

IAEA-TECDOC-1296

Technologies for the treatment of effluents from uranium mines, mills and tailings

*Proceedings of a Technical Committee meeting
held in Vienna, 1–4 November 1999*



INTERNATIONAL ATOMIC ENERGY AGENCY

IAEA

June 2002

The originating Section of this publication in the IAEA was:

Nuclear Fuel Cycle and Materials Section
International Atomic Energy Agency
Wagramer Strasse 5
P.O. Box 100
A-1400 Vienna, Austria

TECHNOLOGIES FOR THE TREATMENT OF
EFFLUENTS FROM URANIUM MINES, MILLS AND TAILINGS
IAEA, VIENNA, 2002
IAEA-TECDOC-1296
ISSN 1011-4289

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Printed by the IAEA in Austria
June 2002

FOREWORD

Effluent treatment is an important aspect of uranium mining and milling operations that continues through decommissioning and site rehabilitation. During the life of a mine, effluent treatment is an integral part of the operation with all effluents either being recycled to the mill or processed through a water treatment plant before being released into the environment. During decommissioning and rehabilitation, effluent treatment must continue either through a water treatment plant or by using passive treatment techniques. In response to increasing public concern and regulatory requirements, new effluent treatment technologies and processes are being developed to improve existing operations and rehabilitation projects.

Effluent treatment spans the uranium production cycle from project development to decommissioning. Therefore, new projects such as the high grade deposits that have recently been developed in Canada as well as decommissioning activities throughout the world require state of the art effluent treatment technology to meet stringent new regulatory requirements. Because of the recent closing of several uranium mines or mining districts, particularly in eastern Europe, effluent treatment is becoming an ever increasing concern. Therefore the IAEA convened a technical committee meeting (TCM) so that experts from different countries could discuss information and knowledge on effluent treatment processes and methods.

The TCM on Technologies for the Treatment of Effluents from Uranium Mines, Mills and Tailings was held in Vienna from 1 to 4 November 1999 and was attended by twelve participants from eleven countries. Ten papers were presented covering effluent treatment at operating mines and at facilities that are being decommissioned. The IAEA is grateful to those participants who contributed papers and took part in the discussions.

The IAEA officers responsible for this publication were J.-P. Nicolet and G. Erdi-Krausz of the Division of Nuclear Fuel Cycle and Waste Technology.

EDITORIAL NOTE

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SUMMARY

Effluent treatment is an important aspect of uranium mining and milling operations that span the uranium production cycle from project development to decommissioning and site rehabilitation. Tailings and mine waste dumps produced during mining and milling are the main source of effluents from a uranium operation, and they continue to be a concern even after a mine is closed. Both tailings and waste rock and low-grade ore dumps represent potentially large volumes of low level radioactive material, which, unless properly disposed of, can have serious environmental impact.

General overview

Typical environmental problems associated with mill tailings and waste dumps are radon emanation and leaching of contaminants, including radionuclides, heavy metals and arsenic, into surface water and groundwater. Radon emission results from the free circulation of air across the tailings and dump surfaces. Contamination of water bodies occurs when infiltration of precipitation is unhindered, bottom liners are absent and no drainage collection is installed. Leaching of contaminants can be exacerbated by acid formation from pyrite oxidation. Contaminants other than radionuclides may be the real problem, so a comprehensive and holistic assessment of tailings impoundments and waste dumps and their remediation processes is typically necessary.

A range of geo-technical engineering measures can be employed to prevent or reduce environmental contamination from effluents derived from tailings and waste dumps. Covering tailings and waste dumps with inert soil, or a similar multi-layer barrier is used to control radon emanation. Low permeability capping also reduces infiltration of precipitation and hence leaching and acid formation from waste materials. Seepage water can be collected at the base of the tailings and waste dumps provided adequate bottom liners and drainage layers are installed. The water can then be treated to remove dissolved radionuclides and/or contaminated colloids and solid particles. This treatment has to be continued until discharge standards set by regulatory bodies are met. Therefore, maintenance activities can extend over decades, requiring active institutional control over a long period of time. The time and expense of monitoring can be significantly reduced, however, if waste treatment and final disposal are integral parts of planning, design and operation of uranium production facilities.

