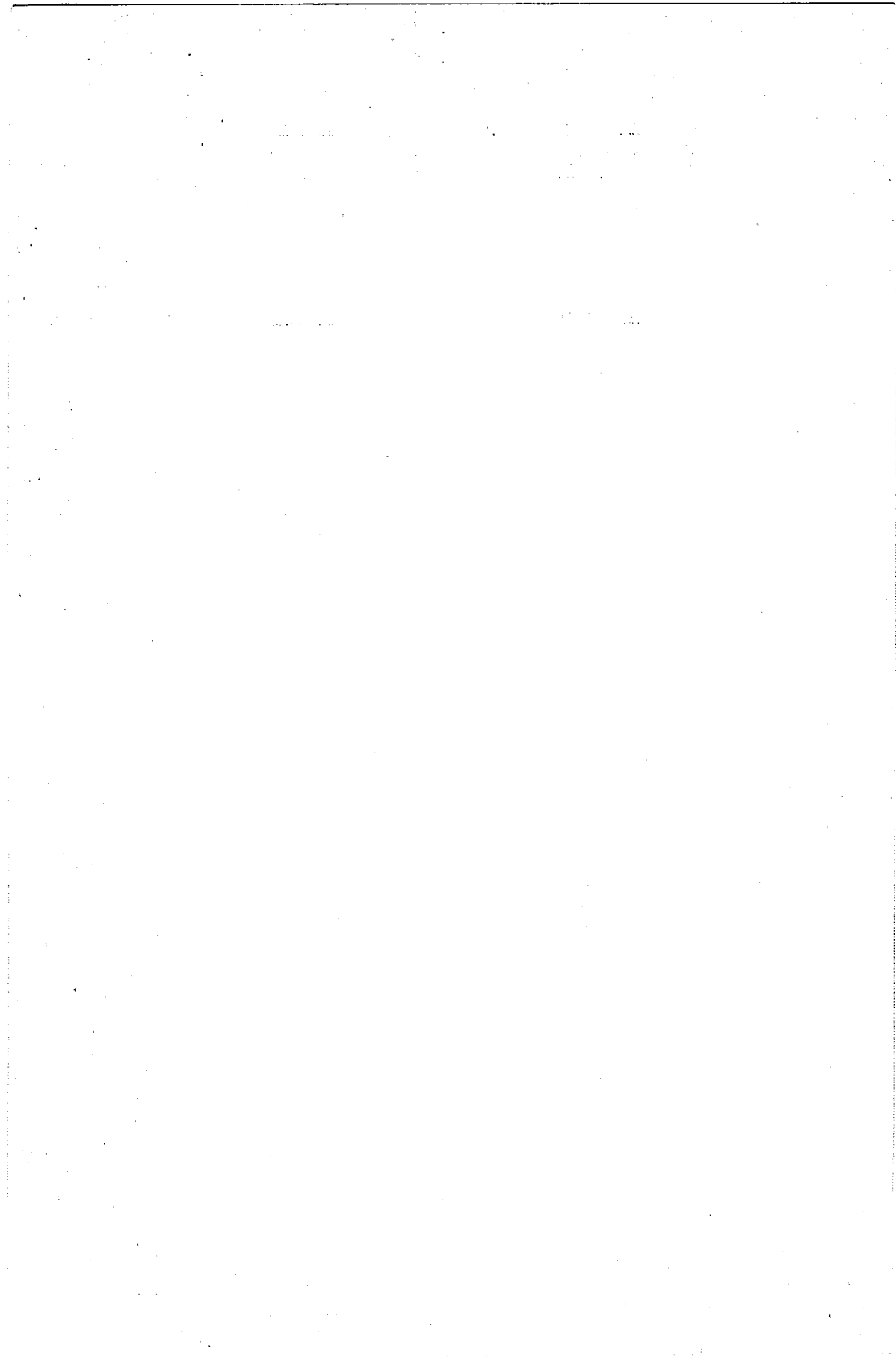


URANIUM ORE PROCESSING

Proceedings
of an
Advisory
Group Meeting
Washington, DC
24-26 November
1975



INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1976



URANIUM ORE PROCESSING

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The Agency's Statute was approved on 23 October 1956 by the Conference on the Statute of the IAEA held at United Nations Headquarters, New York; it entered into force on 29 July 1957. The Headquarters of the Agency are situated in Vienna. Its principal objective is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world".

PANEL PROCEEDINGS SERIES

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FOREWORD

The forecasts of uranium requirements up to the end of the century are very large and it is estimated that the present annual demand for uranium will increase by more than a factor of ten in the next twenty-five year period. There will be a need for a large expansion in milling capacity and an entirely new generation of milling plants within the next decade. This will be in addition to maintaining, modernizing and expanding the production capacity now in operation. All these plants will have to make the most effective use of the various types of uranium resources and must have the advantage of the most efficient processes and technologies that can be made available.

The International Atomic Energy Agency sponsored a panel meeting in Vienna in June 1966 on the Processing of Low-Grade Uranium Ores and followed this with a symposium in São Paulo, Brazil, in August 1970 on the Recovery of Uranium. The proceedings of both meetings were published by the IAEA. The Agency felt that it was timely to convene another meeting on this subject in 1975 and thus, at the invitation of the United States Bureau of Mines, an Advisory Group Meeting on Uranium Ore Processing was held in Washington, DC, from 24 to 26 November 1975.

A total of 49 participants from 17 countries and one international organization took part in the meeting and 18 papers were presented. The technical sessions covered the following topics: Anticipated future demand for uranium and need for increase in milling capacity; uranium ore milling practice; known and partially engineered techniques which have not reached full application in the milling of uranium ores; process problems and developments for some new uranium ore occurrences; processing of low-grade resources; uranium as a by-product and by-products from uranium ores; in situ leaching; and recovery of uranium from sea-water. A panel, appointed from among the participants, summarized the conclusions of the meeting and presented recommendations for future action. The papers, the discussions, and the conclusions and recommendations are presented in these Proceedings.

The Agency wishes to thank all the scientists and engineers who participated in the meeting, not only for the papers but also for the contributions to the discussions. Thanks are especially due to the Chairman of the meeting, Mr. Joe B. Rosenbaum, Consulting Metallurgist, Bureau of Mines, Salt Lake City, Utah, USA, and to the Chairman of the panel group, Mr. F.C. Lendrum, Metallurgical Consultant, King City, Ontario, Canada. Special thanks are extended to Mr. H.E. James of the Atomic Energy Board, Pelindaba, South Africa, who completed the tables given at the end of this book, showing a classification of the available information on commercial uranium extraction flowsheets, new plants and techniques in the countries contributing to the meeting. The Agency also wishes to record its sincere gratitude to the United States authorities and specifically the United States Bureau of Mines for hosting the meeting and providing the excellent conference arrangements and general hospitality to the participants.

CONTENTS

ANTICIPATED FUTURE DEMAND FOR URANIUM AND NEED FOR INCREASE IN MILLING CAPACITY (Session I)

Outlook for uranium demand and production capacity (IAEA-AG/33-8)	3
<i>J.B. Rosenbaum</i>	
Discussion	9

URANIUM ORE MILLING PRACTICE (Session II)

Recent trends in Canadian uranium processing (IAEA-AG/33-9)	13
<i>F.C. Lendrum, H.H. McCreedy</i>	
Discussion	17
Recent trends in research and development work on the processing of uranium ore in South Africa (IAEA-AG/33-5)	23
<i>H.E. James</i>	
Discussion	37
La mineralurgia del uranio en España (IAEA-AG/33-6)	41
<i>J.M. Josa</i>	
Discussion	52
Some recent improvements in a uranium processing pilot-plant at the Ningyo-toge Mine (IAEA-AG/33-1)	55
<i>S. Takenaka, H. Kawate</i>	
Discussion	67
Process development study on uranium-bearing carbonatite (IAEA-AG/33-16)	69
<i>M. Shabbir, Naeem-ul-Zaman</i>	
Discussion	75

KNOWN AND PARTIALLY ENGINEERED TECHNIQUES WHICH HAVE NOT REACHED FULL APPLICATION IN MILLING OF URANIUM ORES (Session III)

Stored technology for possible use in uranium ore processing (IAEA-AG/33-7)	79
<i>D.C. Seidel</i>	
Discussion	89

PROCESS PROBLEMS AND DEVELOPMENTS FOR SOME NEW URANIUM ORE OCCURRENCES (Session IV)

Notes on technical aspects of problems connected with the treatment of several new-generation Australian uranium occurrences (IAEA-AG/33-2)	93
<i>L.H. Goldney, F.R. Hartley, R.G.P. McMahon</i>	
Discussion	105

Treatment of an isolated high-grade, low-tonnage uranium orebody (IAEA-AG/33-13)	107
<i>S. Ajuria-Garza, W.D. Jamrack</i>	
Discussion	117
Environmental problems posed by wastes from the uranium milling industry (IAEA-AG/33-14)	119
<i>P.A. Bonhote</i>	
Discussion	123

PROCESSING OF LOW-GRADE RESOURCES (Session V)

Traitement des minerais d'uranium à faible teneur (IAEA-AG/33-10)	127
<i>P. Michel</i>	
Discussion	137
Переработка бедных урановых руд в СССР (IAEA-AG/33-17)	141
<i>Д.И.Скороваров, Б.Н.Ласкорин, Г.Ф.Иванов, М.Л.Скриниченко, И.П.Смирнов, В.Д.Носов, В.И.Дорофеев</i>	
(Processing of low-grade uranium ores in the USSR: D.I. Skorovarov et al.)	
Discussion	152
Studies on the recovery of uranium from low-grade ores in India (IAEA-AG/33-11)	155
<i>K.M. V. Jayaram, K.K. Dwivedy, A.S. Deshpande, T.M. Ramachar</i>	
Discussion	169
Uranium recovery from bituminous shales at Ranstad (IAEA-AG/33-18)	171
<i>Å. Andersson</i>	
Discussion	176

URANIUM AS A BY-PRODUCT AND BY-PRODUCTS FROM URANIUM ORES (Session VI)

Uranium as a by-product and by-products of uranium production (IAEA-AG/33-3)	181
<i>F.E. McGinley, J.F. Facer</i>	
Discussion	187

IN-SITU LEACHING (Session VII)

In-situ leach mining for uranium (IAEA-AG/33-15)	193
<i>G.R. Davis, R.E. Miller, G.G. Swift</i>	
Discussion	200

RECOVERY OF URANIUM FROM SEA-WATER (Session VIII)

Recovery of uranium from sea-water (IAEA-AG/33-12)	205
<i>G.I.W. Llewelyn</i>	
Uranium from sea-water: Possibilities of recovery, exploiting slow coastal currents (IAEA-AG/33-4)	213
<i>C. Bettinali, F. Pantanetti</i>	

Discussion to papers IAEA-AG/33-12 and IAEA-AG/33-4	231
Conclusions and recommendations	233
List of participants	235

CLASSIFICATION OF URANIUM EXTRACTION PLANTS

Table A—I.	Classification of commercial uranium extraction flowsheets	Attached
Table A—II.	New uranium plants and deposits under investigation	to
Table A—III.	Summary of existing, improved and new techniques	inside
	for uranium ore processing steps	back
		cover

**ANTICIPATED FUTURE DEMAND FOR URANIUM
AND NEED FOR INCREASE IN MILLING CAPACITY**

(Session I)

OUTLOOK FOR URANIUM DEMAND AND PRODUCTION CAPACITY

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Abstract

OUTLOOK FOR URANIUM DEMAND AND PRODUCTION CAPACITY.

Despite appreciable errors in recent forecasts of nuclear power growth rate, the amended estimates still show the required amount of uranium raw material for 1985 to be three to four times larger than the world's existing uranium production capacity, both operating and on standby. By expanding the existing production units and adding new units based on known ore reserves, the uranium demands of 1980 might be met. The number of production units in the world (except for China, the Soviet Union and the Eastern European countries) might increase from the current 45 to about 70. Substantial additional capacity based mostly on newly discovered ore resources would be necessary to meet the uranium demands of 1985. Because a lead time of about eight years is probable to bring a newly discovered uranium deposit into production, a prompt increase in exploration activity is needed. The search for resources and development of reserves is expected to extend to ores containing as little as 0.02% U_3O_8 compared to the average ore grade of about 0.2% U_3O_8 now being processed. Expanded exploration activities require commensurate research and application effort to improve mining and milling practice for efficient and economical utilization of ores that are less accessible, of lower grade, or of different metallurgical response.

INTRODUCTION

The purpose of this paper is to present and assess estimates for uranium demand and production needs to meet the requirements for nuclear power generation. Exclusive of China, the Soviet Union and the Eastern European countries, the world's uranium production capacity, both operating and on standby, is estimated at about 35 000 tons¹ U_3O_8 annually. This is 40% more than the amount required for fuelling nuclear reactors now operating, but it is insufficient for the increasing requirements of reactors being built and planned. Marked errors in the forecast rate of nuclear power growth have occurred because of technological, financial, safety and environmental factors, worldwide economic recession, and efforts to conserve energy. Despite a downward revision of the forecast data, unofficial amended estimates still show the required amount of uranium raw material for 1985 to be three to four times larger than the world's existing uranium production capacity.

DEMAND FOR URANIUM

The anticipated demand for uranium is largely derived from installed and projected nuclear power capacity, but other factors also are of importance. These include reactor type, size, efficiency, operating time and rate, degree of fuel enrichment, percentage of ^{235}U in the enrichment plant tailings, percentage of fuel burn-up in the reactor, availability of spent fuel reprocessing facilities,

¹ Tons are short tons throughout the paper: 1 short ton = 2000 lb = 907.18 kg = 0.90718 tonnes.

TABLE I. URANIUM REQUIREMENTS FOR A SUSTAINED NUCLEAR CAPACITY OF 300 000 MW(e)

Plutonium recycle	Enrichment tailings assay, ^{235}U (%)	Uranium required annually per GW(e) (tons U_3O_8)
No	0.30	190
No	0.36	250
Yes	0.30	162

TABLE II. FORECAST OF UNITED STATES NUCLEAR POWER CAPACITY AND URANIUM REQUIREMENTS

Year	Nuclear capacity (GW(e))	Uranium required annually (10^3 tons U_3O_8)	Uranium required annually per GW(e) (tons U_3O_8)
1975	39	10	256
1980	76	23	303
1985	185	46	249
1990	340	74	218

and whether plutonium recovered from spent uranium fuel is itself used as fuel in nuclear reactors. Breeder-type reactors producing new fuel from uranium or thorium simultaneously with energy are not expected to be in use until after 1990.

The influence of enrichment plant tailings assay and plutonium recycle on uranium requirements is illustrated in Table I (from a paper by Baronowski of the USAEC [1]). These data reflect anticipated conditions of the late 1980s. Increasing the enrichment tailings assay from 0.30 to 0.36% ^{235}U increases the uranium requirements by one-third. With the tailings at 0.3% ^{235}U and plutonium being recycled, the uranium requirement decreases by 14%.

Recently, estimates of the growth rate for nuclear power and concomitant uranium requirements have declined appreciably, especially for the period up to 1990. The forecasters appear to assume that the current obstacles to the development of nuclear power will be resolved over the next 10 to 15 years. Conspicuous among the current obstacles that result in construction schedule errors and deferred construction commitments are technical and financial difficulties, questions of safety, environmental concerns, and a slowdown in the growth rate for electric power generation. Current forecasts of United States installed nuclear capacity and uranium requirements are shown in Table II. The data are from an ERDA (Energy Research and Development Administration) report [2] and reflect a moderate nuclear power growth rate, about 0.3% ^{235}U enrichment tailings, and no plutonium recycle until about 1985. The forecasts assume that nuclear plants operate at 40% of the capacity during a 6-month start-up period, at 65% of the capacity for 2 years, and then at 70% of the capacity. After the 15th year of operation the capacity factor is assumed to drop by 2% per year until a minimum of 40% is reached.

TABLE III. STATUS OF NUCLEAR GENERATING UNITS IN THE USA, 30 JUNE 1975

Status	Number of units	GW(e)
Operable	55	37
Being built	76	77
Planned ^a	112	129
Total	243	243

^a Orders have been placed for 91 units.

During the first six months of 1975, utilities in the USA announced plans for 11 nuclear generating units with a capacity of about 14 000 MW(e). In the same period, three generating units were cancelled and 41 others were delayed, mostly for 1 to 2 years. Table III shows the status of nuclear generating units in the USA as of 30 June 1975.

To fuel the 57 or 58 reactors expected to be operating in the USA by the end of 1975 with a combined capacity of 39 000 MW(e) requires about 10 000 tons U_3O_8 annually. Outside the USA and exclusive of China, the Soviet Union and the Eastern European countries, about 90 reactors with a combined capacity of about 35 000 MW(e) are expected to be operable in 13 countries [3] (see also Ref. [2]). Many of the latter reactors are small and of relatively low efficiency, and their uranium consumption is estimated at about 15 000 tons U_3O_8 in 1975. As compared with possible needs in the USA of 23 000 tons U_3O_8 annually by 1980, 46 000 tons annually by 1985, and 74 000 tons annually by 1990, the rest-of-the-world requirements (except for China, the Soviet Union and the Eastern European countries) might be 40 000 tons by 1980, 80 000 tons by 1985, and 110 000 tons by 1990.

URANIUM PRODUCTION CAPACITY

The uranium production capacity is inextricably bound to the resource base. Measures that would assure an adequate resource base and production capacity to meet the needs of forecast nuclear power generation were examined in a joint OECD-IAEA report issued in 1973 [4]. The report recognized that an eight-year lead time was needed to bring a newly discovered deposit into production and found that a prompt expansion of exploration was desirable to avoid a possible shortage of U_3O_8 early in the 1980s. At that time the uranium prices were believed to be too low to stimulate the needed exploration effort. However, the prices rose dramatically in 1975. A paper by Patterson of ERDA shows that peak prices for delivery in 1975 went up from US \$10 to US \$22 per pound U_3O_8 , and for delivery in 1980 from US \$13 to US \$35 per pound U_3O_8 . Contract prices for delivery in 1982 range from US \$10 to US \$45, and the weighted average is US \$19 [5].

Uranium deposits and how they were formed was the topic of an IAEA symposium held in Athens, Greece, in 1974 [6]. The findings were also published in an abridged form [7]. Again, the long lead time to bring a deposit into production was stressed, and short-term production of uranium from ores averaging only 0.03% U_3O_8 was predicted. In this connection, ore processing at Rossing, South-West Africa, at average grade of 0.037% U_3O_8 , is expected to start at a rate of 25 000 tons per day in 1976.

TABLE IV. UNITED STATES URANIUM PRODUCTION CENTRES — 1975

Company	Location	Nominal capacity (tons ore/d)	Process used	Mining method
Anaconda Co.	Grants, NM	3000	Acid leach, resin-in-pulp	Surface and underground
Atlas Corp.	Moab, UT	1500	Carbonate leach, resin-in-pulp	Underground
Conoco & Pioneer Nuclear, Inc.	Falls City, TX	1750	Acid leach, CCD, solvent extraction	Surface
Cotter Corp.	Canon City, CO	450	Carbonate leach, caustic precipitation	Underground
Dawn Mining Co.	Ford, WA	400	Acid leach, CCD, column ion exchange	Surface
Exxon, USA	Powder River Basin, WY	2000	Acid leach, CCD, solvent extraction	Surface and underground
Exxon, USA	Raypoint, TX ^a	1000	Carbonate leach, caustic precipitation	Surface
Federal-American Partners	Gas Hills, WY	950	Acid leach, eluex	Surface and underground
Kerr-McGee Nuclear	Grants, NM	7000	Acid leach, CCD, solvent extraction	Underground
Petrotomies Co.	Shirley Basin, WY ^a	1500	Acid leach, CCD, solvent extraction	Surface
Rio Algom Corp.	LaSal, UT	500	Carbonate leach, caustic precipitation	Underground
Sohio Petroleum Co.	Grants, NM ^b	1500	Acid leach, CCD, column extraction	Surface and underground
Union Carbide Corp.	Uravan, CO	1300	Acid leach, CCD, column ion exchange	Underground
Union Carbide Corp.	Natrona County, WY	1000	Acid leach, eluex	Surface
United Nuclear-Homestake Partners	Grants, NM	3500	Carbonate leach, caustic precipitation	Underground
United Nuclear Corp.	Gallup, NM ^b	4000	Acid leach, CCD, solvent extraction	Underground
Utah International, Inc.	Gas Hills, WY	1200	Acid leach, eluex	Surface
Utah International, Inc.	Shirley Basin, WY	1200	Acid leach, CCD, column ion exchange	Surface
Western Nuclear, Inc.	Jeffrey City, WY ^a	1200	Acid leach, eluex	Underground
Total		34950		

^a On standby; may require renovation.^b Committed or under construction.

Both resources and production facilities were reviewed by Poole [8]. Owing to the recent sharp escalation in fossil fuel costs, nuclear fuel is now more competitive. It was anticipated that continuing efforts in exploration, ore processing technology and use of secondary sources are necessary.

A report by Klemenic [9] of ERDA identified 19 production units or centres in the USA now operating, on standby, or being built. Each is comprised of a uranium processing mill with ore resources and associated mining facilities. These 19 centres are shown in Table IV. A further unit, of relatively small capacity, Arco's in-place leaching operation in Texas, came on-stream in 1975, and Wyoming Mineral Co.'s in-place leaching operation in Texas, also of relatively small capacity, may come on-stream in 1976. Not shown as a production unit in Table IV because of uncertain ore supply is the TVA mill (formerly Mines Development Co.) in Edgemont, SD, which is on standby.

In 1974, because of slack demand and shortage of underground miners, uranium mills in the USA produced only 11 500 tons U_3O_8 , the lowest production since 1967 [10]. The mills in Table IV have a total nominal capacity of 35 000 tons ore daily. At an assumed average grade of 0.19% U_3O_8 and 95% recovery [10], the production potential is about 22 000 tons U_3O_8 annually. No by-product uranium is recovered currently, but a central finishing mill with one module at a phosphoric acid plant was recently completed [10]. Two more modules are planned at other phosphoric acid plants. The central plant with three modules is estimated to have a capacity of about 500 tons U_3O_8 annually. Stimulated by higher prices and improved technology, the near-term production from phosphoric acid could reach 3000 tons U_3O_8 annually. By drawing on known ore reserves, expanding existing facilities, and adding about six new mills, the United States production capability of 32 000 tons U_3O_8 annually by the early 1980s appears attainable. This is about equal to the forecast demand for 1982 but well under the amended estimate of 48 000 tons U_3O_8 required annually by 1985.

The 1973 OECD-IAEA report [4], the 1975 paper by Poole [8], and the 1975 Commodity Data Summaries of the United States Bureau of Mines [11] show the existing and potential production outside the USA. These data are summarized below. However, the 1974 production statistics of Ref. [11] give only estimated values. The figures given here reflect more recent information.

Australia — No mills operating. The Mary Kathleen plant that has been on standby is expected to resume operation in 1976 and to produce about 750 tons U_3O_8 annually. Two mills are planned, one for 1200 tons U_3O_8 and the other for 3000 tons U_3O_8 annually. Five deposits are known with a near-future potential of about 12 000 tons U_3O_8 annually.

Brazil — No mills operating. One mill is planned to produce 200 tons U_3O_8 annually.

Canada — Three mills operating, five mills on standby. One new mill is planned for 1978.

4700 tons U_3O_8 were produced in 1974. Near-future potential: about 14 000 tons U_3O_8 annually.

France — Three mills operating on uranium ore; one mill operating on low-grade concentrate from Gabon. Mills treating ore have produced 2000 tons U_3O_8 in 1974; they have a potential of 3000 tons U_3O_8 annually.

Gabon — One mill operating, making low-grade concentrate for treatment in France. 850 tons U_3O_8 were produced in 1974.

Germany — Planned near-term production: 300 tons U_3O_8 annually.

India — One mill operating. Production estimated at less than 200 tons in 1974.

Italy — No mills operating. It is intended to produce 120 tons U_3O_8 annually by 1978.

Japan — One experimental mill operating. The production in 1974 is estimated at less than 100 tons U_3O_8 .

Mexico — One mill operating. Less than 100 tons U_3O_8 were produced in 1974. Planned production: 400 tons U_3O_8 by 1978.

Niger — One mill operating. 1200 tons U_3O_8 were produced in 1974. Planned production: 2000 tons U_3O_8 annually by 1976.

Portugal — Two mills operating. Less than 100 tons U_3O_8 were produced in 1974. Planned short-term production: 200 tons U_3O_8 annually.

South Africa — Eleven mills operating (ten on gold ore and tailings and one on copper tailings). 3400 tons U_3O_8 were produced in 1974. Short-term potential estimated at 6000 tons U_3O_8 annually.

South-West Africa — No mills operating. One mill under construction, to produce 2500–5000 tons U_3O_8 annually.

Spain — One mill operating. 50 tons U_3O_8 were produced in 1974. Planned production: 2000 tons U_3O_8 annually by 1980.

Sweden — One experimental mill operating. Less than 100 tons U_3O_8 were produced in 1974. A short-term production of 1500 tons U_3O_8 annually may be possible from shale containing 0.03% U_3O_8 .

To summarize, 26 mills outside the USA, a few of them experimental, produced about 13 000 tons of U_3O_8 in 1974. By drawing on known ore reserves, recommissioning the plants on standby, expanding existing facilities, and adding 8 to 12 new plants, a production capability of 45 000 tons U_3O_8 annually by the early 1980s appears attainable. This is higher than the estimated demand outside the USA of 40 000 tons by 1980 but well under the amended demand estimate of 80 000 tons U_3O_8 by 1985.

SUMMARY

Forecasts of nuclear power capacity, especially for the period until 1990, are in a state of flux because of sharply rising fossil fuel costs, generally escalated construction costs, technological difficulties, and economic and environmental constraints. The net effect has been a downward revision of estimates of installed nuclear capacity and concomitant uranium fuel requirements. The amended estimates, however, still show uranium raw material production requirements in 1985 as three to four times the current production capacity. To meet the increased demands in time, it is necessary to accelerate exploration and resource development, to expand existing facilities and to add new production units, to further research on extraction and process development for known unamenable ores and for newly discovered ores that may be difficult of access, lower in grade, or of different metallurgical response, and to utilize the best technologies that are appropriate for the world's diverse resources.

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DISCUSSION

F.C. LENDRUM: Are those 1975 dollar figures you gave?

J.B. ROSENBAUM: As far as I know, they are 1975 dollars. Of course, the value of the dollar may be quite different in 1982. But the dollar amount shown is without provision for escalation.

R.J. KLOTZBACH: With regard to in-place leaching, I should like to say that Union Carbide will have an in-place leaching facility on stream at Calangat in Texas in February 1976.

When you refer to total uranium requirements are you assuming that all the uranium in the stream is going to be reprocessed?

J.B. ROSENBAUM: I assume that ultimately there will be reprocessing and plutonium recycling. My interpretation of the figures that have been prepared by ERDA is that they do not include substantial uranium reprocessing and plutonium recycling until at least 1985.

URANIUM ORE MILLING PRACTICE

(Session II)

RECENT TRENDS IN CANADIAN URANIUM PROCESSING

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Abstract

RECENT TRENDS IN CANADIAN URANIUM PROCESSING.

Relatively few changes have been made during the past few years in the overall processing technique for Canadian uranium ores. However, there is a continued interest in new techniques and many are being tested, some with encouraging results. Some of these new methods are likely to be applied in the new plants that are being contemplated in response to the improved uranium market.

INTRODUCTION

With the increasing demand for uranium as a source of energy, there is an increased effort towards finding new uranium deposits in Canada and this has met with some success. The three mills operating during 1974/75 are being expanded to help meet the growing demand for uranium, and a few of the idle mills, previously shut down because of a lack of uranium markets, are being readied for production. During 1974/75 the three operating mills produced 8.8×10^6 lb of yellow-cake from a total ore throughput of about 10 000 tons/day.¹

The first new mill in Canada in 20 years started production in 1975 and will add milling capacity of another 2000 tons of ore per day. Currently, uranium ore is being processed by: Denison Mines Ltd., Elliot Lake, Ontario, which is under expansion to handle 7100 tons of ore per day; Rio Algom Mines Ltd. is also under expansion plans and by 1980 will have boosted the output of its Quirke Mill at Elliot Lake to 7000 tons of ore per day. By either reactivating one of their idle mills or building a new mill, they will increase their capacity to 18 000 tons of ore per day. The Eldorado Nuclear milling plant at Uranium City, Saskatchewan, is also under renovation to be able to double its capacity within the next five years. The former Faraday mill in the Bancroft area of Ontario is another operation that is being renovated and should be on stream in 1976 with a capacity of 1500 tons per day.

The following is an updated review of the ore processing steps involved in the recovery of uranium in Canada.

GRINDING

Although there is no new major development in grinding techniques in the Canadian uranium mills, semi-autogenous grinding is being incorporated into the new Gulf Minerals mill in northern

¹ Tons are short tons throughout the paper: 1 short ton = 2000 lb = 907.18 kg = 0.90718 tonnes.

Saskatchewan. This method has been used in other mineral processing plants, but as far as is known there is only one other semi-autogenous mill in the uranium industry in North America, namely in Wyoming, USA.

If this method of grinding proves to be successful in the new uranium plant, one can foresee that it will be incorporated into future plants which are at present on the drawing boards.

PRECONCENTRATION OF URANIUM MINERALS

Preconcentration has been investigated during the past twenty years by methods of flotation, heavy-media separation, radiometric sorting, tabling, high-intensity magnetic separation and photometric sorting. It is realized that preconcentration offers the possibility of reducing the processing cost of present-grade ore and also the capital investment by expanding the production of present plants to meet future capacity requirements. Only radiometric sorting has been used commercially to reject waste rock and acid consumers. Ore Sorters Limited² have recently reported the successful operation of a multi-channel sorter operating on -6 in + 2 in (-150 mm + 50 mm) and -2 in + 1 in (-50 mm + 25 mm) on an American uranium ore.

EXTRACTION METHODS

In Canada, all of the uranium produced is extracted by leaching the ores with sulphuric acid or carbonate solution. By far the largest amount is produced by the sulphuric acid method. The rapidly rising cost of sodium hydroxide has prohibited the application of the basic leach to ores which might have been treated economically by that method only a few years ago.

Whether the sulphuric acid is to be made on the site or purchased ready-made is a case of economy related to the accessibility of the property to established sources. Since many of the newer discoveries of uranium are in remote areas, it is reasonable to assume that, in future, acid production plants will be seen closely associated to mills.

One technique that was borrowed from non-destructive testing and applied to uranium processing is the fluorescent dye technique. This method has been tested recently in the laboratories of CANMET of the Department of Energy, Mines and Resources in Ottawa [1]. The technique can be used to confirm the presence of minute fractures and porous mineral constituents. It is of particular interest when an assessment is being made of the possibility of leaching coarse ore by percolation leaching methods where there is a need for penetration of the lixiviant into the ore. The indicator dye is Zyglo ZL 15³, a water-washable penetrating oil which fluoresces bright green under ultra-violet light. The seepage of the penetrant through the specimen is monitored by observing the amount and level of fluorescence produced on the rough surfaces of the ore specimen.

Kerr Addison Mines Ltd., Agnew Lake Mines Ltd. and the Department of Energy, Mines and Resources undertook a co-operative programme during the past few years to leach coarsely broken ore using a bacterially active sulphuric acid medium. Bench scale tests, a pilot plant test and an underground test on a 50-ton sample of broken ore showed encouraging results [2]. In the pilot plant test, 70% of the uranium in the ore was dissolved in one year from broken ore of -8 in size. This was increased to more than 80% during the first two years. In the underground test, more than 70% of the uranium was dissolved in a little more than half a year. Unfortunately, this underground test had to be discontinued in 1971 when the company decided to put the property on a care and maintenance basis because of market conditions.

² Ore Sorters Ltd., 839 Erskine Street, Peterborough, Ontario, Canada.

³ Manufactured by Magnaflux Corp., 7300 West Lawrence Ave., Chicago, Illinois, USA.

The present improved market for uranium has renewed the interest in using the underground leaching technique as a primary method of extracting uranium from its ore. The most recent test is being done underground at Agnew Lake Mines on a 110 000 ton block of broken ore.

Some interest has been shown in using nitric acid as the lixiviant for uranium ore. This leaching technique, although believed to be more costly than the sulphuric acid method, offers some distinct advantages because of the possibility of recycling and recovering most of the leaching agent.

Strong acid leaching using sulphuric acid, as developed by the United Kingdom Atomic Energy Authority and by Pechiney Ugine Kuhlmann of France, has been examined and shown to be applicable to Canadian ores but as yet has not been incorporated in any of the expansion plans of existing mills or in the plans of the new mills.

SOLID-LIQUID SEPARATION

Two-stage filtration and countercurrent decantation are the methods employed in Canadian mills for solid-liquid separation. Recently, solvent-in-pulp systems have been examined because these offer potential economic advantages of lower operating costs than a conventional mill and lower capital costs for mill expansion.

The Rio Algom Mines have tested their pulp in a pilot plant in a 10 in. dia., 20 ft high, pulsed, perforated column, in series with a solvent stripping stage. The solvent used was 5% V/V Alamine 336 and 5% Isodecanol in kerosene. Solvent loss problems were encountered when sulphides of the ore were present. The problems were minimized when more than 90% of the sulphides were removed by prior flotation. The technique has not yet been incorporated in any of the operating mills in Canada.

The use of the resin-in-pulp ion exchange system is also being considered as a method of treating the slime portion of the pulp. Only a sand-slime separation is required for the resin-in-pulp system.

URANIUM RECOVERY FROM SOLUTION

Another active development in uranium processing is taking place in solution treatment.

Mills built in the late 1950s and employing sulphuric acid leaching used ion exchange exclusively. Plants in remote locations used chloride elution while those nearer a source of supply used nitrate elution, which is faster. Today the use of nitrate is discouraged by the environmentalists, and the question of solution purification is being re-examined. Chloride elution will be used in one new plant and Eluex, sulphuric acid elution followed by amine solvent extraction, is being considered for another.

One of the costly operations is the removal of the finely suspended solids after the primary solid-liquid separation, a process which produces a crystal-clear solution for the conventional ion exchange or solvent extraction processes. The development of a continuous ion exchange with upflow solution movement offers an alternative to the costly clarification procedure.

One such system is the Himsley column⁴. In brief, the special features of this column are as follows:

- (1) A truly continuous countercurrent flow is achieved;
- (2) Clear or turbid liquids can be treated;
- (3) No mixing of resin from one zone to another can occur at shut-down, and rapid start-up can be easily achieved;

⁴ Developed by Himsley Engineering Ltd., 250 Merton Street, Toronto, Ontario.

- (4) No valves close on the resin;
- (5) Minimal loss of resin owing to attrition is achieved by employing low velocities for resin transport;
- (6) Standard grades of resin may be used.

A 24 in dia. Himsley column has been tested recently under plant conditions. The results are encouraging.

Another trend is to use solvent extraction as a means of upgrading and purifying the leach liquor. The new Gulf plant in northern Saskatchewan is using solvent extraction with an amine as the key reactant. The design calls for conventional mixer/settler units.

PRECIPITATION

In the early days of precipitation of yellowcake, magnesium oxide was the main reagent used. This reagent soon gave way to ammonia, which produced a higher grade of product and is more convenient to handle. Today, with the growing awareness of pollution problems, there is a finger being pointed at ammonia as an undesirable constituent in plant effluents. It is not surprising therefore that the precipitation technique is altered in some way in new plants. One plant will revert to magnesia precipitation, and the use of hydrogen peroxide is being investigated by others.

BY-PRODUCT RECOVERY

In Canada, large volumes of barren liquor are produced which are relatively free of uranium but contain thorium and rare earths. During leaching, about 80% of the thorium and about 75% of the yttrium is dissolved, but only 20% of all the rare earths. In the past, both Rio Algom Mines and Denison Mines recovered a bulk rare-earth concentrate by solvent extraction. The limited market has forced the Rio Algom circuit to be closed down. Denison is still producing an yttrium-rich precipitate from the barren solution.

The method [3] involves a pH adjustment of the barren solution to remove thorium and then a readjustment of the pH to allow a solvent extraction step, using Di-(2-ethyl hexyl) phosphoric acid, to effectively separate yttrium and other rare earths from the solution. The loaded organic solution is stripped with nitric acid and the strip solution is neutralized with ammonia. The precipitate is then filtered and dried before packaging.

TAILINGS DISPOSAL AND REVEGETATION

With the present concern about pollution from uranium milling operations, tailings disposal is now considered an important step in processing uranium ore. Every effort is being made by the uranium companies to reduce the impact on the environment from any pollutant in the mining and milling operations. Today's trend is to maximize the recycling of water and solutions, and all uranium companies now use total impoundment of solids.

The operating mills neutralize all the acid with lime and oxidize the ferrous iron to ferric iron by blowing air through the pulp prior to discharging it to the waste disposal area. All the plants use barium chloride to coprecipitate the radium-226 with the tailings. This reduces the radioactivity to a low level. Final clean-up can be done by a secondary treatment, this being double ponding using barium chloride between stages. Other companies neutralize their solid tailings separately from their barren tailings and precipitate the radium from each of these with barium chloride.

A problem that has developed is that of worked-out or stand-by mines where there are large tailings areas from former operations. Two major problems are connected with these areas, one is wind erosion of the solids and transportation to unrestricted areas and the other is percolation of water through the piles into the groundwater or natural drainage.

Efforts are being made to combat wind erosion by revegetation, and although some Canadian companies have up to 15 years experience with tailings revegetation, it is only in the past five years that interest in the problem has become general throughout the industry.

There is considerable work to be done to establish self-sustaining crops, and the ease with which these crops can be developed depends to a large extent on the mineral composition of the tailings deposit.

In some areas the seepage from abandoned tailings areas is being treated with lime to remove the heavy metals and with barium chloride to remove radium-226.

Rio Algom Mines Ltd., in co-operation with CANMET Laboratories of the Department of Energy, Mines and Resources, is now investigating the effect of various ground covers on the quality and amount of seepage from controlled tailings areas. It is hoped by this programme to reduce and possibly prevent further pollution of the environment.

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DISCUSSION

S. AJURIA-GARZA: I would like to ask you about bacterial leaching. Which strains are you using and were these strains obtained from a collection or were they isolated from mine waters? We were trying to develop bacterial leaching in Mexico too but we have had the problem of acclimatizing the strains.

F.C. LENDRUM: We have never used a particular strain of bacteria. We have taken the bacteria that grew in the local mine water and started with them. They are the *Thiobacillus thiooxidans* and *ferrooxidans*. The original strains were isolated in Elliot Lake and I think they are probably still persisting. I do not believe that the bacteria themselves, in a bacterial leach of uranium, have much to do with the dissolution of uranium but rather that they produce ferric iron and sulphuric acid and this lixiviant will certainly dissolve uranium rather quickly. Therefore, you

really have to start with a local strain of bacteria and acclimatize it to your own solutions, to increase the uranium content. Sometimes the bacteria will die and you have to start again. Once continuous mining starts, the bacteria will persist.

K.M.V. JAYARAM: Regarding the use of bacteria, several meetings on this topic have been organized by the International Atomic Energy Agency during the last two or three years. Regarding indigenous bacteria, that is, the bacteria present in the ore itself, they are certainly far superior in their action than inoculated cultures. When you inoculate cultures, the nutrient media required for bacterial growth are likely to be costlier than the reagent savings. So far, it is probably not clearly established whether bacteria have any direct role in dissolving uranium. A recent paper from Yugoslavia has reported a lower-order type of reaction where uranium is oxidized by bacteria, but on an industrial scale no proper picture has so far emerged.

I have one more question. Is there any cost analysis of this type of bacterial leaching technique in comparison to conventional techniques and, if so, how does it compare?

F.C. LENDRUM: It is difficult to give a general answer to the last question. Some feasibility studies have been made but most of them are treated as confidential information by the companies. With the present Canadian approach, where ore does not have to be moved from underground and crushed and agitated, there must be some cost saving.

K.M.V. JAYARAM: Do I understand correctly that hoisting the ore from the underground mine to the surface and leaching it on the surface should be avoided?

F.C. LENDRUM: In the work done by Agnew Lake and the Department of Mines, the concept was that the ore would be developed underground in stopes — a conventional type — but much like what is being done in France. The swell or extra ore would be hoisted to the surface and leached on surface. The other ore would be left in the stope and therefore not have to be moved. This is in itself quite a saving. Neither the ore underground nor the ore on surface is crushed any finer than it is broken underground. Miners will know that when ore is developed underground, the development ore is much finer than would normally be found in stope practice, therefore the ore being hoisted to the surface is somewhat smaller, certainly less than eight inches (approx. 20.3 cm). One of the main things to consider is whether the leaching solution will penetrate these 8-in pieces of ore. I think it is fairly obvious that a solid piece of rock is much like a water glass — it just does not absorb water and water will not penetrate it. The water or lixiviant must penetrate the cracks and microcracks in the rock. In Agnew Lake ore, fortunately, the uranium minerals are also in the cracks. The fluorescent dye technique can be used to assess the possibility of leaching coarse ore by percolation leaching methods (see Kaiman's report, Ref. [1]).

K.M.V. JAYARAM: One more question pertains to the time taken for leaching in this particular process as compared to the cost involvement, annual turnover and crushing. Do you not think that a certain amount of crushing would be advisable rather than prolonging the leaching process?

F.C. LENDRUM: Once again, this is a matter for economics at individual properties. I would be very reluctant to suggest that somebody should take the data for any one property and use it at some other property. This could lead to great trouble.

F.R. HARTLEY: I have three questions. Firstly, I wonder if you could enlarge a little on the semi-autogenous grinding operation as against autogenous grinding which you referred to in connection with Rabbit Lake. Secondly, you made no mention of clarification operations. I think the adoption of the continuous precoat filter probably comes mainly from Canadian operation. I wonder if you still plan to use continuous precoat filters or whether you consider sand clarification or other types of operation in the future. Precoat filters proved fairly expensive in Australia and we certainly think of reverting to sand clarifiers. Thirdly, you made no mention of either the nature of the ion exchange resin or whether any work has been done on the new weak-base macro-reticular type resins.

F.C. LENDRUM: I will answer these questions in a slightly different order. As regards clarification, I would say that we are considering the use of sand clarifiers rather than precoat filters, but furthermore we are hoping that upflow ion exchange (as used in the Himsley Column) will

eliminate the problem. As regards ion exchange in Canada, all the ion exchange plants of which I have knowledge are still using strong-base resins, the IRA 400 or IONAC 590 varieties. Some test work has been done with the weak-base resins Roman Hass 270 and 299, but these resins are very light and they are not applicable to upflow-type columns. We are hoping that a weak-base resin that can be used in upflow columns will come on the market. Not too long ago, such a resin was tested. The loading was excellent, but unfortunately it was almost impossible to remove the uranium, so it did not quite work out the way they had hoped. There are rumours that there are one or two other weak-base resins on the market, but I have not seen any recent work on this subject.

D.C. SEIDEL: I do not know of any major new developments on weak-base resins, but several mills in the USA are using these resins. These mills include the Union Carbide Corporation mill at Uravan, Colorado, and the Utah International mill at Shirley Basin, Wyoming. Both of these mills use conventional column ion exchange systems. I also agree that the specific gravity of the present weak-base resins is relatively low and that they are not heavy enough to use in the upflow column ion exchange systems.

F.C. LENDRUM: We have one more question from Mr. Hartley, about autogenous grinding. Mr. Dakers can answer this.

R.G. DAKERS: There is the autogenous grinding method and the so-called semi-autogenous one, in which steel balls are used. We have employed autogenous grinding in Beaverlodge ores during the last ten years. We attempted originally to use semi-autogenous grinding but we found insufficient coarse material from our underground supply and we added six-inch (approx. 16.2 cm) steel balls to the mill. We shall continue to do this, and it has been the general practice with base metals in Canada to start off with an autogenous mill and end up with a semi-autogenous mill by adding steel.

T.S. ARY: What is the make-up of the ore matrix in the stopes?

F.C. LENDRUM: The ore in the stopes is a typical conglomerate of the Canadian uranium fields. It has quartz pebbles in a quartz matrix with some pyrite (not really large amounts). In the case under discussion, the uranium mineral is mainly uranothorite with some uraninite.

T.S. ARY: The reason for my asking is that, in my experience, when a stope is filled, the fines will settle down, and when any kind of acid-containing liquid runs through it or when the ore contains pyrite and an acidic complex is set up, the stope will become blocked and impervious. Speaking of the typical Canadian ore, how far can it be penetrated by the leach liquor?

F.C. LENDRUM: After two years' work in a simulated stope we found no binding. The pH was held low, at about 1.5 to 1.8. This is published data on the stope. As to how far into the sides of the rock we have penetrated, I can give you some data from Kaiman's report: on one specimen the ore was penetrated 3 in (= 7.62 cm) in about 66 hours and 4½ in (= 11.43 cm) in one month. With other very dense pieces there was literally no penetration. Fortunately the pieces allowing maximum penetration were also those that contained most uranium, and the uranium was in the cracks. I think this emphasizes what I said earlier, that one should not take data from any one mine and try to apply it directly to another mine. One should start with penetration studies first to see whether the system is really feasible.

W.R. FLÖTER: I think I should give some more data on the autogenous grinding that is being used at the Rabbit Lake mill. According to the accounting, the investment costs are reduced to about 40% compared to conventional grinding and milling. The power installed is reduced by 50% and the wear during operation is also reduced by 40–50%. The wear in the Rabbit Lake mill is about 300–500 g/ton of ore with liners, balls and bars. In the rod-mill/ball-mill section it is more than 1000 g/ton. At Gulf Minerals mill, we are feeding run-of-mine ore. This ore is 30% + 20 in. It is passed through a grizzly and then directly fed to the mill. We are very satisfied now, after finishing the start-up period. The grind is very favourable, the sizes are very good, and the circulating load of the mill, which is operating in closed circuit with cyclones, is also working well.

K.M.V. JAYARAM: What is the rock matrix? Is autogenous grinding universally applicable? Our experience has been that we start with it and are obliged to shift from it unless the characteristics

of the ore permit continuation because of harder parts in the ore. Can autogenous grinding be universally applied to all types of uranium ores as is made out in this presentation?

W.R. FLÖTER: I cannot tell whether it can be used for your ore but it could be used in some of the Canadian ores.

F.C. LENDRUM: In the Elliot Lake camp we changed to a semi-autogenous type of grinding in 1959 and 1960 but this was a different type of semi-autogenous technique. The original grinding method used at the Elliot Lake camp was rod-mill grinding, followed by conventional ball-mill grinding. It was found that it was better to take pebbles from the primary crusher and use these pebbles in place of the balls in the secondary grinding. This meant that the mills had to be redesigned. In some cases they were lengthened, in some cases they were made of a larger diameter. This type of semi-autogenous grinding had the advantage of cutting down the amount of iron and therefore the amount of acid and chlorate that had to be used, and the chemical savings were as great as those on steel. I might point out that those plants which could take the pebbles from the primary jaw crusher certainly had less wear than those which had to take them at the secondary stage. I believe that now all crusher houses have been converted so that they can take up pebbles from the jaw crusher discharge. It is necessary to do test work on each ore to check its suitability for forming useful pebbles.

K. PINKAS: You have mentioned the application of strong acid leaching with sulphuric acid. Do you mean that this is an acid cure? What concentration of sulphuric acid do you use for this method and what savings can be achieved by applying it?

F.C. LENDRUM: For strong acid leach testing we used about 10N sulphuric acid. It would probably be advantageous to start with a completely dry grind but it may also be possible to run a grind of, say, 3 to 4% moisture. The curing on some ores can be as short as one hour, on others it may take 6 or 8 hours. I believe that in Niger the curing is short enough to be done in a kiln and I know of Canadian ores that would certainly fit into that category. I know other Canadian uranium ores that would need 6 hours' curing, in which case the handling problems might be quite severe.

K. PINKAS: Do you apply room temperature for the strong acid leaching process?

F.C. LENDRUM: No. Just under 100°C, so that the sulphuric acid will not come off too quickly. With the small amount of water used, the advantage is that the temperature can be raised cheaply.

M. SHABBIR: I would like to know something about the physical upgrading techniques that you have applied. In the light of the increase in the cost of yellowcake, would you say that for processing the low-grade ores these gravity separation techniques are now feasible?

F.C. LENDRUM: With the exception of radiometric sorting, I have never found any pre-concentration or gravity circuit that would be economic, because it is not possible to make a clean enough separation at current uranium prices. As to radiometric sorting, I think each ore would have to be tried individually to find out whether the rejects are completely blank. One interesting point made by the people who built the equipment is that the little uranium left in the rejected waste would cost US\$160 per pound to recover because of the high acid consumption.

F.E. MCGINLEY: You referred to the use of nitric acid as a leaching agent. Is this only for uranium extraction or were you considering the extraction of other metals as well?

F.C. LENDRUM: There have been some discussions in Canada that by using a nitric acid leach, one would be able to recover the radium and keep it out of the tailings. Nobody has explained what to do with the radium when it is isolated. There is one mine in Canada where the throughput would be quite small but the ore grade is extremely high – in the range of 20% U_3O_8 . For this, the nitric acid approach might be quite interesting, but this is an isolated case and I do not know whether it is economically justified. [Note added after the meeting: Later information is that a nitric acid leach will not be used but that gravity concentration will be applied at the mine.]

P.A. BONHOTE: I was interested in your comments on the new method of handling base-metal tailings at high concentrations. Could you comment on the likely environmental benefits resulting from the final form of the tailings disposal area? Would you have a more stable tailings pile than with the conventional type of approach?

F.C. LENDRUM: Dr. Robinsky's approach⁵ should lead to a much more stable tailings area. The danger of movement of the tailings dam is decreased because, with the denser pulp and the fines included in it, a stable hill will result rather than a big mound of tailings. The slopes will be much flatter than the front of a tailings dam, with again less danger of movement. There are no decantation towers or buried pipelines in this type of dam because the pond is at the front or near the low part of the dam. We made the dams of new material and built them to water dam specifications rather than tailings dam specifications. The erosion on the downstream side is practically zero and the fact that the whole tailings area is wet keeps dust and erosion problems to a minimum. We can keep the pond during its operating time at a pH of about 8 or 8.5 and this means that we should have literally no acid seepage. At the end of operative life of the dam it is intended to fill the pond, revegetate the surface and let it regenerate.

⁵ ROBINSKY, E.I., Thickened discharge — a new approach to tailings disposal, Can. Min. Metall. Bull. (Dec. 1975).

RECENT TRENDS IN RESEARCH AND DEVELOPMENT WORK ON THE PROCESSING OF URANIUM ORE IN SOUTH AFRICA

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Abstract

RECENT TRENDS IN RESEARCH AND DEVELOPMENT WORK ON THE PROCESSING OF URANIUM ORE IN SOUTH AFRICA.

The important role that uranium will play in solving the energy problems of the world and the need for a continuing high level of research and development activity in the processing of uranium ore are fully appreciated in South Africa. The rapid increases in the price of gold and uranium in recent years have coincided with an unprecedented increase in working costs at South African gold mines. A re-examination of the existing flowsheets for the recovery of uranium, gold and pyrite from Witwatersrand ores, in the light of these economic trends, has resulted in the identification of a number of profitable areas for research and development. The main topics under investigation in South Africa in the processing of uranium ore are the use of physical methods of concentration such as flotation, gravity concentration and wet high-intensity magnetic separation; the wider adoption of the 'reverse leach', in which prior acid leaching for uranium improves the subsequent extraction of gold; the use of higher leaching temperatures and higher concentrations of ferric ion in the leach to increase the percentage of uranium extracted, including the production of ferric ion from recycled solutions; the application of pressure leaching to the recovery of uranium from low-grade ores and concentrates; the development of a continuous ion-exchange contactor capable of handling dilute slurries, so that simpler and cheaper techniques of solid/liquid separation can be used instead of the expensive filtration and clarification steps, and the improvement of instrumentation for the control of additions of sulphuric acid and manganese dioxide to the leach. A brief description is given of the essential features of the new or improved processing techniques under development that hold promise of full-scale application at existing or future uranium plants.

1. INTRODUCTION

At the IAEA meeting on this subject in 1970 [1] the Panel concluded that, because of the oversupply situation and depressed prices for uranium at that time, the degree of support for research and development of new and improved processing methods was generally low, in spite of predictions of a strong and imminent upsurge in the demand for uranium and hence for new plant design and construction. It was also noted that, because of the time lag of several years between the proving of new processes and the commissioning of new plants, it would be necessary to give urgent attention to the development and testing of novel ideas if these were to be incorporated in the new plants that would be required during the latter half of the 1970s.

In South Africa, everyone concerned with the processing of uranium ore has fully appreciated the important role that uranium will play in solving the energy problems of the world and the need for a continuing high level of research activity in this field. Thus, over the last few years, the Atomic Energy Board, the National Institute for Metallurgy, and the Nuclear Fuels Corporation of South Africa have continued to collaborate very actively on uranium research and development with mining groups and with individual mining companies.

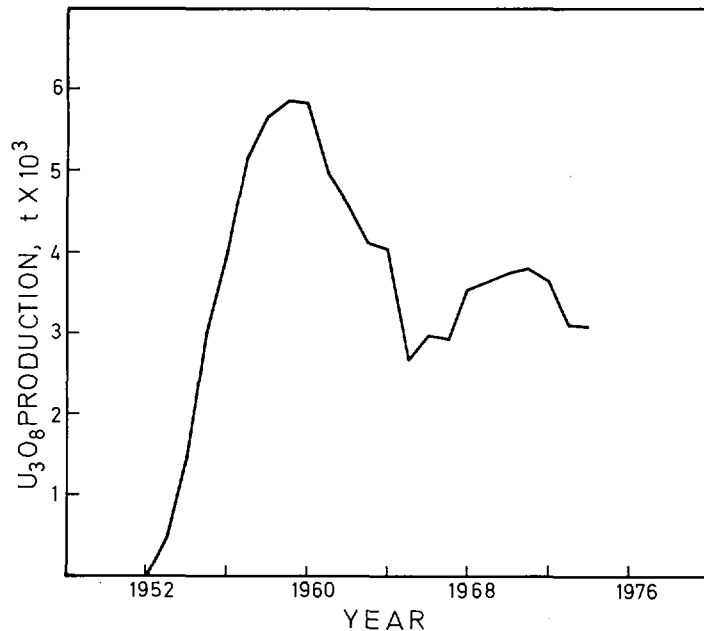


FIG.1. South African uranium production.

The purpose of this paper is to identify and describe briefly the work being done to improve the processing techniques that hold promise of commercial application in the new treatment plants likely to be constructed in South Africa during the next five years for the recovery of uranium, together with gold and pyrite, from Witwatersrand ores.

2. EXISTING METALLURGICAL CIRCUITS

2.1. Influence of changing market conditions on the construction of new plants

The general pattern for the design of uranium plants in South Africa was set during the comprehensive research and development programme of 1946–1956 [2]. The first full-scale uranium plant resulting from this programme was commissioned in 1952, and by 1957 a total of seventeen plants had been erected. Although a considerable amount of individuality was expressed in the design of these plants and the extent of this individuality increased with later plants, they were all essentially based on the results of this initial research programme.

Figure 1 shows how the rapid initial expansion of the uranium industry was dramatically reversed in 1960 by the sudden reduction in the uranium requirements of the Combined Development Agency, an organization representing the governments of the USA and the United Kingdom, with whom supply contracts for the entire South African uranium output had been concluded in the early 1950s. From 1966 on, production gradually improved when numerous small and often short-term contracts were negotiated with electrical utilities as the prospects for nuclear power began to improve [3]. During 1970–1971, three new plants, at Blyvooruitzicht, West Driefontein

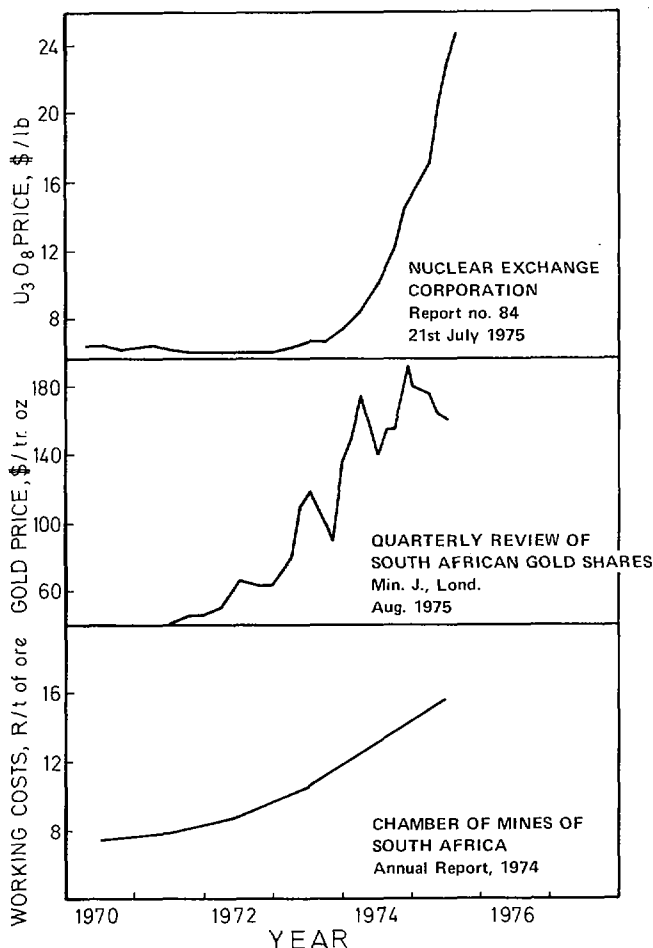


FIG.2. Trends in gold and uranium prices and working costs at South African mines.

and Western Deep Levels gold mines, came into production in anticipation of an upturn in the market, but the expected improved demand did not materialize and the mines were unwilling to continue production at the 1971 level (3800 t of U₃O₈) in the face of continually falling prices. A fourth new plant, at President Brand gold mine, completed in 1971, has not as yet been commissioned. The 1975 production will again be of the order of 3000 t of U₃O₈.

During 1973–1974, there was a spectacular rise in the market price for gold [4] and during 1975 a very rapid increase in the price of uranium [5]. In addition, the demand for pyrite in South Africa, associated with the gold and uranium in Witwatersrand ores, is expected to increase very sharply during the next ten years [6]. However, these market changes have coincided with an unprecedented increase in working costs at South African gold mines [7] (see Fig.2). Also, the gold mines take advantage of a higher gold price by mining ore with a lower grade of gold,

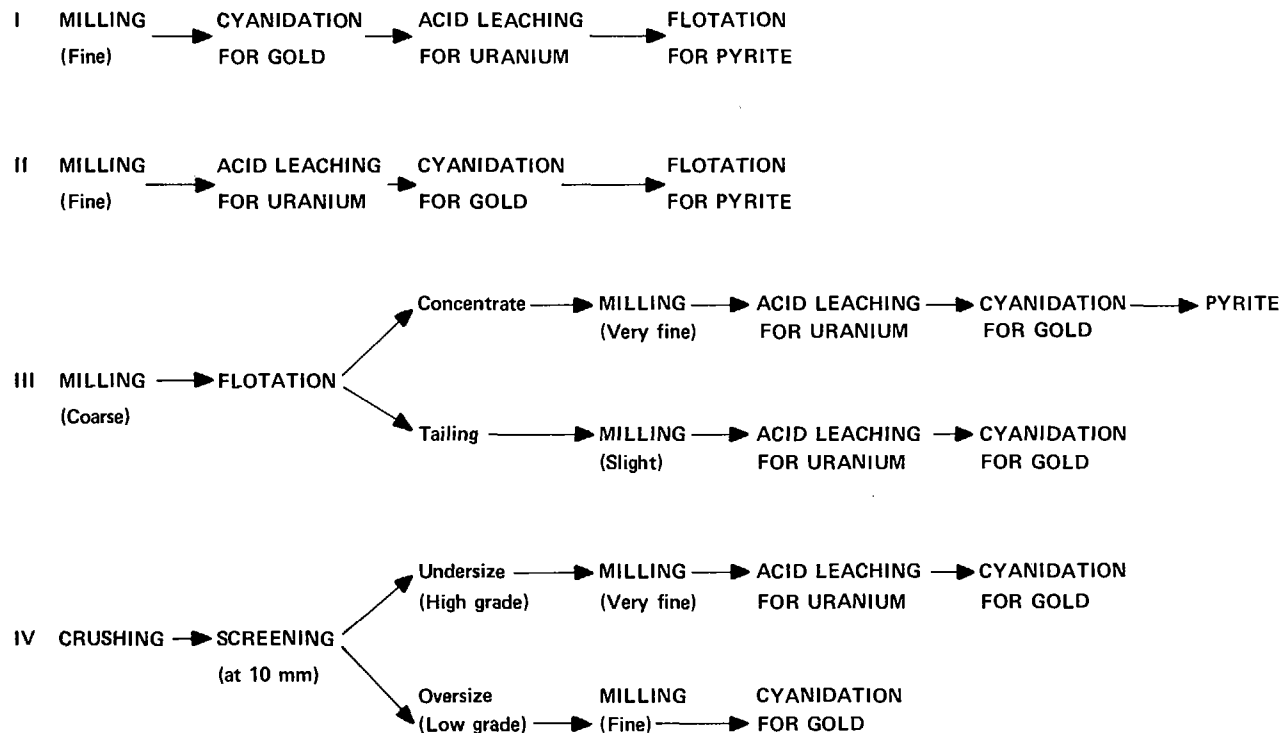


FIG.3. Established procedures for the recovery of uranium, gold and pyrite from Witwatersrand ores.

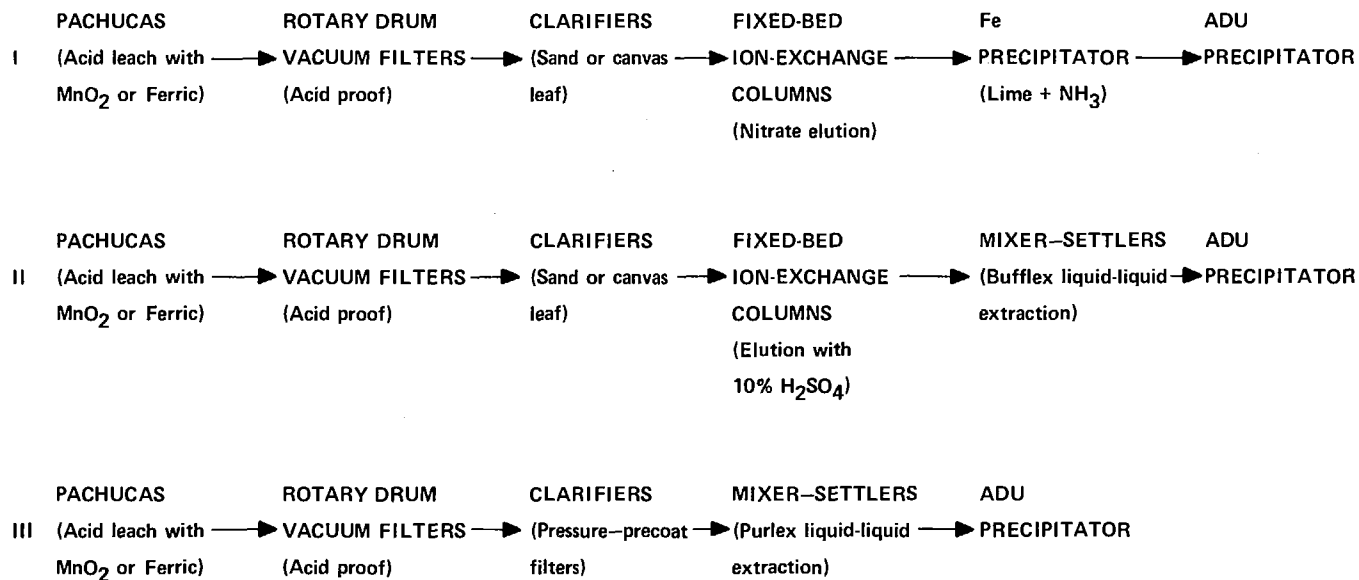


FIG.4. Established flowsheets for uranium plants in South Africa.

which generally also means a lower grade of uranium, and the immediate consequence has been a tendency for the uranium output of existing plants to decrease. By-product uranium contributes only a small part of the profits of the combined gold-uranium operation; at present prices, the revenue from gold is approximately twenty times that from uranium. Thus the economic feasibility of an expansion in uranium production capacity, particularly for the treatment of low-grade slimes dams, is strongly dependent on the development of processes that are cheaper than those employed at present.

2.2. Overall recovery procedures for uranium, gold and pyrite from Witwatersrand ores

The established procedures for the recovery of uranium, gold and pyrite from Witwatersrand reefs can be classified into four main types [8] as shown in Fig.3.

In most of the early uranium plants, acid leaching followed cyanidation (Procedure I), mainly because this involved no change to the existing gold circuits. Prior acid leaching (Procedure II) improves the subsequent extraction of gold (see Section 5). For some ores, prior flotation (Procedure III) can be used to produce a concentrate containing up to 80% of the gold, 80% of the pyrite, and 25% of the uranium in less than 10% of the original mass of ore. Because of the very small mass of this concentrate, it can be milled very finely and treated intensively to yield very high recoveries of gold and uranium. The overall advantage is a saving in milling costs for the bulk of the ore, which does not undergo this intensive treatment. At other mines, a size split of the crushed ore (Procedure IV) results in a fine fraction (approximately 30% of the original mass of ore) that contains two-thirds of the gold and more than half of the uranium. This fine fraction is given an intensive treatment, again with an overall saving in milling costs for the bulk of the ore.

2.3. Flowsheets for uranium recovery

The basic flowsheets that have been used in the design of uranium plants in South Africa can be classified into three main types, as shown in Fig.4.

Flowsheet I was universally adopted for all 17 uranium plants originally constructed. Following pilot-plant demonstration of the Bufflex process [9], the uranium plant at Harmony gold mine changed to Flowsheet II. The successful pilot-plant demonstration of the Purlex liquid-liquid extraction process [10] resulted in the conversion of the Harmony plant and of five of the seven original plants still in operation at that time to Flowsheet III, and the four new plants built in 1970–1971 all used this flowsheet. Two of the original plants are still operating on the ferric-leach variation of Flowsheet I.

3. MOST PROFITABLE AREAS FOR RESEARCH AND DEVELOPMENT

In the light of the extensive research and development already done in South Africa on the processing of uranium ore, it was not immediately apparent five years ago whether any substantial advances could be made by further research. However, a re-examination, in the light of predicted new developments in metallurgical techniques and economic trends, of the basic decisions taken when the early flowsheets were drawn up resulted in the following conclusions on the most profitable areas for research.

(a) The full potential of the physical concentration methods, such as flotation, gravity concentration and wet high-intensity magnetic separation, had not been adequately explored for the upgrading of newly mined ores or accumulated slimes with a uranium grade below the cut-off point for 'economically leachable' material.

(b) The increased attention being given by the gold-mining industry to increasing their overall efficiency of gold extraction indicated that an investigation into the wider adoption of the 'reverse leach' might have considerable merit.

(c) Reliable experimental data were insufficient for an adequate economic evaluation of increased temperature and ferric ion concentration in the acid leach in order to raise the uranium extraction above the level conventionally achieved in South Africa. In particular, the full potential of the ferric leach had not been realized at Purlex plants because there were no successful techniques for the recycling of raffinates.

(d) Although the high capital cost of pressure leaching of Witwatersrand ores had discouraged its introduction in South African uranium plants, the increases in the price of gold and uranium have warranted a re-examination of the process on a pilot scale.

(e) Although the Purlex liquid-liquid extraction process had resulted in significant savings in operating costs compared to those of the original fixed-bed ion-exchange process, its successful operation required a very clear feed solution. The filtration and clarification steps used to produce this clear solution account for a substantial part of the operating and capital costs of a typical South African uranium plant. There was therefore a strong incentive to develop an alternative system of uranium recovery that would be capable of handling the pulp directly from leaching or after partial solid-liquid separation. This would eliminate the expensive filtration and clarification steps or would replace them with simpler and cheaper techniques.

(f) Although extensive use had been made of control instrumentation in the design of existing uranium plants, certain instruments had not performed satisfactorily and substantial benefits were to be gained from the development of improved instrumentation to control the additions of sulphuric acid and manganese dioxide to the leach.

4. PHYSICAL METHODS OF CONCENTRATING WITWATERSRAND ORES

4.1. Gravity concentration and flotation

The mineralogy of Witwatersrand ores and the initial investigations on the application of gravity concentration and flotation to run-of-mine ores and cyanide residues have been authoritatively described [2,8,11,12]. During the past five years, valuable additional data have been accumulated on the amenability of a large number of ores and residues to these two techniques. More attention to the upgrading of the gold and pyrite content, which occurs as a consequence of their close association with the uranium-bearing minerals, has led to a better understanding of the economic potential of these techniques.

Flotation is being examined as a potential procedure for the commercial treatment of old tailings dumps, and it has been reported that a flotation plant with a monthly feed of one million tonnes of slimes is to be erected at South African Lands Gold Mine for this purpose [8]. The large capital expenditure required is economically justified by the values of the pyrite, gold and uranium recovered from these residues, and by the removal of old dumps from areas that have become valuable as real estate to locations of less value.

4.2. Wet high-intensity magnetic separation

It has long been known that some of the uranium-bearing components of Witwatersrand ores are feebly magnetic, but serious consideration has only recently been given to magnetic separation as a means of uranium concentration with the advent of wet high-intensity magnetic separation.

Laboratory results on a large number of low-grade Witwatersrand ores and gold-plant tailings have shown that up to 70% of the uranium and 50% of the residual gold can be recovered in a magnetic concentrate representing, in many instances, less than 10% of the mass of the original feed [13].

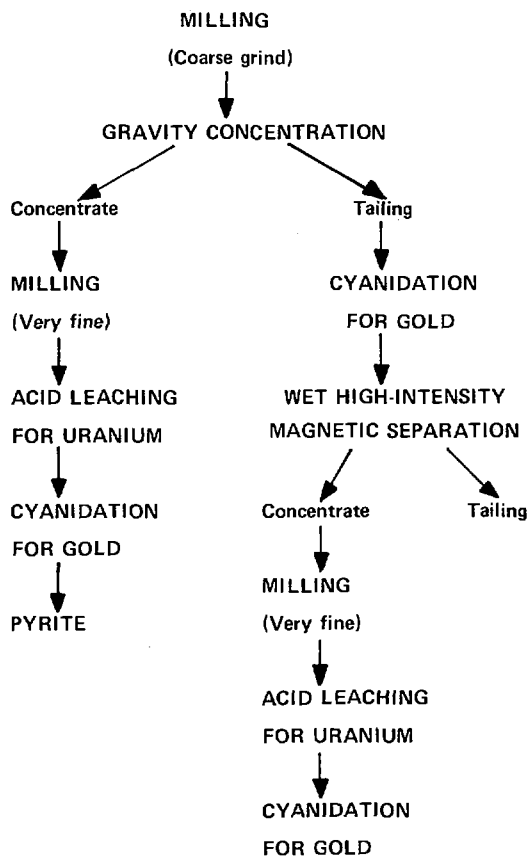


FIG.5. Proposed new procedure for the recovery of uranium, gold and pyrite by the use of a coarse grind with gravity concentration and wet high-intensity magnetic separation.

The selection of suitable industrial equipment is the next step in the work and will be followed by large-scale tests at one of the gold mines. Considerable development work appears to be necessary for laboratory results to be reproduced on an industrial scale, but this technique promises to be a major factor in the development of economic methods for the recovery of uranium from very low-grade ores and gold-plant tailings that were previously considered not to be worth treating for uranium.

