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Decontamination and decommissioning of nuclear facilities

Results of a Co-ordinated Research Programme, Phase II: 1989–1993



INTERNATIONAL ATOMIC ENERGY AGENCY

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FOREWORD

The International Atomic Energy Agency first introduced the subject of decontamination and decommissioning into its programme in 1973, publishing reports reflecting the needs of its Member States on these topics. These reports summarize the work done by various Technical Committees, Advisory Groups and International Symposia.

The decontamination and decommissioning of nuclear facilities is of great interest to many Member States of the IAEA because of the large number of older nuclear facilities which have been or soon will be retired from service. In response to increased international interest in this area and to the needs of Member States, the IAEA initiated the Co-ordinated Research Programme (CRP) on Decontamination and Decommissioning of Nuclear Facilities to create a forum for scientists from different Member States, to bring them into closer contact.

The first phase of the Co-ordinated Research Programme was carried out from 1984 to 1987. 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 related to overall decommissioning activities. The salient features and achievements of the co-ordinated research work performed were summarized in IAEA-TECDOC-511 (1989). It was the recommendation of the participants that the IAEA should extend such initiative and undertake another phase of the CRP on decontamination and decommissioning.

In line with the recommendation of the experts who participated in the first phase of the CRP, the aim of the second phase was to include several areas of decontamination and decommissioning rather than focusing on a single aspect of it. It was also suggested that the number of participants should be increased to include participation from other countries, especially developing Member States. The second phase of the CRP was established in 1989 and concluded in 1993. Experts from 14 Member States participated in this CRP: Canada, China, Czechoslovakia, Finland, France, Hungary, India, Italy, Japan, the Republic of Korea, the Russian Federation, Sweden, the United Kingdom and the United States of America. Research Co-ordination Meetings (RCMs) were held in Vienna (1989, 1991) and in Japan (1993).

The third and final RCM was held in Tokai, Japan, from 22 to 26 February 1993 and was hosted by the Japan Atomic Energy Research Institute (JAERI). The meeting was chaired by D. Mancey (Canada). Concluding reports that summarized the work undertaken within the four year duration of the CRP were presented at the meeting by the chief scientific investigators or their delegates.

During the third RCM, scientific visits were made to several facilities at the Tokai and Oarai sites. The most important one from the decommissioning point of view was the JAERI Japan Power Demonstration Reactor (JPDR), which is currently being decommissioned. In addition, visits were made to the JAERI reprocessing facility (JRTF), which it is planned to dismantle, and to waste management facilities.

The present document summarizes the achievements of the co-ordinated research work performed during the 1989-1993 programme period. The IAEA officers responsible for the meetings were P.L. De (first and second meetings) and K. Shimooka (third meeting and compilation of the report), both of the Division of Nuclear Fuel Cycle and Waste Management.

The IAEA wishes to express its thanks to all the participants in the programme and would like to take this opportunity to acknowledge the excellent co-operation and hospitality of the Japan Atomic Energy Research Institute in hosting the third Research Co-ordination Meeting.

EDITORIAL NOTE

In preparing this document for press, staff of the IAEA have made up the pages from the original manuscripts as submitted by the authors. The views expressed do not necessarily reflect those of the governments of the nominating Member States or of the nominating organizations.

Throughout the text names of Member States are retained as they were when the text was compiled.

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SUMMARY OF THE CO-ORDINATED RESEARCH PROGRAMME (PHASE II)

1. INTRODUCTION

The term 'decommissioning', as used by the nuclear industry, means the actions taken at the end of a facility's useful life to retire the facility from service in a manner that provides adequate protection for the health and safety of the decommissioning worker, the general public and the environment [1]. These actions, which should be in conformity with the country's national laws and regulations, could range from merely closing down the facility and carrying out minimal removal of radioactive material, coupled with continuing maintenance and surveillance, to a complete removal of residual radioactivity in excess of levels acceptable for unrestricted use of the facility and its site. This latter condition, unrestricted use, is the ultimate goal of all decommissioning actions at retired nuclear facilities.

Currently, about 420 nuclear power reactors and 320 research reactors are in operation. According to Refs [2, 3], by the year 2003, 87 nuclear power plants and 248 research reactors will have reached 30 years of age and will therefore become likely candidates for decommissioning by that time. To date, 234 research reactors and 62 nuclear power reactors have already been shut down. In addition, many other non-reactor nuclear facilities will soon require decontamination and decommissioning.

In response to increased international interest and to the needs of Member States, the IAEA activities in these areas have increased since the topic was introduced in the IAEA's programme in 1973. The first phase of the Co-ordinated Research Programme (CRP) on Decontamination and Decommissioning (1984–1987) was organized mainly to promote the exchange of information on the specialized topic of decontamination [4]. The second phase of the CRP was carried out in line with the recommendations of the first phase of the CRP to include topics related to many aspects of decontamination and decommissioning such as chemical and electrochemical decontamination, dismantling techniques and decommissioning strategies. Safety standards and safety guides on decommissioning are being developed in the framework of the Radioactive Waste Safety Standards (RADWASS) Programme [5] and these activities will be enhanced in the future. It may be assumed that the CRP has served, together with other IAEA technical reports and documents, to provide background information for the RADWASS programme.

2. OBJECTIVES AND SCOPE

The objectives of this CRP were to promote the exchange of information on the practical experience by Member States in decontamination and decommissioning.