Current research

The problems related to effluent management at uranium production and decommissioning sites are as varied as the sites themselves. The papers presented at this meeting describe research and pilot testing of effluent control and treatment at uranium production and decommissioning sites in nine countries: Argentina, Australia, Canada, China, Germany, Kazakhstan, Slovenia and Romania. Though these papers describe research specific to the problems at each site, the technologies they describe potentially have much broader application.

Processing high-grade uranium ore and managing the resulting effluents present unique problems for the operating personnel in the Athabasca Basin District in Saskatchewan, Canada. This is particularly true of the multi-element ores that contain anomalously high concentrations of nickel, cobalt and arsenic. Pilot scale leach tests of ore from the Cigar Lake deposit have evaluated the use of pachucas, which have lower operating costs compared to autoclaves that typically have been used to process high-grade ore. The process flow sheet

was modified during the tests to prevent contamination of mill effluent by As and Ni and to ensure that tailings produced from leaching of Cigar Lake ore can be disposed of in compliance with regulatory standards. The methods and techniques developed during the Cigar Lake tests could also be adapted to processing of other uranium ores.

The cost-effectiveness of different effluent treatment processes is a key consideration for active uranium production operations. Nanofiltration, which has lower energy requirements and potentially higher yields than reverse osmosis, is being tested in Australia. In the Australian experiments, 17 commercial filters were evaluated for their effectiveness in removing uranium, manganese and sulphate ions. Preliminary tests reported from these studies underscore the complexity of nanofiltration, but also confirm its potential for cost-effective treatment of uranium mill effluent. The preliminary results indicate that only separation of uranium is generally assured, and that separation of key contaminants such as Mn, Ra and SO_4 depends on the type of membrane used. Further testing of the most promising membranes is ongoing, with the results designed to correlate their performance with industrial membrane elements.

In situ leach (ISL) extraction of uranium accounted for approximately 15% of worldwide uranium production in 2000. As worldwide ISL operations are expanded, predicting the extent of pollution of an ore-bearing aquifer beyond the limits of an ISL operation has important economic and environmental implications. The effectiveness of self-remediation of ISL operations is being evaluated in Kazakhstan. Studies have been conducted to evaluate the extent and characteristics of pollution outside of ISL polygons, and the mineralogical changes in uranium host rocks that have been contacted by leach solutions have been documented. In particular, the sorption coefficients of the host rocks and their capacity to serve as barriers to the spread of contamination from ISL operations is being evaluated. In addition, emphasis has been placed on studying the correlation between physical and mineralogical characteristics of an aquifer as a means to predict the extent and probable direction of the spread of pollution beyond the limits of ISL production.

Research in China has demonstrated the effectiveness of air aeration hydrated manganese hydroxide adsorption to remove radium from uranium effluents, when manganese oxide is used as the oxidant in acid leaching. In effect, waste ions in the effluent are used to treat the effluent. Neutralisation of the effluent discharge from the leach stream precipitates U, Th, Mn and SO_4^{2-} , but radium is still present in the neutralized effluent. Hydrated manganese hydroxide produced by air aeration can be used to absorb radium in the effluent. Since there is no release of radium from the sludge formed by neutralisation and air aeration, it can be disposed of in the tailings dam.

The importance of effluent control and treatment during decommissioning and rehabilitation is demonstrated at a site near Dresden, Saxony, Germany, where uranium was mined for 20 years by the former Soviet-German Mining Company SDAG Wismut. A conceptual hydrogeological model has been developed to predict movement of effluent drainage from a mine waste rock dump into surface water and groundwater systems. This study is being used to predict the efficacy of a soil covering for the waste rock dump to reduce leaching of uranium into the underlying and surrounding hydrologic systems.

Predicting variations in radon gas release from mining dumps is the emphasis of research at another site in Saxony, Germany. Seasonal variations are reported, as are variations that result from changing meteorological conditions. In addition, structural inhomogeneities within mine dumps and underground mine workings beneath the dumps can

have an effect on radon transport. The model for radon transport at this site has broad application in assessing the environmental impact of radon transport processes within and surrounding mine dumps.