4.3. Coarse milling, gravity concentration and magnetic separation

Encouraging laboratory results were recently obtained in an investigation aimed at developing a novel procedure for the treatment of Witwatersrand ores [13] (see Fig.5). It is claimed that this procedure has not only all the advantages of Procedure III (Fig.3), as regards reverse leaching and savings in milling costs on the bulk of the ore, but also the following additional attractions.

TABLE I. CURRENT LEACHING PROCEDURES
AT GOLD-URANIUM PLANTS

Plant	Original plant		Change to reverse leach
	Start-up date	Leaching procedure	
A	1953	Normal	-
B	1953	Normal	1970
C	1955	Normal	-
D	1955	Normal	1957
E	1955	Normal	-
F	1956	Normal	-
G	1956	Normal	1971
H	1957	Normal	1975
I	1970	Reverse	-

(a) If a salable grade of pyrite can be achieved, gravity concentration is a very much cheaper technique than flotation for the recovery of pyrite. (The operating costs for gravity concentration are clearly much lower since no reagents are required and the capital costs can be as little as one-fifth of those for flotation.)

(b) The intensive treatment of the gravity and magnetic concentrates would result in a very high overall recovery of gold.

(c) The comparatively coarse nature of the gravity tailings would endow the cyanide residue with improved filtration characteristics, and soluble losses of gold would be reduced.

(d) The production and intensive treatment of concentrates of low bulk and acceptable grade could make it possible for uranium recovery to be considered at mines where it is currently thought not to be economically feasible.

5. WIDER ADOPTION OF THE REVERSE-LEACH PROCEDURE

Prior acid leaching of Witwatersrand ores improves the subsequent extraction of gold by exposing the gold locked up in acid-soluble minerals, such as uraninite and partly soluble silicates like chlorite, and also, it is presumed, by cleaning the surfaces of gold grains through removing films and tarnishes.

Table I summarizes the present leaching procedures used by uranium-producing gold mines. Plant D has been successfully operating on reverse leaching since 1957 [14] and, when new uranium plants were constructed in 1970–1971 at plants B, G and I, the opportunity was taken to introduce reverse leaching.

With the persistent oversupply situation and the depressed uranium prices of the early 1970s, producers still operating on the normal leach were reluctant to consider a changeover to reverse leaching. However, laboratory testwork undertaken during 1973 on ore from plant H indicated that the undissolved gold in the final residues could be reduced from 0.49 to 0.24 g/t by the use of reverse leaching. These results were confirmed by pilot-scale tests at the mine and it was decided to alter the full-scale plant circuit to reverse leaching. The changeover was completed early in 1975,

and the results obtained during the first six months of operation have confirmed the laboratory predictions. It is estimated that the increased gold revenue to the mine resulting from the change-over will amount to approximately two million rand per annum before tax.

Studies now being undertaken may result in a changeover to reverse leaching at Plants A, C, E and F if similar benefits in increased gold extraction are indicated by the tests.

6. FERRIC LEACHING AND AUTOXIDATION OF RECYCLED SOLUTIONS

The autoxidation process, in which both sulphuric acid and ferric sulphate can be formed by blowing air and sulphur dioxide into solutions containing ferrous sulphate, was studied during the early stages of the development of the acid leaching process for uranium extraction from Witwatersrand cyanide residues [15]. However, the use of manganese dioxide as an oxidizing agent was later found to be more convenient and economic. Uranium extractions with the manganese dioxide leach are generally about 80 to 85%.

High additions of ferric ion, in place of manganese dioxide, were used at two of the seventeen original uranium plants [16]. These high additions of ferric ion are obtained mainly by the dissolution of the iron oxide sinter, obtained from the roasting of pyrite flotation concentrates, in high-strength sulphuric acid. Extra ferric ion is obtained by recycling, via autoxidation with sulphur dioxide, of barren solutions from the fixed-bed ion exchange plants that are still in operation at these two mines.

Following a comprehensive investigation of the reaction mechanisms and kinetics of the uranium-leaching process [17] and in anticipation of an improvement in the uranium market, laboratory and pilot-plant tests were again carried out in 1970–1971 to evaluate the economics of increasing the temperature and ferric ion concentration of the leach as a means of increasing the uranium extraction to as much as 95%. Results showed [18] that a 93% extraction of uranium could be achieved with the so-called Felix leach (64°C for 24 hours with total additions of 6 kg of ferric iron and 14 kg of sulphuric acid per tonne of ore).

It was considered, however, that further work was necessary on the testing of improved equipment for the autoxidation of ferrous to ferric ions in recycled plant solutions, and on the problems associated with the filtration of the pulps resulting from a dilute leach high in ferric ions. Difficulties were also envisaged in the recycling of barren solutions from Purlex plants because of the effect of solvent on the rubber lining of acid-proof pipes and pachucas. Tests were therefore necessary to find techniques for the effective removal of the traces of solvent to permit the recycling of Purlex barren solutions, via autoxidation, to the leach.

In addition, a spiral heat exchanger had come on the market that appeared to be capable of being used for transferring heat from the hot pulp stream leaving the last pachuca to the cold incoming pulp stream. The successful recovery of this heat would further improve the economics of the Felix leach. A detailed investigation of the design and performance of this heat exchanger was desirable [19].

Considerable progress has been made in these investigations during the past five years, and the technical and economic feasibility of the Felix leach is being re-evaluated in the light of the increased uranium demand and price and the use of improved equipment for autoxidation and pulp heat exchange.

A novel technique for the production of ferric sulphate was recently developed in South Africa, in which a moving stream of acidified aqueous ferrous sulphate solution is oxidized by a bacterial film in the presence of dissolved oxygen [20]. This technique eliminates the need for sulphur dioxide and is inherently very cheap. Suitable equipment is now being developed for its application on an industrial scale and, if this is successful, a further substantial reduction in the costs of the autoxidation step, and hence the Felix leach, is envisaged.

It is expected that at least one current uranium producer will change over to a Felix leach and that the wider application of ferric leaching will take place in due course with a resultant increase in the overall efficiency of uranium extraction.

7. PRESSURE LEACHING OF WITWATERSRAND ORES

The treatment of Witwatersrand ores with air or oxygen at elevated temperatures and pressures results in the decomposition of the pyrite to ferric sulphate and sulphuric acid, yielding uranium dissolutions of more than 95%. Pressure leaching was tested on a laboratory scale with complete success in the 1950s [21] but was not considered for the early uranium plants because of its relatively high capital cost. In addition, it was considered essential that further pilot-scale work should be done on corrosion problems to determine the most satisfactory method of transferring abrasive and corrosive pulps at high temperature and pressure and to solve the difficulties of heat transfer.

Recently, a reassessment was made of the additional benefits that might arise from an increasing recovery of gold when pressure leaching is used in a reverse-leaching process, allowance being made for advances in the design of autoclaves and heat exchangers. The results of this study were sufficiently encouraging for a collaborative industry project to be undertaken at one of the mines, involving a demonstration plant comprising a 20 m³ four-compartment mechanically agitated autoclave, an automatic boiler circulating steam through the autoclave and returning it via a spiral heat exchanger, and an oxygen storage and supply system. This plant is at present under construction, and testwork is scheduled to start early in 1976.

8. TREATMENT OF UNCLARIFIED SOLUTIONS OR DILUTE SLURRIES BY CONTINUOUS COUNTERCURRENT ION EXCHANGE

8.1. Development of the NIM CIX Contactor

For a number of years the National Institute for Metallurgy (NIM) has been investigating improvements in ion-exchange engineering technology and in particular the development of an advanced countercurrent ion exchange (CIX) contactor for use in the hydrometallurgical processing of low-grade ores [22]. The multistage fluidized-bed system with reversal of flow for resin transfer, based on the patents of Cloete and Streat, was considered the most suitable for further development. The NIM programme, which has now reached an advanced stage, can be summarized as follows:

- (a) Development of improved engineering components and control methods for the movement of resin into, through, and out of the extraction and elution columns;
- (b) Development of standardized laboratory methods for the study of the capacity, kinetics and hydraulic properties of resins;
- (c) Application of laboratory results to process design and optimization by the use of computer methods and to the analysis of plant performance data;
- (d) Fundamental back-up studies on improvements to the present techniques of process design and analysis.

8.2. Testing of the NIM CIX Contactor for the recovery of uranium

A pilot plant was established in 1972 by the Atomic Energy Board at the uranium plant of one of the gold mines to investigate the application of the NIM CIX Contactor to uranium recovery.

The investigation was designed to meet the following specific objectives:

- (a) To demonstrate the mechanics of the system and its reliability;
- (b) To demonstrate the lower resin inventory of CIX as opposed to fixed-bed ion exchange;
- (c) To test the ability of the plant to handle unclarified feed solutions;
- (d) To develop a process using weak-base resin that would produce material of the same or better purity than that of a Purlex product;
- (e) To undertake a preliminary evaluation of the suitability of the system for handling uranium-bearing solutions with a solid content of up to 10% (by mass).

The investigation has achieved all these objectives, and the results were recently published in detail [23].

8.3. Recovery of uranium by CIX

The successful development and application of the NIM CIX Contactor to the recovery of uranium has opened up the possibility of a new generation of flowsheets for uranium recovery. The basic options in the use of the CIX technique are shown in Fig.6. The main advantage of these flowsheets over established flowsheets is a reduction in overall capital cost as a result of the elimination of the clarification step and the replacement of the filtration step by cheaper and simpler solid/liquid separation techniques.

A comprehensive pilot-plant investigation was recently undertaken on the treatment of material from a slimes dam at one of the gold mines by continuous countercurrent decantation (CCD), followed by CIX with strong-base resin, elution with 10% sulphuric acid, and liquid/liquid extraction. The pilot plant was operated very successfully and a decision has been made to go ahead with the design and construction of a full-scale plant based on this circuit should planned demonstration trials, scheduled for the latter half of 1975 on a full-scale CIX column, be successful. The latter plant comprises an extraction column of 2.5 m diameter and an elution column of 1.2 m diameter, each column being approximately 10 m high.

Work is continuing on the development of alternative techniques for solid/liquid separation such as those using hydrocyclones [24], and attention is being given to the further improvement of circuits using weak-base resins.

9. IMPROVED INSTRUMENTATION FOR URANIUM PLANTS

Extensive use was made of control instrumentation in the design of South African uranium plants, particularly those constructed in 1970–1971. However, many of these instruments were supplied without adequate back-up information on their application, engineering design, installation and commissioning, and consequently they did not always perform satisfactorily.

During the past five years, much progress has been made in South Africa in the development of new instruments and in the adaptation of existing commercial devices for control purposes on uranium plants [25]. Thus, for example, a much improved instrument, based on the principle of electrodeless conductivity measurement, was developed for the control of sulphuric acid addition to the leach. At one of the plants where this instrument was subjected to full-scale trials, previous practice was for the leach operators to maintain the free acid in the leach pachucas between 2 and 10 g/l by manual methods of measurement and control. The new automatic control system using the electrodeless conductivity meter succeeded in maintaining the free acid level at 6.8 ± 0.4 g/l with a considerable reduction in sulphuric acid consumption. Other developments include much improved instruments for controlling the addition of manganese dioxide to the leach by measuring the redox potential as an indicator of the ratio of ferric to ferrous ions and for measuring the pH in organic continuous emulsions in Purlex plants.

All the existing uranium producers have now installed these new instruments with considerable savings in the costs of reagents and labour and improvements in operating efficiency.

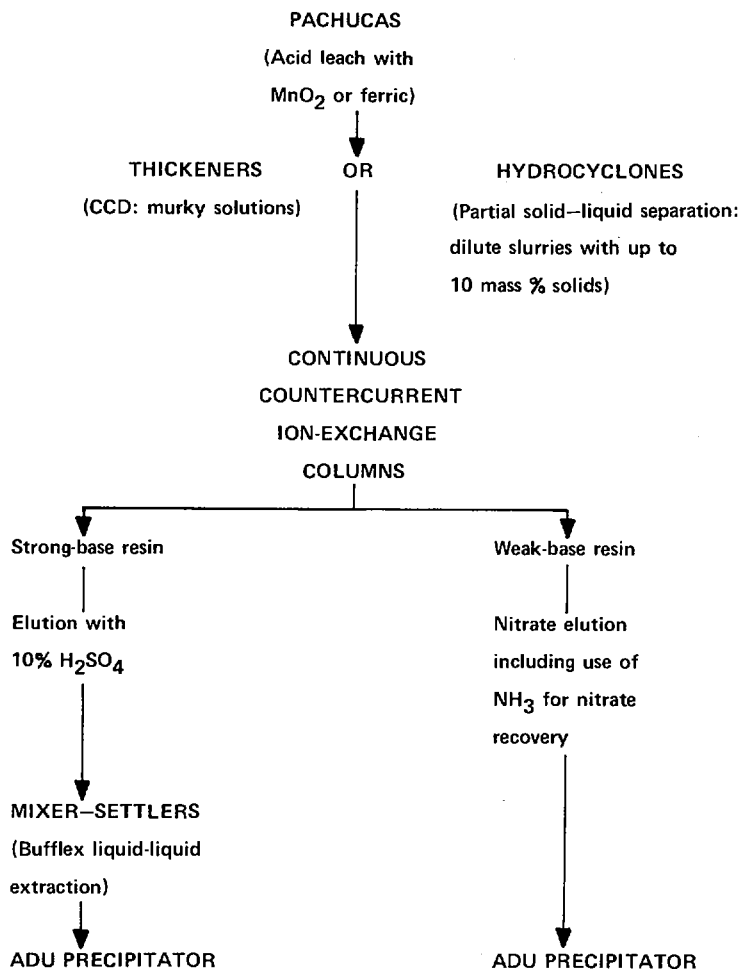


FIG.6. Proposed new uranium recovery flowsheets based on the treatment of unclarified solutions or dilute slurries by CIX.

10. CONCLUSIONS

(a) The rapid increases in the price of gold and uranium in recent years have coincided with an unprecedented increase in working costs at South African gold mines. The economic feasibility of an expansion in uranium production capacity, particularly for the treatment of low-grade slimes dams, is strongly dependent on the development of processes that are cheaper than those employed at present.

(b) Provided that laboratory results can be reproduced on an industrial scale, wet high-intensity magnetic separation may well prove to be a major factor in the development of economic methods for the recovery of uranium from very low-grade ores and gold-plant tailings previously considered not to be worth treating for uranium.

(c) The trend in South Africa is towards higher overall extractions of gold and uranium by a changeover to reverse leaching and by the introduction of more intensive conditions (high concentrations of ferric ion and high temperatures) in the uranium leach.

(d) A reassessment of the economics of pressure leaching in the light of current technology and predicted economic trends has resulted in a collaborative industry project to test it on a demonstration-plant scale. The extent of the improvement achieved in the recovery of gold when pressure leaching is used in a reverse leaching process will be a major factor in any decision to adopt this technique at existing or future uranium plants.

(e) The successful development and pilot-plant application of the NIM CIX Contactor to the recovery of uranium has opened up the possibility of a new generation of uranium recovery flowsheets, which offer a reduction in the overall capital cost of plants by the elimination of the clarification step and the substitution of cheaper and simpler solid/liquid separation for the filtration step.

(f) The successful development of improved instrumentation has resulted in considerable savings in the costs of reagents and labour and an improvement in operating efficiency at South African uranium plants.

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DISCUSSION

F.E. MCGINLEY: With regard to the reverse leach procedure: what pH do you have to attain before you can subject the uranium tailings to gold cyanidation?

M.G. ATMORE: The final pH will be 11–12 for cyanidation, but before transferring the acid-leached pulp it is neutralized to a pH in excess of 7 and conditioned for 2–3 hours to ensure that there is no residual acidity.

K. PINKAS: I would like to know whether you noticed a poisoning of ion exchange resins; I know that silica and other impurities can cause trouble.

H.E. JAMES: In the early days, when we had the normal leach procedure, there was a lot of poisoning of resins with cobaltic cyanides resulting from the prior cyanidation step. In our work on the application of weak-base resins we certainly have experienced considerable problems with silica pick-up on these resins. This work was very adequately described by Haines et al. at the 11th International Mineral Processing Congress (Ref. [23] of the paper) and I shall not go into details here. We are fairly confident that the problems associated with silica pick-up can be overcome. I must point out that the use of weak-base resins is not currently being considered for commercial application in South Africa and a lot of work still needs to be done.

F.R. HARTLEY: First, I should like to follow up the comments just made on silica poisoning in relation to our experience in Australia some years ago with the Mary Kathleen operation. The resin at this plant was particularly prone to silica pick-up. Subsequent experimental work showed quite conclusively that the major portion of the silica was picked up whilst the barren solution was passing through the resin in the trailing column. In fact, at Mary Kathleen, the problem was exacerbated by the fact that a three-column operation was adopted. As a result of that study we believe that the application of continuous moving-bed ion exchange operation would alleviate the problem because it minimizes the volume of barren liquor in contact with the resin. I should like to ask whether you have found that silica pick-up depends very strongly on the barren solution which is passed through the resin.

H.E. JAMES: In the adsorption period a number of competitive mechanisms are in operation of which silica uptake is the slowest. The major influences on silica uptake are the pH changes accompanying the passage of solution through the column, and the total contact time. In a continuous ion exchange column, adsorption contact time is minimized but some silica take-up does occur.

S. AJURIA-GARZA: In your pressure leaching, at what temperature and pressure do you operate the autoclaves?

H.E. JAMES: This particular development is being undertaken by the Anglo-American Corporation and I should like Mr. Atmore to answer the question.

M.G. ATMORE: I think that this depends to a large extent on the type of refractory minerals you are trying to attack. Our aims in pressure leaching have been two-fold: first, to get the best possible uranium recovery when treating the usual South African ores associated with the Witwatersrand gold deposits, and second, to improve the overall recoveries of uranium in those cases where they are very much lower, as with refractory uranium ores which often contain titanium, such as brannerites. The maximum temperatures used are 180–200°C, but some good results have been obtained at lower temperatures, down to 140–150°C, which is very interesting.

F.C. LENDRUM: Was there any indication, in your experience, of increased losses of uranium in tailings when grinding extremely finely? Have you seen indications of excessive or increased losses in the very fine slimes, as if the clay minerals acted as ion exchangers?

H.E. JAMES: Work carried out at the National Institute for Metallurgy about five years ago showed that adsorption of both uranyl and ferric complex species onto quartz and pyrophyllite does occur but is very low, even with very finely ground material. The results indicated that losses of uranium through adsorption in the repulping stage at a typical South African uranium plant amounted to less than 0.5% of the uranium present in the ore.

F.C. LENDRUM: I should like to thank Mr. James for his very enlightened paper. In Canada we feel that if there is to be an intensive investigation in the field of uranium ore processing, it must be one aimed at elimination of clarification and a simplification of the liquid/solid separation steps by the use of upflow ion exchange.

One further question: You mentioned the production of ferric iron by bacteria. I gathered that this is done outside the leaching circuit, i.e. you subject a ferrous solution to bacterial oxidation and then put it back into the leach circuit. Is this correct?

H.E. JAMES: Yes. The idea is to carry out the operation in separate tanks where ferrous solution is passed over plastic packing material covered with a thin film of bacteria. This procedure is the basis for the patent by E.L. Goldblatt of General Mining and Finance Corporation (Ref.[20] of the paper).

F.R. HARTLEY: Not knowing too much about biological systems, I should like to ask whether you have to add an organic nutrient of any sort to the bacterial system for producing ferric sulphate?

H.E. JAMES: No. The ions which already exist in the solution act as nutrients for the bacteria and they stand up to quite severe conditions.

F.R. HARTLEY: Your description of the gravity concentration split brings to mind the commercial application of heavy-media separation at one plant in South Africa. I wonder if you could report about the long-term results of that particular operation.

H.E. JAMES: Again, this is an Anglo-American plant and Mr. Atmore is better qualified to deal with that question.

M.G. ATMORE: At different times we actually had two plants using heavy-media separation. The first was at Free State Saaipiaas where the use of heavy media was primarily aimed at increasing the throughput of a plant which was heavily loaded and operating at designed capacity, without seriously reducing gold recovery. It was an attempt to squeeze the plant. Naturally the effect of heavy media on uranium recovery was investigated and interesting results were obtained. However, great troubles, particularly from abrasion and medium loss, were encountered. In spite of these it was decided to incorporate a heavy-medium stage in an extension being designed at the time for

one of our Vaal Reefs plants. This ran for about two years. The indications were that it was marginally more successful technically but substantially less successful economically, so it was discontinued and we are at present not using this technique on uranium, although it is widely used in the Group elsewhere.

J. CAMERON: You mentioned both flotation and wet high-intensity magnetic separation as potential methods for recovery of uranium from old tailings dumps. Is there a judgement on which of these is likely to be the more productive and what are the different approaches to this?

H.E. JAMES: In my paper I mentioned the work being done by the Anglo-American Corporation at one of the old mines in the eastern Witwatersrand where they are installing a flotation plant with a capacity of one million tonnes of slime per month. This plant is primarily for the production of a pyrite concentrate for sulphuric acid manufacture but gold and uranium will be recovered as by-products. A number of old dumps will be processed through the plant and the residues re-deposited in another area as one large dump. It is only when you take into account pyrite, gold, uranium and property values that this type of operation begins to look economic for the very low-grade dumps. With regard to both flotation and wet high-intensity magnetic separation we have the same experience as in Canada, namely that these techniques do not produce a low enough grade of uranium in the tailings to allow one to discard it. These techniques cannot therefore be used for preconcentration of run-of-mine ores in the classical sense. We are, however, considering their use for the recovery of some of the uranium and gold left in tailings, but applying them at the end of the circuit. With a view to the very long-term future we are hoping that the efficiency of the wet high-intensity magnetic separator and particularly the engineering of large-scale equipment will be improved, allowing us to treat ores with lower uranium grades than are currently being considered economically treatable.

M.G. ATMORE: In relation to the comparison made by Mr. James, I should like to make one point. The wet high-intensity magnetic separation is new as far as we are concerned. On the other hand, the flotation of residues has been practised by us since about 1952. This has been a standard procedure and in our operations we have not been unduly concerned at the low recoveries of uranium, which were 30–35%, since we have been able to obtain high sulphide recoveries and appreciable additional gold. At various times we have used modified procedures but we have always reverted to the straightforward circuit and reagents. It is these that will be used in the Rioden project. This project is located east of Johannesburg in an area where gold mining commenced at an early stage. In many of the old dumps to be reclaimed and treated, the residual gold deposited was higher than it would be now.

F.C. LENDRUM: What are the economics of pyrite in South Africa?

M.G. ATMORE: The economics of recovering sulphur-bearing materials are about the same as they are elsewhere. However, for a number of years the cost of pyrite recovery was very low because other costs were borne predominantly by the gold recovery process or by uranium production itself. Until recently, this very cheap pyrite resulted in an extremely low sulphuric acid production cost, and until two or three years ago this would have been 12 rand or perhaps 13 rand per tonne of sulphuric acid. More recently, this has increased substantially, as have all other operating costs, and in our opinion the price of sulphuric acid now available in South Africa is very similar to that charged almost anywhere else.

LA MINERALURGIA DEL URANIO EN ESPAÑA

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Abstract—Resumen

URANIUM ORE PROCESSING IN SPAIN.

The paper presents a review of the Spanish needs of uranium concentrates and uranium ore processing technology and trends in Spain. Spain produces approximately 200 t U_3O_8/a at two facilities. One plant in the south (Andújar, Jaén) can obtain 70 t U_3O_8/a and uses a conventional acid leaching process with countercurrent solvent extraction. A second plant, situated in the west (Ciudad Rodrigo, Salamanca) has started in 1975 and has a capacity of 120-130 t U_3O_8/a , using acid heap leaching and solvent extraction. There is another experimental facility (Don Benito, Badajoz) scheduled to start in 1976 and expected to produce about 25-35 t U_3O_8/a as a by-product of the research work. For the near future (1978) it is hoped to increase the production with: (a) A new conventional acid leaching/solvent extraction plant in Ciudad Rodrigo; its tentative capacity is fixed at 550 t U_3O_8/a . (b) A facility in the south, to recover about 130 t U_3O_8/a from phosphoric acid. (c) Several small mobile plants (30 t U_3O_8/a per plant); these will be placed near small and isolated mines. The next production increase (1979-1980) will come with the treatment of sandstones (Guadalajara and Cataluña) and lignites (Cataluña); this is being studied. There are also research programmes to study the recovery of uranium from low-grade ores (heap, in-situ and bacterial leaching) and from other industries.

LA MINERALURGIA DEL URANIO EN ESPAÑA.

La memoria presenta una revisión de las necesidades españolas de concentrados de uranio, así como de la tecnología y tendencias existentes en España para su producción. España puede producir actualmente unas 200 t U_3O_8/a ($t=Tm$) en dos instalaciones. Hay una fábrica en el sur (Andújar, Jaén), que puede obtener 70 t U_3O_8/a y que tiene un proceso convencional de lixiviación ácida-decantación en contracorriente-extracción con disolventes. La segunda ha empezado a funcionar en 1975 y tiene una capacidad de 120-130 t U_3O_8/a , está situada en el oeste (Ciudad Rodrigo, Salamanca) y sigue un proceso de lixiviación estática ácida y extracción con disolventes. Existe además otra instalación, experimental (Don Benito, Badajoz), que se pondrá en funcionamiento en 1976 y en la que esperamos obtener de 25 a 35 t U_3O_8/a como subproducto de la investigación. En un futuro próximo (1978) se espera aumentar la producción con: a) Una nueva planta de lixiviación ácida convencional y extracción con disolventes en Ciudad Rodrigo y cuya capacidad será del orden de 550 t U_3O_8/a . b) Una instalación, en el sur, para recuperar unas 130 t U_3O_8/a , a partir del ácido fosfórico. c) Algunas plantas móviles (30 t U_3O_8/a por planta) que se montarán en las minas pequeñas y aisladas. La siguiente expansión de la producción (1979-1980) se logrará con el tratamiento de las areniscas (Guadalajara y Cataluña) y de los lignitos (Cataluña), cuyo estudio está en curso. Hay también otros programas de investigación para estudiar la recuperación del uranio de menas de ley baja (lixiviación estática, in situ y bacteriana) y también para recuperar el uranio de otras industrias.

1. INTRODUCCION

España se ha incorporado a los países que consideran la energía nuclear como una de sus fuentes de aprovisionamiento energético. En la actualidad existen tres centrales nucleares en funcionamiento, con un total de 1120 MW(e). Están en construcción otras cinco centrales con una potencia total de 4695 MW(e), y hay, en fase de contratación, otras cinco (5000 MW(e)) que deberían entrar en funcionamiento antes de 1983. Las previsiones de potencia nuclear instalada para el año 1985 consideran una cifra acumulada de unos 23660 MW(e).

Las exigencias de concentrados de uranio para este programa, referidas a los años 1975, 1980 y 1985, se cifran en 1053 t U_3O_8/a ; 3494 t U_3O_8/a ; y 5900 t U_3O_8/a ; mientras que las cantidades acumuladas de concentrados serían de 1053 t U_3O_8 ; 12 084 t U_3O_8 y 35 824 t U_3O_8 , respectivamente [1].

Las disponibilidades españolas de concentrados de uranio son limitadas. Para hacer frente a esta situación se está tratando de:

- Comprar concentrados de uranio en el exterior.
- Aumentar la exploración y el tratamiento de minerales de uranio en el país.
- Colaborar en programas multinacionales en el exterior, para complementar la producción interior de concentrados de uranio.

En esta memoria el problema se analizará únicamente en su aspecto interno, es decir en el estado actual y perspectivas de la mineralurgia del uranio en España.

2. RESERVAS ESPAÑOLAS DE MINERALES DE URANIO

Las reservas de uranio en España han estado estabilizadas en aproximadamente 8500 tU, para el intervalo de precios de <10 \$/lb U_3O_8 [2]. Sin embargo, considerando los resultados de los trabajos realizados, y a la vista de los nuevos criterios de valoración recomendados por el grupo conjunto de trabajo ENEA/OIEA, las perspectivas son más halagüeñas. España ha aportado [3] a la última reunión del grupo los datos siguientes:

2.1. Recursos razonablemente asegurados en la zona de precios bajos (<15 \$/lb U_3O_8).

10 000 tU

Compuestos por:

Pizarras (Salamanca y Badajoz)	6 500 t
Granitos (Salamanca y Cáceres)	400 t
Areniscas próximas a Mazarete (Guadalajara)	1 700 t
Areniscas terciarias de Vich y Olot (Gerona)	850 t
Conglomerados de Valdemeca (Cuenca)	170 t

2.2. Recursos adicionales estimados en la zona de precios bajos (<15 \$/lb U_3O_8).

8 800 tU

Compuestos por:

Areniscas del área de Mazarete	5 100 t
Areniscas del área de Vich y Olot	3 400 t
Conglomerados de Valdemeca	340 t

2.3. Recursos razonablemente asegurados en el tramo de precios altos (15-30 \$/lb U_3O_8).

93 500 tU

Compuestos por:

Areniscas terciarias (Guadalajara)	4 300 t
Areniscas triásicas zona de Mazarete, Cuenca y Camprodón (Gerona).	8 500 t
Lignitos de la cuenca de Calaf (Barcelona)	15 300 t
Lignitos de Santa Coloma de Queralt (Barcelona)	13 600 t
Lignitos de las cuencas de Fraga (Huesca), Mequinenza (Zaragoza) y Almatret (Lérida)	51 900 t

2.4. Recursos estimados adicionales en el tramo de precios altos (15-30 \$/lb U_3O_8).

97 800 tU

Compuestos por:

Areniscas terciarias (Guadalajara)	17 000 t
Areniscas del área de Molina de Aragón	17 000 t
Lignitos de la cuenca de Calaf	8 500 t
Lignitos de Santa Coloma de Queralt	12 800 t
Lignitos de las cuencas de Fraga, Mequinenza y Almatret	42 500 t

Además se ha puesto en marcha un programa de exploración de 10 años de duración (el Plan Nacional del Uranio) con recursos mucho más amplios que los disponibles hasta el presente (12 500 millones de pesetas). El esfuerzo investigador se va a concentrar sobre 110 000 km² de áreas favorables. La información disponible sobre estas áreas, así como los medios puestos en juego, permiten fundadas esperanzas de éxito.

Desde el punto de vista mineralúrgico, la información más completa se tiene sobre los minerales de pizarras, de los granitos y de las areniscas próximas a Mazarete, que responden bien a diagramas convencionales por vía ácida. Hay también bastantes resultados sobre el comportamiento de los lignitos radiactivos, considerados como mena simple de uranio. Pero las directrices actuales en España son más ambiciosas y quieren abordar también el aprovechamiento de los lignitos como combustibles; en cualquier caso, las perspectivas de trabajo son grandes y el esfuerzo investigador, cara al futuro, muy considerable.

3. TECNOLOGIA ACTUAL

España ha estado produciendo de 55 a 80 t U_3O_8 /a desde 1960. Esta producción se obtenía en una planta comercial (Andújar) [4] y en dos plantas piloto (Don Benito y Ciudad Rodrigo) [5]. A mediados de este año entró en funcionamiento otra planta comercial (Ciudad Rodrigo) y está en avanzado estado de construcción una instalación experimental relativamente grande y que también podrá aportar de 25 a 35 t U_3O_8 /a.

Las técnicas seguidas en todas ellas son procesos de lixiviación ácida, bien en su versión convencional sobre mineral molido, o bien por lixiviación estática sobre mineral grueso. En uno de los casos hay una combinación de ambas modalidades. A continuación se dan las características más sobresalientes de cada una de las instalaciones.

3.1. Fábrica de uranio de Andújar

Fue la primera, y durante varios años la única, instalación comercial para el tratamiento de minerales de uranio en España. Se puso en marcha a finales de 1959, a fin de tratar las menas simples de uranio de la zona de Córdoba — Jaén. Su capacidad es de 200-230 t/d de mineral.

El diagrama original constaba de un circuito convencional de trituración seca en dos etapas, molienda en un circuito doble de barras y bolas, lixiviación ácida, lavado por contracorriente con un sistema mixto de ciclones (para las arenas) y decantadores (para las lamas); luego seguían ajuste de pH, clarificación y cambio de ion en lecho fijo con resina de base de amonio cuaternario; se hacía una doble precipitación con cal (primera fase) y con sosa o amoníaco (segunda fase); los efluentes se neutralizan con cal.

Este diagrama inicial se adecuó (1962) al tratamiento de menas dobles de uranio-cobre. Se introdujeron dos nuevas secciones para la recuperación del cobre, una de flotación para los residuos sólidos y otra de cementación para los efluentes líquidos.

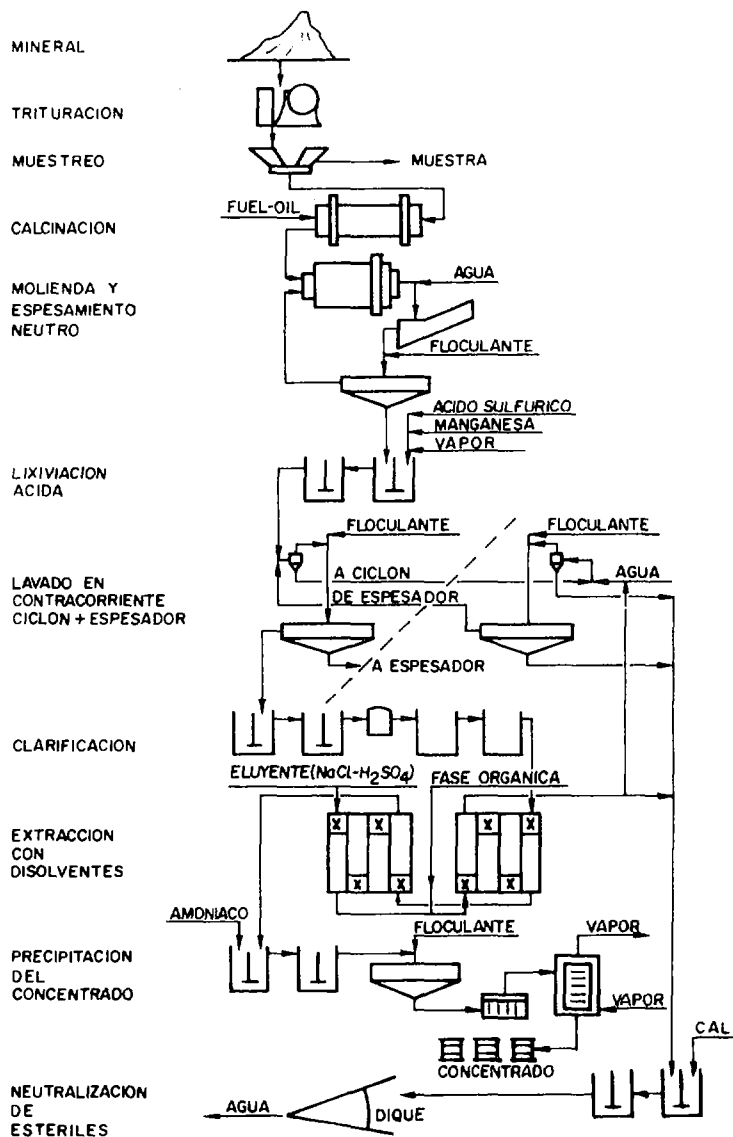


FIG.1. Diagrama de flujo de la fábrica de Andújar.

Posteriormente, se hicieron otras dos innovaciones. En 1970, se introdujo una sección para la tostación a baja temperatura de minerales limoníticos. En 1972, se abandonó el cambio de ion por resinas y se adoptó la extracción con disolventes. El diagrama esquemático actual se indica en la figura 1.

La planta se ha alimentado con minerales muy diversos, y de leyes relativamente bajas (0.9-1,5 kg U_3O_8 /t). El rendimiento medio del proceso ha estado comprendido en el intervalo del 85 al 93%. El concentrado ha tenido leyes comprendidas entre el 80 y el 87% U_3O_8 .

Una distribución porcentual típica de los costos de transformación es: 1973, mano de obra, 47,8%; reactivos y materias auxiliares, 32,0%; materiales de mantenimiento, 8,2%; energía eléctrica, 6,9%; amortización, 1,5% y gastos varios 3,6%.

La contribución porcentual de las diferentes secciones de la fábrica se puede resumir como sigue: recepción, trituración y muestreo, 10,3%; calcinación, 5,5%; molienda, 6,8%; lixiviación, 19,5%; separación sólido-líquido, 10,5%; extracción con disolventes, 7,7%; precipitación y manejo del concentrado, 3,8%; neutralización y eliminación de residuos, 7,3%; servicios auxiliares, 14,0% y gastos generales, 14,6%.

3.2. Instalaciones de lixiviación estática de Ciudad Rodrigo

En la zona próxima a Ciudad Rodrigo están localizadas la mayoría de las reservas de uranio en pizarras. Para esta región existen planes antiguos desde 1963, en que se consideró la posible construcción de una fábrica convencional de lixiviación ácida-cambio de ion. Luego, en 1969, se volvió a considerar el proyecto modificándolo, substituyendo el cambio de ion por la extracción con disolventes, y considerando también una etapa de calcinación a baja temperatura ($\sim 350^\circ C$) para liberar el uranio retenido por las limonitas (10-15% del total); también se consideró el aprovechamiento del uranio de las fracciones marginales por lixiviación estática. No obstante, la planta no se construyó y únicamente se montó una instalación piloto (ELE) para el estudio del mineral marginal. En 1973, y dentro del bache de precios bajos del uranio, se pensó en construir una pequeña instalación para recuperar el uranio de una parte del yacimiento que iba a quedar bajo las aguas de un pantano. Por esas fechas, el Estado español creó una sociedad, la Empresa Nacional del Uranio S.A. (ENUSA), para el suministro de combustibles a los reactores nucleares. La creación de ENUSA y la crisis del petróleo fueron los factores que hicieron que la explotación de los yacimientos de Salamanca entrasen en vía de realización. En la actualidad existe ya una actividad productora de uranio en esa zona, que proviene de las dos plantas que se indican a continuación.

3.2.1. Planta piloto ELE

Es la antigua planta piloto que la JEN había montado para el estudio de la lixiviación estática de minerales marginales [5]. En los últimos tiempos que la tuvo la JEN, trabajó con alimentaciones más ricas, 1-1,5 kg U_3O_8 /t, y se vio que el proceso daba rendimientos aceptables (80-83%), parecidos a los de la lixiviación convencional sobre mineral sin tostar. Ante la posibilidad de transferir la instalación a ENUSA, se la dotó de una segunda unidad de extracción, con lo que su capacidad de tratamiento aumentó a unos 100 m³/d, lo que equivalía a unas 20 t U_3O_8 /a, es decir casi un tercio de la capacidad de la única planta de producción existente (Andújar).

3.2.2. Planta de lixiviación estática Elefante

Está basada en la planta piloto anterior y ha sido fruto de la colaboración JEN-ENUSA, que realizaron todos los trabajos de ingeniería, construcción y puesta en marcha. La planta empezó a producir en el mes de mayo pasado. La capacidad inicial de diseño era de 75 t U_3O_8 /a. Se trató de aumentar esta producción desde el principio, primero a 112 y por último a unas

120 t U_3O_8 /a. Todavía se espera aumentarla algo más cambiando algunos equipos tales como las bombas de la zona de tratamiento de líquidos. La inversión realizada, incluyendo la compra de terrenos y la infraestructura, fue algo inferior a los 100 millones de pesetas. El personal total de la explotación (cantera y planta) es de 64 personas.

El diagrama de flujo de la planta se indica en la figura 2 y consta de las fases de trituración, lixiviación estática ácida, extracción con disolventes, precipitación con amoníaco y neutralización de efluentes con cal. Los aspectos fundamentales de cada fase son:

3.2.2.1. Trituración

Se realiza en una sola etapa. El tamaño nominal de la alimentación es de 300 × 500 mm, pero la mayoría supera a éste, llegando incluso a 0,8 m de dimensión máxima. Hay una pequeña era de recepción. Los camiones descargan en la era o sobre la tolva de alimentación. Un alimentador tipo Apron lleva el mineral a una trituradora de impactos, con rotor de 1,5 × 1,0 m y motor de 150 CV; el producto sale triturado a unos 50 mm. La capacidad tope de la trituradora es de unas 200 t/h. El producto cae sobre una cinta con báscula continua y luego pasa a una tolva. Un desmuestreador lineal toma las muestras, que pasan a un circuito de preparación compuesto de un molino de martillos y de otro desmuestreador lineal. El mineral triturado se lleva a las eras de lixiviación en camiones ligeros (10-12 t).

3.2.2.2. Lixiviación

La zona de lixiviación está en una pequeña vaguada con pendiente suave (8-10%). El suelo se limpió e igualó para evitar salientes; luego se puso una capa de 10 cm de arena y encima una lámina de polietileno de 7,5 m de ancho y 0,3 mm de grueso, que se protegió con otra capa de arena. La zona de lixiviación se divide en cuarteles de 90 × 7 m, mediante caballones. Junto a los caballones y hasta la mitad hay unos tubos de drenaje de PVC, de 100 mm de diámetro.

La primera capa de mineral (~2 m) se coloca con cuidado avanzando con una pala cargadora. Los camiones entran en la era hasta el talud, pero éste se forma con la pala para evitar la segregación de finos y gruesos. Cuando se ha colocado la primera capa, se ara el mineral hasta una profundidad de 0,5 m, y se continúa formando otras capas similares hasta conseguir la altura total, de 6 a 8 m.

El riego se hace con aspersores de plástico según una malla de 4,0 × 5,4 m y una velocidad de 30 l/h/m². El circuito supone una contracorriente con dos etapas. Inicialmente se había previsto sacar gradualmente el uranio usando el ácido con defecto, pero ENUSA ha elaborado una técnica en la que se da un primer riego de acondicionamiento, con aproximadamente 10 kg H_2SO_4 /t, se deja en maduración una semana y luego se inicia la fase de extracción del uranio, con aportación creciente de ácido, procedente de la etapa siguiente. El consumo de ácido es del orden de 40 kg/t y la solubilización alcanza el 80%. Cuando los líquidos decaen en contenido de uranio (150 mg U_3O_8 /l) se suspenden los riegos de la zona correspondiente, que se deja madurar de 2 a 3 semanas, y se vuelve a regar hasta que decae de nuevo la concentración de uranio. Existe además otro conjunto de lixiviación para el mineral marginal de ~400 g U_3O_8 /t de ley.

3.2.2.3. Extracción con disolventes

La solución fértil sale clara y fluye a una balsa de recogida desde la que se envía a otra de almacenamiento. Si la acidez libre es baja (<4 g H_2SO_4 /l) se inyecta ácido en la corriente antes del almacenamiento. Desde la balsa de almacenamiento fluye por gravedad al sistema de extracción con disolventes. Se emplea amina terciaria (2,5%), isodecanol (2%) y queroseno. Se trabaja con el 90% de saturación. La reextracción se hace con cloruro sódico 1,2M y sulfúrico 0,05M, la relación de flujos se ajusta para tener 30 g U_3O_8 /l en el extracto.

La operación se realiza en mezcladores sedimentadores. Se trabaja con orgánica continua, $(O/A)_{ME} = 1,5$ y $(O/\dot{A})_{MRE} = 3$ y se realizan reciclados internos para lograr las relaciones de mezcla establecidas. A fin de reducir las pérdidas de orgánica se ha cuidado el diseño del equipo en el sentido de:

- a) Emplear mezcladores de sección circular.
- b) Usar turbinas de aspiración carradas.
- c) Usar baja velocidad de giro en los agitadores a fin de seguir al criterio de Bellingham [6], es decir, $n^3 D^2 < 20$ ($n = \text{rev/s}$ y $D = \text{diámetro en pies}$). Además los agitadores están provistos de variador de velocidad para reducir ésta al mínimo compatible con las necesidades de bombeo. Un tubo de nivel conectado al doble fondo permite establecer la velocidad de giro adecuada.
- d) En el paso del mezclador al sedimentador se han colocado deflectores que eliminan la energía cinética del líquido y distribuyen la mezcla a lo ancho del sedimentador.
- e) También se ha dotado a los sedimentadores de las barreras estabilizadoras preconizadas por Agers y Dement [7].
- f) La retirada de los líquidos de los sedimentadores se hace por tubos con tomas en canales que se extienden a todo lo ancho de los sedimentadores. El control de interfases, así como la regulación de los reciclados, se hace con tubos telescópicos.

El equipo funciona bien, da refinados bajos ($< 3 \text{ mg U}_3\text{O}_8/\text{l}$), extractos altos ($\sim 30 \text{ g U}_3\text{O}_8/\text{l}$), y bajas pérdidas de orgánica ($\sim 0,1 \text{ l/m}^3$).

3.2.2.4. Precipitación y manejo del concentrado

El extracto acuoso se precipita en continuo (3 tanques) con una mezcla de amoníaco/aire (1/3) hasta un pH final de 7. La operación se realiza a $30-35^\circ\text{C}$. El concentrado se separa de las aguas madres por decantación y se lava en un decantador y un filtro. Luego se seca en una estufa a 250°C y se envasa en bidones de 200 l. El producto es relativamente homogéneo con leyes comprendidas entre el 83 y el 86% de U_3O_8 .

3.2.2.5. Neutralización y eliminación de residuos

Los residuos de mineral estéril quedan sobre el terreno impermeabilizado. En el futuro se espera nivelar el terreno y hacer nuevas eras de mineral sobre los residuos antiguos.

Los efluentes de extracción con disolventes se fraccionan; la mitad se recirculan a lixiviación y la otra mitad se neutralizan con lechada de cal. La sección de neutralización consta fundamentalmente de un silo de almacenamiento de cal triturada, un apagador de cal, un tanque de almacenamiento de lechada y tres reactores de neutralización. Al segundo tanque de neutralización se le inyecta aire. La pulpa de efluentes neutralizados vierte en una vaguada en la que hay una serie de diques para retener los sólidos.

3.3. Planta experimental de Don Benito (proyecto G)

En la zona de Don Benito, en el contacto de granitos y pizarras, existe un mineral fuertemente alterado cuyo tratamiento por lixiviación estática es difícil [5]. Los yacimientos se explotan para enviar a Andújar la fracción más rica. Hay interés en recuperar el uranio de la fracción que queda a pie de cantera; también se piensa en la posibilidad de construir una instalación de mayor capacidad para el tratamiento global del mineral de la zona. El mineral es relativamente difícil, una vez atacado sedimenta mal y la permeabilidad de sus lechos es muy baja.

La JEN decidió la construcción de una instalación experimental (100 t/d) que combinase la lixiviación estática y la convencional. El diagrama comprende las fases de:

- a) Trituración y tamizado.
- b) Separación de gruesos y finos.
- c) Lixiviación estática de los gruesos.
- d) Espesamiento y lixiviación de finos.
- e) Separación arenas/lamas.
- f) Cambio de ion sobre lamas (RIP).
- g) Elución con ácido sulfúrico en columna continua.
- h) Extracción con aminas de la mezcla de las soluciones fértiles de lixiviación estática y de los eluidos de cambio de ion.
- i) Precipitación del concentrado y calcinación continua.
- j) Neutralización y eliminación de efluentes.
- k) Lixiviación estática del todo-uno marginal ($\sim 400 \text{ g U}_3\text{O}_8/\text{t}$, a razón de otras 100 t/d).

La instalación está en avanzado estado de construcción y se espera ponerla en marcha a mediados de 1976. Aparte de obtener información tecnológica para un proyecto más amplio se espera recuperar como subproducto unas 30 t $\text{U}_3\text{O}_8/\text{a}$.

3.4. Posibilidades actuales de producción

Resumiendo lo anteriormente dicho y extendiendo la perspectiva hasta mediados del año próximo, las posibilidades españolas de producción de concentrados de uranio (t $\text{U}_3\text{O}_8/\text{a}$) son:

a) Fábrica de Andújar	60 — 70
b) Planta piloto ELE	20 — 25
c) Planta de lixiviación estática <i>Elefante</i>	120 — 140
d) Instalación experimental de Don Benito	25 — 35
	<hr/>
TOTAL	225 — 270

4. PROYECTOS INMEDIATOS

Ante la situación y perspectivas del mercado de concentrados de uranio, España va a tratar de aumentar su capacidad de producción. En este último trimestre se han iniciado tres proyectos cuyas características esenciales son las siguientes.

4.1. Fábrica de Saelices el Chico

Es la nueva versión del proyecto que la JEN tenía para Ciudad Rodrigo (véase 3.2.). El nuevo proyecto contempla una producción nominal de 550 t $\text{U}_3\text{O}_8/\text{a}$, con una realización en dos líneas y puesta en marcha a mediados de 1978.

El diagrama se está revisando en la actualidad, pero posiblemente se mantenga en sus puntos fundamentales de:

- a) Molienda autógena en seco;
- b) Calcinación;
- c) Lixiviación en una etapa;
- d) Lavado en contracorriente;
- e) Extracción con disolventes y reextracción con sulfato amónico;
- f) Precipitación con amoníaco y calcinación del concentrado y
- g) Neutralización y eliminación de efluentes.

4.2. Recuperación del uranio del ácido fosfórico

En España se producen actualmente unas 400 000 t $\text{P}_2\text{O}_5/\text{a}$ en forma de ácido fosfórico. Este ácido (28-30% P_2O_5) tiene una concentración de uranio de unos 130 g $\text{U}_3\text{O}_8/\text{m}^3$, aunque

depende del origen de la roca fosfática de la que se ha obtenido. En consecuencia, esa producción de ácido lleva consigo la solubilización de unas 150 t U_3O_8/a .

La recuperación del uranio del ácido fosfórico se estudió en España a partir de 1968, primero a escala de laboratorio [8] y luego a escala de planta piloto. El diagrama de tratamiento [9] hace uso de la extracción con disolventes mediante la combinación sinérgica del ácido dietilhexilfosfórico (D2EHPA) y el óxido de trioctilfosfina (TOPO) diluidos en queroseno. El proceso consta de las fases siguientes:

- a) Enfriamiento del ácido fosfórico a 46°C.
- b) Clarificación de este mismo ácido hasta menos de 200 ppm de sólidos.
- c) Recuperación del uranio del ácido por extracción con disolventes.
- d) Reextracción reductora del uranio a temperatura alta y con ácido fuerte para tener extractos de 9-12 g U_3O_8/l .
- e) Oxidación y dilución del extracto de d).
- f) Extracción del uranio con disolventes.
- g) Lavado de la fase orgánica.
- h) Reextracción-precipitación con carbonato amónico-amoniaco para obtener uranil-tricarbonato amónico (UTCA).
- i) Lavado, filtración y calcinación del UTCA.

La planta piloto se montó en la misma fábrica de producción de ácido fosfórico a fin de poder utilizar ácido reciente. Todo el equipo de extracción es de mezcladores sedimentadores. El comportamiento de la instalación fue bueno. La recuperación de uranio alcanzó el 90% y el producto tuvo leyes comprendidas entre el 95 y el 98% de U_3O_8 . El proceso químico no presentó dificultades, aunque la clarificación del ácido fue el paso más delicado; también hubo que introducir medidas correctivas a la formación de incrustaciones de sólidos (fluosilicato) en las turbinas de bombeo.

De acuerdo con los resultados de la planta piloto la aportación de uranio en una instalación comercial podría superar las 100 t U_3O_8/a . Una primera estimación de la inversión para esta planta asciende a unos 500 millones de pesetas. El coste de transformación estimado es del orden de los 10 \$/lb U_3O_8 , pero estas cifras son provisionales. En la actualidad está muy avanzado el manual de diseño de la futura planta.

4.3. Instalaciones móviles

Los resultados obtenidos con los circuitos de lixiviación estática han sido esperanzadores. Por ello se ha pensado en recuperar el uranio de los yacimientos pequeños con plantas de lixiviación estática ácida-extracción con disolventes. En este sentido se han diseñado unas unidades modulares para 150 m³/d de solución fértil.

En la actualidad se está en la fase de compras para dos de estas unidades. Su puesta en marcha podría efectuarse hacia finales del año próximo o a comienzos de 1977. La capacidad de producción de cada una de estas unidades se prevé en 30 t U_3O_8/a .

5. INVESTIGACIONES EN CURSO

Durante los últimos años, los esfuerzos del grupo de mineralurgia de la JEN se han distribuido entre la ingeniería de procesos y la experimentación o investigación. La actividad sobre la primera vertiente se deduce de lo expuesto más arriba. Los trabajos de investigación y desarrollo se centraron en el tratamiento del ácido fosfórico, la lixiviación estática, en mina y bacteriana, el tratamiento de lignitos, el tratamiento de algunas aguas cupríferas y el estudio de los nuevos

minerales. Ante las perspectivas puestas de manifiesto por las exploraciones de uranio, se está reforzando el grupo y se espera hacer un gran esfuerzo investigador en los próximos años. A continuación se indican las investigaciones que están actualmente en curso.

5.1. Tratamiento de las areniscas de Mazarete

Las areniscas de Mazarete tienen una ley del orden de 1 kg U_3O_8 /t. Su estudio está bastante avanzado. Los resultados muestran que se trata de un mineral relativamente normal y adaptable posiblemente a circuitos de lixiviación ácida convencional y extracción con disolventes. La experimentación se continúa en el sentido de ver si existe alguna heterogeneidad del yacimiento respecto al tratamiento.

5.2. Estudio de los lignitos radiactivos

La recuperación del uranio de los residuos de la combustión de lignitos a baja temperatura (400-500°C), se ha estudiado suficientemente. El tratamiento de las cenizas es viable por un esquema de lixiviación ácida-extracción. Las dificultades encontradas son: la tecnología de la combustión a baja temperatura, los elevados consumos de ácido y las exigencias de eliminación de la materia orgánica de los líquidos antes de la extracción con disolventes. En la actualidad se está replanteando el programa de estudio con el ánimo de aprovechar también el potencial calorífico de los lignitos.

5.3. Lixiviación bacteriana

La JEN ha dedicado bastante atención al estudio de la lixiviación bacteriana. Las investigaciones se han centrado en el aislamiento y cultivo de cepas locales. De las aguas de minas graníticas se aislaron tipos muy activos [10], especialmente en cultivos hechos en medios ferrosos. Los trabajos continúan a fin de obtener cultivos importantes para aplicarlos en la lixiviación en mina y en la lixiviación estática de minerales pobres.

5.4. Lixiviación en mina

En España se ha hecho una experiencia de lixiviación ácida del uranio en mina. Los resultados no han sido muy prometedores, posiblemente porque en la matriz de la roca había fosfatos. Estos ensayos se han abandonado. Existe otra mina ya explotada por métodos clásicos, que se podría prestar bien a la lixiviación *in situ*. Se trata de una mineralización de pecblenda y pirita en granito. Algunas pruebas de lixiviación natural, realizadas en el laboratorio, dieron solubilizaciones del orden del 80%.

6. PERSPECTIVAS

En el supuesto de que España pueda llevar a cabo el esfuerzo investigador, tecnológico y económico deducido de su programa nuclear, unas perspectivas razonables, basadas en los conocimientos actuales, para la producción de concentrados de uranio (t U_3O_8 /a) en el país, serían:

Instalación	1980	1985
Proyectos del Oeste (Salamanca)	650	750
Uranio de ácido fosfórico (Huelva)	130	150
Instalaciones móviles (varias)	120	180
Fábrica del Suroeste (Extremadura)	100	100
Mazarete 1 (Guadalajara)	600	600
Mazarete 2 (Guadalajara)	—	600
Lignitos 1 (Barcelona)	300	300
Lignitos 2 (Aragón)	—	300
	— —	— —
TOTAL	1900	2980

De acuerdo con esto, la producción en el país de concentrados de uranio podría cubrir aproximadamente el 50% de las necesidades de uranio. Es evidente que estas perspectivas son relativamente modestas y que se debe hacer un mayor esfuerzo mineralúrgico que considere:

- las necesidades del país en concentrados;
- las reservas de minerales en la zona de precios altos, citados antes (véase 2.);
- los resultados que pueden esperarse del programa de exploración del Plan Nacional del Uranio.

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DISCUSSION

J. CAMERON: At the recent NEA/IAEA meetings on uranium resources, Spain reported new and quite high figures for uranium in lignites, and questions were raised at that meeting about the recovery process. I note here that under Section 5.2 of your paper you mention the recovery of uranium from the residues from the combustion of lignites "a baja temperatura".

The participants of that previous meeting questioned whether a temperature of below 500°C could be maintained in this combustion system. Would you care to comment on that and on the general process of uranium recovery from lignites?

J.M. JOSA: This is a difficult point. When lignites are used in a thermo-plant, the uranium becomes incorporated in the structure of the ash from the lignites and it becomes practically impossible to recover it from this ash. On the other hand, when we tried to recover the uranium from the lignites themselves before combustion, we had very erratic results: one sample gave 90% recovery and another only 50%. Thus we charged liquids with organic material, which we then passed over the resins and through solvents, but we had difficulty in extraction from the solvents. On the other hand, the ash recovered from the lignites calcined at below 500°C gave recoveries of more than 90%. I believe that low-temperature conditions could give us high recoveries, but the experiment was on a small scale. I believe this process was studied in the USA some time ago, using controlled temperature calcination. In Spain, the aim of this investigation is to recover the thermal potential of the lignites as well as the uranium. It is a difficult problem and I do not know whether we shall succeed.

SOME RECENT IMPROVEMENTS IN A URANIUM PROCESSING PILOT-PLANT AT THE NINGYO-TOGE MINE

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Abstract

SOME RECENT IMPROVEMENTS IN A URANIUM PROCESSING PILOT-PLANT AT THE NINGYO-TOGE MINE.

Since 1964 the uranium processing plant at the Ningyo-toge mine has been modified and extended. In 1970 an ammonium diuranate producing process was added to the plant, and a pilot plant with an ore-processing capacity of 50 t/d was brought into operation. As a result of this operation the process which produces high-purity ADU was confirmed. An electrolytic reduction plant for the pilot operation was constructed by the Asahi Chemical Industry Co. in 1972. Experiments on the reduction of uranyl chloride solution were carried out with good results. Hydrofluorination facilities were added to the plant in 1974 and experiments on the hydrofluorination of UCl_4 started. The experiments also studied the characteristics of hydrated UF_4 . Preliminary experiments on dehydration from hydrated UF_4 were carried out to determine the dehydration conditions and the properties of the UF_4 in the laboratory at the Tokai Works, PNC. The results of these dehydration experiments and UF_4 conversion tests were excellent.

1. INTRODUCTION

A uranium processing technique for producing high-purity UF_4 directly from the ore by a wet process (the PNC process) has been developed mainly by the Power Reactor and Nuclear Fuel Development Corporation (PNC). The company's plant has been operated successfully and has undergone several improvements and extensions.

During the first period a pilot plant for treating domestic uranium ores was constructed in 1964 at the Ningyo-toge Mine, and uranyl chloride was produced at the mine. This uranyl chloride solution was sent to the Tokai Pilot Refinery and converted in a wet process into uranium tetrafluoride by electrolytic reduction and hydrofluorination to produce metallic uranium. In 1970 a precipitation process was added to the plant which began to produce ammonium diuranate (ADU) from ores at the mine.

Then UF_4 producing facilities for UF_6 conversion were added to the plant, and research and development for this process has progressed intensively. The research and development of the technique up to the production of UO_2Cl_2 from ores was reported at the 1970 Symposium in São Paulo [1]; this report discusses the research and development for electrolytic reduction, hydrofluorination and UF_6 conversion tests, as shown in Fig. 1.

2. ADU PRODUCTION TEST

In 1970 an ADU precipitation process was added to the Ningyo-toge Plant and research was begun immediately. The aim of this research was to obtain high-purity ADU and stable operation of the pilot plant with an ore treatment capacity of 50 t/d.

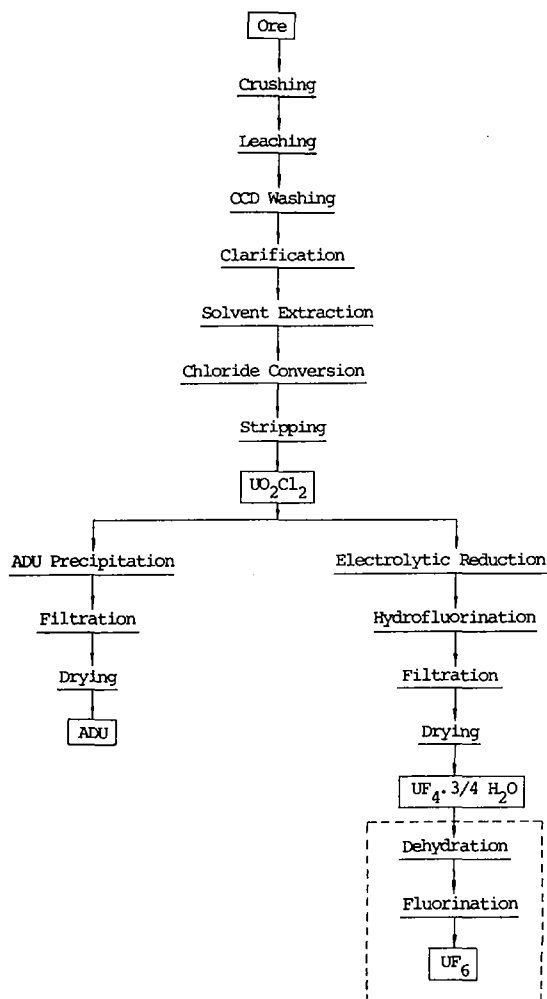


FIG.1. Simplified flow diagram for ADU and UF_6 production.

The pilot plant consists of seven processes: sulphuric acid leaching, countercurrent decantation (CCD), clarification, amine extraction, HCl conversion, stripping and ADU precipitation. Feed ores contain mainly ningyote and/or autunite as uranium minerals, and the grade changes from 0.1 to 0.6% U_3O_8 on average.

This research has resulted in data on producing high-purity ADU. An outline is shown in Table I.

The ADU produced at the Ningyo-toge Plant is converted to UO_2 and then fabricated as experimental fuel for an advanced thermal reactor (ATR) at the plutonium fuel development laboratory, Tokai Works, PNC.

TABLE I. OPERATING DATA OF THE NINGYO-TOPE PILOT PLANT (1971–1975)

Kind of ore		Nakatsugo	Yotsugi
Item			
Ore	Major uranium mineral	Ningyoite	Autunite
	Grade (%U)	0.50	0.09
	Tonnage (t)	5060	13 800
ADU product (kg U)		22 000	9 900
Overall recovery ^a (%)		87.0	79.2
Operating conditions			
Leaching	Pulp density	60% solid	60% solid
	Grinding size	– 3 mm	– 3 mm
	Sulphuric acid	60 kg/t ore	40 kg/t ore
	Temperature	50°C	50°C
	Leaching time	3 h	3 h
Liquid-solid separation	CCD thickener	4 stages	4 stages
	Classifier	2 stages	2 stages
	Pregnant solution	2.4 m ³ /t ore	2.0 m ³ /t ore
Solvent extraction	Pregnant solution	1.70 g U/l	0.37 g U/l
U content	Solvent loading	10 g U/l	10 g U/l
	Strip solution	100 g U/l	100 g U/l

^a Overall recoveries of Nakatsugo and Yotsugi could increase to 92.9 and 86.1% respectively in the case of 4-stage clarifiers and 6-stage CCD thickeners in liquid-solid separation.

3. PILOT OPERATIONS OF THE ELECTROLYTIC REDUCTION

3.1. Historical development of the uranium electrolytic reduction

In 1953 the Asahi Chemical Industry Co., Ltd. (ACI), developed a new type of high-quality ion exchange membrane. Uranium electrolytic reduction in the PNC process has been developed as one of the applications of the membrane, which is called ACIPLEX.

Since 1961 PNC and ACI have been jointly developing a new uranium electrolytic reduction technology. The first series of test runs was conducted at the Tokai Laboratory but the information obtained from the test was not sufficient to design a commercial plant.

In 1972 the second joint study started at the Ningyo-toge Mine and was conducted for two years. The unit used was a model for future commercial-scale plants, as shown in Table II, and its performance was quite satisfactory. Various tests were conducted to improve the electrolytic reduction process and to apply this technology to the various uranium sources that can be expected in the future.

TABLE II. ELECTROLYSER SPECIFICATIONS IN THE NINGYO-TOGE PLANT

Type	Two-compartment cell using bipolar electrodes
Dimension of electrode	48 dm ² /sheet
Effective area of electrode	40 dm ² /sheet
Number of cell pairs	5
Plant capacity	60 t U/a (100 t as UF ₆ /a)

3.2. Results of the pilot operation at Ningyo-toge

The principal object of the pilot operation was to find out the optimum operating conditions. As a result we have successfully operated the electrolyser for more than 1000 hours on a continuous basis. The average current efficiency was 82.3% at a reduction rate of 99.5% or more during this test run. The following items were also objects of the joint study to improve this electrolytic reduction technology:

- (a) Improvement of the structure of the electrolytic cell
- (b) Improvement of the property of ion exchange membranes
- (c) Purification of the feed solution to the electrolyser
(allowable limits of metallic ions other than uranium ion)
- (d) Activating treatment of the cathode plate
- (e) Purification of anolyte
- (f) Improvement of electrodes
- (g) Reduction test of uranyl sulphate

The influence of special metallic ions which affect the current efficiency was completely clarified and the results will be quite useful in the selection of the treatment of uranium ores at various mines in the world.

An improved procedure for plating the electrode with noble metals was developed. As a result thinner and lighter bipolar electrodes were introduced into the cells and lower manufacturing costs are expected for the electrolyser. Improvement in the structure of the electrolytic cell resulted in a decrease in the leakage of current and solutions.

The chemical and physical stability of the ion exchange membrane was improved to achieve high current efficiency at the high reduction rate and the stable performance of the electrolyser.

We believe that these improvements and developments will enable us to design commercial-scale plants utilizing the PNC process.

4. STUDIES ON THE HYDROFLUORINATION OF URANOUS SOLUTION

Since April 1974 the equipment for producing uranium tetrafluoride has been constructed, and various tests on UF₄ production by the wet process have been performed. Recently we succeeded in producing UF₄ with satisfactory characteristics for conversion to UF₆. The details are as follows.

4.1. Equipment

The 400-litre capacity hydrofluorinator is a longitudinal cylinder with cone-type bottom, made of fibre-reinforced plastic (FRP) lined with Furan resin. It has a 50 to 250 rev/min agitator and an electric heater (Fig. 2).

4.2. Feed materials

Feed materials prepared for hydrofluorination are as follows:

Uranous solution

UCl ₄ solution	U	100 g/l	Free Cl	20 g/l
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or

U(SO ₄) ₂ solution	U	100 g/l	Free SO ₄	60 g/l
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Hydrofluoric acid	50% HF solution
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4.3. Unit operation

The uranous solution, heated to above 90°C with a heat exchanger, is continuously fed into the lower part of the hydrofluorinator. In addition, the hydrofluoric acid diluted with the overflow is also fed into the hydrofluorinator in an excess of hydrofluoric acid. The reaction proceeds at the lower part of the fluorinator and UF₄ crystals grow up to 30 to 150 μm in particle size while in suspension in the reaction zone. The conditions for the formation of UF₄ are: (1) the particles must be kept suspended in the reaction zone; (2) the concentration of hydrofluoric acid must be as diluted as possible; (3) the reaction temperature must be kept above 90°C; and (4) the constant speed of agitation must be fast enough to keep the particles in suspension, but not so fast as to let the particles overflow. The UF₄ · $\frac{2}{3}$ H₂O particles are separated from the mother liquor after a few hours' residence in the hydrofluorinator. They are then washed with pure water and dried.

4.4. Specification

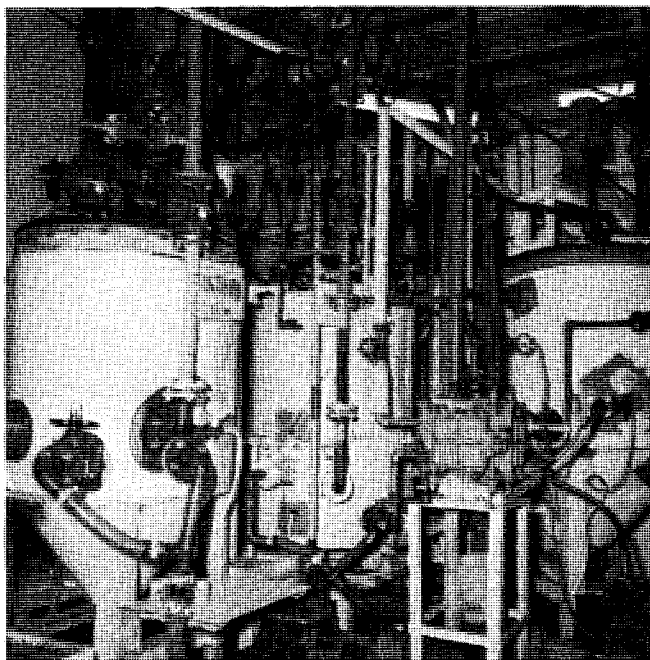
It has been recognized that the hydrated tetrafluoride obtained by the above-mentioned process has excellent washability and is a high-purity product. The decontamination factors for UCl₄ are shown in Table III.

5. STUDIES OF DEHYDRATION AND FLUORINATION

It was pursued to examine basically the properties and the dehydration of hydrated uranium tetrafluorides, and the fluorination of the dehydrated uranium tetrafluorides.

5.1. Properties of hydrated uranium tetrafluorides

The hydrated uranium tetrafluorides produced by reaction with UCl₄ and HF in the PNC process are mainly UF₄ · $\frac{2}{3}$ H₂O crystals, but three crystal structures could be obtained under different conditions. These are the orthorhombic type (UF₄ · 2.5 H₂O: R-type), the cubic type (UF₄ · 1.5 H₂O: C-type), the monoclinic type (UF₄ · $\frac{2}{3}$ H₂O: M-type) and the mixture of these types. The properties of hydrated uranium tetrafluorides are shown in Table IV, and Fig. 3 shows microscopic and scanning electron microscopic photographs of UF₄ · $\frac{2}{3}$ H₂O which has been used for the dehydration in the fluidized-bed reactor.



Hydrofluorinator

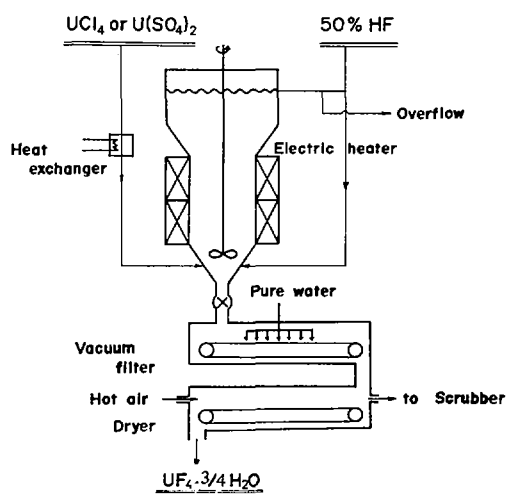


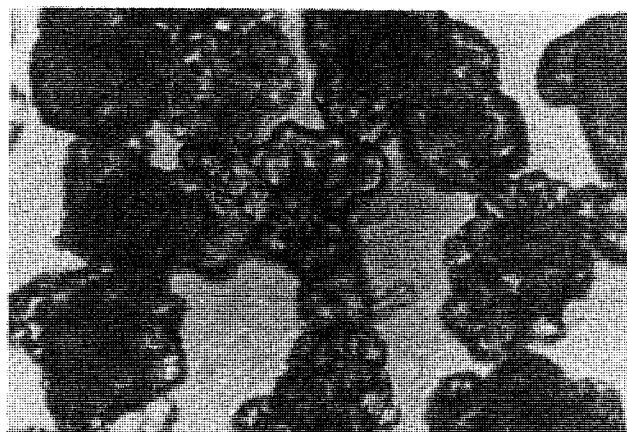
FIG.2. The hydrofluorinator at the Ningyo-toge Plant.

