## IAEA-TECDOC-511

# DECONTAMINATION AND DECOMMISSIONING OF NUCLEAR FACILITIES

FINAL REPORT OF THREE RESEARCH CO-ORDINATION MEETINGS ORGANIZED BY THE INTERNATIONAL ATOMIC ENERGY AGENCY AND HELD BETWEEN 1984 AND 1987



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#### FOREWORD

Since 1973, when the IAEA first introduced the subject of decontamination and decommissioning into its programme, twelve Agency reports reflecting the needs of the Member States on these topics have been published. These reports summarize the work done by various Technical Committees, Advisory Groups, and International Symposia.

While the basic technology to accomplish decontamination and decommissioning (D&D) is fairly well developed, the Agency feels that a more rapid exchange of information and co-ordination of work are required to foster technology, reduce duplication of effort, and provide useful results for Member States planning D&D activities. Although the Agency's limited finanical resources do not make possible direct support of every research work in this field, the IAEA Co-ordinated Research Programme (CRP) creates a forum for outstanding workers from different Member States brought into closer contact with one another to provide for more effective interaction and, perhaps subsequently, closer collaboration.

The first IAEA Co-ordinated Research Programme (CRP) on decontamination and decommissioning was initiated in 1984. Nineteen experts from 11 Member States and two international organizations (CEC, OECD/NEA) took part in the three Research Co-ordination Meetings (RCM) during 1984-87. The final RCM took place in Pittsburgh, USA, in conjunction with the 1987 International Decommissioning Symposium (sponsored by the US DOE and organized in co-operation with the IAEA and OECD/NEA).

The present document summarizes the salient features and achievements of the co-ordinated research work performed during the 1984-87 programme period. It was compiled by Mr. P.L. De (Scientific Secretary) of the Waste Management Section, Division of Nuclear Fuel Cycle.

The Agency would like to take this opportunity in acknowledging the excellent co-operation and hospitality of the CEA, France, and the US DOE in hosting the Second and Third Research Co-ordination Meetings respectively.

## EDITORIAL NOTE

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#### Part I

#### SUMMARY OF THE THREE RESEARCH CO-ORDINATION MEETINGS

#### **1. INTRODUCTION**

Decommissioning, as used in the nuclear industry, means the actions taken at the end of a facility's useful life to retire it from service in a manner that provides adequate protection for the health and safety of the decommissioning workers and the general public, and for the environment.

The decontamination and decommissioning of nuclear facilities are topics of great interest to many of the 113 Member States of the IAEA. A close look at the statistics will show the reasons for this interest. By 1987, 565 research and test reactors had been placed in operation in the world, over 400 of them between 1957 and 1970 [1]. Of the 326 units that are still in operation, many of them are over 20 years old and some could soon become candidates for decommissioning or refurbishment.

To-date, well over 140 nuclear facilities including research, test and prototype reactors have been decommissioned [2,3]. While the number of power reactors which will be decommissioned in the next 10 years will be small, it has been calculated [3] that by the year 2010, reactors equivalent to 200 GWe will be candidates for decommissioning or refurbishment in OECD countries. In addition to reactors, a wide variety of other nuclear fuel cycle and non fuel cycle facilities will have to be decommissioned.

Although no large power reactor has been completely dismantled, technical experts agree that sufficient experience has been gained so far to demonstrate that such dismantlment can be carried out without unacceptable impact on humans and the environment and at a reasonable cost [2,3]. Conceptual studies, projects and research support this viewpoint. However, even though progress has been made in the development of the technology and methodology of decommissioning, further work is required to improve equipment and techniques, reduce costs and exposures, and gain experience with larger facilities.

In response to increased international interest and to the needs of Member States, the IAEA activities in these areas have increased during the past few years and these activities will be enhanced in the future. To assist Member States in the development of the required decommissioning expertise, the IAEA is developing an integrated data base [2, 4 to 16] covering in a systematic way, the wide range of technical, regulatory and safety topics associated with the decontamination and decommissioning (D/D) of all types of nuclear facilities, sites and large contaminated areas.

As part of these activities, the IAEA also organized and sponsored the First Co-ordinated Research Programme on Decontamination and Decommissioning (CRP on D/D) during the period 1984 to 1987. This document is the final report of the CRP on D/D and summarizes the progress made.

#### 2. OBJECTIVES AND SCOPE

The objectives of this CRP were to:

- i) promote the exchange of information gained by different countries in D/D
- ii) stimulate cross-disciplinary and interdisciplinary research on all aspects of these topics
- iii) give the participants an opportunity to visit sites where decommissioning activities were in progress.

The scope of the programme was mainly oriented towards decontamination, however, selected decommissioning projects were included so that the participants could see how their work was related to overall decommissioning activities.

It was decided that this CRP would be somewhat different from other CRP's which tend to focus on narrow topics of research in the classical sense. In this CRP, therefore, the term research was considered in the broader sense to include topics related to many aspects of an integrated D/D programme.

#### 3. ORGANIZATION OF THE RESEARCH CO-ORDINATION MEETINGS (RCM)

#### 3.1 First RCM

The first RCM was held in Vienna from 26-30 November 1984. The meeting, which was chaired by Mr. J.I. Saroudis (Canada), was attended by two contract holders (CSSR and Hungary), seven agreement holders (Belgium, Canada, France, Italy, Japan, UK and USA) and several observers. The representative from India (agreement holder) was not able to attend. A list of participants and observers is given in Table 1, while a list of the formal presentations is given in Table 2. Mr. Cao Guan Ping of China gave a short presentation on the "Reconstruction of the HWRR of China during 1979-80" and Mrs. Dong Yin gave a short review of other decommissioning activities in China.

#### 3.2 Second RCM

The second RCM was held in Marcoule from 12-16 May 1986 and was hosted by the French Government and the Commissariat à l'Energie Atomique (CEA). This meeting was chaired by Dr. R.L. Lurie (CEA). Table 3 is a list of the participants and observers at the meeting. Papers summarizing the research work were presented at the meeting by the chief investigator or his delegate. In addition, papers describing the D/D programme of the IAEA, CEC and NEA (OECD) were presented. Table 4 lists the papers presented and discussed. Belgium and Italy were unable to attend this meeting.

In addition to the formal presentations at the Second RCM, the host institution (CEA-Marcoule) arranged two lectures as follows:

- Special presentation on the overall French decontamination work by Mr. Josso from Cadarache.
- Special presentation on the French programme on computer assisted teleoperators, remote cutting, collision avoidance and robotics by Mr. Clement from Saclay.

The participants were also given tours of several facilities at Marcoule, the most important one from the decommissioning point of view was to the G-2 reactor. In addition, visits were made to the Marcoule Vitrification Plant (AVM) and the TOR reprocessing plant.

#### 3.3 Third RCM

The third and final RCM was held in Pittsburgh from 4-9 October 1987 in conjunction with the 1987 International Decommissioning Symposium which was sponsored by the USDOE in co-operation with the IAEA and the NEA(OECD). Table 5 is a list of the participants and observers at the meeting. Eight of the 10 participants of the CRP presented their papers as part of the Symposium (Table 6). A separate session of the RCM was held on 9 October for presentation of the other two papers, general discussion on all papers and topics related to the CRP.

All participants of the RCM had a tour of the Shippingport reactor site which is currently being decommissioned.

During the closing session of the RCM on 9 October, the following specific items were discussed in detail:

i) Format of the final report of the CRP on D/D:

Since it would not be desirable to include all the papers from the three meetings in an IAEA report, it was agreed that each participant would prepare a 5 to 10 page summary of their work covering the three CRPs using the following format:

-	Introduction	-	Conclusions	and Recommendations
-	Objective	-	Future Work	Planned
-	Results and Discussion	-	References	

The final submissions of the participants in the above format are given in Part 2 of the report.

ii) Need and format of a new CRP on D/D

The need and format for a new CRP on these topics starting in 1989 were discussed.

All the participants felt that the present CRP had been a benefit to them both for the technical discussions and the visits to sites where D/D were in progress. They all stated that a new CRP on D/D should be initiated.

Some participants felt that the number of participants should be increased by three or four to get better participation from other countries including developing countries. Several participants suggested that the CRP should try to focus on one issue which was important to decommissioning e.g. cover all the aspects of decontamination, such as techniques, costs, criteria, secondary wastes, packaging, etc. Others felt that the present format was better. Since no clear consensus was evident as to format, participants were asked to think further on the topic and send suggestions to the Agency early in 1988.

iii) Having the CRP in conjunction with the Symposium

Having the CRP in conjunction with the Symposium was a great benefit to all participants, especially since some would not have been able to attend otherwise. However, the depth of informal discussion between participants on the papers was generally not as much as normally would occur in a regular RCM. Exceptions to this were the poster sessions where the participants had a wider discussion with experts in their particular discipline than would normally occur in a small RCM. It was concluded that although having the RCM in conjunction with the Symposium had some disadvantages, on balance it was a great benefit to all participants and the objectives of the CRP were achieved. Participants and observers at the first Research Co-ordination Meeting (RCM) on Decontamination and Decommissioning held in Vienna on 26-30 November 1984

COUNTRY	PARTICIPANTS	ADDRESS
BELGIUM	Mr. N. Van de Voorde	Centre d'Etudes Nucléaires Boeretang 200, Mol 2400
CANADA	Mr. J.I. Saroudis (Chairman)	Atomic Energy of Canada Limited CANDU Operations 1155 Metcalfe St, 2nd Floor Montreal H3B 2V6
CZECHOSLOVAKIA	Mr. E. Hladky	Nuclear Power Plants Research Inst. Dept. of Research and Racioactive Management Jaslovske Bohunice 919 31
FRANCE	Dr. R. Lurie	CEA, Institute de Protection et de Sureté Nucléaire CEN de la Vallée du Rhône Bagnols-sur-Cèze, Marcoule
HUNGARY	Dr. O. Pavlik	Institute of Isotopes Hungarian Academy of Sciences P.O. Box 77, Budapest 1525
ITALY	Dr. L. Lembo	ENEA Via mazzini 2, Bologna
JAPAN	Mr. T. Kikuyama	Dept. of JPDR, JAERI Tokai Mura, Ibaraki~Ken
UNITED KINGDOM	Dr. M.E. Pick	CEGB, Berkeley Nuclear Laboratories Berkeley, Gloucestershire GL13 9PB
	Mr. J. Stephenson	Risley Nuclear Power Development Establishment, UKAEA Risley, Warrington, Cheshire WA3 6AT
UNITED STATES OF AMERICA	Mr. C.E. Miller Jr.	Surplus Facilities Management Programme Office, US Department of Energy P.O. Box 550, Richland Washington 99352
CANADA	Mr. Balarko Gupta	Atomic Energy of Canada Limited CANDU Operations 1155 Metcale St., 2nd Floor, Montreal
CHINA	Mr. Cao Guan Ping Mrs. Dong Ying	Ministry of Nuclear Industry Bureau of Science & Technology & Nuclear Power P.O. Box 2102, Beijing
CEC	Mr. K.H. Schaller	CEC DG XII-SDM-1/48 Rue de la Loi 200, B-1049 Brussels Belgium
NEA/OECD	Dr. O. Ilari	Radiation Protection & Waste Management Division 38, Boulevard Suchet, F-75016 Paris

(Scientific Secretary: M.A. Feraday)

### <u>Table 2</u>

## Papers presented at the First Research Co-ordination Meeting (RCM on Decontamination and Decommissioning

#### IAEA, Vienna, Austria 26-30 November, 1984

	PAPER/SUBJECT	PARTICIPANT	INSTITUTE	M/S
1.	An overview of the IAEA's programme on decontamination and decommissioning (D/D) of nuclear facilities	Feraday, M.A.	IAEA	-
2.	An overview of the decommissioning and decontamination activities in the Commission's cost-sharing research programme on decommissioning	Schaller, K.H.*	CEC	-
3.	A short summary of NEA activities in the field of decommissioning of nuclear facilities	Ilari, O*	NEA	-
4.	An overview of the Surplus Facilities Management Programme (SFMP) including a presentation on the Shippingport Station (72 MW, PWR) Decommissioning Project. In future years research reports will be presented on topics of timely technical val being carried on in the SFMP decommissionin activities	Miller Jr., C.E. ue g	US DOE	USA
5.	An overview of the status of work associated with the decommissioning of the Windscale Advanced Gas-Cooled Reactor (30 MW) which was shut down in 1981, including an outline of the remote system technology being developed for this task	Stephenson, J.	UKAEA	UK
6.	Methodology of a computerized cost model for the decommissioning of NPP	Gupta, B. Saroudis, J.	AECL AECL	Canada Canada
7.	Measurement techniques of low-level radioactivity concentrations for unrestricted release decommissioning of RITMO and RANA reactors	Lembo, L.	ENEA	Italy
8.	Evaluation of radioactive inventory in in LWR systems	Kikuyama, T.	JAERI	Japan

\* Observer. Invited paper

Table	2	cont	
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	PAPER/SUBJECT	PARTICIPANT	INSTITUTE	M/S
9.	The characterization of radioactive deposits on samples from a variety of different PWR primary circuits to determine the chemical & physical structure of the layers & the distribution of radionuclides	Pick, M.	CEGB	UK
10.	The development of techniques & equipment associated with electrochemical decontam- ination by movable cathode including remotely controlled equipment, associated with future maintenance & decommissioning activities of PWR reactors	Pavlik, O.	Academy of Sciences	Hungary
11.	The characterization of surface films on piping from the G-2 ( $CO_2$ cooled GCR) reactor. Tests of various solutions to determine best method of breaking the film down.	Lurie, R.	CEA	France
12.	Study of decontamination & decommissioning of NPP under IAEA contract	Hladky, E.	NPP Research Inst.	CSSR
13.	The development of plasma torch techniques to decontaminate concrete surfaces & high pressure jet spraying (gas & liquid) with & without solid particles for the decontamination of metal surfaces and the development of remotely operated equipment for these applications	Van de Voorde, N.	CEN	Belgium
14.	Research & development on LWR system decontamiantion	Kikuyama, T.	JAERI	Japan

## Table 3

Participants and Observers at the Second Research Co-ordinated Meeting (RCM) on Decontamination and Decommissioning held in Marcoule on 12-16 May 1986

(Scientific Secretary: M.A. Feraday)

COUNTRY	PARTICIPANTS	ADDRESS
Canada	Mr. G. Pratapagiri	Atomic Energy of Canada Limited CANDU Operations 1155 Metcalfe St, 2nd Floor Montreal H3B 2V6
CZECHOSLOVAKIA	Mr. E. Hladky	Nuclear Power Plants Research Inst. Dept. of Research and Racioactive Management Jaslovske Bohunice 919 31
FRANCE	Dr. R. Lurie	CEA, Institute de Protection et de Sureté Nucléaire CEN de la Vallée du Rhône Bagnols-sur-Cèze, Marcoule
HUNGARY	Dr. O. Pavlik	Institute of Isotopes Hungarian Academy of Sciences P.O. Box 77, Budapest 1525
INDIA	Dr. J.L. Goswami	Bhabha Atomic Research Centre Trombay, Bombay 400 085
JAPAN	Dr. E. Tachikawa	Department of JPDR, JAERI Shirakata, Tokai Mura, Ibaraki
UNITED KINGDOM	Dr. M.E. Pick	CEGB Berkeley Nuclear Laboratories Berkeley, Gloucestershire GL13 9PB
UNITED STATES OF AMERICA	Mr. J. Schreiber	Shippingport Station Decommissioning Project, US Department of Energy Shippingport, Pennsylvania
	OBSERVERS	
CEC	Mr. K. Pflugrad	DG XII-SDM-1/48 Rue de la Loi 200, B-1049 Brussels Belgium

### Table 4

## Papers presented at the Second Research Co-ordination Meeting (RCM) on Decontamination and Decommissioning

## Marcoule, France 12-16 May 1986

	PAPER/SUBJECT	PARTICIPANT	INSTITUTE	M/S
1.	Overview of the IAEA's programme on D/D of Nuclear Facilities	Feraday, M.A.	IAEA	IAEA
2.	The OECD/NEA co-operative programme concerning nuclear installation	Menon, S. presented by	NEA	NEA
	decommissioning projects	Lurie, R.	CEA	France
3.	The EC research activities on the decommissioning of nuclear installations	Pflugrad, K.	CEC	CEC
4.	Report on computer codes for estimating decommissioning costs of NPPs	Pratapagiri, G.	AECL	Canada
5.	Results of the research on decontamination & decommissioning under IAEA contract	Hladky, E.	NPPRI	CSSR
6.	Decontamination of nuclear facilities by by electrochemical methods	Pavlik, O.	Academy of Sciences	Hung <b>a</b> ry
7.	Decontamination tests on the piping of $G-2$ reactor	Lurie, R.	CEA	France
8.	Decommissioning & decontamination of nuclear facilities: Indian programme under IAEA Research Agreement	Goswami, J.L.	BARC	India
9.	Evaluation of radioactive inventory in LWR system & R&D on LWR system decontamination	Tachikawa, E.	JAERI	Japan
10.	Characterization of the radioactive deposits on PWR primary circuit specimens	Pick, M.E.	CEGB	UK
11.	The status of the Shippingport Station Decommissioning Project	Schreiber, J.	USDOE	USA