The scope of the programme included several areas of decontamination and decommissioning rather than focusing on a single aspect of it, in line with the recommendation of the experts who participated in Phase I of the CRP. Experts felt that this format would generate better awareness of decontamination and decommissioning and would be a more effective vehicle for the exchange of information by stimulating broader discussion on all aspects of decontamination and decommissioning. Special emphasis was given to the development of principles and methodologies to facilitate decommissioning and to the new methods and techniques for optimization of decontamination and disassembly of equipment. In particular, the scope included:

- (i) Decommissioning project management: various aspects of a large decommissioning project (power reactor, research reactor or other nuclear facility) being undertaken or planned in the immediate future;
- (ii) Decontamination, disassembly and refurbishment of research reactors and facilities;
- (iii) Decontamination for recycle, reuse, refurbishment or unrestricted release;

- (iv) Application of remotely controlled operation for decontamination, dismantling, packaging and transportation; and
 - (v) Facilitation of decommissioning.

A list of the Chief Scientific Investigators and CRP topics is given in Table I.

TABLE I: CHIEF SCIENTIFIC INVESTIGATORS AND CRP TOPICS

COUNTRY CSI/ORGANIZATION		CRP TOPIC			
Canada	Spekkens, P. Ontario Hydro Research Division Chemical Research Division	Mechanisms of magnetite dissolution in organic acids			
China	Ren, Xianwen China Institute for Radiation Protection	Electropolishing of radionuclide contaminated metal materials			
Czech and Slovak Republic	Blazek, J. Nuclear Power Plants Research Institute	Decontamination for decommissioning of nuclear power plants			
Finland	Salonen, P. Reactor Laboratory, Technical Research Centre of Finland	Long-lived activation products in reactor bioshield material			
France	Roger, J. IPSN/UDIN CEA/CEN Vallée du Rhône	Development of cutting tools for dismantling of nuclear facilities			
Hungary	Pavlik, O. Institute of Isotopes of the Hungarian Academy of Sciences	Electrochemical decontamination for the main pipeline of the primary circuit by movable cathode			
India	Theyyunni, T.K. Process Engineering & Systems Development Division Bhabha Atomic Research Centre	Decommissioning and decontamination studies for nuclear facilities			
Italy	D'Anna, C. ENEL-DPT Unità Attività Nucleari	Project management and general planning of decommissioning activities for the Garigliano (BWR, 160 MWe) and Latina (Magnox, 200 MWe) nuclear power plants			
Japan	Yokota, M. JAERI, Tokai Research Establishment	Decommissioning project management			
Korea, Republic of	Won Zin Oh Korea Advanced Energy Research Institute	Development of decontamination and remedial action technology			
Russian Federation	Kremnev, V.A. Joint Venture DECOM	 Decommissioning project management; Decontamination for recycle, reuse, refurbishment or unrestricted release Facilitation of decommissioning 			
Sweden	Pettersson, S. Swedish Nuclear Fuel and Waste Management Co.	Removal, transport and disposal of reactor vessels in one piece from Swedish nuclear power plants			
United Kingdom	Pick, M.E. Radiological Protection Branch Nuclear Electric plc	PWR oxide characterisation/surface finish improvement and development of decontamination processes for gas and water cooled reactors			
United States of America	Murphie, W.E. US Department of Energy Office of Environmental Restoration and Waste Management	Decommissioning of the experimental boiling water reactor pressure vessel			

3. SUMMARY OF MAJOR TECHNICAL ACHIEVEMENTS

3.1. Chemical and electrochemical decontamination

For the purpose of this CRP, decontamination is defined as the removal of activity from metallic and non-metallic surfaces. There are two general categories of decontamination process, those performed during a plant's operational life, and those undertaken as part of decommissioning. Decontaminations are performed to minimize personnel radiation exposure and to reduce or requalify waste arisings from decommissioning activities.

The projects undertaken as part of the Co-ordinated Research Programme on Decontamination and Decommissioning involve the decontamination of metals by chemical or electrochemical methods. Specifically, the projects were undertaken to address the following issues:

- (a) Advance the understanding of oxide dissolution mechanisms. This work involved the study of the kinetics and mechanism of magnetite dissolution in dilute acidic decontamination reagents. The results of these studies emphasize the electrochemical nature of the process, and quantitatively defined the influence of temperature, pH, potential and hydrodynamics on the kinetics of magnetite dissolution.
- (b) Qualitatively describe the electrochemistry of V(II)/V(III)-picolinate systems, as part of an effort to develop a regenerative decontamination process.
- (c) Optimize decontamination formulations and processes for specific applications. The performance of decontamination reagents based upon various combinations of picolinic acid, EDTA, formic acid, oxalic acid and ascorbic acid, were examined.
- (d) Develop an electrochemical method of decontamination for carbon steels that is capable of reducing the level of contamination to that which will permit the unrestricted release of the steel. Such processes have been investigated in reagents based upon two inexpensive and commercially available chemicals, sulfuric acid and citric acid.

During R&D on decontamination process, it is often difficult to obtain samples that are representative of the materials to be decontaminated. In many instances problems associated with accessibility and radioactivity prohibit the use of specimens taken from actual nuclear facilities. Often decontaminations involve the dissolution of adherent oxide films that have grown, over the course of many years, on metal surface. Practically, the laboratory synthesis of such films is often impossible. For this reason, film dissolution processes are often investigated by performing studies on the dissolution of bulk oxides and clean metal surface. Great care must be exercised when adopting this approach, it must be determined that the metals and oxides used are truly representative of those required. Particularly for the oxide, this necessitates extensive chemical and physical characterization. Further, even if the individual phases are representative of the film components, it is necessary to confirm experimentally that the data obtained yields an acceptable model of film dissolution.

Although advances have been made in decontamination technology, further improvements in decontamination reagent formulation and application must be sought. Among the important issues that require further attention are the following:

- (a) The dissolution kinetics and mechanism of nickel and chromium containing oxides.
- (b) The development of alternative decontamination reagent formulations and processes.
- (c) The formulation of guidelines for the selection, verification and application of decontamination processes.