TABLE III. DECONTAMINATION FACTORS
OF THE HYDROFLUORINATION PROCESS

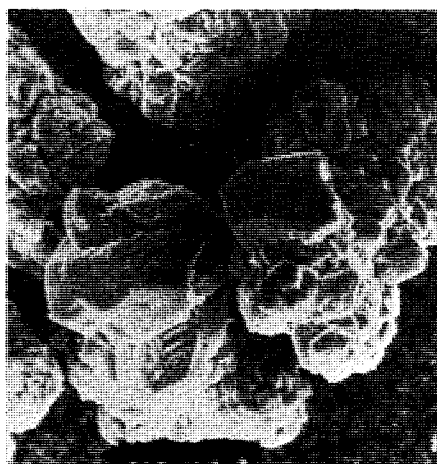
Elements	Decontamination factors
Al	120
B	30
Cd	250
Cr	300
Cu	200
Fe	1000
Mo	3000
V	2000
Ni	400
P	7
Si	5
Na	20

TABLE IV. PROPERTIES OF HYDRATED URANIUM TETRAFLUORIDES

Type	Structure	Property of crystal		Grain size (Å)
		Form	Colour	
UF ₄ · 2.5 H ₂ O (R-type)	Orthorhombic	Needlelike	Light blue	700
UF ₄ · 1.5 H ₂ O (C-type)	Cubic	Needlelike	Light blue	500
UF ₄ · $\frac{3}{2}$ H ₂ O (M-type)	Monoclinic	Granular	Emerald green	1040
UF ₄ (M(s)-type)	Monoclinic	Granular	Emerald green	1050



0 50 μm



0 50 μm

FIG. 3. Microscopic and scanning electron-microscopic photographs of $\text{UF}_4 \cdot \frac{3}{4} \text{H}_2\text{O}$.

TABLE V. GRAIN SIZES OF $\text{UF}_4 \cdot \frac{3}{2} \text{H}_2\text{O}$

Characteristics \ Sample	Before dehydration	After dehydration
Average particle size (μm)	50	48
Particle size distribution (mesh)		
+ 42	—	—
— 42 + 60	—	—
— 60 + 100	—	—
— 100 + 150	—	—
— 150 + 200	3%	7%
— 200 + 250	48	46
— 250 + 300	46	44
— 300	3	3
Specific surface area (m^2/g)	1.6	3.4
Apparent density (g/ml)	2.15	2.25

5.2. Dehydration of hydrated uranium tetrafluorides

As the $\text{UF}_4 \cdot \frac{3}{2} \text{H}_2\text{O}$ indicates excellent fluidizing properties, it is easy to control the fluidizing velocity in the reactor. The grain sizes of the $\text{UF}_4 \cdot \frac{3}{2} \text{H}_2\text{O}$ are shown in Table V.

The dehydration products and the temperature of phase transition were studied by differential thermal analysis (DTA) and thermogravimetric analysis (TGA) in a stream of nitrogen as shown in Fig. 4, and X-ray diffraction as shown in Fig. 5. The forms and crystal system of $\text{UF}_4 \cdot \frac{3}{2} \text{H}_2\text{O}$ before and after dehydration were the same, but the specific surface area was increased after dehydration. The crystal grain sizes estimated with the peak half-width of X-ray diffraction patterns (Warren's method) are shown in Table IV.

The oxygen from the decomposition of hydrated uranium tetrafluorides promotes the formation of UO_2 , and the peaks in the X-ray diffraction patterns demonstrate that uranium dioxide, UO_2 , is formed in a stream of nitrogen above 350°C . In the dehydration test in the fluidized-bed reactor only small quantities of UO_2F_2 and UO_2 are formed, for example, 0.83 wt% of UO_2F_2 and 0.03 wt% of UO_2 under the following conditions:

Fluidizing gas	dried nitrogen gas
Linear velocity of gas	11.4 cm/s
Heating rate	$3^\circ\text{C}/\text{min}$
Maximum temperature	350°C
Holding time at maximum temperature	2 h

The moisture content after dehydration was decreased to several hundred ppm from 4.45 wt% before dehydration.

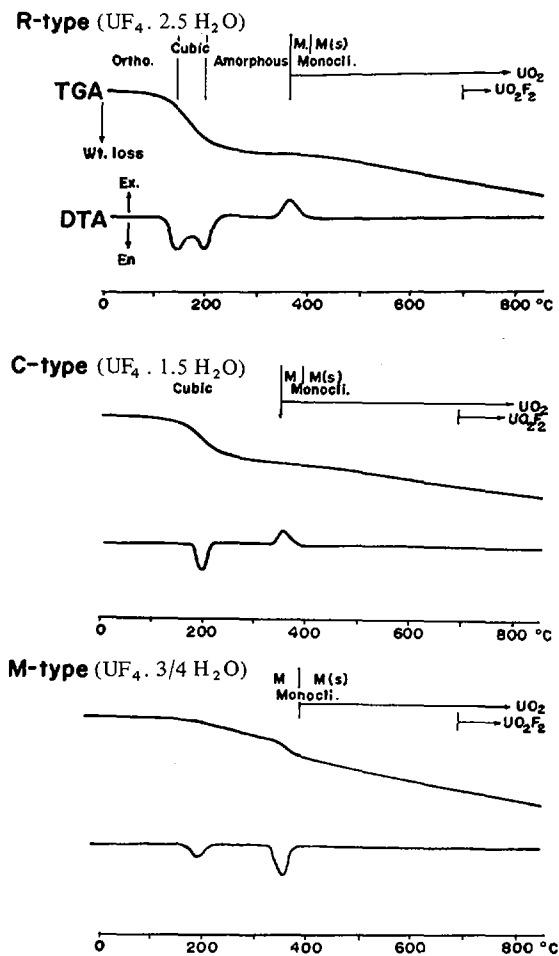


FIG. 4. DTA and TGA curves of hydrated uranium tetrafluorides.

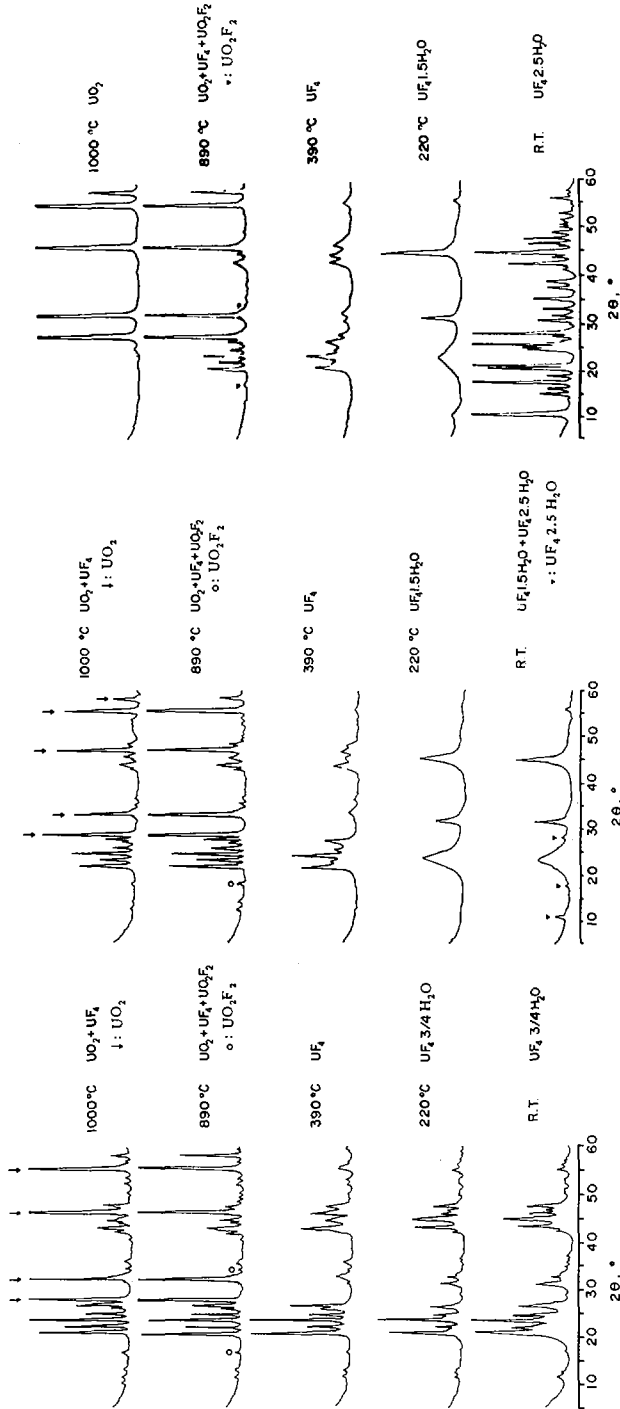


FIG. 5. X-ray powder patterns of hydrated and dehydrated uranium tetrafluorides.

TABLE VI. CHANGES IN IMPURITY
CONTENTS BY FLUORINATION
(in ppm, U basis)

Element	UF ₄	UF ₆ (UO ₂ F ₂ ag)
Al	14	< 14
B	< 0.3	< 0.3
Bi	< 1	< 1
Ca	100	10
Cd	< 0.3	< 0.3
Cu	5	1.4
Fe	< 14	< 14
Mg	2	< 2
Mn	< 2	< 2
Mo	2	2
Ni	12	< 2
Pb	< 1	< 1
Si	< 6	< 6
Sn	< 1	< 1
V	< 3	< 3
Zn	< 10	< 10

5.3. Fluorination of dehydrated uranium tetrafluorides

The physical properties, such as the particle size distribution, remained the same before and after dehydration and therefore the fluidizing properties on UF₄ fluorination were as excellent as in dehydration. In the ordinary temperature range of 400–450°C in the reactor the UF₄ produced by the PNC process had almost the same fluorination reactivity as that produced by the conventional dry process. Continuous fluorination was performed and the maximum value of fluorine utilization efficiency obtained in one stage was about 88%. The behaviour of impurities in the process was also investigated. The changes of impurity content during the conversion are shown in Table VI.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the contributions of their PNC and ACI colleagues and the Mitsubishi Metal Corporation staff members to this work. The authors also thank Dr. T. Kamiyama, Director of PNC, for permission to publish this paper.

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DISCUSSION

S. AJURIA-GARZA: What approximately is the cost of processing, say, one tonne of ore?

S. TAKENAKA: That is very difficult to say, because the cost will depend on the kind of mineral treated.

H.E. JAMES: It would seem to me that production capacities at uranium mills throughout the world tend to be rather too small to support a commercial facility based on the PNC process, at the mine site.

F.R. HARTLEY: To follow up that remark, I should like to ask you to comment on what the potential market is for UF_4 , assuming that a producer at a mill was to install such a plant. There does not seem to me at this moment to be a particularly attractive market which would induce a milling operator to extend his operation to the UF_4 stage.

S. TAKENAKA: Of course, the UF_4 has to be converted to UF_6 . Yellowcake is the current market product produced at mine sites. We intend to produce UF_4 at the site and then convert it to UF_6 somewhere else, at whatever place is convenient.

F.R. HARTLEY: What would the market be in Japan for UF_4 ? That is, if the mill were to put in a plant for UF_4 , would this be a marketable product as far as Japan is concerned?

S. TAKENAKA: PNC has a big project for an enrichment plant in the future. As you know, we have a centrifuge plant in operation. In the future we hope that the UF_4 and UF_6 that we produce will be used as experimental materials for our enrichment plant. As a next stage we are thinking of producing these materials on a commercial basis.

PROCESS DEVELOPMENT STUDY ON URANIUM-BEARING CARBONATITE

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Abstract

PROCESS DEVELOPMENT STUDY ON URANIUM-BEARING CARBONATITE.

Mineralogical and chemical studies on uranium-bearing carbonatites showed the presence of apatite and calcite as major constituents. The presence of uranium in pyrochlore structure was suggested by X-ray diffraction studies. The average uranium content was of the order of 200 ppm. The standard amenability tests indicated the refractory behaviour of the ore with respect to carbonate leaching. A 78% leaching recovery was achieved employing elevated temperature and pressure. The physical upgrading of the ore was possible using conventional gravity separation methods.

INTRODUCTION

The discovery in Pakistan of uranium in carbonatite host rock was reported. Radiometric investigations revealed the presence of low-grade mineral over a considerable area. The low-grade nature of the ore necessitated an early and thorough study from the metallurgical point of view. The first step towards such studies was to find out the type and composition of the ore and to perform amenability tests.

This paper deals with the laboratory and preliminary pilot-plant investigations. The results obtained are to be used in the development of a process flow diagram for the recovery of uranium from this ore.

DESCRIPTION OF THE ORE

Microscopic examination of ore samples showed that it consists predominantly of calcite, apatite and biotite. Sodid hornblende is a subordinate constituent of the sample; magnetite and hydrous iron oxides are present in minor quantities. The radioactive mineral grains appear to be of Metamict origin. The internal structure of the grains has suffered from radiation damage. The change of colour from yellow to yellow-brown and dark brown is indicative of the degree of radiation damage to the mineral. The X-ray patterns resemble those of the pyrochlore and betafide series [1]. Electron probe analysis of the hand-picked pyrochlore grains showed the presence of about 35% uranium, 22% niobium and 7% tantalum.

Spectrographic analysis of the ore showed that Ag, As, Au, B, Ba, Bi, Cd, Ce, Co, Cr, Cs, Ga, Ge, Hg, In, Ir, La, Li, Mo, Ni, Os, Pd, Pt, Rb, Re, Rh, Ru, Sb, Sc, Sn, Sr, Ta, Th, Tl, U, W and Zn are not detectable.

The elements detected by spectrographic analysis, together with their probable contents in per cent, are summarized as follows:

Ca	> 10%
Al, Fe, Mg, Si	1–10%
K, Mn, Na, P	0.1–1%
Nb, Ti	0.01–0.1%
Pb, V, Y, Zr	0.001–0.01%
Cu	0.0001–0.001%

The chemical analysis of a typical sample is (assay per cent):

U ₃ O ₈	HAC CaCO ₃	RCO ₃	Al ₂ O ₃	V ₂ O ₅	Fe	SO ₄
0.023	69.2	67.0	7.6	0.02	3.0	0.07
S	C	OrgC	P ₂ O ₅	K	Mn	SiO ₂
0.01	9.6	0.01	3.77	0.42	0.26	3.0

AMENABILITY TESTS

The standard amenability tests were made, using the following procedure. The ore was ground to -65 mesh and leached at 50% solids in a sodium carbonate and bicarbonate solution for 24 hours. Leach solutions containing (a) 60 g sodium carbonate and 20 g sodium bicarbonate; and (b) 100 g sodium carbonate and 50 g sodium bicarbonate were used. The influence of oxidant was studied with the addition of 10 lb of potassium permanganate per ton of ore.¹ The leaching temperatures were ambient and 80°C.

The leached slurry was filtered and the cake washed with two carbonate washes of 100 ml each and two water washes of 50 ml each. A carbonate solution containing 20 g sodium carbonate per litre was used for washing when the leach solution contained 60 g sodium carbonate and 20 g sodium bicarbonate per litre. When the leach solution assayed 100 g sodium carbonate and 50 g sodium bicarbonate, the wash solution contained 50 and 25 g of sodium carbonate and bicarbonate respectively. Each wash was with half the volume of leach solution. The slurry was readily filtered. The data are summarized in Table I.

The data indicate that the ore responded poorly to the carbonate leaching. At atmospheric pressure the increase in carbonate and bicarbonate concentration gave a small increase in uranium extraction. The extraction is also increased to some extent by the addition of oxidant and an increase in temperature. A maximum of about 25% of the uranium was extracted by the standard amenability testing procedure. For the maximum extraction the leaching temperature was 80°C, and 10 lb of potassium permanganate per ton of ore was used. The carbonate and bicarbonate concentrations were about 60 and 25 g/l respectively. The leaching was done at 50% solids. For 24 hours the bicarbonate decomposition was about 30 lb per ton of ore. This is quite high for the low extraction of uranium.

Pressure leaching was done in a one-litre stainless steel vertical-type autoclave. The autoclave was equipped with a top-mounted, turbine-type agitator. The agitator was powered by an electric motor via a reduction pulley mechanism. The pressure was regulated manually and maintained using an oxygen gas cylinder.

¹ Tons are short tons throughout the paper: 1 short ton = 2000 lb = 907.18 kg = 0.90718 tonnes.

TABLE I. AMENABILITY TESTING DATA OF URANIUM ORE

Test No.	Leach solution concentration (g/l)		Temp. (°C)	Oxidant KMnO ₄ (lb/ton)	Analysis filtrate (g/l)	U ₃ O ₈ residue (%)	Extraction based on residue and feed (%)
	CO ₃	HCO ₃					
1	35	10	80	10	0.012	0.018	22.0
2	35	10	80	0	0.01	0.021	9.7
3	35	10	25	10	0.003	0.021	5.0
4	35	10	25	0	0.003	0.022	5.5
5	59	24.5	80	10	0.024	0.017	26.4
6	59	24.5	25	10	0.004	0.021	9.3

TABLE II. PRESSURE LEACHING DATA OF URANIUM ORE

Test No.	Mesh size	Leaching pressure (lbf/in ²)	Oxidant KMnO ₄ (lb/ton)	Residue analysis U ₃ O ₈ (%)	Filtrate U ₃ O ₈ (%)	Extraction U ₃ O ₈ (%)
1	-200	50	10	0.007	0.06	69.2
2	-200	50	0	0.009	0.054	61.0
3	- 65	50	0	0.012	0.038	48.1
4	-200	90	10	0.005	0.059	78.3
5	-200	90	0	0.005	0.072	78.4
6	- 65	90	10	0.013	0.043	44.0
7	- 65	90	0	0.012	0.044	48.3

Two hundred grams of the ore ground in a disc mill to -65 mesh was leached at 50% solids for eight hours at 120°C in a carbonate solution containing 100 g sodium carbonate and 50 g sodium bicarbonate per litre. A gauge pressure of 90 lbf/in² or 50 lbf/in² was maintained with oxygen. The leached slurry was filtered. The cake was washed twice with carbonate solution containing 50 g sodium carbonate and 25 g sodium bicarbonate per litre using 100 ml of solution for each wash. The carbonate washing was followed by two water washes with 50 ml of water for each wash. The results given in Table II indicate that over 78% of the uranium was extracted. The per cent extraction was pressure and particle-size dependent. The extraction of uranium was not improved by supplemental oxidation with potassium permanganate.

PHYSICAL UPGRADING OF ORE

Uraniferous multiple oxide minerals tend to be metallurgically refractory but can possibly be concentrated rather easily, making the concentrate potentially of economic interest. The investigations made by Dodd [2] on this mineral are suggestive of a uraniferous multiple oxide, thereby directing attention towards physical beneficiation studies. The view is supported by the heavy-media separation tests.

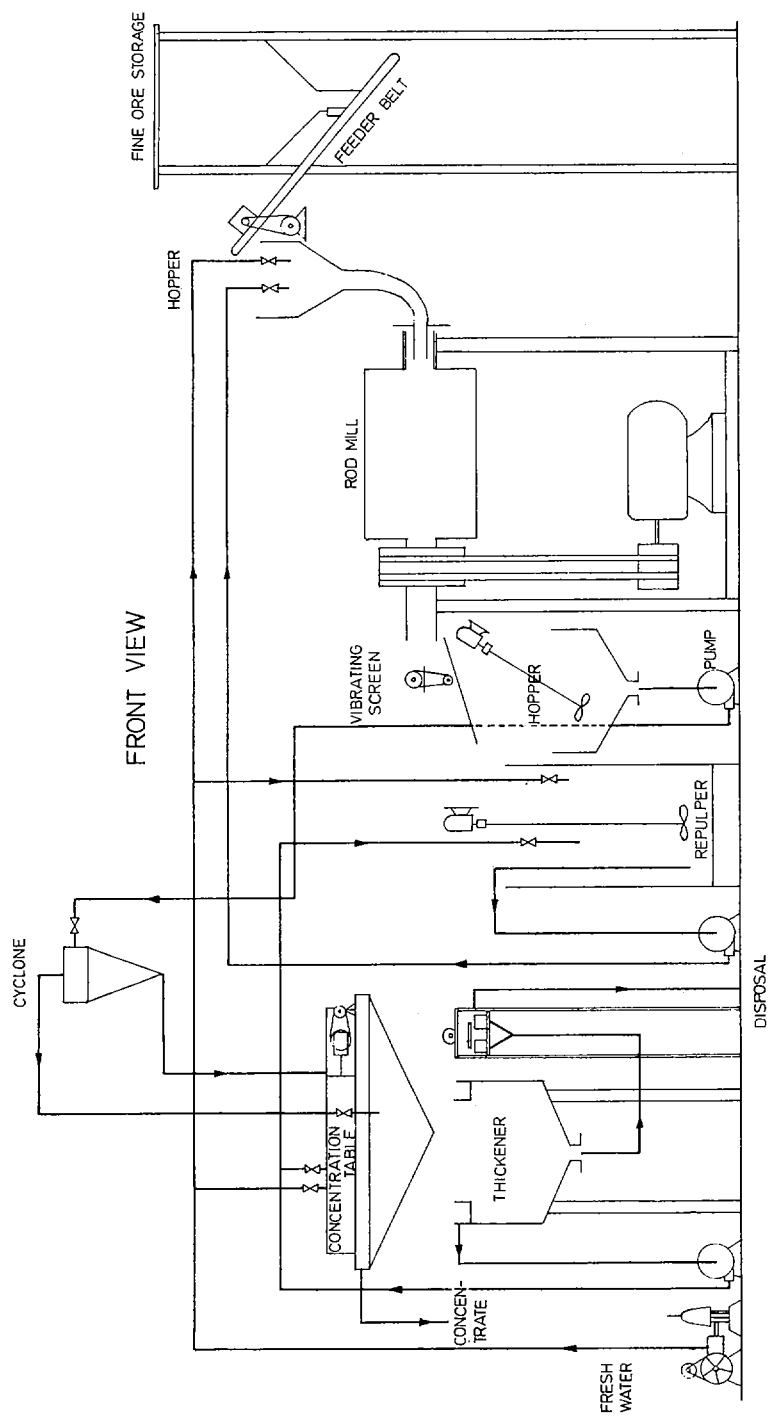


FIG.1. Flow diagram of the pilot concentration unit.

TABLE III. PARTIAL CHEMICAL ANALYSIS OF VARIOUS FRACTIONS OBTAINED BY HEAVY-MEDIA SEPARATION

Fraction	Distribution (%)	Analysis (%)			Distribution (%)		
		U ₃ O ₈	P ₂ O ₅	CaCO ₃	U ₃ O ₈	P ₂ O ₅	CaCO ₃
-325 mesh	11.96	0.019	8.72	18.6	11.3	24.0	3.5
+325 mesh lights	77.66	0.003	1.48	77.8	11.5	26.5	94.6
+325 mesh heavies	10.38	0.15	20.7	11.4	77.2	49.5	1.0

A sample was screened through 325 mesh and the +325 mesh was then separated into light and heavy fractions with tetrabromoethane (specific gravity 2.94). The results of the chemical analysis of the three products are shown in Table III. This analysis shows that approximately a seven-fold concentration of U₃O₈ has been achieved in the heavy mineral fraction, while maintaining a 77% recovery.

A batch test employing a gravity separation technique using a Wilfly shaking table proved effective.

To obtain technical data for the physical upgrading of the ore a pilot concentration unit was designed which consists of the following:

- Crusher
- Fine ore storage bin
- Conveyor belt
- Continuous discharge wet grinding mill
- Vibrating screen
- Wilfly shaking table
- Thickeners
- Storage tanks for concentrate, middling and slimes.

The flow diagram of the pilot concentration unit operating in the PAEC Laboratories is shown in Fig. 1.

The ore is crushed in the jaw crusher to $\frac{1}{2}$ in size and fed manually to a 5 ton hopper. A 9 in wide feeder belt operating at a speed of 2 ft/min takes the charge from the hopper through an adjustable gate and discharges it into the grinding mill. The 24 in dia. by 48 in long grinding mill is operated at 60 rev/min powered by a 20 h.p. motor. The grinding media (1 in dia. rods) and mill lining are of high manganese steel. The grain size is controlled by the charge of the grinding media and the solid-liquid ratio.

The grinding mill discharge falls onto a vibrating screen fitted with a 30 mesh wire gauze. The oversize particles are recycled and the rest pumped to the table through a 1 in cyclone separator. The slimes are separated with overflow of the cyclone and a slurry of the required density is obtained as underflow by varying the vortex opening. The liquid-solid ratio on the table is controlled by the addition of wash water. Separation on a concentration table depends on the density difference and on the grain size of the mineral and the gangue. The screen analysis of the ground ore (Table IV) shows that the grinding characteristics of the mineral are nearly the same as those of the ore and the distribution of uranium is uniform in each fraction.

TABLE IV. SCREEN ANALYSIS AND URANIUM DISTRIBUTION
SCREEN

Screen size (mesh)	Weight (%)	U ₃ O ₈ (%)	
		Analysis	Distribution
-10 +20	25.8	0.01	12.5
-20 +35	27.5	0.019	25.2
-35 +48	10.1	0.034	16.6
-48 +65	5.9	0.040	11.5
-65 +100	5.5	0.038	9.4
-100 +200	9.0	0.026	11.5
-200 +325	4.9	0.015	3.6
-325	11.2	0.018	9.7
Composite	100.0	0.021	100.0

The following are the conditions of a typical test run:

Ore grade	0.02% U ₃ O ₈
Physical appearance of ore	Sugary, yellowish
Feed to the table	100% -30
Solids slurry	15%
Wash water	1 gal/min
Table strokes	300/min
Stroke length	3/4 in

The discharge was split into three fractions and the results obtained are as follows:

	wt%	U ₃ O ₈ (%)
Heavy	10	0.12
Middling	70	0.005
Slimes	20	0.02

These results indicate a 70% recovery for a concentration ratio of six and a relatively high loss of uranium in slimes.

The recovery figure and the concentration ratio can be increased by employing multistage concentration. The high loss of uranium in slimes is due to the fine grinding of a high fraction of feed. It is anticipated that more rigorous control of the grinding conditions would minimize both the formation of slimes and the loss of uranium in slimes.

ACKNOWLEDGEMENTS

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DISCUSSION

S. AJURIA-GARZA: What are the total estimated reserves and the average grade of this deposit which is in process of being developed?

M. SHABBIR: The total amount of the reserves is not yet known, since the geologists are still at work, so it is very difficult for me at this moment to give you an estimate. The grade is low, being of the order of 0.02% U_3O_8 .

**KNOWN AND PARTIALLY ENGINEERED TECHNIQUES
WHICH HAVE NOT REACHED FULL APPLICATION
IN MILLING OF URANIUM ORES**

(Session III)

STORED TECHNOLOGY FOR POSSIBLE USE IN URANIUM ORE PROCESSING

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Abstract

STORED TECHNOLOGY FOR POSSIBLE USE IN URANIUM ORE PROCESSING.

Although no major flowsheet changes have been made in recent years, the uranium industry has produced a number of processing innovations which have not been fully utilized. It is probable that several of these techniques can offer improvements over current processing methods. Recently, development studies and applications of continuous ion exchange have increased; also, renewed interest in leaching modifications and autogenous grinding have become evident. In general, uranium processing has matured and is effectively used with ores now being fed to mills throughout the world. Much of the current development work relates to unit operations and processes for lower-grade ore.

INTRODUCTION

In recent years, uranium processing technology has matured and is effectively used with ores now being fed to mills. Most uranium operations have processed relatively high-grade ores, but the picture is changing. The 1974 United States uranium mill feed grade averaged about 0.18% U_3O_8 ; this is 10% lower than that fed to the mills in 1973. Decreasing ore grades are also being encountered in other countries.

Current research and development work in the uranium industry reflect the anticipation that lower-grade resources will provide a significant portion of future uranium production. A major segment of both United States and worldwide exploration programmes is directed toward areas that may contain uranium mineralization in the 0.01 to 0.1% U_3O_8 range.

Nearly every uranium milling operation has been developed to meet the particular economic needs and processing characteristics of the deposit being mined. Future operations on lower-grade resources will require even more thorough evaluation to determine where process innovations and modifications can be used.

Past research has developed a fund of potential uranium ore processing techniques that have been applied only sparingly or not at all. Some of these procedures may be useful in processing newly discovered resources. This paper identifies such stored techniques that may have future applicability in uranium ore processing.

SIZE REDUCTION

Most uranium mills throughout the world have used conventional wet grinding circuits. Recently, however, autogenous or semi-autogenous grinding on run-of-mine ore has been adapted to uranium operations in the USA. At Shirley Basin, Wyoming, the Utah International Inc. uses

a cascade mill in closed circuit with a 20-mesh DSM (Dutch State Mines)¹ screen in place of the more conventional multistage crushing and grinding circuits. Run-of-mine ore is fed directly to the mill by an apron feeder. Western Nuclear, Inc., is installing a similar grinding circuit at their Jeffrey City, Wyoming, operations. This mill is scheduled to start up in 1976.

When processing sandstone ores, the semi-autogenous mills operate at about 80% solids with a 2% to 4% ball charge. Since the Wyoming sandstone ores disintegrate easily, the mills produce rapid breakage to grain size which is followed by an intensive scrubbing action. Plugging of the grinding circuit screens or cyclones can be a significant problem if the feed contains appreciable wood or other fibres. In some instances wire has also apparently caused plugging and tangling.

Semi-autogenous dry grinding has been used successfully in the Union Carbide uranium-vanadium mill at Uravan, Colorado. These air-swept mills use a dry product classification and sizing system [1].

Both wet and dry autogenous grinding are applicable for uranium milling, and it seems likely that the technique will be carefully considered for future uranium operations.

BENEFICIATION

For many years, the uranium industry has carefully investigated the preconcentration of ores, but except in isolated instances, the results have not met the necessary uranium recovery and cost criteria. Canadian and French operations have used radiometric sorting on +1-in material while heavy-media and other gravity separation techniques have been applied in South Africa and France [2-5]. In the USA several operations preconcentrated sandstone ores by a combined desliming-leaching technique. The uranium-enriched slime fraction was separated and the remaining sand fraction was acid leached. The uranium was precipitated from the leach solutions and combined with the slimes for shipment to a finishing plant. These operations were most often associated with ores containing both uranium and vanadium. Recently, a radiometric sorting unit has been installed at the Schwartzwalder mine near Golden, Colorado, but operating information on this installation has not been published.

Flotation can be used to concentrate the uranium-bearing refractory carbonaceous fractions of some United States ores. These concentrates are not amenable to conventional processing but can be treated by roast-leach techniques or, in some instances, by autoclave leaching.

A significant amount of unreported flotation work has also been conducted on high-lime uranium ores. One objective of this work was to produce high and low lime fractions that could be treated separately. Although previous results have not been particularly favourable, the changing energy situation has created some new interest in this approach.

Past experience indicates that for uranium ores the probability of achieving both a high recovery and a high concentration ratio is low, but partial beneficiation may be possible. The potential benefits, however, are great enough so that the beneficiation characteristics of each new uranium source should be carefully investigated.

LEACHING TECHNIQUES

Ore characteristics and economics have made agitation leaching the most widely used extraction technique for uranium ores. The technique has proved applicable for a wide variety of time, temperature, and reagent addition requirements. Equipment development has also produced reliable operation with minimum maintenance and operating manpower requirements.

¹ Reference to specific equipment, trade names, or manufacturers does not imply endorsement by the Bureau of Mines.

Both mechanical and Pachuca leaching vessels have been used successfully. United States and French operations have preferred mechanical agitation whereas Pachuca units have been widely used in Canadian and South African mills. Without doubt, past experience and local preference have influenced the selection of leaching equipment for any given uranium milling operation. In recent years, equipment developments have probably favoured mechanical agitation; for example, the adaptation of large axial flow propellers has significantly reduced the horsepower requirements for many leaching operations. Manufacturers have also improved the wear resistance of both agitators and leaching vessels.

A modified agitation leaching technique is currently being studied by Marconafllo, Inc.² The system is an outgrowth of the ore handling technology which Marconafllo has been marketing for a number of years. The system leaches fine-crushed ore ($-3/8$ by 0) in a countercurrent series of reactors. The lixiviant enters each reactor through specially designed jets which fluidize the coarse mass and permit the continuous movement of solids from one leach stage to the next. Extensive tests in a 25-tons/day³ pilot plant have shown the technique to be applicable for copper oxide ores, and the principle is now being tested on uranium ores.

As the need for processing in future lower grade and more refractory ores has become evident, the interest in alternative extraction techniques has revived. Environmental considerations and energy requirements also have influenced this current interest. The strong-leach or acid-cure technique is one of the extraction procedures that is receiving additional consideration. This procedure solubilizes the uranium by contacting the dampened ore with concentrated sulphuric acid and then allowing the mixture to cure. Various washing techniques have been used to recover the solubilized uranium for subsequent purification by ion exchange or solvent extraction.

Leger and Boutonnet [6] have described the strong acid leach plant which was constructed in 1970 at the Arlit operations in Niger. The Arlit ore contains organic compounds of tetravalent uranium which require a residual acid concentration of nearly 100 g/l H_2SO_4 to achieve adequate uranium extraction. Arlit data indicate that the strong acid process reduces the acid required to less than 50% of that required for a conventional single-stage leach. The strong acid leach also exhibits enough oxidation potential to eliminate the need for additional chemical oxidation.

The United Kingdom Atomic Energy Authority (UKAEA) also has developed and patented a strong acid leaching process [7-9]. The ore is dry ground to about -1 mm, mixed with 6N H_2SO_4 and cured for 12 to 24 h at 75°C. Test data indicate a potential acid saving of about 20% for some ores. An evaluation by UKAEA shows that the capital costs for a strong acid leach plant would be of the order of 25% lower than those of a conventional design.

Western Nuclear Corp. operated an acid cure/percolation leaching plant in Converse County, Wyoming, during the early 1960s [10]. The operation which was known as the Spook uranium plant processed about 400 tons/day of ore averaging between 0.12% and 0.14% U_3O_8 . The unconsolidated sandstone ore was crushed to $-3/4$ in and fed to an agglomerator where it was mixed with H_2SO_4 and water; pellets of about 1 in dia. were formed. These pellets, containing 10 to 12% moisture, were bedded in percolation leach tanks and allowed to cure for approximately 14 h. A three-stage washing technique was used to recover the solubilized uranium; the total leach cycle time was about 72 h. Uranium was recovered from the leach solution by ion exchange. The plant continued to operate until the ore bodies in the area were depleted.

In the early 1950s the Kerr-McGee Corp. operated an acid-cure leach system on low-lime, high-vanadium ores at Shiprock, New Mexico. The ore was pugged with 10% water and 400 to 500 lb of concentrated sulphuric acid per ton of ore. A six-hour cure converted the uranium and vanadium into water-soluble salts. Clegg and Foley [11] indicate that the acid-cure operation was discontinued after about one year, principally because of changing ore characteristics.

² See footnote 1.

³ Tons are short tons throughout the paper: 1 short ton = 2000 lb = 907.18 kg = 0.90718 tonnes.

The strong-leach or acid-cure process appears to offer significant reductions of added acid for ores that require relatively high terminal acid concentrations. The system also offers the potential for minimizing crushing and grinding requirements and may facilitate subsequent solid-liquid separation.

IN SITU AND HEAP LEACHING

The unconsolidated sandstone deposits of Wyoming and Texas have been the prime target for in situ leaching development in the USA. Initial investigations were primarily acidic leaching operations, but applications of carbonate leaches have recently been expanded. Both fixed-bed and continuous expanded-bed ion-exchange systems have been investigated for recovery of uranium from the recirculating leach solution. Patterns for injection wells have been the subject of a number of patents [12-15].

In early 1975, a seven million dollar operation started up near George West, Texas, utilizing in situ leaching to feed a plant rated at 250 000 lb/a of yellowcake [16]. The plant is a joint venture of Atlantic Richfield, US Steel, and Dalco. A proprietary alkaline leach solution extracts the uranium from the ore strata through a pattern of 66 injection wells and 46 recovery wells. This leaching system covers a surface area of a little less than 3 acres. Solution circulates through the leach system at approximately 2000 gal/min, and the uranium is recovered from the pregnant solution by a conventional column ion-exchange system (CLX). Details of the leaching oxidation requirements for the George West operation have not been disclosed, but a United States patent recently assigned to US Steel teaches the use of oxygen as an oxidant for the in situ leaching of uranium with carbonate lixiviants [17].

A number of other companies are also actively involved in evaluation of in situ leaching of uranium. Wyoming Minerals, a subsidiary of Westinghouse, plans to start up a 250 000-lb/a operation near Bruni, Texas, during the latter part of 1975. Exxon, Union Oil, and Rocky Mountain Energy are other companies investigating in situ leaching technology.

Heap leaching of lower-grade uranium ores is being applied or considered for treatment of submill feed grade ores at operating mines and mills. However, a number of companies are re-evaluating heap leaching as an extraction method for relatively small, lower-grade deposits.

A significant number of moderate-scale heap leaching operations and experiments have been performed, but relatively little information on the results has been published. The Bureau of Mines has initiated studies to gather fundamental and comparative data on the heap leaching characteristics of ores from the New Mexico, Wyoming and Colorado uranium districts. It is anticipated that the first phase of this work will be completed by mid-1976.

SOLID-LIQUID SEPARATION

Vegetable gum and polyacrylamide flocculating agents have played an important role in the development of uranium processing flowsheets. These flocculants, which were developed during the early 1950s, permitted the use of reasonably sized thickeners and filters. No major new flocculant developments have occurred in recent years, but the industry has continued to improve the application efficiency. Again, each milling operation has developed techniques that fit its own specific requirements.

Currently, a modified thickener which was initially developed for waste treatment is being evaluated for solid-liquid separation in uranium circuits. Figure 1 shows a generalized view of this patented unit which has been designated the Enviro-Clear thickener. The flocculated feed slurry is introduced into a previously formed sludge bed rather than applied above the bed as it is in a conventional thickener. The technique is said to promote the agglomeration of smaller

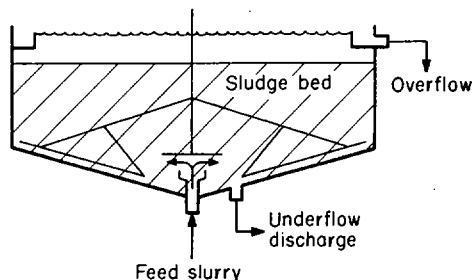


FIG.1. Enviro-Clear thickener.

particles and thereby reduce the required settler area. A five-fold or greater reduction in required settling area has been reported for some materials. Underflow densities, however, may be somewhat less than those from a conventional thickener. Various feed streams such as clay slurries and paper industry waste streams have been tested in Enviro-Clear units, but specific evaluations for each new application are recommended.

ION-EXCHANGE DEVELOPMENTS

The uranium industry has used a variety of ion-exchange techniques. The first ion-exchange operations adapted packed columns similar to those used for many years in water softening. A significant number of mills continue to use various modifications of the column exchange technique. Several types of semi-continuous or continuous countercurrent ion-exchange systems have also been installed. These systems were designed to permit resin-in-pulp (RIP) ion exchange and thereby eliminate the need for producing clarified leach liquors. For example, the basket RIP system developed during the 1950s [18–20] can handle slime pulps containing up to about 10% solids while slime slurries containing 25% solids have been ion-exchanged in the screen-mix systems used by Union Carbide, Federal-American Partners, and Western Nuclear in their Wyoming operations. Figure 2 illustrates two units of a screen-mix system; normally, five or six units are used in the absorption circuit. In the system shown, the ~200-mesh slurry is moving continuously from left to right. The resin and slime slurry contact in the air-agitated mixing tank which has a retention time of about 20 min. The mixture of resin and slime slurry overflows and is air-lifted to a 65-mesh screen which separates the resin from the slime slurry. The separated slime slurry flows to the next unit on the right. The resin stream is split; a part goes to the next unit on the left and the remainder recirculates to the mixing unit. This resin recirculation permits adjusting the resin/slurry ratio in the mixing tank to give optimum resin retention times. All splitters operate on the same time cycle to assure uniform movement of resin through the circuit.

Similar equipment arrangements have also been used for the elution circuits, but the number of stages is normally 10 to 14. The larger number of stages permits the production of relatively high-grade eluates containing 10–12 g U_3O_8 per litre.

Several continuous or semi-continuous upflow ion-exchange columns have continued to receive additional study during recent years. These systems include the United States Bureau of Mines column [21, 22], the Clote-Street column developed in England [23], and the Himsley column, a Canadian development [24].

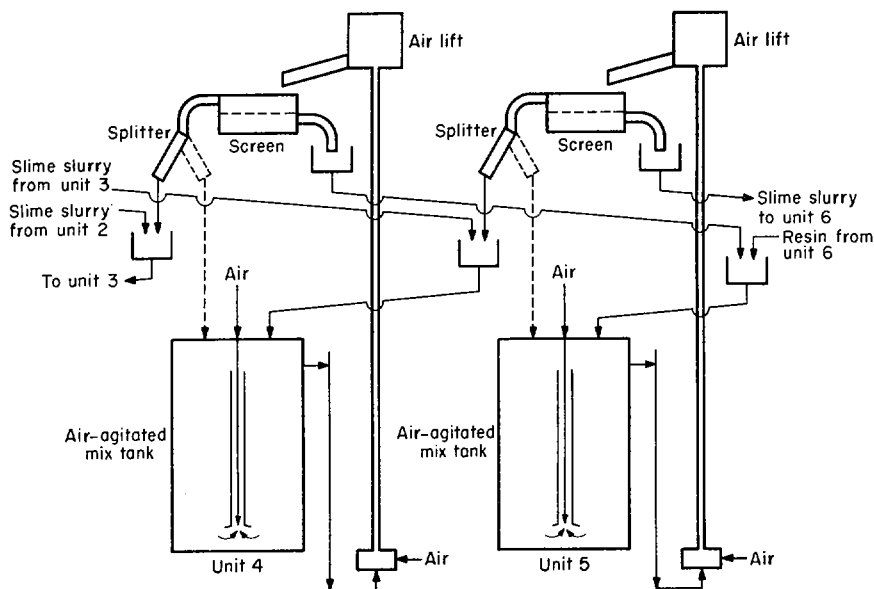


FIG.2. Screen-mix resin-in-pulp system (two units).

In these columns, the upflowing feed solution fluidizes the resin in a series of separate compartments. A generalized schema of the United States Bureau of Mines and the Clote-Street absorption column operation is shown in Fig. 3. Orifice plates divide the column into a series of compartments, the orifice openings constitute about 5% of the column cross-sectional area. The feed solutions flow upward through the column at rates of about 10 to 20 gal/min per square foot. This fluidizes the resin in each compartment, but the flow through the orifices prevents the resin from dropping into a lower compartment. Periodically, the feed stream is momentarily interrupted, a discharge valve is opened, and an increment of resin is discharged. This operational cycle permits the increments of resin to move down through the column countercurrent to the solution flow. A similar column arrangement can also be used for elution of the loaded resin. The Himsley column uses an external piping arrangement that permits resin transfer between the sections without interrupting the feed solution flow.

With the presently available resins all of these columns can handle slime slurries containing up to about 7–8% solids. The upper limit is reached when the combination of slime slurry density, viscosity and velocity causes the resin to overflow with the slurry leaving the top of the column.

Multiple-compartment ion-exchange columns are being considered for use in conjunction with the processing of gold-uranium ores in South Africa and the recovery of uranium from the low-grade solutions produced by in situ mining ventures in the USA.

A multiple-tank fluidized ion-exchange system has been developed by R.R. Porter. Counter-current resin solution contact is made in a series of individual absorption vessels. A schema of the Porter system is shown in Fig. 4. The solution flows continuously from left to right through a series of five absorption vessels; either gravity flow or pumping can be used. Periodically, the loaded resin in unit 1 is transferred by airlift to the resin holding tank. The sequence valve then

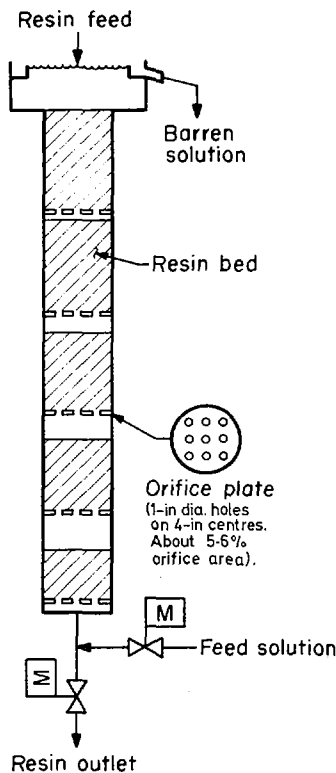


FIG. 3. Multiple-compartment ion-exchange column.

shifts and the resin in unit 2 is moved into unit 1. The cycle continues until the resin has been emptied from unit 5; freshly eluted resin is then transferred into unit 5 from the elution system. Packed-column downflow elution is used to recover uranium from the loaded resin. Normally, a three-column moving-bed elution system is used to minimize resin inventory.

Since the feed solution continues to flow during resin transfer, some back-mixing does occur but apparently this produces only a minimal decrease in efficiency when relatively low-grade feed solutions are used.

A modified version of the system is being installed at the Rossing plant in Southwest Africa. The flow to this five-stage unit will be about 10 000 gal/min. Startup is anticipated during 1976.

The potential for processing low-grade uranium ores is also of renewed interest in applying the Higgins column. During recent years, this column, which was initially developed at the Oak Ridge National Laboratory in about 1951, had been used for high flow-rate water softening. Rates in excess of 2000 gal/min have been accommodated in single units. A schema of this pulsed fixed-bed system together with an operating cycle description is shown in Fig. 5.

The present Higgins units can be classified as third-generation equipment. Significant innovations have been engineered into the system. As with other continuous exchange units, experience has shown that pilot scale studies are desirable for each individual metallurgical application.

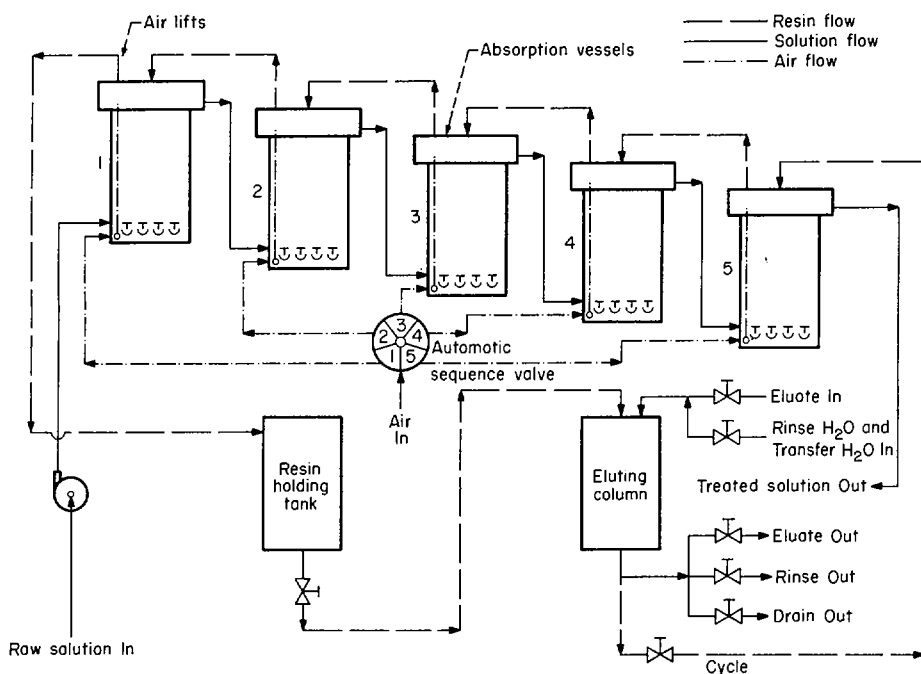


FIG. 4. Porter fluidized-bed ion-exchange system.

SOLVENT EXTRACTION

The uranium industry uses mixer-settler equipment for nearly all of its solvent extraction operations. The mixer-settler offers appreciable flexibility and relatively few operational problems. The systems, however, do require a relatively large solvent inventory, and emulsion or sludge formation problems occur in some operations. Research and development studies during recent years have attempted to minimize these drawbacks. This work includes impeller design studies and the use of coalescers to improve phase disengagement [25]. Ritcey has presented a review which discusses the selection and application of solvent extraction contactors [26].

A variety of columns and other solvent extraction equipment has been proposed for uranium extraction, but the scale-up data for many of these units have been minimal. This has without doubt limited the application of these units for mills built during the past five years.

Solvent extraction has proved to be an economical uranium recovery method and no doubt will be used in a significant percentage of future uranium mills.

YELLOWCAKE PRECIPITATION

Over the years, the specifications for uranium yellowcake concentrates have gradually become more restrictive. Many mills have found it necessary to install supplemental purification operations to meet the specifications for contaminants such as molybdenum, phosphorus, sulphate, vanadium and sodium. Both auxiliary solvent extraction and multistage precipitation flowsheets have been

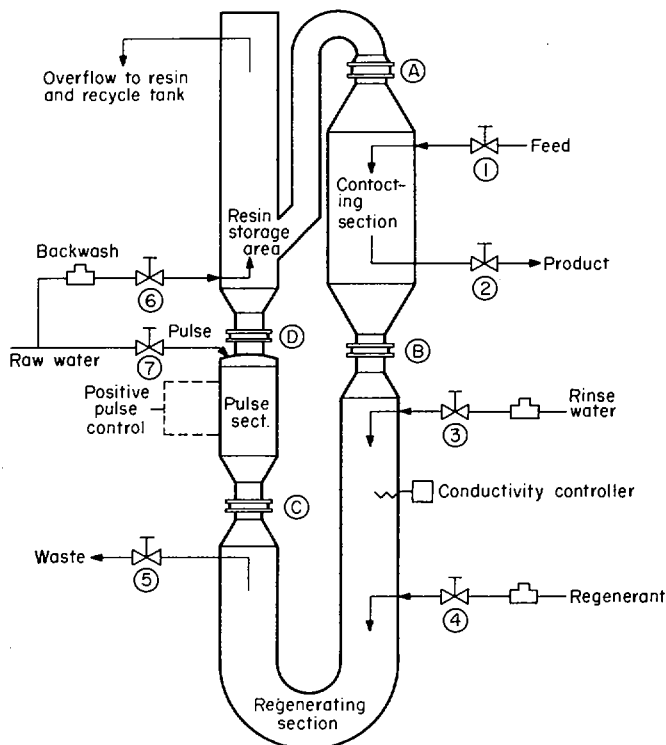


FIG.5. Higgins column.

Operating cycle for water softening: The system operates by continuous cycling between a 'run mode' and a 'pulse mode'. During the run mode of 5 to 20 min duration, the main valves A, B and C are closed, effectively dividing the loop into three sections. In the contacting section, raw water is fed down through the resin bed (valve 1) and treated product water is delivered from the bottom of the bed (valve 2). Simultaneously, regenerant is fed (valve 4) through loaded resin in the regenerating section at the bottom of the loop and exits as waste after stripping the resin free of unwanted ions (valve 5). At the same time, loaded resin is dropping through open main valve D into the pulse section. The positive pulse-control sensors close valve D when the pulse section is properly filled. Meanwhile, clear rinse water (valve 3) washes regenerant traces from the stripped resin (next to enter the contacting section). Valve 3 is operated by a conductivity controller that senses the interface of rinse and regenerant solutions to ensure that all regenerant traces are removed before rinsing is terminated and positions interface properly. During the pulse mode, main valve D is closed. Main valves A, B, and C are open. Valves 1, 2, 3, 4 and 5 are closed. A hydraulic pulse of raw water (valve 7) transfers resin around the loop at a pre-selected distance. This movement puts loaded resin from the pulse section into position for regeneration while pushing stripped and rinsed resin into the contacting section and moving loaded resin from the contacting section around to the storage area. A backwash from valve 6 cleans resin fines and foreign particles from the loaded resin which is fluidized at this point. The pulse mode takes about 30 seconds.

adopted. At least three United States uranium mills have also used modifications of the hydrogen peroxide precipitation technique. This procedure effectively controls the phosphorus, vanadium, sulphate and sodium contents of the yellowcake product. The yellowcake product is normally shipped to refining operations for conversion to a fluoride product.

A number of flowsheets for producing UF_4 at uranium mills have been proposed but not yet adopted. This hesitancy probably has occurred because the production of nuclear grade products at remote mining and milling operations can present particular difficulties. Also, if further purification of the fluoride product was necessary, this would significantly reduce any advantage for mill-site UF_4 production.

Relatively little research has been directed toward the final product recovery steps. Additional studies should be considered because future recovery from lower-grade ores may require even more selective product precipitation techniques.

SUMMARY

Uranium ores throughout the world vary considerably. Almost all recent processing developments have attempted to build a process around the requirements of a given feed material or to minimize economic and environmental constraints that develop in operating mills. Each uranium deposit presents a relatively unique combination of processing requirements, and this situation will probably continue to exist in the future. Lower-grade ores will supply an increasing percentage of the world uranium needs and the uranium processing industry will no doubt concentrate on developing unit operations which will minimize the cost of treating these resources. Design criteria for new uranium milling operations will also place more emphasis on processing energy requirements. Developed techniques such as those discussed here may be applicable and appropriate for some of the future uranium resources.

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DISCUSSION

F.C. LENDRUM: In the three papers from Canada, South Africa and the United States, the upflow ion exchange column has received a lot of attention. In your paper you mention the Porter column which in some respects is similar to the Himsley column in that a plug of resin is moved forward through the system. I should like to ask whether in the work carried out in the USA and in South Africa any tendency was noticed for the resin to segregate in the upflow columns, i.e. for the fine resin to work towards the top. If such a situation developed one might expect, particularly if a lot of attrition occurred which resulted in a high proportion of fine resin, that the barren liquor would be contacting loaded resin rather than fresh resin. Any comments?

D.C. SEIDEL: We have noticed segregation of resin, but apparently the density change from unloaded to loaded resin is great enough so that the particles do work down through the column. In our operations, the unloaded resin will have a specific gravity of about 1.1 or less, whereas the loaded resin will have a specific gravity approaching 1.25. This difference is apparently enough to allow the relatively fine resin to move down through the column. The very fine resin is elutriated from the column, and we have not encountered any build-up of an intermediate-size fraction.

H.E. JAMES: To Mr. Lendrum's question I should like to make the point that you cannot really have any confidence about resin attrition and distribution of the resin in these columns until you have tested fairly large units. The upward movement of fine resin inside the column has been the least of our problems in South Africa. The main difficulties that we have experienced relate to maintenance of a stable resin inventory at each stage and constantly moving resin in and out of the column. Both problems have been overcome, the former by development of a patented design of the bottom distribution tray, the latter by careful design of the hydraulics for resin movement.

Liquid flow distribution has also been put forward as a problem with fluidized bed columns. Again, however, with good hydraulic design, even liquid flow distribution and hence even fluidization can be achieved. We have in fact observed the 2.5 m diameter column to be better in this respect than the smaller pilot units that we previously tested.

D.C. SEIDEL: I agree with your comments. Our experience and the information we have indicate that extrapolating resin breakage from small-scale equipment may be risky. We have also observed that specific resin characteristics must be considered. Some resins crack or break in half while others tend to exfoliate in layers. The exfoliated shells may be very thin but they do present a large cross-sectional area. The shape factor has a major influence on the elutriation characteristics of the particles. We think that one should examine not only the equipment but also the particular resin that is being considered.

A.W. LANKENAU: I would like to comment on two aspects of the paper. At the Shirley Basin Plant of Utah International, we use semi-autogenous wet grinding. We follow this with wet pulp storage in air-agitated tanks, and as a consequence we are trying to screen the slurry for the solids content of the mill discharge, which is +70%. We are doing this on fixed static screens of DSM type, but we have modified them to straight decks. We have found that we do not get a very good screen efficiency and we have also tested high-frequency Derrick screens. It seems that they are very good and we are proceeding to install a Derrick screen in the plant for our plant test; we are very confident that these screens will solve our problem. Anyone who attempts to use this system should be aware of this problem.

I should also like to mention that we have tested the Enviro-Clear thickener at the Lucky Mac plant of Utah. We found it excellent, but it is very important that no air is entrained into the feed. If it is, the system acts like a big flotation machine. Mr. Seidel's comment on the underflow density being somewhat less is correct. We also found that there was some increase in flocculant consumption.

F.R. HARTLEY: I would like to ask whether you have had any experience or whether you know of any experience with the operation of lamellar thickeners, particularly for product thickening before dewatering. Secondly, I am rather surprised that there has not been any mention of the use of the Asahi continuous ion-exchange process. We have a pilot plant which is capable of operating at about 7000 gallons of water per hour. Although we have not tried it for the recovery of uranium we have successfully operated for quite a number of months on lime slurries and we have found that there is no resin attrition at all owing to mechanical handling of the resin. We found that resin attrition is about the same as for a fixed-bed system.

D.C. SEIDEL: I am not familiar with the thickener applications that you have mentioned and appreciate your bringing it to our attention. I would also be very interested in any reports or other information that you might have on this technique.

F.R. HARTLEY: The Asahi system is not dissimilar to the Higgins column, except that there are no valves at all which close on resin; all the valves close on clear water. The resin moves very smoothly, and very fast and quite short cycle times can be used so that there is virtually no entrainment of solids.

P.A. BONHOTE: Do you know of any processes that can be used for removing radium from tailings. As you know, the radium content of tailings is a major cause of the environmental problems. I was wondering if any research work had been done that you are aware of.

D.C. SEIDEL: The Bureau of Mines has done some work at the Salt Lake City Metallurgy Research Center (see Report of Investigations RI-8099, US Bureau of Mines).

P. MICHEL: I was very astonished at the efforts being made on continuous ion exchange. It seems that solvent extraction has not received as much attention. I understand that mixer-settlers work quite well and that solvent inventory is not considered a problem in most industrial countries, but we have had problems in this regard. For these applications we would prefer a solvent extraction system which is less operator-prone and has a lower solvent inventory. We have therefore begun in France some work on the development of large pulse columns for use in uranium solvent extraction. So far we have only carried out extraction tests but not stripping tests. In the extraction tests we have obtained specific flow rates of about four litres per square centimetre per hour and we hope to have a solvent inventory of a third or a quarter of the solvent inventory we would have had with mixer-settlers.

**PROCESS PROBLEMS AND DEVELOPMENTS
FOR SOME NEW URANIUM ORE OCCURRENCES**

(Session IV)

NOTES ON TECHNICAL ASPECTS OF PROBLEMS CONNECTED WITH THE TREATMENT OF SEVERAL NEW-GENERATION AUSTRALIAN URANIUM OCCURRENCES

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Abstract

NOTES ON TECHNICAL ASPECTS OF PROBLEMS CONNECTED WITH THE TREATMENT OF SEVERAL NEW-GENERATION AUSTRALIAN URANIUM OCCURRENCES.

The paper has three distinct parts. The first describes characteristics of the Nabarlek orebody as they affect any proposed mining and treatment process. The mineralogy of the Nabarlek orebody is well suited to the application of traditional acid leach solvent extraction technology, and parameters have now been defined for a satisfactory flowsheet. However, the unique feature of this orebody is its high grade, and this has presented several problems of significance, which form the main subject of this note. The second part of the paper describes the technical feasibility of recovering uranium from a carnotite-bearing ore by pelletizing and percolation leaching with sodium carbonate/sodium bicarbonate solutions. The investigation was prompted by a particular problem presented by this orebody in the very poor settling characteristics displayed during pilot plant operations using more conventional agitation leaching/countercurrent washing. Using lime as a cementing agent, pellets were prepared and laboratory and pilot-scale percolation leaching indicated that uranium extraction in excess of 97% could be achieved without mechanical breakdown of the pellets. The paper is rounded off by a note on the problems of extracting uranium from a third type of uranium mineralization which occurs over a relatively wide area in central Queensland, where uranium appears in brannerite and zircon in a matrix which includes apatite and carbonate minerals. This note refers to work carried out specifically on a 'new generation' find — the Valhalla Deposit adjacent to Mt Isa.

INTRODUCTION

The mineralogical environment and grade of uranium deposits which have been found in Australia probably have a wider range of compositions and values than occurs in most other countries. The 'first generation' deposits included the refractory davidite ore of Radium Hill, the association of uraninite with the highly basic alanite and related calc-silicates of Mary Kathleen, uraninite and secondary uranium minerals in Rum Jungle ores which contained carbonaceous and other shales which were very prone to 'slime' badly during leaching, the rich uraninite-containing ores of the South Alligator region, where one small mine treated run-of-mine ore at 1% U_3O_8 , and finally, the low-grade refractory deposits around the Mt Isa region, typified by Anderson's Lode, which defied all attempts to develop a route for economic processing. Each deposit presented specific problems and each treatment plant was presented with ore of unique characteristics, as reflected in the various descriptions which have been published (Almond, J.N. (1958), Couche, R.E. and Hartley, F.R. (1958), Fitzgerald, M.L. and Hartley, F.R. (Collaters) (1965), Harris, J.W., Boag, J. and Hemmingway, P. (1961), Hartley, F.R. (1968)).

Although one uranium 'province' has now been discovered in the Northern Territory of Australia, of such magnitude as to be able to support several operations treating quite similar types of ores, nevertheless the 'new generation' of uranium discoveries in Australia has continued to show a wide range of mineralogical composition and grade. Among these discoveries are the Westmoreland deposits in north-eastern Queensland, the discoveries of low-grade deposits in the northern Flinders Ranges of South Australia, later discoveries of apparently roll-front type of deposits near Lake Frome in South Australia, further discoveries of the Anderson's Lode variety such as the Valhalla deposit in the Mt Isa region, the very significant discovery at Yeelirrie in Western Australia - the first substantial deposit in Australia which will require alkaline leaching for economic processing - and, of course, the very large deposit in and adjacent to Arnhem Land in the Northern Territory.

One paper has appeared, describing investigations into the treatment of some of these ores (L.H. Goldney, R.G. Canning and J.E.A. Gooden (1972)). However, much of the information required for detailed comparative review has not yet been released for publication. Lacking the ability to produce such an overall review, it was decided to present what amounts to three notes on three specific investigations. The first outlines particular opportunities and problems associated with treating the high-grade Nabarlek ore. The second note outlines an investigation into percolation leaching of pellets as a possible solution to the problem of treating an ore showing poor filtration and thickening characteristics. The third note refers to the still intractable problem of the refractory ores for the Mt Isa region, typified here by the more recent Valhalla discovery.

OPPORTUNITIES AND PROBLEMS WHEN CONSIDERING THE TREATMENT OF A HIGH-GRADE URANINITE DEPOSIT

The Nabarlek orebody, located approximately 300 km east of Darwin in the Northern Territory, is of exceptionally high grade. The primary ore consists essentially of pitchblende in a matrix of chlorite and muscovite. The pitchblende is variable in habit, with a significant proportion of massive, high-grade occurrences, with lesser amounts finely dispersed down to the micron level, interleaved with flakes of silicate gangue. Some massive pitchblende extends to within 2 metres of the surface, although above the water-table, at approximately 20 metres, extensive weathering has occurred with secondary ore containing sklodowskite and rutherfordine in a matrix of muscovite, chlorite, quartz and hematite/goethite. There are minor amounts of other minerals, including kasolite, curite and pitchblende.

Although the total geological reserve and average grade have been assessed with reasonable confidence, there is considerable uncertainty as to the distribution of grade within the deposit, due to the random distribution of massive, high-grade material. Some indication of the quality of the grade distribution can be obtained when one considers that at a cut-off of 0.1% U_3O_8 , total geological reserves are estimated to have an average grade of the order of 2.4% and a U_3O_8 content of approximately 9500 tonnes, whilst at a cut-off of 2.5%, geological reserves are estimated to have an average grade of approximately 9.3%, yet contain approximately 8250 tonnes of U_3O_8 .

Investigations into the treatment of composite fractions of the primary and secondary materials have demonstrated that the ore is readily amenable to conventional acid leaching and subsequent extraction processes, as might be anticipated from its mineralogy. However, the massive form of much of the pitchblende - particularly in the primary ore - and the intrinsic

high-grade nature of the ore present several interesting opportunities for alternative methods and a few unusual problems in the application of the conventional extraction processes.

Gravity Concentration

Because of the small capital investment required and the very low costs for operating a gravity separation plant, one attractive option was to consider the possibility of producing material from such a treatment of a sufficient grade to market directly. This would enable an early generation of cash for investment in the subsequent chemical treatment plant, retaining a high local equity and requiring little, if any, loan funds.

Experimental work was therefore conducted, using shaking tables, with sized fractions of drill core samples. These yielded, from the primary ore, approximately 60% of the U_3O_8 in a concentrate of 50% U_3O_8 . For the secondary ore, a concentrate of 30% U_3O_8 for a recovery of 45% was achieved. Concentrate grades and recoveries from a friable surface secondary ore sample, in which a high proportion of the uranium values were in the finer size fractions, were even less. Some improvements could probably be obtained by more sophisticated procedures, particularly by the application of heavy-media cycloning, although the inherently fragile nature of most of the secondary uranium minerals suggests that recoveries from the secondary part of the orebody would remain relatively poor for any simple gravity separation technique. The results indicate that several thousand tonnes of uranium could be won into a concentrate of the order of 50% U_3O_8 . Whether the economic objective which suggested this study would be met could only be discovered by careful economic analysis, after determining the likelihood of a market and price for the concentrate from this gravity operation, together with marketing information for the yellow cake, which would be subsequently produced from the tailings.

Selective Mining and Treatment of High-Grade Ore

The geological mining studies indicated that about 80% of the total uranium values should be able to be mined selectively to yield a high-grade ore of approximately 8.5% U_3O_8 grade. A decision to use this option would imply production of a low-grade stockpile containing approximately 20% of the total uranium for subsequent treatment, probably by blending with other lower-grade ores in the adjacent region. The choice of this option then gave two options for the subsequent chemical treatment. Acid leaching of ore of this grade yields a liquor with a high concentration of U_3O_8 , and experiments were therefore conducted on winning a yellow cake by direct precipitation from this liquor. Using a two-stage process, a yellow-cake product of acceptable purity for refinery feed was produced (90% U_3O_8 after calcination). Overall recovery from the feed material into the yellow cake was 97%. The first-stage precipitate, which contained 1% of the total uranium extracted in leaching, would need to be added to the low-grade ore stockpile, where its ferric iron content may be beneficial in reducing the amount of oxidant required in the subsequent leaching.

However, the inclusion of an amine extraction stage, with ammonia sulphate stripping and ammonium precipitation circuit, yielded a much higher-grade product (99.5% U_3O_8 after calcination). Although the avoidance of the more sophisticated solvent extraction process has some attractions, the economic advantages which might accrue to the first option depend very much on the acceptability of the lower-grade product on the market and, to some extent, on the cost of raffinate neutralization prior to tailings disposal - a cost which is totally offset by the first option.

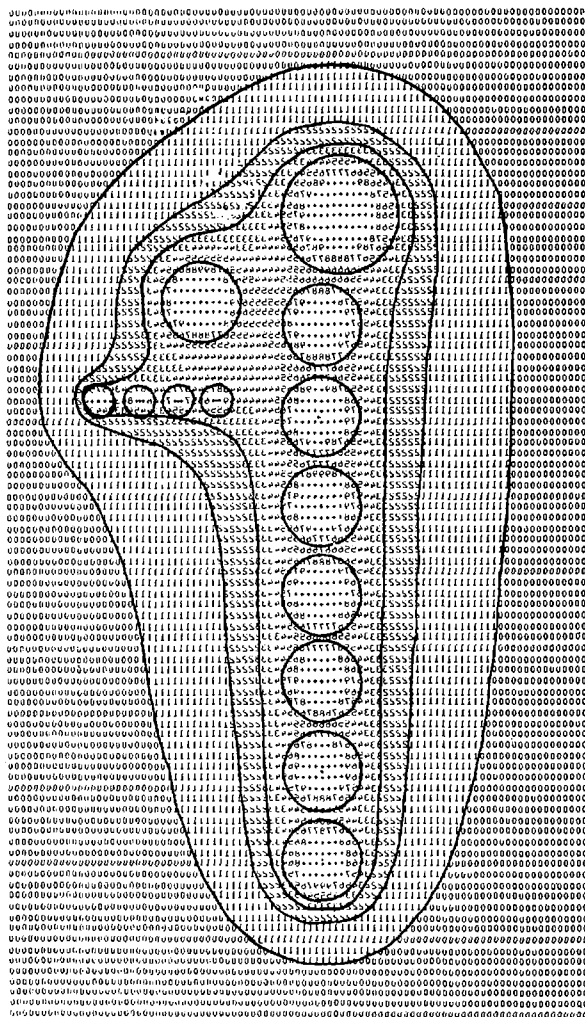


FIG.1. Contours in ranges of mR/h around a conceptual plant layout -- illustrative model.

Specific Problems Associated with Higher Grades

It is apparent that particular attention is required to provide adequate protection from gamma radiation and radon gas released in handling this ore. This will apply to mine-face and through the transport, stockpile and blending stages, when there could be occasions where pods of pitchblende containing in excess of 50% U_3O_8 will be handled.

TABLE I. GAMMA-RADIATION DOSE-RATES
IN PILOT TESTS

(From 260 litres of 40 wt% solids slurry of
8.0% U_3O_8 ore in 0.7 m dia. rubber-lined
steel tank)

Condition	Dose-rates (mR/h)	
	Side surface	1 m away (horizontal)
Neutral slurry	8.0	0.6
After 5 h leaching	3.9	0.2
After 10 h leaching (Liquor, 67 g/l U_3O_8)	2.5	0.1
Partly washed solids (Liquor, 22 g/l U_3O_8)	7.0	0.6
Fully washed solids (Liquor, zero U_3O_8)	8.0	0.6

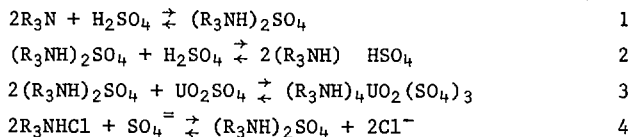
Adequate precautions must also be extended to all the extraction plant operations where either the original ore or leached tailings are still in the process vessels. A computer program has been designed to give basic dose-rate contours around a proposed plant layout. The information feed-in includes equipment, unit position and characteristic dose-rate and range-law for each unit, giving a total dose-rate calculated in the horizontal plane. Contours in ranges of mR/h are shown in Figure 1, for 8.5% U_3O_8 ore processing. This is a preliminary figure, produced mainly to illustrate the procedure, and refinements are to be made to the basic program to include the effects of process streams in transit and effective shielding by the equipment materials of construction. Radiation and radon release measurements are in process of being made for inclusion in these programs. One interesting facet already observed is the self-shielding against gamma radiation which will be available over the top of the high-grade thickeners and final leach tanks. Table I illustrates the gamma-radiation conditions during an experimental leaching operation.