## <u>Table 5</u>

Participants and Observers at the Third Research Co-ordinated Meeting (RCM) on Decontamination and Decommissioning held in Pittsburgh, USA on 4-9 October, 1987

(Scientific Secretary: M.A. Feraday)

COUNTRY	PARTICIPANTS	ADDRESS
Canada	Mr. G. Pratapagiri	Atomic Energy of Canada Limited CANDU Operations 1155 Metcalfe St, 2nd Floor Montreal H3B 2V6
CZECHOSLOVAKIA	Mr. E. Blazek	Nuclear Power Plants Research Inst. Dept. of Research and Racioactive Management Jaslovske Bohunice 919 31
FRANCE	Dr. R. Lurie	CEA, Institute de Protection et de Sureté Nucléaire CEN de la Vallée du Rhône Bagnols-sur-Cèze, Marcoule
HUNGARY	Dr. O. Pavlik	Institute of Isotopes Hungarian Academy of Sciences P.O. Box 77, Budapest 1525
JAPAN	Dr. E. Tachikawa	Department of JPDR, JAERI Shirakata, Tokai Mura, Ibaraki
UNITED KINGDOM	Dr. M.E. Pick	CEGB Berkeley Nuclear Laboratories Berkeley, Gloucestershire GL13 9PB
UNITED STATES OF AMERICA	Mr. J. Schreiber	Shippingport Station Decommissioning Project, US Department of Energy Shippingport, Pennsylvania
	OBSERVERS	
CEC	Mr. B. Huber	DG XII-SDM-1/48 Rue de la Loi 200, B-1049 Brussels Belgium
NEA/OECD	Mr. S.K. Menon	38, Boulevard Suchet F-75016 Paris France

## <u>Table 6</u>

#### Papers Presented at the Third Research Co-ordination Meeting (RCM) Pittsburgh, USA: 4-9 October 1987

	PAPER/SUBJECT	PARTICIPANT	INSTITUTE	M/S
1.	Decontamination for decommissioning of NPP A-1	Blazek, J.	NPPRI	CSSR
2.	Decontamination of nuclear facilities by electrochemical methods	Pavlik, O	Academy of Sciences	Hungary
3.	Characterization of radioactive deposits on PWR primary circuit surfaces	Pick, M.E.	CEGB	UK
4.	Computer programme for estimating decommissioning costs of NPP	Pratapagiri, G.	AECL	Canada
5.	Shippingport Station Decommissioning Project	Schreiber, J.J.	USDOE	USA
6.	Decontamination and dismantling of G-2 reactor circuits*	Lurie, R.	CEA	France
7.	Research and development on LWR system decontamination, mechanical and Redox decontamination methods*	Tachikawa, E.	JAERI	Japan
8.	IAEA activities in decommissioning and decontamination	Feraday, M.A.	IAEA	IAEA
9.	International co-operation on decommissioning within the OECD/NEA	Menon, S.K.	Studsvik	Sweden
10.	Advances in the EC programme of research on decommissioning	Huber, B.	CEC	CEC

All papers except those marked \* were presented at the 1987 International Decommissioning Symposium in Pittsburgh, USA.

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#### 4. REVIEW OF THE SCIENTIFIC AND TECHNICAL PAPERS

#### Canada

Work was reported on the development and enhancement of a cost estimating program (called DECOM) using a microcomputer (IBM PC or compatible). The program is user-friendly, menu driven and written in the dBase III format.

With the help of this program, costs can be estimated for Stage 1, Stage 2 or Stage 3 decommissioning. The program is based on the generally accepted concept of unit cost factors.

The computer program is suitable as an aid in the selection and planning of a decommissioning alternative. Apart from its application in the Gentilly-1 decommissioning project, the program had been used in the preparation of cost estimates for both CANDU (<u>Canada Deuterium Uranium</u>) and light water reactors. In order to validate the code, samples of actual cost and manhour data from the Gentilly-1 project were processed through the DECOM code. It was observed that the total costs were accurate within 20%, though the costs for individual activities in some instances varied significantly due to differing project parameters.

#### Czechoslovakia

As part of a programme carried out in Czechoslovakia in connection with decommissioning of the Nuclear Power Plant A-1, a study has been conducted on decontamination of materials and their possible reuse. The objectives of this study were to select suitable decontamination agents, decontamination efficiency by use of ultrasonic and electrochemical techniques, and to develop a computer code applicable for reuse of materials. The study was focused on carbon steel surfaces of the secondary circuit and stainless steel of the explosive mixture combustion system of the NPP A-1.

Metallographic examinations of construction materials have shown that in case of carbon steel surfaces the corrosion products are formed by two layers, the upper layer contained mainly magnetite with high amounts of copper, manganese and zinc, the lower one was composed of hematite, goethite and small oxide particles. Corrosion products on stainless steel surfaces were represented by various ferrous and ferric compounds; presence of ferrous oxalate has also been proved by X-ray analysis.

From a series of decontamination tests run with various solutions it has been shown that the decontamination efficiency could substantially be enhanced by simultaneous application of an ultrasonic treatment. This is valid for low-alloy steel surfaces treated with a mixture of formic acid and di-sodium-EDTA for 0.5-2 hours at  $50^{\circ}$ C, followed by removal of the residual contamination with the same solution or with the help of sulphuric or phosphoric acids. As regards the stainless steel, the 0.8% nitric acid, and a mixture of 1.5% formaldehyde with 9% formic acid and 0.33% of di-sodium EDTA were successful for decontamination prior to dismantling of equipment. Electrochemical decontamination of low-alloy steels showed that this technique can be applied for reduction of contaminated concentrations to the levels, allowing unrestricted use of the material. Electrolytes such as nitric, sulphuric, phosphoric and oxalic acids were used. It has been confirmed that the time period necessary for decontamination depends mainly on current density and thickness of the layer to be dissolved. For the given material and current density of 150 A.dm<sup>-2</sup>, necessary times ranged between 30 and 60 minutes.

Electrochemical decontamination tests of stainless steel showed that anodic oxidation and regimes with reverse polarities were more effective than cathodic reductions.

In developing a computer code for unrestricted release of materials on the basis of the individual exempt dose criterion 10  $\mu$ Sv/a, the following conclusions could be drawn:

- 1) off-site storage of material can enhance the total risk of the practice and should, therefore, be minimized
- 2) important risk is represented by slag material
- 3) the material can be released without any risk at radionuclide activity of  $1 10 \text{ Bq.g}^{-1}$  and at total value of surface contamination of 4 Bq.cm<sup>-2</sup>
- 4) value of collective dose equivalent will be decisive for assessment of the total amount of material that can be released to the environment.

#### France

The decommissioning programme for the gas-cooled graphite reactor G2 at Marcoule, France includes among others, decontamination of the facility in such a manner that the solutions are as small as possible and the process is carried out remotely. Low contamination levels (in average 33 Bq.cm<sup>-2</sup>) and large amounts of construction materials (2500 tons) led to the selection of decontamination procedures which would permit the reuse or recycle of the construction material.

To reach these goals, a study composed of the following steps has been carried out:

- review of various decontamination techniques applicable for the given facility and selection of the most appropriate one
- testing of the selected method under laboratory conditions
- full scale testing on a first part of the circuit.

Two approaches have been considered, the first consisting of in-situ decontamination, the second of dismantling the materials first and decontaminating them subsequently in a central decontamination facility. The final decision, however, has not yet been taken.

Laboratory testing included application of various acids (HF,  $HNO_3$ , HCl and  $H_2SO_4$ ) and their mixtures of gels and foams, high pressure jets, and electrochemical decontamination methods. With acidic solutions decontamination factors over 100 were obtained in time spans less than 2 hours; with gels and foams similar results could be reached, the latter method proving to be less aggressive and resulting in removal of lower amounts of material treated.

Experiments with high pressure jets were successful at pressures of about 45 MPa, flows up to 20 m<sup>3</sup>.h<sup>-1</sup> and distances from 2.5 to 10 cm between the nozzle and the sample. Electrochemical decontamination was tested using a movable electrode, composed of stainless steel, glass fibers and felt. It was noticed that good efficiency can be obtained with sulphuric and phosphoric acids as electrolytes.

Full scale tests were performed in 1986 and 1987 with 30 tons of steel  $(300 \text{ m}^2)$  using the gel technique which during laboratory testing offered most promising results. Both steps, spraying of the gel and rinsing, were done by hand. Initial activity of 200 Bq.cm<sup>-2</sup> dropped to 2 Bq.cm<sup>-2</sup>, thus obtaining the decontamination factor of 100. Volume of liquid effluents decreased after some improvements of the technique to 12 L.m<sup>-2</sup>.

The results confirmed that the gel technique can successfully be used, where contaminated surface is accessible for gel spraying. In order to adapt the reagent to the type of base material and to the contaminant, thorough laboratory tests have to be carried out.

#### Hungary

Work was reported on electro-chemical decontamination methods for maintenance of the Paks nuclear power plants in Hungary. For insitu decontamination of large components, for example, main circulating pump, gate valves, steam generator collectors, methods using remotely operated movable electrodes were implemented successfully.

For the main circulating pump, the electrolyte contained phosphoric, sulfuric and oxalic acids. Before decontamination, the inlet nozzle for reactor coolant was plugged in such a way that the electrolyte could not enter the connecting pipe. The spent electrolyte accumulated on the bottom of the treated pump case and was drained into the radioactive effluent stream. After electrochemical decontamination the inner surface of the pump case was rinsed with a 12 g.1<sup>-1</sup> boric acid solution. The concentration of the radioactive isotopes and the corrosion products was measured in the spent electrolyte and in the rinsing solution too. About 150 1 liquid radioactive waste (together with rinsing water) was produced during the decontamination procedure. The duration of the decontamination was 6-8 hours. The procedure was carried out by 3-4 workers, their collective dose was 2-3 mSv. The decontamination was followed by chemical and radiometric analyses.

The criteria for the selection of electrolytes were:

- high decontamination factor
- low corrosion rate
- does not develop aggressive gas
- high electric conductivity
- relatively low electrolyte feed rate and low concentration of chemical;
   does not produce a big quantity of radioactive waste.

The electrolyte used was  $H_3PO_4$  40 g.1<sup>-1</sup>, +  $H_2SO_4$  30 g.1<sup>-1</sup>, +  $H_2C_2O_4$  20g.1<sup>-1</sup>, or citric acid 20 g.1<sup>-1</sup> +  $H_2C_2O_4$  20 g.1<sup>-1</sup> electrolytes. This was the result of a compromise.

The electrolyte feed rate was 20-40  $l.h^{-1}.dm^{-2}$ .

The remotely controlled electrochemical decontamination method using movable cathode has the following advantages:

- high decontamination factor (20-500)
- short application time
- produces smooth surface which reduces recontamination
- low collective dose
- low volume of liquid radioactive waste.

#### Disadvantages:

- The movable cathode cannot clean the whole surface to be decontaminated, if the surface has a complicated geometry or profile
- The above-mentioned remotely operated movable heads can decontaminate only equipments with well defined dimensions.

#### Japan

In order to satisfy requirements for efficient crud removal from contaminated surfaces and an easy subsequent treatment of spent decontamination solutions, two methods have been developed, tested and applied in decommissioning of the Japan Power Demonstration Reactor plant. The specimens of pipes and tubes of the primary system were thoroughly examined with use of chemical analysis, X-ray, activity measurement and electron microscope. High content of Cr in the crud was ascribed to poor quality control of cooling water in the early stage of plant operation.

Since decontamination with common chemical reagents was not effective, further tests have been modified in two ways. The first method consisted of a mechanico-chemical procedure, in the second various redox decontamination reagents were used. Subsequently, laboratory testing was replaced by large scale tests performed in decontamination loops, designed and constructed for the given purpose. The following results were obtained:

The combined mechanico-chemical process, composed of treatment with an abrasive (boron carbide 0.5 mm in diameter) suspended in water up to 20 wt% and with a circulating time of 48 hours, proved to be an efficient method. The main features observed were as follows:

- at 12 hours decontamination, more than 97% of the activity was removed from the sample specimens.
- by continuing the decontamination up to 18 hours after replacement of the abrasive, the removed fraction reached to 99%
- increase of the flow rate from 4.8 m.sec<sup>-1</sup> to 6.7 m.sec<sup>-1</sup> positively affected the detached fraction, although the extent was not significant
- at 35 hours decontamination, the removed fraction was about 99.9%, further removal seemed to need much longer decontamination times.

The rate of weight decrease of the samples during the decontamination was more or less linear with decontamination time, but the rate depended upon the flow rate and increased with increasing flow rate, as was expected.

The ratio of the activity removed to the weight-decrease during the decontamination continuously decreased with time. This might be understood to show that during the decontamination rather uniform surface removal occurs, while the activity becomes sparse as the removal proceeds.

It could therefore be concluded, that high DF's can be obtained as follows:

- a high DF can be obtained in a relatively short decontamination time, being independent of the chemical composition of crud
- the resulting waste solution can be easily treated
- decontamination efficiency is almost independent of temperature and decontamination can be carried out under the atmospheric conditions
- decontamination procedure is relatively simple.

The disadvantages are:

- a positive counterplane is needed to eliminate the trapping of abrasives during decontamination, particularly when the item to be decontaminated has a complexed structure
- a relatively large capacity circulation pump is needed to obtain a flow rate sufficient to circulate the abrasives.

The study of a suitable redox decontamination process consisted in the examination of various redox reagents tested under different conditions. The results led to  $Ce(IV) - H_2SO_4$  solution as the most promising candidate for successful decontamination. It dissolves the crud through oxidizing its components by contemporaneous reduction of Ce(IV) to Ce(III) which, later on, can be reoxidixed by an electro-chemical method to its original state.

In-loop experiments led to the following results:

- activity removed: 3.8 mCi (Calculation gives 2.3 mCi).
- decontamination factor: 300 1800
- metal removed: 10 kg
- solid waste: three 100 liter drums (for concentrated waste solutions and sludges)
   two 200 liter drums (for IX resins).

In decontamination of samples from some NPPs by various decontamination reagents it has been observed that the redox method gives a satisfactory decontamination factor, regardless of the Cr-content and can be successfully used as a decontamination method for pre- and/or post-dismantling of a reactor.

In examining decommissioning strategies for LWR's and the possible role of decontamination, characterization of the radioactive deposits on circuit surfaces is required to provide information on the radioactive inventory and the type of oxide on the surface. Knowledge of the latter will determine which is the most appropriate decontamination process to use and its potential efficiency. Results from examinations performed on a number of Inconel 600 steam generator (SG) and stainless steel PWR specimens and also a limited number of BWR and CANDU speciments are summarized. A variety of techniques have been utilized including: gamma spectrometry, alpha spectrometry, scanning electron microscopy and wet chemical analysis. In addition, preliminary studies using secondary ion mass spectrometry (SIMS) have been performed The sources of the major radionuclides present on circuit surfaces are considered

From the results of the study the following conclusions could be drawn:

The compositions of oxides formed on Inconel SG tube and stainless steel PWR specimens are very similar. Both are enriched in chromium by about a factor of two over the base metal chromium concentration of 16-18%. The other major elements in the oxide are iron and nickel. In addition, manganese, titanium, silicon, cobalt, copper and zinc are present at levels of a few per cent or less.

Oxide thicknesses on stainless steel specimens are greater than on Inconel SG tube after a similar EFPY, this may be due to the much rougher surface on the stainless steel specimens.

The predominant gamma-emitting radionuclide on the speciments is  ${}^{60}$ Co, formed from  ${}^{59}$ Co. It is considered that the major source of the  ${}^{60}$ Co is the high cobalt alloy Stellite. Most of the other gamma-emitting radionuclides measured in significant quantities on the specimens, e.g.  ${}^{54}$ Mn,  ${}^{58}$ Co,  ${}^{65}$ Zn,  ${}^{106}$ Ru,  ${}^{125}$ Sb are shorter-lived than  ${}^{60}$ Co, which will therefore dominate radiation fields in the immediate period after shutdown;  ${}^{94}$ Nb, which is likely to be the dominant contributor to radiation fields after long decay periods, was detected on one of the specimens examined.

Alpha emitting actinides deposited as a result of fuel failures were detected on nearly all the specimens examined; typical levels ranged up to 5  $Bq.cm^{-2}$  of longer lived actinides on PWR specimens and 100  $Bq.cm^{-2}$  on BWR specimens. Clearly, their presence on out-of-core surfaces must be taken into account in assessing decommissioning scenarios.

Developments by the CEGB have led to improved decontamination processes for use on PWR and BWR surfaces. These processes based on NP or AP and LOMI reagents have been used in over 20 major applications to date mainly on reactors and components being returned to service. However, the possible application of these processes in a multi-cycling process, to provide high DF's has been investigated and DF's of over 100 on stainless steel specimens have been obtained, on Inconel SG tube DF's are less satisfactory but this is probably due to radioactivity present in grain boundaries up to 10  $\mu$ m or so into the metal.

<u>UK</u>

A general project overview of the Shippingport Station decommissioning programme was given. This includes the background of the project, and the development and implementation of plans for the management, engineering and site operations. The technical objectives of the project are to:

- remove equipment and material so that the site can be released for unrestricted use;
- assure the transfer of decommissioning technology to as many U.S.
   companies as possible through sub-contracts;
- document project data for long-term storage and retrieval for use in future decommissioning projects.