3.2. Dismantling techniques

The following describes the major technical achievements on dismantling techniques, difficulties encountered during the CRP and future work required:

(a) Major technical achievements

The Japan Power Demonstration Reactor (JPDR) Decontamination and Decommissioning (D&D) Project has made good progress in developing new technologies for reactor decommissioning and collecting of various data on project management and the performance of the developed tooling. The experiences gained on the project will serve to contribute to future decommissioning of commercial nuclear power plants. Of specific interest was the development, deployment and field testing of tools to perform the following: removal of the reactor vessel by arc saw cutting, removal of the reactor vessel internals by plasma cutting, removal of biological shield by abrasive water jet cutting and diamond saw cutting, and controlled explosive cutting of piping connected to the reactor vessel.

The Experimental Boiling Water Reactor (EBWR) Decommissioning Project has made good progress in preparing for the removal of the EBWR reactor vessel. During this period, removal of the reactor systems and components was completed and the wastes disposed of at the Department of Energy (DOE) Richland disposal site. Facility modifications for the removal of the reactor vessel have been completed including : installation of a new High-Efficiency Particulate Air (HEPA) ventilation system, and installation of a water filtration and transfer system for treatment of and processing of spent fuel pool water. The latter system is critical for size reduction operations on the reactor internals of the EBWR. Work is now starting on the removal of the reactor vessel has made progress during this period, including: design of the lifting frame and backing plate for the reactor vessel (RV) removal, check-out of the testing abrasive water jet unit on steel plate simulating the EBWR reactor vessel and check-out of the water jet carrier unit.

The work carried out by the French participants consisted of testing tools suitable for use on D&D projects in order to make the tools more efficient and for better use in remote control operations. The work carried out involved: performing comparative tests of tools for metal cutting during D&D, and performing comparative tests of two tools for concrete cutting during D&D. The plasma torch is the most rapid tool, and shows the least tool wear. Mechanical tools (milling etc.) are the slowest tools, but are also very interesting, because they: show low tool wear, produce the least aerosols, produce the metal chips that are very easily to collected and cause no heating of the work piece. Work has been completed on the diamond saw cutting technique and work is underway on developing the shot blasting machine. Active trials with it look promising.

(b) Difficulties encountered

The following is the summary of difficulties encountered during work on dismantling techniques. The difficulties encountered during these efforts are as follows:

- Regulatory changes or guidelines being developed can cause a need to revise a previously planned method for a particular D&D activity or even cause work to stop after an implementation plan is developed/ approved.
- Generation of secondary wastes was a common problem. Further work is required to minimize waste arisings and to efficiently treat these secondary wasters. The methods for efficient treatment were not as readily available as desired.

(c) Future work required

The following is a summary of the areas in which future work is required:

- Concrete structure dismantling techniques;
- Dismantling techniques that include all processes in using the D&D tools: preparation, set-up, cutting and clean-up from the operation;
- Methods to minimize secondary wastes and treat wastes in order to minimize all wastes generated during D&D activities;
- Development of tools for D&D with multi-purpose capabilities on a number of nuclear facilities: hot cells, reactors, fuel reprocessing, etc. These tools should be simple, yet multi-purpose in nature;
- Continued development of methods to minimize radiation exposures and continue good, safe decommissioning operations; and
- Further exchange of technical information on D&D activities of various Member States to facilitate development of new, innovative and better dismantling techniques.

3.3. Decommissioning strategies

The development of a safe, effective and cost efficient strategy for the decommissioning of each Member State's nuclear facilities has to be done within the overall framework of its nuclear policy and of its safety and regulatory requirements. The approaches to this end vary significantly depending on factors such as :

- the availability of waste storage or disposal capacity;
- future use of the site;
- the types of facilities;
- the financial resources needed to complete the task;
- whether the facilities are private or state owned; and
- the availability of the necessary technology and skilled workforce.

Although the contents of national decommissioning policies vary among Member States, they in general all include the following:

- Waste Management. The availability of storage and/or disposal facilities for the spent fuel and waste arisings from decommissioning dictates the overall decommissioning strategy, and affects the costs. If, for example, there is no offsite spent fuel storage, or reprocessing or disposal capability, then the release of reactor sites will be delayed;
- Financial Assurance. Provisions for assuring that sufficient funds are available for performing the necessary decommissioning work are required by most Member States; and
- The recycling and re-use of material recovered in decommissioning work. This may be an attractive alternative to waste disposal. The decision whether to use this approach depends on many factors such as : technical feasibility; the availability of accepted residual activity levels; economic implications and public acceptance. At present few Member States have issued firm criteria for applying this alternative.

The choice of a preferred decommissioning strategy is in general based on an optimization study which includes radiation protection amongst many other factors. In some cases the choice is not obvious; decision making techniques such as cost-benefit or multi-attribute analysis may be used. There are many factors to be considered. The impact parameters to be considered generally fall into four groups:

- safety and environmental factors;
- technical factors;

- cost; and
- social and political considerations.

In order to permit advanced (timely) provisions of waste disposal facilities and dismantling resources, the decommissioning plans and strategies should cover nuclear plants that are already shut down, as well as those which are still in operation. These plants and strategies should be reviewed regularly in the light of R&D progress, regulatory changes, political developments and economic situation.

It is our opinion that there is much to be gained from international cooperation on issues related to waste treatment and decommissioning technologies. Priority should be given to the following:

- (a) agreement on regulatory exemption limits and the recycling of materials;
- (b) the sharing of major recycling facilities, such as melters; and
- (c) co-operative support for the development of the relevant technologies.

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THE INFLUENCE OF POTENTIAL, TEMPERATURE, pH, AND HYDRODYNAMICS, ON THE KINETICS OF MAGNETITE DISSOLUTION

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Abstract

The dissolution of oxides is a pre-requisite to successful decontamination by chemical means. Frequently, the principal oxide encountered in nuclear power station cooling circuits is magnetite, or a related oxide. Consequently, the formulation and application of decontamination reagents can benefit from an detailed understanding of the magnetite dissolution reaction (MDR). The influence of potential, pH, temperature and hydrodynamics upon the kinetics of magnetite dissolution. The mechanism of the MDR, combined with the kinetic data, leads to a quantitative expression for the dissolution rate of magnetite as a function of the four investigated variables (potential, pH, temperature and electrode rotation frequency). The predictions of this quantitative model are shown to be in good agreement with the experimental observations.