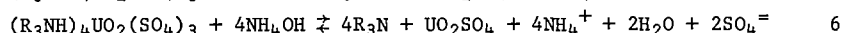
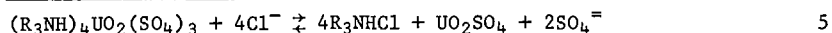
Implications of High-Grade Pregnant Uranium Liquors in Tertiary Amine Extraction

With high-grade ores containing 8-10% U_3O_8 , the conventional sulphuric acid treatment route should produce a pregnant liquor of approximately 25 g/l U_3O_8 feeding the solvent extraction circuit. This concentration is considerably higher than that from typical ore grades which give liquors of 1 to 2 g/l U_3O_8 .

The following reactions typify the processes proceeding in the extraction and stripping stages, numbers 1 to 4 occurring during extraction, number 5 in chloride stripping and number 6 in ammonium sulphate stripping.

Extraction Reactions:



Stripping Reactions:

Reaction 5 shows that for every uranyl ion stripped, four chloride ions are transferred to the organic phase. These chloride ions are subsequently transferred to the aqueous phase during extraction and pass out of the circuit in the raffinate. The phase ratios of aqueous:organic in extraction circuits treating liquors containing between 1 to 2 g of U_3O_8 per litre are between 5 and 2.5:1, and the chloride ion concentration does not exceed about 1 g/l in the raffinate. However, at a phase ratio of aqueous over organic of 0.2, as required for treating the liquor containing 25 g of U_3O_8 per litre, the chloride ion theoretically can reach a concentration of 13 g/l. In fact, before reaching this concentration, it interferes with the extraction of uranium. If chloride stripping is contemplated, a separate solvent sulphating stage must therefore be incorporated.

Using ammonium sulphate stripping, reaction 6 indicates that the free amine species is produced for recycle, and no deleterious anion species is introduced into the extraction circuit. However, the free amine must be converted to sulphate in the extraction circuit, and equations 1 to 3 show that 2 moles of sulphuric acid are extracted with 1 mole of uranyl sulphate. Since the amine typically loads uranium to about 70% of the theoretical value, and sulphate to 100%, the actual acid consumption is about 3 moles to 1 mole of uranium. Pregnant liquors typical of most acid leach circuits contain between 0.05 and 0.025 moles per litre of sulphuric acid, and it can be seen that at the phase ratios normally used (5 to 2.5:1 A/O) there is sufficient acid to completely sulphate the solvent. However, when treating pregnant liquors from Nabarlek high-grade ore containing 25 g/l U_3O_8 , phase ratios are only of the order of 0.2, and there is a net deficit of sulphuric acid. This deficit must be added to the solvent extraction circuit in a separate acidification stage. However, the aqueous phase acid concentration must not be too high, otherwise reaction 2 will predominate, producing a bisulphate form of the solvent, which reaction consumes twice as much acid.

PELLETIZING AND PERCOLATION LEACHING OF A URANIUM ORE

Laboratory and pilot-scale investigations into the alkaline leaching of a carnotite uranium ore revealed that a serious problem arose from difficulties in the settling and filtration of the leached slurry. Guided by previous experience in Amdel on the percolation leaching of green pellets to extract copper from its ore, and on publications relevant to this technique (Mashbir, D.S. (1964), Crosby, J.W. (1955), Keyes, H.E. (1930), Sullivan, J.D. and Towne, A.P. (1930), Lodding, W. (1967)), an alternative treatment process was investigated involving pelletizing the ore and percolation leaching of the pellets with sodium carbonate/sodium bicarbonate solutions at temperatures in the range 90°C to 100°C.

The ore examined contained approximately 0.24% U_3O_8 , the only uranium mineral of any consequence present being carnotite. The gangue was principally dolomite, with small amounts of clay.

The pelletizing process was investigated. A number of variables were studied, including the use of various binders such as Portland cement, Wyoming bentonite and sodium silicate. The binder which produced the strongest pellet was hydrated lime. More detailed studies of the variables affecting the quality of the pellets were then investigated, using hydrated lime as the binder.

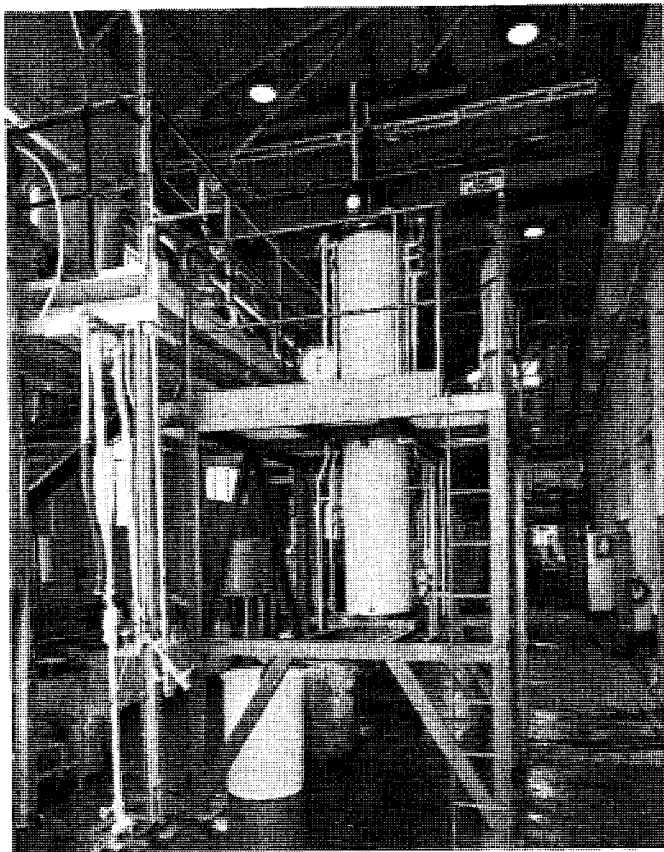


FIG.2. Equipment for percolation under pressure.

Apart from the pelletizing conditions themselves, other significant variables were the curing time and the conditions under which the pellets were cured. The main parameters by which the pellets were judged were the compressive strength of the green pellet and the resistance of pellets to break-down in a solution containing 40 g/l of sodium carbonate at 90°C.

The load on a 5 mm pellet at the bottom of a 9 m column was calculated to be approximately 0.25 kg, ignoring wall effects. The best-quality pellet was obtained using 4% of lime, ore which was pre-dried before blending with lime, and pellets which were cured under sealed conditions for 11 days. These conditions yielded pellets with a compressive strength of 1.74 kg when freshly prepared, and 1.34 kg after soaking for one hour.

Having in mind the cost of drying and of storage capacity, a compromise was made, and for the pilot plant study pellets were made under the following conditions. The ore was ground to approximately 70% -45 μm without drying,

and 4% lime was added. Pellets were produced on a 1 m diameter disc, with a rim height of 280 mm, an angle of inclination of 47° and a rotation speed of 18 rev/min. The freshly prepared pellets had a mean moisture content of 21.8% and compressive strength of 0.27 kg force, with diameters varying from 3 to 5 mm. After curing for seven days, they had a moisture content of 21% and a mean compressive strength of 0.95 kg force. The U_3O_8 content was 0.28%.

Laboratory percolation leaching experiments were conducted in glass columns 50 mm in diameter, fitted with steam jackets. Results indicated that uranium extractions of 93 to 94% could be achieved using leaching temperatures in the range of 90–93°C. Pellets remained intact after 35 hours immersion.

Pilot-Plant Experiment

Encouraged by these results, pilot-plant investigations were undertaken. The test equipment designed and constructed for the work is shown in Fig. 2. It consisted of a stainless steel column 0.3 m in diameter and 3.1 m high, capable of holding approximately 230 kg of pellets (containing 21% moisture). The column was fitted with a steam jacket surrounded by asbestos fibre insulation and metal cladding. A double-ended hydraulic cylinder and piston was fitted to the top of the column. A perforated stainless steel plate 0.29 m in diameter was fitted to the lower end of the piston. When actuated by the cylinder, the pressure plate forced down on the contained pellets, giving a load on the pellets equivalent to that calculated to exist at the bottom of a 10.7 m high column of pellets and a total simulated load of 13.1 m of pellets. Compression of the pellets was indicated by a pointer attached to the upper end of the piston. Liquor was pumped through the pellets by a positive displacement pump with a variable speed drive. Pipework was arranged to permit upward and downward percolation, and the liquor delivery line was fitted with a double tube heat exchanger in which steam was used to maintain the liquor temperature in the range 95–100°C.

After loading the pellets and the application of the simulated load, the pellets were subjected to leaching and washing cycles using a number of liquors with varying concentrations of Na_2CO_3 , $NaHCO_3$, $NaOH$, $NaCl$, Na_2SO_4 , and U_3O_8 . The cycles were designed to simulate countercurrent operation in a plant with a number of leaching vats and comprised the following:

- (a) Pre-washing
Upward percolation, temperature between 95 and 100°C.
- (b) Leaching (in 3 stages)
Upward percolation, temperature between 95 and 100°C.
- (c) Washing with barren liquor
Downward percolation at ambient temperature.
- (d) Washing with water
Downward percolation at ambient temperature.

The length of the overall cycle was 43 hours, during which time approximately 2000 litres of solution were passed through the pellets.

During the experiment there were no difficulties associated with pellet break-down. Percolation rates were adequate, effluent liquors contained very little fines, and compaction of the 2.4 m high column of pellets was only 128 mm. At the end of the test the pellets were removed through a gate in the bottom of the column and inspected. Figures 3 and 4 show the condition of the pellets.

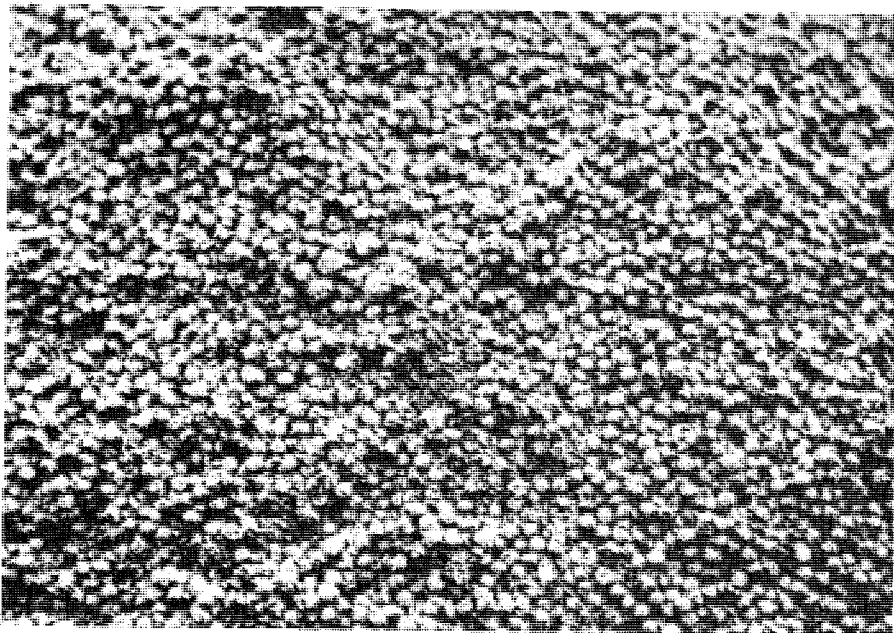


FIG.3. Pellets after leaching.

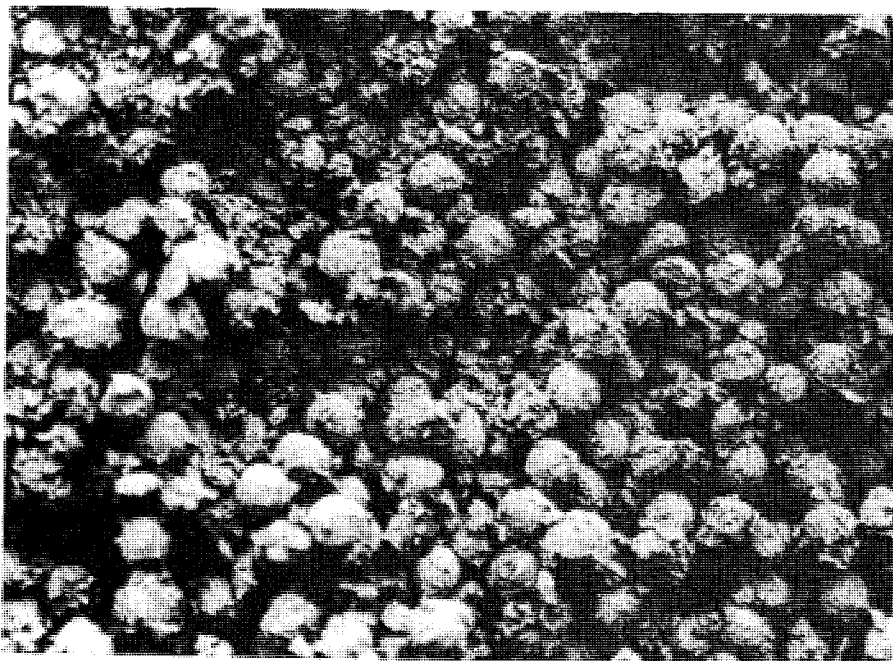


FIG.4. Close-up pellets after leaching.

On removing the pellets it was found, from the difference in the tightness of packing, that the load from the ram had been supported largely by the pellets in the upper part of the column. Although there had been some break-down, as evidenced by the adhering fines, pellets from all levels were substantially intact. However, having in mind the observation of the wall effect, it would be relevant to consider that for large-scale operations there could be advantage in considering a large column packed, in turn, with a nest of smaller-diameter columns, and a suitable distributor to ensure effective liquor distribution throughout the nest. A typical sample of pellets taken from the column had the following characteristics: mean moisture content 25%, mean compressive strength 0.2 kg force, percentage U_3O_8 (no further washing of pellets) 0.013, percentage U_3O_8 (after washing on a Buchner) 0.012. These results indicate a uranium extraction of 95-96%.

Subsequent calculations show that the cycle time of 43 hours used in the investigation was longer than necessary, 30 hours apparently being adequate.

THE BRANNERITE/ZIRCON/DOLOMITE ASSOCIATION - AN INTRACTABLE PROBLEM

The third note in this paper refers to the as yet intractable problem of treating the deposits referred to as 'Skal type', after one occurrence of that name. Anderson's Lode was another of several first-generation uranium discoveries of this type, occurring in the general region of Mt Isa, Queensland. The following is a brief account of results of work on another deposit discovered in the late 1960s, known as Valhalla. Occurring alongside a main highway only 50 km from Mt Isa, it had considerable economic virtue by way of location. A study was made of leaching conditions, and results have been reported in more detail elsewhere (Goldney, L.H., Canning, R.G., Gooden, J.E.A. (1972)).

Mineralogy

The mineralogy of the ore is generally indicative of the problem. The majority of the uranium occurs in a brannerite, which microprobe analysis has shown to contain up to 50% U_3O_8 . A minor but significant amount of uranium is contained in a metamict zircon, the microprobe analysis indicating between 4% and 8% U_3O_8 content. The proportion of these minerals is variable, some core samples being primarily zircon type mineralization. The gangue minerals are predominantly albite with dolomite, sodic pyroxenes and amphiboles, and small to trace amounts of calcite, apatite, brannerite and metamict zircon. The uranium mineralization occurs generally in very fine-grained, intergranular matrix between more granular albite and dolomite.

A composite sample of the drill cores contained 0.16% U_3O_8 , 0.21% Zr, 9.2% CO_2 and 0.78% P_2O_5 .

That these combinations of mineralogy and chemical composition present a formidable problem for economic solution has been borne out by the results of leaching experiments.

Leaching

Leaching with acidities of less than 0.2 moles per litre of sulphuric acid was too slow for any practical consideration - an observation generally in agreement with conditions found elsewhere for leaching brannerites. Figure 5, reproduced from Goldney et al., shows the effect of acid concentration on leaching rate and also the variation of acid consumption with time, when leaching with 0.3 moles per litre sulphuric acid.

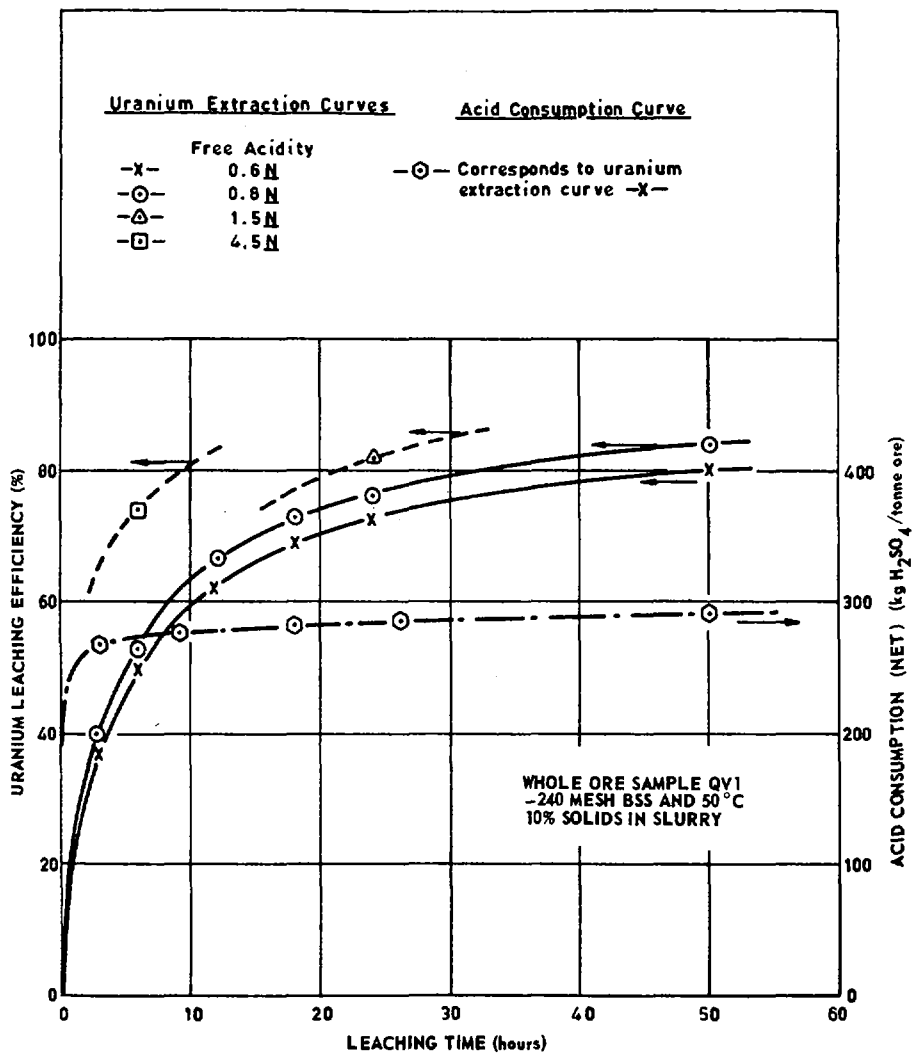


FIG.5. Sulphuric acid leaching of Valhalla ore. Effect of free acidity level on rate of leaching.

The presence of the carbonate at the concentration shown is apparent as the major problem, consuming the bulk of the acid during the initial period of the leach.

The difference between the granularity of the dolomite and the intergranular matrix containing the uranium suggests that physical beneficiation is possible. However, the only technique showing promise is flotation. Using fatty acid as collector for flotation of the dolomite, concentrates have been produced with CO_2 content reduced to 3%. The uranium recovery has varied with the samples examined between approximately 60% and 80%, which indicates appreciable losses before leaching.

The following conditions were technically suitable for leaching the ore:

Grind size	-240 BSS (0.063 mm)
Free acidity maintained while leaching	0.25 moles per litre H_2SO_4
Oxidant	10 kg MnO_2 per tonne of ore
Ferric iron	3 g/l in initial solution
Temperature	50°C
Time	30 hours.

Experimental work showed that a relatively high ferric iron concentration throughout the leaching was advantageous. This effect is thought to be due to the high P_2O_5 content, with a resulting relatively high concentration of PO_4 ions in solution, which depress the activity of ferric ions due to complexing action.

Experiments with Valhalla material indicated that the uranium in the zircon was leached almost as effectively as the brannerite - an unexpected result - but subsequent studies have indicated that this result does not necessarily apply to all materials with similar mineralogy, as some of the metamict zircon may be more refractory.

ACKNOWLEDGEMENTS

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DISCUSSION

H.E. JAMES: I have two questions relating to carnotite deposits. First, you described a pelletizing and percolation leaching technique. How were the pellets prepared — was there any crushing and milling or size separation done before you actually produced pellets? The point of my question is that usually, in this type of deposit, there is a sort of barren grit which does not contain uranium; the uranium tends to be in the material around the grit. I thought that one possible approach would be to remove a very fine slime fraction, to pelletize and at the same time to upgrade the feed. The other question relates to whether you considered solution mining for this particular deposit as a possible approach?

F.R. HARTLEY: We made a preliminary size distribution study on the ore. There is a tendency for differential distribution of the uranium but it was considered, in consultation with the particular company, that size separation would be inadequate — it certainly did not appear to improve greatly the settling characteristics of the remaining ore and so the whole ore was treated; it is the whole-ore treatment which is described in the paper.

As regards your second question, solution mining has been considered. The deposit is very extensive, in fact it is probably the second largest now of our uranium provinces in the country, and I believe that solution mining would not be particularly satisfactory.

K.M.V. JAYARAM: We have copper deposits associated with uranium and we tried to apply gravity separation. Originally it was all right but of late, because of the price variation for copper, the cut-off grades for copper changed so much that uranium recovery by gravity separation became difficult. Also, a considerable amount of uranium is lost with the slimes. Do you think that pelletizing of these tailings and subsequent percolation leaching will help?

F.R. HARTLEY: It depends. In your case, of course, the problem is the cost of dewatering the slimes produced. I presume that you actually apply desliming and your question is whether this process can be applied to a specific fraction. I cannot give you an answer to this. We have tried this type of technique in several copper operations and in this uranium operation. The nature of the clay is important, its base exchange properties; you could only try. If you actually apply wet desliming, you are faced with the necessity of dewatering and drying back to about 20–21% moisture to make effective pellets.

K.M.V. JAYARAM: The problem is that the feed would be of the order of 5000 tonnes per day when all the copper plants go into operation and the low-grade copper tailings are of different sizes. Uranium appears to be mostly associated with the finer sizes. So instead of trying to segregate either on the size basis or on the gravity basis, could we not pelletize the tailings as such and use percolation leaching?

F.R. HARTLEY: I would suggest that it should be tried — it could be a very satisfactory procedure but there is no way of predicting without trying it.

S. AJURIA-GARZA: Regarding solvent extraction of high-grade pregnant liquors, what amine concentration are you using in the organic solvent?

F.R. HARTLEY: This is a standard amine assessing 0.1 molar amine in the kerosene solution — nothing different from the normal except the procedure.

S. AJURIA-GARZA: We have a somewhat similar programme. We feed 7–9 grams per litre to our solvent extraction system but the amine concentration used is higher than normal.

F.C. LENDRUM: I was interested in your discussion of radiation dose rates at the plant. In Canada, at the old Port Radium plant, we had this type of ore. A study is being made in Canada now on the health of those who worked at Port Radium between 1933 and 1950.

F.R. HARTLEY: This problem is crucial in the design of the operation. The vigilance required now on this sort of operation — quite apart from any moral obligations — is so tight that any information at all that we could get would be very valuable.

T.S. ARY: What is the vanadium content of carnotite ore?

F.R. HARTLEY: I could not give you the figure off-hand. It is relatively low and it has not been considered at all in the economic studies.

R.H. KENNEDY: In the past, we have of course received large quantities of high-grade pitchblende type concentrates from both the Congo and from the Great Bear Lake region in Canada. These were shipped directly to our refining plant where they were processed by nitric acid digestion followed by TBP extraction and water stripping of the TBP. Of course, this produces directly a nuclear-grade uranium trioxide which can then be processed. Although the amine system has some advantages it would seem to me that to try the TBP route would be very worthwhile in connection with these extremely high-grade ore products. One of the advantages of TBP is that it can handle directly the slurry that results from the digestion of ore with nitric acid whereas the amine type solvent requires a much clearer solution to work from.

F.R. HARTLEY: The feasibility studies associated with the development of these particular deposits have included considerations of refinery installations at various places within the country and indeed also some consideration of markets outside the country.

TREATMENT OF AN ISOLATED HIGH-GRADE, LOW-TONNAGE URANIUM OREBODY

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Abstract

TREATMENT OF AN ISOLATED HIGH-GRADE, LOW-TONNAGE URANIUM OREBODY.

A completely integrated process leading to a commercial-grade uranium concentrate has been developed for the El Nopal orebody which contains 115 000 t of ore with an average grade of 0.283%, equivalent to 325 t of U_3O_8 . The process consists of crushing (from -12 in to -1.5 in), heap leaching with recirculation, a special type of countercurrent washing (also in the heap), solvent extraction to give an exceptionally high uranium concentration, re-extraction, precipitation with ammonium hydroxide, filtration and calcining. The main factors that influence heap leaching are analysed in detail: heap geometry, crushed ore size and size distribution, base design and construction, method of heaping, acid feeding method, flow of liquors through the heap and washing procedures. Leaching efficiencies range from 80 to 85% and washing efficiencies from 96 to almost 100% with an overall extraction efficiency of 77 to 85%. Acid consumption is usually less than 25 kg/t of ore. The leaching and washing processes described are designed to use an overall solid-to-liquid ratio of about 3:1 thus producing very concentrated liquors with a U_3O_8 content of about 7 g/l. These liquors are filtered and fed directly to a solvent extraction system using Alamine 336 (tri-capryl amine) and isodecanol in kerosene. The high feed concentration makes it possible to use a high concentration of amine. The organic solvent contains 120 g/l of Alamine and 98 g/l of isodecanol. Re-extraction can be with a saline solution containing 100 g/l NH_4Cl and 250 g/l $(NH_4)SO_4$. The strong liquor obtained from re-extraction contains 70-75 g/l U_3O_8 . The remaining steps of precipitation, filtration and calcining can then follow general practice and lead to a product which meets currently accepted commercial specifications. As an alternative, nitrate re-extraction has been shown to produce a solution of about 70 g/l which will feed directly to tributyl phosphate (TBP) refining without the necessity of ever producing a concentrate.

1. INTRODUCTION

A process with some novel features has been developed specifically to treat the El Nopal ore. The El Nopal orebody is located about 40 miles north-east of the city of Chihuahua in Northern Mexico. It contains about 300 t of U_3O_8 with an average grade of about 0.28%. Being quite permeable, the ore responds well to percolation leaching, as it does also to conventional acid leaching. However, it was calculated that a process based upon percolation leaching with an efficiency of, say, 70% would be substantially cheaper than one based upon conventional leaching at 90% efficiency.

Percolation leaching of low-grade uranium ore (below 0.1%) is currently in use in several countries: the United States of America, France, Spain, Portugal, Argentina and possibly others. As far as is known, the El Nopal process would be the first to use percolation leaching for an ore of normal grade (between 0.1% and 0.3%) and therefore new emphasis had to be placed upon some of the factors involved, such as leaching efficiency, washing efficiency, uranium concentration in the product liquor and absence of liquor losses. Since a highly efficient recovery of uranium proved possible with a combined leach and wash liquor at a concentration of about 7 g/l, an

amine extraction process was developed, using a high amine concentration, which gave a high product concentration. Throughout this paper, all concentrations described as uranium refer to U_3O_8 .

2. LEACHING AND WASHING

2.1. Experiments in 1 m columns

A large number of leaching experiments with 11 kg samples were carried out in 1 m by 10 cm glass columns with a standard representative sample of ore containing 0.239% U_3O_8 . They showed the optimum leaching conditions to be as follows: particle size up to 1.5 in, sulphuric acid concentration between 25 and 50 g/l, and recirculation of the acid at a flow rate of about 100 l/h · m² for up to 1000 hours. Oxidants had no detectable effect. The efficiencies were a few per cent lower at 0°C. Using recirculation, the optimum leaching efficiencies were about 80 to 85% and the uranium concentrations in the liquor were between 7 and 11 g/l. Leaching for 100 hours instead of 1000 hours reduced the efficiency by about 2%. Impurity contents of the leach liquor were relatively low. The volume of leach liquor in the recirculating system could be reduced to the minimum practicable without affecting efficiency but it was important to maintain the free acid above 25 g/l.

Washing with water was as effective as washing with dilute acid. Recirculation during washing would not, of course, remove the uranium efficiently and a very large volume of wash water was used in a single pass in these early experiments to recover the uranium completely. It was noticed that an interruption of washing led to an increase in the uranium concentration in the next portion of wash liquor. This led to the idea that a longer time was needed for the rich liquor coating or impregnating the ore particles to mix with or diffuse into the wash water. A reduced flow rate or intervals with no flow should therefore be beneficial.

2.2. Experiments on the 500 kg and 900 kg scale

In the 500 kg experiments the ore was leached and washed in wooden boxes of 60 cm square section by 1 m in height. It was assumed that the increased area would eliminate any 'wall effect' of a narrow column. Irrigation was by means of a perforated flexible PVC tube laid over the surface of the ore.

The first five experiments were with the optimized leaching conditions established earlier, but for only 110 hours. A large volume of wash water was used, and the combined leach and wash liquor had a final uranium concentration of 1 to 2 g/l. The only variable was the flow rate. In the five successive experiments, flows of 166, 166, 139, 111 and 111 l/h · m² gave leaching efficiencies of 72.7, 79.7, 78.8, 78.8 and 78.5% respectively, based upon analysis of the solid residues. Except for the first anomalous result, the leaching efficiencies were quite close to those obtained in the glass columns after only 100 hours.

A representative 11 kg sample of the residue from the last experiment was leached for an additional 36 hours in a glass column under comparable conditions. The additional 3.4% efficiency (for a total of 81.9%) was presumably due to redistribution of the ore particles and to the difference in scale. Conventional acid leaching of the twice leached residue, after grinding to below 100 mesh, only gave an additional 0.5% yield.

The next six 500 kg experiments allowed a comparison to be made of three recirculation leaching flow rates with and without periods of interruption, but all continued for about 1000 hours, as shown in Table I. Optimum conditions were used, with temperature fluctuations between 10°C and 20°C. This table shows clearly that the highest leaching efficiency is obtained by using the highest flow rate with a continuous flow, as in experiment 6. The loss in efficiency

TABLE I. LEACHING RESULTS ON 500 kg SCALE

Experiment	Flow rate (l/h · m ²)	Cycle	Efficiency ^a (%)	Acid consumed (kg/t)	Total acid used (kg/t)
6	127	A	88.0	19.0	29.0
7	54	A	84.8	17.8	24.6
8	18	A	85.9	18.6	24.6
9	127	B	82.6	18.6	27.0
10	54	B	82.6	18.2	25.0
11	18	B	81.8	17.2	22.8

^a Calculated from solid residues.

A – Leach liquors flowing continuously for 1000 hours.

B – Four cycles of 2 days' leaching and 6 days without leaching.

TABLE II. WASHING RESULTS ON 500 kg SCALE

Experiment	Flow rate (l/h · m ²)	Cycle	Efficiency ^a (%)	Uranium concentration ^b (g/l)	Uranium concentration ^c (g/l)
6	100	A	99.3	2.8	0.05
7	25	B	96.1	2.4	0.04
8	25	C	93.1	2.5	0.04
9	125	D	79.7 (95.3)	1	1 (0.37)
10	54	E	90.0	1	0.23
11	18	F	82.7	0.87	0.075

^a Per cent of dissolved uranium recovered.

^b Concentration in mixed leach and wash liquors.

^c Concentration in last liquor to leave column.

A – 0.5 h wash, 7.5 h no flow, 8 d total.

B – $\frac{1}{2}$ h wash, $1\frac{1}{2}$ h no flow, 8 d total.

C – 12 h wash, 36 h no flow, 8 d total.

D – 12 h continuous (+second 12 h).

E – 48 h continuous.

F – 8 d continuous.

when the flow is interrupted for long periods is not justified by the reduced cost of pumping. It is interesting to note that all the efficiencies with interruption of flow are intermediate between all those with no interruption at 110 hours and 1000 hours. This corresponds with the fact that the periods of actual flow totalled about 250 hours. Acid consumptions were all similar, at about 18 kg/t, which compares favourably with about 60 kg/t for conventional leaching of this ore.

The washing results of these six experiments are shown in Table II. This table demonstrates clearly the superiority of washing with a high flow rate for successive short periods of time with long interruptions, as in experiment 6. All results with flow interruption are better than all those without. The fairly high uranium concentration (2.8 g/l) in the combined leach and wash liquors of experiment 6, combined with the high wash efficiency, provided grounds for the belief that a countercurrent washing process with flow interruption might be highly effective.

The 900 kg experiments were carried out in rubber-lined steel tanks at flow rates of 400 l/h·m². In one experiment, 816 hours of continuous leaching gave a leaching efficiency of 83.2%. In the other experiment the ore was leached for 1008 hours, but with interruptions, in regular cycles lasting 75% of the time. Leach solution was actually flowing for 264 hours giving an efficiency of 78.7%. This difference again confirmed the results that continuous flow was justified during leaching (but not during washing).

2.3. Experiments on the multi-ton scale

Five large-scale experiments were performed to study the leaching process and the mechanical construction of the heaps. The dissolved uranium was always washed out with an excess of water and the washing process itself was not optimized. The object of the first two experiments was mainly to determine the best method of constructing large heaps and therefore the leaching results were not typical.

The third experiment, with 10 t of ore, gave a good leaching efficiency of 71%, considering the very short leaching time of 64 hours.

The fourth experiment, with 28.5 t of ore and a grade of 0.204%, gave quite a high efficiency of 78% in 492 hours, even though a high proportion of the ore leached was in the shape of a regular slope. The method of construction, using a concrete base (and walls on the pilot scale) with PVC sheet bonded to it by bitumen, was cheap and very satisfactory. The temperature range of 1–30°C was typical of the El Nopal location.

The fifth experiment, with 72.5 t of ore in a tank, was leached for 1520 hours with leach liquor recirculated for 25% of the time, the flow rate being increased in stages from 12 to 100 l/h·m². The gradient of the graph of efficiency against time increased at each stage, showing that the highest flow rate was best. This was assumed to be equivalent to 380 hours continuous leaching, and the time was then increased by further continuous leaching at 100 l/h·m² to a total of 1000 hours.

During leaching of the 72.5 t heap, several arrangements of perforated hose and sprays were used to obtain good liquor distribution above the heap and, for 1000 hours, it is believed that any loss of efficiency owing to maldistribution of the liquor would be small.

A possibly more serious defect was noticed towards the end of leaching, by digging into the top surface of the heap. A single channel appeared to have been created which took a high proportion of the liquor flow. This maldistribution of liquor was confirmed by later studies in large wooden boxes. It is believed that the problem can be alleviated by removing the fines and treating them in a conventional leaching plant to be built later.

2.4. Complex leaching and countercurrent washing experiment

Complex experiments involving a number of columns were needed to simulate a four-stage, countercurrent washing system. Table II shows that washing is most efficient with a number of cycles, each consisting of a short washing period followed by a long period without flow.

Fifteen 6 m high columns, 10.6 cm in diameter, were erected and each was filled to a height of 4.5 m with 55 kg of typical crushed ore of grade 0.232%.

All 15 columns were leached together, at $70 \text{ l/h} \cdot \text{m}^2$, for 672 hours. By analysis of the washed leached ore residues the leaching efficiencies varied from 76.1% to 87.2%, with an average of 82.9%, and the uranium concentration of the leach liquor was 8.43 g/l.

A true four-stage countercurrent washing system was used in this experiment, gradually building up to equilibrium. The various washing routes in sequence were as follows:

Columns 1, 2, 3, 4	Columns 9, 10, 11, 12	
Columns 2, 3, 4, 5	Columns 10, 11, 12, 13	
Columns 3, 4, 5, 6	Columns 11, 12, 13, 14	
Columns 4, 5, 6, 7	Columns 12, 13, 14, 15	
Columns 5, 6, 7, 8	Columns 13, 14, 15	} End of system only
Columns 6, 7, 8, 9	Columns 14, 15	
Columns 7, 8, 9, 10	Column 15	
Columns 8, 9, 10, 11		

Thus, from 15 columns, 12 solutions were available which had passed through the full set of 4 columns. Each of these was mixed with one fifteenth of the total volume of leach liquor from the fifteen columns, to represent the final product liquor from one column (although actually each liquor represented part of the uranium from four identical columns).

Each of the wash solutions had been applied to each column in its route in the form of 36 portions. Each portion was applied over $\frac{1}{2}$ hour to the first column in its route and the liquor passed immediately to the next column and so on, down the series of four columns. Each half-hour period of flow at $100 \text{ l/h} \cdot \text{m}^2$ was followed by 7.5 hours without flow. Thus each washing route required 288 hours (i.e. 12 days). As expected, the concentrations of uranium leaving the columns were rather uniform. These were, in sequence, 8.49, 9.33, 11.66, 9.83, 10.33, 10.66, 10.33, 10.83, 10.70, 9.33, 8.08 and 7.95 g/l. The twelfth liquor should be regarded as fairly typical of the equilibrium condition.

From all fifteen liquors, 84.4% of the total uranium in the system was recovered at 8.44 g/l, which is nominally slightly more than the amount leached, so the washing efficiency is presumably near to 100%. The countercurrent washing system was therefore fully justified.

3. AMINE EXTRACTION

3.1. Design of the system

Several solutions of Alamine 336 (tri-capryl amine) were made up, of different concentrations. In each case isodecanol was added to the extent of 67% of the amine concentration. The distribution coefficients of uranium were then determined between each of these treatments, with typical leach liquors at pH 0.7, 1.0 and 1.7. The results obtained at pH 0.7 were a little higher than those at the other pH values and the equilibrium lines obtained are shown in Fig.1. It is clear from the graphs that an amine concentration as high as 120 g/l in kerosene is beneficial for extraction of uranium from a leach liquor of a concentration of about 7 g/l. An equilibrium diagram is shown in Fig.2 for this amine concentration and uranium feed concentration (and an alternative of 2 g/l). A washing section is included. This shows that virtually complete extraction of the uranium will take place in four extraction stages and that three stages of washing with dilute sulphuric acid can be allowed. The flow ratios required are 20 : 7 : 2.5, for leach liquor, solvent and wash solution respectively. The concentration of uranium in the solvent will be 20 g/l from a 7 g/l feed (or 15 g/l from a 2 g/l feed). The high ratio of aqueous to solvent phases with the 2 g/l feed (8 : 1) in the extraction section is probably on the border of practicality.

This high-amine concentration flowsheet is suitable for the proposed El Nopal beneficiation factory because of the high uranium concentration and low concentration of impurities in the leach

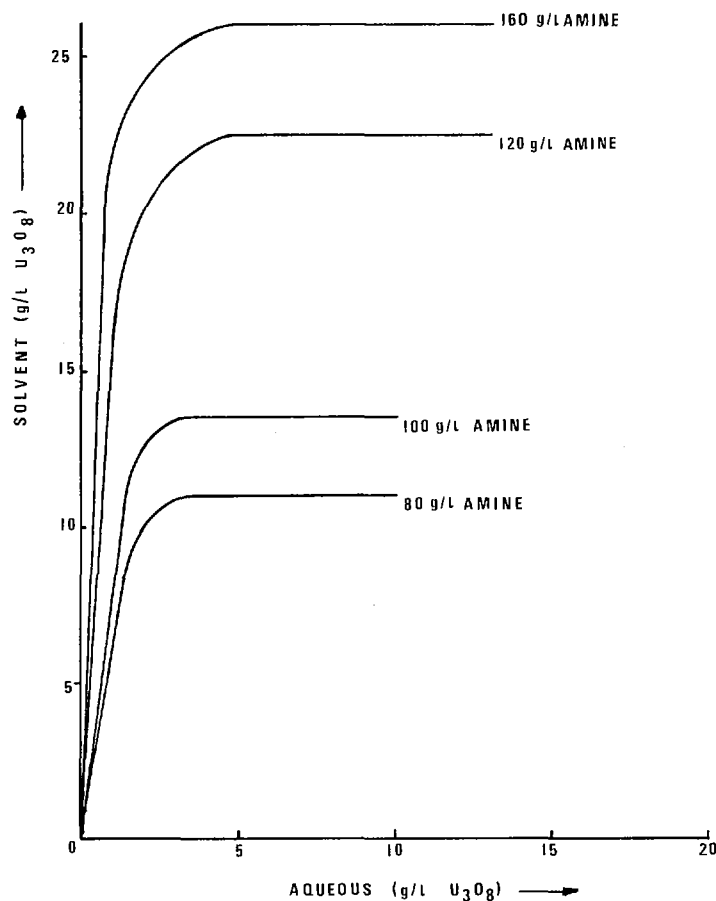


FIG.1. Uranium distribution coefficients at pH 0.7.

liquor. It may not be suitable for other ores which cannot give a high uranium concentration in the leach liquor and where the impurity contents may be greater.

Re-extraction of the uranium from the solvent into an aqueous phase can be by the use of ammonium chloride solution if it is decided to produce a normal 70% uranium concentrate by subsequent precipitation, filtration, washing and drying. Distribution coefficients determined by successive re-extractions on a laboratory scale gave the graphs shown in Fig.3. An ammonium chloride concentration of 80 or 100 g/l is clearly very suitable for complete re-extraction. The equilibrium diagram is constructed for 80 g/l and it shows that in only two or three stages a uranium concentration of 70 g/l can be obtained in the aqueous phase from 20 g/l in the solvent. If the uranium concentration in the solvent was only 15 g/l (from leach liquor at 2 g/l), then the re-extraction would be to a uranium concentration of 52 g/l.

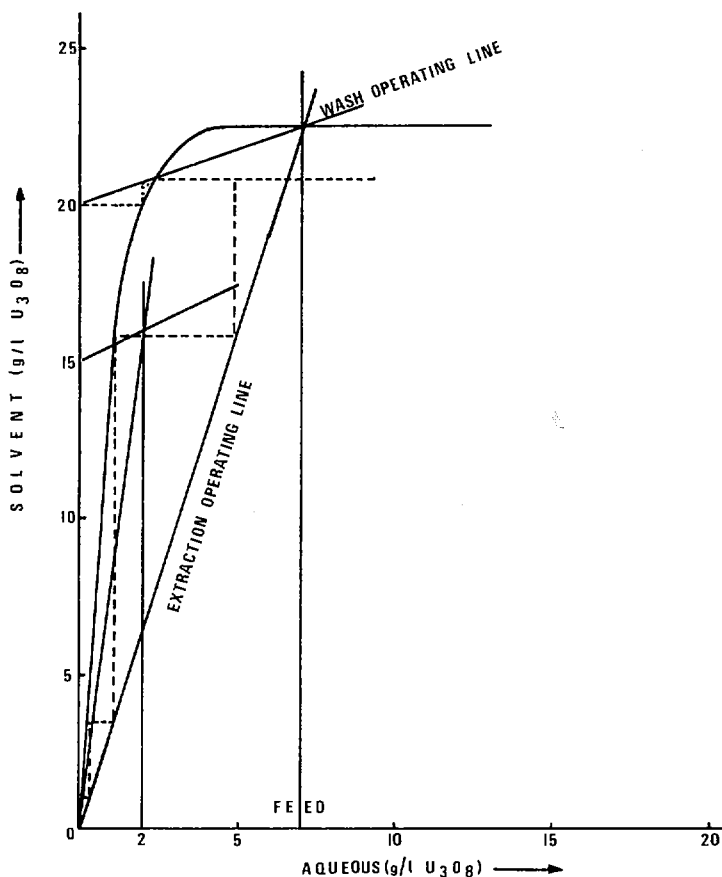


FIG.2. Equilibrium diagram for uranium extraction.

It might be possible to re-extract directly with ammonium nitrate, or to use a sulphate system with careful pH control. In view of the high uranium concentration this might then allow the uranium to be purified by direct extraction with 20% or 30% tributyl phosphate (TBP) to give a nuclear-grade product without the necessity of ever producing a concentrate.

The required control of pH could not easily be obtained with the sulphate system, so re-extraction with ammonium nitrate solution was next examined. The equilibrium diagram for an ammonium nitrate concentration of 100 g/l is shown in Fig.4. This indicates that, with about five stages for complete re-extraction, a nitrate re-extraction solution containing 50 g/l of uranium can be obtained from a solvent containing 20 g/l uranium.

A few distribution coefficients are available for the direct extraction into 30% TBP of the uranium from a solution produced by ammonium nitrate re-extraction of the amine after

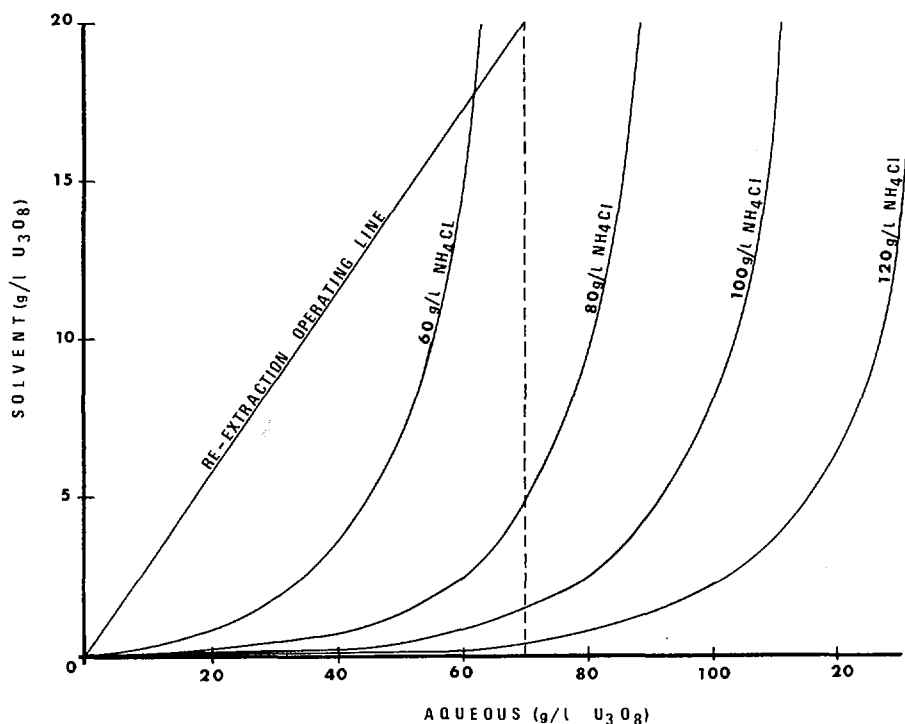


FIG.3. Equilibrium diagram for uranium re-extraction with ammonium chloride.

acidification to 3N with nitric acid. These are given in Fig.5 which shows an equilibrium diagram for a feed of 50 g/l uranium. With six or seven stages, the uranium can be completely extracted up to a concentration of about 77 g/l in the solvent. This is a standard system including washing with a small amount of the recycled pure uranyl nitrate solution (at 25 g/l uranium). The ratio of the flow rates of aqueous feed to solvent to wash solution is 6.5 : 5 : 2.5.

A TBP re-extraction process, using a solvent-to-aqueous ratio of about one, is known to remove the pure uranium completely into the aqueous phase, at a concentration of about 77 g/l. This would be of nuclear grade, but it could be precipitated, filtered, washed and calcined to the pure oxide in the same way as a crude concentrate.

3.2. Pilot plant operation of the amine extraction system

An experiment was performed with a set of mixer-settlers, following several preliminary trials where the equipment was not ideal.

Four stages of extraction were used with a feed varying between 6.5 and 7 g/l in uranium content and an amine concentration of 140 g/l, together with a single water washing stage. The flow rates were 8.1 l/h of feed, 2.7 l/h of solvent and 2.7 l/h of wash water. The wash water passed through the extraction stages to produce only a single raffinate. The extraction efficiency was

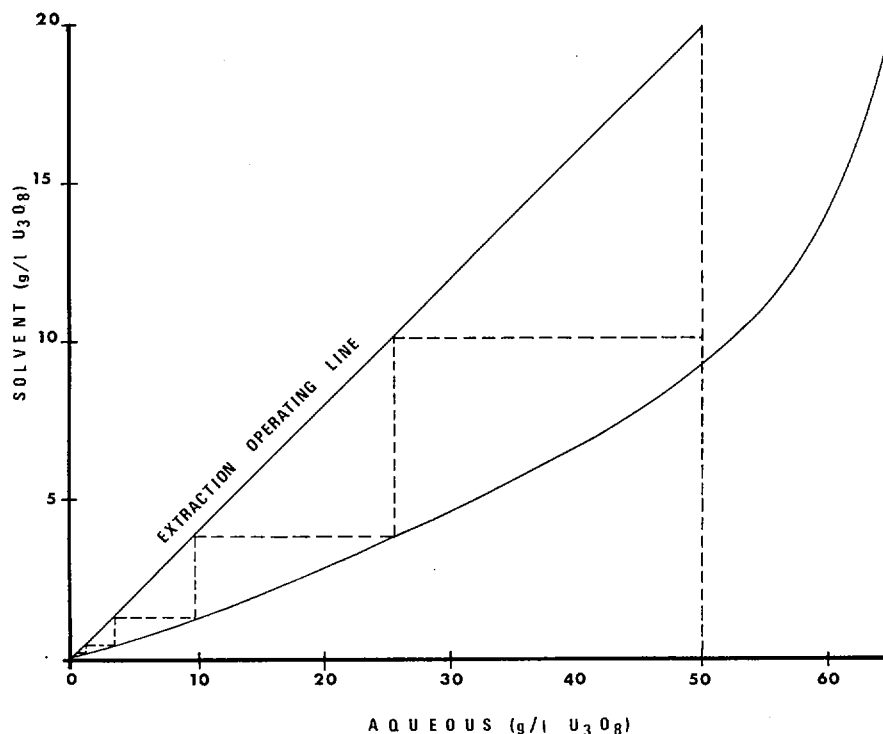


FIG. 4. Equilibrium diagram for uranium re-extraction with ammonium nitrate.

consistently above 99.8% and the concentration of uranium in the solvent was between 19 and 21 g/l, as expected. Re-extraction was with a solution containing 100 g/l ammonium chloride and 250 g/l ammonium sulphate, at a flow rate of 0.7 l/h, using three mixer-settler stages. The re-extraction efficiency was over 99.7% and the concentration of uranium in the re-extraction liquor was between 70 and 74 g/l. The barren organic solution was recycled continuously back to the extraction section without treatment, but its high chloride content of 10 to 12 g/l apparently had no detectable effect upon the extraction of uranium.

4. CONCLUSIONS

The leaching experiments on scales of from 11 kg to 72.5 t demonstrated clearly that a leaching time of about 1000 hours is necessary, with recirculation and a free acid concentration between 25 and 50 g/l. Oxidant did not improve the leaching efficiency, and interruption of liquor flow caused a reduction of efficiency being about equivalent to the time lost.

Sloping heaps were leached almost as effectively as horizontal heaps, which will allow the production heap to be free-standing without any retaining walls. Leaks were prevented by the use of a concrete base to which PVC sheet was bonded with bitumen. Even irrigation at the top

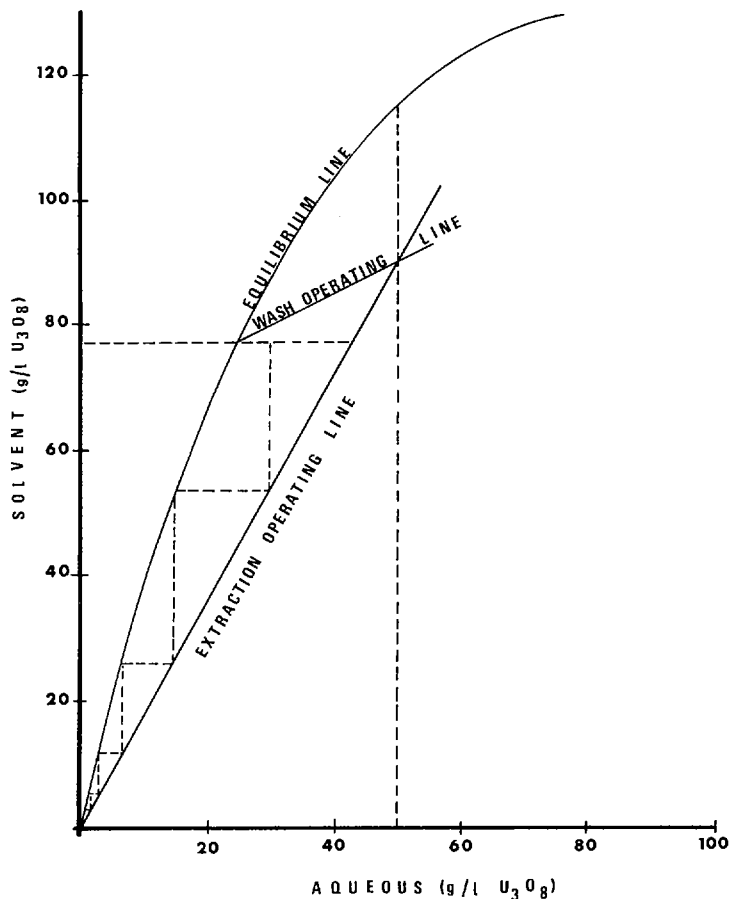


FIG.5. Equilibrium diagram for TBP extraction after amine re-extraction with ammonium nitrate.

of the heap is desirable and practicable. Some loss of efficiency may occur as a result of poor distribution of liquor within an extensive heap unless this can be improved by the removal of fines for processing in a conventional leach plant to be built later. The efficiencies did not appear to be affected by differences in vertical height of the heap of between 1 m and 5 m.

A very effective four-stage, countercurrent washing process was developed in which a number of washing cycles were used, involving a high flow for a short time followed by no flow for a longer time. Leaching efficiencies of about 84% were obtained in 5-m column experiments, combined with countercurrent wash efficiencies of 98% and the recovery of all the uranium at a high concentration of over 7 g/l. There is considerable evidence that the overall recovery efficiency of 82% will not fall as low as the design figure of 70% on the production scale.

A novel amine extraction process has been developed, using a high amine concentration, to match the high uranium concentration of 7 g/l in the leach liquor. This results in a uranium

concentration of about 20 g/l in the solvent and 70 g/l in a chloride re-extraction liquor. These figures have been confirmed in continuous mixer-settler trials, with extraction and re-extraction efficiencies both exceeding 99.7%.

An alternative re-extraction procedure using a nitrate system appears promising. This gives a re-extraction solution that is only slightly lower in uranium concentration but allows direct extraction with TBP to give a nuclear-grade product without the necessity of ever producing a concentrate.

ACKNOWLEDGEMENTS

The experimental work was performed at the Villa Aldama Pilot Plant in Chihuahua under the direct supervision of Salvador Patiño (now at the University of Guanajuato) and Benjamín Blanco. Arturo Navarro and Arturo Olivero were responsible for the pilot plant experiments, José González and Sonia Sánchez for the analytical work.

DISCUSSION

K.M.V. JAYARAM: The feed grade of about 0.28% U_3O_8 is fairly high. Do you not think that, even with 90% extraction by heap leaching, the tailings might constitute an ore grade of the future?

S. AJURIA-GARZA: Yes, this is correct. Our problem is one of time. We are in dire need of uranium concentrates — as a matter of fact, we needed them yesterday. We have already purchased two nuclear reactors and we have to supply enough uranium for the first cores in a very short time. This plant was originally conceived and engineered three years ago under very different conditions. At that time the current price of uranium was about \$5 per pound and this orebody was all we had. We are fully aware of the new conditions and we expect that the residue left after our first operation can be recovered in later operations. We believe that the project is fully justified under the prevailing economic and political conditions but we are by no means prepared to let all that uranium go to waste. Most certainly it will be processed in a new plant which is now being engineered.

K.M.V. JAYARAM: I would like to know whether any combination of processes has been tried. For instance, with ore of this particular type, could you separate it into a very low-grade ore for heap leaching and a very high-grade one for conventional leaching?

S. AJURIA-GARZA: No, that cannot be easily done. The orebody is of hydrothermal origin! It is a chimney, about 100 m high and about 25 m in diameter, and there is a very marked gradation of ore grade from the bottom to the top. At the very bottom of the chimney we have a grade of only about 500 g U_3O_8 per tonne and at the top we have about 10–15 kg. We took a cut-off point at about 55 m, roughly halfway from the bottom of the chimney, and we are going to process the top part as described. The bottom part probably will be processed by in-situ leaching. We are now doing experiments on a process for the lower part. This is rather refractory. We have also divided the main body from the halfway point to the surface into six main blocks. If we can proceed very quickly with the new plant design, which will be a conventional agitated leach plant, we may be able to reserve some of the higher-grade pockets for conventional processing. This is a fairly complex situation. The real problem is that this is not a purely technical affair; it also has political and economic implications which we cannot overlook. If we were dealing with a purely technical problem, it would be easier.

R.J. KLOTZBACH: I think that the high recoveries obtained in the laboratory will never be reached in the actual plant. Especially if you have a free-standing heap, you must have a barren matrix to support it and sides to prevent breakthrough. There is a section of the heap

that never gets in contact with the acid so the overall recovery will certainly be lower. In your experiments you used plastic sheets. When we build big heaps, we always use clay because we can run heavy equipment on it and still have an impervious liner. With a uranium price of US \$25/lb, I would not put a high-grade ore on a heap leach. We are trying a thin leach, that is, we fix with acid and spread it out in real thin layers and then wash it. With this method we are almost sure that we can get 90% recoveries.

S. AJURIA-GARZA: We are aware of this; thank you for your comments. The plant efficiency has been taken at 75%, which is approximately 10% lower than the efficiency from our laboratory and pilot plant work. We are taking into account a reduction in efficiency owing to the increased scale. As for heavy equipment, we do not plan to run any over the heap. The ore will be piled up using an overhead crane. We are also searching for different materials to use as a heap base. As of now, a combination of concrete, bitumen and plastic has been shown to be quite satisfactory and the losses of leach liquor have been minimal. Regarding the use of heap leaching for a high-grade ore, we have had strong and heated debates over this topic at our own headquarters. We have decided to go ahead in the way it was originally planned because of the urgent need to provide concentrates to feed our reactors. This plant was designed as a solution for a problem posed in 1972. Now, in 1975, the problem is different. But we have already gone too far down the road; we are committed. It is a take-it-or-leave-it proposition: either we go ahead and do this as planned or forget about it. If we shelve this project we shall not have any domestic uranium concentrates until about 1980 and this could jeopardize our nuclear programme as a whole.

F.R. HARTLEY: I would like to ask whether you have any figures on solvent loss, particularly the amine loss from the high amine solvent?

S. AJURIA-GARZA: I do not have any figures with me now, but they are not particularly high. We are studying this very carefully and we have developed an analytical procedure whereby we take the raffinate and make a solvent extraction with ether. Then we collect a fraction and analyse it by gas chromatography. This is a rather precise procedure that gives us quick information. The other way is to run a mass balance over an extended period of time, but the problem with a mass balance is that you have to wait for a long time to find out that you have lost some material. With the procedure we are now using we can tell at once, with fair certainty, where our losses are.

ENVIRONMENTAL PROBLEMS POSED BY WASTES FROM THE URANIUM MILLING INDUSTRY

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Abstract

ENVIRONMENTAL PROBLEMS POSED BY WASTES FROM THE URANIUM MILLING INDUSTRY.

The processing of uranium ores has many effects on the environment. These effects can be caused by outside influences as well as by the actual milling operations. The major impacts associated with milling are connected with the waste products produced from uranium extraction. Solid, liquid and gaseous wastes arise during processing. The environmental problems posed by wastes from uranium milling are receiving attention by the industry and by national authorities. Air pollution from mills is not a serious problem, water pollution is potentially a greater problem; but methods are available to help us overcome this problem. The problems posed by the disposal of solid tailings have not been completely resolved. Some of these problems are discussed in the paper.

1. INTRODUCTION

The processing of uranium ores has many effects on the environment, as have all other metal extraction operations. These effects can be caused by outside influences and the provision of transport and other services as well as by the actual milling operations. The major impacts associated with the latter are in connection with the wastes produced at uranium extraction.

Solid, liquid and gaseous wastes arising during uranium processing pose the normal problems associated with all ore processing wastes and, additionally, health problems owing to the radiotoxicity of the various radionuclides occurring in uranium ores. The radiotoxicity of these radionuclides is due to their potential for producing injury arising from their emitted radiation when such radionuclides are incorporated into the body or externally irradiate it.

The wastes originating during the processing of uranium ores and the treatment they are given before discharge are described in an Agency publication [1].

2. ENVIRONMENTAL EFFECTS OF AIRBORNE WASTES

Various contaminants are released to the air at various stages of uranium mining and milling, but those having the greatest potential for environmental impacts are the dusts from tailings disposal areas, especially in dry climates, and the radon that is released at all stages.

Dusts are a health hazard to man and can also smother vegetation and, especially when they come from tailings piles, can spread radioactive contamination over a large area.

The radon that is released in effluents from mills does not appear to be significant, but that released from large tailings piles can pose a significant hazard. This problem is further discussed in Section 4.

3. ENVIRONMENTAL EFFECTS OF LIQUID EFFLUENTS

The potential effects on man and his environment from liquid effluents may range from minimal effects for mines and mills operating on an essentially closed circuit, as in dry climates,

to substantial effects for those which discharge large amounts of insufficiently treated water to natural waterways [2, 3]. Where liquid effluents are wholly discharged to the waste retention system, the effects are due to the substances dissolved in, and carried by, the final outfall water. Such substances may have been originally present in the mine/mill discharge or, less predictably, may arise from solubilization of solid wastes.

Radium-226 and its long-lived daughters, which are discharged from operational tailings ponds through overflow or seepage or leached from abandoned tailings, do constitute a potential risk to members of population groups who may use the contaminated water. Surveillance is indicated and appropriate remedial measures should be taken when necessary.

In the case of acid leaching, the use of solvent extraction is tending to come into general use. In this case the effluents contain organic products, either in solution or in emulsion, such as amine solvents, secondary alcohols, tributyl phosphate and kerosene, in addition to other dissolved mineral elements.

When liquid effluents are discharged to the environment, adequate treatment may be required to ensure that there are no long-term adverse effects. When treatment is absent or grossly inadequate, the effects may be immediately evident by a change in colour or the turbidity of the receiving water. Less evident may be the effects on aquatic life, fish in particular. There may be outright fish kills or more subtle but deleterious effects on reproduction, size and growth rate and species diversification. The potential harm of relatively low levels of the heavy metals, organic compounds and ammonia is well documented. The effects may be seen directly where there is commercial fishing. In areas of recreational value, this may be destroyed or rendered unattractive if angling is poor and the waters uninviting.

The radionuclides naturally associated with uranium and thorium are potential sources of contamination. The isotope radium-226 has received greatest attention because of its long half-life of 1620 years and the potential harm from its alpha radiation after ingestion, as it is incorporated in the bone structure. Specific steps are taken to remove this radioisotope from liquid wastes.

The safe disposal of radioactive wastes into rivers, lakes and estuaries is the subject of an IAEA Safety Series publication [4]. The necessity of controlling releases of radioactivity from the uranium industry is just as cogent as for any other source of radiation.

The need for close control of deleterious impacts on the environment has encouraged the development of improved practices. As any degradation of the environment affects man's well-being and ultimately his health, it is essential that the best technology be followed and that the necessary research and development be pursued.

4. ENVIRONMENTAL EFFECTS OF SOLID WASTES

4.1. Mill tailings

The tailings release radioactive material to the air as radon gas and airborne particulates, to waterways as particulates that may settle out in certain locations, and as radionuclides that may be leached out by precipitation, surface run-off and the waste solutions. The tailings are sufficiently radioactive to create a weak field of gamma radiation in their immediate vicinity. Chemical pollutants including various heavy metals, sulphates and sulphuric acid may also be released from them.

Dust is referred to in Section 2. Wind and water erosion of tailings heaps depends on the physical properties of the waste and the climate of the area.

The sulphide tailings released from some uranium mills will produce acid seepage and oxidation of the surface layer. This surface oxidation will hinder the development of vegetation.

The decay of radon gas within the tailings and the atmospheric dilution of radionuclides released are usually sufficient to keep the radon concentrations, at some distance from operational and non-operational waste retention systems, sufficiently low so as not to constitute a significant health hazard.

TABLE I. URANIUM MILLING: AIRBORNE RELEASES OF RADIOACTIVE MATERIALS

Radionuclide	Release rate ($\mu\text{Ci/d}$)	Site boundary ^a air concentration ($\mu\text{Ci/ml}$)
Natural uranium	500	7.9×10^{-14}
Thorium-230	270	4.3×10^{-14}
Radium-226	270	4.3×10^{-14}
Radon-222	13×10^5	1.1×10^{-11}

^a Distance to site boundary assumed to be 600 m.

However, in houses built in the immediate vicinity of or directly on tailings heaps, an elevated radon concentration can be expected. Significantly elevated radon daughter concentrations as well as increased dose rates have been observed in houses built over fill material containing uranium mining and milling waste [5].

The estimated maximum release rates of airborne radionuclides and conservative estimates of air concentrations at the site boundary for a model uranium mill producing 960 t U/a are given in a recent survey [6].

The source of the radionuclides is the plant and the ore pile and tailings system (see Table I). The radionuclide concentrations constitute only a fraction of those considered acceptable for individual members of the public.

Radon-222 dosimetry is a complex and difficult subject. The radiation dose caused by radon is a function of the equilibrium state between radon and its daughters at the time of exposure and therefore it is not enough to know the radon-222 concentration alone. Long-term exposure occurs because mill tailings contain long-half-life ^{226}Ra , the parent of radon, so that radon will be continually produced and will emanate from the pile indefinitely.

A ^{226}Ra concentration of 1 pCi/l above background has been used as the recommended limit for continuous exposure to individuals in the general population [7]. It should be noted that background levels may range from 0.1 to 1.0 pCi/l in the free atmosphere and from 1 to 10 pCi/l in granite dwellings.

Wind-borne particulate material from waste retention systems may also be of some significance because of the ^{230}Th and ^{226}Ra content. It appears that radon releases, which are the most difficult to control, are of most significance but that radioactive particulate releases also may sometimes be significant [8].

The United Nations Scientific Committee on the Effects of Atomic Radiation [9] found that the contribution of uranium mining and milling operations to the radiation dose commitment is probably small but should be evaluated.

Radon emanation and the release of radioactive particulates from a given amount of tailings is reduced when:

- (a) The tailings area exposed is decreased;
- (b) The tailings are under water, ice or snow;
- (c) The tailings are covered with a thick cover of earth or an impermeable membrane.

The radiation dose to the public can be reduced by:

- (a) Siting tailings dams as far as possible from established residential areas;
- (b) Restricting the development of new residential areas close to operating or non-operating tailings areas.

In spite of some uncertainty regarding the actual health effect of radon and particulates released from tailings, it appears prudent to utilize all the above measures to the maximum extent possible to limit a potential future detriment.

Radium and heavy-metal accumulation on the surface of sulphide and non-sulphide wastes may hinder or inhibit seed germination and plant establishment. The impact of these accumulated metals is widespread and not clearly defined. Several plant species can tolerate a high accumulation of metals with no apparent damage, but they may be toxic to grazing animals. Other reactions in the food chain to man are not clearly known. When the chemical substances that inhibit vegetation establishment are treated, more waste results with similar detrimental properties.

Owing to the size of some large tailings areas the complete isolation of the waste from water courses will be impossible. Filling a lake or valley with tailings will result in a greater moisture reserve than previously existent whereas constructing a tailings dam on a flat surface and raising it by perimeter dams will maintain a drier surface.

When tailings are deposited near population centres or in a scenic area, it is essential to fit them aesthetically into the surroundings. These considerations will influence the extent of reclamation to be carried out.

4.2. Sludges

Precipitated sludges will normally remain insoluble and will not present an immediate hazard. However, as the operations proceed, or if the nature of the liquid flowing over the precipitate changes — either because of process changes or simply because of abandonment of the operation — problems may be encountered caused by slow release of the ^{226}Ra content.

The accumulated sludge is considerably more radioactive than solid tailings and may become a source of radium contamination. Under unusual conditions, such as a 10-year storm or in the case of a main dam or settling dam failure, the sludge could be washed downstream into public receiving waters. Should this occur, it will present a radiological hazard both as regards the ingestion of particulate matter and the possibility of dissolution because of the composition of the receiving water.

4.3. Non-operating mill sites

Very few countries have any experience in the decommissioning of uranium mill sites. The USA does have certain regulations concerning the clean-up levels necessary to decontaminate abandoned buildings and the surrounding area. These criteria are also utilized at abandoned uranium ore-buying stations. The prescribed procedures are necessary especially in areas where the mill site may be utilized after the mill has been shut down. In the western USA where over twenty mills have been closed down, the local residents are often interested in utilizing the remaining buildings. Extensive decontamination procedures have been necessary to reduce the contamination levels from the uranium tailings and former ore stockpiles.

5. CONCLUSIONS

The environmental problems posed by wastes from the uranium milling industry are receiving attention by the industry and by national authorities. Air pollution from mills is not a serious

problem. Water pollution is potentially a greater problem, but methods are available to treat liquid effluents released from operational mills so that their impact need not be significant.

The problems posed by the disposal of tailings in engineered surface waste retention systems are not completely resolved at the present time. Some of these problems, such as contaminated seepage, revegetation and stabilization, are common to other mineral extraction operations. The unique problems arise from the long half-life of the radionuclides (notably ^{230}Th and ^{226}Ra) present in the tailings, which have the potential to lead to radiation exposure of both present and future populations. The actual significance of these potential radiation exposures has not yet been convincingly demonstrated. Bearing in mind the fact that these exposures are caused by natural radioactivity to which mankind has always been exposed, it is believed that the total radiation detriment caused by the extraction of uranium from ores can be shown not to be unjustifiable in relation to the benefits from such extraction.

ACKNOWLEDGEMENT

The information reported here is drawn extensively from the work of two IAEA meetings convened to consider waste management in the uranium and thorium mining and milling industry. The first meeting was held in Ottawa, 8–12 July 1974, under Mr. K. Hester as Chairman, the second was convened in Vienna, 12–16 May 1975, with Mr. D. Nussbaumer as Chairman. The author wishes to thank all members of those meetings for their active participation.

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DISCUSSION

F.E. MCGINLEY: I want to thank Mr. Bonhote for an excellent paper. There is a very good compilation of information and data accumulated throughout the years in the United States which I can recommend to those interested in this particular subject, namely the Oak Ridge National Laboratory

Report TM-4903, Volumes 1 and 2 ("Correlation of Radioactive Waste Treatment Costs and the Environmental Impact of Waste Effluents in the Nuclear Fuel Cycle for Use in Establishing 'As Low As Practicable' Guides — Milling of Uranium Ores", by M.B. Sears et al., May 1975). The report is available from the National Technical Information Services, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, Virginia, 22161.

W.R. FLÖTER: Is it related to the USA or worldwide?

F.E. MCGINLEY: This is specific to the USA but it does contain a compilation of just about all the information that has ever been published in the USA on this particular subject — it is a very good reference work.

PROCESSING OF LOW-GRADE RESOURCES
(Session V)

TRAITEMENT DES MINERAIS D'URANIUM A FAIBLE TENEUR

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Abstract--Résumé

PROCESSING OF LOW-GRADE URANIUM ORES.