Removal of reactor and internals in one-piece is a special feature of this project. Physical work of decommissioning started in 1985 with the site release scheduled for 1990.

<u>USA</u>

### 5. CONCLUSION AND RECOMMENDATIONS

Based on the discussions at the three RCMs and subsequent communication with the experts who attended the RCMs it was concluded that the present CRP on D/D was a real benefit to all participants both from the technical discussions and from the site visits where D/D were in progress. It was recommended that:

- it would be worthwhile to initiate another CRP. Although the theme and membership in the CRP was not yet decided, it was suggested that the number of participants should be increased by 3 or 4 to include participation from other countries, especially developing Member States.
- ii) the new CRP should include topics related to many aspects of D/D, such as decommissioning project management, dismantling, decontamination, etc., instead of focussing on one technique only. This format would generate better awareness in D/D and would be a more effective vehicle for exchange of information by stimulating broader discussion on all aspects of D/D.

#### **ACKNOWLEDGEMENTS**

The organizers and particpants of the RCMs would like to acknowledge the excellent co-operation and hospitality of the CEA France (second RCM) and the USDOE (third RCM) in hosting the RCMs away from the IAEA headquarters in Vienna. The participants would like to thank the IAEA for organizing and funding the 1984-87 CRP on D/D.

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Part II

## FINAL SUBMISSION BY PARTICIPANTS ON THE RESEARCH WORK PERFORMED DURING 1984–1987

## FINAL REPORT ON COMPUTER CODES FOR ESTIMATING THE DECOMMISSIONING COST OF NUCLEAR POWER PLANTS

G. PRATAPAGIRI Atomic Energy of Canada Limited, Montreal, Canada

#### Abstract

Work is reported on the development and enhancement of a cost estimating program (called DECOM) using a microcomputer. The program is user-friendly, menu driven and written in the dBase III format. With the help of this program, costs can be estimated for Stage 1, Stage 2 or Stage 3 decommissioning. The program is based on the generally accepted concept of unit cost factors. The computer program is suitable as an aid in the selection and planning of a decommissioning alternative. Apart from its application in the Gentilly-1 decommissioning project, the program had been used in the preparation of cost estimates for both CANDU (<u>Canada Deuterium</u> <u>Uranium</u>) and light water reactors. In order to validate the code, samples of actual cost and manhour data from the Gentilly-1 project were processed through the DECOM code. It was observed that the total costs were accurate wihin 20%, though the costs for individual activities in some instances varied significantly due to differing project parameters.

#### 1.0 INTRODUCTION

As many nuclear power plants around the world approach the end of their expected lives, decommissioning has taken on more than an academic interest. Worldwide, a number of plants have been offered for various stages of decommissioning in recent years. These include Gentilly-1 in Canada, the power demonstration reactor (JPDR) in Japan, Windscale (WAGR) in the United Kingdom, Shippingport in the USA, and several others within the European Community.

Currently, there are several decommissioning alternatives that are technically, socially, and politically acceptable. A reliable cost estimate is essential to assist in the planning and selection of the most suitable decommissioning programme from among the options available, and to establish a practical funding mechanism for it.

The preparation of such cost estimates is a complex task because there are a large variety of plant inventories, radioactivity levels, waste categories and decommissioning options. In addition, a number of "what-if" questions regarding cost-benefit analysis must be answered before a decision is made as to which stage a plant will be decommissioned.

Atomic Energy of Canada Limited (AECL) faced this situation in 1983 during the initial decommissioning studies on the Gentilly-1 nuclear station and came to the conclusion that a computerized cost model was essential to permit the analysis of numerous decommissioning scenarios and for optimization purposes. To address this requirement, a computer program called DECOM was developed for use on an IBM main frame computer using a utility program called ADRS (<u>A</u> Departmental Reporting System).

Under the Coordinated Research Program (CRP) Agreement number 3960/CF, in Decommissioning and Decontamination, organized by the International Atomic Energy Agency (IAEA), Vienna, further development work was undertaken on the DECOM Code (Ref. 1, 2 and 3).

#### 2.0 OBJECTIVES OF THE WORK

The objectives of the work is to improve the flexibility, capability and versatility of the computer code by implementing the following requirements:

- Enhancing all capabilities of the code to integrate all elements of costs, i.e., Activity and Period Dependent costs, Special Items and Dormancy (storage) Period costs and produce a final cost report.
- Transfer the code from main frame computer (ADRS format) to micro-computer (IBM-PC with dBase III format).
- Selection of disposal containers as per volume, weight and density of different levels of radioactive waste generated.
- Analysis of costs based on various scenarios of decommissioning (stage 1, 2 or 3 as per IAEA guidelines).
- Flexibility to consider esscalation and discount factors for upto 200 years while planing cash flow requirements for a decommissioning project.
- Improvements to make the code menu driven and user friendly.

#### 3.0 RESULTS AND DISCUSSION

In 1985, in order to provide more flexibility and ease of operation for the users of the DECOM computer program, the DECOM Code was converted for use on an IBM-PC type microcomputer.

By the beginning of the third CRP meeting held at the 1987 International Decommissioning Symposium held in Pittsburg, U.S.A., the AECL DECOM, as it is now called, has been fully enhanced to take into consideration costs for decommissioning a nuclear station to all three stages of decommissioning i.e. stage 1, stage 2 and stage 3 as defined by IAEA, along with "on site storage" costs of radioactive waste and long term storage costs of nuclear stations where a delayed dismantling is envisaged.

#### 3.1 COST ESTIMATING METHODOLOGY

The methodology used in the AECL DECOM computer cost estimating program has been developed based on generally accepted principles of decommissioning cost estimating. (Ref. 4 and 5).

The components that make up the total cost for decommissioning a nuclear plant have been grouped into four categories, each of which need to be handled in a slightly different fashion. These are:

- a) activity dependent costs
- b) period dependent costs
- c) special item costs (or collateral costs)
- d) dormancy period costs (if delayed dismantling is envisaged)

Activity dependent costs are those associated with tasks that are discrete, measurable and of a repetitive nature and can thus be analysed by developing typical Unit Cost Factors (UCF)\* which can be applied to the category of equipment that they represent (e.g. cutting pipe, removing pumps, dismantling structural steel). The type and category of equipment which are to be dealt with dictates the type and number of cost factor models that need to be developed for a given nuclear facility.

Period dependent costs are those associated with the durations of different phases of the project such as engineering, project and construction management, licensing, quality assurance and security.

Special item costs are split into two categories: special items that are non-repetitive such as the reactor vessel removal and miscellaneous items such as the operation and maintenance cost, cost of energy and the like.

Dormancy period cost are those associated with long term storage of a Nuclear Station which may be between 40 years to 100 years (typically). These may be; costs for security and maintenance, energy, periodic surveys etc.

3.2 GENTILLY-1 PLANT

During the studies associated with the G-1 plant, decommissioning cost estimates for stages 1, 2 and 3 were prepared, using the DECOM computer program (ADRS Version).

This section describes the step by step approach which was pursued to prepare the G-l stage 3 decommissioning cost estimate and cash flow. This approach can be applied to any nuclear power plant. Major steps were as follows:

- Survey of equipment inventory
- Application of a computer code
- . Survey of radioactive inventory
- . Radiological exposure to workers (man-rem)
- . Development of unit cost factors
- PERT/CPM network to determine critical path
- Manpower requirements
- . Integration of cost and schedule
- . Summary of costs
- . Financial analysis and cash flow

The physical inventory of all the plant components (equipment, structures, etc.) was obtained from a room by room survey. All the components were grouped first into major equipment categories such as pumps, tanks, heat exchangers. Each major equipment group was further divided into subgroups which could represent a component for any type of plant. All component items were then entered into the database of the DECOM computer program.

In order to estimate radiological doses to the workers, a survey of radioactive inventory was done for each component in every room of the plant. For each component, two dose readings were taken: at contact (1 cm away) and at one meter distance. The background radiation in the centre of the room was also measured. These data were entered in the DECOM to calculate man-rem exposure to the workers based on the number of workers and the duration they handled the components.

A detailed CPM was developed from a master schedule and was computerized through a CPM/PERT program. Each activity on the schedule was given an identification number (1, 2, 3, etc.). All cost items in the data base associated with one scheduled activity carried the same number, as a link between the CPM program and the cost estimate code. For example, all cost items associated with activity no. 3 - "clear feeders and steam drums", were grouped together and summarized to facilitate accurate cash flow computation. The activity dependent costs associated with the schedule were then added to the period dependent and special item costs to obtain total costs. Several alternative decommissioning scenarios were then studied.

The experience from the G-l study suggests that although total cost is an important factor in choosing a preferred decommissioning alternative, it may not necessarily be the most dominant one.

Other considerations such as the annual cash availability, future use of the site, availability of a radioactive waste disposal facility play important roles in selecting a decommissioning alternative. Each plant should be treated individually.

It must be emphasized however that the estimates helped in optimizing the decommissioning decisions given the financial and other constraints imposed on the project.

#### 3.3 OTHER APPLICATIONS

Apart from use on the the Gentilly-1 project, the DECOM program has been used in the past few years in the preparation of decommissioning cost estimates for both CANDU (<u>Canada Deuterium Uranium</u>) and PWR (Pressurized Water Reactor) type reactors. Specific Applications are for SAN ONOFRE Units 1, 2, 3 in the USA, Point Lepreau-2 (hypothetical case) and NPD (Nuclear Power Demonstration Station) in Canada.

The estimates have been found to be within the range reported in the OECD/NEA decommissioning cost surveys (Ref. 6). It can be adapted to other types of reactors and may be extended to non-nuclear facilities with suitable modifications of the cost codes.

#### 3.4 VALIDATION OF THE CODE WITH SITE DATA

In order to test the validity of the cost estimates prepared using DECOM, a sample of actual cost and manhour data from the Gentilly-1 Decommissioning operation were processed through the DECOM code and it was observed that the total cost figures were accurate within a 20% range, though costs for individual activities in some instances differed significantly due to changes in the project technical concepts, reduction in the production work day because of unanticipated clothing changes, showers or breaks, increased radiation protection coverage provided, and special features such as asbestos removal. However, sufficient confidence has been developed in the capabilities of the DECOM Code through this experience. UCFs built into the code have been updated based on this bench marking exercise. The accuracy of the estimates can be further improved by constantly reviewing the UCF's in the DECOM code and adjusting them as required.

#### 4.0 CONCLUSIONS AND RECOMMENDATIONS

For effective pre-planning for decommissioning, the need for a credible cost estiamte cannot be over-emphasized. A logical, standardized and consistant estimating method will assist greatly in the decision making process leading up to the selection of a decommissioning alternative and/or in establishing a decommissioning estimate for ratebase setting purposes.

The large amounts of data to be considered, the issue of radiation exposure to workers, the waste categories generated and the large number of decommissioning scenarios that can be analyzed make this a time consuming and tedious process if done manually. Computerization, using a code like AECL-DECOM, represents a feasible and attractive alternative to preparing such cost estimates manually.

The AECL DECOM computer program is a versatile tool for applications in decommissioning studies.

It has already been used successfully to estimate costs for two CANDU type reactors and three PWRs. It can be adapted to any type of nuclear reactor.

The IAEA coordinated research program provided the opportunity to develop and enhance the computer program to its present form. The AECL DECOM provides even greater flexibility and acceptability than its predecessor (DECOM), since it is menu driven, user friendly and in a dBase III format that can be run on any IBM PC-XT or equivalent compatible microcomputer.

#### 5.0 FUTURE WORK PLANNED

The following additional features are being considered for the AECL DECOM.

- Unit costs of decontamination
- Unit costs of asbestos removal
- Resource based period dependent costs
- Further improvements in program flexibility to make the system user friendly

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## DECONTAMINATION FOR DECOMMISSIONING OF NUCLEAR POWER REACTORS

E. HLADKÝ, J. BLAŽEK,
D. MAJERSKÝ, V. ŘEHÁČEK
Nuclear Power Plants Research Institute,
Jaslovské Bohunice, Czechoslovakia

#### Abstract

As part of a programme carried out in Czechoslovakia in connection with the decommissioning of the Nuclear Power Plant A-1, a study has been conducted on decontamination of materials and their possible reuse. The objectives of this study were to select suitable decontamination agents, assess decontamination efficiency by use of ultrasonic and electrochemical techniques, and develop a computer code applicable for reuse of materials. The study was focused on carbon steel surfaces of the secondary circuit and stainless steel of the explosive mixture combustion system of the NPP A-1. From a series of decontamination tests run with various solutions it has been shown that decontamination efficiency could substantially be enhanced by simultaneous application of an ultrasonic treatment. Electrochemical decontamination of low-alloy steels showed that this technique can be applied for reduction of contamination to the levels suitable for unrestricted use. Electrolytes such as nitric, sulphuric, phosphoric and oxalic acids were used. It has been confirmed that the time period necessary for decontamination depends mainly on current density and thickness of the layer to be removed. Electrochemical decontamination tests of stainless steel showed that anodic oxidation and regimes with reverse polarities were more effective than cathodic reductions.

#### 1. INTRODUCTION

Nuclear Power Plant A-1 /NPP A-1/ with CO<sub>2</sub> cooled and heavy water moderated reactor with 150 MWe output was definitely shut/down in Febr. 1977, after 5 years of operation. Considering existing technological base, availability of waste management and disposal facilities in Czechoslovakia the NPP A-1 is currently under decommissioning to the first upgraded stage /according to the IAEA classification/, with partial dismantling of the secondary and selected auxiliary circuits. Among operative decommissioning problems the issues concerning decontamination of materials from the circuits and metallic LLW management have to be studied.

#### 2. OBJECTIVES OF THE WORK

The main objectives of our research work within this Contract were:

- choice of suitable solutions for the chemical decontamination of both carbon and stainless steels from the NPP A-1 circuits including assessment of their efficiency in the decontamination of real samples as well as the possibility to intensity chemical decontamination by means of ultrasound /US/
- development of electrochemical processes for decontamination of materials from the NPP A-1
- development of the model and computer code for determination of residual contamination limits allowable on metals from the NPP A-1 designated for reuse.

#### 3. RESULTS AND DISCUSSION

For decontamination experiments carried out in the laboratory and also for characterization of corrosion products layer by optic microscopy, SEM, X-ray diffraction and Moessbauer spectroscopy samplex were taken from different parts of secondary circuit /carbon steel/ and from the inlet pipe of the explosive mixture combustion system /stainless steel/. The samples were cut to the size of 30 x 30 mm.

Decontamination efficiency was evaluated by decontamination factor /DF/.

### 3.1. Characteristics of contaminated corrosion layer

Results of phase analysis have proved <sup>1</sup> that corrosion layer on carbon steel surface can be divided into two specific parts:

- a/ upper layer is thin, compact. It consists of substituted magnetite with higher content of Cu, Mn and Zn,
- b/ lower layer, adhering to the base material represents a bigger part of the entire corrosion layer. This part consists of compact, mechanically firm magnetite /thickness from 0.1 to 0.3 mm/ with different degree of nonstoichiometry and smaller portion /up to 30 %/ of hematite, goethite and small particles of oxides. The whole layer is unhomogeneous.
A part of the material has contaminated layer loosely bound to the base material.

Contamination is represented by at least 94 % of  $^{60}\mathrm{Co}$  and the rest is  $^{137}\mathrm{Cs}_{\bullet}$ 

The contaminated layer of stainless steel contains a small portion of iron oxides /cca 4 %/ and from 40 % to 50 % of another compounds of Fe<sup>2+</sup> and Fe<sup>3+</sup>. A presence of  $\text{FeC}_2O_4 \cdot 2H_2O$  was proved by X-ray analysis. On a basis of analyses results and determination of metals in decontamination solutions we can presume that contamination layer is formed by metallic compounds /Fe, Ni/ with radiation-chemical products of D<sub>2</sub>O and entraining CO<sub>2</sub> radiolysis.

3.2. Chemical decontamination

For experimental verification of efficiency have been 2-4 chosen decontamination agents referred to in the literature 2-4 as successful for low-alloy and stainless steels and solutions whose composition was based on better familiarity with processes of corrosion layers dissolving. An influence of ultrasonic /US/ impact on decontamination efficiency was experimentally verified at simultaneous treatment with ultrasound and the agent and also the impact of ultrasound after chemical decontamination of samples.

<u>Decontamination of Low-Alloy Steels.</u> The decontamination efficiency by oxalic acid of  $10 - 50 \text{ g.dm}^{-3}$  concentration at  $T = 90 \,^{\circ}\text{C}$  and exposure up to 3 hours, was low and DF < 3. The efficiency was increased by addition of  $\text{H}_2\text{O}_2$  or  $\text{H}_2\text{SO}_4$  respectively, but in spite of this, it was still low.

The DF  $\leq$  2.8 was obtained with the mixture of acids, such as: HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>, H<sub>3</sub>PO<sub>4</sub>, concentrations within 0.2 - 1.0 mol.dm<sup>-3</sup>, at T = 50 °C and 3 hours exposure.