1. INTRODUCTION

Decontamination reagents function by dissolving oxide films and particles. This dissolution process liberates the radioactive constituents of the oxides, facilitating the removal of the activity from the system undergoing decontamination. The nature of the oxides that form in the primary coolant circuits of water cooled nuclear reactors depends both upon the materials of construction and operating conditions. In most instances, these oxides are predominantly magnetite or a related ferrite. This report describes a fundamental investigation of the mechanism and kinetics of magnetite dissolution. The work discussed in this paper emphasised the electrochemical nature of the magnetite dissolution reaction (MDR). This report describes the effect of temperature, pH, decontamination reagent formulation and mass transport upon the MDR.

2. EXPERIMENTAL

2.1. Magnetite electrodes

Rotating disc electrodes (RDE) and rotating ring-disc electrodes (RRDE) were made from magnetite of geological origin. The magnetite discs were obtained from the specimens of magnetite by a process of cutting and polishing. The magnetite discs were recessed in a Teflon housing to produce a coplanar surface. This surface was then polished until the magnetite surface exhibited a mirror finish. The disc electrode assembly was attached to the end of a cylindrical shaft that permitted rotation of the magnetite disc at speeds of between 0 and 10,000 rpm.

2.2. Magnetite dissolution experiments

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All the dissolution experiments were performed in a glass electrochemical cell. The electrolyte contained in the cell was continuously purged with argon. For the experiments performed at elevated temperatures a jacketed cell was used. Water from a thermostatically-controlled water bath was constantly circulated through the jacket to maintain the desired temperature.

The ring electrode of a RRDE can function as a quantitative in-situ detector for the soluble species generated by reactions on the disc electrode. RRDEs have been used to study the dissolution of magnetite of geological origin in various electrolytes [1]. The use of RRDEs allowed the quantitative measurement of both the Fe²⁺ ion dissolution flux (v_{Fe}^{2+}) and the Fe³⁺ ion dissolution flux (v_{Fe}^{3+}). The total iron release rate (v_{Fe}) is obtained by the addition of the two ion fluxes:

$$v_{Fe} = v_{Fe^{2+}} + v_{Fe^{3+}}$$
 (1)

Practical difficulties were encountered when attempting to use RRDEs at elevated temperatures. When a RRDE could not be employed, a RDE was used instead. In these cases v_{Fe} was calculated from the net disc electrode current observed. This requires a knowledge of the overall dissolution reaction, and use of Faraday's law.

During controlled-potential dissolution studies a potentiostat was used to control and systematically vary the potential of the magnetite electrode. At each value of potential investigated, the steady-state currents were measured. In this way, the relationship between potential and the steady-state dissolution rate was obtained point by point within the potential range of interest.

The dissolution fluxes of ionic species are expressed in chemical (nmol.cm⁻².s⁻¹) rather than electrochemical (μ A.cm⁻²) units. This dissolution flux is the rate of iron released to solution, and not rate of magnetite dissolution. Rates expressed in the former manner exceed those expressed in the latter manner by a factor of three. This factor arises from the stoichiometry of magnetite (Fe₃O₄).

The electrode surface areas used to calculate the dissolution fluxes and current densities were the simple geometric surface areas: no attempt was made to measure or correct for the effects of surface roughness.

Many individual controlled-potential experiments were performed using different electrode rotation rates and in electrolytes of different; composition, pH and temperature. In this way, this set of data allowed the determination of the effects of these parameters and potential upon the kinetics and mechanism of magnetite dissolution.

3. RESULTS

3.1. Influence of potential and electrolyte composition upon the kinetics of magnetite dissolution

The effects of potential upon the electrochemical behaviour of magnetite electrodes in both complexing and non-complexing electrolytes have been described in detail elsewhere [1]. Consequently, only a summary of the findings will be provided in this report.

3.1.1. Magnetite dissolution in non-complexing (perchlorate) electrolytes

Figure 1 indicates the dependence of v_{Fe}^{2+} for a magnetite RDE upon potential in a perchlorate electrolyte at a pH of 2.8 and at 20°C. The value of v_{Fe}^{3+} remained below detection limits within the potential range investigated, and v_{Fe}^{2+} could not be reliably



Figure 1. Potential dependence of the dissolution flux (v_{Fe}) generated by a magnetite RDE in a perchlorate electrolyte, pH = 2.8, 20±2°C, f = 33.3 Hz.

measured at potentials greater than 50 mV. The dependence of $v_{Fe^{2+}}$ upon potential exhibits three maxima at potentials of -100±10 mV (E₁), -970±10 mV (E₂) and -1100±10 mV (E₃).

Clearly, $v_{Fe^{2+}}$ is strongly dependent upon potential in a rather complex manner. The results of these experiments indicate that, within the potential range of -800 to 0 mV, the only Faradaic reaction of significance is the reductive dissolution of magnetite, equation (2).

$$Fe_3O_4 + 8H^+ + 2e^- \rightarrow 3Fe^{2+} + 4H_2O$$
 (2)

Under these circumstances, the iron in magnetite is released exclusively as ferrous ions, v_{Fe}^{3+} is zero, and v_{Fe} is equal to v_{Fe}^{2+} .

At potentials negative of about -800 mV, the hydrogen evolution reaction becomes significant, and it is likely that magnetite is reduced to metallic iron.

The experimental studies performed do not clearly identify the nature of the reactions that define E_{oc} for magnetite immersed in perchloric acid.