The papers presented in this document describe techniques for treatment of effluents from uranium production operations — both past and present. As shown from these papers, effluent treatment is as varied as are the sources of the effluents. The goal, however, is the same regardless of the type of treatment — minimising environmental impact. While these papers present examples of effluent treatment for specific operations or problems, the solutions have potentially broader application.

List of relevant IAEA publications

IAEA-TECDOC-492: In situ leaching of uranium: Technical, environmental and economic aspects, IAEA, 1989;

STI/DOC/10/314: Guidebook on design, construction and operation of pilot plants for uranium ore processing, TRS No. 314, IAEA, 1990;

STI/DOC/10/359: Uranium extraction technology, TRS No. 359, IAEA, 1993;

STI/DOC/10/362: Decommissioning of facilities for mining and milling of radioactive ores and closeout of residues, TRS No. 362, IAEA, 1994;

IAEA-TECDOC-979: Environmental impact assessment for uranium mine, mill and in situ leach projects, IAEA, 1997;

IAEA-TECDOC-1059: Guidebook on good practice in the management of uranium mining and mill operations and the preparation for their closure, IAEA, 1998;

A Joint OECD-NEA/IAEA report: Environmental activities in uranium mining and milling, published by the OECD-NEA, 1999;

IAEA-TECDOC-1244: Impact of new environmental and safety regulations on uranium exploration, mining, milling and management of its waste, IAEA, 2001.

METHODS OF EVALUATING ORE PROCESSING AND EFFLUENT TREATMENT FOR CIGAR LAKE ORE AT THE RABBIT LAKE MILL

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Abstract. Cigar Lake is the second-largest, high grade uranium orebody in the world. Mineable reserves for Cigar Lake Phase 1 are estimated at 191 million pounds U_3O_8 with a grade of 25.6% U_3O_8 . Subject to regulatory approval, Cameco intends to process the majority of ore from Cigar Lake in the Rabbit Lake mill. Cameco initiated a programme to study the processing of Cigar Lake ore and the treatment of the resulting waste streams. Laboratory and follow-up pilot scale ore leaching tests with Cigar Lake ore samples were performed. Tailings and effluents were generated from the products of the pilot scale leach tests. Mill process tailings were blended with ground waste rock. Using these materials, geotechnical and geochemical properties, including long term tailings pore water characteristics, will be evaluated. In addition, proposed changes to the mill waste treatment operations were developed to deal with increased levels of arsenic and radium in the waste streams. This paper describes the methods and techniques Cameco used in this programme.

1. INTRODUCTION

The Cigar Lake orebody is located in the Athabasca Basin in northern Saskatchewan, about 660 km north of Saskatoon, see Figure 1. The Cigar Lake project, currently operated by Cigar Lake Mining Corporation, is owned by Cameco Corporation (50.025%), Cogema Resources Inc. (37.100%), Idemitsu Uranium Exploration Canada Ltd. (7.875%) and TEPCO Resources Inc. (5.000%). Upon the joint venture decision to proceed with development, Cameco will assume the role of operator at the Cigar Lake minesite.

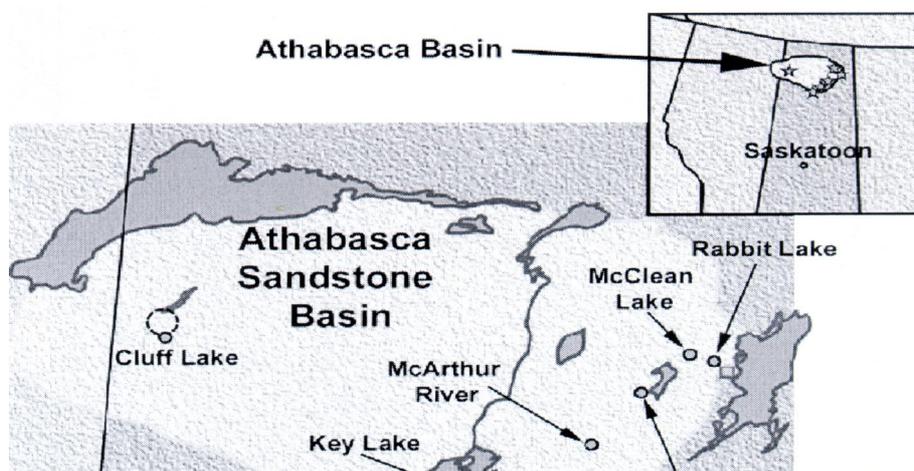


FIG. 1. Project location map.