From the DF values obtained after decontamination in the solutions of  $H_2SO_4/0.5$  and 1 mol.dm<sup>-3</sup>/and  $H_3PO_4$  /1 mol.dm<sup>-3</sup>/ with or without such additives as  $Na_2S_2O_3$ , sulphur, EDTANa<sub>2</sub> we can further state that in general it is possible to achieve DF = 100 - 1000 after 3 - 5 hours decontamination. Residual level of contamination  $n_{res} \notin 0.37$  can by achieved on that material by alternative action of  $H_2SO_4$  or  $H_3PO_4$  solution and ultrasound or by utilization of fresh solutions at repeated

decontamination. Decontamination in  $H_2SO_4$  or  $H_3PO_4$  solution is faster at simultaneous ultrasound treatment but acceleration depends on ultrasound field intensity.

The results of performed decontamination tests in the reducing mixture of  $CH_2O$ -HCOOH are reported in Tab. I and Tab. II.

Solution conc.	t = 3 hours	$t \approx 5$ hours	+ 0.5 hour US in $H_2^0$
∠_mol.dm_3_7	<b>▲</b> m <b>/</b> <sup>-</sup> %_7 DF	Am [~%_7 DF	am [%_7 DF
сн <sub>2</sub> 0 <u>/</u> 0.4 <u>8</u> 7 нсоон <u>/</u> 0.4 <u>8</u> 7	l 2.97	1.8 25.8	1.81 60
сн <sub>2</sub> о <u>/</u> б.4 <u>8</u> 7 нсоон <u>/</u> б.2 <u>4</u> 7	0.48 2.6	1.85 42.5	1.94 112
сн <sub>2</sub> 0 <u>/</u> 0.2 <u>4</u> 7 нсоон <u>/</u> 0.4 <u>8</u> 7	1.08 2.86	1.9 45.7	1.94 86.8

Table I Decontamination tests in  $CH_2O$ -HCOOH mixture at 50  $^{O}C$  with subsequent ultrasonic treatment

Am - mass loss of a sample after decontamination as percentage of the initial mass

Table II DF values in the solutions during decontamination at 50 °C at simultaneous effect of ultrasound

Solution conc. mol.dm <sup>-3</sup> _7	t = 30 min DF	t = 60 min DF	t = 120 min DF
нсоон /0.4/	1.42	1,82	3.6
HCOOH /0.19/ EDTANa <sub>2</sub> /0.01/	5,8	142	297
HCOOH /1.07/ HCOONa /0.05/ EDTANa <sub>2</sub> /0.01/	6 371	1 851	
HCOOH /1.07/ CH_0 /0.4/ HCCONa /0.05/ EDTANa2/0.01/	24.3	349	1 658

In the process of chemical decontamination not only dissolving of corrosion layer occurred.

The decontamination efficiency of  $CH_2O-HCOOH$  solution can be enhanced 2 - 3 times by subsequent ultrasonic treatment of samples in pure water after their chemical decontamination.

The decontamination efficiency of this solution was also very good at the laboratory temperature but the long exposure of samples was necessary.

Decontamination to  $n_{res} \le 0.37$  Bq.cm<sup>-2</sup> is considerably supported in some solutions by simultaneous ultrasound treatment as can be seen from Tab. II. Comparing the decontamination efficiency it seems suitable to use for decontamination of the secondary circuit material simultaneous treatment of ultrasound and HCOOH + EDTANa<sub>2</sub> solution for 0,5 - 2 hours at 50 °C with or without additives as CH<sub>2</sub>O, HCOONa, followed by residual contamination removal in the same solution or in H<sub>2</sub>SO<sub>4</sub> or H<sub>3</sub>PO<sub>4</sub> solutions with or without ultrasound treatment.

<u>Decontamination of Stainless Steel.</u> With regard to the characteristics of contaminated corrosion layer on stainless steel from the explosive mixture combustion system, the oxidizing and oxidizing-reductive agents such as:  $HNO_3$  /8 g.dm<sup>-3</sup>/,  $CH_2O$  /15 g.dm<sup>-3</sup>/ + CHOOH /10.34 g.dm<sup>-3</sup>/ + EDTANa<sub>2</sub> /3.36 g.dm<sup>-3</sup>/ were successful for decontamination prior to dismantling.

## 3.3. Electrochemical Decontamination

Experiments were carried out in standard arrangement. After electrolysis the electrodes were pulled out under voltage from the solution, rinsed with water, dried and activity was measured.

<u>Decontamination of Low-Alloy Steels.</u> During anodic oxidation and cathodic reduction in the electrolytes on the basis of  $HNO_3$ ,  $H_2SO_4$ ,  $H_3PO_4$  and oxalic acid, decontamination efficiency was observed depending on the process main parameters, as e. g. electrolyte concentration and temperature, current density and duration of electrolysis. Cathodic reduction in the 10 %  $H_2SO_4$  and in the mixture of 10 %  $H_2SO_4$  with 10 %  $H_3PO_4$  was successful in the aspect of decontamination. Regarding the properties, in the further experiments  $H_3PO_4$  was used as an electrolyte.

Increase of  $H_3PO_4$  conc was demonstrated in increased rate of corrosion layer dissolution and decontamination efficiency. DF = 258 was reached in 40 % /vol/  $H_3PO_4$  after 20 min. of electrolysis.

From the effect of electrolyte /20 %  $H_3PO_4$ / temperature, at current density i = 0.5 A.cm<sup>-2</sup>, on the time dependence of the sample residual activity  $/n_t$ / it follows that the most suitable electrolyte temperature was about 80 °C. Non-linear regression analysis of different curves  $n_t = f/t$ / showed that at given flow density and material, at 80 °C, independently on acid electrolyte type and its concentration, the following polynomial best fitted the curves

$$\log n_t = a - ct^2 + et^4 \qquad /1/$$

The shape of time dependence of residual activity was changing at lower temperature.

Results from the performed experiments showed <sup>1</sup> that it was possible by use of electrochemical decontamination to reduce the residual contamination level up to the level of specific activity, allowing unrestricted use of the material off the controlled zone. Necessary time of the electrochemical decontamination for given electrolyte depends mainly on the size of current density and thickness of the corrosion layer that is to be dissolved.

For the given material and current density of  $150 \text{ A.dm}^{-2}$  this time was 30 - 60 min. At the given current density, the DF value was increasing with time and for the time given it was increasing with the current density, but the dependence was not linear.

Decontamination of Stainless Steels. From the results obtained at anodic and cathodic polarization of samples as well as in regime with polarity reversation in one minute intervals was obvious that anodic oxidation and regime with polarity reversation are for decontamination rate more effective than regime of cathodic reduction. For samplex previously not decontaminated, background level at anodic oxidation or polarity reversation was reached after 5 - 6 min, for samples chemically decontaminated it was reached later, after minimum 14 min.

3.4. Release of materials from nuclear installation decommissioning to the environment

Decision on equipment or material reuse can be based on assessment of acceptable limits for radionuclides activity in material given either per weight and/or surface units.

There are the following basic aspects for assessment of the acceptable limits of radionuclide activity in material released for reuse /recirculation/:

- assessment of limit for contribution to the total dose to public /annual effective dose equivalent/ caused by this performance
- preparation of "scenarios" describing the most probable pathways for material release to the environment /recirculation procedure/
- calculation of maximum permitted activity of individual radionuclides in the released material, by which any limits of annual dose equivalent will not be exceeded at any stage of its use according to considered scenarios.

Regarding the potential increasing amount of released material from the decommissioning of nuclear installations it seems useful to regulate even the total amount of released material /total amount of radionuclides/. Assessment of limit collective dose equivalent from material release seems to be very suitable for this purpose.

In the first stage we considered the following scenario: transportation of material from NPP - interim storage at municipal site - melting in the furnace - use of remelted material in reinforcement of concrete foundations. The main attention was focused on evaluation of the risks to the public from the material on the site and remelting in furnace 1, 5. In spite of interim character of calculations, following conclusions can be formulated from the results obtained /on the relation to the annual limit of the effective dose equivalent of 10/uSv/:

- a/ In order to lower risk annual dose equivalent limit it seems suitable to eliminate interim off-site storage of material prior its processing.
- b/ Important risk is represented by slag material.
- c/ The material can be released without any risk at radionuclide activity of 1 - 10  $\text{Bq.g}^{-1}$  and at total value of surface contamination of 4  $\text{Bq.cm}^{-2}$ .
- d/ Value of collective dose equivalent will be decisive for assessment of the total amount of material that can be released to the environment.

## 4. CONCLUSIONS AND RECOMMENDATIONS

Paper gives short survey of works and results achieved on phase analysis of corrosion products on carbon steel from the secondary circuit and stainless steel from the explosive mixture combustion system in NPP A-1, together with results of research in the area of decontamination efficiency dependence of various procedures of chemical and electrochemical decontamination of given materials. It also shows approach to acceptable residual contamination assessment for metallic materials from NPP A-1 decommissioning and their introduction to the environment after smelting.

## RECOMMENDATION

It should continue the work on the research and development of cost effective decontamination methods as well as on the development of appropriate criteria for choice and application of the decontamination processes within decommissioning of nuclear power plants.

Models and related computer code for assessment of acceptable residual activity limits on materials released into environment from decommissioning of nuclear facilities should be refined. Experimental data on radionuclide behaviour during material smelting are required as an input data for computer code and for evaluation of all issues related to this attractive technology of radwaste volume reduction.

Elaboration of methods supporting various issues of nuclear power plants decommissioning cost estimates and that of cost-benefit-risk analysis application for these activities.

We can participate on the solution of these issues within following CRP.

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# DECONTAMINATION OF THE MAIN CIRCUITS OF THE G2 GAS-GRAPHITE REACTOR

R. LURIE
Institut de protection et de sûreté nucléaire,
Commissariat à l'énergie atomique,
Bagnols-sur-Cèze, France

#### Abstract

The decommissioning programme for the gas-cooled graphite reactor G2 at Marcoule, France includes decontamination of the facility in such a manner that the solutions are as small as possible and the process is carried out remotely. Low contamination levels and large amounts of construction materials led to the selection of decontamination procedures which would permit the reuse or recycle of the construction material. Two approaches were considered, the first consisting of in-situ decontamination, the second consisting of dismantling the materials and subsequent decontamination in a central decontamination facility. Laboratory testing included application of various acids and mixtures of gels and foams, high pressure jet, and electrochemical decontamination methods. With acidic solutions decontamination factors over 100 were obtained in less than two hours; with gels and foams similar results could be reached, the latter method proving to be less agressive and resulting in removal of lower amounts of contaminated material. Full scale tests were performed in 1986 and 1987 with 30 tons of steel (300  $m^2$ ) using the gel technique which during laboratory testing offered most promising results. The results confirmed that the gel technique can successfully be used, where contaminated surface is accessible for gel spraying. In order to adapt the reagent to the type of base material and to the contaminant, thorough laboratory tests have to be carried out.

## a - INTRODUCTION

The G2 reactor located on the Marcoule establishment started operation in 1958 and reached full power : 260 Mwth and 40 Mwe in July 1959 has been devinitively shut down in February 1980 after 21 years of operation.

## Reactor core

The core is composed of a pile of 15,000 graphite bars. Its section is quite octogonal of 9.5 m of axis and 9 m long. This pile is crossed by 1200 horizontal channels for the fuel elements and 51 vertical channels for the control rods. This active part is surrounded by a graphite reflector of about 0.8 m thick. To lower the heat to the concrete, the graphite is entirely shielded by 12 cm thick steel plates and by an insulation. This block is enclosed in a prestressed concrete vessel in form of an horizontal cylinder of 14 m internal diameter and 18 m long closed at each end by an hemispherical dome. The concrete thickness of the cylinder and the domes is 3 m. In order to withstand the 15 bars in service pressure and the 30 bars test pressure the vessel is prestressed by 161 cables tightened each one at 1200 tons.

#### Circuits

Numerous and complex carbon dioxide circuits are used : to cool the reactor during normal operation or incidental situations, to maintain the concrete shielding, the thermal screen and the internals at a reasonable temperature. They involve 1700 m of pipes, 1000 m of which is over 1 m in diameter.

#### Steam generators

G2 is equiped with four steam generators of 3.5 m in diameter and 32 m long disposed vertically outside the building. They worked in parallel and include an economiser, HP and LP vaporizer and HP overheater composed of finned tubes. Each one weighs 300 T.

## b - OBJECTIVES OF THE WORK

The decommissioning programm of the facility has been divided in two main steps :

- Dismantling all equipments and circuits outside the reactor vessel spread over a period of four years.

- Dismantling the reactor core and vessel.

Each step necessitate some research and development work to choose the techniques and to design the equipments. Among these we can quote the followings for the first step :

- Décontamination process producing the smallest volume of effluents.
- Automatic implementation of the decontamination process.
- Automatic cutting of large pipes and cylindrical vessels.
- Activity measurement of very low activities on large amount of metallic waste.

The first item has been choosen to be reported to the CRP.

#### Detailed objective

Let aside a few hot spots, due to debris activated into the core and spread in the circuits, these equipments have a very low activity. The mean value is  $10-3 \mu \text{Ci/cm2}$  (33 Bq/cm2) due to activation products such as Co 60 and fission products such as Cs137. This low contamination and the large amount of steels

1100 tons for the circuits
1200 tons for the steam generators
200 tons for the ancillary equipment.

lead to the choice of decontaminating down to a level low enough to allow reuse or resale as scrap iron.

The aim of the research program is to prepare the industrial decontamination step. It will involve :

- a review of the decontamination technics which can be used on the circuits,
- the choice among these technics of the best process taking in account the base metal (mild steel) and the type of contamination,
- the test of these technics on samples taken out from the circuits,
- the final choice and testing on a first part of the circuit.

Two implementation ways have been investigated : either decontaminate the pipes in situ, this will help the further job of cutting and reduce the man-rem exposure but gathering the effluents will be more difficult ; or cut the pipes and treat them in a central decontamination work-shop, which implies more carefull cutting to avoid spreading of contamination.

According to the diameter of the pipes and their location (horizontal or vertical), in situ decontamination needs the development of special machines.

The implementation of a technic can be easy with an horizontal large pipe for example and very difficult for vertical or smaller one. Even if the choice is to decontaminate in situ it will be necessary to have an additional decontamination shop with another process to treat specific parts such as valves, bends with flanges ...

Any decontamination technic must also be consistent with the effluent treatment station available on site.

## c - RESULTS AND DISCUSSION

1 - FIRST STEP : Test of different decontamination process.

Samples of 160 mm x 160 mm have been cut out from different parts of the primary circuit, some of them coming from the hot leg before the steam generator, the others from the cold leg after the steam generator.

The analyses showed activities due to  $\beta$  and  $\tau$  emiters ranging from 10-4 $\mu$  Ci/cm2 to 2x10-3 $\mu$ Ci/cm2 (3,3 to 67 Bq/cm2), 90 % of the activity coming from Co60 and Cs 137.

In order to decontaminate down to unrestricted release (even if actually in France there are not fixed values for unrestricted release) a decontamination factor larger than 100 must be obtained to reach a surface activity lower than 3 10-5  $\mu$  Ci/cm2 (1 Bq/cm2) and mass activities lower than 1,5 10-5  $\mu$  Ci/g (0,5 Bq/g).

#### DECONTAMINATION PROCESSES

#### A - Soaking in chemical baths

Different types of acids and acids mixtures have been tested such as

- hydrochloric, sulfuric + nitric, nitric, hydrochloric + nitric, fluoridric, whith times ranging up to two hours. The decontamination factors and the layers removed have then been measured.

The results showed that the hot leg was easy to decontaminate, DF>150 with nitric + sulfuric acid, the decontamination occurs in less than 1 hour with a removed layer of  $\sim 130 \ \text{Mm}$ .

Hydrochloric + nitric acid is also efficient in less tan two hours with a removed layer of 20 to 40  $\mu$  m.

The cold leg decontamination was more difficult. Sulfuric + nitric acid was efficient in 1 hour with DF  $\sim$  100, and 120  $\mu$  m erosion. Hydrochloric + nitric acid was also efficient with a very small erosion.

#### B - Gels and foams

These technics are used with the same chemical products as for soaking. For gels, the mixture is spread on the surface let one hour then rinsed. For foams, the mixture is send on the surface by air compressed system then pumped and recycled.

The gels were made from silica gels with fluoridric + nitric, sulfuric + nitric, hydrochloric + nitric acids. The amount spread on the surface is about 200 g/m2.

As for soaking the hot leg is easier to decontaminate. The most efficient are hydrochloric or hydrochloric + nitric gels with length of time of three hours. The erosion is in the range of 30  $\mu$ m for the hot leg and 15  $\mu$ m for the cold leg with DF over 150.

Compared with soaking these methods are less agressive need a longer time but are producing a smaller amount of reagents.

## C - High pressure jets

The tests were performed on 20 mm x 80 mm samples in a special cell, different parameters have been checked such as :

- pressure : 150, 450 and 600 bars,
- flow : up to 20 m3/h,
- distance between nozzle and sample : 2,5 5 10 cm.

The samples were first degreased.

It was noticed that the cold leg was easy to decontaminate when the pressure was rised up to 450 bars. But a 600 bars pressure with a nozzle to sample distance of 2,5 cm was not able to decontaminate the hot leg.