The low-grade ores are defined as ores which cannot be economically processed in the existing plants and for which conventional processing plants cannot be built. In the report four types of low-grade ores are studied: (1) Low-grade ores that must be extracted because they are enclosed in a normal-grade deposit. Heap leaching is the processing method which is largely used at present in many countries. It allows to obtain solutions or preconcentrates which may be delivered at the nearest plant. The main features of this method are developed in the report. (2) Normal-grade ores contained in low-amplitude deposits. They can be processed using in-place leaching as far as the operation does not need any large and expensive equipment. (3) Medium-grade ores in medium-amplitude deposits. A simplified conventional process can be applied using fast heap leaching. An example of a practical application is given in the report. (4) Low-grade ores in large deposits. Great attention must be paid to this type of ore. The report explains processing possibilities leading in most cases to the use of in-place leaching. The operating conditions of this method are laid out, especially the selection of the leaching agents and the preparation of the ore deposit to obtain a good tightness with regard to the hydrological system and a good contact between ore and reagent. Conclusions are supposed to be drawn during the panel discussions. Nevertheless it appears that using in-situ leaching is very attractive, but it must be adapted to each case. Many experiments will be needed to overcome all the difficulties.

TRAITEMENT DES MINERAIS D'URANIUM A FAIBLE TENEUR.

Les minerais à faible teneur sont définis comme des minerais qui ne peuvent être économiquement traités dans les usines existantes ou ne justifient pas la construction d'une installation classique pour leur traitement. On distinguera donc dans le rapport quatre catégories de minerais: 1) Les minerais pauvres extraits obligatoirement, parce qu'ils sont inclus dans un gisement normal. La méthode de traitement largement utilisée à l'heure actuelle dans de nombreux pays est la lixiviation en tas, qui permet de préparer des solutions ou des préconcentrés livrables à l'usine la plus proche. Les caractéristiques principales de cette méthode sont développées dans le rapport. 2) Les minerais riches en gisements de faible importance. Ils sont passibles de la lixiviation *in situ* pour autant que la mise en oeuvre ne nécessite pas d'investissements importants. 3) Minerais en gisements d'importance et de richesse moyenne. On peut appliquer un procédé classique simplifié en attaquant le minerai par lixiviation accélérée. Un exemple de mise en oeuvre est donné dans le rapport. 4) Minerais pauvres en gisements étendus. C'est le cas qui doit attirer le plus l'attention. Le rapport expose les possibilités de traitement qui conduisent à l'emploi de la lixiviation *in situ*. Les conditions d'application de cette méthode sont étudiées dans le rapport, en particulier le choix des réactifs d'attaque, et la préparation du gisement pour un bon isolement vis-à-vis du réseau hydrologique et pour une bonne mise en contact. Le rapport laisse à la table ronde le soin de tirer des conclusions. Il apparaît cependant que l'emploi de la lixiviation *in situ* est très séduisant, mais qu'il nécessite des adaptations à chaque cas. De nombreuses expériences seront nécessaires pour pouvoir répondre à toutes les difficultés.

1. INTRODUCTION

Les réunions précédentes, tenues à Vienne (1966) et São Paulo (1970), avaient montré l'intérêt que tous portaient au traitement de minerais pauvres. Toutefois, la dépression du marché de l'uranium avait freiné le développement à grande échelle de la plupart des méthodes étudiées. Le nouveau départ de la production d'énergie électrique d'origine nucléaire a entraîné en particulier

des besoins accrus en uranium. L'augmentation de la demande en uranium a également pour conséquence la recherche de nouvelles sources, en particulier les minerais pauvres jusque-là inexploités ou mal reconnus. Il est très probable que dans les 10 années à venir certains gisements pauvres commenceront à être exploités.

2. CLASSIFICATION DES MINERAIS PAUVRES

Presque tous les minerais d'uranium sont peu riches, avec des teneurs nettement inférieures à 1% en uranium. Un minerai n'est donc réputé *pauvre* que par référence économique. C'est un minerai qui, exploité et traité seul par des méthodes classiques, fournirait de l'uranium à un prix supérieur au prix du marché.

Dans un pays industriel moderne, la teneur de coupure variera avec les difficultés d'exploitation et de traitement, mais elle sera certainement inférieure ou voisine de 1‰ si l'on est près d'une usine de traitement ou de 2‰ si l'on est à plus de 200 km d'une usine.

Dans un pays plus isolé industriellement (Afrique par exemple) où l'on doit amener de loin tous les éléments nécessaires au traitement, la teneur de coupure peut être plus élevée (3‰ par exemple).

Les critères sont également différents si le minerai est extrait obligatoirement (zones marginales d'une exploitation minière). On a alors tendance à traiter les minerais au maximum dans l'usine si elle n'est pas saturée, ou par d'autres méthodes.

Une autre variété de minerais pauvres sont les gisements de teneur très satisfaisante (plusieurs ‰) mais de faible réserve. Le plus souvent les travaux d'exploitation (carrière ou mine) sont trop chers à eux seuls et on est amené à concevoir leur mise en valeur en place.

En résumé, on considérera dans ce qui suit les quatre catégories suivantes de minerais pauvres:

- les minerais pauvres exploités obligatoirement,
- les minerais normaux en faibles amas,
- les minerais ni très pauvres, ni en amas très faibles mais à distance notable d'une usine de traitement,
- les minerais réellement pauvres en amas importants.

Dans l'exposé, on n'a pas pris en compte, bien entendu, l'uranium récupérable comme sous-produit, par exemple du traitement des minerais d'or ou des phosphates.

3. TRAITEMENTS APPLICABLES AUX MINERAIS PAUVRES

3.1. Minerais pauvres exploités obligatoirement

Il s'agit donc de minerais habituellement traités dans une usine classique relativement proche des gisements et qui se trouvent à des teneurs inférieures à la teneur de coupure jugée économique. Cette teneur peut varier de 0,8‰ comme dans les exploitations françaises à 2 ou 3‰ comme en Afrique.

Une méthode couramment appliquée maintenant en France et dans de nombreux pays est la lixiviation en tas de ces minerais marginaux. Elle a été fréquemment décrite [1-5] et on n'en rappelle ici que les caractéristiques principales (fig.1).

- Il faut créer une *aire étanche*. En règle générale pour ces teneurs en uranium on cherche à réduire au maximum les frais d'investissement. Aussi les aires seront constituées de la manière la plus rustique possible; par exemple le sol sera nivelé avec une légère pente, un revêtement étanche en feuilles de chlorure de polyvinyle sera posé et une couche protectrice de gravier ou de sable stérile déposée sur les feuilles étanches.

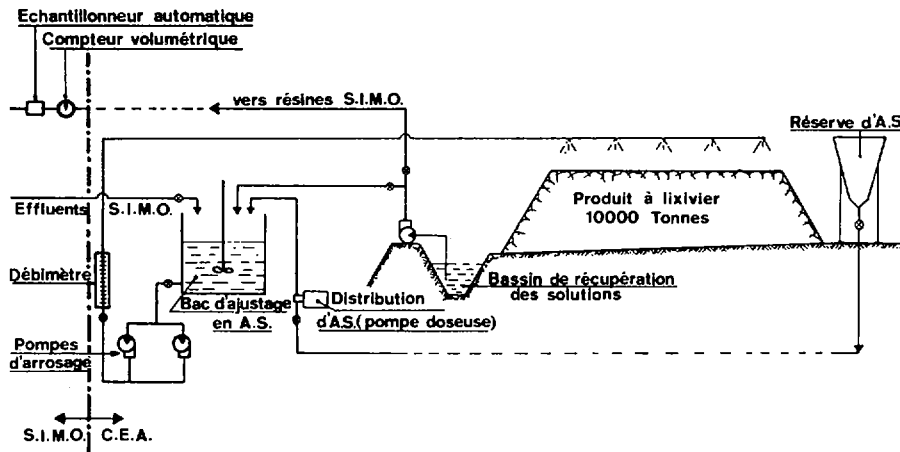


FIG.1. Lixiviation en tas, schéma de fonctionnement.

Le choix d'une telle aire rustique suppose implicitement que l'on travaille en «aire perdue», ce qui évite les frais de reprise des stériles. Ce choix nécessite en revanche la disponibilité de terrains importants.

Si l'on ne dispose pas de surfaces suffisantes et si les minerais que l'on veut traiter ne sont pas trop pauvres, il peut être encore rentable de construire des aires récupérables avec revêtement durable et d'enlever les stériles en fin d'opération.

— L'aire étanche étant établie, elle doit être *chargée*, en évitant de tasser le minerai, par exemple en faisant rouler des véhicules sur le tas pendant sa constitution, surtout si le minerai contient des fines abondantes (argiles par exemple). Si le minerai est vraiment pauvre, on le chargera tel quel. S'il est moins pauvre, il peut être intéressant de lui faire subir un concassage primaire qui augmentera notablement le rendement d'attaque.

La hauteur possible du tas dépendra essentiellement de sa nature; un minerai fortement argileux à travers lequel la percolation des liqueurs est difficile sera de préférence disposé en tas peu épais (quelques mètres). Si au contraire, comme à Bessines (France), le minerai est favorable, on peut constituer des tas de 10 ou 20 m de haut.

— Le chargement étant fait, on dispose sur le haut du tas des tubes d'arrosage qui peuvent être simplement des tubes plastiques perforés, et qui sont amovibles. On envoie par ces tubes le réactif convenable (le plus souvent de l'acide sulfurique) à un débit variable selon les possibilités de percolation. Les débits peuvent ainsi varier de 1 à 25 l/h·m². Les liqueurs chargées sont recueillies dans des bassins, constitués aussi de façon rustique: trous dans le sol recouverts de plastique (caoutchoucs synthétiques le plus souvent). Une partie des liqueurs va à la production et est compensée par des réactifs neufs. Le reste des liqueurs et les réactifs neufs sont recyclés sur le tas. Selon le minerai traité on peut obtenir des solutions titrant de 100 mg/l à 2 g/l.

— La solution enfin doit être *traitée*. Si les tas sont voisins de l'usine de traitement, la solution peut directement être injectée dans les circuits de purification (résines ou extraction par solvant). C'est le cas à Bessines et à l'Ecarpière (France). Si au contraire on est à une certaine distance de l'usine, il faut savoir s'il vaut mieux transporter des liqueurs ou des minerais. Les premières ne sont intéressantes que si leur teneur est notablement supérieure à celle des minerais.

Dans certains cas, on a essayé d'obtenir sur place un concentré intermédiaire pour réduire les frais de transport. On précipitait le plus souvent un uranate de calcium très impur titrant de 4 à 15% en uranium. L'inconvénient est qu'on doit créer sur place une infrastructure de traitement (cuves de précipitation, décanteurs ou filtres, préparation de réactifs, traitement d'effluents éventuellement). Le choix ne peut être fait que par un bilan économique comparatif.

On peut aussi proposer de fixer l'uranium sur résine et transporter soit la résine chargée, soit un éluat concentré. Dans les deux cas, il faut aussi créer une infrastructure pour traiter les effluents et disposer d'une petite aire d'épandage.

Dans ce qui précède, on suppose implicitement que le traitement par lixiviation est compatible avec celui effectué à l'usine et en particulier que le réactif est semblable voire identique de nature, ou que le concentré obtenu peut être facilement repris par l'usine. Il y a toute chance pour qu'il en soit normalement ainsi et que le minerai soit de même nature et seulement de teneur différente. Si toutefois il en était autrement (par exemple minerai marginal plus calcaire que le minerai riche traité normalement par l'acide sulfurique) il est clair que, dans la plupart des cas, ce minerai marginal ne devrait plus être traité, sauf si les quantités disponibles permettaient de justifier un traitement différent du traitement principal. Là encore, un bilan économique peut seul permettre une décision.

La lixiviation peut donc être d'un très grand secours dans ce cas, mais peut-on concevoir l'emploi d'autres méthodes?

Des méthodes d'enrichissement physique qui fourniraient un concentré physique au moins aussi riche que le minerai nominal seraient assez séduisantes. Trois voies ont été explorées et peuvent être envisagées:

- *Le triage radiométrique* [6]. Il consiste à trier les grains de plus ou moins grande taille au moyen d'un comptage qui commande un système d'élimination des grains refusés ou des grains acceptés. Ce procédé a été pratiqué à Bessines (France) sur le minerai tout-venant avec un certain succès. Toutefois, on a considéré que l'enrichissement obtenu (1,25) n'était pas en rapport avec les dépenses engagées (investissement et exploitation). En outre, ce système est *aveugle* vis-à-vis des morceaux de minerai où l'uranium est disséminé dans la masse stérile.

- *La flottation* [7] a été essayée ou pratiquée dans de nombreux pays. Cependant l'ensemble des résultats est assez décevant car il n'y a finalement que très peu d'espèces minérales d'uranium qui soient flottables (l'autunite par exemple) et l'enrichissement n'est souvent obtenu *a contrario* que par élimination de substances gênantes (sulfures par exemple). Le taux d'enrichissement, hormis quelques cas d'espèce, est généralement faible.

- *Les méthodes gravimétriques* [8] ont donné quelques résultats satisfaisants mais sont aussi limitées à quelques cas favorables. Si l'uranium est sous forme de pechblende ou d'uraninite assez bien libérée, on peut obtenir des résultats parfois spectaculaires. Dans la majorité des cas, cependant, l'enrichissement obtenu est faible par rapport aux moyens mis en œuvre.

On peut donc dire en résumé que, dans quelques cas bien déterminés, un procédé d'enrichissement physique peut permettre de bien améliorer un minerai pauvre, mais cela ne peut en aucun cas constituer une méthode générale.

3.2. Les minerais riches en faibles amas

Il s'agit de gisements dispersés qui ne justifient pas, même réunis ensemble, l'installation d'une usine de traitement et qui sont relativement loin des usines existantes. Plus précisément, il existe des amas riches ou relativement riches de quelques dizaines de tonnes d'uranium et trop profonds pour une exploitation économique par travaux miniers traditionnels. Il faut donc trouver une méthode rustique qui permette d'obtenir des concentrés d'uranium transportables. Ce ne

peut être que la lixiviation *in situ*, utilisant des réactifs classiques avec des minerais dans des structures géologiques elles aussi classiques. Il ne semble pas que ces gisements aient jusqu'ici fait l'objet d'une exploitation de ce genre bien que, souvent, ils puissent représenter pour tout un pays des milliers de tonnes d'uranium. En France, une étude a été entreprise pour exploiter les nombreux petits gisements en terrain cristallin par lixiviation sulfurique *in situ*. Les caractéristiques et contraintes du procédé sont les suivantes:

- Le schéma général est l'injection, par des forages de diamètre relativement faible, d'une solution lixivante. La solution résultante est recueillie ensuite, par d'autres forages, et doit subir un traitement concentrant.

- Le gisement doit en général être préparé pour permettre un bon contact entre minéral et solution lixivante. Cette préparation consiste essentiellement en une fragmentation qui peut être obtenue par charges explosives ou par fracturation hydraulique.

Ces possibilités ont notamment fait l'objet de communications au récent *Dallas Solution Mining Symposium* [9–11].

- Des travaux préparatoires doivent également être faits pour éviter la fuite de solutions dans le réseau hydrologique souterrain.

Ces précautions ont naturellement deux buts: d'abord et au premier degré, éviter la pollution, essentiellement chimique, de l'environnement; ensuite, évidemment, obtenir une récupération aussi complète que possible de l'uranium.

Dans un terrain cristallin, il semble que ce but puisse être généralement assez facilement atteint au prix d'une faible dilution de la solution résultante.

- La solution doit être traitée pour obtenir un facteur de concentration convenable, par exemple par fixation sur résine. Il faut transporter à une usine soit la résine chargée, soit l'éluat. Il se posera le problème de traiter au moins la partie non recyclée des effluents de fixation et de l'épandage des résidus solides (essentiellement le sulfate de chaux).

Il faudra attendre les expériences dans les conditions réelles pour savoir si ces méthodes sont applicables.

3.3. Gisements de teneur et d'importance intermédiaires

Les minerais traités en France et aux Etats-Unis, par exemple, sont pour la plupart à des teneurs comprises entre 1‰ et 3‰, avec des valeurs moyennes qui varient de 1,5 à 2‰. Quand les gisements dont ils proviennent contiennent des réserves importantes, il est justifié économiquement de créer une usine classique appliquant un procédé adapté aux minerais à traiter. Quand ce n'est plus le cas et que les réserves permettent une exploitation à la capacité de 50 à 200 t d'uranium par an, une usine classique est difficile à amortir. On se trouve donc dans un cas intermédiaire où le minéral est trop pauvre pour payer les frais d'extraction, de transport et de traitement et trop peu abondant pour justifier une usine classique qui lui soit consacrée.

De tels cas existent en France. Ainsi la Compagnie française des minerais d'uranium (CFMU) a été conduite à adopter une technique intermédiaire qui évite les investissements lourds et les coûts d'exploitation élevés. Il faut pour cela supprimer au moins les opérations de fragmentation fine (broyage), de séparation solide-liquide (décantation ou filtration) et de clarification. A Langogne (France), l'attaque des minerais les moins pauvres est réalisée sur le tout-venant (<300 mm) par lixiviation sulfurique en stalles de 2000 t [12]. Elles sont protégées du froid et des intempéries par des hangars mobiles. La durée du cycle de chargement, attaque, lavage et déchargement du minéral est de 1 mois environ (fig.2).

Les solutions sont purifiées par solvant aminé et l'uranium finalement produit sous forme d'uranate de magnésium. Les minerais marginaux sont traités selon les principes exposés au paragraphe 3.1. L'unité peut ainsi produire une centaine de tonnes d'uranium par an à des prix compétitifs.

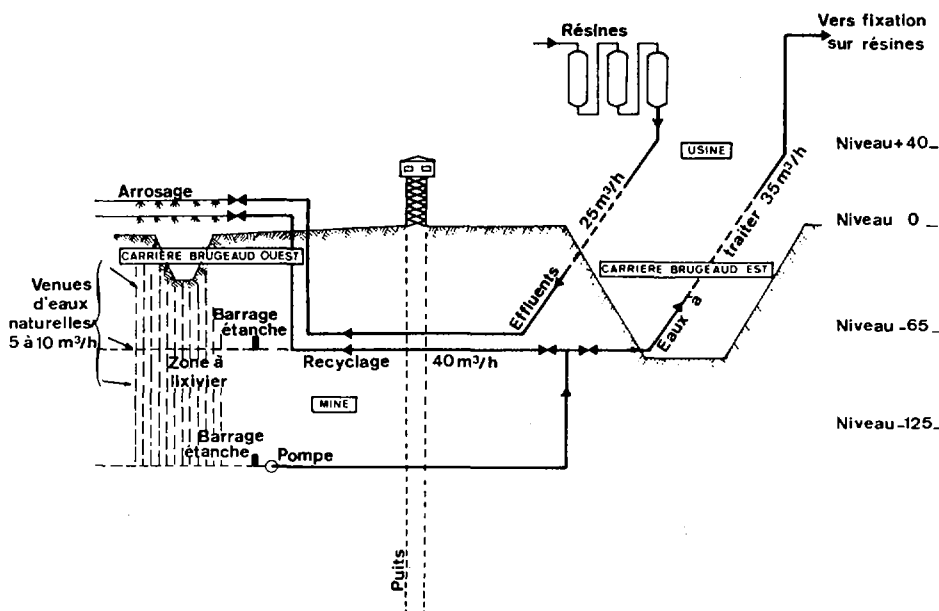


FIG.2. Lixiviation au fond (exemple de Brugeaud ouest).

Certains minerais sont difficiles à lixivier à cause de la présence de fines argileuses qui gênent la percolation. On peut alors envisager de faire une séparation de ces fines par une méthode physique telle qu'un cyclonage. La sous-verse serait traitée par lixiviation. Mais la surverse peut contenir une proportion importante d'uranium et il est exclu de la rejeter. On pourrait alors la traiter par attaque classique en petites cuves et traiter ensuite la pulpe sans séparation solide-liquide. On utiliserait par exemple le système appelé «résine en pulpe», qui est déjà pratiqué industriellement [10]. Un tel ensemble, s'il est réalisable, est plus coûteux que celui qui n'utilise que la lixiviation mais peut encore permettre de rentabiliser un gisement moyen.

3.4. Minerais pauvres en grands gisements

Il s'agit des minerais en amas abondants en quantité, mais de faible teneur en uranium.

Il apparaît à tous, au premier examen, que la rentabilisation de tels gisements, pour un cours donné d'uranium, n'est possible qu'avec des travaux miniers minimaux et un traitement chimique simple.

Pour minimiser les travaux miniers, le procédé le plus simple qui vienne à l'esprit est la mise en solution *in situ*. On a déjà vu (paragraphe 3.2) les conditions de mise en application de cette méthode, mais le problème était plus simple à résoudre du fait de la faible extension du gisement. Dans le cas d'un gisement étendu, les problèmes de confinement vis-à-vis des réseaux hydrologiques naturels sont plus difficiles:

- pollution du réseau hydrologique, et accessoirement pertes de rendement en uranium,
- à l'inverse, dilution excessive de l'uranium par les venues d'eaux souterraines.

Avant d'aller plus loin, il convient de classer les cas qui peuvent se présenter. Cette classification apparaît dans le tableau I. On rencontre trois grandes catégories de problèmes:

- choix du réactif d'attaque,
- préparation du gisement pour une bonne récupération des liqueurs,
- obtention d'une bonne mise en contact.

Avant d'étudier un peu plus en détail ces trois aspects il faut d'abord vérifier qu'on ne se crée pas dans certains cas des problèmes qui n'en sont pas. Il n'est pas exclu en effet de trouver des gisements pauvres pratiquement en surface. On pourrait alors concevoir de les exploiter en carrières simples, avec un coût direct relativement faible. A cette conception s'oppose la volonté, de plus en plus répandue dans les pays économiquement développés, de ne pas altérer irréversiblement l'environnement. De grands gisements pauvres deviennent ainsi inexploitable en carrières parce que cela conduirait à détruire le paysage; et même si l'on admettait cette destruction transitoirement, il faudrait certainement procéder à une reconstitution dans laquelle tous les profits passés seraient probablement absorbés. Il faut pourtant noter que la Suède a ainsi abordé le problème du traitement de ses schistes pauvres [13] (0,3% avec un rendement de 70%). Une production de 500 t/an d'uranium dans ces conditions conduirait à sortir de 2 500 000 à 3 000 000 t de minerai.

La recherche de méthodes de traitement *in situ* apparaît donc indispensable, comme elle l'est pour le cuivre [14]. Plusieurs sociétés envisagent de l'utiliser en grand, telle la *Wyoming Mineral Corporation* à Bruni (Texas) [15].

3.4.1. Choix du réactif

En supposant, ce qu'on verra en 3.4.2, que la récupération des liqueurs soit bonne, il faut utiliser soit des réactifs connus et assez efficaces tels que l'acide sulfurique ou les carbonates alcalins, soit des réactifs a priori plus coûteux mais qui seraient à la fois sélectifs et aisément récupérables.

L'acide sulfurique doit en général être accompagné d'un oxydant parce que l'uranium est souvent à l'état réduit dans le minerai. Pour la lixiviation (en tas ou *in situ*) il doit être soluble. Le moins cher est assurément le fer à l'état ferrique pour autant qu'on puisse le trouver dans le minerai et qu'on puisse l'oxyder sans frais en utilisant par exemple des bactéries du type ferro-oxydants [16]. Si on ne trouve pas de fer dans le minerai, il faut ajouter un oxydant classique, les moins polluants en cas d'incident étant les chlorates alcalins et l'eau oxygénée.

La lixiviation alcaline ne doit être évidemment envisagée que si les constituants du minerai sont la cause d'une consommation exagérée d'acide sulfurique, par exemple si la teneur en carbonates est trop élevée. Là aussi un oxydant est souvent nécessaire et il est plus difficile d'en trouver qui soient efficaces et stables en milieu alcalin. Certains ont proposé les chlorates ou l'eau oxygénée [15]. La discussion reste ouverte à ce sujet.

Restent les agents de lixiviation nouveaux qui seraient sélectifs et récupérables. Théoriquement ils devraient déjà exister et être utilisés dans des usines classiques. En fait, l'acide sulfurique et les carbonates alcalins ont jusqu'à présent permis de résoudre tous les problèmes d'une manière assez économique et cela n'a évidemment pas stimulé la recherche de tels agents. Ils devraient être probablement organiques et complexants comme le DTPA, l'EDTA, d'autres dérivés de l'acide acétique ou des acides organiques habituellement complexants (tartrique ou citrique). Ils seraient pour la plupart déplaçables par un acide fort minéral.

Ce pourrait être un des sujets de discussion de cette réunion. Il ne faut pas terminer cette revue sans rappeler que de très nombreux travaux ont porté sur l'utilisation des bactéries. Deux cas sont possibles:

- La présence de pyrites dans le minerai permet à des bactéries du type thiooxydants et ferroxidants de créer le réactif d'attaque: ions sulfates et ions ferriques [17].

TABLEAU I. CLASSIFICATION DES CAS POSSIBLES

Nature du sol	Méthode applicable	Conditions d'application
1) Granits ou grès partiellement dégradés	Lixiviation sulfurique <i>in situ</i>	Étanchéité naturelle ou facilement obtenue pour le gisement Uranium d'accès facile ou rendu tel par ébranlement (explosion,hydraulique)
2) Granits ou grès très dégradés (argiles)	Lixiviation sulfurique <i>in situ</i>	Percolation obtenue par des additifs
3) Schistes	Lixiviation sulfurique <i>in situ</i> ou lixiviation alcaline <i>in situ</i>	Comme en 1), étanchéité renforcée dans le cadre de la lixiviation alcaline
4) Minerais très pauvres du type granit et schiste durs	A déterminer	

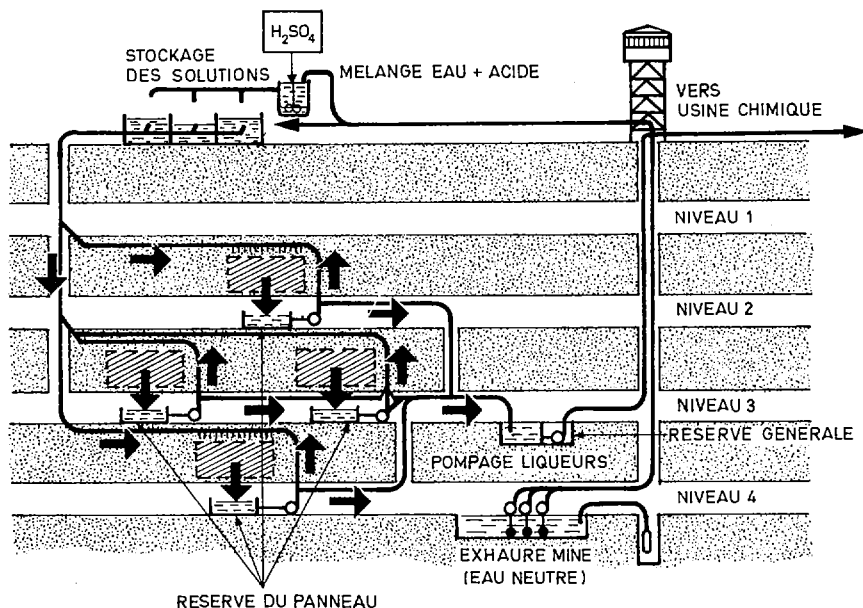


FIG. 3. Préparation du gisement, schéma général de la mine.

— Des bactéries spécifiques susceptibles de provoquer la mise en solution de l'uranium ont été également étudiées [18,19]. Elles agissent d'ailleurs en synthétisant des complexants organiques tels que des acides aminés.

Les bactéries demandent dans tous les cas des conditions de développement très particulières et, en définitive, cela revient à utiliser des réactifs très dilués, donc à lixivier très lentement.

3.4.2. Préparation du gisement

Le gisement doit être préparé pour permettre de récupérer les liquueurs chargées en uranium. A cet égard plusieurs possibilités peuvent s'offrir:

- a) Le gisement pauvre est le reste d'une exploitation classique en mine ou carrière. On peut alors bénéficier de tous les travaux antérieurs, et de modestes travaux complémentaires permettent:
 - d'empêcher les venues d'eau trop importantes,
 - d'empêcher les fuites de liquueurs vers le réseau hydrologique naturel,
 - de créer les points bas et d'installer les pompes nécessaires pour remonter les liquueurs à la surface (fig.3).

b) Le gisement est d'une teneur insuffisante pour une exploitation classique mais suffisante pour permettre de payer des travaux miniers, par exemple en créant quelques galeries simples [20]. On serait techniquement à peu près ramené au cas a) ci-dessus.

c) Le gisement ne permet que le percement de quelques puits de diamètre juste suffisant pour implanter les pompes destinées à remonter les liquueurs. Il n'y a pas de règles générales et chaque gisement sera un cas particulier, selon son isolement, naturel ou non, vis-à-vis du réseau hydrologique et sinon, selon les méthodes possibles pour créer une étanchéité artificielle. Ce pourrait être un second sujet de discussion de cette session.

De toute façon, les travaux minimaux comporteront le percement de puits d'injection et de puits de pompage.

3.4.3. Obtention d'un bon contact

Sauf cas particulier, le minerai n'est pas assez divisé pour obtenir un bon contact. Il semble admis généralement qu'une fracturation préalable des zones minéralisées est nécessaire. Les méthodes proposées dans la littérature peuvent être classées en deux catégories principales:

- fracturation par explosion, qui a été particulièrement étudiée par l'USBM [9] et Du Pont [10],
- fracturation hydraulique, inspirée des méthodes pétrolières et qui a été exposée au *Dallas Solution Mining Symposium* [11].

La comparaison de ces deux méthodes de fracturation et éventuellement l'examen de méthodes qui n'ont pas encore fait l'objet d'une publication pourraient constituer un troisième sujet de discussion à cette session. Ces procédés ne sont en fait applicables avec efficacité qu'à des terrains granitiques ou gréseux. Dans le cas de gisements argileux, la fracturation mécanique est inutile. La percolation est très mauvaise [21] dans un tel terrain. Il faudrait user d'adjuvants particuliers (agents mouillants ou coagulants) permettant une bonne pénétration des réactifs dans les argiles. Ce pourrait être un quatrième sujet de discussion pour cette session.

Il faut enfin citer les procédés utilisant l'électromigration [22] qui pourraient accélérer la mise en solution de l'uranium dans une roche poreuse ou rendue telle. La validité technique et économique de ce procédé reste à démontrer.

Certains gisements, toutefois, peuvent rester très peu perméables aux réactifs malgré l'emploi des méthodes précitées de fracturation. Il peut s'agir de zones dont le minerai principal est un schiste dur ou un granit sain. Dans ce cas, une étude plus précise doit être entreprise pour savoir si on peut économiquement appliquer:

- une méthode de préparation du gisement par travaux souterrains simplifiés, comme exposé au paragraphe 3.4.2.b),
- une méthode d'exploitation classique du minerai avec traitement chimique simplifié, comme exposé au paragraphe 3.3.

Sinon, il faudra attendre alors la mise au point de méthodes originales qui n'en sont peut-être encore qu'au stade d'études préliminaires.

Un dernier mot concernant les minerais très pauvres. Il existe, notamment aux Etats-Unis et en Europe, des zones granitiques contenant de l'uranium dispersé à des teneurs comprises entre 1 et 25 ppm avec une valeur moyenne de l'ordre de 5 à 10 ppm. Une étude poussée avait été publiée en 1966 par l'ORNL [23]. Les résultats montraient la possibilité d'obtenir des rendements de l'ordre de 70% à l'attaque par l'acide sulfurique (10 à 20 kg/t). L'attaque par lixiviation *in situ* n'a pas été étudiée, mais certains résultats laissent apparaître cette possibilité sous réserve d'une très forte fragmentation. Toutefois, on rencontrerait deux obstacles principaux dans l'exploitation de telles ressources:

- une très forte fragmentation, même en laissant le minerai en place, constituerait une forte atteinte à l'environnement,
- la rentabilité de la récupération de cet uranium ne paraît pas pouvoir être démontrée actuellement.

4. CONCLUSIONS

Les conclusions ne pourront être tirées qu'après la discussion de la table ronde. On voit cependant apparaître les lignes générales suivantes:

- Le traitement des minerais marginaux est déjà largement pratiqué, en utilisant la lixiviation en tas et d'une manière souvent rentable.
- Le traitement des minerais pauvres au moyen de la lixiviation en place est proposé très généralement, mais il pose des problèmes hydrogéologiques, de préparation du gisement et de chimie de mise en solution. L'économie de l'ensemble est évidemment fonction de l'importance des dépenses à faire pour résoudre chacun de ces problèmes.

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DISCUSSION

K.M.V. JAYARAM: What is the approximate proportion of solution loss in in-situ leaching?

P. MICHEL: In fact, we have very few results for in-situ leaching. We have used it in two cases. In the first one, it was used to exhaust an old uranium mine. In this case we were able to do some preparatory work to avoid any loss of solution because we had galleries and also the bedrock was quite tight by itself. In the second case, we tried to apply in-situ leaching in a mine that was still operated by traditional methods. So we had two different methods there and it was very difficult to acquire sufficient control. We were obliged to stop the experiment because solution penetrated to lower galleries, which was very dangerous for the miners.

K.M.V. JAYARAM: Am I to understand that the ore is hoisted and then leached by heap leaching?

P. MICHEL: No, it is in-place leaching. Small explosions were used to fracture the ore in Bessines.

S. AJURIA-GARZA: If I understand correctly, the tanks for leaching intermediate-grade ore are used several times. How do you take the ore out from the vats without damaging the rubber lining?

P. MICHEL: The rubber lining is protected with wood. Another precaution is to leave a small part of the exhausted ore in the vat to avoid damaging the lining. With these precautions, there was no accident in three years.

S. AJURIA-GARZA: Can you remove the ore by mechanical means?

P. MICHEL: We use a mechanical shovel because the vats are relatively small — they contain only 2000 tonnes. I think about half a week is needed to unload the vats.

F.C. LENDRUM: Did I understand correctly that there are bacteria that attack the uranium minerals?

P. MICHEL: You mean bacteria other than thiooxidans or ferrooxidans?

F.C. LENDRUM: Yes.

P. MICHEL: Two references in my paper concern studies that were made in a school laboratory in France on bacteria that generate amino acids. These were able to solubilize a certain amount of uranium in ores. The amino acids were at very low concentrations. The studies were made partly to explain certain geological phenomena and why it is that sometimes we can find uranium in places where it was not expected.

B. ROSNER: When applying in-situ leaching methods or heap leaching or stope leaching, did you experience any clogging? Was there a stoppage of leaching after a while, for instance by formation of gypsum or colorite, and if so, what did you do to start leaching again?

P. MICHEL: With granite rock, which is the main ore type in France, we had no difficulties with percolation, for instance from calcium sulphate, except if the calcium carbonate content was high. In this case, a large amount of calcium sulphate is formed and sometimes the percolation rate is slightly decreased. But we had other experiments, for instance at a small clay deposit, where in-situ leaching was applied for uranium extraction. There, percolation was very poor and the experiment was discontinued. But we hope to test again in-situ leaching of this clay deposit by using flocculating or wetting agents to improve percolation.

T.S. ARY: It seems that the heap leach pile contains rather large-size rock fractions. What percentage of the estimated uranium content did you actually recover from the leach pile?

P. MICHEL: It is not easy to estimate the leach yield because we do not sample the ore before constituting the heap. But by inventory of the solutions, the yield is estimated at about 70–80%.

T.S. ARY: Did you say you did not sample the ore before it went into the heap?

P. MICHEL: We did not.

T.S. ARY: Did you have an estimate as to what amount of uranium went into the heap?

P. MICHEL: Yes, we have an estimate of the uranium content of the ore because it was under the cut-off grade. If we suppose that it was at the middle of the scale, we can say that we have recovered something like 70–80% of this uranium. This is just an estimate by inventory of the solution obtained.

T.S. ARY: So you really do not have a good idea of the economics that are involved?

P. MICHEL: We can estimate the economics involved only by considering how much uranium we have recovered and what are the operating costs and the fixed costs.

T.S. ARY: In Gabon, COMUF makes a semi-concentrate of magnesium uranate. Do you make this particular product in order to satisfy the feed for the plant in France?

P. MICHEL: No. I think this middle product was made because, at the time when the plant in Gabon was built, it was wiser to have a very simple process. At present, the feasibility of a

solvent extraction unit or perhaps an ion exchange unit is studied to be able to make an international grade product. It is intended to have such a unit in operation in the middle of 1977.

J. CAMERON: I should like to draw your attention to the 1966 IAEA panel¹ where heap leaching of uranium ores at Urgeiriça, Portugal, was described by de Lacerda. The actual operation was done by the company I worked for in Portugal until 1962. It may be of interest that this was a normal mine like yours in France with a nearly vertical vein structure and mined in the normal way. The ore was sorted radiometrically on the surface, high-grade ore going to the normal treatment plant, low-grade ore being piled up in heaps. This was probably where the first bacterial leaching was identified in 1953–54. After 1962, a point was reached where there were no more reserves in that mine, and the Junta de Energia Nuclear started leaching the old stopes and recovering uranium from the backfill which had come off the stope walls. They put acid in on one level, letting it come down through channels in the fill and using explosives to break up the stopes. The operation gradually proceeded up the mine, ruining it, but taking out just about all the uranium that could be got.

K. PINKAS: What is the uranium concentration in leach solutions for in-situ leaching, obtained by means of ion exchange or any process, and what kind of ion exchange do you use in this case?

P. MICHEL: For in-situ leaching in an old mine we obtained solutions containing about 100 mg U per litre. The solutions are sent to the nearest uranium plant and are processed on ion exchange columns.

¹ Processing of Low-Grade Uranium Ores (Proc. Panel Vienna, 1966), IAEA, Vienna (1967).

ПЕРЕРАБОТКА БЕДНЫХ УРАНОВЫХ РУД В СССР

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Abstract—Аннотация

PROCESSING OF LOW-GRADE URANIUM ORES IN THE USSR.

The paper is a further contribution to the study of the technology of uranium ore processing in the USSR, including that of low-grade ores. It describes a radiometric method of uranium ore enrichment, the use of autoclaves in processing technology, underground leaching and the sorption and extraction of uranium from pulps and solutions. It emphasizes the scientific and technical level achieved in the USSR, which has enabled a wide variety of problems in the processing of uranium raw materials and in environmental protection to be effectively solved.

ПЕРЕРАБОТКА БЕДНЫХ УРАНОВЫХ РУД В СССР.

Доклад посвящен дальнейшему рассмотрению вопросов технологии переработки урановых руд в СССР, и в том числе бедных урановых руд. Дается описание радиометрического метода обогащения урановых руд, применения автоклавов в технологии переработки, подземного выщелачивания, а также сорбционного и экстракционного извлечения урана из пульпы и растворов. В докладе подчеркивается достигнутый в СССР научный и технический уровень, позволяющий эффективно решать самые разнообразные задачи в области переработки уранового сырья и предотвращения загрязнения окружающей среды.

ВВЕДЕНИЕ

Для удовлетворения возрастающих потребностей в уране возникает необходимость расширения мощностей по переработке руд и вовлечения в эксплуатацию различных видов урановых сырьевых материалов, в том числе бедных, упорных и комплексных руд. Вопросы переработки урановых руд в СССР, включая извлечение урана из бедных руд, подземное выщелачивание и комплексное использование урансодержащих руд, освещались в докладах, представленных СССР на совещании экспертов [1,2] и на симпозиуме [3-5], которые проводились Международным агентством по атомной энергии соответственно в 1966 и 1970 гг.

Возможность вовлечения в переработку бедного уранового сырья будет зависеть от прогресса в технологии переработки урановых руд и развития новых направлений. Поэтому обмен опытом и достижениями в данной области представляет значительный интерес.

Настоящий доклад посвящен дальнейшему рассмотрению вопросов технологии переработки урановых руд в СССР, и в том числе бедных урановых руд.

РАДИОМЕТРИЧЕСКОЕ ОБОГАЩЕНИЕ УРАНОВЫХ РУД [6-9]

Одним из методов механического обогащения, имеющим наибольшее практическое значение применительно к урановым рудам, является метод автоматической сортировки, основанный на использовании естественной радиоактивности.

Радиометрическое обогащение может применяться как единственный метод предварительного обогащения, так и в комбинации с другими методами обогащения и технологическими процессами, например с гравитационными процессами, флотацией, кучным выщелачиванием и другими.

Стандартной технологической схемой радиометрического обогащения является двухстадийная сортировка:

1. Вся добытая горнорудная масса, погруженная в емкости (вагоны, автомашины и др.), направляется на радиометрическую станцию (РКС) и, в зависимости от содержания урана, разделяется на пустую породу, фабричную и богатые руды.

2. Последующая более глубокая покусковая сепарация выделенных на РКС обогащенных сортов руды (рис. 1) производится на радиометрических обогатительных фабриках (РОФ), обычно расположенных непосредственно на месторождении.

В ряде случаев непосредственно на гидрометаллургических заводах на специализированных установках производится дополнительная операция сортировки после среднего дробления.

К основным технологическим характеристикам урановых руд, влияющим на эффективность радиометрического обогащения, относятся: контрастность, гранулометрический состав и распределение урана по классам крупности, наличие корреляционной связи урана с сопутствующими полезными компонентами.

К специфическим свойствам урановых руд, определяющим возможность применения к ним радиометрического метода обогащения, относятся радиологические свойства (радиоактивное равновесие, эманирование, присутствие других радиоактивных элементов (K_{pp} , K_z), особенности распределения урановой минерализации в кусках).

На практике верхняя граница крупности сортируемых на РОФ классов составляет 300-100 мм и нижняя граница — 25-15 мм.

Мелкая фракция, не подвергаемая радиометрической сепарации, в зависимости от ее характеристики либо направляется на обогащение другими методами, либо объединяется с соответствующими продуктами радиометрической сортировки и направляется в гидрометаллургическую переработку.

Существенное различие в размерах кусков в сортируемом классе может оказывать неблагоприятное влияние на процесс радиометрической сепарации, приводить к нарушению принципа разделения по относительному содержанию урана и ухудшению технологических показателей.

Это отрицательное влияние ограничивается путем применения грохочения на сравнительно узкие классы по крупности при модуле шкалы грохочения 1,5-2,0 или путем использования сепараторов, производящих сортировку с учетом веса кусков.

Как правило, обязательным элементом технологических схем радиометрического обогащения является операция промывки или обдирки машинных классов перед их поступлением на сепараторы, что позволяет в ряде случаев в 1,5-2 раза увеличить выход отвалных хвостов.

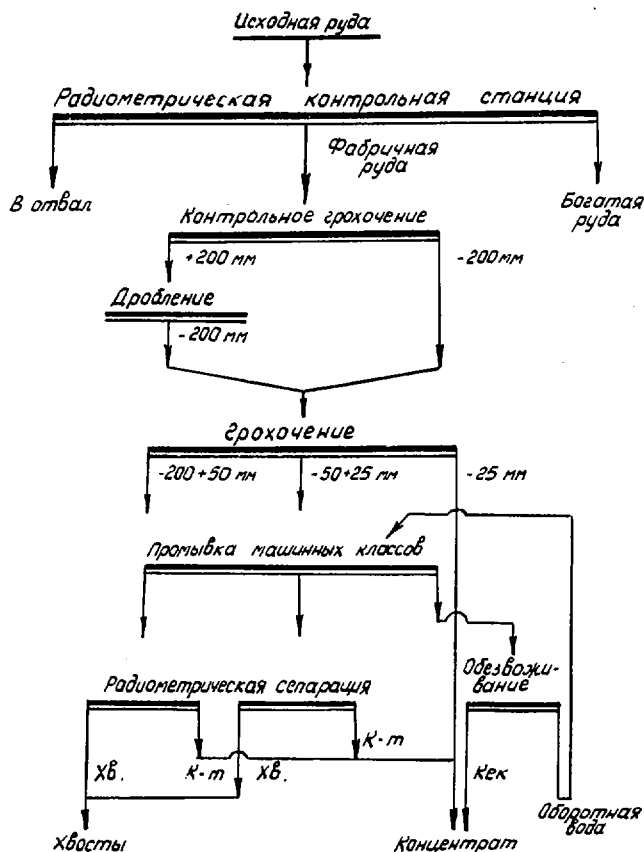


Рис.1. Типовая технологическая схема радиометрического обогащения руд.

В тех случаях, когда сортировка осуществляется с целью выделения и сопутствующего элемента, на качество разделения, кроме рассмотренных выше факторов, определяющее влияние оказывает степень постоянства корреляционной связи между содержанием основного и сопутствующего элементов.

Радиометрическая сортировка радиоактивных руд осуществляется на сепараторах, конструктивное решение которых в разных странах различно, но все конструкции сепараторов имеют одни и те же узлы: питатель, подающее устройство, радиометр, разделяющий механизм, вспомогательные устройства.

Различия в конструктивном исполнении датчика, размерах и числе используемых в датчике счетчиков (кристаллов), различия в расстояниях от датчика до куска, различия в степени экранировки датчиков от излучений, регистрируемых от рудных кусков, и от посторонних радиоактивных излучений (фона), а также различие естественных флуктуаций

гамма-излучения приводят к тому, что на датчик воздействует та или иная доля общей радиации, излучаемой куском при том или ином соотношении между полезным сигналом и фоном. В итоге рудосортировочная аппаратура не точно определяет содержание в данном рудном куске. Это определение в условиях автоматической сортировки носит не достоверный, а вероятностный характер.

В связи с этим граница содержания полезного компонента, по которому данный автомат производит разделение рудных кусков, всегда в той или иной степени размыта и точнее следует говорить о полосе или интервале разделения. Чем уже интервал разделения (т.е. интервал содержаний), в пределах которого куски с той или иной вероятностью могут направляться в оба конечных продукта, тем лучше разрешающая способность автомата.

Для обеспечения большой точности разделения стремятся увеличить долю излучения, регистрируемого от рудного куска, в суммарной величине регистрируемого излучения за счет повышения чувствительности датчиков радиометра, увеличения времени измерения рудного куска и удлинения геометрии измерения, снижения уровня фона.

Количественно разрешающая способность сепаратора представляет разность между относительными содержаниями полезного компонента в рудных кусках равного веса, отбираемых при одной и той же настройке в одноименный продукт (концентрат или хвосты) с вероятностью 95 и 5%.

Если обозначить через α_{95} содержание полезного компонента в рудном куске, отбираемом в концентрате с вероятностью 95%, а α_5 — содержание в куске, отбираемом с вероятностью 5%, то разрешающая способность

$$Z = \alpha_{95} - \alpha_5 \quad (1)$$

Порог разрешающей способности определяется выражением

$$Z = \frac{330 \sqrt{n_{\Phi} t} + 272}{g s t} \quad (2)$$

где n_{Φ} — скорость счета фоновых импульсов, имп/с;

t — время измерения, с;

g — вес рудного куска, г.

Разрешающая способность сепаратора в зависимости от крупности сортируемого класса изменяется от 0,1-0,8% на мелком классе и до 0,02-0,04% на крупном, что вполне достаточно для качественной сортировки большинства радиоактивных руд.

Схема сепаратора "Гранат" изображена на рис.2.

Производительность сепараторов, используемых в настоящее время для переработки классов -200 -50 мм и -50 -25 мм, составляет 40-60 и 10-15 т/ч. На классах -25 -15 мм и менее, в зависимости от принятого режима сортировки, производительность колеблется от 20 до 2-5 т/ч.

Действующие в Советском Союзе урановые радиометрические обогатительные фабрики выводят в отвал до 35 и более процентов отвальных хвостов (без учета отвалов, выделяемых на РКС при рудниках).

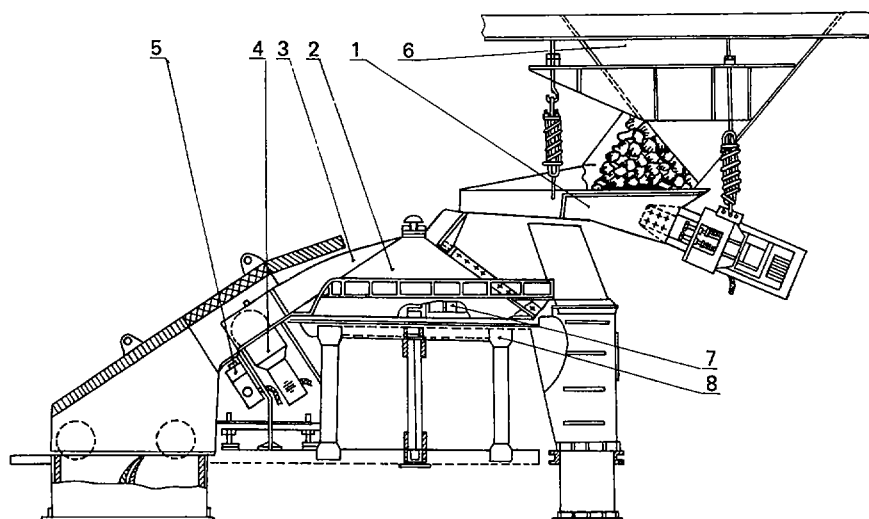


Рис.2. Схема РСА "Гранат": 1-вибропитатель; 2-конус; 3-направляющая спираль; 4-датчик импульсов (радиометр); 5-разделяющий механизм; 6-бункер; 7-электропривод конуса; 8-рама.

Из бедных руд выход отвальных хвостов составляет 40-60%.

Дальнейшее снижение стоимости сортировки и улучшение технологических показателей возможно путем применения новых, более совершенных высокопроизводительных сепараторов и освоения методов, основанных на использовании искусственной радиоактивности руд.

Промышленной практикой, полупромышленными испытаниями, лабораторными исследованиями и теоретическими расчетами доказана возможность эффективного применения радиометрического обогащения к большинству бедных урановых руд.

ПРИМЕНЕНИЕ АВТОКЛАВОВ В ТЕХНОЛОГИИ ПЕРЕРАБОТКИ УРАНОВЫХ РУД

В СССР проблему переработки бедных, упорных и комплексных руд в целом ряде случаев успешно решают, используя автоклавную технику. Применение повышенных температур и давлений, наряду с наиболее дешевым из окислителей — кислородом воздуха, позволяет организовать рентабельную переработку рудного сырья и получить высокое извлечение урана при сокращении расхода реагентов (например, серной кислоты) и энергетических затрат (пар).

В СССР разработана, серийно изготавливается и успешно применяется в промышленности эффективная автоклавная аппаратура, обеспечивающая проведение процессов окислительного выщелачивания урана в широком диапазоне температур, давлений и концентраций реагентов.

В зависимости от коррозионных свойств пульпы и парогазовой смеси при повышенных температурах в качестве конструкционного материала

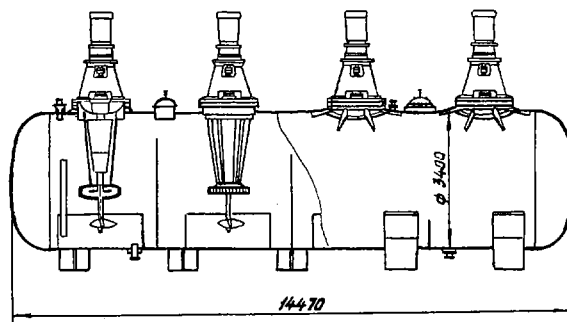


Рис.3. Автоклав с механическим перемешиванием пульпы.

для изготовления корпуса, перемешивающих устройств и других деталей, соприкасающихся с агрессивной средой, используют нержавеющие стали (X18H10T, X17H13M2(3)T и др.), титан (BT1-O) или биметалл -Ст.3 - нержав. сталь (только для корпуса). Положительно зарекомендовала себя и защита корпуса автоклава футеровкой (кислотоупорный кирпич по андезитовой плитке и полиизобутилену).

В отечественной практике для выщелачивания урана получили распространение автоклавы двух типов - горизонтальные четырехкамерные с механическими перемешивающими устройствами (рис.3) и, в последнее время, вертикальные с пневматическим перемешиванием пульпы (рис.4).

Промышленные горизонтальные автоклавы имеют емкость 125 м³ (коэффициент заполнения - 0,8) и габариты - 3,6 X 13 м. Перемешивающие устройства автоклава - 2-ступенчатые турбинные мешалки - аэраторы консольного типа [10].

Каждое перемешивающее устройство засасывает и диспергирует в пульпе около 300 м³ газа (на 1 м³ пульпы). Мощность индивидуального электропривода 75 кВт. Уплотнение вала перемешивающего устройства торцового типа (пара трения: графит-хастеллой), охлаждаемое водой. Защита корпуса автоклава от истирающего действия пульпы достигается установкой броневых плит под мешалками (если корпус не футерован). Наружная поверхность корпуса имеет теплоизоляцию.

Среднее время безотказной работы аппарата около 3000 ч.

Как показала практика работы горизонтальных автокладов, турбинная мешалка-аэратор является эффективным перемешивающим устройством, обеспечивающим высокий коэффициент использования кислорода. Однако применение подобного типа перемешивающих устройств повышает стоимость изготовления и эксплуатации автоклава. Так, например, 40% стоимости изготовления автоклава емкостью 125 м³ приходится на его перемешивающие устройства.

Этих недостатков лишены автоклавы вертикального типа с пневматическим перемешиванием пульпы. Отсутствие вращающихся перемешивающих устройств делает их конструкцию проще, облегчает вес аппарата, снижает его металлоемкость и стоимость, облегчает ремонт автоклава. Кроме того, уменьшаются энергетические затраты, так как удельный расход электроэнергии в этом случае на 60% ниже, облегчается ремонт автоклава.

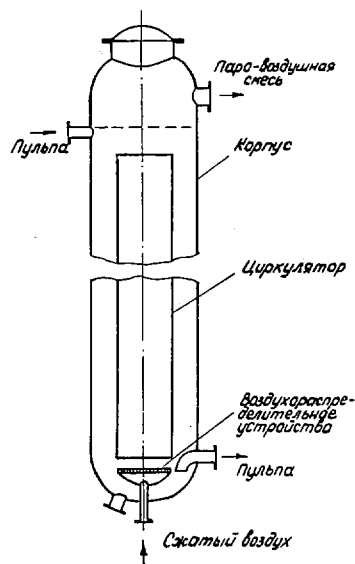


Рис. 4. Схема автоклава с пневматическим перемешиванием пульпы.

Вертикальный автоклав с пневматическим перемешиванием пульпы фактически представляет собой пачук емкостью 100–200 м³, рассчитанный на работу при повышенных температурах (до 180°C) и давлениях (до 16 атм). Габариты автоклава полезной емкостью 100 м³: диаметр – 3 м, высота – 18 м. По центральной оси аппарата расположена аэролифтная труба, служащая для циркуляции пульпы (при необходимости может быть выполнена в виде теплообменника). Воздух поступает снизу через распределительное устройство – перфорированную плиту (отверстия диаметром 5 мм) с колпачками, установленное под аэролифтной трубой. Расход воздуха на перемешивание (при плотности пульпы 1,6–1,8 т/м³) – 15–20 нм³/м³ пульпы. Наружная поверхность корпуса автоклава для снижения тепловпотерь имеет изоляцию.

Применение процесса окислительного выщелачивания в автоклавах позволяет осуществлять эффективную переработку уранового сырья.

ПОДЗЕМНОЕ ВЫЩЕЛАЧИВАНИЕ УРАНОВЫХ РУД

За последние годы в СССР для извлечения урана из бедных руд во все больших промышленных масштабах находит применение метод выщелачивания полезного компонента из руды на месте ее залегания. Уран в этом случае извлекается из недр на поверхность не в виде руды, а в виде раствора.

Метод подземного выщелачивания позволил уменьшить капитальные затраты на организацию производства, снизить стоимость получения урана и существенно улучшить условия труда. Это дало возможность

вовлечь в переработку забалансовые руды месторождений, отработанных обычным методом механической добычи, целики и крылья балансовых рудных тел, а также вовлечь в переработку месторождения, залегающие в сложных горногеологических условиях, расположенные в отдаленных географических районах, и обрабатывать локальные рудные залежи небольшого масштаба.

Метод подземного выщелачивания в промышленном масштабе осуществляется по разным технологическим схемам: с помощью скважин, пробуренных с поверхности в проницаемых обводненных месторождениях пластового типа, и в горных выработках, пройденных в месторождениях, сложенных массивными, плотными горными породами.

Подземное выщелачивание массивных пород требует проведения комплекса подготовительных горных работ, необходимых для разрушения массива и магазинирования отбитой крупнокусковой руды в подземные блоки.

Выщелачивание проницаемых месторождений не требует проведения горных работ для проходки горных выработок и разрушения руды, так как подача выщелачивающего и откачка урансодержащего товарного раствора осуществляются через скважины.

В настоящее время в СССР подземное выщелачивание представляет собой самостоятельный химико-технологический процесс.

Процесс подземного выщелачивания урана из проницаемых руд пластовых месторождений состоит из следующих стадий:

- 1) подачи выщелачивающего раствора в пласт с целью обеспечения его контакта с частицами рудной массы, находящейся в пласте;
- 2) выщелачивания урана в процессе движения раствора вдоль пласта;
- 3) выдачи раствора на поверхность для извлечения из него урана (методами ионного обмена или экстракции). Растворы после выделения из них урана доукрепляются выщелачивающим реагентом и снова направляются в пласт на выщелачивание.

Для правильной организации технологического процесса подземного выщелачивания обычно проводится ряд предварительных исследований:

- технологическое опробование кернов на моделях с целью определения основных технологических и технико-экономических показателей (извлечение, расход реагентов, продолжительность процесса и т.д.);
- геологическое и гидрогеологическое изучение обстановки на месторождении с целью определения геометрии рудного тела, его мощности, условий залегания, водоносности пласта и окружающих пород;
- определение скорости и направления естественного водного потока в пласте.

Выщелачивающими реагентами могут быть растворы кислот или соды.

Извлечение урана в раствор зависит от вещественного состава руды, выбранного реагента и кинетики процесса. Обычно оно достигает 70-90%.

Подземное выщелачивание массивных руд, требующих предварительного разрушения и магазинирования, осуществляется по методу капиллярного выщелачивания.

Раствор просачивается через слой руды и собирается на днище блока, оттуда насосом откачивается на поверхность для извлечения из него урана. Отработанный раствор после доукрепления реагентом возвращается для повторного выщелачивания.

Разрушение рудного массива осуществляется буро-взрывными работами.

Крупность дробления определяется составом рудовмещающих пород, их плотностью, пористостью, текстурой и структурой рудных монолитов.

По интенсивности и полноте извлечения урансодержащие породы можно расположить в следующий ряд: фельзит-порфир > гранит > аргиллит > базальт > трахидацит.

Дробление массива должно быть проведено таким образом, чтобы в руде преобладали классы -100+20 мм, а содержание классов +150 мм не превышало 15-25%.

При выщелачивании таких руд достигается извлечение урана до 75-90% за 6-9 месяцев. Наиболее экономично обрабатывать таким способом ореолы забалансовых рудных тел в отработанных рудных месторождениях.

Опыт промышленной эксплуатации показывает, что методом подземного выщелачивания могут быть обработаны различные виды бедных урановых руд.

СОРБЦИОННОЕ И ЭКСТРАКЦИОННОЕ ИЗВЛЕЧЕНИЕ УРАНА ИЗ ПУЛЬП И РАСТВОРОВ

Новые возможности в решении проблемы переработки бедных и комплексных руд открывают сорбционные процессы, в основе которых лежат физико-химические закономерности ионного обмена.

Неоспоримые преимущества таких процессов обуславливаются как агрегатным состоянием зернистых ионообменников, что позволяет легко проводить процесс разделения, так и высокой обменной емкостью большинства типов смол. Эти обстоятельства еще в начале пятидесятых годов позволили осуществить процесс сорбции из пульп, который стал доминирующим в урановой промышленности СССР и играет важнейшую роль в решении проблемы переработки бедных руд.

Схема аппаратного оформления процесса сорбционного извлечения урана из плотных пульп показана на рис.5 [11].

Промышленные марки выпускаемых в СССР ионитов, содержащие как слабокислые, так и сильноосновные обменные группы, могут быть использованы практически для любых, в том числе и сложносолевых систем.

Создание пористых ионообменников, обладающих высокими кинетическими свойствами, повышенной сорбционной способностью и необходимой прочностью, привело к расширению областей эффективного использования ионообменных процессов.

Создание новых типов ионитов — амфолитов позволило осуществить комплексное извлечение сопутствующих элементов.

В СССР созданы ионообменные смолы для сорбции из пульп и растворов и освоено производство сильноосновных анионитов в гелевой форме АМ, АМП, ВП-1А, ВП-3А и на той же основе макропористых анионитов АМ(п), АМП(п) и ВП-1А(п), би- и полифункциональных анионитов АМ-2Б, ЭДЭ-10п, АН-2Ф, среднеосновных анионитов АМ-3, ВП-1П(п), а также весьма перспективных карбоксильных амфолитов АМК, АМК-2, ВПК и различных фосфор- и фосфор-азотсодержащих ионитов (амфолиты АФИ-5, АФИ-7, ВПФ-1 и ВПФ-2).

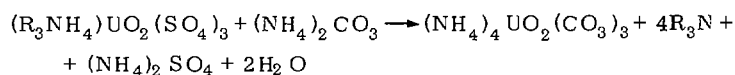
Сравнительная оценка процессов сорбции и экстракции позволила определить наиболее эффективные и оптимальные области применения каждого из этих процессов. Сорбционное извлечение урана из пульпы обычно сочетается с экстракционной переработкой десорбционных растворов.

В качестве экстрагентов используются соединения, содержащие активные группы $P=O$, $\equiv N$, и др. (трибутилфосфат, фосфиноксид, ди-(2-этилгексил) фосфорная кислота, триалкиламин).

В зависимости от солевого состава растворов и поставленной задачи может быть выбран соответствующий экстрагент.

Для реэкстракции наиболее целесообразным является метод, позволяющий получать непосредственно из органической фазы соли урана.

В общем виде реакция может быть представлена так:



Полученное соединение легко может быть превращено в окислы урана.

Создание аппаратов большой производительности позволяет осуществить процесс экстракции непосредственно из рудных растворов.

Два типа экстракторов по-прежнему занимают доминирующее положение в аппаратном оформлении процессов экстракции — это смешительно-отстойные экстракторы и экстракторы колонного типа.

Основной принцип дальнейшего усовершенствования экстракторов — определение оптимальных условий перемешивания, т.е. создание определенного равновесия в системе диспергация-коалесценция.

Значительное снижение капитальных и эксплуатационных затрат может быть достигнуто в случае использования процесса экстракции непосредственно из плотных пульп и использования процесса неводного выщелачивания. Однако эти работы не вышли еще из стадии лабораторных и стендовых испытаний.

Новые возможности в гидрометаллургии открываются при создании процессов, сочетающих преимущества сорбционных и экстракционных методов. Такими комбинированными методами являются использование импрегнированных органическим растворителем пористых гранул и процессы десорбции твердых ионитов жидкими ионитами или нейтральными экстрагентами.

В заключение следует подчеркнуть, что достигнутый в настоящее время в СССР научный и технический уровень позволяет эффективно решать самые разнообразные задачи в области переработки уранового сырья и предотвращения загрязнения окружающей среды.

Широкие возможности в деле вовлечения в переработку бедного урансодержащего сырья открываются при извлечении урана в качестве побочного продукта или совместного продукта в сочетании с другими полезными компонентами. В этом направлении СССР располагает большим производственным опытом в области извлечения урана и других ценных компонентов из фосфатного сырья, а также в области комплексного использования урано-молибденовых руд.

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DISCUSSION

F.R. HARTLEY: I have two questions. First, is the radiometric sorting equipment a single-stream or multiple-stream type equipment? Second, could you give some indication of the size distribution, or the distribution of the uranium within the different size fractions? Do you find that the ore being treated has in fact a quite distinct cut-off at a given size so that the tailings are substantially lower in uranium than the concentrate?

G.F. IVANOV: The radiometric sorting equipment used in industry is a multiple-stream type equipment. Very often, the greater the size of the ore fractions, the less uranium is contained, so that fractions of -25 or -15 mm are separated from the ore before radiometric sorting and added to the concentrate. By means of radiometric sorting, tailings with very low uranium content may be separated.

F.R. HARTLEY: Is the machine itself handling a single stream and are the particles put in a line, or do you have a multiple stream that is being sorted while passing through the machine? From the illustration it would appear that it is a single-line machine with a cone distributor.

G.F. IVANOV: Figure 2 shows the distributing cone, which lays the pieces of ore in lines. They are then sent to individual detectors by means of which the sorting of the ore is achieved. So the process is in fact a multiple-stream one.

W.R. FLÖTER: From our experience, there is some danger in using radiometric sorting. Because of airborne dust there is precipitation on the lamps or on the detector and so there are several discharges into the wrong product. What do you do to prevent this?

G.F. IVANOV: To prevent this, we give a preliminary washing to the ore.

W.R. FLÖTER: So you are sorting wet ore?

G.F. IVANOV: Yes. This is not a wet process but the ore is washed to remove dusty particles from the pieces. This helps to achieve better results.

W.R. FLÖTER: Do you also wash or spray the apparatus?

G.F. IVANOV: The apparatus is also periodically washed.

H.E. JAMES: May I ask you to explain the resin-in-pulp system shown in Fig.5. What kind of mechanism is used for the transfer of resin from the various tanks?

G.F. IVANOV: The resin is transferred by means of air lifts and the separation of resin from the transferring pulp is achieved by means of screens. The air lifts are shown on the right-hand section of Fig.5.

STUDIES ON THE RECOVERY OF URANIUM FROM LOW-GRADE ORES IN INDIA

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Abstract

STUDIES ON THE RECOVERY OF URANIUM FROM LOW-GRADE ORES IN INDIA.

Known uranium reserves in the \$15/lb U_3O_8 category, even when augmented by future discoveries, are insufficient for the projected demand. Since India has only about 3000 t uranium in the \$10–15/lb U_3O_8 category, investigations were carried out to utilize the available para-marginal and low-grade ores – chlorite schists, amphibolites, carbonate ores, clays and quartzites – analysing between 0.027 and 0.08% U_3O_8 . In addition, tests were undertaken on the technical and economic feasibility of recovering uranium as a byproduct from the copper flotation tailings and phosphorites. Since samples of quartz-chlorite schists from the Singhbhum district, Bihar, analysing about 0.03% U_3O_8 , yielded low economic recoveries by using ore dressing and hot agitation leaching techniques, heap and bacterial leaching tests were conducted. Studies also showed that the ores harbour active *Ferrobacillus ferrooxidans* and that the addition of 9K nutrients helps to reduce acid consumption and leaching time and increase uranium solubilization. These tests also confirmed that periodic withdrawal of leach liquor increases uranium leachability when compared to continuous percolation. Studies on 10-mesh samples of amphibolites from Inderwa, Bihar, (0.08% U_3O_8) showed that only 32.8% recovery could be obtained by wet tabling and 85% by agitation leaching, while static leaching tests yielded 81% recovery in 24 hours of contact time. Similar tests on calcareous phyllites (0.05% U_3O_8) with 30 kg/t Na_2CO_3 and 8 kg/t $NaHCO_3$ yielded 86% uranium leachability at ambient temperature. Biogenic uraniferous clay from Udaisagar (0.029% U_3O_8) yielded 43.3% uranium recovery using 1000 l/t of neutral water for 6 h. Although hot agitation leaching indicated 94% leachability from hard quartzites (0.06% U_3O_8), percolation leaching tests were conducted in view of the small, pockety nature of the deposit. Results showed that 81% uranium could be recovered in 24 days. Although preliminary ore dressing studies on tailings obtained from the copper flotation (0.013% U_3O_8) at Surda established the technical feasibility and yielded a concentrate analysing 0.063% U_3O_8 at 66% recovery, recent tests on the tailings from the copper concentrator indicated only 48% recovery at a grade of 0.112% owing to decrease in the feed grade. Studies on the utilization of large-capacity gravity machines and selective mining of uranium-rich copper lodes may render this source economic. Preliminary studies on a phosphorite sample containing 22.0% P_2O_5 and 0.04% U_3O_8 from the Mussorie area in Uttar Pradesh on calcination followed by scrubbing yielded a sand enriched in P_2O_5 values (33.7% P_2O_5 at 92.5% recovery) but not in U_3O_8 values. Bulk tests conducted on large tonnages of sample confirmed the data obtained in the laboratory tests. In two of the places more than thousand tonnes ore were treated by heap leaching and ion-exchange methods and the contained uranium was recovered as NaDU cake. While large high-grade ores can be treated by any of the known and proved extraction techniques, small, pockety deposits and large low-grade deposits require simple and cheap methods. Ore dressing methods do not hold promise for treating low-grade ores in the present context of cost and conservation, although they can be used for byproduct recovery. The conventional agitation leaching method is also not suitable for treating low-grade ores because of the high capital input, high process cost and relatively high boundary imposed on the cut-off grade. Heap and bacterial leaching and solution mining techniques are not yet developed as independent process techniques, and are riddled with many plant engineering problems. Bacterial leaching, although encouraging, requires careful study. Large-scale experimentation is needed before it can be accepted as an independent process technique.

1. INTRODUCTION

The anticipated cumulative world demand for uranium by the year 2020 is 10 to 12×10^6 t [1] as against the reasonably assured and the additional estimated resources of 3.083×10^6 t [2]

in the categories of $< \$ 10/\text{lb}$ and $\$ 10\text{--}15/\text{lb}$ U_3O_8 . Since the annual mean discovery rate of new deposits is only 65×10^3 t, there is a considerable gap between the projected demand and the known supply. In India we have 3.384×10^3 t of reasonably assured resources and another 846 t of additional estimated resources in the price range of $\$ 10\text{--}15/\text{lb}$ U_3O_8 [1]. Though some of our recent finds are anticipated to be of better grade, resources available in our para-marginal (0.05% U_3O_8) and low-grade ores (0.03% U_3O_8) require exploitation. Large inputs of research and development effort are called for in this direction. Investigations were, therefore, carried out on samples of ores from Narwapahar, Bhatin, Keruadungri and Nandup in the Singhbhum and Inderwa in the Hazaribagh districts of Bihar, Umra and Udaisagar in the Udaipur district of Rajasthan, and Chhinjra in the Kulu district of Himachal Pradesh.

Recovery of uranium as a byproduct or co-product of metallic and non-metallic ores is of considerable importance. As uranium occurs associated with phosphorites and copper ores in India, investigations were conducted to recover uranium from Surda copper ore in the Singhbhum district of Bihar and from phosphorites from Chamansari in the Saharanpur district of Uttar Pradesh.

2. TESTS ON LOW-GRADE ORE SAMPLES

2.1. Quartz-chlorite schists

Samples of quartz-chlorite schists from Narwapahar, Bhatin, Keruadungri and Nandup, analysing 0.03, 0.033, 0.035 and 0.027% U_3O_8 respectively, were studied for uranium recovery. Mineralogically, the samples consist of quartz (40–52%), chlorite (38–44%), apatite (2–5%), magnetite (5–10%), opaques (3–3.5%), together with accessory amounts of amphiboles, tourmaline etc.

2.1.1. Ore dressing and agitation leaching tests

Physical beneficiation studies carried out on these samples by wet tabling at 55% –200 mesh size yielded concentrates analysing 0.07, 0.088, 0.10 and 0.09% U_3O_8 , representing a recovery of 51.3, 72.8, 33.4 and 40% from Narwapahar, Bhatin, Keruadungri and Nandup, respectively. Hot agitation leaching tests (55°C) conducted at 60% P.D. (solids) and 60% –200 mesh yielded a recovery of 79.6, 88.7, 86.9 and 55% respectively from the above samples by using 22 kg/t H_2SO_4 and 5 kg/t pyrolusite for 12 hours.

2.1.2. Heap and bacterial leaching tests

Since ore dressing and hot agitation leaching methods did not yield economic recoveries, heap and bacterial leaching studies were conducted on samples of ores drawn from ore dumps lying stacked outside the pit mouth. Geo-microbiological studies conducted on surface and sub-surface samples and also on mine waters confirmed the presence of *Ferrobacillus ferrooxidans* and also other anaerobic and heterotrophic bacteria (Fig. 1).