Subject to regulatory approval, Cameco intends to process 57% of the Cigar Lake ore in the Rabbit Lake mill. (Processing of the rest of the ore is proposed for the McClean Lake mill, subject to regulatory approval). The Rabbit Lake operation is 90 km northeast of Cigar Lake, see Figure 1. The Rabbit Lake project is 100% owned and operated by Cameco. It is planned to mine and grind the ore at the Cigar Lake site and truck ore slurry to Rabbit Lake for processing.

The Cigar Lake ore is high grade, with the Phase 1 mineable reserve averaging 25.6% U₃O₈. The ore also contains significant quantities of arsenic and nickel, with the Phase 1 ore expected to average 3.0% As and 1.5% Ni. The scheme for processing Cigar Lake ore in the Rabbit Lake mill must include methods to prevent contamination of mill effluent discharge by As and Ni in particular, but other metals also. For the Rabbit Lake operation, the present Saskatchewan Environment and Resource Management waste water quality limits include the following criteria:

Substance	Maximum Monthly Arithmetic Mean Concentration	Maximum Grab or Composite Sample Concentration
Arsenic (mg/L)	0.5	1.0
Copper (mg/L)	0.3	0.6
Lead (mg/L)	0.2	0.4
Nickel (mg/L)	0.5	1.0
Zinc (mg/L)	0.5	1.0
Uranium (mg/L)	2.5	5.0
Total Ra-226 (Bq/L)	0.37	1.1
Total Suspended Solids (mg/L)	25.0	50.0
Total Cyanide (mg/L)	1.0	2.0
Th-230 (Bq/L)	1.85	3.7
Pb-210 (Bq/L)	0.92	1.84
Un-ionized Ammonia (mg/L)	0.5	1.0

2. LABORATORY SCALE LEACHING TESTS

Prior to the tests undertaken by Cameco in this programme, the process design for leaching Cigar Lake ore used autoclaves. Cameco has operated uranium leach autoclaves at the Key Lake mill since 1983. Since capital and operating costs for autoclaves are relatively high, it was decided to see if the Cigar Lake ore could be leached in pachuca.

The laboratory scale testing was begun in late 1997 at the Key Lake metallurgical laboratory. Pachuca leaching was tested using specially-designed leach vessels which were developed in the late 1970s by Eldorado Nuclear R&D to enable a bench scale simulation of the operation of the 5.5 metre diameter by 17 metre tall leach pachuca at the Beaverlodge mill. These laboratory leach vessels are essentially low pressure autoclaves with automatic temperature control, continuous oxygen addition at a controlled pressure, and large, slow-moving mixing blades to simulate the relatively mild agitation in pachuca. See Figures 2 and 3.

Because there was no Cigar Lake ore sample available, the first laboratory scoping tests used a hand picked, high grade (28% U₃O₈) ore sample from the A-zone stockpile at Rabbit Lake. Leach conditions kept constant were: 35% solids, 450 kg/t sulphuric acid addition, and 60°C temperature. Three tests were run at 150, 200 and 250 kPa, equivalent to pachuca 11, 15 and 19 metres tall. Results were very promising:



FIG. 2. Pachuca leach test apparatus.



FIG. 3. Test apparatus internals.