#### D - Two steps process

In order to reduce the volume of effluents, it seemed interesting to test a two step process : first to spray a gel and then to rinse with high pressure jet.

The cold leg did not bring problems and is decontaminated with a small flow  $1 \text{ m}_3/\text{h}$  and a low pressure (150 bars).

The hot leg decontamination necessitates 600 bars and a 5 m3/h flow. To improve this hot leg decontamination, it was decided to alternate gel sprays and high pressure rinsing.

With 150 bars and 2 m3/h two gel sprays were necessary. With 600 bars and two gel sprays only 1 m3/h was necessary to achieve decontamination.

From these results it appeared that hot leg contamination is linked to surface corrosion or chemical actions whereas the cold leg one is linked to plate out and adsorption.

#### E - Electrochemical decontamination

Electrochemical decontamination was tested with a movable electrode, composed of stainless steel, glass fibers and felt. The electrolyte is send through the felt, flows on the sample and then is recycled. The following electrolytes have been tested : sulfuric acid, phosphoric acid, sulfuric + phosphoric acids, sodium hydroxyde with a current density of 50 A/dm2.

It was noticed that the cold leg was easy to decontaminate with sulfuric and phosphoric acids.

Sodium hydroxyde acts very quickly without achieving a high DF.

It was then suggested to start with sodium hydroxyde and achieve decontamination with sulfuric or phosphoric acid.

#### CONCLUSION OF THIS FIRST STEP

All these tests proved that the following processes were efficient

for the decontamination of G2 pipings :

- hydrochloric gel followed by low or high pressure rinsing,

- high pressure jets (450 bars, 5 m3/h),

- soaking into hydrochloric, hydrochloric + nitric or sulfuric + nitric acids,

- electrolytic decontamination with sulfuric acid.

Among these possibilities the process producing the smallest amount of effluents was chosen. A two step gel process : a basic gel of sodium hydroxide and an acid gel of phosphoric + sulfuric acids followed by high pressure rinsing. 2 - SECOND STEP : Test on different samples.

Large samples of  $500 \times 500$  mm were cut on different parts of the circuit with different cutting technics, in order to have a better knoledge of the efficiency, the volume of effluents produced and to see if the contaminants were trapped into the melted zone (for thermal cuttings) and less easily removed by the decontamination process.

Location of samples :

-	Hot leg east	area	3 3	samples samples	with with	plasma torch grinder.
-	Hot leg west	area	2 2	samples samples	with with	oxyacetylene grinder.
-	Cold leg		4 3 3	samples samples samples	with with with	grinder oxyacetylene plasma torch.

#### Conclusion and results

The different sequences were as follows :

spray of basic gel,
rinsing,
spray of acid gel,
rinsing,

or : two sprays of acid gel followed by rinsing.

An amount of 150 to 200 g/m2 of gel were sprayed on the surface and let 40 minutes to operate. The amount of rinsing water was between 20 and 30 1/m2.

- Cold leg :

The contamination of the samples was ranging between 25 and 40 Bq/cm2. After decontamination by a two step process basic + acid or acid + acid gel the remaining contamination was below 0.3 Bq/cm2. No significant difference was observed between the different cutting processes.

- Hot leg :

The initial contamination was ranging from 90 to 120 Bq/cm2 and needed a tree steps process to come down to a contamination below 0.3 Bq/cm2.

The basic gel had only a small decontamination factor but degrease the surface and improved the decontamination factor of the next acid step.

3 - FULL SIZE TESTS ON THE CIRCUIT

Full size tests on the circuit started in sept. 1986 up to sept. 1987. Spraying the gel and rinsing was done by hand. Thus only a part of the larger pipes (1,6 diameter) was treated.

The gobal results coming from the decontamination of  $300 \text{ m}^2$  of pipes (30 tons of steels) were as follows :

- initial mean activity : 5 10-3 Ci/cm2 (200 Bq/cm2)

- final mean activity : 10-5 Ci/cm2 ( 2 Bq/cm2)

- decontamination factor : 100

- mean mass activity : 200 Bq/Kg

- liquid effluents : during the implementation of the process improvements were effected and the volume of effluents fall from  $30 \ 1/m^2$  down to  $12 \ 1/m^2$ .

#### d - CONCLUSIONS AND RECOMMENDATIONS

Decommissioning a nucleal facility needs generaly a decontamination of the main components. Before cutting or dismantling in order to lower the radiation field and the integrated radiation dose to the workers, and sometimes after cutting in order to keep the waste inside the limits fixed by the repository sites or to resale the materials as scraps.

The gel process which has been developped with full success to the decontamination of large pipes can be used for any component or item, when the contaminated surface is accessible for gel spraying.

Laboratory tests must be done prior to any decontamination in order to adapt the reagents to the type of base material and to the contaminants.

#### e - FUTURE WORK PLANNED

An automatic machine has been designed to spray the gels and the rinsing water inside the pipe, and will be used during 1988. This machine is necessary for smaller pipes where man work cannot be done.

Another decontamination problem is the washing of large vessels housing equipments such as heat exchangers or steam generators.

Filling the vessel with decontaminating solutions will give rise to a very large volume of effluents increasing the overall cost of decontamination.

Two process are actually studied decontamination by foam or mist including the reagents.

# DECONTAMINATION OF NUCLEAR FACILITIES BY ELECTROCHEMICAL METHODS

O. PAVLIK Institute of Isotopes, Hungarian Academy of Sciences, Budapest, Hungary

## Abstract

Work was reported on electro-chemical decontamination methods for maintenance of the Paks nuclear power plants in Hungary. For in-situ decontamination of large components, for example main circulating pump, gate valves, steam generator collectors, methods using remotely operated movable electrodes were implemented successfully. For the main circulating pump, the electrolyte contained phosphoric, sulfuric and oxalic acids. The concentration of the radioactive isotopes and corrosion products was measured in the spent electrolyte and in the rinsing solution. About 150 l liquid radioactive waste (together with rinsing water) was produced during the decontamination procedure. The duration of the decontamination was 6-8 hours. The procedure was carried out by 3-4 workers, their collective dose was 2-3 mSv. The decontamination was followed by chemical and radiometric analyses.

## INTRODUCTION

In Hungary the Paks Nuclear Power Plant is operating since the end of the year 1982. It has pressurized water reactors of VVER-440 type. Four units of four hundred forty MWs are in operation this year. The long-term plans include the installation of two one-thousand MW units, also pressurized water types. Systematic and efficient maintenance and inspection are essential conditions for the safe operation of nuclear power plants, however, maintenance and supervision are dangerous because of the personnel exposure hazard. Radiation doses can be reduced by the decontamination of the primary circuit. The exposure of the decontamination staff can be reduced using remotely operated devices. The primary circuit of the VVER-440 type reactor is made of stainless steel. Therefore agressive physicochemical processes can be used thus increasing the efficiency of decontamination.

In the last three years the development of electrochemical decontamination was carried out as a part of a coordinated research program of the International Atomic Energy Agency on the development of decommissioning and decontamination of nuclear facilities.

## OBJECTIVES OF THE WORK

Contamination in water-cooled reactors is essentially governed by the behaviour of iron, chromium, nickel and cobalt ions in the coolant. Since cations can easily be built into metallic surface, the radioactive isotopes, fission and corrosion products, will be part of the structural materials.

On metallic surfaces decontamination can only be achived by the removal of the upper layer of the metallic surface containing the contaminants. Successful decontamination removes all contamination,

without excessive corrosion of the substrate. In many cases the decontamination of the complete primary circuit is not necessary, • the partial - component or subsystem - decontamination my be sufficient, while it is cheaper, quicker and produces less liquid wastes.

The electrochemical decontamination is an application of electropolishing of metal surfaces. The oxid-layers can be removed from the surface of metal, using a direct electric current between the workpiece and a cathode in an acidic electrolyte. Generally the workpiece to be decontaminated is immersed in an electrolyte tank as an anode in an electrolitic cell. When contaminated large-sized equipments should be sunk into a

tank for electropolishing a big volume of liquid radioactive waste will be produced.

Some years ago in -situ decontamination process was developed to decontaminate large or inmobile equipments, that cannot be immersed in a tank. Electrochemical decontamination using movable cathod does not use big volume of electrolyte and does not produce big quantitites of liquid wastes. The method can be seen in Fig.1.



Fig. 1. Decontamination my movable cathode

High personnel exposure could be taken account if the cathode was moved by an operator (manhandling). The exposure of the decontamination staff can be reduced using remotely operated equipments.

## RESULTS AND DISCUSSION

Laboratory experiments

Corrosion and decontamination tests were carried out by a laboratory scale movable cathode to optimize the electrolyte composition.

Electric power supply:	battery charger
Potential:	6-30 V. D. C
Current density	25 A. $dm^{-2}$
Contact time: Ó	2x2Os (Deco tests)
Contact time:	10 min (Corrosion tests)

Comparing the ratio of DF and corrosion rate the electrolyte containing citric acid 600 g/l, sulfuric acid 150 g/l proved to be the best.

Components /g/1/	Corrosion rate /g.cm <sup>-2</sup> .min <sup>-1</sup> /	DF / /	DF/corr.r.
H <sub>3</sub> PO <sub>4</sub> 40	2,3.10 <sup>-</sup> 4	174	7,6.10 <sup>5</sup>
H <sub>2</sub> SO <sub>4</sub> 30			
H <sub>2</sub> C <sub>2</sub> O <sub>4</sub> 20			
н <sub>з</sub> РО <sub>4</sub> 75	_		_
H <sub>2</sub> SO <sub>4</sub> 5	3,9.10 <sup>-5</sup>	162	4,2.10 <sup>6</sup>
Cr 0 <sub>3</sub> 6			
H <sub>3</sub> PO <sub>4</sub> 1600	4,9.10 <sup>-5</sup>	107	2,2.10 <sup>6</sup>
C <sub>2</sub> H <sub>5</sub> -OH 80			
H <sub>2</sub> SO <sub>4</sub> 150	1,4.10 <sup>-5</sup>	90	<b>6</b> ,4.10 <sup>6</sup>
citric-acid 600			
citric-acid 20	1,2.10 <sup>-5</sup>	28	2,3.10 <sup>6</sup>
<sup>H</sup> 2 <sup>C</sup> 2 <sup>O</sup> 4 20			

Table I.: Corrosion rate and DF versus electrolyte composition

The rate of electrolyte feed was also investigated.

The electrolyte content of the felt increases with increasing rate of electrolyte feed. Meanwhile the current density also increases up to the full saturation of the felt. After safuration the current density will be constant. On the other hand the felt temperature decreases with increasing electrolyte rate, since the flowing electrolyte has a significant cooling effect (Fig. 2.)



Fig. 2.: Current density and felt temperature versus electrolyte rate

## REMOTELY OPERATED EQUIPMENTS FOR DECONTAMINATION

Three electrochemical, remotely operated decontamination equipments were developed for the Paks NPP.

The first one was constructed for the decontamination of the main circulating pump case. Figure 3. shows the principle of operation. The equipment consists of the support and the traversing mechanism of the movable decontamination head and the supply unit.



Fig. 3. Electrochemical decontamination equipment for the main circulating pump case

The supply unit is carried by a small truck and can be craned. A feed pump provides the accurate dosage of the electrolyte for the movable decontamination head. The traversing mechanism is equipped with a multi-jointed arm moved by pneumatic cilinders on the inner surface of the pump case. The central bearing axle can be rotated by a rotative gear right and left 360 degs. The multi-jointed arm pulls away the movable decontamination head from the decontaminated surface after one turn-around, moves it 45 mm down and presses on the surface again. The compressive force can be regulated between 40-400 N. The position of the movable decontamination head always follows the curving of the pump case and the felt fits close to the surface. The uniform contact between the felt and the surface is ensured by pressure springs. The movable decontamination head is equipped with mechanical sensing device to detect if the head reaches the pipe connection of the primary circuit.

## Results Obtained From Plant Decontamination

Effective decontamination procedures were conducted at the Paks Nuclear Power Station during the shutdown periods by the remote controlled electrochemical decontamination equipment for the main circulating pump case.

The electrolyte contained phosphoric, sulfuric and oxalic acids. Before decontamination the inlet nozzle for reactor coolant was plugged in such a way that the electrolyte could not enter the connecting pipe. The spent electrolyte accumulated on the bottom of the treated pump case and was drained into the radioactive sewage. After electrochemical decontamination the inner surface of the pump case was rinsed with a 12 g/l boric acid solution. The concentration of the radioactive isotopes and the corrosion products was measured in the spent electrolyte and in the rinsing solution too. About 150 l liquid radioactive waste (together with rinsing water) was produced during the decontamination procedure. The duration of the decontamination was 6-8 hours. The procedure was carried out by 3-4 workers, their collective dose was 2-3 mSv. The decontamination was followed by chemical and radiometric analyses.

Table II. shows the removed activity, the removed corrosion product and the decontamination factor (DF).

Date	and	location	DF	removed activity (MBq)	removed corrosion products (g metal)
1985.	<u>I.</u> 4. 5.	unit MCP MCP	27 1800	1500	
	<u>2.</u> 6. 3.	unit MCP MCP	490 11	1850 4200	52 42
1986.	$\frac{1}{1}$	unit MCP MCP	110	3800 2200	76 47
	$\frac{2}{1}.$ 4.	unit MCP MCP	46 52	2600 2200	54 70

The second remote operated decontamination equipment, showed in Fig. 4. was constructed for the <u>collectors of the steam generator</u>. The equipment consists of the support, the traversing mechanism of the decontamination head and the control unit including electrolyte and voltage supply.

The whole mechanism can be craned into the collector and the control unit inflates the inner tube of the plug. The tube is fixed by friction and seals between the pipewall and the plug. The decontamination head is fitted swinging on the axle of the pneumatic cylinder which presses the head on the surface to be decontaminated. The head moves down at a speed of 20 mm/min. The whole surface of the collector can be treated in twelve hours. In the bottom position the control unit stops rotating, moving and feeding the electrolyte too.

The control unit is carried by a small car and can be craned. A piston type pump feeds the electrolyte from a tank to the movable decontamination head. The spent and contaminated electrolyte accumulates on the bottom of the treated collector and is removed by a plunger pump. The bottom of the collector is closed by a plug.



Fig. 4. Decontamination equipment for the collectors of the steam generator

The guide rail of the support and traversing mechanism of the decontamination head is fitted with bearings and is free-wheeling. The lower bearing socket is built in the plug at the bottom of the collector. The upper bearing socket is built in a bridge placed on the upper plane of the collector. The turning is performed by a gear fixed on the bridge. The decontamination head returns after one turning to avoid any damage to the vertical rails. The speed of the turning is 0.4 turn/min during continuous moving. The cathode can also be moved step by step to reduce the damaging of the felt. The third remotely controlled electrochemical decontamination equipment was constructed to treat the inner surface of the main gate valve case. /Fig. 5./

The decontamination of the gate valve case can be conducted only after removing the cut - off slide - valve. First the two pipe-ends must be plugged to prevent from the electrolyte or other impurities getting in . The feeding and removing of the electrolyte is performed as in the case of the two electrochemical decontamination processes mentioned before.

The support and the traversing mechanism is craned on the flange of the gate valve case. The decontamination equipment has two movable decontamination heads. One of them /head II./ moving around the vertical symmetry axis of the case treats the upper part of the case. The central bearing axle is only rotated right and left 180 degs to avoid damaging the wires and conduits. The lower spherical part of the case is treated by head II. moving round the horizontal axis of the centre of the pipe ends.



Fig. 5. Decontamination equipment for the main gate valve case

The spherical surface of the case and the outer surface of the pipe ends is simultaneously treated by cathode I. The moving of both movable heads is complicated because they have to go round the two guide rails of the cut-off slide valve as well. About 80 percent of the contaminated surface can be treated in such a way. The guide rails, the inner and the sealing surface of the pipe-ends can only be decontaminated by manually moved cathods.

## CONCLUSIONS AND RECOMMENDATIONS

Influence of the <u>electrolyte composition</u>. The points of view of the electrolyte selection: - high decontamination factor

- HIGH GECONLAMINATION IACT
- low corrosion rate
- does not develop agressive gas
- high electric conductivity
- relatively low electrolyte feed rate end low concentration of chemicals /does not produce big quantity of radioactive waste/

We use the H<sub>3</sub>PO<sub>4</sub> 40 g/l, + H<sub>2</sub>SO<sub>4</sub> 30g/l, + H<sub>2</sub>C<sub>2</sub>O<sub>4</sub> 20 g/l, or citric acid 20 g/l + H<sub>2</sub>C<sub>2</sub>O<sub>4</sub> 20 g/l electrolytes. This was the result of a compromise.

The electrolyte feed rate is  $20-40 \text{ l.h}^{-1}.\text{dm}^{-2}$ .

The remotely controlled electrochemical decontamination method using movable cathode has the following advantages:

- high decontamination factor (20-500)
- short application time
- produces smooth surface which reduces recontamination
- low collective dose
- low volume of liquid radioactive waste

Disadvantages:

- The movable cathode cannot clean the whole surface to be decontaminated, if the surface has a complicated geometry or profile
- The above-mentioned remotely operated movable heads can decontaminate only equipments with well-defined dimensions.