3.1.2. Magnetite dissolution in complexing (EDTA) electrolytes

The dependence of v_{Fe} upon potential observed during the dissolution of magnetite in EDTA electrolytes is very similar to that during dissolution in perchlorate electrolytes. However, the composition of the dissolution flux in EDTA varies markedly from that observed in perchlorate electrolytes. Figure 2, shows the dependence of v_{Fe} , v_{Fe}^{2+} and v_{Fe}^{3+} upon potential for a magnetite electrode in an EDTA electrolyte at a pH of 3.0 and at 20°C. In contrast to magnetite dissolution in perchlorate electrolytes, the dissolution flux is composed of both ferrous and ferric ions, and the relative proportions of which change as a function of potential. Detailed analysis of these results [1] reveals that, in EDTA-containing electrolytes, the Fe²⁺/Fe³⁺ couple generated by the dissolution process is, by virtue of a electron-transfer process, in equilibrium with the magnetite. This electron transfer process serves to define E_{oc} for magnetite in EDTA-containing electrolytes.

3.2. Influence of pH upon the kinetics of magnetite dissolution

Figure 3 contains a semi-logarithmic plot of the relationship between current density and potential observed during a number of studies of magnetite dissolution in perchlorate electrolytes of differing pH. It is apparent that at any given pH, the relationship between logarithm of current density and potential is linear on both the positive and negative sides of peak E_1 , with slopes of -80 ± 10 mV.decade⁻¹ and 170±10 mV.decade⁻¹, respectively. As discussed in a number of publications [1,2],



Figure 2. Potential dependence of the dissolution fluxes generated by a magnetite RDE in EDTA, pH = 3.0, $20\pm2^{\circ}$ C, f = 33.3 Hz. (o) Ferrous ion dissolution flux, v_{Fe}^{2+} ; (\oplus) ferric ion dissolution flux, v_{Fe}^{3+} ; (\oplus) total iron dissolution flux, v_{Fe} .



Figure 3. Potential dependence of the logarithm of the dissolution flux (v_{Fe}) generated by a magnetite RDE in perchlorate electrolytes as a function of pH, pH = 1.06 to 4.70, 20±2°C, f = 33.3 Hz.

these observations are consistent with the the theories of the dissolution of ionic solids developed by Engell [3] and Vermilyea [4]. In these theories the potential dependence of the dissolution rate is interpreted in terms of rate control by either anion (O^{2-} , OH^{-}) or cation (Fe²⁺, Fe³⁺) interfacial ion-transfer at potentials positive and negative of E₁, respectively. These theories indicate that linear relationships between logarithm of current density and potential will be observed when a single ion-transfer reaction is rate determining, and in the absence of mass-transfer effects.

From Figure 3 it is apparent that potentials less than E_1 , a single straight line satisfactorily describes the relationship between the logarithm of current density and potential at all values of pH investigated. This indicates that the kinetics of the iontransfer reaction involving dissolution of the iron cations is independent of pH. Conversely, at potentials greater than E_1 , the kinetics of the ion-transfer reaction involving dissolution of the oxygen-containing anions exhibits a strong pH dependence.

The quantitative relationship between pH and the rate of the individual rate determining reactions, free from mass-transport effects, was determined in the following manner. Straight lines, with the aforementioned slopes, were fitted to the current density data on the positive and negative sides of the peak centred at E₁. For all the experiments performed at different pH, extrapolated or interpolation of these lines allowed the evaluation of the current density at some fixed value of potential. This data was then assembled into a figure that expresses the effect of pH upon the kinetics of an individual rate-determining ion-transfer reaction at a constant potential, Figure 4. Consequently, each point on this figure was produced from an separate experiment. The magnetite electrode was repolished prior to each experiment. Recognizing the inherent inhomogeneities that must be present in geological specimens of magnetite, some of the scatter in the experimental data was undoubtedly due to variations in the properties of the magnetite electrodes. Additionally, as the pH was varied the total extent of magnetite dissolution varied by several orders of magnitude, and therefore changes in the surface state of magnetite were slight at high pH, but quite extensive at low pH. Consequently, the apparent dissolution rates of experiments performed at low pH may have been increased due to surface roughen effects.

To address the aforemention experimental limitations, a second investigation of the pH effects on the magnetite dissolution rate was performed using a different experimental strategy. From the many complete investigations of the effects on potential upon kinetics of magnetite dissolution, it was possible to identify the relatively narrow range of potential on the positive side of peak E_1 in which exhibited the linear relationship between current density and potential. Thus, using a single electrode, the polarization behaviour of magnetite was examined only in these restricted ranges of potential. Without removal of the electrode form the electrolyte, the pH as adjusted, and the electrochemical measurements repeated. In this manner the pH was first decreased



Figure 4. pH dependence of the logarithm of the dissolution flux (v_{Fe}) generated by a magnetite RDEs in perchlorate electrolytes, E = 0.1 V vs SCE, 20±2°C, *f* = 33.3 Hz.

from 4.8 to 1.3 in 10 steps, and then increased again to 4.6 in 5 steps. The results from this experiment are shown in Figure 5. The current densities, obtained using these two different experimental methodologies, are shown together in Figure 6.

From Figure 6 it is apparent that, in the pH range of 1.0 to 3.5, the reaction order with respect to hydronium ions for the charge-transfer reaction that is rate determining on the positive side of peak E_1 is unity. From Figure 3, it is apparent that within this same pH range, this reaction order for the rate determining reaction on the negative side of peak E_1 is zero.

3.3. Influence of mass transport upon the kinetics of magnetite dissolution

The use of RDE and RRDE electrodes allowed the investigation of the effects of mass transport of upon the kinetics of magnetite dissolution, by means of varying the rotation frequency of the electrode. The dependence of the current density upon potential and electrode rotation frequency (f) for a magnetite RDE in perchloric acid is shown in Figure 7. From this figure it is apparent that mass-transport effects are observed, the current density increases as f increases, but that these effects are limited to potentials close to E₁. The dependence of current density upon f, for potentials close to E₁, is further analysed by the methods described in Appendix A. Figure 8 shows the



Figure 5. pH dependence of the logarithm the dissolution flux (v_{Fe}) generated by a single magnetite RDE in a perchlorate electrolyte, E = 0.1 V vs SCE, 20±2°C, f = 33.3 Hz.