For the purpose of the investigation, samples of ore were crushed to about 1 cm size and placed in sterilized glass columns to a predetermined height. One hundred to 150 l/t per week of dilute H_2SO_4 at pH 1.8 to 2.0 were then percolated three to four times daily through the column at a rate of $4 \text{ cm}^3/\text{cm}^2 \cdot \text{min}$. The volume and pH of the leach liquor was adjusted daily before recirculating the leach liquor, since any rise in the pH beyond 2.4 causes precipitation of ferric hydroxide on the mineral grains and prevents further solubilization of uranium. To avoid excessive build-up of ferric sulphate and other bacterial toxins the liquor was withdrawn after 6 days and circulation was resumed with a fresh batch of dilute H_2SO_4 after allowing a curing time of one day.

Tests were carried out simultaneously under the following conditions: (i) leaching of sterilized ore with dilute H_2SO_4 at pH 1.8 to 2.0; (ii) leaching of ore as such, to which 9K nutrients were added; (iii) leaching of ore as such, but without nutrients; (iv) leaching of ore with dilute H_2SO_4 at pH 1.8 to 2.0 after inoculation with autotrophic bacteria; and (v) leaching with the addition of pyrite as energy source in place of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ in the 9K nutrients.

2.1.3. Results

The results of these tests, given in Tables I and II, show that: (i) the indigenous population, when energized with 9K nutrients, decreases the leaching time from 84 to 56 days, (ii) leachability also increases by using 9K nutrients (Fig.2); (iii) the acid consumption was reduced from 3.8 to 1.6 kg/t in the case of Narwapahar ore; from 9.83 to 8.80 kg/t in the case of Bhatin ore and from 6.75 to 4.0 kg/t in the case of Keruadungri ore; no such reduction was noticed for Nandup ore; (iv) the use of pyrite in place of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ reduces the leachability marginally and increases the leaching time; (v) although the overall rate of uranium solubilization decreases after the fourth week in the case of bacterial leaching in comparison to percolation leaching, the solubilization of uranium is greater in the former than in the latter when the head assay at the start of the period is considered (Table II); (vi) periodic withdrawal of leach liquor helps to increase the leachability as it prevents build-up of iron when the leach liquor is continuously recycled [3]. This is confirmed by 13.9 g/l Fe and 31.4% U_3O_8 solubilized in continuous recycling against 2.5 g/l Fe and 78.7% U_3O_8 leached when weekly batch withdrawal was carried out [4]; (vii) the stack height for obtaining the optimum recovery is 75 cm.

2.2. Amphibolites

Two samples of amphibolites analysing 0.08% U_3O_8 were studied for uranium recovery. Mineralogically, the samples consist of hornblende, quartz and biotite as major minerals with accessory amounts of magnetite, apatite, calcite and covellite. Uraninite occurs free, and also as inclusions in hornblende.

Wet tabling of these samples, at 55% -200 mesh, yielded a recovery of 32.8% at a concentrate grade of 0.092% U_3O_8 . Agitation leaching tests at 60% -200 mesh size, 60% P.D. (solids) and 60°C indicated a leachability of 85% in 12 hours of leaching time using 27 kg/t H_2SO_4 and 2.5 kg/t MnO_2 .

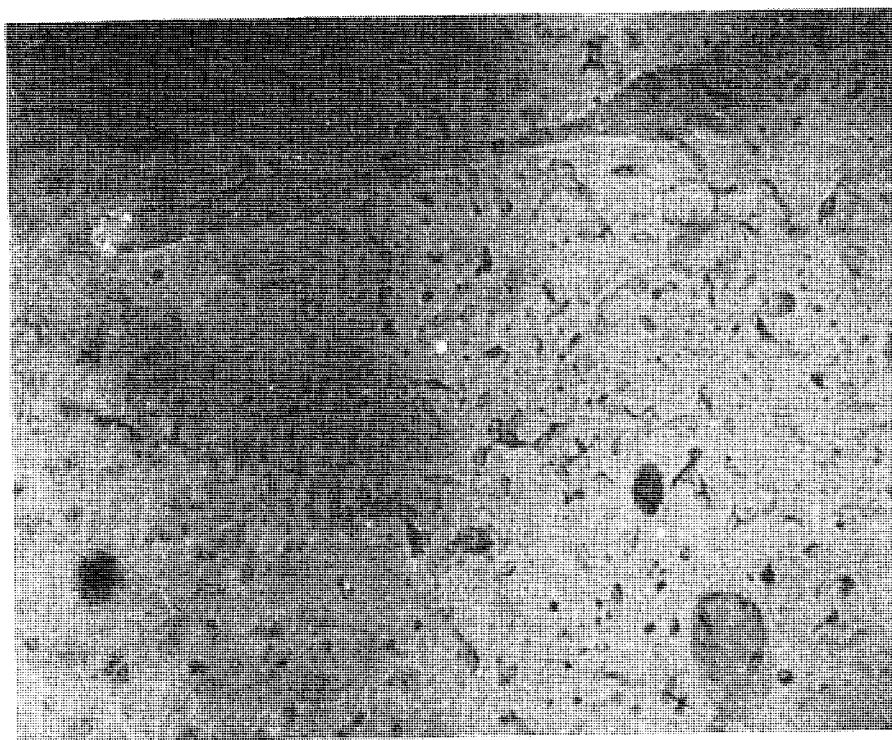
Stack leaching tests were conducted on samples of ore mixed with an oxidant and placed in a container with a filter bed of quartz pebbles. Dilute H_2SO_4 (different dilutions) was then added and the stack was allowed to cure. At the end of a predetermined leaching period the liquor was withdrawn through the filter bed and the stack washed with 200 l/t dilute H_2SO_4 at pH 2.0

Static leaching was preferred to percolation leaching, since the ore contained acid-consuming gangue minerals like calcite, which in percolation leaching caused high acid consumption owing to the removal of the calcium sulphate. In static leaching, on the other hand, the insoluble calcium sulphate remained undisturbed.

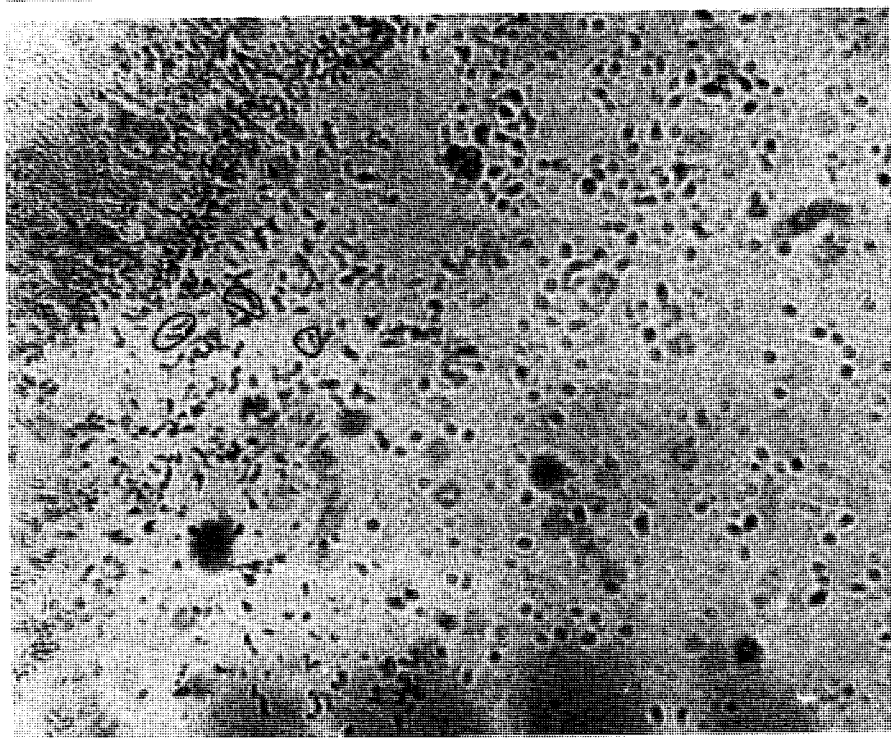
The results of tests on three samples are given in Table III. These results show that uranium leachability increases with increasing acid loading, pulp density, contact time and oxidant addition. It decreases with increasingly finer grinding owing to the increased liberation of calcite [5].

2.3. Carbonate ores

Samples of uraniferous calcareous shale, analysing 0.05% U_3O_8 , were investigated by the static leaching method. Since high pulp density (80% solids) acid leaching yielded only 33.1% leachability, alkali leaching tests using Na_2CO_3 and NaHCO_3 at ambient temperature were conducted.



(a)



(b)

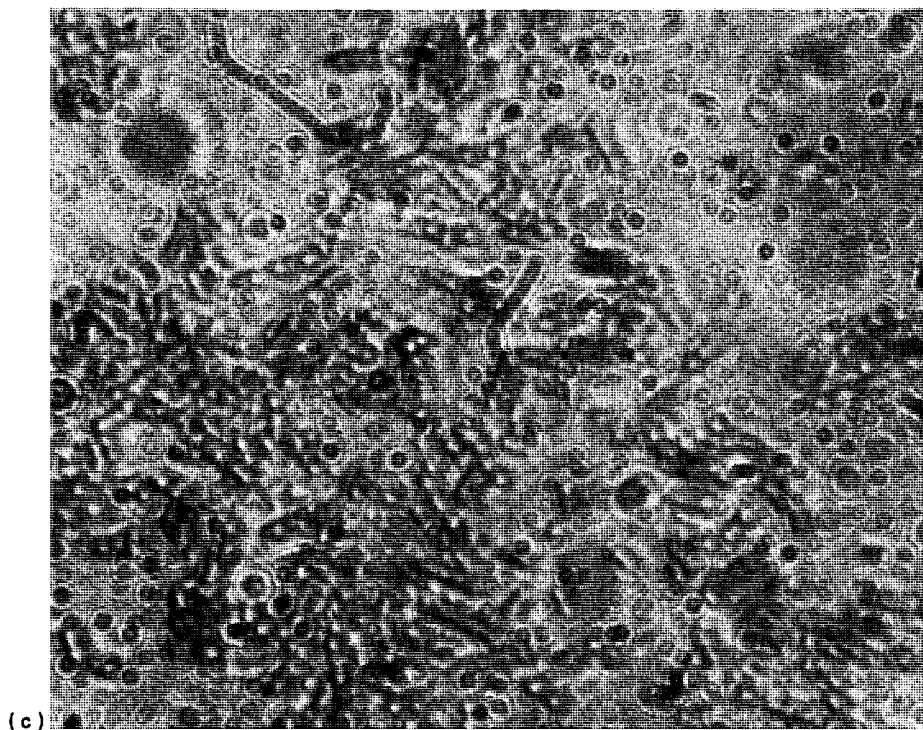


FIG.1. The presence of bacteria on various samples: (a) *Ferrobacillus ferrooxidans* from Bhatin leach liquor; (b) *Desulfovibrio desulphuricans* from Narwapahar; (c) *Bacillus subtilis* from Keruadungri.

TABLE I. HEAP AND BACTERIAL LEACHING TESTS, pH 2.0

	Time (d)	Narwapahar		Bhatin		Keruadungri		Nandup ^a	
		Acid (kg/t)	Leach (%)	Acid (kg/t)	Leach (%)	Acid (kg/t)	Leach (%)	Acid (kg/t)	Leach (%)
Control	84	3.87	69.8	9.83	60.0	6.75	62.6	—	—
	56	3.37	66.2	8.78	49.7	5.50	55.9	2.1	56.7
Ore + 9K	84	4.40	84.2	10.93	85.0	4.34	94.2	—	—
	56	1.62	77.4	8.80	66.4	4.00	91.4	2.6	63.5
Ore + 9K + T.f. ^b	84	4.40	79.8	7.54	72.7	4.8	94.2	—	—
	56	3.9	76.8	6.75	60.0	4.0	88.3	2.3	67.5 ^c
Ore + 9 K + pyrite - FeSO ₄	84	4.1	76.8	3.64	69.9	4.36	91.6	—	—
	56	3.4	67.4	3.60	65.0	3.90	83.3	—	—
Control, sterilized ore	84	3.87	68.2	9.1	59.8	5.95	59.8	—	—
	56	3.40	56.5	8.5	46.7	5.10	54.7	2.5	57.7

^a 33 days leaching time.

^b *Thiobacillus ferrooxidans* culture.

^c Leach liquor aerated.

TABLE II. RATE OF LEACHING

Period (weeks)	Leachability with respect to calculated head assay at the start of period (%)						Leachability with respect to original head assay (%)					
	Narwapahar		Bhatin		Keruadungri		Narwapahar		Bhatin		Keruadungri	
	A	B	A	B	A	B	A	B	A	B	A	B
0-2	19.8	22.8	15.1	3.2	28.9	4.9	19.8	22.8	15.1	3.2	28.9	4.9
2-4	46.1	23.1	22.1	22.1	57.3	19.2	36.7	17.9	18.8	21.0	31.2	18.3
4-6	34.3	28.9	27.3	20.2	52.2	24.5	15.0	17.1	18.1	15.4	15.4	18.8
6-8	20.6	20.0	30.0	16.6	30.1	24.0	5.0	8.4	14.4	10.1	5.9	13.9
8-10	12.8	19.7	33.5	13.0	20.0	15.0	2.9	6.6	11.3	6.6	1.8	6.6

A: leaching with nutrients; B: leaching without nutrients.

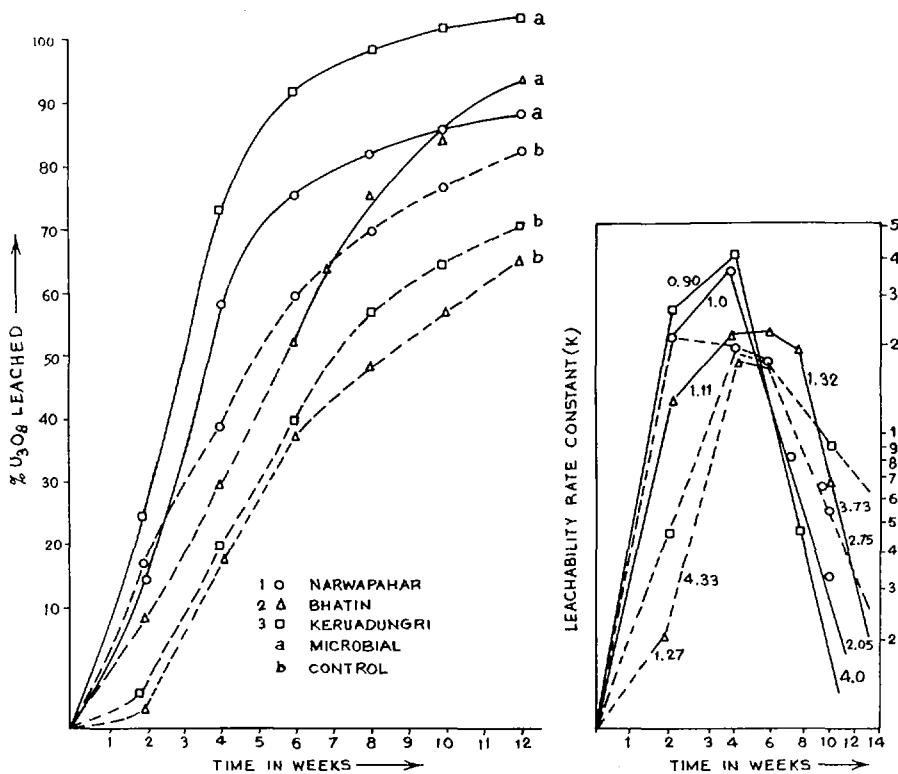


FIG. 2. Bacterial leaching of Narwapahar, Bhatin and Keruadungri ores. Feed assay: $0.032 \pm 0.003\%$ U_3O_8 .

TABLE III. STATIC LEACHING OF AMPHIBOLITES
FROM INDERWA
Feed assay 0.08% U_3O_8 ; time 24 h.

Size	Acid (kg/t) (vol.%)		Oxidant (kg/t)	Leachability (%)
$-\frac{1}{8}$ in	25.0	7.5	7 MnO_2	36.5
$-\frac{1}{4}$ in	25.0	7.5	7 $KClO_3$	71.3
-10 mesh	32.0	10.0	5 $KClO_3$	88.0
-20 mesh	25.0	7.5	7 $KClO_3$	84.3
-20 mesh	32.0	10.0	7 $KClO_3$	85.8

TABLE IV. STATIC LEACHING OF UMRA ORE
Feed assay 0.05% U_3O_8 .

No.	P.D. % (solids)	Time (d)	End pH	Na_2CO_3 (kg/t)	$NaHCO_3$ (kg/t)	Leachability (%)
1	60	30	11.0	25	—	34
2	60	30	11.2	25	6	49.6
3	70	20	11.5	25	1	58.5
4	70	20	10.2	30	7	85.9
5	70	20	10.2	30	7	86.8
6	50	20	10.2	30	7	71.8
7	60	15	10.0	35	7	61.7

As the ore contained secondary uranium minerals, no oxidant was used in these tests. The results given in Table IV show that up to 86% uranium could be solubilized in 20 days at -5 mesh size (8% -200 mesh).

2.4. Quartzo-feldspathic ores

2.4.1. Clays

Samples of kaolinized and brecciated fault gauge (0.03% U_3O_8), from Udaisagar in the Udaipur district of Rajasthan, consisting of specks of uraninite and sooty pitchblende together with chalcopyrite and pyrite in a matrix of abundant clay gauge and fragments of phyllite, quartz and feldspar, were tested for uranium solubilization.

Since physical beneficiation methods yielded only 40% recovery with an enrichment factor of 1.5, static leaching tests were conducted using dilute H_2SO_4 of 0.1–2.0 vol.% strength at different pulp densities. Results indicate that: (i) 43.3% of the contained uranium could be leached in 6 hours with 1000 l/t H_2O at ambient temperature; (ii) the addition of 4.0 kg/t H_2SO_4 increases uranium solubility to 81.2%; and (iii) neither increase in time to 72 hours nor increase in acid loading to 11.0 kg/t increases the leachability [6].

TABLE V. LEACHING OF CHHINJRA ORE
Feed assay 0.06% U_3O_8 .

Size	Time (d)		Acid (kg/t)	MnO_2 (kg/t)	$FeSO_4 \cdot 7H_2O$ (kg/t)	Leachability (%)
	Curing	Leaching				
$-\frac{1}{4}$ in	18	8	26	5	—	48.5
$-\frac{1}{4}$ in	19	8	26	5	—	58.0
— 5 mesh	11	13	26	5	—	71.5
— 5 mesh	11	13	26	5	2.0	80.0
— 10 mesh	11	13	26	5	2.0	81.0
— 10 mesh ^a	4	2	27	8	2.0	67.9
— 20 mesh ^a	28	13	26	5	—	75.0
— 20 mesh ^a	11	13	27	5	—	65.6

^a Magnetic minerals removed from the ores.

2.4.2. Quartzites

Small pockets of uranium occur in the hard, impermeable quartzites (0.06% U_3O_8) of the Chhinjra area of the Kulu district of Himachal Pradesh. Mineralogically, the ore consists of veins and veinlets of uraninite as well as replacements of quartz grains. Secondary uranium minerals and small amounts of chalcopyrite, pyrite, magnetite and haematite also occur.

Laboratory tests showed that ore dressing methods are not suitable, since a recovery of 70% only could be obtained by wet tabling at 65% –200 mesh grind. Although hot agitation leaching (60°C) tests at 70% –200 mesh yielded recoveries of 94% in 12 hours using 35 kg/t H_2SO_4 and 5 kg/t MnO_2 , this method was discarded because of the small and sporadic nature of the deposit located in an inaccessible area.

Percolation leaching tests conducted on samples of this ore (Table V) showed that:

(i) a leachability of 81% could be obtained in 24 days of leaching with 26 kg/t H_2SO_4 and 5 kg/t MnO_2 [7]; (ii) since the ore does not contain sufficient iron, 2 kg/t $FeSO_4 \cdot 7H_2O$ must be added; and (iii) as the ore is hard and impermeable, size reduction to –10 mesh is necessary to obtain optimum recovery.

Bulk tests conducted on samples of this ore confirmed the necessity of adding iron to improve recovery and reduce leaching time.

3. BYPRODUCT RECOVERY

3.1. Recovery from copper ores

Large quantities of copper ore from the Surda, Mosabani and Rakha mines in the Singhbhum district of Bihar are known to contain significant amounts of uranium, the grade varying from 0.009 to 0.017% U_3O_8 . Since the tailings obtained after sulphide flotation are a possible source of uranium, tests were conducted to assess the technical and economic feasibility of obtaining a preconcentrate.

The mineralogical composition of the Cu-flotation tails from Surda is: quartz 55.0%, chlorite 22.0%, apatite 5.0%, magnetite 3.2% opaques 12.1% and others 6.0%.

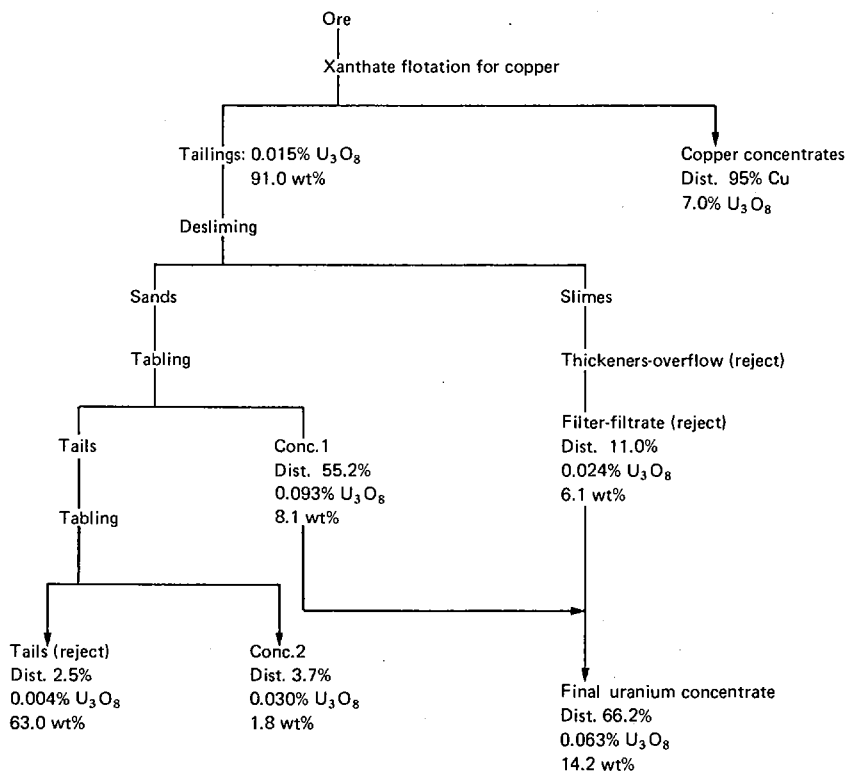


FIG.3. Uranium recovery flowsheet.

After xanthate flotation at 50% – 200 mesh (for recovery of Cu) the tailings were subjected to wet tabling (Fig.3). First results on a representative sample showed that a concentrate analysing 0.063% U_3O_8 could be obtained at 66% recovery [8].

Agitation leaching tests conducted on samples of this concentrate (68% – 200 mesh) yielded a leachability of 94% with 27.0 kg/t H_2SO_4 and 12 kg/t MnO_2 after 12 hours leaching time.

Although the above investigations suggested the technical feasibility of recovering uranium as a byproduct, recent studies on the flotation tailings from the 400 t/d copper concentrator, which has since been commissioned, yielded only a recovery of 48.5% in concentrates (6.2 wt%) analysing 0.112% U_3O_8 . This is due to a drop in the feed grade from 0.015% U_3O_8 to 0.011% U_3O_8 and a reduction in grind size from 50% – 200 mesh to 35% – 200 mesh. Therefore, static leaching studies at ambient temperature on deslimed and undeslimed flotation tailings were conducted (Table VI). These results indicate that, while recovery of uranium from the copper ore is technically feasible, it would be economic only if the comparatively uranium-rich copper lodes are selectively mined and treated.

3.2. Recovery from phosphorites

A dolomitic phosphorite sample analysing 37.0% CaO, 6.9% MgO, 22.2% P_2O_5 , 0.24% Mo, 0.23% Ni and 0.034% U_3O_8 from Chamansari in the Saharanpur district of Uttar Pradesh was

TABLE VI. STATIC LEACHING TESTS – SURDA Cu TAILS

	Feed assay (% U_3O_8)	Acid (kg/t)	Stack height (cm)	Leachability (%)	Remarks
1 A	0.06	13.5	20	41.3	Table concentrates
B	0.06	13.5	20	63.8	Table concentrates
2 A	0.009	7.0	20	53.1	Cu tails
B	0.009	7.2	20	62.3	Cu tails
3 A	0.010	7.5	20	57.4	Deslimed sand
B	0.012	7.5	20	72.4	Deslimed sand
4 A	0.012	7.3	40	70.5	Deslimed sand
B	0.012	7.5	60	61.9	Deslimed sand

A: control, B: with 9K nutrients.

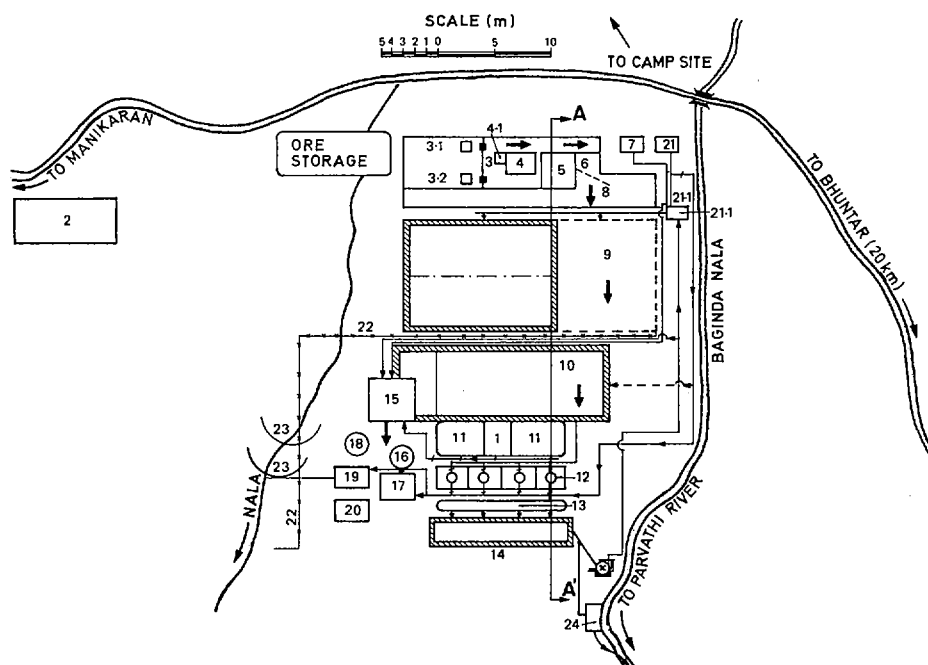


FIG. 4. Layout of the Baginda heap leaching plant. Capacity 25 t/d.

1 - Laboratory, 2 - generator shed, 3 - counter shaft, 3.1 and 3.2 - motors, 4 - jaw crusher of 12 in X 6 in, 4.1 - feeder, 5 - roll crusher of 24 in X 16 in, 6 - screen of 5 mesh, 7 - acid tank, 8 - crushed ore yard, 9 - leaching tank of 14 m X 7 m X 15 m, 10 - leach liquor tank of 69 m³, 11 - working platform, 12 - resin columns, 13 - surge tank, 14 - barren liquor tank, 15 - eluate tank of 3000 litres, 16 - NaOH tank of 200 litres, 17 - precipitation tank of 3000 litres, 18 - surge tank for eluate, 19 - filtration assembly, 20 - cake packing area, 21 - water tank, 21.1 - dilute acid tank, 22 - tailings disposal line, 23 - tailings dam, 24 - neutralization pond.

investigated for recovering uranium as a byproduct. The samples consist of calcite, chert, haematite, quartz, magnetite and apatite, set in an amorphous matrix of collophane. Uranium is very finely dispersed in the collophane.

Although no preferential concentration was possible by wet tabling, flotation using oleic acid, ethyl alcohol and diesel oil yielded a concentrate analysing 34.4% P_2O_5 and 0.045% U_3O_8 at recoveries of 79.8% and 63.3%, respectively. Calcination of the ore (29.6% P_2O_5 and 0.04% U_3O_8) crushed to -6 mm size at 900°C followed by scrubbing yielded concentrates analysing 35.7% P_2O_5 and 0.048% U_3O_8 at recoveries of 92.5% and 75.0%, respectively [9]. Thus, while physical beneficiation studies were found useful to enrich the P_2O_5 value, the uranium values could not be sufficiently upgraded.

Agitation leaching tests at 80% -200 mesh size and 60°C showed that: (i) increasing the acid loading from 360 to 720 kg/t increased the leachability of uranium from 27.0 to 66% and that of phosphorus from 23.0 to 60.0%; (ii) uranium leachability increases from 53.0% at 25% P.D. (solids) to 82% at 50.0% P.D. (solids) and, similarly, solubilization of phosphorus increases from 38.0 to 80% [10].

Static leaching tests conducted at -5 mesh size using 800 kg/t H_2SO_4 solubilized 43.1% U_3O_8 , 38.5% P_2O_5 , 80.3% Ni and 78.9% Mo, as compared with 45.4% U_3O_8 , 34.8% P_2O_5 , 64.5% Ni and 68.6% Mo solubilized with 400 kg/t H_2SO_4 .

4. BULK TESTS

Following the encouraging results obtained in the laboratory, bulk tests were conducted at two of the mine sites, Umra and Chhinjra. Uranium was recovered from the pregnant liquor by an ion-exchange method using De-acidite FF-530, suitably arranged in 4 FRP, fixed-bed columns. Uranium from the chloride eluate was then precipitated using NaOH. Figures 4 and 5 show the plant layouts. Results of these tests are shown in Table VII [11].

5. DISCUSSION

While extraction methods for treating large deposits of high-grade ore are well known, small high-grade deposits and large but low-grade deposits require considerable laboratory and pilot-plant experience before finalizing the process design. Diverse techniques are needed to exploit ores varying in mineralogical complexion and physical characteristics. Any process designed for a given ore type may not hold good for another and mineralogical variations even in the same lode are not uncommon. Since the exploitation of any deposit depends on its economic viability, it is imperative that para-marginal and low-grade ores are treated by cheaper techniques requiring lower capital input.

Physical beneficiation of low-grade ores is being attempted in several countries. Although heavy-media separation, jigging, tabling, flotation and radiometric lump or car sorting are carried out both at the pilot-plant level and in large-capacity mills, these methods have not found favour since size reduction becomes unavoidable for preconcentration. This, together with the considerable loss of uranium values in the tailings, which is too high to be neglected, renders these techniques unsuitable. Further, if the cost of uranium increases as expected, ore dressing steps, due to lower overall recoveries, become more and more uneconomic. However, ore dressing steps can be gainfully used for byproduct recovery from multi-metallic or non-metallic ores, using selective mining if necessary.

Barring a few isolated cases, uranium ores are primarily treated by the hot agitation leaching technique. The recoveries by this method are usually of the order of 90% and uranium from the pregnant liquors is extracted either by resin ion exchange or by solvent extraction techniques. Although almost all the present production is obtained by this technique, the capital required in

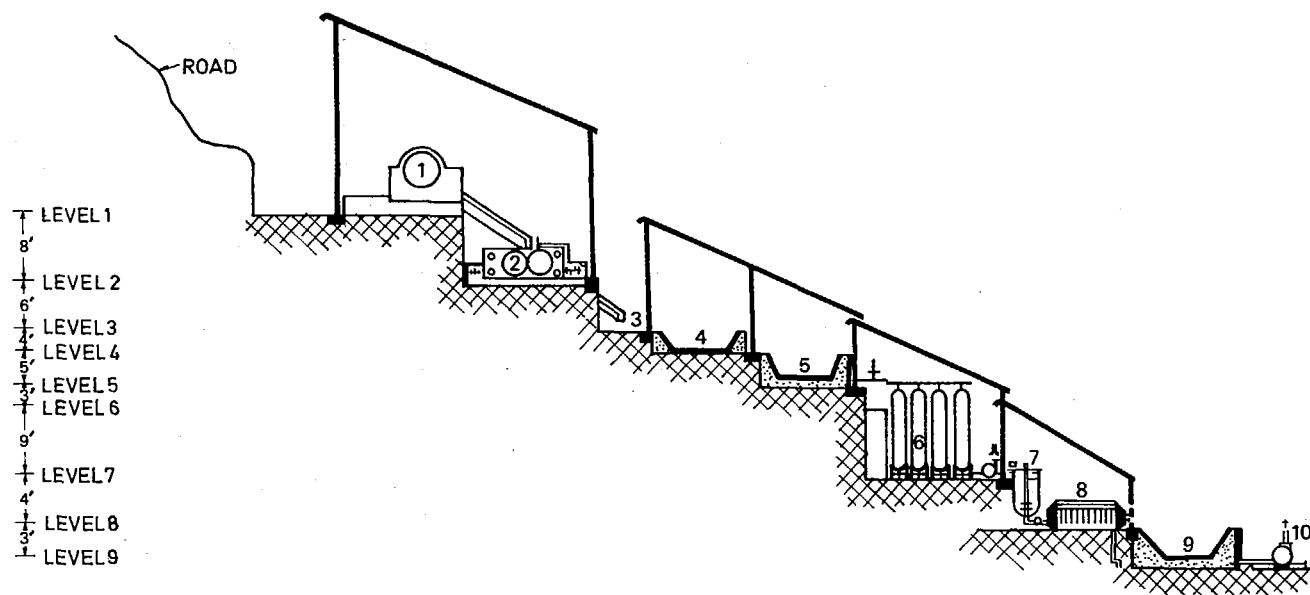


FIG. 5. Layout of the Kulu heap leaching plant — SE-NW elevation. Capacity 25 t/d.

Level 1 — water tank, ore yard, jaw crusher, acid tank; level 2 — roll crusher; level 3 — crushed ore yard; level 4 — leaching tank; level 5 — leach liquor tank, NaOH tank, Na₂CO₃/NaCl tank; level 6 — working platform for extraction unit; level 7 — resin column/countercurrent unit; level 8 — precipitation unit, filtration unit; level 9 — barren liquor tank and pump.

TABLE VII. BULK TESTS

No.	Locality	Feed assay (% U ₃ O ₈)	Leaching time (d)	Size	Amount treated (t)	Acid used (kg/t)	MnO ₂ (kg/t)	Leachability (%)	Cake grade (% U ₃ O ₈)	Recovery (%)
1.	Umra	0.03 ^a	3	-12 mm	830	27.0	—	72.3	70.1	70.5
2.	Chhinjra	0.042	38	-10 mesh	6	32.0	2.0	76.86	13.22	65.8
		0.06	75	-10 mesh	10	32.0	4.0	83.30	3.40	81.0
		0.23	24	-10 mesh	5	25.0	5.0	86.40	17.90	84.1
		0.125 ^a	40	-10 mesh	135	13.0	4.0	77.60	69.8	77.0
		0.975 ^a	80	-10 mesh	170	31.7 ^b	10.8	70.8	71.2	70.0
3.	Bhatin	0.053 ^c	40	-5 mesh	2.0	5.8	—	91.9	—	—
		0.053 ^d	39	-5 mesh	2.0	6.5	—	90.7	—	—
4.	Keruadungri	0.043 ^c	36	-5 mesh	2	6.5	—	75.0	—	—
5.	Udaisagar	0.029	2	as mined	163	4.0	—	79.0	6.5	69.0
		0.040	27	-5 mesh	—	5.5	2.0	55.0	—	—
6.	Narwapahar (bore hole core)	0.040 ^c	27	-5 mesh	—	4.0	—	59.5	—	—

^a Heap leaching and ion-exchange method; chloride-free ion-exchange barren is used.^b 3.0 kg/t FeSO₄·7H₂O added to the ore.^c Bacterial leaching using 9K nutrients.^d Chloride-free ion-exchange barren.

the main process and also in the accessory supports, such as acid plants, oxidant grinding mills, steam raising equipment, etc., renders the process uneconomic for para-marginal ores. Another limitation is the relatively high boundary imposed on the cut-off grade, due to the high mining, grinding and filtration costs. Unless research and development work is increased to design new equipment and to reduce the process cost (such as solvent in pulp or resin in pulp), low-grade ores cannot be treated by this conventional process.

Several countries are actively engaged in adopting heap leaching, bacterial leaching and solution mining techniques for extracting uranium from low-grade ores. With rising reagent and labour costs this method holds promise for the exploitation of small, pockety, high-grade deposits and large low-grade deposits, since it has the advantage of being simple and cheap. The limitations, however, are the prolonged leaching time, comparatively lower recovery of 50 to 80% and the high mining cost. Further, it may be necessary to crush the ore to about 5 mm size for obtaining competitive recoveries from the disseminated type of ores. Thus heap leaching, bacterial leaching and solution mining techniques are not yet developed as independent process techniques, and are riddled with many plant engineering problems.

Bacterial leaching, although encouraging, is not an end in itself. Careful experimentation and interpretation is required, since all the ore may not harbour indigenous bacteria and inoculation of cultures in large ore dumps may offset the cost saved on the leachants, etc. The utilization of barren liquor, attempted in bulk tests carried out in India, has proved that it is not necessary to seed bacteria or feed the nutrients continuously. It would be enough to hasten the reaction by using the nutrients in the initial batch and to use the ion-exchange barrens in subsequent cycles.

Large para-marginal deposits which do not justify a conventional plant can be treated by a judicious process mix of the different techniques.

6. RESEARCH ORIENTATIONS

Detailed investigations into the mineralogical, chemical and locational aspects of each individual occurrence and a judicious process mix may render the para-marginal and low-grade ores amenable to exploitation at competitive prices. A rough cost analysis of different unit operations shows that about 20% of the extraction cost and 30–40% of the direct cost is accounted for by milling and leaching respectively. The other major costs are for filtration and waste disposal. Research is, therefore, required to minimize these costs.

The mills as are currently operated and designed are bulky and heavy, and consume a disproportionately large portion of the power when compared to the actual power required for grinding the ore itself. Added to this is the imperfect classification of the mill discharge owing to the variation in the mineralogical characteristics of the diverse ore-gangue complexes. The need to study the behaviour of fine particles (slimes) is to be kept in view. Studies on flocculation, ore agglomeration, gravity treatment and flotation of ultrafines may enable the utilization of low-grade ores. Similarly, studies to develop and perfect large-capacity machines and to reduce reagent consumption and increase specific ore particle/reagent interaction are necessary.

In the case of dump leaching, investigations to reduce leaching time and return of the tailings to the mine (as backfill) are necessary before this method could be accepted in full.

The tailings obtained by chemical extraction methods would leave enough free acid and other toxic products. Development of cheap reagents to fix these poisonous products need be achieved early. For this also, the role of anaerobic and heterotrophic bacteria needs understanding.

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DISCUSSION

T.S. ARY: You mention that in one case you had a shale with 0.02% U_3O_8 and the uranium was leached by a bacterial process from the black shales and remobilized and deposited in the clay. What evidence do you have of this remobilization and redeposition?

K.M.V. JAYARAM: A paper on this topic has been published by the IAEA.¹ The main evidence is as follows: 1. The uranium available in the black shales surrounding the fault zone is depleted compared to the uranium in the general black shales in the area. 2. The hydro-geological studies using tracer technology have shown a migration of the waters, hydrological waters, from the black shales into the fault zone. 3. The fault zone itself contains clay which is a neutral exchanger in itself for uranium. There is a profuse growth of *Desulfovibrio desulphuricans*, which is a reducing bacterium in this zone, and a profuse growth of the oxidating bacteria in the black shale zone. Other evidence is that there is a seasonal fluctuation of uranium content in the wells around and in the shaft. During the rainy season we find that the water contains a little more uranium than during the dry season. About 40–50% of the uranium is water-soluble. When clay is put into water and left there for an hour, 50% of the uranium is extracted.

¹ JAYARAM, K.M.V., DWIVEDY, K.K., BHURAT, M.C., KULSHRESTHA, S.G., "A study of the influence of microflora on the genesis of uranium occurrences at Udaisagar, Udaipur district, Rajasthan", Formation of Uranium Ore Deposits (Proc. Symp. Athens, 1974), IAEA, Vienna (1974) 89.

T.S. ARY: I should like to point out the different outlook of the mining man and the milling man. In the USA we have a lot of promotional activity with respect to our minerals and our uranium boom, our silver boom and what have you, and it seems to me that the promoters are always trying to build mills before they find an orebody. In this particular case I would suggest that more time be spent on geology and exploration work until you find an orebody that it is worthwhile building a mill on. I do not know how extensive the geological and exploration effort has been, but my information is that there is a great deal still to be done in India. This is surely a more important matter than how to handle a 0.02% ore and whether the miners should mine it and whether you should leach it in place.

K.M.V. JAYARAM: Certainly one would like to discover more and much richer deposits. I think it is in an IAEA publication where the annual mean discovery rate is put at only 65 000 tonnes. Of late, but for the recent Australian finds, I do not think that we have come across deposits that are really rich. Generally, in the base metal industry, we find that the grades do go down and, after all, mineral wealth is a wasting asset. One day we shall have to take up low-grade ore utilization. Low-grade utilization and cost structure differ from place to place. In geological terms, there are areas which are completely devoid of a particular metal. For instance, the USA is rich in uranium but lacking in certain other metals such as manganese. In India there are many metallogenic provinces that are rich in thorium but poor in uranium. We are doing our best and some recent finds are encouraging. We have a large area to investigate and in the coming two decades or so, considering the forecasts for uranium supply and demand, we shall have to go down in grade. For instance, an eight-step lowering in copper grades has taken place even in the USA. Taking 0.25% U_3O_8 as the grade of ore which was treated in the USA several years ago and assuming an eight-step lowering by analogy with copper, we come to an ore grade of 0.02 or 0.03% U_3O_8 .

URANIUM RECOVERY FROM BITUMINOUS SHALES AT RANSTAD

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Abstract

URANIUM RECOVERY FROM BITUMINOUS SHALES AT RANSTAD.

LKAB is planning a new plant for the recovery of uranium from low-grade bituminous shales at Ranstad in south-western Sweden. The processes have been developed at the existing plant which has served as a testing facility since 1968. After mining in open-pit and underground mines the ore is treated by heavy-media separation to separate the shale from limestone inclusions. The shale is crushed to -2 mm and acid-cured with sulphuric acid before percolation leaching. Seven large vats will be used for a leaching time of 6 days, including 1 day of washing. Acid consumption is about 60 kg/t of shale, giving a uranium recovery of 80%. The uranium is recovered from the leach solution by solvent extraction followed by precipitation and drying. Mining of a low-grade shale deposit will be a disturbing element to the environment. Open-pit and underground mining and tailings disposal will affect the scenery as well as the groundwater table. There is always a danger that water seepage will pollute both surface and groundwaters. The radiotoxicity of the tailings is carefully studied but, as yet, no great difficulties are foreseen.

1. THE RANSTAD PROJECT

A small uranium mill at Ranstad was built and taken into operation in 1965. The plant, which was constructed with government funds, was closed down shortly after start-up, as a result of the low demand for uranium caused by delays in commercial reactor programmes. The facilities have since been used for pilot-plant testing. New and more efficient processes are being studied and developed.

Since the beginning of 1975, LKAB has taken over the facilities and is now planning a new mine and mill at Ranstad for the recovery of 1500 t of uranium oxide per year.

2. GEOLOGY OF ORE DEPOSIT

In Sweden, large but low-grade uranium ore reserves are found in the extensive formations of bituminous shales. At Ranstad, in the south-west of Sweden, the uranium-bearing shale is a part of the practically horizontal series of sediments belonging to the upper Cambrian Era (Fig.1). The horizon of interest covers an area of 500 km². The seam is 2.5–4.0 m thick with a grade of 250–325 g of uranium per metric ton. The total uranium content is about 1×10^6 t, of which it should be possible to recover at least 0.3×10^6 t on a purely technical and economic basis.

The bituminous shale contains about 22% organic matter and about 13% pyrite. Quartz and silicates such as illite and feldspars are the main constituents. The shale is very fine grained with a natural particle size of less than 0.01 mm. About 90% of the uranium is evenly distributed in the seam. The remaining 10% is concentrated in high-carbon "kolm" inclusions. The seam also contains about 6 wt% of lens-formed inclusions of bituminous limestone.

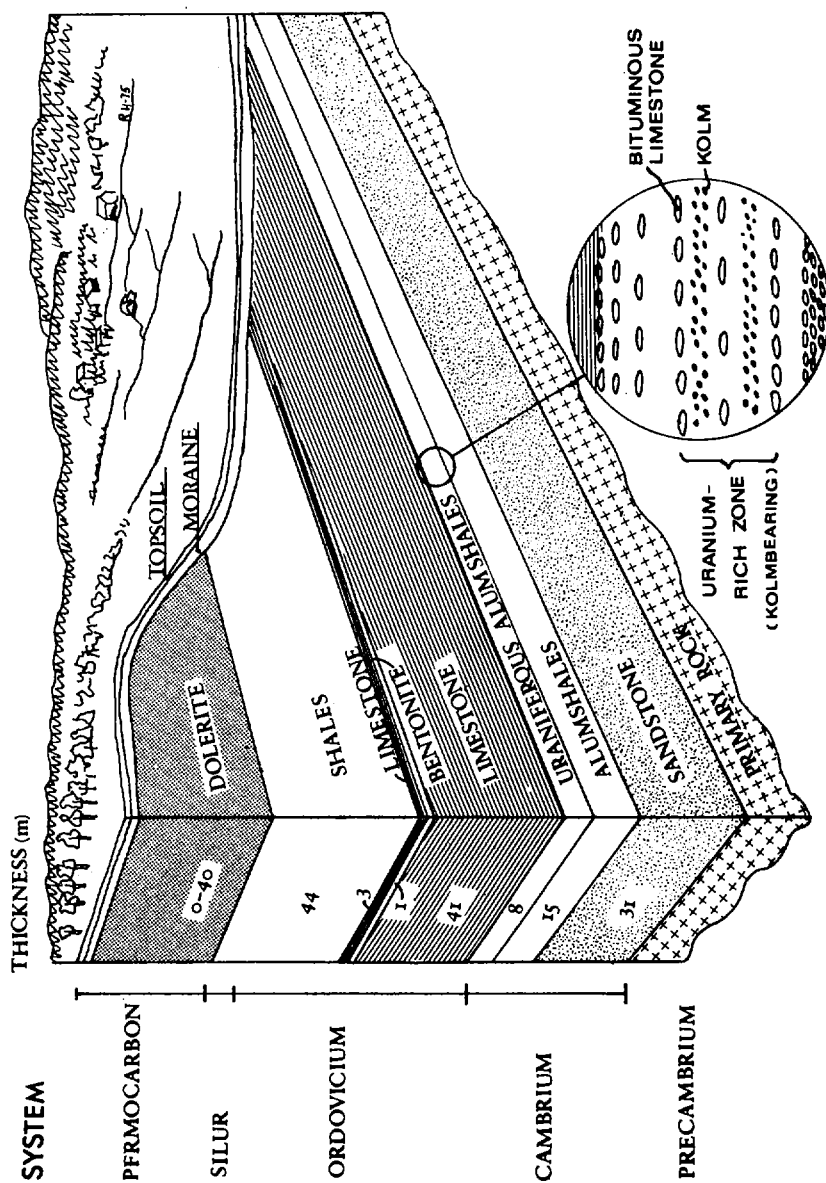


FIG.1. Section of the Billinge mountain.

3. MINING OF 6×10^6 t OF ORE

The mining rates in open pit and underground mines exceed 6×10^6 t of ore per year. The open pit will be designed as a strip mining operation with a capacity of 3×10^6 t per year. The ratio of overburden to ore will be about 4 to 1.

Underground mining will be started at a capacity of 3×10^6 t per year. When the open pit is exhausted after about 15 years of operation the capacity underground will have to be raised to 6×10^6 t. Mining by the room and pillar method will take place at 100–150 m below the surface using 5×5 m square-set pillars. The width of the room will be 5 m and the height 3.5 m. Mining operations are carried out as conventional drifting with drilling and blasting. The ore is loaded by front and end loaders and hauled to the primary crusher station by underground diesel trucks.

Methane as well as radon gas emission from the bituminous shale is anticipated. With regard to these and to diesel exhausts an efficient ventilation system is required. Mined-out areas will be closed off and filled with tailings.

4. URANIUM EXTRACTION PROCESS

The proposed plant will include facilities for ore preparation, percolation leaching, solvent extraction and precipitation of sodium or ammonium uranate. The flowsheet of the proposed process is shown in Fig.2.

4.1. Ore preparation

After primary crushing in a gyratory crusher located underground, the ore is transported by belt conveyors to a 3-day capacity storage bin. From here the ore is conveyed to the heavy-media separation plant where limestone inclusions are separated from the shale. Material coarser than 25 mm is processed in drawboy machines and material finer than 25 mm is treated in hydro-cyclones. In both cases a magnetite-water suspension at an apparent density of 2.4 will be used. The uranium-bearing shale turns up as float product at a high yield containing about 1% residual CaO.

The coarse float product is crushed in hammer mills and combined with the fine float before final crushing in impactors. The crushed material is passed over vibrating resonance screens with a 2 mm square mesh working in closed circuit with the impactors. The -2 mm material is contacted with sulphuric acid in curing drums.

Crushing and screening operations produce dust which is sucked off by air and precipitated in filters to a quantity of about 10% of the total ore production. For recovery, the dust is first treated in an air classifier where 30% of material coarser than $60 \mu\text{m}$ is separated and fed directly into the main flow ahead of the curing drums. The remaining dust material, which is finer than $60 \mu\text{m}$, is pelletized to 10 mm dia. balls. Before agglomeration the dust is wetted with sulphuric acid which acts as binder. The pellets, which are fed to the main flow after curing, have proved to be strong enough to resist mechanical as well as chemical treatment in the further processing. The crushed shale is stockpiled in an open storage where oxidation and bacterial processes occur which may have a positive effect on the leaching process.

4.2. Percolation leaching

The uranium is extracted from the ore by countercurrent percolation leaching with sulphuric acid. Using a leaching time of 6 days at a temperature of about 60°C the uranium recovery will reach 79% (the maximum yield during extreme conditions using sulphuric acid is 85%). Acid consumption is expected to be 6% of shale weight.

The plant will consist of seven leaching vats with a capacity of 15 400 t each. The vats must be lined with acid-resistant material. At the bottom they will be provided with a gravel filter.

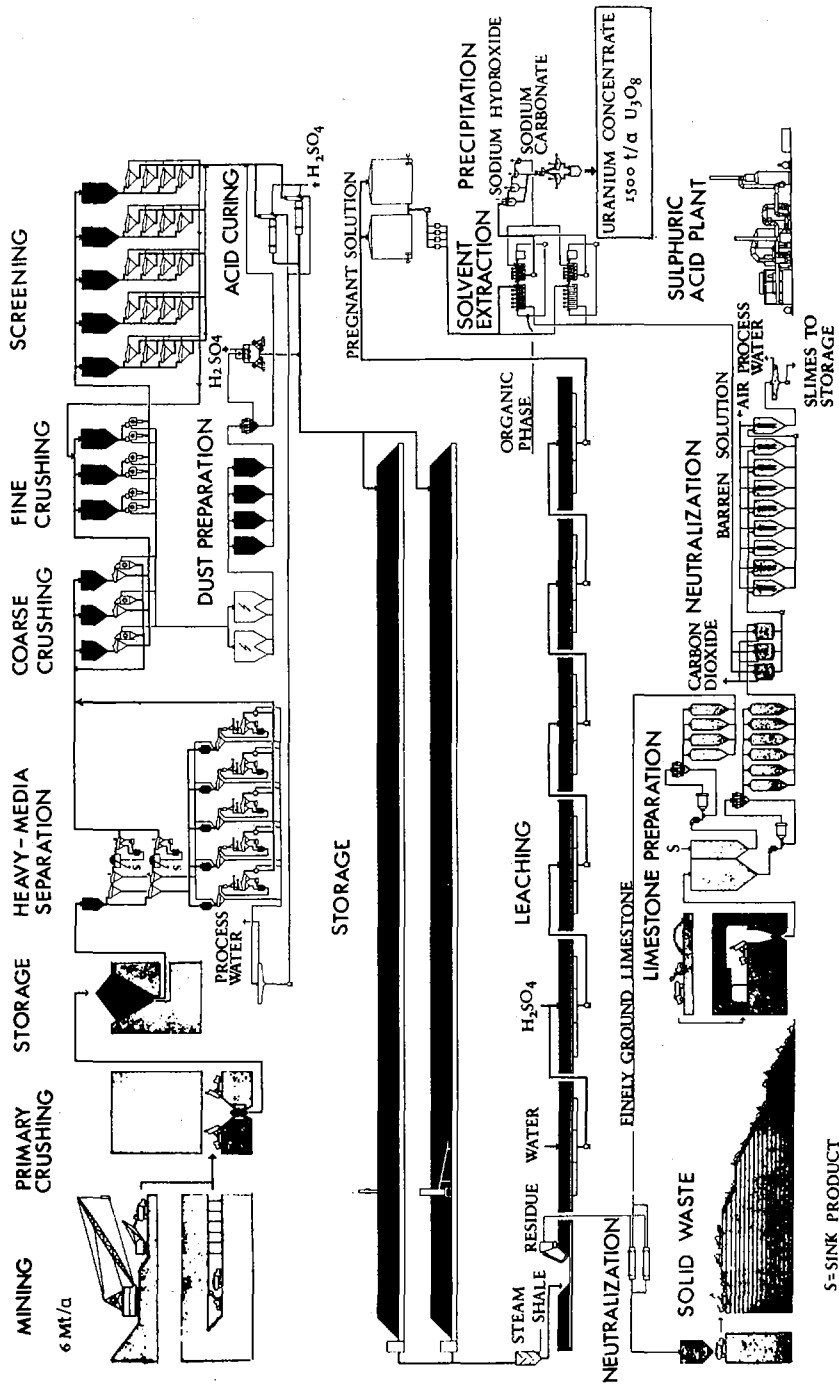


FIG. 2. Flowsheet of the Ranstad uranium plant.

4.3. Solvent extraction of uranium

The pregnant leach solution is composed approximately as follows:

U	0.7	g/l
Al	17	g/l
Fe	10	g/l
K	8.5	g/l
Na	5.8	g/l
Mo	0.04	g/l
V	0.3	g/l
Ni	0.2	g/l
SO ₄	133	g/l

After filtration of the solutions the uranium is separated by solvent extraction using a tertiary amine (Alamine 336) dissolved in kerosene. At the addition of sodium carbonate (or acidified ammonium sulphate) to the organic solution the uranium is stripped and a yellowcake is precipitated by the addition of sodium hydroxide (or ammonia). Pilot-plant tests have proven a yield of over 99.8%.

5. TAILINGS

The solid residue from the leaching process will be deposited in the depleted open-pit area. As pyrite still remains in the residue, certain care has to be taken in order to prevent weathering.

6. BARREN ACID SOLUTION

The barren solution from uranium extraction is neutralized. Finely ground limestone is used for the process in which gypsum and metal hydroxides are precipitated. The slime is thickened to about 35% solids and pumped to mined-out areas or tailings ponds for storage.

7. PRODUCTION OF SULPHURIC ACID

About 325 000 t of sulphuric acid per year is consumed in the leaching process. A factory based on burning elemental sulphur supplied with double catalysis is being planned at the mine site.

8. OTHER PRODUCTS THAT MAY BE PRODUCED

The uranium extraction process described above has been optimized with regard to uranium. The shale as well as the leach solutions contain many other valuable products that may be of interest such as vanadium, nickel and aluminium. On a long-term basis, the energy content of the bituminous shale (1800–1900 kcal/kg) as well as its content of sulphur may be of interest for the production of electric power and sulphuric acid.

9. POLLUTION PROBLEMS MAY CHANGE THE PROJECT

The orebody is located in a densely populated and rich farming district. Mining, processing and tailings disposal will be a disturbing element to the environment and the considerations by

government and local authorities may have an influence on the total project and may also change mining and process particulars. Some of the more important environmental problems are listed below:

(a) Open-pit mining and tailings disposal will change the scenery. The depleted open-pit area as well as the tailings piles and ponds will have to be arranged to fit into the surroundings. The surface will have to be recultivated.

(b) Mining will lower the groundwater table. There is also a danger that water seepage from mining operations and tailings disposal may pollute the groundwater.

(c) Emissions of sulphur dioxide and dust will have to be closely controlled. The plant will have to be equipped with efficient means for cleaning of exhaust air and stack gases.

(d) Further and more efficient treatment of barren solutions is being investigated. It is considered desirable to close the process and balance the solutions to avoid a contaminated effluent to the recipient.

(e) The radiotoxicity of the tailings is carefully studied. Radon emission will be reduced to low values by covering the tailings with densely packed soil. No great difficulties are, as yet, foreseen. However, a large and detailed testing and sampling survey is under way.

DISCUSSION

A.W. LANKENAU: Is this ore similar to the lignite shales in the USA?

Å. ANDERSSON: I think there is some similarity but the organic content is different. Also, this orebody is flat and quite easily mined. I do not know much about your lignites.

A.W. LANKENAU: I was thinking about chemical characteristics.

Å. ANDERSSON: The organics are different, but otherwise I think the chemical characteristics are fairly similar.

A.W. LANKENAU: I would like to know the BTU content of this material.

Å. ANDERSSON: I cannot give you any BTU values. The ore contains 2000 kilocalories per kilogram, which is a little more than about one fifth of the value of a good coal or of oil.

K. PINKAS: Do you intend to recover from your leaching solution besides uranium other precious metals, perhaps to better the economy of the whole process?

Å. ANDERSSON: We intend to recover vanadium and nickel for a start.

R.H. KENNEDY: In the original Ranstad plant, I believe you have had a very difficult time handling the fines from the crushing operations. How is that to be handled in the future plant?

Å. ANDERSSON: Fine dust will be pelletized with sulphuric acid. In heavy-media separation we can also handle finer material using hydrocyclones, down to approximately 1/32 in (≈ 0.7938 mm).

A.W. LANKENAU: Have you experienced any difficulty with the organic content in the ore in your solvent extraction?

Å. ANDERSSON: No.

S. AJURIA-GARZA: How long do you store the ore in the storage bins?

Å. ANDERSSON: We have found that storage before leaching does not give us better uranium recovery. We store for about 10 days only to ensure feed for the leach vats.

P. MICHEL: Do you have a large amount of carbonates in your shales?

Å. ANDERSSON: Before heavy-media separation the carbonate content is approximately 6–7%. Afterwards it is about 1% CaO. In curing with sulphuric acid this carbonate is eliminated. Curing has made it possible to avoid intermediate storage if this is desired. Before curing was introduced the ore had to be stored for at least 10 days. During this storage time acid was formed by bacterial attack on pyrite. This acid reduced the carbonate content.

J.A. PATTERSON: One comment. I think the concept of pelletizing fines has much more significance than may be realized from the casual mention that was made here. Shales are a very fine-grained material with attendant dust and slime problems. There is the problem of processing the fines in the plant and a waste disposal problem. Pelletizing provides an opportunity to handle the fines as a desirably sized mineral material. Instead of making processing difficult, the fines become an asset as an additional useful mill feed. In addition, regarding waste disposal, instead of dealing with a slimey liquid material we now have a strong material that can be formed into piles and will drain. I think this is a very interesting and significant development.

G.F. IVANOV: I should like to know whether self-burning of the shale does occur during storage.

Å. ANDERSSON: If the ore is not carefully handled, it may start burning. This is actually a matter of grain size — coarse ore does not burn easily while fine material may catch fire if not properly stored.

**URANIUM AS A BY-PRODUCT AND
BY-PRODUCTS FROM URANIUM ORES**

(Session VI)

URANIUM AS A BY-PRODUCT AND BY-PRODUCTS OF URANIUM PRODUCTION

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Abstract

URANIUM AS A BY-PRODUCT AND BY-PRODUCTS OF URANIUM PRODUCTION.

There has been no large-scale production of uranium as a by-product in the United States of America. During the 1950s and early 1960s small tonnages of uranium were recovered as a by-product of the phosphate industry. With increasing demand and correspondingly higher prices for uranium there is a renewed interest in uranium recovery from phosphates. The possible recovery of uranium from porphyry copper waste dump leach liquors is also being re-examined. Because of the concern about the adequacy of uranium resources in conventional-type deposits, other low-grade sources of uranium such as the Chattanooga shale (60 ppm U) are being re-evaluated. Most uranium recovered in the USA comes from sandstone-type ores with little or no other metals of commercial interest. Some ores, however, contain sufficient vanadium to warrant recovery. Nearly all ores contain traces of other metals, many of which either have been recovered on a small scale at one time or another or the possibilities of recovery have been investigated. They include molybdenum, rhenium, selenium, copper, silver, scandium, arsenic, mercury, thorium, ionium, protactinium and radium. Uranium ore processors should be alert to the possibility of recovering these and other metals which may occur in uranium ores.

INTRODUCTION

By-product uranium production in the United States of America has been very modest so far, amounting to somewhat less than 1000 tonnes of uranium. However, with the projected increasing demand for uranium and correspondingly higher prices, by-product sources of uranium are receiving renewed attention. This is especially true of uranium from phosphates and from porphyry copper waste dump and ore leaching operations. The construction of facilities was recently completed in central Florida to resume the recovery of uranium from one wet-process phosphoric acid plant. Other phosphoric acid producers can be expected to commence uranium production in the next few years.

In every uranium recovery operation, whether by conventional ore processing or as a by-product of some other industry, consideration should be given to recovery of other metals or elements which may accompany the uranium. In some cases, recovery of other metals can be quite profitable besides conserving natural resources. This paper reviews the current status of by-product uranium recovery and the recovery, both past and present, of other metals or elements occurring in USA uranium ores.

BY-PRODUCT URANIUM FROM PHOSPHATES

The recovery of uranium from wet-process phosphoric acid has become more attractive as the price for uranium has increased and as process technology has improved. Phosphoric acid

produced from central Florida land-pebble phosphate rock generally contains more uranium (100 to 200 mg/l) than acid from other phosphate rock in the USA, so currently this acid is the most attractive source of uranium [1]. Recently, the Uranium Recovery Corporation (URC) completed the construction of an 8 million dollar complex in central Florida to commence uranium production using an exclusive process developed by URC [2]. Unlike earlier solvent extraction plants that operated in the 1950s and early 1960s, the URC concept is to split the process into two parts, with only the initial solvent extraction and stripping operation at the phosphoric acid plant site [3]. The strip solution from the first stage is transported by truck to a central processing plant where the second solvent extraction and stripping step produces specification grade uranium concentrate. This approach has the advantage of requiring only simplified facilities or 'modules' at each phosphoric acid producing plant, whereas the more complex but smaller-scale downstream processing is accomplished at a single centrally located facility or 'refinery'. Assuming early success in the operation of the initial module, it is expected that additional modules soon will be constructed at other nearby phosphoric acid plants. Although the economics are yet to be fully developed, it currently appears that it would not be profitable to construct modules at plants with an annual uranium production potential of less than about 50 tonnes or at a distance of more than about 80 kilometres from the central plant.

Several other phosphoric acid producers are seriously considering recovering uranium and some have operated or are operating pilot plants of 10–20 l/min acid flow rate to determine the extraction characteristics of their acids and to investigate process modifications or improvements. In general, these efforts appear to be directed primarily toward a two-cycle solvent extraction process similar to that developed by the Oak Ridge National Laboratory (ORNL) a few years ago [4]. In the ORNL process, the uranium is extracted by the synergistic combination of di(2-ethylhexyl) phosphoric acid (D2EHPA) plus trioctylphosphine oxide (TOPO) in an aliphatic diluent such as kerosene. The extractant is stable and can be recycled extensively, and solvent losses are low.

Currently, the uranium production potential from wet-process phosphoric acid produced from central Florida land-pebble phosphate rock is about 3000 tonnes of uranium per year. One would expect this potential production capability to increase as the acid-producing industry expands, which in the recent past has grown at a rate exceeding 10% per year. Also, the tendency is to construct larger phosphoric acid plants which make uranium recovery more attractive.

Past studies have shown that wet-process phosphoric acid also contains rare earths and other elements that possibly could be recovered [5]. Such possibilities should not be overlooked in conjunction with uranium recovery.

BY-PRODUCT URANIUM FROM COPPER WASTE DUMP LEACH SOLUTIONS

The leach solutions from many copper waste dumps in the western United States contain 1–10 ppm uranium [6]. The solution flows are large enough for 800–900 tonnes of uranium per year to be potentially available, but none is being recovered.

The US Bureau of Mines and Kennecott Copper Corporation ran ion-exchange pilot plant studies on uranium recovery from the dump leach liquor at the Bingham Canyon mine near Salt Lake City [7]. These studies confirmed laboratory tests that the uranium is easily recovered, but there was some concern that the uranium concentration of the recycled leach liquor would decrease to an uneconomic level. More recently, Westinghouse Corporation has run some recovery tests and announced tentative plans to build a small uranium recovery unit near the copper cementation plant of a large copper producer.

Recent increases in the price of uranium have prompted other copper producers with leaching operations, both ore and waste, to re-examine this potential source of uranium.

BY-PRODUCTS OF URANIUM PRODUCTION

Vanadium

Vanadium and uranium have been produced since 1900 in the Uravan Mineral Belt of Colorado and Utah and to a lesser extent in adjoining portions of the Colorado Plateau extending into New Mexico and Arizona [8]. Much of the vanadium came from uranium-vanadium ore such as carnotite which contained three to five times as much vanadium as uranium. Initially uranium was in demand for ferro-uranium alloys and pigments for ceramics, and vanadium was needed for ferro-vanadium alloys, for use as a mordant and as an oxidizer.

Vanadium production in the Uravan Mineral Belt from 1911 through 1937 was cyclic, averaging 1000–2000 tonnes per year with no significant growth. Uranium production was not important and when uranium was recovered, it was a by-product of vanadium or radium. (Radium was an important product from uranium-vanadium ore in the Uravan Mineral Belt from the early 1900s until 1923.)

Beginning in about 1946, the uranium production in the USA came almost entirely from the tailings of old vanadium and radium mills in the Uravan Mineral Belt and as a by-product of operating vanadium mills. In the 1950s the demand for uranium exceeded that for the vanadium associated with it, so some vanadium concentrate was stockpiled for later use and some vanadium was discarded with the mill tailings.

Since 1960 there have been seven uranium mills in the USA which have recovered vanadium as a co-product. A few of these mills have also recovered uranium as a by-product from vanadium ore which contained too little uranium to be recovered economically in the absence of vanadium. Production has been a few thousand tonnes of vanadium (1000–6000 t) per year.

Generally it has not been economic to recover the small amounts of vanadium present in uranium ore from outside the Uravan Mineral Belt; however, two uranium mills have recovered these small amounts of vanadium. One of these mills, Mines Development, Inc., in South Dakota, had a circuit for recovering vanadium from iron slags and used this circuit to recover vanadium from uranium plant tailings [9]. The other mill, United Nuclear-Homestake Partners in New Mexico, precipitates a sodium diuranate concentrate containing about 3% vanadium [10]. This concentrate would not be acceptable at the plants which convert concentrate to uranium hexafluoride, so United Nuclear-Homestake Partners calcines the impure concentrate and quenches the calcine in water to dissolve out most of the vanadium. The resulting sodium vanadate solution is concentrated and sold to a uranium-vanadium mill for vanadium recovery.

Until recently, most of the vanadium from uranium-vanadium ore was recovered by salt roasting and then water leaching the salt-roasted ore to dissolve the sodium vanadate. Some additional vanadium dissolved during subsequent acid leaching of the ore to recover uranium, and this was also recovered. Today salt roasting is not practiced at uranium-vanadium mills in the USA. Instead, the ore is given a hot oxidizing leach, and uranium and vanadium are recovered in separate ion exchange and/or solvent extraction circuits.

Enough vanadium is available from non-uranium sources so that currently there is no incentive for uranium ore processing plants to recover vanadium unless the mill feed contains an average of more than 0.2% vanadium. In the Grants area of New Mexico, most uranium ores contain only about 0.05% vanadium or less, and in Wyoming the ores contain less than 0.01% vanadium.

Molybdenum

Molybdenum occurs in at least trace amounts in all domestic uranium ores and some ores contain a few tenths of a per cent or more. Although not much study has been devoted to the mineralogy or mode of occurrence of the molybdenum in these ores, it generally is thought to be present as a sulphide, sulphate, a hydrated molybdate or adsorbed on carbonaceous materials or on hydrated iron oxides [11].

Usually, more molybdenum is extracted by alkaline carbonate than by acid leaching solutions. In either case, means are available for removing or controlling the molybdenum concentration in solutions. When uranium is precipitated from alkaline carbonate solutions with caustic, the molybdenum remains in solution and is recycled to the leach. Eventually, a solution bleed is necessary to avoid an excessive molybdenum build-up and molybdenum recovery is possible. Another effective control method where molybdenum is present in the ore as a sulphide is to float and separate the sulphides ahead of alkaline leach. The sulphide concentrate can be acid leached for uranium and molybdenum recovery, if warranted.

In acid solutions, various molybdenum control or recovery procedures are available such as sulphide precipitation, adsorption on activated carbon, and purification by partial neutralization. Molybdenum has been recovered in several uranium mills and usually it has been marketed as a calcium molybdate or an ammonium phosphomolybdate containing 40–50% Mo. Total production to date is estimated at one million kilograms molybdenum. A substantial portion of this resulted from the processing of uraniferous lignites which contained almost as much molybdenum as uranium. No lignites have been processed for several years but small quantities of molybdenum are still recovered at a few mills. At one mill, molybdenum is recovered primarily because of environmental considerations.

Rhenium

Uranium ores containing molybdenum also contain rhenium in amounts of from less than 0.3 to 3 ppm [12]. In alkaline leaching, most of the molybdenum and rhenium are dissolved along with the uranium and if it were economically attractive, both metals could be recovered. In acid leach liquors, the rhenium content ranges from 0.03 to 1 ppm. About one-half of the rhenium can be removed from the leach liquor by passing it through a bed of activated charcoal, by treating it with ion exchange resins, or by solvent extraction with secondary or tertiary amines.

Uranium mills that recover by-product molybdenum have the best prospects of recovering rhenium. Only one mill has recovered rhenium, as a perhenate solution, but others have investigated the possibility.

Uraniferous Chattanooga shale from Tennessee contains 0.05% molybdenum and about 1 ppm rhenium. Vanadiferous shales from Wyoming also contain about 1 ppm rhenium. Because of the complex mineral assemblage in the shales and the large amount of organic material present, efforts in the laboratory to collect an enriched rhenium product by roasting and collecting the fume were unsuccessful.

Selenium

Selenium has long been known to occur in sandstone-type uranium deposits, but the selenium content does not correlate with the content of any other analysed constituent in the deposits [13]. The concentrations of selenium range from less than 1 ppm to as much as 0.1% in the immediate vicinity of uranium deposits, and some western soils, such as the Pierre, Niobrara and Mancos shales, contain concentrations of as much as 200 ppm.

In the Grants Mineral Belt, selenium is found in the native state as gray or red selenium and as ferroselite (FeSe_2), and many of the uranium ores in the area will average about 0.03% selenium [14].