## FUTURE WORK PLANNED

Our intention for the next years is:

- to increase the decontamination factor with new composition of electrolyte. The damage of the electrolyte absorbing material (felt) can be avoided by the application of teflon felt, therefore more aggressive electrolyte might be used;
- to extend the application of the decontamination equipments. Now the movable heads can decontaminate only surfaces with well-defined dimensions. We would like to develop devices suitable for the decontamination of equipments with different dimensions.
- to construct movable head for material testing (ultrasonic) of the inner walls of different equipments of the primary circuit.

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# **RESEARCH AND DEVELOPMENT OF LWR SYSTEM DECONTAMINATION: MECHANOCHEMICAL AND REDOX DECONTAMINATION METHODS**

M. KAWASAKI, E. TACHIKAWA, H. YASUNAKA, T. SUWA, T. GORAI Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken, Japan

#### Abstract

In order to satisfy requirements for efficient crud removal from contaminated surfaces and easy subsequent treatment of spent decontamination solutions, two methods were developed, tested and applied in the decommissioning of the Japan Power Demonstration Reactor (JPDR) plant. The specimens of pipes and tubes of the primary system were thoroughly examined with use of chemical analysis, X-ray, activity measurement and electron microscope. High content of Cr in the crud was ascribed to poor quality control of cooling water in the early stage of plant operation. Since decontamination with common chemical reagents was not effective, further tests were modified in two ways. The first method consisted of a mechanico-chemical procedure, in the second various redox decontamination reagents were used. Subsequently, laboratory testing was replaced by large scale tests performed in decontamination loops, designed and constructed for this purpose. In decontamination of samples from NPPs by various decontamination reagents, it was observed that the redox method gave a satisfactory decontamination factor, regardless of the Cr-content, and could be successfully used as a decontamination method for pre- and/or post-dismantling of a reactor.

1. Introduction

The Japan Atomic Energy Research Institute(JAERI) started the "Reactor Decommissioning Technology Development Programme" in 1981 under contract with the Science and Technology Agency, one of the Goverment organizations, to provide information for development of technical guidelines for the decommissioning of nuclear power plants. The programme consisted of two phases. The phase 1 was the development stage of various techniques necessary for reactor dismantling, including estimation of radioactive inventory, decontamination, disassembly, waste management, radiation control, and system engineering, and terminated in 1986, followed by the phase 2 programme of actual dismantling of the Japan Power Demonstration Reactor(JPDR) plant, aimingto demonstrate dismantling technology developed and to acquire nuclear power plant dismantling experience.

Decontamination prior to reactor dismantling must satisfy several requirements, which differ somewhat from those for decontaminatin of nuclear facilities under operation. At JAERI efforts have been paid to develop decontamination for dismantling, as part of the JPDR-decommissioning project activity, with emphasis on that the method provides a high DF for any kinds of CRUD, and that the waste solution resulting from decontamination can easy be treated. Two decontamination methods have been developed, mechanochemical method and redox method, and successfully applied for the part of the primary system of JPDR. The former is based on the additive effects obtained by combining mechanical cleaning and chemical one, while the latter on the strong oxidizing power of Ce(IV) and its regeneration.

2. Basic Experiments for Decontamination

2.1. Characterization of CRUD

Specimens of pipes and tubes, taken from various parts of the primary system of the JPDR were subjected to chemical analysis, activity measurement, X-ray diffraction analysis, and electron microscopic measurements. The amount of CRUD and its activity, deposited on the pipe surfaces, are in the range of 0.3 - 1.0 mg/cm<sup>2</sup> and  $(3 - 6)10^{-2}$  uCi/cm<sup>2</sup>, respectively. The Crcontent amounts roughly to 22% at the reactor water cleaning line and 13% at the recirculation line, similar to the CRUD in a PWR rather than in a BWR. This is possibly due to rather poor guality control of the primary water in the early stage of JPDR operation.

2.2. Chemical decontaminatin of sample specimens

Decontamination experiments were conducted under static and dynamic conditions with various decontamination methods.

2.2.1. Decontamination with existing chemical reagents

The decontamination factor(DF) was measured as a function of reagent concentration, temperature, time, and flow rate. Although the DF varies depending upon the decontamination conditions, the highest value attained by such a reducing chemical reagents as LND-101A(Can Decon), NS-1, GE-dilute, ED-40, and KD-203 was 4. This simply was ascribed to the high Cr content of the CRUD. A high DF was only obtained when the chemical decontamiation was preceded by a preoxidation treatment(NP-process).

2.2.2. Mechanochemical decontamination

When small grains of abrasives are suspended in a flowing chemical cleaning solution (or in water), the grains hit or rub the inner surfaces of the pipes, leading to mechanical release of the CRUD. Thus, both mechanical release and chemical dissolution of the CRUD can be anticipated.

Throughout the basic experiments, the following conditions are chosen for the planned system decontamination,

o Abrasive: Boron carbide( $B_4C$ ) of ca. 0.5 mm in diameter.

o Fluid: Abrasive is suspended in pure water up to 20 wt%.

o Decontamination: Fluid is circulated at a flow rate of ca. 4.8m/sec at room temperature for 48 hrs.

2.2.3. Redox decontamination process

Since the oxidation of the CRUD is essentially required in order to attain a high DF, redox reagents were examined under various conditions. The results obtained, together with thermodynamic considerations, lead to  $Ce(IV)-H_2SO_4(SC \text{ solution})$  at the most promising condidate as a chemical reagent for reactor

dismantling. In the solution, the CRUD dissolves according to the equation,

 $CRUD + Ce(IV) = Fe^{+2} + Cr^{+6} + i^{+2} + Ce(III)$ The resulting Ce(III) can be reoxidized to Ce(IV) by an electrochemical method at any desired rate. After decontamination the waste solution is treated by electrodialysis, and then by a deep bed ion-exchange column.

## 3.System Decontamination

## 3.1. Construction of system decontamination loops

Applicability of the newly developed decontaminatin methods can be examined through decontamination of a part of the primary system of the JPDR. The primary line was refashioned to provide 4 system decontamination loops: Two of them from the reactor water cleaning line and the others from the reactor water recirculation line. By decontaminating the each loop with different methods, can-Decon(LND-101), modified Dow Chemical(NP/NS-1), mechano-chemical, and redox methods, the comparative results can be obtained.

## 3.2. Decontamination by Can-Decon process

Figure 1 shows a schematic diagram of the loop subjected to the Can-Decon method. In the figure, the bold lines are the part of the reactor water cleaning line and the others the additional lines to complete the decontamination loop. The characteristics of the loop and the decontamination conditions are also included. Total amounts of activity and metals recovered throughout the decontamination were 1770 uCi and 1167g, respectively. The DF obtained varied from place to place, ranging from 3 to 90. High DF was found only in and near the heat exchangers, DF at the pipe lines was limited from 3 to 11. This was ascribed to a large difference in chemical composition of the CRUD: Near the heat exchangers Cu was the major element of the CRUD, reaching to as high as 90%.

## 3.3. Decontamination by modified Dow Chemical process

Schematic diagram of the second loop is shown in Fig.2. The Dow Chemical process was preceded by the NP-process. In order to avoid production of extra amounts of waste solutions by this preoxidation process, the solution resulting from the preoxidation process was subjected for a reverse osmosis treatment. The NP-1 reagent was added to the processed water. After the subsequent decontamination, the waste solution was again processed with the reverse osmotic treatment.

In Table 1 summarized are the results of decontamination. Activity recovered by this NP/NS-1 process was roughly 6.5 mCi. The DF along the line differed ranging from 90 to 740. Average corrosion rate, measured using coupon specimens in the line during the decontamination, was between 200 and 300 mdd for stainless steel.



opectrication of acconcamination syste	Specification	of	decontamination	svste
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System area	Surface area	Volume
Decon. area Additional area	16.8 m <sup>2</sup> 20.9 m <sup>2</sup>	$0.08 m^3$ 0.64 m <sup>3</sup>
Total	37.7 m <sup>2</sup>	0.72 m <sup>3</sup>

FIG.1. Decontamination test by the CAN-DECON process.



Decontamination test loop diagram (recirculation line)

Specifi	icatio	n of decontamir	nation system
System	area	Inner surface area	Volume in water
Decon	area	9 21 m <sup>2</sup>	1 09 m <sup>3</sup>

Decon. area	9.21 m <sup>2</sup>	1.09 m <sup>3</sup>
Additional area	30.27 m <sup>2</sup>	1.20 m <sup>3</sup>
Total	39.48 m <sup>2</sup>	2.29 m <sup>3</sup>

	<ol> <li>Preoxidation: reagent and its concentration</li> </ol>	KMnO <sub>4</sub> lg/l HNO <sub>3</sub> 5g/l	
-	2) Reduction deco	ontamination	1
	Decontamination: reagent and its concentration	NS-1 0.7wt	- 8
	Duration	24hr	
	Temperature	120 C	
_	Velocity	0.3m/sec	

Test condition

FIG.2. Decontamination test by the modified NS-1 process.

Decontamination process	Modified NS-1 process (Recirculation system)		
Decontamination effect	Place measured		DF
	Upper side of horizontal line at pump discharge		205 - 350
	Bottom side of horizontal line at pump discharge		95 - 121
	Side part of horizontal line at pump discharge		91 - 108
	Vertical line at pump		456 - 738
Activity removed	-	Ci	
Metal removed	Total	360 gr	
	Fe	150 gr	:
	Ni	150 gr	
	Cr	130 gi	:
	Cu	_	
	Со	10 gi	: :

3.4. Mechanochemical decontamination process

In Fig.3 is shown the flow diagram in the mechano-chemical decontaminatin loop. The experimental procedures is in Fig.4 During the decontamination, sample specimens were taken out at appropriate intervals for activity- and weight-measurements. At 12 hours decontamination the abrasives were replaced with fresh one, followed by the additional decontamination.

3.4.1. Results:

Changes in activity and weight of sample specimens during the decontamination are graphically summarised in Fig. 5. The main features are,

- o At 12 hours decontamination, more than 97% of the activity was removed from the sample specimens.
- o By continuing the decontamination up to 18 hours after replacement of the abrasive, the removed fraction reached to 99%.
- o Increase of the flow rate from 4.8m/sec to 6.7 m/sec positively affected the detached fraction, although the extent was not significant.
- o At 35 hours decontamination, the removed fraction was ca.99.9%, further removal seemed to need far prolonged decontamination hours.

As for the weight decrease of the samples during the decontamination,

o It increased more or less linearly with decontamination time, but the rate depended upon the flow rate and increased with increasing flow rate, as was expected.

	Surface area(m <sup>2</sup> )	Inner volume(1)
Objected line to be cleaned	1.8	18
Supplemental line	6.7	92
Total	8.5	110





FIG.3. Schematic diagram of the system decontamination loop (by the mechanochemical method).



FIG.4. Mechanochemical decontamination procedures.



FIG.5. Activity and weight loss during decontamination.

The ratio of the activity removed to the weight-decrease during the decontamination continuously decreased with time, as seen in Fig.5. This might be understood to show that during the decontamination rather uniform surface removal occurs, while the activity becomes sparse as the removal proceeds.

## 3.4.2. Treatment of waste suspended solution:

After decontamination the solution was passed through the abrasive recovery system, where the gralular abrasive was collected, and then through the cartridge filter(10um) at a flow rate of 20 m<sup>3</sup>/hr. The final purification was by ultrafiltration. In the present case the activity and the solid concentration of the solution was reduced from  $6\times10^{-5}$ uCi/ml and 1000 ppm to  $10^{-6}$ uCi/ml and 13 ppm, respectively, and no further treatment was needed.

3.4.3. Summary:

The results are summaried as follows,

- o Activity removed: 540uCi
- o Decontamination factor: 200 1650
- o Metal removed: 2150g (including crud's, metal chips, and crushed abrasives)
- o Abrasives recovered: 37.6 kg (Recovery ratio: ca.80%)

oResulted solid wastes: mainly consisted from cartridge

filter, ultrafiltering module and abrasive and contained in a waste-drum(inner volume : 50 liter)

The present method has the following advantages:

oA high DF can be obtained in relatively short decontamination time, independent of the chemical composition of crud's.

o The resulting waste solution can be easily treated.

o Decontamination efficiency is almost independent of temperature 0 and can be carried out under the atmospheric conditions.

- o Decontamination procedure is relatively simple.
- The existing disadvantages are,
- o A positive counterplane is needed to eliminate the trapping of abrasives during decontamination, particularly when the line to be decontaminated has a complexed structure.
- o A relatively large capacity of the circulation pump is needed to obtain a flow rate sufficient to circulate the abrasives.

3.5. Redox decontamination process Main features of the SC method are,

- o Dissolution of crud proceeds by the oxidation of Fe(II) to Fe(III), and Cr(III) to Cr(VI). Similarly, stainless steel also corrodes by the reactions of Fe to Fe(III), Ni to Ni(II), and Cr to Cr(VI). Thus the amount of Ce(IV) consumed is equal to the sum of these oxidations.
- o Dissolution rate is proportional to the Ce(IV) concentration.
- o Dependence of the dissolution on the acid concentration is not significant above 0.25M  $\rm H_2SO_4.$
- o Both temperature and acid-concentration affect the dissolution rate of crud.
- o Addition of Ce(III) ion to the SC solution lowers the oxidation potential of the solution, leading to the decrease of the corrosion rate of steels.
- During the decontamination, the Ce(IV) concentration can be maintained at a desired value by electrochemically regenerating Ce(IV) from Ce(III).

## 3.5.1 Decontamination loop:

One of the system decontamination loops, constructed from the reactor coolant recirculation line, was subjected for the decontamination by the present SC method. The whole system is shown in Fig. 6. Upper half of the figure shows the main circulation line of SC solution, and the lower part includes the Ce(IV)-regeneration line(right hand side) and the waste-solution process line. In the main line test section was attached, in which sample specimens from both coolant cleaning line and recirculation line are loaded: Chemical composition of the crud in the former line was Fe:50%, Ni:27%, and Cr:22%, while that in the latter Fe:65%. Ni:20%, and Cr:12%. The characteristics of the loop are summarized in Table 2.



FIG.6. Decontamination test loop diagram of the SC process.

	Surface area	Inner volume
Objtxted line to be cleaned	9.2 m <sup>2</sup>	1.1 m <sup>3</sup>
Supplemental line	33.5 m <sup>2</sup>	1.2 m <sup>3</sup>
Total	42.7 m <sup>2</sup>	2.3 m <sup>3</sup>

# Table 2 Water holding and inner surface area of system decontamination loop (Redox method)



FIG.7. Schematic of the process for sulfuric acid-cerium (SC) decontamination of the JPDR recirculation system.

Step I : Injection of decontamination reagents

Step II : Decontamination with electrolytic regeneration

Step III: Electrolytic reduction of decontamination solution

Step IV: Purification with electrodialysis and ion-exchange resins.

## 3.5.2. Decontamination procedures:

Whole the procedure was consisted from decontamination and waste-solution treatment, and is shown in Fig.7: After adjusting the  $H_2SO_4$ -concentration of the solution in the line to ca. 0.25M, Ce(III) ion was added. Subsequently the solution was heated up to 80<sup>0</sup>C, to which Ce(IV) ion was introduced. Electric generation of Ce(IV) was continued till its concentration reached to around 2 mM. Decontamination was lasted to the end, while checking the Ce(IV) concentration in the solution. After the decontamination, the residual Ce(IV) in the solution was reduced to Ce(III) electrochemically and then the solution was treated by electrodialysis, and then by a deep-bed ion-exchange column. The concentrated waste solution was solidified after being neutralized.



FIG.8. Relation between decontamination factors of specimens and decontamination time.

## 3.5.3. Results:

In the early stage of the decontamination, burst of scale in the line occurred, which consumed Ce(IV) in the system, at a much faster rate than the regenerating rate, resulting to the very small effective concentration of Ce(IV) in the line. Thus the decontamination was once stopped, and the scale was filtered off from the solution by inserting an ordinal filter. The decontamination was further continued. The DF of the sample specimens during and after the decontamination are plotted as a function of time in Fig.8. The final DF was roughly  $3\times10^2$  for the recirculation line and  $1\times10^3$  for the coolant cleaning line.

3.5.4 Summary:

The results are summarised as,

o Activity removed: 3.8 mCi(Calculation gives 2.3 mCi).

o Decontamination factor: 300 - 1800

	vertical pipe	horizontal pipe	elbow pipe	pump-casing
TLD Test pieces	5 - 9 300	10	30 - 50	3 - 5
Sampling	1800	360 -1000		

o Metal removed: 10kg

oSolid waste: 3 drums of 100 liters (for concentrated waste solutions, and sluges)

2 drums of 200 liters (for IX resins)
## 3.5.5 Remarks:

During the reactor operation, various kinds of scales are deposited in various places in the line. These are easily released to the solution in the beginning of the decontamination, as was observed as the burst in the present case, and reacted with Ce(IV), leading to the decrease of the effective concentration of Ce(IV). Since the scales can be released out even in a 0.25 M  $H_2SO_4$  solution, these should be collected by inserting a filtering device, before the addition of Ce(IV). Otherwise, a large capacity of the Ce(IV)-regenerator has to be equipped to dissolve the scale in a short time.