Figure 6. pH dependence of the logarithm of the dissolution flux (v_{Fe}) generated by a magnetite RDEs in perchlorate electrolytes, E = 0.1 V vs SCE, 20±2°C, f = 33.3 Hz. A combination of the data shown in Figures 4 and 5.



Figure 7. Potential dependence of the dissolution flux (v_{Fe}) generated by a magnetite RDE in a perchlorate electrolyte as a function of RDE rotation frequency, pH = 2.80, 20±2°C, f = 1.7, 3.3, 6.7, 13.3, 26.7, 53.3 Hz.



Figure 8. Dependence of the inverse of dissolution flux (v_{Fe}) generated by a magnetite RDE in a perchlorate electrolyte upon the inverse square root of RDE rotation frequency as a function of potential, pH = 2.80, 20±2°C, E = -85, -105, -125 mV vs SCE.

dependence of the inverse current density upon the inverse of the square root of electrode rotation frequency. The average value of the slope of the straight lines is $0.138 \text{ nmol}^{-1}.\text{cm}^2.\text{s}^{3/2}.$

3.4. Influence of temperature upon the kinetics of magnetite dissolution

Figure 9 contains a semi-logarithmic plot of the relationship between current density and potential observed during a number of studies of magnetite dissolution in perchlorate electrolytes of $pH = 2.9\pm0.1$ at differing temperatures in the range 2 - 81°C. An analysis, similar to that used to determine the pH-dependence, was used to determine the temperature-dependence of the MDR. Figure 9 illustrates the fitting of straight lines to the experimentally observed data a potentials > E₁. It should be noted in these instances the slope of the lines used is 3.12 RT / F, which at 25°C corresponds to 80 mV.decade⁻¹. Although, not shown on Figure 9, a similar analysis was performed the data at potentials < E₁. It is important to recognize that the activation energy is a function of both potential and temperature. Consequently, the value of the activation energy produced from a Arrhenius plot will depend upon the value of potential at which the dissolution rates were evaluated. The potentials used were selected to be within the potential ranges used for the fitting process.



Figure 9. Potential dependence of the dissolution flux (v_{Fe}) generated by a magnetite RDE in perchlorate electrolytes as a function of temperature, pH = 2.9±0.1, *f* = 33.3 Hz, temperature = 2, 25, 40, 61, 81°C.

4. DISCUSSION

4.1. Mechanism of magnetite dissolution

The semi-logarithmic representation of the dissolution flux dependence upon potential indicates that there are three different charge-transfer reactions involved in the MDR, and that identity of the rate limiting charge-transfer reaction changes as a function of potential. The kinetics of each of these three charge-transfer reactions will be described in turn, starting with the charge-transfer reaction that is rate determining at the most positive potentials, and ending that which is rate determining at the most negative potentials. For convenience, these three charge-transfer (CT) reactions will be designated CT-1, CT-2, and CT-3, respectively.

4.1.1. Charge-transfer reaction 1 (CT-1)

Reaction CT-1 is rate determining for the MDR at potentials greater than E_1 . The studies performed indicate that reaction CT-1 exhibits a reaction order of unity with respect to protons. For reaction CT-1 at 25°C, the relationship between the logarithm of flux and potential is linear with a slope of -80 ± 10 mV.decade⁻¹. The activation energy of reaction CT-1 was found to be 58 kJ.mol⁻¹ at a potential of 0 mV vs SCE. As discussed previously, reaction CT-1 is believed to be a ion-transfer reaction involving the dissolution of the oxide ions. In accord with the proposals of others [2], these observations suggest that the process CT-1 involves two consecutive reactions:

$$O^{2-}(ads) + H^{+}(aq) = OH^{-}(ads)$$
 (3)

$$OH^{-}(ads) + H^{+}(aq) \rightarrow H_2O(ads)$$
(4)

Equation (3) is a protonation-deprotonation equilibria. It is likely that under the acidic conditions used, the surface coverage of $OH^-(ads)$ will have obtained a limiting value. Consequently, the reaction order with respect to protons would be unity, rather than two. However, it would be anticipated that as the pH was increased, the reaction order of one would eventually change to two.

At potential close to E_1 , the overall rate of the MDR was under mixed control by both reaction CT-1 and a mass-transfer process. If it is assumed that the mass-transfer process involves protons, then a quantitative relationship can be developed that describes the influence of RDE rotation rate upon the dissolution flux, Appendix A.

This theoretically derived relationship indicates that a plot of $(v_{Fe})^{-1}$ vs $f^{-1/2}$ should yield a straight line with slope of 0.122 nmol⁻¹.cm².s^{3/2}. The average experimental observed value is 0.138 nmol⁻¹.cm².s^{3/2}, see Figure 8. The good agreement between experiment and theory strongly supports the assertion that the mass-transport process involves the transport of protons to the magnetite surface. Hence, these findings provide additional support for the reaction scheme described by equations (3) and (4).