Selenium is leached from some uranium ores in alkaline circuits, and along with molybdenum it will tend to concentrate in the recirculated solutions. This build-up is usually controlled by bleeding a small portion of the solution to waste and this bleed stream conceivably could be treated for selenium and molybdenum recovery. The addition of sodium sulphide will precipitate selenium directly from the alkaline solution [15]. Cotter Corporation recovered selenium from old residues from Congo uranium ores by sulphide precipitation [16].

Copper and silver

Some of the uranium ore from Utah and Arizona contains 0.15–2.0% copper and trace amounts of silver. Four uranium ore processing plants (El Paso Natural Gas Co. [17] at Tuba City, Arizona; Texas-Zinc Minerals Corp. [18] at Mexican Hat, Utah; Cotter Corp. at Canon City, Colorado; and Atlas Minerals Corp. [19] at Moab, Utah) recovered copper from copper-uranium ore. These mills recovered copper by flotation of sulphides from the ground ore and by cementation of solubilized copper from the effluents from uranium ion exchange or solvent extraction. The sulphides were acid leached to extract the uranium.

Copper recovery was 75–80%, including 85–92% of the sulphide copper. Each mill recovered 400–800 t of copper per year as a concentrate containing 18–36% copper, less than 12% water, 0.01–0.03% uranium and 70–400 g of silver per tonne of copper.

Today, only the Atlas Minerals plant still has copper recovery facilities. These operate for a few days every year, because very little copper-uranium ore is received. This ore is set aside in stockpiles until there is enough accumulated for processing in a short campaign.

Cotter's flowsheet was unique in that, in addition to copper, cobalt and nickel were recovered as sulphides from the acid leach solution while processing old residues from Congo uranium ores [16, 20].

Scandium

Trace amounts of scandium found in most uranium ores dissolve during an acid leach, yielding solutions containing up to one milligram Sc_2O_3 per litre [21]. In the Vitro Chemical Company uranium mill the scandium followed the uranium into the organic extractant (dodecyl phosphoric acid) and remained there during subsequent stripping with hydrochloric acid. The concentration of scandium built up along with thorium and titanium. A hydrofluoric acid strip of the organic removed and precipitated a scandium-thorium fluoride cake containing about 10% Sc_2O_3 and 20% ThO_2 . This concentrate was processed at another location to produce a high-purity scandium oxide (more than 99.5% Sc_2O_3). Special precautions were necessary in handling the scandium-thorium concentrate because of high radioactivity owing to the thorium-230 (ionium) and thorium-234.

Subsequently, the US Bureau of Mines, Salt Lake City Metallurgy Research Center, developed an improved solvent extraction process for scandium recovery which was tested at the Vitro pilot plant [22].

Arsenic and mercury

No attempt has been made to recover arsenic or mercury from uranium ores in the USA although several vein-type ores have contained arsenic and mercury as sulphides. The recovery of mercury from mill tailings was proposed at the Lakeview mill, Oregon, but never undertaken. Data collected at the Gunnison mill, Colorado, showed that mill effluents contained 10 to 20 ppm arsenic which necessitated effluents being retained indefinitely to prevent chemical pollution of the nearby river and wells [23].

In countries where uranium ores contain high sulphide concentrations, recovery of arsenic and mercury might be worth studying. This has not been done in the USA because most of the uranium ore being processed is sandstone-type which does not contain any significant concentration of arsenic or mercury.

Thorium-232

At present no uranium ore being processed in the USA contains enough thorium to be a problem or to be worth recovering. Earlier, a few thousand tonnes of uranothorianite ore from

Alaska were processed in several US mills. These mills did not recover thorium but found it necessary to modify their processes to prevent the thorium from contaminating the uranium concentrate. At the Texas Zinc Minerals Corp. plant this was accomplished by changing from a tertiary amine to a secondary amine for solvent extraction. The Dawn Mining Co. added lime and then phosphoric acid at a pH of 3.5 to 4.0 to the pregnant liquor from ion exchange for its first-stage precipitation of calcium sulphate, ferric hydroxide and thorium phosphate. This 'white cake' was removed by filtration before the uranium concentrate was precipitated.

Thorium has been recovered as a by- or co-product of uranium in other countries, but the demand for thorium is not great enough to make its recovery attractive at most operations today.

Ionium (thorium-230) and protactinium-231

These radionuclides are desired for special laboratory studies, but ores in the USA do not contain significant amounts of either. However, the waste material accumulated from processing very high-grade uranium ore from the Belgian Congo did contain appreciable amounts of ionium (^{230}Th) and protactinium-231.

The US Atomic Energy Commission arranged to have Cotter Corporation, while processing the Congo residues to recover other elements, set aside the uranium concentrate for possible recovery of the contained ^{230}Th and ^{231}Pa . The recovery ranged from 10 to 50% of the ^{230}Th and from 20 to more than 50% of the ^{231}Pa [24]. The uranium, thorium, protactinium and many impurities were precipitated from the carbonate strip solution by adding excess sodium hydroxide. This crude cake containing 50–150 ppm ^{230}Th and about 1 ppm ^{231}Pa has not been processed to recover these radionuclides in more concentrated forms.

Other countries may wish to investigate the recovery of ^{230}Th and ^{231}Pa if they are fortunate enough to find very high-grade (more than 20% uranium) ore to process. However, at this time we are not aware of any particular demand for these elements other than for research purposes.

Radium

For about 10 years (1913–1922) the USA dominated the world radium market [25]. This condition changed abruptly in 1923 when Belgium dropped the price of radium after discovery of the rich uranium deposits in the Congo (Katanga). Since 1923 very little radium has been produced in the USA. Nearly all production came from a relatively small area in south-western Colorado and south-eastern Utah where high-grade (1–2% uranium) carnotite deposits were mined. Before World War I, much of the ore was exported to England, France and Germany for processing, but subsequently all domestic production was treated in the USA. For many years radium was used as a source of radiation in the treatment of cancer but it outlived its usefulness as better, safer radioactive materials became available.

Today there is little or no market for radium so its recovery from uranium ores is not considered worthwhile. The one exception would be to render uranium mill tailings less hazardous to the public through radium removal but this would be costly, especially if very high recovery is required.

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DISCUSSION

S. AJURIA-GARZA: Do you know of any report on zirconium associated with uranium mining? We have at least one orebody which contains zirconium.

F.E. MCGINLEY: Several ore samples were brought to us for examination that did contain zirconium, but to my knowledge there has never been any significant tonnage of that type of mineralization in the USA.

F.R. HARTLEY: In my paper I pointed out that we have large reserves of uranium in ores around the Mount Isa area, much of the uranium being associated with zircon, but at this point in time they are quite uneconomical for treatment.

One other comment. The Port Pirie Plant, which was shut down some years ago, had tailings in which the barren liquid contained 0.03 g/l scandium. We operated a pilot plant for some time

and extracted quite a large amount of scandium — something like 550 lb of scandium assaying +99.9% Sc. It was quite an extensive programme to see how much interest there was in purchasing scandium, and we made some rather exotic compounds for at least 15 universities and various other research institutes scattered throughout the world, none of which had any feedback whatever.

F.C. LENDRUM: I have one comment on the discussion about the possibility of recovering radium from the tailings. Consider the sandstone ores. What is the difference between the radium in mill tailings and the radium that is just below the surface in the original orebody? This is something I have never been able to understand. In the orebody underground nobody worried about it, but when it shows up in mill tailings it becomes a big problem. If anybody has any comments as to the difference between radium when it is in the orebody and radium when it is in the tailings, I would appreciate it.

P.A. BONHOTE: I must admit the same thought has occurred to me on several occasions. One of the problems of course is that, finely ground and brought up on the surface, it is a massive dump, with some attraction for various purposes for people if they have access to it. I feel that the radon release to the atmosphere from a tailings pile is much greater than the radon release from an underground orebody. Actually, we are only dealing with natural radioactivity; we have not added any radioactivity, but we have made it more accessible and more of a hazard. Nevertheless, I still feel that the actual hazard is not very significant compared with all the other environmental hazards that we are exposed to.

F.C. LENDRUM: I will just add one comment. I was specifically referring to the open pits rather than underground mines because, looking at the hills in Wyoming or Australia, surely someone must have dug up some for cement blocks at some time. At a later time they made cement blocks out of the old tailings — one becomes a hazard, one nobody worries about. There cannot be that much difference. I merely want to make this comment for the record.

F.R. HARTLEY: I would like to come in on this as well. The problem is certainly pre-occupying our attention very considerably in view of the ore grades that we are handling. We have been led to believe that the solubility of the radium in the tailings is much higher than in the ore. Radium is present in the slime, much of it as a sulphate, and there generally it is more soluble and more liable to penetrate to groundwaters. Another problem is that the tailings — in Australia at any rate — will have to be placed below ground level; we cannot afford to build tailings heaps above ground level because of the risk that they may be damaged or washed away in the rainy season. At present we are considering using sand/slime separation and deliberately placing the tailings so that the slimes are always covered with a fairly substantial amount of sand. The ratio will be about 10:1.

R.H. KENNEDY: Since this subject happens to be my particular responsibility at the present time, maybe I can make a few comments here for you to consider.

Recently, Dr. Victor Archer reported a total of about 277 lung cancer deaths in the uranium-producing industry in the USA. I recently talked with Dr. Saccomanno in Grand Junction who carries out sputum cytology studies for the uranium miners. He says that if they had been able to keep track of the total number of people who had been working in the industry, he thinks that the total number of lung cancer deaths on record would probably have been about 420. So we have here an industry in which the number of deaths attributable to radiation far exceeds that in all other parts of the nuclear industry put together. The environmentalists are going to take this point up — in fact they are taking it up — and they are regarding the uranium-producing industry as perhaps the Achilles heel of the whole nuclear programme in the USA. I am sure that this will be a problem in other countries as well. It is one thing to talk about exposures to radon and daughters in mining operations, and it is certainly another thing to talk about the exposures that exist in mills and exist for the general public as a result of radon emanating from tailings piles. Let us not forget that the general method of determining the public hazard these days is a straight-line extrapolation from the high exposure rates in mines, and it still calculates out to a significant number of so-called health effects or expected cancers that would

come from exposures to tailings. I think that although the radiation levels may not be significant compared to the other health hazards encountered in industry and by the general public, nevertheless they are not going to be neglected, certainly not by the environmental group. I think, in its own defence, the uranium-producing industry should also take these calculations seriously, because they are going to be a problem to the industry in the future. Although it may or may not seem reasonable to think that the small amount of radon gas released from a tailings pile is a health problem, it is going to be a problem to the industry in general if it is neglected. We do have instances in the USA where tailings piles have been built in fairly sizeable metropolitan areas and the dust blown from these tailing piles constitutes a considerable nuisance if not a public health hazard. Our Environmental Protection Agency has been making radiation measurements around these piles for some time. We have also done this ourselves, and the evidence is that elevated radon levels can be readily detected within half a mile (≈ 0.8 km) of an existing tailings pile. This regards non-stabilized piles or those stabilized simply by a vegetative cover or something like that, as is used today.

The calculations that have been made, based on diffusion models, indicate that about 20 to 25 ft of cover material would be required to reduce the radon emanation rate by about 95%. In many instances this amount of cover material would be more than the amount of tailings accumulated on the site. Putting an ordinary cover on top of a tailings pile is not a very attractive proposition from the producer's point of view. Therefore, we are trying to institute a small research and development project to try to develop much more effective radon cover materials than the normal dirt cover that has been used so far. We have some indications already that it might be possible to use much more dense materials and much more water-bearing materials, for instance, to effectively reduce the radon emanation rates. Radon emanation is lower through water than through any other potential cover material that I know of and that is readily available. This is the line of research that we follow. It is a pretty serious proposition with us because in Grand Junction, for instance, the Federal Government has already put up US \$5 $\times 10^6$, and the State of Colorado US \$1.7 $\times 10^6$, to correct a very undesirable situation that had developed when tailings were used in building habitable structures. We are now considering measures to be taken at Salt Lake City. The early cost estimates for controlling the tailings pile are well into the millions of dollars. I am sure this is more than any of the uranium producers are prepared to spend to control the further spread of radiation from their operations when they are closed down. It is a serious and difficult problem and I expect that the industry is going to get hit with it, so it would be well for uranium producers to plan in advance to fix their tailings in such a way that the subsequent radiation exposure or off-site radiation discharge is reduced to an absolute minimum.

F.C. LENDRUM: I think that this points up, once again, that the sand/slime separation in the tailings pile may be part of the problem we get with high concentration in the slimes or the pools in the middle.

J. CAMERON: If I may take up another problem, do you have information about what is happening in the rest of the world regarding by-products and co-products? Particularly I had in mind the phosphates. We get increasing enquiries about uranium recovery from phosphates and if there are any developments on this available, I would like to hear about them. I also would like to ask a very speculative question. I heard recently that a new idea was being entertained on the recovery of uranium from phosphates by a German group, I believe, in Morocco, using sea-water and an electrolysis process to concentrate the phosphate and then recover uranium through ion exchange. Has anyone any comment on this — is it something that might be important in the future?

F.E. MCGINLEY: I am not aware of the work that the Germans are doing. In regard to work that is going on in the USA, I do know that a number of phosphate companies are very active in this field. They are working at uranium recovery from a proprietary point of view with the thought that the technique or the process that each company is developing or utilizing might be of interest to other companies. Some of these companies are actively discussing their processes

with other potential producers in the USA. I think, as time goes on, more and more of the process information will be revealed. Another source of information, of course, are the patents that have been or will be applied for in the future.

W.R. FLÖTER: With regard to Mr. Cameron's question, I do not know very much about the project going on in Morocco, but I do believe that it is a process which is dealing with the recovery of secondary uranium after weathering of the phosphates from the ore; a study was made and it was found that the chlorine ion is useful in this process. Uranium would then be recovered by solvent extraction in a second step. I am not informed in detail about what is going on but I do know the gentleman who is working on that; his name is Kronjäger.

K. PINKAS: Concerning the recovery of uranium from raw phosphoric acid, a new beneficiation method has been developed (USA-Canadian patent) by which it is possible to precipitate uranium-IV in the ordinary way from barren solution after crystallization of mono-ammonium phosphate.

IN-SITU LEACHING
(Session VII)

IN-SITU LEACH MINING FOR URANIUM

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Abstract

IN-SITU LEACH MINING FOR URANIUM.

The first commercial mine for production of uranium by dissolving the element in chemical solutions injected into and produced from wells has been opened in Texas. This paper describes the geology of the region, the operation of the processing plant and the development of the mine. Injection wells, production wells and disposal wells are described. Hydrology studies conducted prior to start-up are outlined and environmental considerations and the development of baseline data are discussed in detail.

INTRODUCTION

Atlantic Richfield Company, Dalco Oil Company and U.S. Steel Corporation jointly own the Clay West mineral properties in Live Oak County, Texas. Over the last five years, Dalco and U.S. Steel Corporation have developed and pilot tested the successful uranium in-situ leach technology presently employed at the Atlantic Richfield operation. Atlantic Richfield is experienced both in Frasch sulphur mining, which employs water injection for recovery of sulphur from wells completed in the mineralized zone, and in conventional oil industry water flooding techniques. A commercial plant and field facility was designed and has been built for 250 000 lb of U_3O_8 per year with plans to expand production to 1×10^6 lb U_3O_8 per year after operating experience was gained. This is the only operating commercial sized uranium solution mining facility.

This paper deals with the design and operation of the quarter million pounds U_3O_8 per year unit. The facility is ten miles south-west of George West, Texas. Its cost was US \$7 $\times 10^6$ and the staff consists of 50 persons at the plant and additionally 12 supporting staff members at the Corpus Christi office.

ADVANTAGES OF LEACH MINING

Solution mining permits the exploitation of uranium reserves without disposing of overburden and without creating the open pits associated with strip mining. Solution mining is adaptable to exploitation of reserves below the water table, whereas this is difficult or impossible with underground mining. Solution mining does not have the depth limitations of strip mining in terms of overburden to ore ratio. Solution mines can be brought on stream in much less time than conventional mines and are not as labour-intensive or as fuel-intensive. We have found solution mining to be more environmentally acceptable than either strip or underground mining.

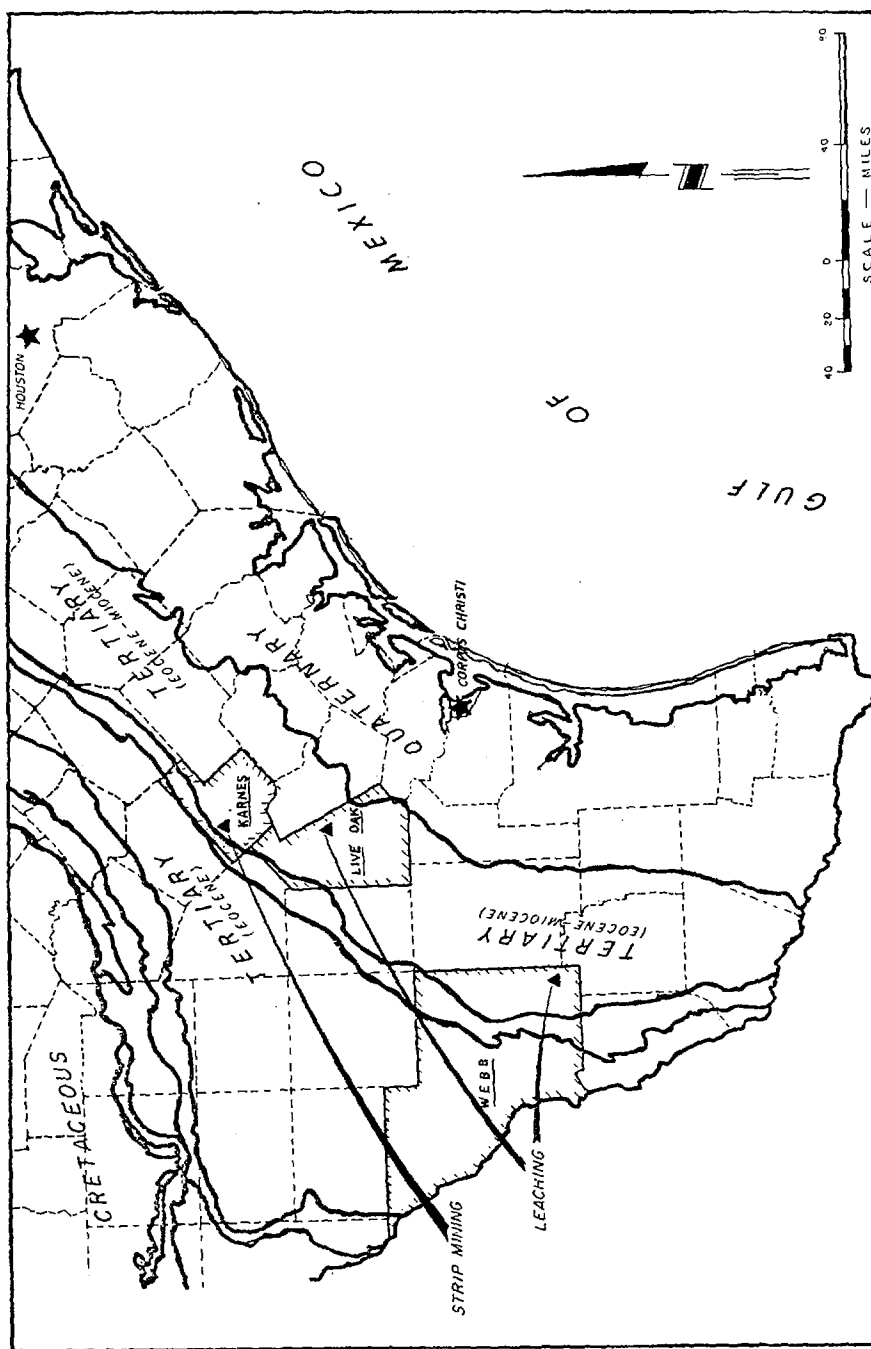


FIG. 1. Uranium exploration area in South Texas.

ENVIRONMENTAL CONSIDERATIONS

The Oakville formation is a freshwater aquifer and supplies the City of George West and other communities with drinking water. In the vicinity of the uranium reserve, the water is non-potable because of the naturally occurring high levels of radioactivity. It was necessary to develop safeguards which would assure that no irreparable damage to the aquifer occurred. The Operator worked very closely with the Texas Water Quality Board, the Texas Department of Health Resources, the Texas Water Development Board and the Texas Air Control Board to establish guidelines that would permit the production to proceed while fully protecting the environment.

Continuous monitoring of the zones around and above the leaching operation is required to assure that no solutions leave the leach area. When the area is exhausted, the reservoir solution must be returned to substantially its original condition.

An environmental task force was organized within Atlantic Richfield Co. to determine the overall baseline and monitoring requirements. The task force included chemical and petroleum engineers, geologists and hydrologists, zoologists, biologists and radiologists. The baseline study performed by an outside consultant included bacteria, insects, birds, amphibians and fish, roots and stems, grasses and shrubs, and soils and streams.

All water wells within a 5-mile radius of the operation were sampled and analysed before operations began. For the duration of the project, one-eighth of these wells will be analysed every three months, permitting a complete re-analysis of all wells every two years. These analyses include calcium, sodium, iron, molybdenum, bicarbonate, sulphate, chloride, nitrate, nitrite, ammonium, pH, and total dissolved solids. This list of ions is referred to as *List A*. The wells are also analysed for gross alpha and gross beta. A barium coprecipitation for radium-226 is performed whenever gross alpha values exceed 3 pCi/l. A lead-210 determination is performed whenever the gross beta exceeds 50 pCi/l.

Before operation, 28 wells in the area to be leached were analysed for List-A ions. Additionally, ten wells were analysed radiometrically and for heavy metals. These latter analyses were for magnesium, fluoride, arsenic, barium, boron, cadmium, copper, chromium, lead, manganese, mercury, nickel, selenium, silver, uranium, zinc, simple cyanide, P-alkalinity and total alkalinity.

Surface drainage is monitored by analysing chemically and radiometrically the local drainage (Spring Creek) above and below the extraction area and the Nueces river above and below its confluence with Spring Creek.

The present plan for restoring the orebody after leaching is completed is to pump contaminated water from the ore zone for disposal into a deep disposal well. This will allow the fresh water native to the aquifer to sweep the leached area, thus reducing ion concentrations to original levels.

GEOLOGY

Uranium was discovered in South Texas during an air reconnaissance survey in 1954. A radioactive anomaly near Tordillo Hill in Karnes County opened up a large uranium trend (Fig.1). This area had not previously been considered by geologists to be favourable for uranium exploration. The trend is approximately 500 km long. It parallels the coast in Texas and extends southward across the Rio Grande into Mexico.

The host rocks are sands and sandstones, generally containing some volcanic ash. Some lignites are present and permeability generally is high. The host rocks represent flood plain deposits, deltaic deposits or shallow marine lagoonal deposits.

A geologic cross-section is shown in Fig.2. The figure relates the Jackson formation, where strip-mining is currently taking place, to the Oakville and Catahoula formations, which are being leached. The Oakville formation has been strip mined at various places in Live Oak County. The regional dip of all shallow formations is approximately 80 feet per mile eastward toward the coast.

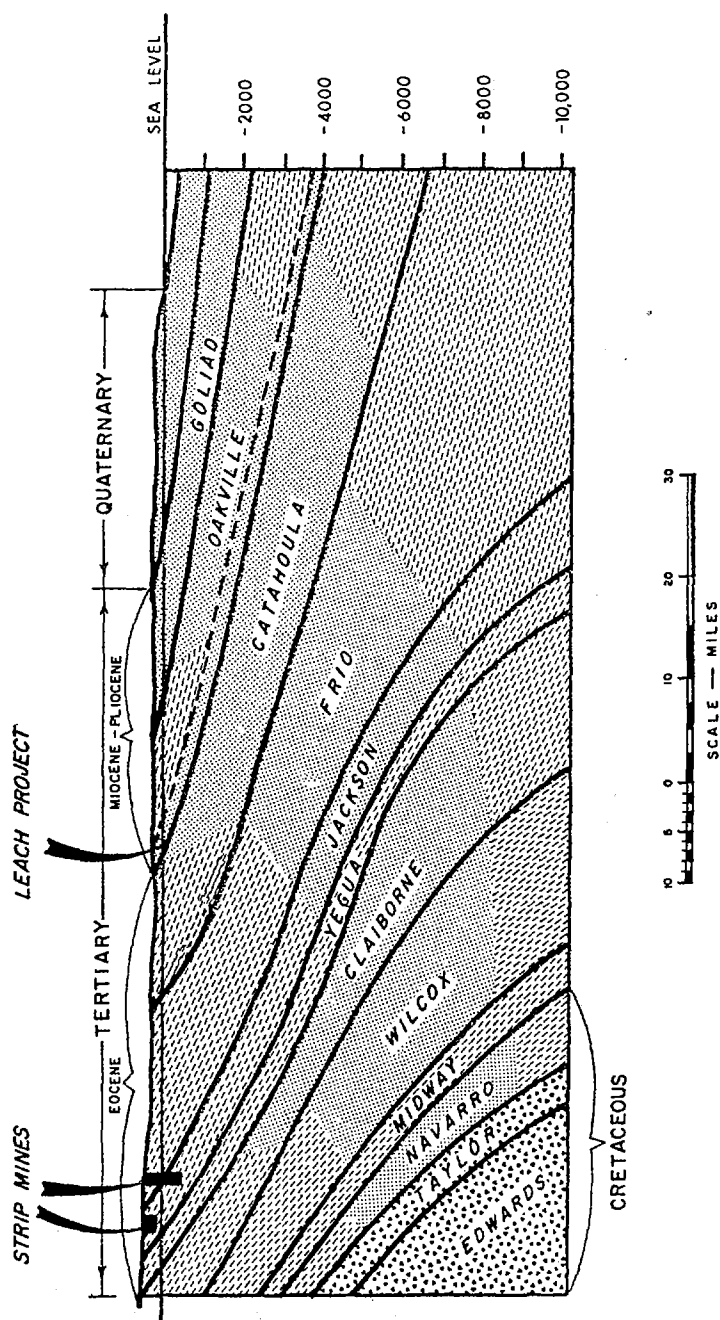


FIG. 2. Geological cross-section of uranium exploration area in South Texas.

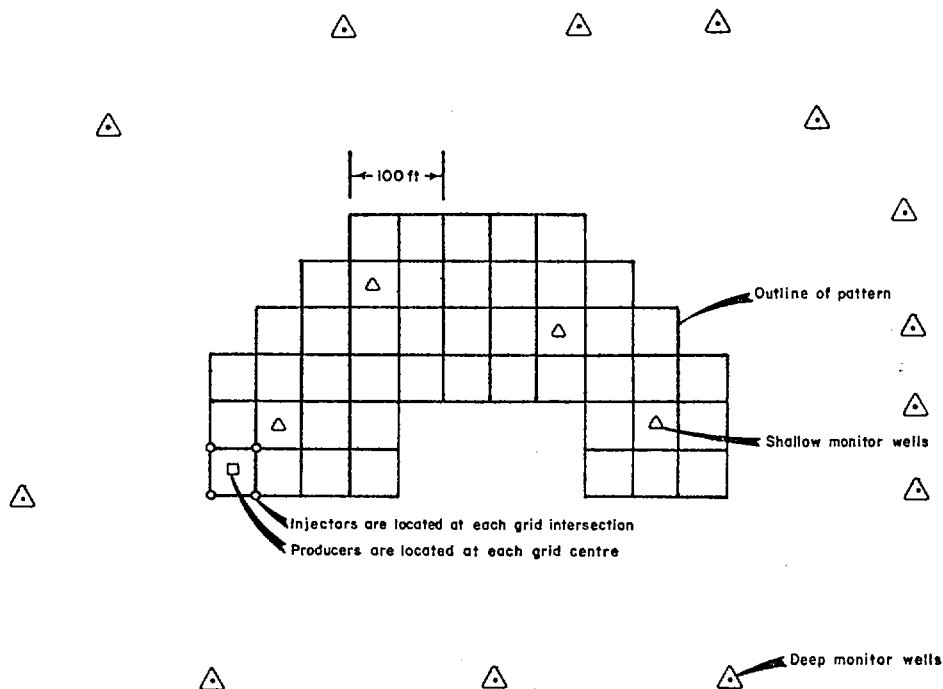


FIG.3. Clay West uranium leaching pattern.

FIELD DEVELOPMENT

The injection and production wells are arranged in a geometrical pattern as shown in Fig.3. The wells are about 450 feet deep on the average. The injection wells are located in a square pattern on 50-ft centres. The production wells are situated at the centre of each square formed by four injection wells. The injection wells are completed using 4-in. PVC schedule-40 plastic pipe. The production wells are 6 in. in diameter. The larger casing in production wells accommodates down-hole pumps. Both injection and production wells are equipped with well screens opposite the ore zones. These screens permit barren injection water to enter the formation, and solution rich in uranium to enter the well-bores of the production wells. The 46 patterns cover approximately three acres. The leach field is outlined by a ring of monitor wells which are sampled routinely to determine whether leaching solution has migrated away from the leach area. Four shallow monitor wells are placed above the orebody to detect the presence of leach solutions in shallower aquifers. Monitor wells are sampled and analysed for total dissolved solids, pH, uranium and fluid level.

If leach solution is found in a monitor well, the injection in that area of the field will be decreased while the production is continued. Hydraulics of the reservoir will then allow native reservoir fluid to sweep past the monitor well into the leach area, returning the migrated fluid to the patterns.

A diagram showing the construction of the field wells is shown in Fig.4. It is seen that the well casing is cemented from just above the top perforations to the surface of the ground. This cement seal prevents migration of leaching solutions up the borehole.

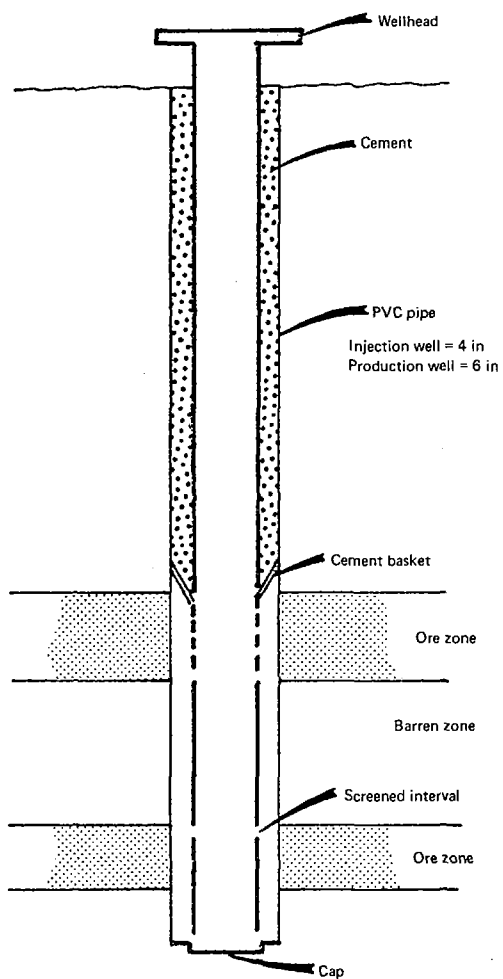


FIG.4. Injection and production wells.

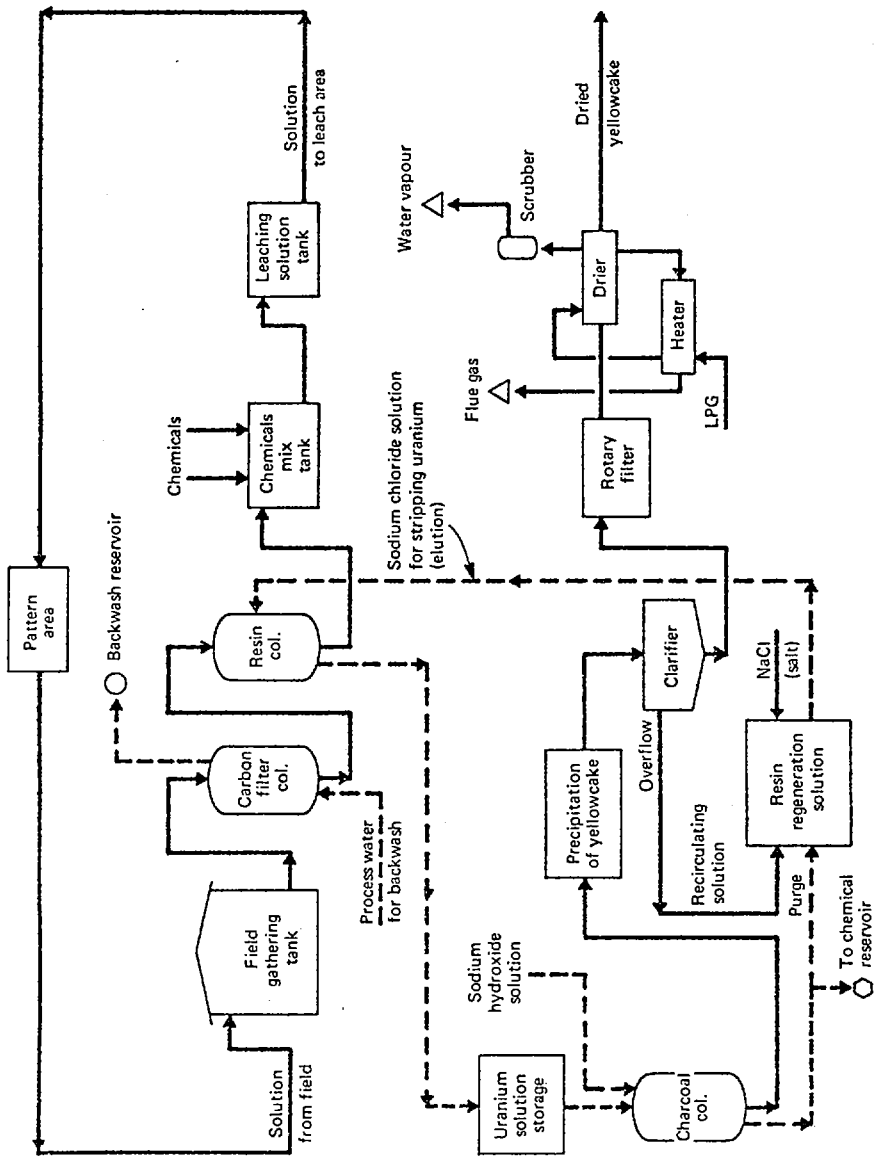


FIG.5. Layout of uranium recovery plant.

The production wells are connected by a system of lateral pipes and headers to a 12-in. dia. PVC pipeline, 6000 ft long, and to the uranium recovery plant. Barren solution from the plant returns to the field through a second pipeline where it is distributed to the injection wells through a system of headers and laterals. All headers and laterals are fabricated from PVC pipe because of its corrosion resistance, lower cost and ease of installation.

Injection and production volumes are carefully monitored and adjusted so that the injection into each five-spot pattern is balanced with the production from that pattern. Thus injection equals production for the entire set of 46 patterns minimizing leakage away from or into the leach area.

After leaching is complete and the aquifer has been fully restored, the wells will be filled with cement and the well casing cut off below plow depth. The area will be reseeded and in a short time no signs of a mining venture will be evident.

RECOVERY PLANT DESIGN

The block diagram of Fig. 5 represents the flow of material within the plant boundary. The field gathering tank is actually located at the field patterns. It provides surge capacity between the plant and field. The carbon filters in the plant remove silt from the well stream. Ion-exchange resin beads in the resin column remove dissolved uranium from the field stream. After removal of uranium from the circulating stream, it is reconstituted with chemicals for return to the leach area. The chemical treatment is proprietary and will not be discussed in detail.

Periodically, the flow is interrupted in each ion exchange column in rotation to permit elution of the mineral from the resin. Rich eluate is passed over charcoal to remove undesirable contaminants before the uranium is precipitated as yellowcake. Sodium hydroxide is used periodically to remove molybdenum from the charcoal. The yellowcake slurry is thickened, filtered, dried and placed into barrels for shipment. Propane for the dryer is the only fuel required for the plant.

TECHNICAL SUPPORT

A staff of reservoir and chemical engineers, geologists, environmental and administrative personnel are assigned as support for the project. Additionally, this support group can call on research chemists, computer analysts and other specialists as needed. Because of the newness of the technology, the availability of such support personnel contributes significantly to the success of the process. Operating experience and know-how gained during this start-up phase are extremely important.

DISCUSSION

A.W. LANKENAU: I have two questions. First, can you tell me the concentration of the solutions fed to the ion exchange columns and in the raffinate? Second, how do you remove the mud from your carbon clarification columns?

G.R. DAVIS: The uranium concentration will vary from well to well in the pattern area from sometimes probably as low as ten parts per million to several hundred. The mix of materials will vary from time to time in the 46 patterns. I think we have had concentrations as low as maybe 25 to 30 ppm and probably as high as 100 ppm. There are some operators who think that this can be increased very substantially and I think great progress can be made. As to the second question, how we handle the removal of the mud and the sand from the carbon filters. We have a backwash

system with a reinforced concrete reservoir and we pump about 800 gallons of water per minute from that reservoir into the bottom of the columns, countercurrent to the direction of flow. The material is washed back into the carbon backwash pond where it settles. The water in the carbon backwash pond stays there and we find that this is effective in removing the materials.

A.W. LANKENAU: Do you have screens on top of the column?

G.R. DAVIS: Yes we do.

A.W. LANKENAU: What is the concentration of the raffinate from the ion exchange columns?

G.R. DAVIS: The uranium leaving the columns varies from about 1 ppm to as high as 8 to 10 ppm.

P.A. BONHOTE: I was wondering why you put your plant so far away from the field. Is this because you are going to develop further areas at a later date? Secondly, do you have to pump the raffinate into the wells or is the pressure in the distribution line sufficient to force it back into the wells?

G.R. DAVIS: The plant site was selected to be fairly central to a number of known and future exploitable orebodies. The plant is a mile from this site in one direction but there are orebodies in the opposite direction also. On the second question, the plant site happens to be at a fairly high elevation. I think there may be 60 or 80 ft of elevation difference so the material enters the wells by residual head. It is pumped from the plant to the pattern area but there are no pumps in the field to inject it into the ground.

F.R. HARTLEY: I should like to ask whether you include in the lead time the development time for the hydrological and other investigations involved in the concept of in-situ leaching. Can you give us a figure of the development costs for the time between the start of investigations and the beginning of production?

G.R. DAVIS: The process had been tested on a pilot plant in 1973 and 1974 by Dalco Oil Co. and US Steel Co. on property belonging to Atlantic Richfield Co. I do not know the total development costs involved. There was of course laboratory leaching of ores that had gone on over the years; there was the pilot plant operation which probably lasted about a year. Once the decision was made to proceed, all the wells were drilled, and all the hydrological work was done, which was very short, incidentally. The hydrologists considered they had the most ideal arrangement for their studies. There were 100 plus wells with no injection and no production, so they could perform their pumping, make draw-down tests and injection tests and study the transients across the field with no interruption from any operation. For an expansion of the operation it will be different, because there will be ongoing operation then.

R.G. DAKERS: Have you experienced any plugging or reduced flow due to calcium precipitation in your production wells?

G.R. DAVIS: We have had worries about that, but we can calculate the solubility products to find out the saturation point. We expected to have problems but, so far, there have been none. However, I think there will be a time when we shall have them.

F.C. LENDRUM: Do you monitor gross beta and gross alpha radiation continuously and did you find any high radium values in the outlying wells which you surveyed?

G.R. DAVIS: The high radium values are of course associated with the uranium occurrence itself. I believe if we found high radium values in wells away from the pattern area we would consider that a potential mining area. We do not monitor gross alpha and gross beta continuously. This is done periodically.

W.R. FLÖTER: I have two questions. First, what acid concentration do you use? Second, what is the amount of horse power at the plant and in the field?

G.R. DAVIS: The formation is sufficiently carbonaceous for us not to use acid. This is an alkaline leach. I regret that I have to decline talking specifics on chemistry. On the second question: The pumps that are down holes have $7\frac{1}{2}$ hp each and we have 46 of them. The main pumps pumping from the field pumps and into the plant have 200 hp each, and we run only one at a time.

J.A. PATTERSON: Could you comment on the natural hydraulic gradient that exists in the area? Also, can you say something about the applicability of the method to different types of uranium ore deposits?

G.R. DAVIS: In the Oakville formation we expected the hydraulic gradient to be down to the coast, but it is not. It is parallel to the coast in this area and it is very slight and amounts to a movement of perhaps 40 ft per year within the reservoir. You can almost forget about it, I think. There is a difference of about $1\frac{1}{2}$ ft in static elevation from one end of the field to the other.

As regards the general applicability of the method, we have little experience other than with sandstones of relatively constant permeability. There have been some experiments in the Rocky Mountain area by other companies.

H.E. JAMES: Can you give us more details on the ion exchange system? Am I correct in assuming that you are using a fixed-bed column or is this a continuous ion exchange application?

G.R. DAVIS: It is a fixed-bed column, Rohm & Hass IRA 430. The reason why we are using the downflow fixed bed is that this was the technique used in pilot-testing the process. The mesh size of the resin is about $\frac{1}{16}$ in ($\frac{1}{16}$ in = 1.5875 mm). Our engineering people do not believe that there is any substantial advantage in deviating from downflow for a different facility.

H.E. JAMES: Would you not consider continuous ion exchange an advantage in a situation where there are various satellite plants around a central area with uranium being loaded onto resin which is then transported to a central elution system? And would this not change the economic picture?

G.R. DAVIS: Yes, it certainly could. We have considered trucking the resin from the satellites, physically moving the resin; we have considered changing to rich eluate and physically trucking the rich eluate, and we have also considered changing to slurry and physically trucking the slurry. It seems that moving the resin is about as economical as it is to carry out processing at the satellites.

M. SHABBIR: Could you please comment on the technique for recovery of uranium from the eluate?

G.R. DAVIS: We precipitate the uranium as ammonium diuranate. We transport it into a storage tank — a thickener — and then pump it from the bottom of the thickener through filters and washers. We repulp ahead of the drier.

M. SHABBIR: Do you have any molybdenum and vanadium present in your leach liquor or in the eluate? How about eliminating them?

G.R. DAVIS: We have a large amount of molybdenum in the circulating solutions. It is probably considerably less than the uranium, but because we remove the uranium, the molybdenum builds up preferentially in the circulating solutions. The molybdenum is quite concentrated in the rich eluate from the IX columns. The rich eluate is then denuded of molybdenum by being passed over a carbon bed. From time to time the carbon bed is regenerated to remove the molybdenum so that it can be used again and again for molybdenum removal.

RECOVERY OF URANIUM FROM SEA-WATER
(Session VIII)

RECOVERY OF URANIUM FROM SEA-WATER

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Abstract

RECOVERY OF URANIUM FROM SEA-WATER.

The possibility of extraction of uranium from sea-water on a sufficiently large scale to contribute significantly to national UK requirements is placed in perspective. It seems unlikely that there are sites around the UK coast where this could be achieved, and insufficient work has been done to be confident that sites exist anywhere to enable uranium extraction to be carried out on a large scale. Process techniques have been developed on a small scale, but extensive further research work would be necessary to reduce appreciably the present uncertainties. It would be unwise to expect uranium from sea-water to contribute significant amounts to the world's uranium demand for thermal reactors on an acceptable timescale.

INTRODUCTION

The extraction of uranium from sea-water offers the possibility of a virtually limitless supply of that mineral, with far-reaching strategic and other implications. It could be argued that as the grades of orebodies being exploited on land decrease, the prospects for sea-water uranium become enhanced. This review attempts to place those prospects in perspective.

SUMMARY OF EARLIER WORK

Following initial comments made at the third Geneva Conference on Peaceful Uses of Atomic Energy in 1964, work at AERE, Harwell, on the extraction of uranium from sea-water was reported by Davies et al. [1], including a scheme proposed by Hill and Johnson of the Engineering Group of the UKAEA. This was based on a hypothetical plant in the Menai Straits to produce 1000 t U/a, and the estimated production cost was such that interest has intermittently arisen in the possibilities. Later work by Keen [2] at AERE illustrated the feasibility of the absorption process on a small scale. Further process development work done at AERE on the absorption and elution stages has not been published.

Research and development in this area has been done by some other countries, notably by the Japanese who have been examining methods of extracting minerals from sea-water since the early 1960s. Progress has been made in the development of improved absorbents and it has been decided to proceed with a basic study aimed at the design of a pilot plant. As well as a tidal scheme, the recovery of uranium from the effluent from nuclear power stations is under consideration [3].

The original Menai scheme discussed by the UKAEA authors was subjected to critical appraisal in 1967 in the USA by a team of workers at the Oak Ridge National Laboratory but the report was only released in 1974 [4]. Assumptions different from those made in the original study resulted in very much higher production cost figures being deduced and suggested that the prospects of a large and economical supply of uranium from the sea were remote.

The status of the technology of uranium extraction from sea-water has recently been reviewed by Hill [5].

THE SEA AS A SOURCE OF URANIUM: AN EXAMINATION OF POSSIBLE SCHEMES

The concentration of uranium in the oceans appears to be reasonably constant at about 3 ppb (3×10^{-9}), representing a total uranium content in the sea of 4000 Mt. To extract amounts of uranium which would be significant in terms of consumption in a thermal reactor programme, enormous volumes of sea-water would have to be processed so that if pre-treatment of the water was deemed necessary (either chemical or physical), there would be major engineering difficulties. The dominant requirement in any scheme will be the ensurance that the plant is always supplied with new sea-water and that the processed water is not inadvertently recycled.

Three possible schemes which have received attention for the treatment of large quantities of sea-water are discussed below in order of potential extractive capacity, but it should be noted that a number of others on a large and small scale have also been proposed.

(a) The possibility of a combined desalination and uranium extraction plant was assessed by Khan [6]. The uranium production of the largest desalination plant under consideration is only 1 t/a.

(b) Recovery of uranium from the cooling water used in coastal power stations could yield up to 50 t/a from the UK fossil-fired stations and up to 23 t/a from the UK Magnox stations. Consideration of the more generalized data given by Scriven et al. [7] indicates that the uranium content of the cooling water of nuclear power stations represents about 5% of the uranium used in that reactor. Some Japanese workers have recently quoted a possible production of up to 15% of the uranium input into their reactors, which is interpreted as meaning that their cooling water usage is much higher.

(c) It has been suggested that uranium extraction could be associated with tidal power facilities. Nearly 100 sites around the world have been identified as suitable for the construction of such sites. Ignoring all technical aspects of the extraction process, the total uranium content of the volume of sea-water involved in these schemes has been estimated as 2.5×10^4 t/a [8], which is a significant amount, but it is only about 10% of the anticipated annual world demand by the year 2000. Power schemes require a large tidal range and large barrage capacity — common features with any extraction scheme — but the factors which provide large tidal ranges tend to conflict with the water flows necessary to ensure adequate replenishment. The UK tidal schemes which have been proposed are poor in this respect.

The estimated conceptual capacities of combined schemes are summarized in Table I. It seems from the above that combined schemes are inadequate to meet the uranium requirements of a thermal reactor network and that purpose-designed and constructed projects using either pumped or natural flow in well-selected locations must be considered.

Other more imaginative schemes have been given some preliminary thought, for example, in connection with floating and island concepts, including the use of thermal and wave power. Insufficient consideration has been devoted to such ideas to enable a realistic assessment to be made of them, but the development periods required would undoubtedly be long.

ENERGY BALANCE ANALYSIS OF THE EXTRACTION OF URANIUM FROM SEA-WATER

Some recent debate in the United Kingdom has centred on the energy balance of large projects and although the discussion has brought out many limitations in the approach, it was felt that it might be useful to carry out a rough energy balance of the extraction of uranium from sea-water as a guide for the justification or otherwise of further consideration of the feasibility of the scheme. It might also enable a comparison to be made between sea-water extraction and the mining of uranium in low-grade ore deposits. Sensitivity analysis of the figures obtained could also give some

TABLE I. ESTIMATED CONCEPTUAL CAPACITIES OF COMBINED SCHEMES FOR URANIUM EXTRACTION

	Water flow (m ³ /d)	Uranium production (t/a)
Desalination	1×10^6	1
'2000 MW' station (fossil)	5×10^6	5
All CEBG Coastal (fossil)	—	50
Magnox — Dungeness (410 MW)	2.5×10^6	2.5
All Magnox	—	23
Severn barrage	1.6×10^9	1.6×10^3
World tidal power projects (except USSR)	—	2.5×10^4
Anticipated world demand		3×10^5 t/a by end of century

indication of the most important components of the scheme and illustrate how further work might be oriented in order to reduce the uncertainties attached to these aspects. The energy analysis was carried out by Taylor and Walford of the Programmes Analysis Unit and a report on this work will be generally available [9].

Two energy accounting approaches were adopted: (i) energy costs, determined in kW·h of primary energy per £ value of the inputs from the main industrial sectors involved in the scheme, based on conversion factors derived from the UK Input-Output Tables for 1968 [10], and (ii) a method based on the energy cost of materials, calculated by integrating the inputs of primary energy required to produce these materials. Both tidal and pumped schemes, based on the earlier UKAEA work on a 1000 t U/a plant, were considered in the analysis. Many of the estimates were extremely crude but this is not considered a serious disadvantage in view of the uncertainties attached to so many aspects of the engineering, design and operation of the schemes. Considering the approximations used, satisfactory agreement was achieved between the two approaches and it was possible to interpret to some extent the discrepancies observed. The best estimates of the energy costs of extracting uranium from sea-water for a pumped scheme and a tidal scheme were then deduced and are given in Table II.

It must be stressed that these figures are based on many assumptions regarding the civil engineering aspects and process technology. Although information exists on absorber performance on a small scale, an engineering study has not been done. It is interesting to compare the total cost figures given in Table II with the energy cost of 15.8 GW·h(th)/t U (later amended to 7.1 GW·h(th)/t U) extracted from Chattanooga shales derived by Chapman and Mortimer [11].

The energy costs of inputs into the nuclear power cycle are summarized in Table III for a SGHWR system, no plutonium credit, and uranium extracted from sea-water.

The thermal output of 1 tonne of uranium fuel of this enrichment (see Table III) is 22 000 MW·d, which equals 528 GW·h(th) so that the energy gain ratio for uranium from sea-water then becomes 6.4 for the pumped scheme and 11.7 for the tidal scheme. This indicates a marked net energy gain which can be achieved using the process, but we can take the argument one stage further. At 30% steam conversion efficiency and 15% internal electricity consumption, including distribution costs, the electricity output from 1 tonne of enriched uranium in a SGHWR is $528 \times 0.3 \times 0.85 = 135$ GW·h(e). Therefore, for every kW·h(th) of fossil fuel used in the construction of the total system for the production of uranium from sea-water and its subsequent

TABLE II. ENERGY COST ESTIMATES FOR URANIUM EXTRACTION FROM SEA-WATER^a

	Energy costs (GW·h(th))	
	Tidal scheme	Pumped scheme
Capital		
Construction	8414	4556
Initial absorber	<u>826</u>	<u>826</u>
	<u>9240</u>	<u>5382</u>
	Energy costs (GW·h(th)/t U)	
	Tidal scheme	Pumped scheme
Production		
Capital/Maintenance	0.510	0.317
Absorber	0.083	0.083
Eluent	3.900	3.900
Pumping	<u>0.017</u>	<u>10.661</u>
	<u>4.510</u>	<u>14.961</u>

^a For an explanation of this cost base see text.

TABLE III. ENERGY COSTS OF INPUTS INTO THE NUCLEAR POWER CYCLE FOR A SGHWR

Inputs	Energy costs (GW·h(th)/t enr. U)	
Enrichment	22	
Materials	1	
Capital	6	
	Pumped scheme	Tidal scheme
Ore (yellowcake)	53	16
Total	82	45

use in the nuclear reactor, $135/82 = 1.65 \text{ kW} \cdot \text{h(e)}$ (pumped scheme) or $135/45 = 3.0 \text{ kW} \cdot \text{h(e)}$ (tidal scheme) of electricity is available. This compares with $\sim 0.25 \text{ kW} \cdot \text{h(e)}$ per $\text{kW} \cdot \text{h(th)}$ as the average ratio of output to input for the UK generating system in 1972, as calculated by Chapman et al. [12].

From Table II it can be seen that the most sensitive aspects of the two extraction schemes are the amount of eluent required and the pumping power needed in the pumping scheme. Even with a doubling of the energy costs under these two categories the process still exhibits favourable gain characteristics.

The rough energy accounting exercise carried out, therefore, indicated that a considerable energy gain might be achieved through extraction of uranium from sea-water, but further work is necessary to verify that the assumptions made can be attained in practice and on the required scale.

POSSIBLE EXTRACTION PLANT SITES

When a site is being sought that would contribute a significant proportion of the uranium needed in the United Kingdom or a similar proportion of that required in any other country, a number of stringent conditions have to be met. A large net flow of constant current would be necessary, equivalent to an amount greater than the combined flows through the Straits of Dover and the Irish Sea for a plant of 10 000 t U/a. The overall plant layout would have to ensure that there is an adequate inflow of fresh sea-water and no likelihood of the 'depleted' sea-water being recirculated through the plant. The performance of the absorber bed would be affected by the solids content of the sea-water, in the form of silt and organic matter, and by temperature fluctuations in the water. Other aspects which would need consideration in the determination of a suitable site are discussed elsewhere in this paper.

A review of the available oceanographic information was carried out by Haigh of the CEGB [8] in order to identify sea areas which meet some or all of these criteria. Possible sites in British coastal waters were examined in some detail and land masses in the North and South Atlantic, the Indian Ocean, and the North and South Pacific were also studied.

Haigh concludes that the low surface current velocities around the British Isles and the strong dependence of the surface current direction on the prevailing wind probably precludes the existence of UK sites meeting the sea-water replenishment criterion, at least for a plant of large capacity. On a world basis, it is suggested that further consideration could be given to sites in the Bahamas where the constant Florida and Antilles currents have favourable characteristics and where the relatively high temperature would assist the absorption stage. Such sites could be particularly relevant to proposals for ocean-located concepts. Much of the available oceanographic data on the North Pacific Ocean indicates that the Kuro Shio current meets many of the criteria.

Hill [5] has recently argued that insufficient oceanographic data are publicly accessible to evaluate systematically the availability of promising sites and pointed out that most circulation data are in the macroscale whereas extraction plants require data going beyond the existing microscale data.

It seems that sites probably do exist where conditions are favourable for the extraction of large amounts of uranium from sea-water, but insufficient evaluation work on candidate areas has been done to be confident that this is so, particularly in relation to the characteristics which would ensure that an adequate inflow of fresh sea-water will be available. Political factors will affect the eventual decision on the large investment that will be needed.

ENVIRONMENTAL ASPECTS

Any project likely to produce significant amounts of uranium from the sea would involve the enclosure of large areas of sea-water, probably adjacent to a considerable stretch of coastline. It would be unrealistic not to expect strong opposition on general environmental grounds, but no methodical assessment of the possible effects in this category seems to have been made. Some preliminary discussions have been held in the United Kingdom and a number of aspects have emerged from which some general conclusions can be made.

(a) It has been estimated that for a tidal lagoon scheme the total area of enclosed water for the production of 10 000 t U/a could be 300–400 square miles. The walls forming the seaward

boundary of the lagoon would be low and would have little visual impact, but the larger dams containing the absorber beds would be more intrusive. These dams might also accommodate the chemical plants for elution operations, which, alternatively, could be sited nearby. Such a large complex could have effects on inshore fishing, shipping channels, established currents, erosion, and sedimentation over a wide area. For a pumped-water scheme, the number of dams required would be less and the beds could be deeper in view of the greater head of water available, thereby reducing the total area affected by the scheme. On the other hand, there would be visual disamenity owing to the power supply required, especially if this was provided by an on-site generating station.

(b) There will be interactive effects between the process and the 'biological productivity' of the sea-water, which can vary by three or four orders of magnitude over the year at certain points of the UK coast. The biological activity in the relatively stationary water of a lagoon scheme could be significantly different from that in the surrounding open sea and could result in embarrassing build-up of plankton and other material on the beds. Elution would kill the plankton but the accumulated solids and the associated growth would still require removal (and disposal) to ensure maintenance of the necessary pressure drop across the bed. If growth of plankton blooms occurred in the stationary water the position would be aggravated.

(c) The environmental effect of discharging these residues would probably be small and the preliminary opinion is that destruction of the plankton would not have a serious ecological effect as it would be capable of rapid replacement.

(d) Effluents from the process will consist of finely divided hydrous titanium oxide, calcium and magnesium carbonates, dilute sodium carbonate, bicarbonate, and nitrate solutions. None of these effluents is highly toxic and it is not envisaged that they will be persistent although it will probably be important that the effluents are well mixed with large volumes of sea-water to avoid local high concentrations. Both the economics of the process and the environmental problems would provide strong incentives to reduce these effluents as the embryonic technology develops. With the technology in its present state of development, it has been estimated that perhaps as much as 10^7 t/a of finely divided solids could be deposited over a wide area of sea-bed for a plant of 10 000 t U/a capacity. Although the total solid discharge is much higher, the total area of sea-bed involved is so large that the effects from the wastes per unit volume of sea-water are not altogether dissimilar to those previously experienced by industry when the main effects were to smother the sea-bed, thereby making it less suitable for burrowing organisms and sea-bed feeding fish. The consequences included marked local effects on the flora and fauna but there is no evidence of physical damage to fish as a consequence of particulate suspension.

Summarizing, it seems probable that a large-scale uranium extraction plant of this nature would involve a major biological management effort both within the process and in its environmental consequences. The extent of the measures required could only be assessed in relation to a specific site but there is no reason to believe that there would be any biological problems that could not be overcome.

STATUS OF PROCESS DEVELOPMENT

Many of the uncertainties associated with the project have been discussed above, viz. environmental aspects, energy requirements, site characteristics and availability, etc., all of which would need further research before a decision to proceed with a large project could justifiably be made. Not unexpectedly, there are also major uncertainties associated with the extraction/elution process itself.

(a) Keen has developed a form of hydrated titanium oxide, following the work of Davies et al. This material absorbs the uranyl ion; its performance, although dependent on temperature, is satisfactory over the probable operating temperature range, and it has a low solubility in water. The new material for preparing the absorber is available at a realistic cost and in plentiful supply. Further

study is necessary to determine the optimum size of absorber particles and whether there would be chemical or physical advantages in using large coated particles. Other absorbers have also been suggested, particularly by Japanese workers.

(b) The size of absorber particles will affect the operation of the bed in several important respects, viz. whether it will blind, with ensuing loss of extraction efficiency, and whether a pre-filter will be necessary to remove plankton and other fine solids. The particle size will govern the optimum design and arrangement of the beds.

(c) Take-up of calcium and magnesium ions on the bed could constitute a serious problem, especially at the elution stage when finely divided calcium and magnesium carbonates are precipitated and could be washed out into the environment in large amounts.

(d) The best elution chemical has to be determined. Early work at Harwell indicated that ammonium carbonate was a satisfactory eluant but later studies suggested that sodium bicarbonate could give improved operating characteristics. As seen earlier, the loss of the elution chemical could be one of the factors of major economic sensitivity and it will be important to optimize the efficiency of the elution stage.

CONCLUSIONS

(a) Process techniques have been developed on a small scale which demonstrate the possibility of extraction and subsequent recovery of uranium from sea-water under the right conditions.

(b) There are a number of uncertainties associated with the process and extensive further research work would be necessary to reduce appreciably the level of these uncertainties.

(c) Uranium extraction combined with other proposed schemes handling large volumes of sea-water (desalination plants, coastal power stations, barrages) would only have a marginal effect on the expected demand of uranium to feed thermal reactors.

(d) On the assumptions made, a rough energy accounting exercise indicates that a considerable gain might be achieved through extraction of uranium from sea-water, even for a pumped system.

(e) It seems unlikely that there are sites around the UK coast where uranium extraction could meet UK demands.

(f) Some sites probably exist, around the world, where conditions are reasonably favourable to the extraction of uranium from sea-water on a scale significant in relation to large thermal reactor programmes. However, insufficient work has been done to be confident that this is so, particularly in relation to the characteristics determining whether adequate inflow of fresh sea-water will be available.

(g) It seems probable that a large-scale uranium extraction plant would involve a major biological management effort both within the process and in its environmental consequences. The extent of the measures required could only be assessed in relation to a specific site but there is no reason to believe that there would be any biological problems that could not be overcome.

(h) Most of the work done to date must be regarded as of a preliminary nature and there are so many uncertainties associated with the project that it would be unrealistic to attempt to quote even an approximate estimate of the likely cost of extraction.

(i) It would be unwise to expect uranium from sea-water to contribute significant amounts to the world's uranium demand for thermal reactors on an acceptable time-scale. New deposits must be found elsewhere, including low-grade orebodies which will be a more certain source of supply in the foreseeable future than sea-water uranium.

(j) In the long term, assuming the adoption of the fast reactor, uranium from sea-water could contribute to the much lower anticipated uranium requirements, possibly through a number of small schemes combined with power production. Such requirements could be the uranium needs of a decreasing thermal reactor component within an expanding world fast reactor system or applications for which uranium has special advantages, e.g. small power plants.

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B. Cutts	UKAEA
J.W. Davies	UKAEA
C.P. Haigh	CEGB
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URANIUM FROM SEA-WATER: POSSIBILITIES OF RECOVERY, EXPLOITING SLOW COASTAL CURRENTS

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Abstract

URANIUM FROM SEA-WATER: POSSIBILITIES OF RECOVERY, EXPLOITING SLOW COASTAL CURRENTS.

The authors analyse the interest in uranium recovery from sea-water within the framework of uranium world supply problems. The most reliable methods proposed for recovery are summarized and discussed, both from the chemical and the plant project points of view. Tides as a source of energy for water movement cannot be used in the Mediterranean and therefore only currents can be taken into account. The acceptable cost of an exchanger, in relation to the uranium price, is considered and related to known exchangers.

The characteristics of exchanging elements are examined and the influence of the speed of sea currents discussed. The extractable uranium is a function of the exchange rate and of the speed of the flow inside the exchanging system; therefore it is quite clear that the current speed is not a prerequisite and that coastal currents around Italy are suitable. Exchanging elements built with sheets parallel to the flow, exchanging pans containing granular or fibrous exchangers have been considered. The main characteristics of a 1000 t/a plant are discussed considering different possibilities. The most acceptable seems to be the continuous extraction system. The parameters needed to calculate the dimensions of such a plant are given and the relation between the length and speed of the moving chain discussed. A rough economic evaluation of the plant cost — starting from known technologies — and of the final cost of the uranium oxide produced is made.

1. INTRODUCTION

The year 1973 can be considered from an economic point of view the end of a cycle if not of a historical period. The epoch of low-priced raw materials for energy is now over and so are the low price of gold and the cheap and steady price of industrial animals, vegetables and mineral materials. The market is now a seller's market. Such events not only had a serious impact on the stability and type of economy but even on the model of society we are now living in and on the relations between individuals and on the role of the individuals in that society. World economy has been obliged to react to the new situation by taking into account new industrial processes and technologies and sometimes even decreasing levels of production.

Electrical utilities in countries like Italy, strongly dependent on crude oil as a means of generating electricity, have been obliged to absorb an increase in costs of 400% for fuel and therefore to find short and middle-term alternative sources to overcome the present difficulties and to be protected in the future from events such as those of 1973. Apart from coal, the most widely accepted solution by far to the energy problem is massive electronuclear production. It is not our aim to analyse the different aspects of such a choice, but it must be borne in mind that the nuclear fuel — uranium — is so widely diffused over the earth and seas that practically every nation with a developed technology and the available capital can make itself independent of foreign sources.

This communication is a part of a series of studies undertaken by our institution (CNEN) on the long-term possibilities of the extraction of uranium from sources with a wide geographic distribution and on the methods that are, at the present stage of technical development, more reliable and economically acceptable.

We have examined in detail the work done by different laboratories to develop methods of extracting uranium from sea-water, considered as a low-grade source — a class-B source according to the accepted terminology. The exploration of this source is primarily justified by the following considerations:

- (a) The price of uranium is continuously and constantly increasing and is now around \$25/lb U_3O_8 .
- (b) Since 1 October 1975 nuclear reactors with a total power of 370 000 MW are in operation, construction or firmly ordered. If we take as a rough basis a cumulative need of 5 tonnes of uranium for the inventory and refuelling over 30 years for each MW installed, this results in an overall requirement of 1.85 million tonnes. This amount must be related to the known (measured and assumed) reserves that can be mined at a cost of up to \$30/lb, amounting at present to 3.4 million tonnes, half of which is assumed.
- (c) The recovery of uranium from sea-water, as we will discuss later in some detail, is mainly a problem of exchangers and of engineering and it is highly probable that a careful assessment of the parameters of the problem could reduce the production costs to levels not far from the present market prices and very likely to the levels attainable in the next few years.
- (d) The availability of a reliable technology would enable our country to produce uranium and, moreover, to exchange such a technology with other countries, since such technical operations could easily be applied to the recovery of other metals from dilute solutions.
- (e) Finally, the cost of recovery of the huge uranium reserves in the sea will be the maximum attainable price to be considered in future forecasts of the price of uranium. Furthermore, one must not overlook the possibility of social and political events prevailing on economic factors and so blocking other uranium sources.

The above-mentioned considerations can be made more clear by observing that the amount of uranium carried by a current of 1 knot (2 km/h) flowing across a section of 1 km² is about 6 tonnes per hour. That is to say, more than 50 000 tonnes of uranium flow over the same section in one year — sufficient to supply ten 1000 MW(e) nuclear reactors throughout their life. The state of the art in the field of exchangers is such that exchangers are now available, the cost of whose basic materials is in the range of 16–63 £/kg and which can fix from 0.5 to 4 kg of uranium per tonne. values that are similar to the grade of conventional uranium ores. As a consequence it seems reasonable to affirm that countries with no significant uranium resources could produce at least part of their uranium requirements by exploiting sea-water, utilizing partially new technologies. In our opinion the main problem is now the applied technological and engineering research needed to lower the costs.

Many methods to recover uranium from sea-water have been proposed by different research groups:

- (a) Precipitation in open basins saline-like, or biological depuration systems using hydroxides or phosphates of aluminium and iron as precipitating agents, or using synthetic or natural exchangers in suspension;
- (b) Liquid-liquid extraction by organic solvents and foams;
- (c) Living marine organisms;
- (d) Ionic exchange with selective organic or inorganic exchangers submerged in moving sea-water.

In this paper we shall discuss features and engineering problems related to method (d) because such exchangers have been well studied and the proposed applications are at the pilot-plant level, while the first three types are still at the stage of basic research. At this point it seems useful to recall the main research groups that are, or have been, engaged on this subject.

(A) United Kingdom

A series of research projects with essays in a pre-pilot plant were carried out between 1962 and 1968 by the Atomic Research Establishment, Harwell, and Engineering Group, Risley. A plant with a forecast production of 1000 t U_3O_8 per year has been projected utilizing the tides and current of Menai Straits near Anglesey Island (North-West Wales). The exchanger selected was titanium hydroxide, but many others have been tested. The projected plant includes a series of dams like that of the tidal power plant at Rance in France. Amortization periods are fairly long and the production costs are evaluated at \$11–22/lb. However, a more recent evaluation made by British researchers in 1968 showed double these costs.

In the same period the UKAEA suspended such research not strictly related to reactor activities because of the lack of demand on the uranium market, the presence of USA uranium stockpiles and the dubious competitiveness of nuclear-produced electric power. Obviously, today such considerations seem merely historical.

(B) Japan

In Japan the Reactor Institute in Tokyo, the Institute of Industrial Sciences of the Tokyo University and the Japan Monopoly Corporation began systematic research in 1966. The Japanese have re-examined globally various problems such as solvent extraction, foams, biological and low-cost exchangers such as activated coals, lignin and galena, and improving the methods of preparing titanium hydroxides. A recent cost estimate gave a range of \$46–76/lb. The Japan Monopoly Corporation is now studying the problem in a small pilot plant.

(C) Federal Republic of Germany

West Germany has supported the activity of some research groups linked to the Jülich GmbH. The use of synthetic ultramarine has been studied, but this material requires a slightly different pH from that of sea-water and its use is therefore restricted to closed basin systems.

The same centre has supported research on select strains of algae able to concentrate uranium and promising results seem to have been obtained.

(D) United States of America

In the USA the ORNL published a report on the same subject in 1963. Very recently private US industries have re-examined the problem and indicate a wide cost range of \$30–1000/lb, according to method with a good probability for the lower level.

(E) International organizations

The OECD, and more recently the IAEA, have shown interest in promoting such studies.

The problem of Italy, to which our paper mainly relates, is strongly conditioned by the fact that tides are very low and the contact between the uranium-bearing marine waters and the plant must be obtained by other means than tides and dams.

Many possibilities theoretically exist:

- (a) Waters of the cooling systems of coastal industries and nuclear reactors
- (b) Low-cost pumping by means of solar or other low-cost energy
- (c) Floating structures in slow movement
- (d) Marine currents.

In case (a) the amount of uranium recoverable can be of only a few tens of tonnes per year. Roughly speaking, it is possible to obtain 1 tonne of U per year for each 10 m³/s of flow. In the

case of fast breeder reactors this amount would be enough to provide for the refuelling of the plants. Case (b) is related to progress in the utilization of solar energy. Case (c) has never been considered and is outside the scope of this paper.

We now shall examine in detail case (d) in relation to Italy in particular, and in general to every country with the same marine conditions. The following sections will examine:

- (1) The amount of uranium carried by the marine current surrounding the Italian Peninsula
- (2) Technical and economic factors conditioning the possibilities of recovery, i.e. the exchangers and the type of plant
- (3) The technological research needed to improve the basic understanding and to realize a pilot plant.

2. URANIUM RESOURCES IN ITALIAN SEA CURRENTS

The Mediterranean Sea, with an area of 3.77 million km³, is the second largest land-locked sea in the world and practically a closed basin. Assuming an average uranium content of 3 t/km³, it contains 11 million tonnes of uranium, which are renewed by the current flowing through the Straits of Gibraltar (1.75 million m³/s inflow and 1.68 million m³/s outflow) at a rate of 0.21 kg/s, that is to say 6.630 t/a. The balance of water exchange with the Black Sea is practically negligible, being less than 0.7% of that through the Straits of Gibraltar. The above-mentioned calculations and considerations clearly show that even the extraction of some thousand tonnes of uranium a year will not significantly alter the chemical composition of the sea-water and therefore no ecological problem can be foreseen. Moreover, uranium does not seem very important in the metabolism of any known species of sea animals or algae: only a few selected strains of them seem to concentrate uranium.

Uranium occurs in sea-water mainly in solution, in the chemical form of a tricarbonat complex ion, and its distribution in different seas is rather constant. The diffusion coefficient of uranium in sea-water, as measured by different authors, ranges from 1.3×10^{-6} to 3×10^{-6} cm²/s. This means that the mixing of uranium is rather fast.

As a basis for further calculations Fig. 1 shows the amount of uranium brought from a sea current of a given speed. The ordinate gives the amount of uranium in kg/m²·a and the abscissa the current speed up to 1.5 m/s (about three knots). This speed range was chosen because the average speeds of the Adriatic and Tyrrhenian current loops are included in this range. Current flows between 0.2 and 6 knots are reported for some Italian Straits in the specialized literature.

The values of Fig. 1 are related to a uranium content of 3×10^{-3} g/m³ to take into account anomalous values and non-exchangeable uranium, although the most reliable analysis gives a content of 3.3×10^{-3} g/m³. The flux of sea-water that can be utilized is that inside the exchanging system and therefore the true current speeds are only an indication for any real calculation.