Pressure, kPa	% U ₃ O ₈ Extraction at 20 h	% U ₃ O ₈ Extraction at 40 h
150	79.5	99.8
200	93.2	99.7
250	94.7	96.2

Follow-up tests to confirm these results with a Cigar Lake ore sample (28.4% U₃O₈) were done in early 1998. In these tests the leach feed slurry density was maintained at 35% solids, while sulphuric acid addition, temperature and pressure were all varied. Leach pressure was kept below 150 kPa, since higher pressures gave no apparent benefit. The success of pachuca leaching was indeed confirmed by the results:

Acid Addition kg/t	Temperature °C	Pressure kPa	Leach time h	U ₃ O ₈ Extraction %
450	53	150	20	99.83
450	56	150	30	99.83
450	57	150	40	99.84
450	65	100	40	99.80
450	56	75	40	99.82
350	53	150	40	99.52
250	57	150	40	94.92
350	65	75	40	99.58
250	56	75	40	85.17
350	50	75	40	95.24
350	40	75	40	99.58

In each of these laboratory tests, all the sulphuric acid was added in a single dose at the start of the leach. In contrast, in full scale operation sulphuric acid would be added continuously to maintain a target free acidity; in fact, slurry conductivity (directly proportional to free acidity) is sensed and controlled. To simulate acid addition in full scale operation, two more bench scale tests were performed with the free acidity controlled to 100 g/L at the start of the leach, gradually reducing to 20 g/L at the end.

Acid addition kg/t	Temperature °C	Pressure kPa	Leach time h	U ₃ O ₈ Extraction %
304	60	75	36	99.11
310	45	75	36	97.87

The encouraging results of the laboratory scale leaching tests justified continuing on to pilot scale tests.

Small tailings samples were made using the products from the leaching tests. Leach residues from individual tests were mixed together into one sample, then slurried and neutralized with lime. Undiluted tailings were prepared by mixing the neutralized residue slurry with gypsum solids and solids from solvent extraction raffinate neutralized in the Rabbit Lake mill. Combining a portion of the undiluted tailings with ground till or ground waste rock, both from the Rabbit Lake site, provided the final diluted tailings samples. The purpose of this dilution was to lower the tailings grade and improve geotechnical properties — final settled density, for example.

A great deal of experience in splitting and blending tailings slurry samples was obtained generating these laboratory scale diluted tailings samples. This experience was invaluable later in preparing the larger pilot scale tailings sample. It was expected that the solute concentrations in the till-diluted tailings samples would be lower than in the waste rock-diluted tailings samples because of the presumed absorptive properties of the clays in the till. In fact, there was no significant difference found. Both till-diluted and waste rock-diluted tailings had improved geotechnical properties. The Rabbit Lake site has waste rock already mined. However, till for tailings dilution would have to be mined, with a potential for increased environmental impact. Thus the till-dilution option was dropped in favour of waste rock-dilution.

It was also observed that the geotechnical properties (e.g., solids settling rate, final settled density) of the undiluted tailings sample were not optimal for subaerial or subaqueous deposition in the Rabbit Lake in-pit tailings management facility. Geotechnical properties of undiluted tailings were considered potentially effective for deep injection into the previously deposited tailings.

3. PILOT SCALE LEACHING TESTS

In mid-1998 Cameco asked Cigar Lake Mining Corporation to supply a sample of Cigar Lake Phase 1 ore sufficient for pilot scale testing. A drilling programme was initiated and completed by October 1998. 575 kg of drill core were collected and stored in bags. Of the 212 individual bagged samples, 89 were ore and 123 were waste. Each individual bagged sample was dried, crushed and assayed. A pilot plant composite sample was mixed from selected individual samples to approximate as closely as possible the expected average composition of the Cigar Lake Phase 1 ore. The resulting pilot plant composite sample closely matched expected ore composition:

	% U ₃ O ₈	% As	% Ni	% Fe
Expected Cigar Lake Phase 1 Ore	25.6	3.0	1.5	6.0
Pilot Plant Composite	24.6	2.7	1.8	4.6

The pilot plant consisted of a grinding, classification and thickening circuit, a leach feed conditioning tank, and a pilot pachuca. Grinding equipment included a 0.6 m³ feed hopper with a vibratory pan discharge, a variable speed feed conveyor, and a 41 cm diameter by 41 cm long ball mill. Classification was provided by a vibrating screen with 500 micrometer openings. Thickening after grinding was performed in a single 91 cm diameter by 91 cm deep thickener. Leach feed conditioning was done in a 76 cm diameter by 122 cm tall polyethylene tank. Various pumps, piping and ventilation hoods and fans were fitted as required. See Figures 4 and 5.