4.1.2 Charge-transfer reaction 2 (CT-2)

Reaction CT–2 is rate determining for the MDR at potentials slightly less than E_1 . The studies performed indicate that reaction CT–2 exhibits a reaction order of zero with respect to protons. For reaction CT–2 at 25°C, the relationship between the logarithm of flux and potential is linear with a slope of 170 ± 10 mV.decade⁻¹. The activation energy of reaction CT–2 was found to be 69 kJ.mol⁻¹ at a potential of -300 mV vs SCE. As discussed previously, this charge-transfer is believed to be a ion-transfer reaction involving the dissolution of the iron cations. The simplest representation of these reactions are given in equations (5) and (6)

$$Fe^{2+}(ads) \rightarrow Fe^{2+}(aq)$$
 (5)

$$Fe^{3+}_{(ads)} \rightarrow Fe^{3+}_{(aq)} \tag{6}$$

4.1.3 Charge-transfer reaction 3 (CT-3)

While the studies performed provide a reasonably complete quantitative description of the kinetics of CT–3, the nature of this charge-transfer reaction has yet to be identified. The influence of pH upon the kinetics of this charge-transfer reaction have not been analysed in detail. However, preliminary indications are that this reaction exhibits a reaction order of one half with respect to the proton concentration. For reaction CT–3 at 25°C, the relationship between the logarithm of flux and potential is linear with a slope of -340 ± 50 mV.decade⁻¹. The activation energy of reaction CT–3 was found to be 41 kJ.mol⁻¹ at a potential of -600 mV vs SCE.

4.2. Quantitative model of the kinetics of magnetite dissolution

A complete description of the kinetics of the MDR can be achieved once the nature, and number, of elementary reaction steps that are involved in the overall reaction are determined. As has been demonstrated, there are three charge-transfer reactions, each of which becomes rate determining within certain potential limits. The rate of each of these charge-transfer reactions exhibits a unique dependence upon pH, potential and temperature. Additionally, mass-transfer of protons can also become rate determining under certain conditions. The kinetic study under taken, permits expressions to be developed for the rates of each of the charge-transfer reactions as a function of pH, potential and temperature. The rate of the mass transfer process can be expressed as a function of pH.

$$Log\{v_{ct-1}\} = 3.45 - pH - (E F) / ((1.35)(2.303 R T)) + 3042 (1/295 - 1/T)$$
 (7)

$$Log\{v_{ct-2}\} = -0.1 + (E + 0.3) F / ((2.70)(2.303 R T)) + 3614 (1/295 - 1/T)$$
 (8)

$$Log\{v_{ct-3}\} = 1.2 - pH/2 - (E + 0.6) F/((5.40)(2.303 R T)) + 2161(1/295 - 1/T) (9)$$

The information provide in Appendix A yields:

$$v_{\rm mt} = 5.16 \times 10^3 \ (f)^{1/2} \ \rm C_i$$
 (10)

The temperature dependence of the rate of the mass-transfer process has not been included in the quantitative treatment at this stage. Such an enhancement of the model is quite straightforward.

Using the standard methods for treating parallel and consecutive steps in a multi-step reaction, an expression for the dissolution flux (v_{Fe}) can be obtained in terms of the rates of the four elementary processes:

$$v_{Fe} = v_{ct-3} + ((v_{ct-1} v_{ct-2} v_{mt}) / ((v_{ct-1} v_{mt} + v_{ct-2} (v_{ct-1} + v_{mt}))))$$
(11)

Consequently, the combination of equations (7) - (11) yields an expression for v_{Fe} in terms of potential, temperature, pH and electrode rotation frequency. This expression is not given because it is rather unwieldy.

The use of equations (7) - (11) allows the generation of plots that illustrate the relationship between dissolution flux (v_{Fe}) and potential, Figure 11. This figure illustrates how the overall reaction rate is influenced by each of the four elementary



Figure 10. Arrhenius plots of the dissolution flux (v_{Fe}) generated by a magnetite RDE in perchlorate electrolytes as a function of potential, pH = 2.9±0.1, f = 33.3 Hz, E = 0, -300, -600 mV vs SCE.



Figure 11. Calculated potential dependence of the logarithm of v_{Fe} , v_{ct-1} , v_{ct-2} , v_{ct-3} and v_{mt} for a magnetite RDE in a perchlorate electrolyte, pH = 3.0, 25°C, f = 33.3 Hz.

reaction steps at 25° C, pH = 3.0, 2000 rpm. Similarly, these equations allowed the calculation of plots that illustrate the influence of pH, electrode rotation frequency, and temperature, upon the kinetics of the MDR, Figures 12 - 14, respectively.

A comparison of Figures 12 - 14, with Figures 3, 7 and 9, respectively, permits a comparison of the predictions of the dissolution model with experimental observations. Qualitatively, the predictions are in good accord, suggesting that the the basis for the model is sound. Additionally, there is reasonable quantitative agreement between the models predictions and experimental observations. This model will be subjected to further refinement in order to improve the reliability of its quantitative predictions.

It is important to recognize the limitations of this model. The limits on the values of the parameters, that can be meaningfully used in the dissolution model, are given in Table 1.

The experiments described in this work were all performed in perchlorate electrolytes. Other studies [1] have demonstrated that the presence of complexing species, such as ethylenediaminetetra-acetic acid (EDTA), at millimolar concentrations, do not significantly alter the kinetics of the dissolution reactions. Consequently, these results should be generally applicable to acidic electrolytes, if the concentration of complexing species does nor exceed the millimolar level.



Figure 12. Calculated potential dependence of the logarithm of the dissolution flux (v_{Fe}) for a magnetite RDE in perchlorate electrolytes as a function of pH, pH = 1.0, 2.0, 3.0, 4.0, 25°C, f = 33.3 Hz.



Figure 13. Calculated potential dependence of the dissolution flux (v_{Fe}) for a magnetite RDE in a perchlorate electrolyte as a function of RDE rotation frequency, pH = 3.0, 25°C, f = 1.7, 3.3, 6.7, 13.3, 26.7, 53.3 Hz.



Figure 14. Calculated potential dependence of the logarithm of dissolution flux (v_{Fe}) for a magnetite RDE in perchlorate electrolytes as a function of temperature, pH = 3.0, f = 33.3 Hz, temperature = 2, 25, 40, 61, 81°C.