# SUMMARY OF WORK ON CHARACTERIZATION OF THE RADIOACTIVE DEPOSITS ON PWR PRIMARY CIRCUIT SURFACES

M.E. PICK Berkeley Nuclear Laboratories, Central Electricity Generating Board, Berkeley, Gloucestershire, United Kingdom

#### Abstract

In examining decommissioning strategies for LWR's and the possible role of decontamination, characterization of the radioactive deposits on circuit surfaces is required to provide information on the radioactive inventory and the type of oxide on the surface. Knowledge of the latter will determine which is the most appropriate decontamination process to use and its potential efficiency. Results from examinations performed on a number of Inconel 600 steam generator and stainless steel PWR specimens and also a limited number of BWR and CANDU specimens are summarized. A variety of techniques have been utilized including: gamma spectrometry, alpha spectrometry, scanning electron microscopy and wet chemical analysis. In addition, preliminary studies using secondary ion mass spectrometry (SIMS) have been performed. The sources of the major radionuclides present on circuit surfaces are also considered.

#### 1. INTRODUCTION

The major constructional materials used in PWR primary circuits are austenitic stainless steels and high nickel alloys (eg Inconel and Incoloy). In addition, there are small areas of various other alloys in the circuit, eg the high cobalt material, Stellite, which is used as a hard facing in pumps, valves and various other components. These circuit materials corrode slowly during normal operation forming surface oxides and also releasing soluble and particulate material to the circulating coolant. Deposition of this material on the Zircaloy clad can occur where it is neutron activated; re-release and deposition of this activated material onto out-of-core surfaces can then take place. In the majority of LWR's (PWR and BWR) activated corrosion products are the major contributors to radiation fields and doses. Fission products generally contribute < 10% of station dose. The presence of high radiation fields on LWR's has prompted research into decontamination processes, experiences with processes developed in the UK are reviewed together with a summary of the main IAEA agreement work on the characterisation of the oxides formed on out-of-core surfaces.

#### 2. OBJECTIVES OF THE WORK

In examining decommissioning strategies for LWR's and the possible role of decontamination, characterisation of the radioactive deposits on circuit surfaces is required to provide information on the radioactive inventory and the type of oxide on the surface. Knowledge of the latter will determine which is the most appropriate decontamination process to use and its potential efficiency. Results from examinations performed on a number of Inconel 600 steam generator (SG) and stainless steel PWR specimens and also a limited number of BWR and CANDU specimens are summarised. A variety of techniques have been utilised including: gamma spectrometry, alpha spectrometry, scanning electron microscopy and wet chemical analysis. In addition, preliminary studies using secondary ion mass spectrometry (SIMS) have been performed. The sources of the major radionuclides present on circuit surfaces are considered.

#### 3. RESULTS AND DISCUSSION

#### 3.1 Gamma Spectrometry

The predominant isotope on nearly all the specimens examined was  ${}^{60}$ Co formed from an n,  $\gamma$  reaction on  ${}^{59}$ Co, results from a typical reactor are shown in Table 1. The majority of the  ${}^{60}$ Co was associated with fixed (grown-on) oxide on the specimens. The other isotopes detected on all the specimens were  ${}^{54}$ Mn (from n,p on  ${}^{54}$ Fe) and  ${}^{125}$ Sb (n,  $\gamma$  on  ${}^{124}$ Sb and n,p on  ${}^{125}$ Sn). Additional radionuclides detected on some of the specimens included the fission products  ${}^{106}$ Ru,  ${}^{137}$ Cs and  ${}^{144}$ Ce and the activation products  ${}^{94}$ Nb (n,  $\gamma$  on  ${}^{93}$ Nb), the europium isotopes  ${}^{152}$ Eu (n,  $\gamma$  on  ${}^{151}$ Eu),  ${}^{154}$ Eu (n,  $\gamma$  on  ${}^{153}$ Eu) and  ${}^{155}$ Eu (n,  $\gamma$  on  ${}^{154}$ Eu).

The <sup>60</sup>Co radioactivities on SG tube are plotted in Fig. 1 versus effective full power years of operation (EFPY). A reasonable fit of the data points beyond 1.5 EFPY is provided by a simple <sup>60</sup>Co build-up/decay equation  $\frac{R}{\lambda}$  (1-e<sup>- $\lambda$ t</sup>), with a <sup>60</sup>Co deposition rate/of 48 kBq cm<sup>-2</sup> per EFPY. This leads to a saturation level of <sup>60</sup>Co on SG tube surfaces of about 350 kBq cm<sup>-2</sup> after 25 years.

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# TABLE 1

# Radioactivity Levels on a Specimen from a

	Total Activity kBq cm <sup>-2</sup>			
Radionuclide	Inconel 600 SG Tube 5.4 EFPY	Stainless Steel Manway Insert 5.8 EFPY		
<sup>6 0</sup> Co	253	958		
<sup>54</sup> Mn	4.4	26.3		
57 <sub>Co</sub>	0.5	1.7		
<sup>58</sup> Co	170	433		
<sup>65</sup> Zn	2.2	5.2		
<sup>9 4</sup> Nb	0.012	1.7		
106 <sub>Ru</sub>	0.15	0.3		
110m <sub>Ag</sub>	0.015	-		
<sup>125</sup> Sb	0.46	0.2		
<sup>137</sup> Cs	0.0037	-		
<sup>144</sup> Ce	0.24	0.8		
<sup>152</sup> Eu	0.0078	-		
<sup>154</sup> Eu	0.011	0.06		
155 <sub>Eu</sub>	0.011	0.04		





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On stainless steel PWR specimens the major isotopes were again  ${}^{60}$ Co,  ${}^{58}$ Co and  ${}^{54}$ Mn; for similar EFPY on Ringhals 2 specimens (Table 1)  ${}^{60}$ Co levels were about a factor of 4 greater than on SG tube. It was notable that  ${}^{60}$ Co levels on Loviisa PWR specimens were about 2 orders of magnitude lower than on Westinghouse PWR specimens such as Ringhals. Low levels of  ${}^{60}$ Co were also found on CANDU reactor specimens (Table 2). On BWR specimens  ${}^{60}$ Co was also the predominant isotope, although as with the CANDU specimens a number of other radionuclides were present in appreciable amounts, in particular  ${}^{65}$ Zn. On BWR circuits this is thought to arise from the brass condensers and other components in the feed train.

#### TABLE 2

Reactor (EFPY)	Radio- nuclide	Total Activity (kBq cm <sup>-2</sup> )
Bruce 3 (5.9)	<sup>60</sup> Co <sup>54</sup> Mn <sup>65</sup> Zn 106Ru 124Sb 125Sb 144Ce	0.22 0.05 0.15 2.0 0.70 0.16 3.9
Bruce 4 (3.5)	60Co 54Mn 65Zn 106Ru 125Sb 137Cs 144Ce	1.7 0.81 18.9 0.73 0.027 0.005 0.77

Radioactivity Levels on CANDU SG Tube Specimens

#### 3.2 Alpha Spectrometry

Results from direct alpha spectrometry of the PWR specimens are summarised in Table 3. Two of the reactors showed < 0.05 Bq cm<sup>-2</sup> of alpha radioactivity. It is possible that this level may represent the quantity expected from uranium impurities in the circuit, eg tramp uranium on fuel. On the other specimens, the total alpha radioactivity varied from 0.9-26 Bq cm<sup>-2</sup>, although if short-lived <sup>242</sup>Cm data are neglected the range is reduced to 0.9-4.9 Bq cm<sup>-2</sup>. The reactors examined had experienced up to

#### TABLE 3

	Bq cm <sup>-2</sup>
Total Alpha Activity	0.022 - 26
$^{239}$ Pu + $^{240}$ Pu	0.0078 - 1.7
$^{238}$ Pu + $^{241}$ Am	0.013 - 2.9
<sup>244</sup> Cm	0.0015 - 0.63
<sup>242</sup> Cm	0.0 - 23

Actinide Radioactivities Measured on PWR Specimens

No of reactors examined = 10EFPY range = 1.2 - 6.8

6.8 EFPY of operation. It is difficult to predict levels on circuit surfaces at the end of reactor life since these will depend upon the number of fuel failures which are probably the major source of actinides. Calculation of the actinide inventory at the end of life also requires an assessment of the impact of the growth of daughter products. The major change will result from decay of the weak beta ( $\beta$ ) emitter <sup>241</sup>Pu (T½,15y) to <sup>241</sup>Am (T½,433y). Actinide concentrations on three BWR specimens were also measured, levels recorded were in the range 81-174 Bq cm<sup>-2</sup>, ie somewhat higher than on the PWR specimens.

# 3.3 Beta and X-Ray Spectrometry

A number of activation products and fission products which do not emit gamma-rays and decay by either pure  $\beta$ -emission or electron-capture (EC) will be present on circuit surfaces. These are likely to include the following radionuclides:

<sup>55</sup>Fe (EC, T<sup>1</sup>/<sub>2</sub> 2.7y) from n,  $\gamma$  on <sup>54</sup>Fe <sup>59</sup>Ni (EC, T<sup>1</sup>/<sub>2</sub> 7.6 x 10<sup>4</sup>y) from n,  $\gamma$  on <sup>58</sup>Ni <sup>63</sup>Ni ( $\beta$ , T<sup>1</sup>/<sub>2</sub> 100y) from n,  $\gamma$  on <sup>62</sup>Ni <sup>90</sup>Sr ( $\beta$ , T<sup>1</sup>/<sub>2</sub> 29y) fission product <sup>93</sup>Mo (EC, T<sup>1</sup>/<sub>2</sub> 3.5 x 10<sup>3</sup>y) from n,  $\gamma$  on <sup>92</sup>Mo In Fig. 2 the calculated variation in primary circuit contamination radioactivity with isotope and decay time is shown. It is apparent that <sup>63</sup>Ni becomes the predominant isotope for decay periods beyond 30 years.





## 3.4 Chemical Analysis

Results from chemical analysis of Inconel SG tube oxides for iron, nickel, chromium and cobalt using techniques described in Ref 1 showed that with respect to the base metal the fixed oxide was always enriched in chromium which varied from 30-55% compared with 16% in the base metal. Iron concentrations were in the range 21-38% and nickel 23-48% The loose particulate oxide was lower in chromium than the fixed oxide and higher in iron and nickel. Cobalt concentrations in the fixed oxide ranged from 0.46-1.7% and showed a considerable enrichment (7-30) over the cobalt in the base metal. Analyses of oxides on stainless steel specimens showed similar results to the SG tube with enrichment of chromium and cobalt. A notable exception was specimens from Loviisa where no enrichment of cobalt was observed. Cobalt levels in the oxide (0.06-0.08%) were identical to those in the base metal (0.06-0.085%). The Russian designed Loviisa reactors are reported to contain no Stellite. This suggested strongly that the high cobalt concentrations observed in the oxides from Western type PWR's arose from Stellite wear and corrosion. This conclusion was also supported by a mass balance study (Ref 2). On the specimens from BRUCE-CANDU reactors (Table 2) very low  $^{60}$ Co arisings were also noted. On these reactors, steps have been taken to replace the majority of the Stellite by other hard-facing materials and steam generator tubing containing a low concentration of cobalt (<0.015%) has also been used.

#### 3.5 Metallographic Examination

Sections taken through the oxide on the SG tube specimens showed that in general it was less than 1  $\mu$ m thick. The surface of the tubes was very smooth although in some cases there was evidence of penetration of oxide into the metal up to a depth of about 10  $\mu$ m. In contrast the surface of the stainless steel specimens examined was generally rough with typical peak to valley distances of 20  $\mu$ m. Oxide thicknesses were greater than on the SG tube specimens and ranged up to 5  $\mu$ m.

#### 3.6 Scanning Electron Microscopy

An electron-micrograph of the oxide on Ringhals 2 SG tube is shown in Fig. 3a. There are a number of small particles on the surface but they only occupy a small proportion of the surface area. A more interesting electron-micrograph is shown in Fig. 3b which is of the underside (ie oxide-metal interface) of the oxide stripped from the tube using bromine/methanol solution. The pattern shown corresponds to where oxide has penetrated down grain boundaries to a depth of about 10  $\mu$ m. Electronmicrographs of the stainless steel specimens examined tended to show a rather rougher surface than the SG tube, as expected from the metallographic examinations.

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(b) Underside Oxide – Metal Interface

20 µ m

FIG.3. ELECTRON - MICROGRAPHS OF PRIMARY SIDE OXIDE ON RINGHALS 2 (1983) SG TUBE.

#### 3.7 Secondary Ion Mass Spectrometry

A number of the reactor specimens have been examined using a SIMS system with an argon ion beam to produce depth profiles through the oxide. A typical SIMS spectrum obtained from a Ringhals 2 specimen is shown in Fig. 4. Although SIMS is a highly sensitive technique the SIMS response to different elements varies markedly; for instance, by three orders of magnitude between alkali metals and transition metals and to a lesser extent between elements in the same group. However, the SIMS signals can be processed to produce semi-quantitative depth profiles. One of the main points to emerge from the SIMS study was that there is no clear evidence of a layer-type structure for the PWR fixed oxides; for instance, an inner chromium-rich oxide layer and an outer layer consisting mainly of iron and nickel oxides. A large number of minor elements (0.01-3% level) were easily detected in the oxides by SIMS including lithium, sodium, aluminium, titanium, zinc and zirconium. In general, their concentrations fell as sputtering proceeded through the oxide.



4. EXPERIENCE WITH CHEMICAL DECONTAMINATION PROCESSES DEVELOPED BY THE CEGB IN THE UK

The work described in this paper shows that the radioactive oxides on PWR circuit surfaces can be considered to be of two types: (a) the chromium-rich grown-on oxides which are strongly adherent to the base metal and (b) loose deposited oxide containing more iron and nickel and less chromium. On PWR's the bulk of the radioactivity is associated with the grown-on oxide. On BWR's the oxides are mainly iron based although on some specimens an inner chromium rich layer is present. Historically, the oxides on PWR surfaces have been treated by using a process known as APAC (alkaline permanganate followed by ammonium citrate). This process and various versions of it have been widely used but have the drawback of producing large quantities of liquid waste due to the rinses between stages. Research work over the last several years has focused on the development of more dilute decontamination processes, including dilute versions of the APAC process. At BNL research followed two main routes:

- development of oxidising systems: NP (nitric permanganate) and the POD (PWR oxidative decontamination) procedure.
- development of novel reducing systems: LOMI (low oxidation state metal ion) reagents.

These developments are reviewed in reference 3 and will not be elaborated upon here.

#### 4.1 UK - Winfrith SGHWR

The first application of LOMI to a reactor was on the South circuit of the WSGHWR in 1980. Since then the reagent has been used on both circuits including fuel, each year. The BWR type deposits and fuel surface deposits have dissolved rapidly in LOMI. In some areas of the plant chromium-rich oxides were also present and on occasions LOMI has been combined with NP to improve the decontamination of these areas. The annual LOMI decontaminations of WSGHWR coolant have reversed the upward trends of gamma fields around certain areas of the circuit, without which increasingly demanding safety-related inspections and maintenance work would have become extremely difficult. Annual net savings in dose amount to several hundred man-rems.

#### International

The NP/LOMI and AP/POD processes were first used on a large scale outside the UK in 1982 at Battelle Northwest Laboratories in the USA for decontamination of the hot leg of the Surry 2 SG channel head. Both NP and AP based processes were used in order to maximise the decontamination factors which reached 50 on stainless steel surfaces. Since this first demonstration there have been about 15 further applications of LOMI and NP/LOMI to operating BWR's and PWR's, mainly in the USA (Table 4). On BWR's, LOMI has proved very effective, giving DF's of 10 or more in most cases. In cases where a significant proportion of chromium was present in the oxide the NP/LOMI process was used. The latter process and also AP/NP/LOMI have been used on the PWR applications. The savings in dose from these decontaminations have been considerable. For instance, at Dresden 3 the decontamination operation is estimated to have saved 2,000 man-rems and at Indian Point 3 over 400 man-rems.

#### 4.2 Application of CEGB Processes for Decommissioning Purposes

In principle for decommissioning purposes concerns over corrosion of circuit materials are reduced and aggressive reagents such as mineral acids could be employed for decontamination. However, the problems of treating the radioactive waste solutions remain. An alternative is the use of a multi-stage cycling process based on the dilute reagents used for operating plant. For instance, initially AP or NP solution could be applied for a few hours and then drained to a holding tank. LOMI or citrox solution would then be added and circulated before being drained to a holding tank or ion-exchanged. The original AP or NP solution would then be fed back into the system for a second application followed by a further application of LOMI or citrox and so on.

The cycling procedure was tested with the reagent combinations: NP/Citrox, NP/LOMI, AP/Citrox and AP/LOMI on stainless steel and Inconel SG tube specimens. The NP or AP reagent was employed for a total of 12 hours which was divided into 2, 3, 4 and 6 hour applications; each application was followed by a 2 hour application of either LOMI or citrox. Results showed that an increased number of cycling steps led to an improved DF. With stainless steel specimens, DF's of > 100 were consistently obtained after 6 x 2 hour cycles of NP compared with only ~ 20 after 2 x 6 hour cycles.