The pilot pachuca was fabricated at Rabbit Lake. Figure 6 provides details of its design. Based on the laboratory scale leach tests, the slurry depth in the pilot pachuca was set at 11 metres. Pilot pachuca instrumentation included ORP and conductivity probes to permit control of oxidizing potential and conductivity (i.e., free acidity). A thermocouple placed in the top overflow lateral enabled leaching temperature control. An oxygen gas flowmeter and totalizer was installed to determine instantaneous flow rate and total consumption.

In outline, the pilot pachuca leach test procedure was as follows:

- 110 kg of ore were ground and thickened
- Ore slurry was pumped into the leach feed conditioning tank and the initial addition of sulphuric acid was made
- The conditioned ore slurry was pumped into the pilot pachuca
- Oxygen gas flow was set at the operating level; this was taken as time zero for the leach test
- Slurry temperature was increased as rapidly as possible, then held at 50°–60°C
- Leaching was continued for 40 hours
- Pilot pachuca contents were pumped into the conditioning tank, flocculated, and allowed to settle

- The clear pregnant aqueous solution was collected from the top of the tank and stored for later use
- To simulate CCD washing, acidified wash water (pH~2) was added to the tank, the slurry remixed and allowed to settle, and the supernatant solution was drawn off; this was done five times to simulate the Rabbit Lake CCD circuit
- The washed leach residue solids were collected and stored for later use

Two pilot pachuca leach tests were performed. In the first, U_3O_8 extraction was 99.7%. This extraction was reached after just 16 hours of leaching. In the second, U_3O_8 extraction was 99.8%. Once again, this extraction was reached after just 16 hours of leaching.

Once metallurgical tests were completed the test products were used to prepare tailings samples.

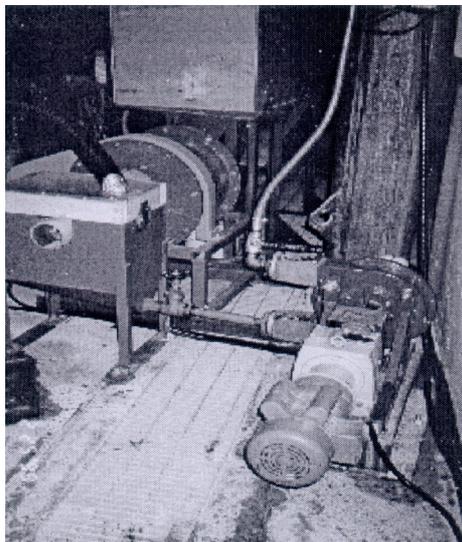


FIG. 4. Pilot plant ball mill, pump box and classification screen feed pump.

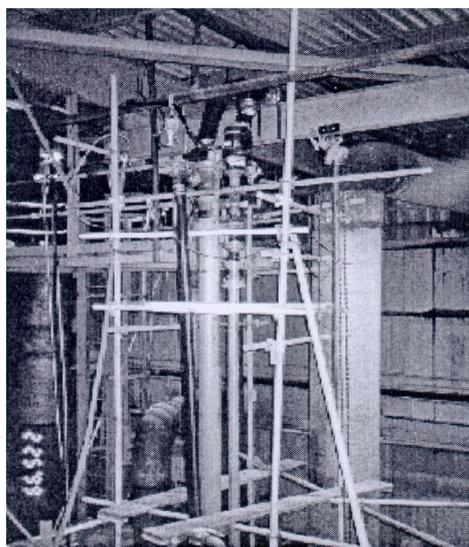


FIG. 5. Top and upper section of the pilot plant pachuca

Increasing pH further, to between 8 and 9, provided an eight-fold decrease in dissolved arsenic relative to pH 3. However, increasing pH even further appeared to cause the dissolution of the basic ferric arsenates; pH 11.3 gave a ten-fold increase in dissolved arsenic. Equally importantly, it was found that at pH 8.5 the solution concentrations of U_3O_8 and Ni were within acceptable levels. Based on these results, an adjustment in the pH profile of the solution neutralization pachucas has been proposed:

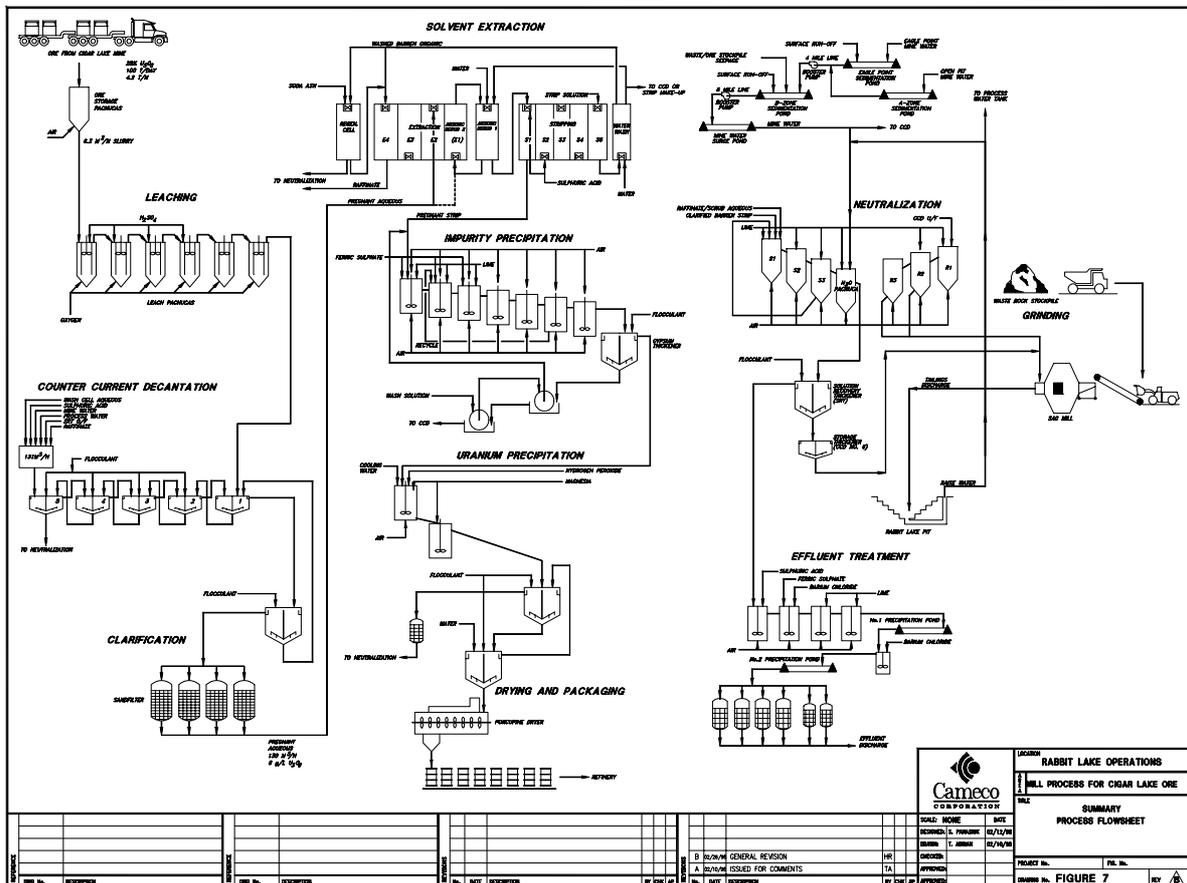


FIG. 7. Rabbit Lake operations, mill process for Cigar Lake ore, summary process flowsheet

Solution Neutralization Pachuca	pH
1	2.0
2	3.0
3	8.5

It was decided that the residue neutralization pachuca profile should mirror the solution neutralization pachuca profile. In the residue neutralization pachucas, lime is added for pH control only in the first two pachucas. The pH in the final pachuca matches that of the second pachuca. The following profile has been proposed:

Residue Neutralization Pachuca	PH
1	3.0
2	8.5
3	8.5

5. ENHANCED RADIUM PRECIPITATION

It was noted in early leach residue neutralization trials that the dissolved radium (Ra-226) concentration was increasing as the slurry pH was increased from less than 2 to 8.5. This situation had to be corrected to prevent unacceptably high dissolved Ra-226 levels in the tailings pore water. The standard method of removing radium from solution is to add barium chloride solution and thus precipitate barium-radium sulphate. However, “conventional wisdom” held that barium chloride addition is only effective in clear solutions, and ineffective in slurries. Nevertheless, it was decided to test barium chloride addition to the acidic leach residue slurry as the first step in the residue neutralization process. Happily, the tests showed that addition of 25 to 50 grams of barium chloride per cubic metre of slurry, followed by neutralization to the above proposed pH profile, was effective in reducing the dissolved Ra-226 concentration to below acceptable levels.

6. TAILINGS PREPARATION

Tailings samples were needed to provide geochemical and geotechnical data to enable evaluation of three proposed tailings deposition concepts: subaerial, subaqueous and deep injection.

The tailings samples prepared were comprised of:

- Neutralized leach residue
- Neutralized raffinate solids (i.e., solution recovery thickener underflow; see Fig. 7)
- Gypsum thickener underflow solids (see Fig. 7)
- Ground waste rock

The first three constituents were mixed in the ratios dictated by pilot test results combined with historical operating data from the Rabbit Lake mill. Undiluted tailings made from these constituents were mixed with ground Rabbit Lake waste rock to give diluted tailings. The waste rock addition rate diluted the tailings to an equivalent 4% U_3O_8 ore grade.

Neutralized leach residue was prepared by mixing together the leach residue slurries from the two pilot pachuca leach tests, adding barium chloride, and neutralizing with milk of lime to the above proposed pH profile.

To prepare neutralized raffinate solids, the combined pregnant aqueous solution from the two pilot pachuca leach tests was subjected to solvent extraction to produce the required raffinate. The raffinate was then neutralized with milk of lime to the above proposed pH profile and the resultant slurry of precipitated solids was thickened.

During a period when the Rabbit Lake mill was processing ore with high As: U_3O_8 and Ni: U_3O_8 ratios (indeed, higher ratios than those expected when processing Cigar Lake ore), gypsum thickener U/F solids were collected from the mill circuit. These were subjected to a wash simulating the mill CCD circuit wash.

Waste rock slurry for tailings dilution was prepared in the pilot plant grinding, classification and thickening circuit.

The pilot plant waste rock-diluted tailings sample was mixed according to the following “recipe”:

Neutralized Leach Residue, weight %	Neutralized Raffinate Solids, weight %	Gypsum Thickener U/F Solids, weight %	Ground Waste Rock, weight %
7.5	20.5	12.0	60.0

The pilot plant undiluted tailings sample was mixed according to this “recipe”:

Neutralized Leach Residue, weight %	Neutralized Raffinate Solids, weight %	Gypsum Thickener U/F Solids, weight %	Ground Waste Rock, weight %
18.8	51.2	30.0	0.0

Completed tailings slurry samples, diluted and undiluted, were thickened, packaged, and shipped to consultants for geochemical (e.g., solids assays, initial and aged porewater assays) and geotechnical (e.g., grain size distribution, specific gravity of particles, settled density, hydraulic conductivity) testing.

7. RECOMMENDATIONS

In principle, the methods and techniques described for evaluating ore processing and effluent treatment could be adapted for other uranium ores. The exact methods and techniques would have to be tailored for the specific unique properties of the ore in question. For example, arsenic and nickel might not be of concern, but other metals such as vanadium, selenium or molybdenum could well pose ore processing and effluent treatment challenges.

ACKNOWLEDGEMENTS

The essential contributions of the following Cameco personnel in planning, executing and analyzing these tests are hereby gratefully acknowledged: C. Rodgers, A. Sarion, N. Chauvet, T Davis, metallurgists at Key Lake; B. Bharadwaj, J. Jarvi, S. Mason, X. Li, metallurgists at Rabbit Lake; N. Holl, P. Landine, hydrogeologists, Engineering & Projects; K. Dyck, S. Panasiuk, metallurgists, Engineering & Projects. Title of figures.