The seasonal and daily variations of the currents in the seas adjacent to the Italian Peninsula are not well known. The values indicated have been given to compare them with the operating parameters of our working hypothesis, and to give a rough idea of their quantitative effects. It is, however, evident that a plant planned to extract 1000–2000 t/a of uranium will be considerably undersized in respect to the uranium flowing across it if the plant is placed in a strait with a flow of 10 000–15 000 t/a of uranium.

3. ECONOMICS OF THE PROCESS

For the recovery of uranium from sea-water many ion-exchanging materials have been studied with a fixation time varying from a few seconds to 30 days according to the physical form of the exchanger. The concentration can reach a ratio of 0.5–4 kg U/t exchanger. This ratio is similar to the grade of the best minerals found.

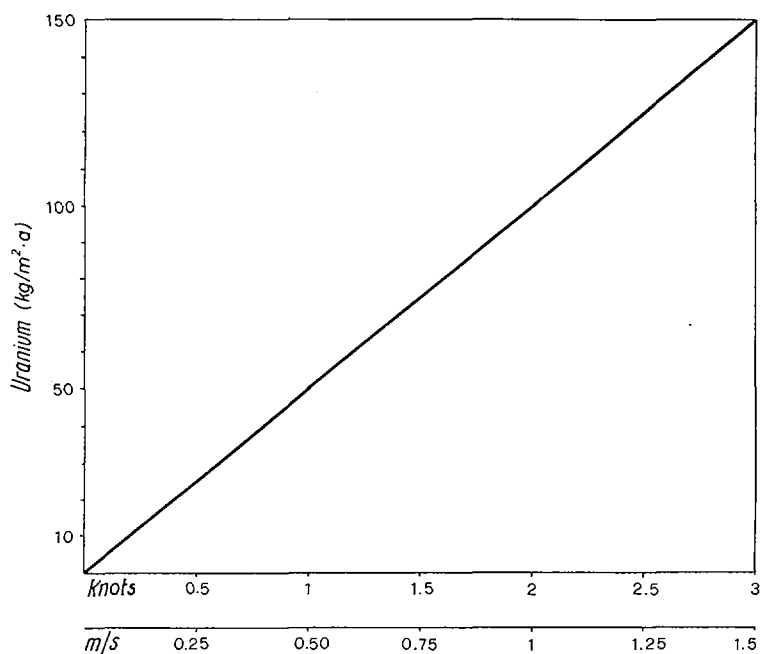


FIG. 1. The amount of uranium passing over a surface of 1 m^2 , as a function of the current speed.

The economic viability of such synthetic minerals as against normal ones is mainly related to factors as the cost of the raw materials from which the exchanger is synthesized, the cost of the process needed to activate or improve it, the type of plant needed to realize the contact exchanger/sea-water, the time of contact water/exchanger needed to obtain an economically profitable extraction and an optimization of the whole extracting cycle. To roughly evaluate such costs we shall examine at first some factors and in particular the amount of exchanger needed to recover a given amount of uranium, the amount of investment available per tonne of exchanger if an economically valid process must be set, the characters of the plants to be built and the likely yield.

Exchanger

Let Q_v be the amount in kg/m^3 of exchanger to be set in a section perpendicular to the flux of sea-water. We then have:

$$Q_v = Q \cdot V / C_s \cdot C \cdot n \cdot h$$

where Q is the amount of uranium flowing in one time unit through a surface unit carried by a current of one speed unit. V is the speed of the current calculated at the outlet of the exchanging system, which we assume to be 70% of the speed of the sea current; C_s is the ion-exchanging capacity of the materials utilized in kg of uranium per kg of exchanger; C is the percentage of

TABLE I. CHARACTERISTICS OF PROPOSED EXCHANGERS

Exchanger	Exchange capacity	Granulometry (mm)	Saturation value (d)	Elution	Regeneration	Cost (\$/kg)	Remarks
Titanium hydroxide	0.55 1.5	0.15-0.1 0.15-0.1	30 2	(NH ₄) ₂ CO ₃ -1.5M	- -	1.2-1.5	Flux 0.42 cm/s
Galena	3.8	0.1-0.076	<1	Na ₂ CO ₃ -10%	HCl pH 5	0.22	
Lead pyrophosphate	0.5	0.15-0.1	21	HCl	-	0.44-0.51	
Activated coal	0.5	0.15-0.1	1	-	-	0.3-1	
Cellulose	0.3	0.15-0.1	1	-	-	0.25-0.26	
Zeolites ultramarine	0.4-4	-	<1	Na ₂ CO ₃ -1.1M	-	0.22-0.30	pH 7-5
Diatomite	?	-	?	?	?	0.02-0.09	
Pozzolan	?	-	?	?	?	0.01-0.02	
Ca phosphate	0.1-0.05	-	1	?	?	0.26-0.28	

the exchange capacity utilized during the time of contact water/exchanger; n is the number of annual cycles (charge + elution + regeneration) obtainable with a given amount of exchanger, and h is the thickness of the exchanging bed.

The decrease in C_s during the repeated cycles is, as a first approximation, neglected; the relation between C_s and \bar{C} is really of an exponential type, being related to the kinetics and to the type of exchanging process: ionic, absorption or partition. Table I reviews some characteristics of the materials proposed as exchangers for uranium in sea-water, and of low-cost materials known for their exchanging power that could be studied or improved. For some materials we have not been able to find reliable data.

Market prices of the exchangers have not been reported in literature but, to give a rough idea, Table I reports some prices of the raw materials used to synthesize the exchangers. These prices must be increased for chemical and physical treatments. A very important factor in the calculation of plant performance is the thickness of the exchanging bed. In fact no exchanger studied has a molecular network wide enough to allow the penetration of uranyl tricarbonate ions at a pH similar to that of sea-water and therefore none is fit for use in bulk. Therefore we must devise some arrangement with the greatest specific surface possible. The exchange rate is another important factor conditioning the number of cycles and the ratio of exchange capacity really used. This factor varies according to the particle size of the exchanger and its type. Table I also gives some data on the time needed to reach 90% saturation. Practically the only materials that have been sufficiently studied are titanium hydroxide and galena.

Another problem to be considered is the ratio between the economically bearable price of the exchanger and the price of the uranium on the market. The ratio between these two monetary quantities is rather complicated but we shall try to relate them to technical parameters. As a first approximation this ratio is linearly related to the percentage of the market price of uranium that can be allotted to the exchanger, to the ion exchange capacity, to the per cent of the ion exchange capacity really used during the cycle, to the number of working cycles of the same amount of exchanger performed in one year and to depreciation for the exchanger.

A simple calculation leads to the conclusion that the amount of money to be paid for the exchanger is strongly dependent on the annual number of cycles, the other parameters being related on the one hand to the time of charge and on the other to some physico-chemical characteristics of the exchanger, which are very difficult to vary.

4. PLANT PROJECT REQUIREMENTS

The plant proposed by the Harwell Group envisages two solutions:

(a) The exchanger is layered on a muslin support and suspended below the water level. The uptake of uranium is fast, but under these conditions the amount of uranium recovered from the total carried by the current is very small.

(b) Exchanger-impregnated asbestos or glass wool frames placed normally to the current as filters.

To maintain the pressure head on one side of the frame it is possible to exploit the tides by building a series of basins, provided the tide level can reach some metres in height.

Such a solution is obviously useless for a plant off the Italian coast as the average height of the tides is some tens of centimetres. There remains only the possibility of utilizing currents or some method of low-price pumping. We shall consider briefly a plant able to extract 1000 t U/a with the following techniques:

(a) Absorbing sheets set parallel to the flow;

(b) Filtering frames perpendicular to the current (granular or fibrous exchanger).

We shall also discuss the plant arrangement for discontinuous or continuous cycles, and resin-in-pulp and static beds.

4.1. Laminar sheet parallel to the current

Let us examine the hypothesis of a plant made of laminar sheets set parallel to the current flow as mentioned in project (a) of the Harwell Group. As an example, we imagine depositing on a rigid surface a layer of galena or other exchanger with a thickness fit to obtain a fast and complete exchange that is 3×10^{-3} cm (30 μ m).

Let us consider an exchanging element with a front section of 1 m² made by 100 sheets of 1 m² surface each at a distance of 9 mm; the amount of the layered galena is about 45 kg, which is able to fix 136 g/kg, the exchange capacity being 3 g/kg. This amount of uranium is brought by a 0.5 m/s sea-water flow across 1 m² in 23 hours. We can therefore perform 381 cycles a year. To produce 1000 t U/a will require about 2×10^4 exchanging elements and 7.5×10^6 total cycles each year.

Even if the exchanging surface is increased, the number of cycles required cannot be decreased by more than one order of magnitude. Such a type of plant will pose many and difficult problems, both economic and operational, depending on whether a single or a continuous operating system is designed.

4.2. Exchanging elements with granular exchanger

To evaluate the characteristics of an exchanging element made from a granular stratum and set perpendicular to the current, we must first examine some considerations.

For an industrial plant the time during which the exchanger must stay in the water is of prime importance. It is quite clear that long contact times increase the amount of capital locked up and decrease the number of cycles in a given time, thereby lowering the maximum economically acceptable cost of the exchanger. As said before, only materials with costs in the range of a few tens of cents per kilogram can be used.

The time of charge depends on many factors such as the flow of current through the exchanger and the exchange ratio, which we will discuss in some detail.

4.2.1. Flow through an exchanging granular frame

To evaluate a current flux through a granular bed the well-known Darcy's relation is applied

$$q = \frac{KS}{\mu} \frac{dP}{dh} \quad (1)$$

where q is the flow, K is the permeability, S is the section crossed by the flux, μ is viscosity, and dP/dh is the pressure gradient. Other parameters to be considered are the porosity B , that is to say the ratio between the volume of the empty spaces and the total volume in per cent (with values ranging between 0.99 and 0.01), and the specific surface S_0 given in m²/g or cm²/g.

Porosity and specific surface are as a rule measured experimentally. The specific surface can also be calculated as in the case of homogeneous diameter spheres and assuming the maximum packing factor. In other cases, such as those of an exchanger fixed on sheets or fibres, the specific surface calculation can be reliable. There are in fact semi-empirical relations, as that of Kozeny, between permeability K , porosity B and specific surface S_0 :

$$K = \frac{gB^3}{5 S_0^2 (1-B)^2} \quad (2)$$

With g the gravity constant = 980 cm/s⁻² and S_0 in cm²/g, the values of K from Eq. (2) will be 10⁻⁵ Darcys.

To evaluate the behaviour of a granular exchanging stratum set normally to the marine current we must consider, as a first approximation, the behaviour of an incompressible fluid in a constant flow. From Eq.(1) and separating the variables and integrating we have

$$V = \frac{K}{\mu} \frac{P_1 - P_2}{h} \quad (3)$$

where V is the speed inside the porous strata, and P_1 and P_2 are the hydrodynamic pressures on the front and rear of the element of thickness h . Now we can evaluate the term $P_1 - P_2$, setting P_1 as the hydrodynamic pressure related to the values of the speed of the marine current and $P_2 = 0$.

In the case of frames not too small and with low viscosities, the hydrodynamic pressure can be evaluated from

$$P = \frac{D}{A} = C_d A \frac{\rho}{2} V_0^2 \quad (4a)$$

$$V_0 = \sqrt{\frac{2P}{C_d \rho A}} \quad (4b)$$

where D is the resistance, that is to say the force acting on the surface. A is the surface and C_d the resistance coefficient; ρ the density and V_0 the speed of the element in the fluid. The coefficient C_d is 0.25 in the case of square sections. Inserting Eq.(4a) into Eq.(3) we obtain the speed of the current inside the granular exchanging sheet.

It is evident that in practice the speed V inside a granular or fibrous frame cannot be higher than V_0 . Figure 2 gives the values of V obtained from Eq.(3) as a function of P_1 and for strata with $h = 1$ cm and $S_0 = 80$ cm²/g, corresponding to a galena bed made with cubic elements of 0.1 mm side. The abscissa gives the values of V_0 obtained from Eq.(4b).

It is evident that there are values of K giving $V > V_0$, which means that the potential speed of the flux that can pass through the exchanger is greater than that of the marine current. In such a case the real speed through the exchanging strata must be evaluated with other relations. From Fig. 2 it is also possible to evaluate how much the thickness of the strata must be increased or decreased to optimize the flux through the exchanger. Knowing K we can find the corresponding value of V related to V_0 , the current speed. If V is greater or lower, it is necessary to change the value of K . This can be achieved by increasing B (Eq. (2)), decreasing S_0 (specific surface) or varying the thickness of the exchanging strata. S_0 cannot be easily changed in a given material so we must vary h , the thickness of the exchanging strata.

4.2.2. Exchange rate

To complete the survey of the parameters influencing the absorption of uranium from seawater by means of exchangers we shall consider the exchange rate.

So as not to decrease the yield of the process too sharply, the exchange rate must be greater than the transport speed of uranium inside the exchanging system. The exchange rate is a property of the exchanger itself and is dependent on both the chemical composition and the volume properties. Therefore two factors must be taken into account: the exchange on the surface and the diffusion of the exchanged ions inside the exchanger.

These values are insufficiently well known for the various exchangers and it is worth improving our knowledge in this field. In the case of our two hypotheses (a layered exchanger and a granular stratum) the time of contact T_c of the solution with the exchanger varies with the speed of the current V inside the exchanger, that is to say $T_c = h/V$, where h is the thickness of the exchanging

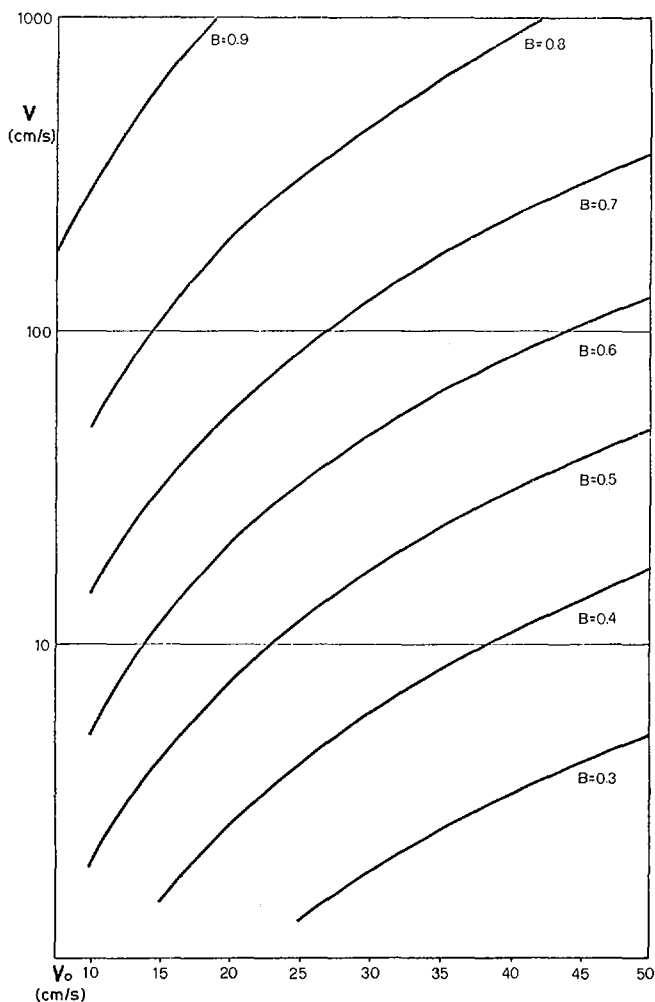


FIG.2. The flow of sea-water inside an exchanging bed as a function of the current speed and the porosity. Thickness of the exchanging strata 10 cm.

strata. In the case of a flux along a layer of an exchanger 1 m thick and set parallel to the current, the time of contact assuming a speed of 40 cm/s will be about 2.5 s. The low values of the contact time are probably a critical factor in an economically productive plant.

The afore-mentioned contact time T_c is the time in which a stream filament of water remains in contact with the exchanger and influences the percentage of extraction from a given flow of sea-water. In the case of an industrial plant the time of contact exchanger/sea-water is equal to the time during which the exchanging element remains submerged in the water. We must therefore consider that if the exchange rate is slow, a fast current is practically useless: for each type of plant there is an optimum value between the current flow and the rate of exchange.

The optimum would be an exchanger with a very high porosity and large water/exchanger contact surface. As an example, a granular exchanger with a high porosity has a small contact surface. The same is true for a laminar element. Probably the best system would be an exchanger fixed on glass wool or asbestos fibre.

4.2.3. *Exchanger on a fibrous framework*

We shall now calculate, as an example, some characteristics of an exchanger layered on fibre. We shall consider a fibre diameter of 0.02 cm covered with a 0.003 cm thick layer of exchanger material, such as titanium oxide, lead sulphide, etc. The diameter of the whole will be 0.026 cm. The surface of one metre of fibre will be $8.16 \times 10^{-4} \text{ m}^2$. To obtain 200 m^2 of exchange surface, as in the laminar elements, $2.5 \times 10^5 \text{ m}$ of fibre will be needed. The volume occupied, if the wire is set in a cubic metre, will give a framework with 98.7% of voids. It is quite possible to increase the amount of wire 10 times, even maintaining a high porosity ($B = 0.87$).

Calculating the specific surface of the exchanger and assuming it to be galena, we shall have $7.5 \times 10^3 \text{ kg/m}^3$. The weight of the galena will therefore be 45 kg and the specific surface $4.44 \text{ m}^2/\text{kg}$. The value of K will be 4.09. If B is taken as 0.87, as previously stated, the charge of galena can be as high as 450 kg/m^3 .

4.3. Industrial plants

4.3.1. *Discontinuous plant producing 1000 t U/a*

The number M of m^2/a of an exchanging element, set normal to the current flow and required to produce 1000 t U/a can be found from the relation

$$M = \frac{10^6}{Q_v C_s h} \quad (5)$$

where Q_v is the weight of the exchanger set in a unitary volume of the exchanging element, C_s is the exchange capacity in kg U/kg exchanger, and h the thickness in metres of the exchange strata. As an example, if Q_v is 3000 kg/m^3 , h is 1 cm and $C_s = 3 \times 10^{-3} \text{ kg/kg}$, M will be $1.1 \times 10^7 \text{ m}^2/\text{a}$ or $1250 \text{ m}^2/\text{h}$ of fully loaded exchanging elements. If we assume as operating under these conditions a system that uses elements of 10 m^2 , 3000 of them would be exhausted daily. If boats are to be used in such operations with a working capacity of 30 cycles a day, that is to say to handle 30 elements, we shall need 100 boats. If the exposed surface of the elements is increased 10 times, some tens of boats will be enough.

Obviously such a type of facility, even if rather flexible, gives rise to a series of problems related to the use of a specially built dock, boats and trained sailors, and many other operating problems such as weather conditions, positioning problems, etc.

4.3.2. *Continuous plant to produce 1000 t U/a*

A continuous plant can be planned for various technologies but it is necessary to underline that practically no studies or operating experience are yet available in this field. We shall therefore now examine some possibilities:

- (a) Exchanging elements of the filtering type (see Section 4.2) or the laminar type set parallel to the current (see Section 4.1) or a fibrous framework (see Section 4.2.3) suspended on a wire rope moving between metallic platforms set on the sea bottom like a cableway. The elution and regeneration facilities can be installed on the platforms. If the elution terminal is placed on an artificial island, several cableways can reach it in a starfish arrangement.

- (b) Another system can be a light pier, along the sides of which the exchanging elements can move. This type of plant will be strongly dependent upon the sea conditions and will need to be placed in a protected bay that does not hamper the coastal currents owing to the need for good water exchange.
- (c) If submarine engineering work can be undertaken, at least in the near future, the pier can be built on the sea bottom, thereby decreasing the effect of the sea waves. A plant like this can pose serious problems in the realization of the elution terminal.

We shall now discuss and try to quantify some aspects of a continuous plant, such as the moving speed and the length, and outline some possible structural arrangements.

The elements, whether of the sheet, fibrous or granular type, are lowered into the sea-water and connected to a rope with a translational movement. After a given time of contact T with the water, which depends on the length L of the implantation and on the translational speed V_t , the elements are taken out of the sea-water and placed in tanks containing the elution and regeneration solutions before being replaced in the sea. The data needed to evaluate the performance of a continuous cycle 1000 t U/a plant are therefore the length L and the translational speed of the elements V_t . Such data can be evaluated from the time of contact T needed to load the exchanging material. These data can be related by means of the simple equation:

$$L = T \cdot V_t \quad (6)$$

where V_t is given in m/s and T in s. T can be calculated bearing in mind that the value cannot be less than the time needed by the flow to load the exchanger. Therefore

$$T = \frac{Q_v \cdot h C_s}{V \alpha 3 \times 10^{-6}} \quad (7)$$

where Q_v is the apparent density of the exchanger in kg/m^3 ; h the thickness of the exchanging strata in m; C_s the exchanging capacity in kg/kg ; V the speed of the current flow through the exchanger; 3×10^{-6} is the amount of U in kg/m^3 sea-water, and α is the yield of the exchanger. The quantity V_t , as T , cannot assume arbitrary values but must be such as to relate the amount of exchanger (in kg) that must be worked each second to accomplish the production of 1000 t/a. We therefore have

$$V_t = 3.2 \times 10^{-2} \frac{1}{Q_v h A C_s} \quad (8)$$

where A is the working surface of the exchanging element given in m^2/m ; this value depends on the shape of the element; and 3.2×10^{-2} is the amount of uranium to be produced each second (in kg) to obtain 1000 t/a. Introducing Eqs (7) and (8) into Eq. (6), we have

$$L = \frac{10^4}{V A \alpha} \quad (9)$$

The length of the plant depends only on the speed V of the current inside the exchanger, on the percentage of uranium actually fixed, α , and on the surface A (in m^2/m) of the exchanging elements.

In Fig. 3 is shown the behaviour of the values T and L as a function of the current speed V for different values of α . It is evident that for a continuous cycle plant, to avoid a huge length, V and α must not be too low.

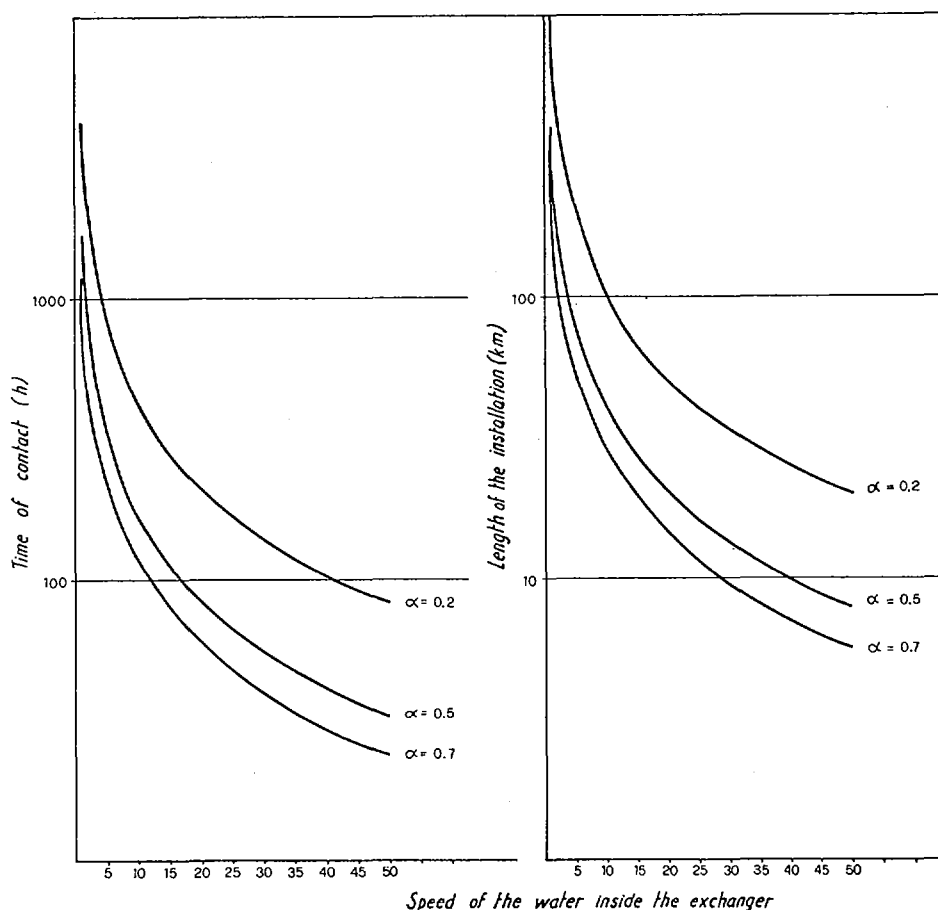


FIG. 3. Contact time T and length L of a continuous plant as a function of the current speed inside the exchanger V for different values of the total yield.

To obtain realistic L values (i.e. less than 10 km) the product of $(V\alpha A)$ must be > 1 . Practically, V can vary between 0.1 and 0.4 m/s and α will be at maximum 0.5. The values of A must be between 20 and 5 m² for each linear metre of implantation. The plant therefore, even if complex, is not impossible to realize.

As an example, let us examine how to increase the permeability of the exchanging elements using well-fitting exchanger-support combinations. As a working hypothesis we imagine a series of exchanging elements (it does not matter whether they are set parallel – lamellar elements – or perpendicular – filtering ones – to the current flow). The elements are, as stated before, suspended from a rope set between fixed platforms and moving at V_t m/s. The effective speed of the flux through the elements will be a vectorial composition of the current speed and translational speed V_t of the elements. As we shall see later, the second term is much lower and can as a first approximation be ignored. Each exchanging element will fix an amount of uranium

proportional to the weight of the exchanger. Therefore the annual production will be given by the product of the number of elements exhausted multiplied by the yield α of the whole process, $P_{\text{tot}} = C_s Q_v h \alpha N$, where P_{tot} is the total annual production, N the number of elements that can be exhausted each year and the other symbols are the same as before. N is very great and therefore a multistage implantation is needed.

Just to fix some numerical values, we assume elements with a parallelepiped shape, so as to obtain the greatest panel surface for each linear metre of implantation. As an example, panels of $3 \times 4 \times 0.5$ m with a centre distance of 3.5 m will have an A-value of $3.43 \text{ m}^2/\text{m}$. We assume that each panel is filled with glass fibre loaded with lead sulphide as in the previous example (Section 4.3), i.e. 450 kg/m^3 of PbS and a porosity of 87%.

Each element could be loaded with 8.1 kg of uranium. To produce 1000 t/a with $\alpha = 0.5$ it is necessary to exhaust 123 460 elements each year. From Eq. (8) under such conditions ($Q_v = 450$, $h = 0.5$, $A = 3.43$ and $C_s = 0.003$) the translational speed of elements will be 50 m/h, and if $V = 0.30$, using Eq. (7) T will be 420 h and the implantation length 19.5 km.

To reduce the length of a single-section plant, five sections could be arranged in a star formation set in a suitable location taking into consideration the wind and sea current directions and with a central platform for elution and regeneration. To change the plant dimensions we can vary V , which depends on the permeability of the exchanging element. A is limited by the encumbrance while Q_v influences the permeability. Thus many parameters must be considered in optimizing the plant.

4.4. Submerged resin-in-pulp plant

A submerged plant could be formed, in principle, by boxes, like containers, shaped for use on the sea bottom. The shape of the boxes must be such that the external flow can produce current eddies that stir the pellets of the exchanger inside the structure. A grid at the outlet would retain the pellets. The dimensions of the pellets and their specific weight must be such as to favour the maximum mixing and contact with the water. To avoid too small a pellet size the exchanger can be fixed on the surface of inert spheres. Porous exchanger pellets could also be studied. If the dimensions of the pellets are too great, the following phase, that of pumping the loaded exchanger to the surface for elution and regeneration, could pose some difficulties.

To evaluate the dimensions of such a plant let us again postulate a 1000 t/a plant. The section through which 10^3 t of uranium can flow utilizing a current of 0.5 m/s, as above, is $2 \times 10^4 \text{ m}^3$, as can be seen from Table I. If the elements are 50 m^2 in section, assuming 100% extraction, 400 elements will be needed. In practice this number must be multiplied at least 5 times to take into account the decrease in the speed of the current inside the box. This decrease is due to Darcy's law and, moreover, helps to optimize the time of water/exchanger contact and consequently the extraction yield.

4.5. Static bed extraction

We assume a static bed with a very low-cost exchanger. The problem is practically that of synthesizing an artificial mineral and treating it in a normal extraction process. The material must be spread on the bottom of lagoon-like shallow coastal basins with good water exchange with the open sea. First of all, we must evaluate the time needed to load the exchanging material. This time depends on the diffusion of uranium carbonate ions inside the exchanging material. The complex diffusion laws are not easily predictable. The second factor is the amount of sea-water necessary to load the exchanging strata.

We assume a lagoon of some tens of square kilometres of surface and a few metres deep, and a very cheap inorganic exchanger suitable for loads of 1 kg/t, e.g. some kind of volcanic ash. According to the average values of natural water exchange in some coastal lagoons some hundred

years would be needed to load the exchanger. This time could be considerably reduced if the rate of water exchange can be increased by artificial means, but in this case the problem is the availability of a low-cost pumping system. As an indication, the cost of the exchanger must be in the range of a few dollars a tonne. This method in our opinion is not, at present, viable.

5. CONCLUSION

In our opinion, the great interest in recovering uranium from sea-water lies mainly in the fact that, in view of the enormous amount of uranium contained in the oceans, which is estimated to be around 4–5 billion tonnes, the cost at which the uranium is recovered would also be the maximum cost of uranium considered in future estimations.

The possibility of recovering uranium from sea-water has been considered on the basis of existing literature, which is mainly English and Japanese. Some kinds of exchangers have been reviewed, the more interesting among them appearing to be titanium hydroxide, which can be charged up to 1.5 kg/t, and galena, which can be charged up to 3.5 kg/t. Both show extremely interesting concentration factors with respect to the uranium content of sea-water, which – as known – averages about 3 ppb. Research into exchangers is being carried out with particular care by Japanese workers.

As regards the method of achieving the sea-water/exchanger contact, until now two different methods have been studied: the use of water heads due to tides, and the recovery of uranium from industrial plants that use sea-water for cooling. In the latter case, according to the Japanese researchers, the high temperature of water would have a positive effect on the exchanging ratio of the exchangers.

As regards our country, the problem appears to be rather complex. First of all, sufficient information is not available to show that the sea around Italy has the same uranium content as the mean calculated for the ocean, although four analyses seem to confirm that. Secondly, and most importantly, no recorded tide around Italy is of such amplitude as to be usable to fill basins, even at low elevation. The cooling waters of industrial plants could be taken into account if it were possible to set up low-price modular plants that could be easily connected to the existing discharge channels.

We presume that in our country we could recover about a hundred tonnes of uranium per year with the latter method. This amount would not cover our supply needs, but would be of interest from an economic point of view (at present, one tonne of uranium, in the form of yellowcake, has a value of about \$65 000). As this problem is easier to solve from the technological point of view, it seems obvious that first research will be aimed at its solution.

With regard to greater production in the range of 1000 t/a, we have rejected the solution of low-cost pumping (which would lead to pumping about 10 000 m³ of water per second) and would wait for technological improvements in this field, such as, for instance, heat pumps. We have then examined in some detail the possibility of utilizing currents to achieve the contact between sea-water and the exchanger. We have discarded the possibility of utilizing static exchanging elements on account of the huge number of both elements and elution-regeneration cycles needed.

More promising appears to be the possibility of utilizing continuous systems with elements hanging on a cable, slowly moving under the sea surface and anchored to a couple of platforms resting on the bottom of the sea. The whole plant length, i.e. the span between the two platforms, is in the range of 15–25 km, and there would be the possibility of dividing the span into segments of lesser length. The cost of a plant of this kind should be about 195–225 million dollars, while the overheads would range between 93 and 180 million dollars, depreciation allowance included.

In conclusion, the prime cost should be in the range of 65–120 \$/lb U₃O₈, the same order of magnitude as that calculated in British and Japanese research. As a basis we have assumed a cost of money of 12%, and a percentage of 15% to 25% depreciation on the total cost.

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DISCUSSION

(Papers IAEA-AG/33 - 12 and IAEA-AG/33 - 4)

J.A. PATTERSON: I wonder whether you could tell us something about the research and development programmes that you intend to carry out for recovery of uranium from sea-water.

G.I.W. LLEWELYN: The work in the UK was carried out by the United Kingdom Atomic Energy Authority and this programme ceased a few years ago. At present we are doing no practical work on the extraction of uranium from sea-water. What I have described was a paper study carried out by a Working Party that was set up about a year ago to investigate the overall strategic implications of this project. The resources that would be used in further practical work in this area are currently being used on aspects of the nuclear fuel cycle which are of rather higher priority than extraction of uranium from sea-water. The UKAEA would be pleased to discuss the results of this work on uranium extraction, which has not been published, with staff from other laboratories who are engaged in similar studies.

F.R. HARTLEY: I would like to raise the question of environmental effects. From the point of view presented by the environmental group in our country, it seems to me that it can certainly not be answered by the fairly cursory comment that removing uranium from sea-water would have no environmental effect. I imagine that several thousand tonnes of suspended solids would be involved and these would be biologically quite important. I was wondering to what extent this was taken into account.

F. PANTANETTI: In the first phase of our work we have not taken into account the environmental aspect; this will be studied later. We have only seen that the extraction of uranium from sea-water does not change the chemical composition of sea-water because the amount of uranium to be extracted is so small in respect to the total quantity in the sea that it is of no importance at all.

G.I.W. LLEWELYN: In the UK, preliminary discussions have been held with marine biologists and their feeling is that, although there would be an environmental management problem, there is nothing involved in this process which would be insurmountable.

CONCLUSIONS AND RECOMMENDATIONS

CONCLUSIONS

1. There is a true spirit of co-operation between the countries present to share technical information and data on uranium ore processing.
2. Every effort must be made to improve existing processes and to encourage the finding and development of new orebodies so that the world will be able to meet the obvious increase in demand expected for the years 1980–1985 and onward.
3. Because of the 8 to 9 years' lead time required to bring a large deposit on stream, more research, development and encouragement is required on the following items:
 - (a) Small, unsophisticated processes for small or isolated deposits.
 - (b) In-situ type operation which can be brought on stream quickly.
 - (c) Preconcentration methods to allow existing plants to expand.
 - (d) Heap leaching with or without bacterial assistance.
 - (e) Realistic environmental standards for radium-226 in effluents and radon emanations are required so that operators may be aided in planning processes that will not harm the environment.

This work is essential to allow the member countries to meet their growing demands.

4. This conference emphasizes the significant results that uranium producers have accomplished in reducing the environmental impact of uranium mining and milling operations.

Nevertheless, the environmental impacts associated with the processing of uranium ores using the best currently available techniques do not appear to be unjustifiable in relation to the potential benefits from the uranium produced. However, in view of the public concern about the environment, these impacts should be fully considered at the planning stage so that they may be reduced to the lowest reasonably achievable levels.

5. Further development is required on methods for the recovery of uranium from currently uneconomic low-grade ores or as by-products from other processes.
6. Further research is required on the extraction of uranium from sea-water although this will be in the longer term.

RECOMMENDATIONS

The Advisory Group recommends that:

1. The Agency encourage and support continuing research and development on uranium ore processing with particular attention to low-grade, complex, and refractory resources, and that it facilitate the exchange of uranium ore processing information among its Member States.
2. As a means of encouraging research and development and facilitating information exchange, a broadly based symposium or conference should be held at least every 5 years, and advisory or study group meetings every 2 or 3 years.
3. Standing committees or working groups should be appointed to monitor (and assess) technological developments in uranium extraction and provide prompt notification to the Agency and its members of important developments as these occur. Subgroups might be established for certain resource type or process areas such as: shales, lignites and other carbonaceous ores; phosphates; refractory and complex ores; in-situ leaching.

4. Continuing attention be given by member nations to the development of simplified flow-sheets for small or isolated deposits using, for example, such techniques as improved resin-in-pulp, solvent-in-pulp, autogenous generation of acid with bacterial assistance, and in-situ methods.
5. Information and data should be collected and disseminated on the occurrence and mitigation of radiation hazards and chemical pollution in uranium ore processing and tailings disposal. Attention should be given to improved techniques of tailings dam construction, tailings containment and stabilization, and treatment of tailings solutions.
6. A tabulation of the known uranium ore processing plants and the processes utilized has been compiled and it is recommended that the Agency should revise, up-date and publish this list at regular intervals. The complete tabulation will be found attached to the inside back cover of this book.

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CLASSIFICATION OF URANIUM EXTRACTION PLANTS

TABLE A—I.	CLASSIFICATION OF COMMERCIAL URANIUM EXTRACTION FLOWSHEETS
TABLE A—II.	NEW URANIUM PLANTS AND DEPOSITS UNDER INVESTIGATION
TABLE A—III.	SUMMARY OF EXISTING, IMPROVED AND NEW TECHNIQUES FOR URANIUM ORE PROCESSING STEPS

TABLE A—I. CLASSIFICATION OF COMMERCIAL URANIUM EXTRACTION FLOWSHEETS

BASIC PROCESS TYPE	LOCATION OF URANIUM PLANT			COMPANY	START-UP DATE	ORE CHARACTERISTICS		MINING	MILLING		LEACH				SOLID/LIQUID SEPARATION	CLARIFICATION	CONCENTRATION AND PURIFICATION			
	Country	State/Province	Mill			Type	Main uranium mineral		Type	Degree	Type	Temperature (°C)	Oxidant	Equipment			Equipment	Resin/Solvent	Elution/Stripping	Precipitation
Acid leach Countercurrent decantation Ion exchange	USA	Washington	Ford	Dawn Mining Company	1957	Vein	Autunite Uraninite	Open pit	Ball	Coarse -28 mesh	Two-stage, acid	50	Sodium chlorate	Mechanical agitators	4-stage CCD thickeners	Granular activated carbon clarifier	I.X. columns (fixed bed)	Strong base	Nitrate	Two-stage — lime — ammonia
		Colorado	Uran	Union Carbide Corporation	1950	Sandstone	Tyuyamonite	Underground	Dry autogenous aerofall	Coarse -28 mesh	Two-stage, acid	80	Sodium chlorate	Mechanical agitators	8-stage CCD thickeners	Sand filters	I.X. columns (fixed bed)	Weak base (XE 299)	Chloride	Ammonia
		Wyoming	Shirley Basin	Utah International	1971	Sandstone	Uraninite	Open pit	Semi-autogenous cascade	Coarse -28 mesh	Single-stage, acid	35	Sodium chlorate	Pachucas	Cyclone circuit for sand-slime separation and sand washing, 6-stage CCD	Anthracite coal clarifiers	I.X. columns (fixed bed)	Weak base (XE 270)	Chloride	First and second eluates ppt separately ammonia
	CANADA	Ontario (Elliot Lake)	Quirke	Rio Algom Mines	1956	Quartz-pebble conglomerate	Uraninite Brannerite	Underground	Rod + pebble	Medium -65 mesh	Single-stage, acid	70	None	Pachucas	5-stage CCD thickeners	Leaf clarifiers	I.X. columns (fixed bed)	Strong base	Nitrate	Two-stage — lime — ammonia
Acid leach Filtration Ion exchange	CANADA	Ontario (Elliot Lake)	Denison	Denison Mines	1957	Quartz-pebble conglomerate	Uraninite Brannerite	Underground	Rod + pebble	Medium -65 mesh	Single-stage, acid	75	None	Pachucas	Wash thickener and 2-stage rotary filters	Leaf clarifiers	I.X. columns (resin transfer type)	Strong base	Nitrate	Two-stage — lime — ammonia
			Stanrock	Denison Mines	1958 (closed down in 1964)	Quartz-pebble conglomerate	Uraninite Brannerite	Underground	Rod + ball	Medium -65 mesh	Single-stage, acid	65	Sodium chlorate	Mechanical agitators	Wash thickener and 2-stage rotary filters	Leaf clarifiers	I.X. columns (fixed bed)	Strong base	Nitrate	Two-stage — lime — magnesia
			Nordic	Rio Algom Mines	1957 (in moth-balls since 1970)	Quartz-pebble conglomerate	Uraninite Brannerite	Underground	Rod + ball	Medium -65 mesh	Single-stage, acid	65	None	Pachucas	Wash thickener and 2-stage rotary filters	Leaf clarifiers	I.X. columns (fixed bed)	Strong base	Nitrate	Two-stage — lime — ammonia
			Panel	Rio Algom Mines	1958 (closed down in 1961)	Quartz-pebble conglomerate	Uraninite Brannerite	Underground	Rod + ball	Medium -65 mesh	Single-stage, acid	65	Sodium chlorate	Pachucas	Wash thickener and 2-stage rotary filters	Leaf clarifiers	I.X. columns (resin transfer type)	Strong base	Nitrate	Two-stage — lime — ammonia
			Preston	Preston Mines	1958 (closed down in 1961)	Quartz-pebble conglomerate	Uraninite Brannerite	Underground	Rod + ball	Medium -65 mesh	Single-stage, acid	65	Sodium chlorate	Pachucas	Wash thickener and 2-stage rotary filters	Leaf clarifiers	I.X. columns (fixed bed)	Strong base	Nitrate	Two-stage — lime — ammonia
	SOUTH AFRICA	Transvaal	Hartebeestfontein	Anglo-Transvaal Consolidated Investment	1956	Quartz-pebble conglomerate	Uraninite	Underground	Rod + pebble	Fine -200 mesh	Single-stage, acid	60	Manganese dioxide + ferric	Pachucas	2-stage rotary filters	Sand clarifiers	I.X. columns (fixed bed)	Strong base	Nitrate	Two-stage — lime — ammonia
		Orange Free State	Virginia	Rand Mines	1955	Quartz-pebble conglomerate	Uraninite	Underground	Ball + pebble + rod	Fine -200 mesh	Single-stage, acid	75	Ferric	Pachucas	2-stage rotary filters	Sand clarifiers	I.X. columns (fixed bed)	Strong base	Nitrate	Two-stage — lime — ammonia
	FRANCE	Vendée	L'Ecarpière	SIMO	1956	Vein in granite	Pitchblende	Underground and open pit	Rod + ball	Medium -35 mesh	Single-stage, acid	50–60	NaClO ₃	Mechanical agitators	Wash thickeners for fines Rake classifiers for sands	Sand filters	I.X. fixed bed eluex (start 1976)	Strong base Tertiary amine	H ₂ SO ₄ (NH ₄) ₂ SO ₄	MgO then NH ₄ OH
Acid leach Countercurrent decantation Solvent extraction	USA	Texas	Falls City	Continental Oil and Pioneer Nuclear	1972	Sandstones (Tordilla) Lignites	Uraninite	Open pit	Rod (preroast possible for lignite ores)	Coarse -28 mesh	Single-stage, acid	60	Sodium chlorate	Mechanical agitators	5-stage CCD thickeners	Downflow sand filters	Mixer-settlers	Tertiary amine	Chloride	Ammonia
		Wyoming	Shirley Basin	Petrotomics Company	1962 (shut down 1974)	Sandstones	Uraninite	Open pit	Rod	Coarse -28 mesh	Single-stage, acid	Ambient	Sodium chlorate	Mechanical agitators	6-stage CCD thickeners	Pressure precoat filters	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
			Powder River Basin	Exxon Company	1972	Sandstones	Uraninite	Open pit and underground	Rod	Coarse -28 mesh	Single-stage, acid	Ambient	Sodium chlorate	Mechanical agitators	5-stage CCD thickeners	Pressure sand clarifiers	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
		New Mexico	Ambrosia Lake	Kerr-McGee Nuclear Corporation	1958	Sandstones	Coffinite	Underground	Rod	Coarse -28 mesh	Single-stage, acid	60	Sodium chlorate	Mechanical agitators	Cyclones for sand/slime sep. Sands: 5-stage rake classifiers Slimes: 6-stage CCD thickeners	Pressure precoat filters	Mixer-settlers	Tertiary amine	Chloride	Ammonia
	FRANCE	Limousin (La Crouzille)	Bessines	SIMO	1958	Vein (granite)	Pitchblende	Underground and open pit	Rod + ball	Medium -35 mesh	Two circuits	40–50	NaClO ₃	Mechanical agitators	CCD wash thickeners	Sand filters	I.X. fixed bed solvent extraction in mixer-settlers. Eluex (start 1976)	Strong base Tertiary amine	H ₂ SO ₄ (NH ₄) ₂ SO ₄	NH ₄ OH
											Filter acid for fines									
											Strong acid for sand	70	NaClO ₃	Revolving drums	Rake classifiers					
	AUSTRALIA	Northern Territory	Rum Jungle	Australian Government	1954 (closed 1971)	Vein	Uraninite	Open pit	Rod	Medium -65 mesh	Batch acid leaching	Ambient	Manganese dioxide	Pachucas	4-stage CCD thickeners	Sand filters (old I.X. columns)	Mixer-settlers	Tertiary amine	Chloride	Magnesia
		Queensland	Mary Kathleen	Mary Kathleen Uranium (Rio Tinto)	1958 (closed 1974 new plant 1976)	Breccia conglomerate	Uraninite	Open pit	Rod + ball	Fine -200 mesh	Single-stage, acid	40	Manganese dioxide	Mechanical agitators	Cyclones for sand/slime sep. Sands: cyclones Slimes: CCD thickeners	Rotary precoat filters	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
	SPAIN	Jaén	Andújar	Junta de Energía Nuclear (JEN)	1959	Vein (granite)	Pitchblende	Open pit and underground	Preroast of limonites, rod + ball	Medium -48 mesh	Single-stage, acid	40	Manganese dioxide (eventually)	Mechanical agitators	Cyclones for sand, 4-stage CCD for slimes	Leaf pressure filters	Mixer-settlers	Tertiary amine	Chloride	Ammonia
Acid leach Filtration Solvent extraction	SOUTH AFRICA	Transvaal	Blyvooruitzicht	Rand Mines	1953 (closed 1964 new plant 1970)	Quartz-pebble conglomerate	Uraninite	Underground	Rod + ball	Fine -200 mesh	Single-stage, acid	Ambient	Manganese dioxide	Pachucas	2-stage rotary filters	Old I.X. columns	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
			Buffelsfontein	General Mining and Finance Corporation	1957 (I.X.) 1968 (S.X.)	Quartz-pebble conglomerate	Uraninite	Underground	Rod + pebble	Fine -200 mesh	Single-stage, acid	55	Manganese dioxide	Pachucas	2-stage rotary filters	Sand clarifiers + pressure sand filters	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
			West Rand Consolidated	General Mining and Finance Corporation	1952 (I.X.) 1970 (S.X.)	Quartz-pebble conglomerate	Uraninite	Underground	Ball + pebble	Fine -200 mesh	Single-stage, acid	55	Manganese dioxide	Pachucas	2-stage rotary filters	Sand clarifiers	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
			West Driefontein	Gold Fields of South Africa	1956 (closed 1962 new plant 1972)	Quartz-pebble conglomerate	Uraninite	Underground	Ball + pebble	Fine -200 mesh	Single-stage, acid	60	Manganese dioxide	Pachucas	2-stage rotary filters	Pressure precoat filters	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
			Vaal Reefs	Anglo American Corporation	1956 (I.X.) 1970 (S.X.) 1972 merged with Western Reefs	Quartz-pebble conglomerate	Uraninite	Underground	Ball + pebble	Fine -200 mesh	Single-stage, acid	50	Manganese dioxide	Pachucas	2-stage rotary filters	Leaf clarifiers and old I.X. columns	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
			Western Deep Levels	Anglo American Corporation	1970	Quartz-pebble conglomerate	Uraninite	Underground	Pebble	Fine -200 mesh	Single-stage, acid	50	Manganese dioxide	Pachucas	2-stage rotary filters	Leaf clarifiers	Mixer-settlers	Tertiary amine	Sulphate	Ammonia
		Orange Free State	Harmony	Rand Mines	1955 (I.X.) 1970 (S.X.)	Quartz-pebble conglomerate	Uraninite	Underground	Ball + pebble	Fine -200 mesh	Single-stage, acid	Ambient	Manganese dioxide	Pachucas	2-stage rotary filters	Sand filters (old I.X. columns)	Mixer-settlers	Tertiary amine	Sulphate	Ammonia

TABLE A–I (cont.)

BASIC PROCESS TYPE	LOCATION OF URANIUM PLANT			COMPANY	START-UP DATE	ORE CHARACTERISTICS		MINING	MILLING		LEACH				SOLID/LIQUID SEPARATION	CLARIFICATION	CONCENTRATION AND PURIFICATION													
	Country	State/Province	Mill			Type	Main uranium mineral		Type	Degree	Type	Temperature (°C)	Oxidant	Equipment			Equipment	Resin/Solvent	Elution/Stripping	Precipitation										
Acid leach Filtration Lime precipitation Solvent extraction	FRANCE	Forez	Saint-Priest la Prugne	Owner CEA Operator SIMO	1960	Vein (granite)	Pitchblende	Underground and open pit	Rod	Medium -35 mesh	Single-stage, acid	50–60	NaClO ₃	Mechanical agitators	Continuous belt filters	None	Intermediate lime preconcentrate precipitated, press-filtered and redissolved in nitric acid													
																	Mixer-settlers and columns	Tributyl phosphate	Water	None Concentration by evaporation										
Acid leach Countercurrent decantation Ion exchange Solvent extraction	USA	Wyoming	Gas Hills	Utah International	1958	Sandstones	Uraninite	Open pit	Rod	Coarse -28 mesh	Two-stage, acid	35	Sodium chlorate	Mechanical agitators	4-stage CCD thickeners	Activated carbon bed clarifiers	I.X. columns (resin transfer type)	Strong base	10% Sulphuric acid	Ammonia										
																	Mixer-settlers	Tertiary amine	Sulphate											
Acid leach Sand/slime separation Ion exchange (RIP)	USA	New Mexico	Bluewater	Anaconda Company	1956	Sandstones	Uraninite Coffinite	Open pit	Rod	Coarse -28 mesh	Single-stage, acid	Ambient	Manganese dioxide	Mechanical agitators	Multistage cyclones and rake classifiers	None	Basket RIP system	Strong base	Chloride	Two-stage — lime — magnesia										
Acid leach Sand/slime separation Ion exchange (RIP) Solvent extraction	USA	Wyoming	Gas Hills	Federal American Partners	1959	Sandstones	Uraninite	Open pit	Ball	Coarse -28 mesh	Single-stage, acid	45	Sodium chlorate	Mechanical agitators	Multistage cyclones and spiral classifiers	None	Screen-mix RIP system	Strong base	10% Sulphuric acid	Ammonia										
																						Mixer-settlers	Tertiary amine	Sulphate						
					Gas Hills	Union Carbide Corporation	1960	Sandstones	Uraninite	Open pit	Rod	Coarse -28 mesh	Single-stage, acid	50	Sodium chlorate	Mechanical agitators	Multistage cyclones and spiral classifiers	None	Screen-mix RIP system	Strong base	10% Sulphuric acid	Ammonia								
																					Mixer-settlers		Tertiary amine	Sulphate						
			Jeffrey City	Western Nuclear	1957	Sandstones (cemented)	Uraninite	Open pit and underground	Semi-autogenous cascade	Coarse -28 mesh	Single-stage, acid	40	Sodium chlorate	Mechanical agitators	Multistage cyclones and drag classifiers	None	Screen-mix RIP system	Strong base	10% Sulphuric acid	Ammonia										
																	Mixer settlers	Tertiary amine	Sulphate											
Strong acid-cure leach Countercurrent decantation Solvent extraction	NIGER	Agades	Arlit	SOMAIR	1971	Sandstones (clays)	Pitchblende Coffinite	Open pit	Dry autogenous cascade	Coarse -28 mesh	Granulation by spraying water + acid onto dry ore	90	None	Horizontal revolving drum	Rake classifier for sand/slime separation Sands: 5-stage rake classifiers Slimes: 6-stage CCD thickeners	Sand filters	Mixer-settlers	Tertiary amine	Carbonate	Sodium hydroxide										
Acid heap leaching	SPAIN	Salamanca	Elefante	Empresa Nacional del Uranio, S.A. (ENUSA)	1975	Schists	Pitchblende Coffinite	Open pit	Impact crushing	Coarse -50 mm	Acid heap leach	Room temperature	None	Garden nozzles	None	None	Mixer-settlers	Tertiary amine amine	Chloride	Ammonia										
Alkaline (carbonate) leach Filtration + countercurrent decantation Caustic precipitation	USA	Colorado	Canon City	Cotter Corporation	1959	Vein	Pitchblende	Underground	Ball	Fine -200 mesh	Both pressure and atmospheric carbonate	80	Oxygen (from air)	Pachucas + autoclaves	Single-stage rotary filters and 2-stage CCD thickeners	Modified sand filters (fine coal)	4-stage caustic precipitation and dissolution, and reprecipitation with ammonia													
			Utah	La Sal	Rio Algom Corporation	1972	Sandstones	Uraninite	Underground	Ball	Fine -200 mesh	Both pressure and atmospheric carbonate	110	Oxygen (from air)	Autoclaves + pachucas	3-stage rotary filters	None	Caustic precipitation and dissolution, and reprecipitation with ammonia												
Alkaline (carbonate) leach Filtration Caustic precipitation	USA	New Mexico	Grants	United Nuclear Homestake Partners	1958	Sandstones (Limestones)	Coffinite	Underground	Ball (in carbonate solution)	Medium -48 mesh	Pressure, carbonate	95 (70 lb/in ² (g))	Oxygen (from air)	Autoclaves	3-stage rotary filters	Settling tank	8-stage caustic precipitation with SDU recycle, calcining and water quench to remove vanadium from yellowcake													
																									Pachucas					
	CANADA	Saskatchewan	Beaverlodge	Eldorado Nuclear	1953	Vein	Pitchblende	Underground	Ball (in carbonate solution)	Fine -200 mesh	Atmospheric, carbonate (small acid circuit for flotation conc.)	90	Oxygen (from air and O ₂ plant)	Mechanical agitators	2-stage rotary filters	Leaf clarifiers	Caustic precipitation													
Alkaline (carbonate) leach Sand/slime separation Ion exchange (RIP)	USA	Utah	Moab	Atlas Corporation	1956 (acid) 1961 (alkaline) 1975 (alkaline + acid)	Sandstones (Limestones)	Uraninite Tyuyamonite	Underground	Ball (in carbonate solution)	Medium -65 mesh	Pressure, carbonate (acid leach for flotation conc.)	120 (50 lb/in ² (g))	Oxygen (from air)	Autoclaves	4-stage drag classifiers 4-stage cyclones	None	Basket RIP system	Strong base	Chloride	Sulphuric acid to drive off CO ₂ peroxide precipitation										
																													Mixer-settlers	Tertiary amine
Solution mining (dilute alkaline leach) Clarification (carbonate) Ion exchange	USA	Texas (Live Oak County)	George West	Atlantic Richfield, U.S. Steel, Dalco Oil	1975	Sandstones (Tordilla)	Uraninite	Solution mining (injection wells)	None		Dilute carbonate in situ	Ambient	Oxygen	Injection wells	Not applicable	Downflow carbon bed filter columns	I.X. columns (fixed bed)	Strong base	Chloride	Ammonia										
Gravity concentration Acid leach Filtration Solvent extraction	SOUTH AFRICA	Transvaal	Phalaborwa	Palabora Mining Company	1971	Carbonatite (Copper ore)	Urano-thorianite	Open pit	Flotation concentrate from copper plant fed to Reichert cone concentrators. Heavy minerals conc. to shaking tables		Hot nitric acid leach				Tilting-pan filters	Sand clarifiers	Mixer-settlers	Tributyl phosphate	Warm water	Ammonia										

TABLE A–II. NEW URANIUM PLANTS AND DEPOSITS UNDER INVESTIGATION

LOCATION OF URANIUM PLANT			COMPANY	APPROXIMATE AVERAGE GRADE (% U ₃ O ₈)	ORE CHARACTERISTICS		MINING	EXTRACTION PROCESS ADOPTED OR PROPOSED
Country	State/Province	Mill/Prospect			Type	Main uranium mineral		
USA	New Mexico	Gallup (Church Rock)	United Nuclear	0.15–0.20 range	Sandstone: Fine to medium grained, feldspathic	Coffinite	Underground	Acid leach, solvent extraction
		Laguna	Sohio Petroleum and Reserve Oil	0.15–0.20 range	Sandstone: Fine to medium grained, feldspathic	Coffinite	Open pit + underground	Acid leach, solvent extraction
	Texas	Three Rivers	Wyoming Mineral	—	Sandstone	Uraninite	Injection wells	Dilute carbonate leach, proprietary ion exchange extraction system
	Florida	Mulberry	Uranium Recovery Corporation	—	—	—	—	Uranium recovered as a by-product of phosphoric acid manufacture — solvent extraction process
CANADA	Saskatchewan	Rabbit Lake	Gulf Minerals	0.17–0.34 range	Vein: Replacement type	Pitchblende	Open pit	Semi-autogenous milling, acid leach, CCD thickeners, solvent extraction
		Cluff Lake	Amok Limited	0.17–0.34 range	Vein: Replacement type	Pitchblende	Open pit	Under investigation
	Ontario	Faraday (Bancroft)	Madawaska Mines	0.09–0.17 range	Pegmatite	Uraninite Uranothorite	Underground	Plant built in 1957. Closed 1964. To be reopened 1976/77. Rod + pebble milling, acid leach, two-stage filtration, ion exchange with chloride elution, magnesia precipitation
		Agnew Lake	Agnew Lake Mines	0.09	Quartz-pebble conglomerate	Uranothorite	Underground	Going into production. In-situ underground leaching and surface leaching of blasted ore using a bacterially assisted ferric sulphate leach, Himsley upflow ion exchange columns and eluex with ammonia precipitation
SOUTH-WEST AFRICA	Swakopmund	Rossing	Rossing Uranium Limited	Confidential	Granite: Syntectic alaskite host rock, pyroxene-garnet gneiss, amphibole-biotite schist	Uraninite and alteration products Betasite	Open pit	Open-circuit rod mill, acid leach at 40° C, sand/slime split, washing of sands and slime, strong-base resin ion exchange (Porter system), solvent extraction and ammonia precipitation
AUSTRALIA	Northern Territory	Ranger	Geopeko Limited	0.2–0.3 range	Disseminated: Chlorite schist host rock and chlorite gangue	Pitchblende	Open pit	Acid leach proposed
		Jabiluka	Pancontinental Mining Ltd. and Getty Oil/Development Co. Ltd.	0.39	Vein/Disseminated: quartz, chlorite and graphite schists	Pitchblende Uraninite	Open pit + underground	No metallurgical work done yet
		Koongarra	Noranda Australia Limited	0.50	Vein/Disseminated: quartz, chlorite and mica schists	Uraninite/Pitchblende Secondary uranyl phosphates and silicas	Open pit + underground	Conventional milling and acid leach (heap leach to be investigated as a possibility)
		Nabarlek	Queensland Mines Limited	2.37	Vein: chlorite, mica and quartz-mica schists	Pitchblende Sklodowskite	Open pit	Acid leach under investigation
	Western Australia	Yellirrie	Western Mining Corporation Limited	0.15	Calcrete: quartz, dolomite, calcite, clays, feldspar, gypsum and celestite	Carnotite	Open pit	Alkaline leach under investigation
	South Australia	Beverley	Western Nuclear, Oilmin, Transoil, Petromin	0.265	Unconsolidated to semi-consolidated sand	Not identified owing to fineness of uranium minerals	Open pit	Acid leach under investigation
		North Flinders Range Hodgkinson Mt. Gee Armchair Radium Ridge	Oilmin, Transoil	0.25 0.10 0.10 0.06	Breccia rock, hematite and chlorite gangue	Uraninite	Open pit + underground	Back-up resources for Beverley prospect
SWEDEN	Västergötland (Billingen)	Ranstadt	LKAB	0.025–0.0325 range	Shales: Bituminous, 22% organic, 13% pyrite, limestone, illite, feldspar	Organouranium complex	Open pit + underground	Under investigation. Proposed process: primary crushing, heavy-media separation of limestone, hammer mills and impactors, vibrating screens, sulphuric acid curing, storage in open stockpiles, percolation leaching in vats, clarification, solvent extraction, yellowcake precipitation
SOUTH AFRICA	Orange Free State	President Brand	Anglo American Corporation	Confidential	Quartz-pebble conglomerate	Uraninite	Underground	Plant built in 1971 but not started up. To be recommissioned 1976/77. Acid leach, two-stage filtration, clarification, tertiary amine solvent extraction, ammonia precipitation
	Transvaal	Randfontein Estates	Johannesburg Consolidated Investment Company	Confidential	Quartz-pebble conglomerate	Uraninite	Underground	Technical and economic feasibility study in progress
SPAIN	Salamanca	Queros	ENUSA	0.08	Mainly schists	Pitchblende Coffinite	Open pit	Primary crushing, cascade mills, roast fluid bed, single acid leach with mechanical agitation, 6-stage CCD thickeners, sand filters, tertiary amine solvent extraction, ammonium sulphate stripping, ammonia precipitation, and limestone + lime neutralization
	Badajoz	Lobo G	JEN	≤ 0.08	Altered schists	Pitchblende	—	Start-up 1976. Primary crushing, sand and fines separation: acid heap leach for sand and single acid leach for fines; sand/slime separation with cyclones, RIP (contactors and screen), acid elution, solvent extraction heap leaching pregnant liquors and eluate, chloride stripping, ammonia precipitation and lime neutralization
	Guadalajara	Mazarete-1	?	0.06–0.1	Sandstones	Pitchblende	Open pit	Under investigation. Rod and ball mill, single acid leach (CCD + SX versus sand/slimes separation and CCIX) ammonia precipitation and effluent neutralization. Start-up: 1979
	Huelva	ONIX	FESA?	—	—	—	—	Uranium recovered as a by-product of phosphoric acid manufacture, solvent extraction process. Start-up: 1979
FRANCE	Hérault	Lodève	?	0.2–0.3	Shale with limestone and sulphides	?	Underground + open pit	Probably alkaline leaching
NIGER	Agades	Akouta	Niger Government CEA-OURD ENUSA COMINAK	0.3–0.5	Sandstones	Pitchblende Coffinite	?	Start-up in 1978. Dry autogenous milling. Acid oxidant curing. Continuous belt filtering. Amine extraction. Uranium precipitation with MgO
		Imouraren	Niger Government CEA CONOCO	0.2–0.5	Sandstones	Pitchblende Coffinite	?	Under investigation
		Djado	Niger Government CEA Urangesellschaft	?	Sandstones	?	?	At present not investigated
GABON	Franceville	Mounana	COMUF	0.3–0.5	Sandstones	Francevillite	Open pit	Preconcentrates precipitated with MgO produced since 1961. Major expansion of plant, including solvent extraction unit to produce yellowcake, to start up in 1978.

TABLE A—III. SUMMARY OF EXISTING, IMPROVED AND NEW TECHNIQUES FOR URANIUM ORE PROCESSING STEPS

UNIT OPERATION	DESCRIPTION OF TECHNIQUE	INDUSTRIAL APPLICATIONS	REMARKS ON RECENT DEVELOPMENTS AND FUTURE WORK
MILLING	Wet grinding in ball and rod mills	Widely practised in USA, Canada, South Africa, France and others	Much attention is being given in South Africa, Canada, the USA and Australia to automatic control of semi-autogenous and autogenous mills. These techniques are being carefully considered for future uranium ore processing plants
	Wet semi-autogenous and autogenous grinding in pebble mills	Widely practised in South Africa and Canada	
	Dry and wet semi-autogenous and autogenous grinding in cascade mills	Used at Shirley Basin (Utah Int.), Jeffrey City (W. Nuclear), Uravan (Union Carbide) and Arlit (SOMAIR)	
BENEFICIATION (Preconcentration)	Radiometric sorting	Widely used in the USSR. Used at Bessines (SIMO), France, and in Bancroft area, Canada	Studies are continuing in the USSR on improvements to the sensitivity of radiometric sensing heads, etc. Ore Sorters Limited of Canada have successfully tested a multi-channel radiometric sorter on an American uranium ore
	Flotation	Widely used in South Africa for concentration of uranium gold and pyrite from Witwatersrand ores. Used at Moab (Atlas Corp.) for separating a copper concentrate and Beaverlodge (Eldorado Nuclear) for production of low-time concentrate for acid leaching	Although past experience has shown that flotation and gravity concentration does not achieve a discardable tail when applied to a wide variety of uranium ores, testwork is continuing in a number of countries since the potential benefits are great enough to warrant consideration of these techniques for each new uranium source
	Gravity concentration	Used at Phalaborwa (Palabora Mining) to produce heavy minerals concentrate	
	Heavy-media separation	Previously used at Vaal Reefs and Free State Saaiplaas (Anglo American)	Under consideration at Ranstadt (LKAB) for removing limestone from uranium-bearing shales
	Wet high-intensity magnetic separation	No industrial applications on uranium ores	Laboratory and pilot plant investigations are being conducted in South Africa to test the amenability of Witwatersrand ores to concentration by this technique. A major problem to be overcome is the development of large industrial-scale machines
LEACHING	Single-stage dilute acid leach in pachucas or mechanical agitators	Widely practised in the USA, Canada, South Africa, L'Ecarpière, France	Marginal improvements continue to be made in optimizing leach conditions and in automating the addition of reagents. The use of large axial flow propellers has reduced horsepower requirements for mechanical agitation. Marconaflo are developing a modified agitation leaching technique involving the use of specially designed jets for the introduction of lixiviants
	Two-stage acid leach in pachucas or mechanical agitators	Used at Lucky Mc (Utah Int.), Uravan (Union Carbide), Spokane (Dawn Mining) and Bessines (SIMO)	
	Strong acid cure leach	Used at Arlit (SOMAIR)	The process was developed independently from UKAEA for the Arlit mill in Niger, but UKAEA has also developed and patented a strong acid leach process. The success of the Arlit operation has stimulated interest in this technique, particularly for refractory ores
	High-pressure, high-temperature acid leach in autoclaves	No industrial applications in Western countries. Used industrially in the USSR	NUFCOR/Anglo American Corporation are testing acid pressure leaching of Witwatersrand ores on a demonstration scale
	Alkaline (carbonate) leach (atmospheric or pressure) in pachucas, mechanical agitators or autoclaves	Used at Beaverlodge (Eldorado Nuclear), Canon City (Cotter Corp.), Grants (United Nucl. - Homestake) and Moab (Atlas Corp.)	Marginal improvements continue to be made in optimizing leach conditions and equipment
	Dilute alkaline in-situ leaching with injection wells (solution mining)	Used at George West (Atlantic Richfield and Dalco) and Bruni and Three Rivers (Wyoming Minerals)	Exxon and Mobil are investigating solution mining technology
	Underground in-situ acid leaching with or without bacterial assistance	Recently used on an industrial scale in the USSR. Practised at Brugeaud in France	Agnew Lake Mines are undertaking testwork on underground in-situ leaching with a bacterially active acid-sulphate medium
	In-situ, acid, heap (or percolation) leaching	Practised in France at Bessines, L'Ecarpière and Langogne. Proposed for industrial plant at El Nopal in Mexico. Widely used in the USA	The United States Bureau of Mines at Salt Lake City has initiated heap leaching studies on ores from New Mexico, Wyoming and Colorado. The use of pelletizing and alkaline percolation leaching at an Australian calcrete-carnotite ore is investigated by AMDEL
SOLID/LIQUID SEPARATION	CCD in thickeners	Widely practised in the USA and used at Quirke (Rio Algom), Mary Kathleen (Rio Tinto) and Arlit (SOMAIR)	A fivefold reduction in settling area requirements is obtained on some pulps using the Enviro-clear thickener. Marginal improvements continue to be made in the effectiveness of flocculating agents. CCD in conventional thickeners is being considered for the first time in South Africa as an alternative to rotary filters
	Staged filtration	Widely practised in South Africa and Canada. Used at Forez, France, and Grants (United Nucl.-Homestake), USA	Belt filters are being tested at Forez as an alternative to rotary drum filters
	Sand/slime separation and separate washing of sands and slime using cyclones and classifiers (rake, spiral or drag)	Used at Bluewater (Anaconda), Gas Hills (Federal American Partners), Gas Hills (Union Carbide), Jeffrey City (Western Nuclear), Shirley Basin (Utah International) and Moab (Atlas Corp.). Widely used in the USSR. Also used at L'Ecarpière and Bessines, France	Experimentation is continuing at existing plants to arrive at the optimum configuration of cyclones and classifiers — in some instances changes in the physical properties of the ore have necessitated changes in operating conditions
CONCENTRATION AND PURIFICATION	Column ion exchange (fixed-bed and resin-transfer type)	Still used at a number of plants in the USA, Canada, South Africa and France	Recently used at George West (Atlantic Richfield) but not generally favoured for new uranium plants
	Solvent extraction in mixer-settler units	Widely practised in the USA and South Africa and used at Bessines and Forez (SIMO), Arlit (SOMAIR), Rum Jungle and Mary Kathleen (Australia), and recently introduced in Canada at Rabbit Lake (Gulf Minerals)	Solvent extraction has proved to be an economical uranium recovery method and no doubt will be used in a significant percentage of future uranium plants, either directly on pregnant solution (Purlex type) or in conjunction with ion exchange (Eluex or Bufflex type). Improvements continue to be made in the design of mixer-settler units and in the development of equipment for recovery of solvent lost in raffinates
From unclarified solutions (up to 1000 ppm solids)	Continuous countercurrent ion exchange (CIX) (upflow column and multicompart-ment types)	The first industrial application of CIX on unclarified solutions will be the use of the Porter system at Rössing. The NIM CIX system will be used industrially in the near future at a Western Transvaal gold mine	Considerable development work is in progress on the application of CIX techniques to the recovery of uranium from unclarified solutions. USA: Porter, USBM, Higgins systems; South Africa: NIM CIX system; Canada: Himsley system; Japan: Asahi system
From dilute pulps (up to 25% solids)	Basket RIP ion exchange	Used at Bluewater (Anaconda) and Moab (Atlas Corporation)	Superseded by the Screen-Mix RIP ion exchange system for newer plants
	Screen-Mix RIP ion exchange	Used at Gas Hills (Fed. American Partners), Gas Hills (Union Carbide) and Jeffrey City (Western Nuclear)	More successful than Basket RIP system, but the operating costs of RIP plants appear to be slightly higher than those of solvent extraction plants
	Continuous countercurrent ion exchange (CIX) (upflow column and multi-compartment types)	Not used industrially on dilute pulps	The NIM CIX system has been shown capable of handling up to 10% solids on a pilot scale. Testing of Porter and Himsley systems on dilute pulps of up to 8% solids is under consideration
From thick pulps (up to 50% solids)	Pachuca type RIP ion exchange system developed in the USSR	Widely used in the USSR on an industrial scale for thick pulps (up to 45% solids)	The development of an RIP system capable of handling leach pulps directly without any sand/slime separation is a very worthwhile objective, but there is no evidence to suggest that any success is being achieved in this direction in any Western country
	Solvent-in-pulp systems	Not used on an industrial scale anywhere	Various pilot-scale studies have been undertaken in the USSR, Australia and Canada, but solvent loss problems have been severe
PRECIPITATION	Acid leach circuits	Ammonia precipitation very widely practised	Magnesia and hydrogen peroxide are being considered as an alternative to ammonia for new plants in the USA and Canada
	Alkaline (carbonate) leach circuits	Caustic precipitation widely practised	Some plants have additional processing steps (calcining and water quench or dissolution and reprecipitation with ammonia) to improve product purity

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