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CEGB D	econtamination	Processes	_	Reactor	Applications	11D	to	Mav	1987

REACTOR	TYPE	COMPONENT	PROCESS	AVERAGE DF
Surry	PWR	Channel head	ad NP/LOMI AP/POD	
Monticello	BWR	Recirculation Pipe LOMI/NP/LOMI RWCU LOMI:NP/LOMIx2		27 22
Dresden 3	BWR.	Recirculation Pipe LOMI/LOMI RWCU		12
Quad Cities 1	BWR	Discharge system suction & RWCU	LOMI/NP/LOMI LOMI	8
Indian Point 3	PWR	4 channel heads	AP/NP/LOMI	3 - 7
Conneticut Yankee	PWR	2 channel heads	AP/NP/ Candeconx2	6
Oyster Creek	BWR	Recirculation system	LOMI/NP/LOMI	10
HP Robinson	PWR	Reactor coolant pump	NP/LOMIx2	30
Browns Ferry	BWR	Reactor coolant pump	LOMI NP/LOMI	n.a.
Quad Cities	BWR	Fuel element	AP/LOMI	100 ci removed
Quad Cities 2	BWR	Recirculation pipe + RWCU	LOMI	4.5 4.3
Dresden 2	BWR	Recirculation pipe + RWCU	LOMI	10 n.a.
SGHWR 1980	PTBWR	South primary circuit + fuel	TURCO 4521 LOMI	
SGHWR 1981	PTBWR	Both primary circuits + fuel	LOMI	
SGHWR 1982	PTBWR	Both primary circuits + fuel	LOMI	
SGHWR 1983	PTBWR	Both primary circuits + fuel	LOMI	
SGHWR 1984	PTBWR	South circuit + fuel North circuit + fuel	LOMI NP/LOMI/NP/LOMI	
SGHWR 1985	PTBWR	South circuit + fuel North circuit + fuel	LOMI LOMI/NP/LOMI	
SGHWR 1986	PTBWR	South circuit + fuel North circuit + fuel		
SGHWR 1987	PTBWR			
Dodewaard	BWR	RWCU let-down pipe	LOMIx2	20
Millstone 1	BWR	Recirculation Pipe	LOMI	n•a•
		······································	4	

n.a. = not available

The Inconel SG tube specimens proved more difficult to decontaminate, DF's ranged from 2-30 after 6 x 2 hour cycles. The low DF's on some specimens were thought to be due to penetration of oxide and  $^{60}$ Co down grain boundaries up to a depth of about 10  $\mu$ m. Hence, a more corrosive decontamination reagent would be required for the SG tubes.

#### 5. CONCLUSIONS

The composition of oxides formed on Inconel SG tube and stainless steel PWR specimens are very similar. Both are enriched in chromium by about a factor of two over the base metal chromium concentration of 16-18%. The other major elements in the oxide are iron and nickel. In addition, manganese, titanium, silicon, cobalt, copper and zinc are present at levels of a few per cent or less.

Oxide thicknesses on stainless steel specimens are greater than on Inconel SG tube after a similar EFPY, this may be due to the much rougher surface on the stainless steel specimens.

The predominant gamma-emitting radionuclide on the specimens is  ${}^{60}$ Co, formed from  ${}^{59}$ Co. It is considered that the major source of the  ${}^{60}$ Co is the high cobalt alloy Stellite. Most of the other gamma-emitting radionuclides measured in significant quantities on the specimens, eg  ${}^{54}$ Mn,  ${}^{58}$ Co,  ${}^{65}$ Zn,  ${}^{106}$ Ru,  ${}^{125}$ Sb are shorter-lived than  ${}^{60}$ Co, which will therefore dominate radiation fields in the immediate period after shutdown;  ${}^{94}$ Nb, which is likely to be the dominant contributor to radiation fields after long decay periods, was detected on one of the specimens examined.

Alpha emitting actinides deposited as a result of fuel failures were detected on nearly all the specimens examined; typical levels ranged up to 5 Bq cm<sup>-2</sup> of longer lived actinides on PWR specimens and 100 Bq cm<sup>-2</sup> on BWR specimens. Clearly, their presence on out-of-core surfaces must be taken into account in assessing decommissioning scenarios.

Developments by the CEGB have led to improved decontamination processes for use on PWR and BWR surfaces. These processes based on NP or AP and LOMI reagents have been used in over 20 major applications to date mainly on reactors and components being returned to service. However, the possible application of these processes in a multi-cycling process, to provide high DF's has been investigated and DF's of over 100 on stainless steel specimens have been obtained, on Inconel SG tube DF's are less satisfactory but this is probably due to radioactivity present in grain boundaries up to 10  $\mu$ m or so into the metal.

#### 6. RECOMMENDATIONS AND FUTURE WORK PLANNED

It is recommended that steps should be taken to reduce the potential for <sup>60</sup>Co generation from <sup>59</sup>Co BWR and PWR circuits by replacing Stellited components wherever possible and reducing <sup>59</sup>Co impurities in stainless steel and Inconel.

The levels of pure beta-emitting and electron capture radionuclides have not yet been determined on the specimens available. However, it has been calculated that these radionuclides, eg  $^{63}$ Ni and  $^{59}$ Ni, may well dominate residual radioactivity, although not dose-rates, over long decay periods. Hence measurements to determine these radionuclides are planned.

It is hoped to elucidate the corrosion and oxide formation mechanisms and to help achieve this goal it is planned to examine the reactor specimens in more detail using techniques such as X-ray photoelectron spectroscopy, Auger, transmission electron microscopy and also further SIMS studies.

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# SHIPPINGPORT STATION DECOMMISSIONING PROJECT: OVERVIEW AND PROGRESS REPORT FOR THE FISCAL YEARS 1984–1985, 1986 AND 1987

W.E. MURPHIE Division of Facility and Site Decommissioning Projects, United States Department of Energy, Washington, D.C., United States of America

#### Abstract

A general project overview of the Shippingport Station decommissioning programme is given. This includes the background of the project, and the development and implementation of plans for the management, engineering and site operations. The technical objectives of the project are highlighted. Removal of reactor and internals in one-piece is a special feature of this project. Physical work of decommissioning started in 1985 with the site release scheduled for 1990.

## REPORT PRECIS

The technical objectives of the project are as follows:

- 1. Removal of all government-owned facilities and radioactive portions of the Shippingport Station.
- 2. Optimization of subcontractor participation.
- 3. Documentation of management, planning, engineering, and operations.

The following project milestones are complete:

- . Award DOC contract March 1984
- . DOE-HQ approval to start decommissioning operations June 1984
- . Shippingport site turnover and initiation of caretaker status-September 1984
- . Initiate Decommissioning Operations January 1985
- . Start physical work of decommissioning September 1985
- . Complete transfer of irradiated components to RPV March 1986
- . Complete removal of AC and BD Chamber primary components March 1987
- . Complete removal of pipe and equipment from AC and BD Chambers April 1987

At the end of FY 1987, decommissioning Operations are 55.2 percent complete versus 57.4 percent planned, according to present target schedule. The target schedule does reflect a completion earlier than forecast.

At the end of FY 1987, budgeted cost of work scheduled is 58.9 million; budgeted cost of work performed is 56.9 million; and actual cost of work performed is 56.1 million. This is an unfavorable schedule variance of \$2.5 million (1%) and a favorable cost variance of \$.8 million (1%). Both are considered to be acceptable.

# HISTORY OF THE SHIPPINGPORT ATOMIC POWER STATION

The Shippingport Atomic Power Station is located on the south bank of the Ohio River at Shippingport (Beaver Valley), Pennsylvania, on approximately seven acres of land leased from Duquesne Light Company (DLC) by the U.S. Department of Energy (DOE). This location is approximately 25 miles northwest of Pittsburgh, Pennsylvania.

The Station was constructed during the mid-1950s as a joint project of the Federal Government and DLC. The purposes of the project were to develop and demonstrate pressurized water reactor (PWR) technology and to generate electricity. The reactor and steam generating portions of the Station are owned by DOC, and the electrical generating portion is owned by DLC.

The Station achieved criticality in December 1957 and was operated by DLC under supervision of DOE-Naval Reactors (NR) until operations were terminated on October 1, 1982. End-of-life testing as well as reactor defueling were conducted in the following two years. The Station utilized three cores of reactor fuel. The first two cores were PWR cores, and the last core was a light water breeder reactor (LWBR) core. The LWBR core was installed in 1977 for the purpose of demonstrating the thermal breeding principle in a light water reactor. Responsibility for the Station was transferred from Naval Reactors to Richland, within DOE, on September 6, 1984, with GE replacing DLC as Operations Contractor at that time.

Figure 1 displays summary operational data and a chronological historical sequence of operations at Shippingport.

Pressurized Water Reactor Project Authorized	July 1953
Ground Broken	September 6, 1954
Construction Started	March 1955
Initial Core 1 Criticality	December 2, 1957
Core 1 Operations Complete	February 9, 1964
Initial Core 2 Criticality	April 9, 1965
Core 2 Operations Complete	February 4, 1974
Initial Light Water Breeder Reactor Criticality	August 26, 1977
Light Water Breeder Reactor Operations Complete	October 1, 1982
Defueling and End-of-Life Testing Complete	September 6, 1984
Turnover for Decommissioning	September 6, 1984
Physical Decommissioning Operations Started	September 17, 1985

	Total Operation (EFPH)	Total Gross Generation (kW-HR)
Core 1 (PWR)	27,780.9	1,793,581,700
Core 2 (PWR)	23,812.0	3,476,592,300
Core 3 (LWBR)	28,730,4	2,103,833,029
Totals	80,323.3	7,374,007,029

EFPH = Effective Full Power Hours

FIG.1. Shippingport atomic power station: chronological sequence of operations and summary operational data.

# PROJECT TECHNICAL OBJECTIVES

The technical objectives of SSDP decommissioning operations are:

- Removal of all government-owned facilities, components, and other radioactive portions of the Shippingport Station necessary to meet standards for unrestricted use of the site.
- Optimization of the number of subcontractors used for decommissioning work in order to increase the number of qualified, available decommissioning contractors; thereby assuring the transfer of decommissioning technology to the U.S. nuclear industry.

3. Documentation of the management, planning, engineering and operations aspects of the project in a appropriate manner to assure long-term preservation and retrieval of performance data so that the experience gained on Shippingport can be readily transferred to industry for use in future decommissioning projects.

The accomplishment of the above technical objectives will demonstrate to the public and power generation industry that nuclear power reactors can be safely decommissioned at reasonable cost.

Another important fact that follows from these objectives is that the SSDP is not defining new technology, but rather drawing from current knowledge. The project is planned to demonstrate decommissioning operations within an environment of current industry practice (e.g., dismantlement and other SSDP operational procedures are based upon current construction, operation, maintenance, and demolition practices).

## PROJECT SCHEDULE OBJECTIVES

The final key schedule objective is to release the Shippingport Station site by April 1990.

# PROJECT COST OBJECTIVES

The total project cost is estimated at \$98.3 million.

## PROJECT SCOPE

The scope of the SSDP, established in the Project Management Plan, includes two major phases:

#### <u>Phase 1</u>

The first phase began in FY 1979 with the signing of a Program Management Agreement between the Division of Naval REactors and the Nuclear Waste Management Office at DOE-Headquarters (DOE-HQ). A Decommissioning Assessment and an Environmental Assessment were completed in FY 1979, and a Final Environmental Impact Statement (EIS) was issued in May 1982. The preferred EIS alternative called for immediate dismantling, following defueling of the Shippingport Station reactor; and this decision was published in the DOE Record of Decision in the Federal Register on August 7, 1982. the SSDP is identified as Major Project Number 118.

Engineering and planning for the SSDP began in 1980 and was completed in September 1983 upon DOE-RL issuance of an approved "Decommissioning Plan for the Shippingport Station Decommissioning Project" (RL/SFM-83-4). This comprehensive twelve-volume plan was compiled by Burns & Roe Industrial Services Corporation and includes specifications for the dismantling work as well as supporting engineering studies and other technical documents. A separate detailed cost estimate for the project was also provided at that time.

#### <u>Phase 2</u>

The second phase began in the second quarter of FY 1984 and will be completed in FY 1990 when the Shippingport Site has been fully dismantled and the DOE-HQ Assistant Secretary for Nuclear Energy has approved release of the site for unrestricted use.

Phase 2 of the SSDP is termed "Decommissioning Operations" and is divided into a caretaker period followed by the physical decommissioning. The caretaker period involved approval to initiate decommissioning operations, award of the contract, training, document development (detailed plans, specifications, and procedures), and maintenance, surveillance and site operations work. Physical decommissioning involves preparations for decommissioning, decontamination, and dismantling of designated facilities, waste packaging and transportation, and certification of the site for release.

The general decommissioning approach is to first perform site preparation work and remove all asbestos insulation, followed by radioactive an non-radioactive piping systems, components and structural material. Next, the remaining steel and concrete structures are to be surveyed, decontaminated as necessary, and released for further dismantlement by standard demolition methods. Finally, the site is to be restored, approved for release and returned to the custody of DLC. Buildings and chambers which form contamination control boundaries are not to be dismantled until all internal work which may have potential for contamination release is completed.

#### CHARACTERIZATION OF PROJECT WORK

The decommissioning work is characterized by the following features:

- . The reactor vessel, internals, and the neutron shield tank are to be removed from the plant as one package and shipped for burial by barge. Shielding is to be provided by installing an engineered fill material in the neutron shield tank.
- . The steam generator heat exchangers, the pressurizer, and other major contaminated plant components are to be shipped as their own containers for burial.

- . The primary systems are not to be given a general decontamination prior to dismantling since a study showed this was not cost effective.
- . Liquid wastes are to be processed by filtration and dilution. Filters and resin columns are to be used to process the existing liquid inventory. Final liquid waste quantities are to be processed by a small temporary filtration system when dismantlement of radioactive waste facilities begins.
- . Tank bottoms and sludge are to be solidified with cement in steel drums. Spent resins are to be placed in high integrity containers and dewatered.
- . Underground structures, after decontamination to unrestricted release levels, are to be removed to three feet below grade. Underground spaces are to be backfilled. The top six inches are to be filled with topsoil. The ground surface is to be contoured for drainage, and erosion control vegetation is to be planted.

Figure 2 gives estimated summary technical statistics for the project that indicate size of the tasks involved in decommissioning, and Figure 3 is a site plot plan which shows the structures to be removed.

Reactor Vessel Package	870 Tons
Radwaste Volume	3000 Cubic Yards
Radioactive Contents	13,500 Curies
Vessels/Tanks	130
Chamber Steel	22,400 Tons
Contaminated Concrete	50 Cubic Yards
Non-Contaminated Rubble	15,000 Cubic Yards
Contaminated Pipe	56,000 Linear Feet
Non-Contaminated Pipe	55,000 Linear Feet

FIG.2. SSDP estimated summary technical statistics (1985).



FIG.3. Shippingport station site plan.

## PRIMARY PARTICIPANTS AND CONTRACTORS

## U.S. Department of Energy

The SSDP is a major project within DOE under the Assistant Secretary for Nuclear Energy with program responsibilities assigned to the Division of Facility and Site Decommissioning Projects in the Office of Remedial Action and Waste Technology at Headquarters.

The management of the SSDP has been assigned to DOE-Richland Operations Office (DOE-RL). The Project Manager at the on-site Shippingport Station Decommissioning Project Office (SSDPO) is responsible for project execution, implementation, and on-site administration of the SSDP.

# Technical Support Contractor (TSC)

The SSDPO includes a dedicated TSC. UNC Nuclear Industries, now Westinghouse Hanford Company, serves as TSC under its basic operating contract with DOE-RL. In this role, the TSC provides direct support to DOE site management for technical, cost, and schedule management of the SSDP.

# Decommissioning Operations Contractor (DOC)

General Electric Company, with its integrated subcontractor, MK-Ferguson Company, is the DOC. The DOC is responsible for site operations and management support, directing decommissioning activities, and obtaining and managing the decommissioning subcontractors.

# PROJECT COST AND SCHEDULE STATUS

The overall Project Summary Schedule (Figure 4) details the project activities to Work Breakdown structure level 2, and depicts project milestones. Milestones and timelines have been shaded to indicate status. During FY 1987, the project milestone for the removal of AC and BD Chamber primary components was completed on schedule in March, along with removal of pipe and equipment from the AC and BD Chambers, which was completed in April.



FIG.4. Shippingport station decommissioning project: project summary schedule.

The SSDP's total estimated cost is \$98,300,000. Figure 5 represents budgeted cost of work scheduled (BCWS), budgeted cost of work performed (BCWP), and actual cost of work performed (ACWP) through FY 1987.



FIG.5. FY 1987 project performance chart.

At fiscal year end, the project's BCWS was \$58,971,000; BCWP was \$56,997,000; and ACWP was \$56,138,000. This represents a favorable cost variance of \$859,000 and an unfavorable schedule variance of \$2,449,000. This schedule variance equates to approximately three weeks behind schedule for the project. In terms of Phase 2 activities, the project progress levels are 55.2 percent actual versus 57.4 percent planned on the accelerated schedule which is attempting to foreshorten the project by six months.

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