Table 1.	Limits on the	values of the	parameters that	t can	be use	d in the	MDR	rate
	eq	uation derived	I from equations	s (7) -	(11).			

Parameter	Lower limit	Upper limit		
Potential (mV vs SCE)	-800 mV	0 to 200 mV		
pH	1	4		
Temperature	2°C	80°C		
Electrode rotation rate	100 rpm	3200 rpm		

In the model developed, the rates of mass-transfer processes are described in terms of the rotation frequency of a RDE. Under the conditions in which the experiments were performed the electrolyte flow at the surface of the magnetite disc electrode is laminar. Consequently, the quantitative treatment of the mass-transfer process would have to be modified in order use the model to predict the rate of magnetite dissolution from pipe walls subject to turbulent flow.

Scanning electron microscopy was performed on the magnetite electrode surfaces at the conclusion of a number of dissolution experiments. The surface roughening of magnetite electrodes, particularly in experiments performed at low pH and high temperature, was considerable. As described in this report, efforts were made to minimize the effects of surface roughening during the investigation of the kinetics of the MDR. Hence, the rate information provided in this paper, that is normalized per unit surface area , is likely to be reasonably accurate.

When contemplating the application of this dissolution model to magnetite dissolution processes during decontaminations, it is necessary to address the issue of quantifying the magnetite surface area. It is evident that, in addition the the difficulties involved in estimating the initial surface area of magnetite, the surface area is likely to significantly altered by roughening processes caused by dissolution. The uncertainties in the values of these surfaces areas is likely to be much greater than the uncertainties in the kinetic parameters employed in the model.

5. CONCLUSIONS

The results presented in this paper serve to emphasize the electrochemical nature of the MDR. Electrochemical techniques were used to determine the influence of temperature, pH, potential and RDE rotation frequency upon the kinetics of the MDR. The findings of this study are both internally consistent, and consistent with other published results [1,2]. The mechanism of magnetite dissolution was identified, permitting the development a quantitative model of the kinetics of the MDR. The predictions of this model are shown to be in good accord with experimental observations. The development of this model is an important step towards the goal of achieving a quantitative model of decontamination processes.

APPENDIX A: MASS-TRANSPORT LIMITED REACTIONS AT ROTATING DISC ELECTRODES.

The Levich equation describes current observable at a disc electrode in the masstransfer-limited condition.

$$i_{mt} = 6.2 \times 10^{-4} \text{ n F } (D_i)^{2/3} (2 \pi f)^{1/2} (v)^{-1/6} C_i$$
 (A.1)

Symbol	Meaning	Units
i _{mt}	Mass-transport limited current density	A.cm ⁻²
Vmt	Mass transport limited flux	nmol.cm ⁻² .s
V _{ot}	Charge-transfer limited flux	nmol.cm ⁻² .s
V _{total}	Total flux	nmol.cm ⁻² .s
n	Equivalents per mole	equiv.mol ⁻¹
F	Faraday constant	C.equiv ⁻¹
Di	Diffusion coefficient of species i	cm ² .s ⁻¹
f	Rotation frequency	revs.s ⁻¹
v	Kinematic viscosity	cm ² .s ⁻¹
C:	Bulk concentration of species i	mol.dm ⁻³

Table A1. Definition of symbols and units used in the equations given in Appendix A.

Using Faraday's Law, this equation can be readily reformulated to provide an expression for the mass-transport limited flux of species i:

$$v_{mt} = 6.2 \times 10^5 (D_i)^{2/3} (2 \pi f)^{1/2} (v)^{-1/6} C_i$$
 (A.2)

At 25°C, the kinematic viscosity of water is approximately 10^{-2} cm².s⁻¹. Hence, the general expression for the mass transport limited flux of species i, in dilute aqueous solutions at 25°C, is as follows:

$$v_{mt} = 3.35 \times 10^6 (D_i)^{2/3} (f)^{1/2} C_i$$
 (A.3)

The diffusion coefficient of protons in water at is $9.31 \times 10^{-5} \text{ cm}^2 \text{.s}^{-1}$ at 25°C . Hence, for a perchlorate electrolyte at a pH of 2.80, the mass transport limited flux of protons is given by:

 $v_{\rm mt} = 10.9 \ (f)^{1/2}$ (A.4)

It is necessary to consider the relationship between the value of v_{mt} calculated for proton diffusion to the electrode, and the values of v_{Fe} that are experimentally observed. As discussed in the Section 4, the rate determining reaction at potentials greater than E_1 is given by equation (4). From equation (2), it is apparent that the rate determining reaction must occur four times in order to release three ferrous ions, leading to a stoichiometric factor of 0.75. Consequently, the mass transport limited value of v_{Fe} is given by the following expression:

$$v_{\text{Fe-mt}} = 8.2 \ (f)^{1/2}$$
 (A.5)

For a electrochemical reaction that involves consecutive reaction steps that are mass-transport and charge-transfer controlled, v_{total} is related to the v_{mt} and v_{ct} in the following way:

$$1 / (v_{total}) = 1 / (v_{ct}) + 1 / (v_{mt})$$
 (A.6)

Consequently, for the case of the dissolution flux arising from magnetite dissolution (v_{Fe}) , the use of equations A.5 and A.6 leads to:

$$1 / (v_{Fe}) = 1 / (v_{Fe-ct}) + 0.122 f^{-1/2}$$
 (A.7)

Consequently, if the assumptions concerning the identity of the species involved in the mass-transfer processes, and the value of the stoichiometric factor, are correct, then a plot of $(v_{Fe})^{-1}$ vs $f^{-1/2}$ should yield a straight line with slope of 0.122 nmol⁻¹.cm².s^{3/2}. The intercept of this plot yields the value of v_{Fe} that would be observed in the absence of mass-transfer effects.

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ELECTRODECONTAMINATION OF TOOLS AND INTERNAL SURFACE OF METAL PIPE

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Abstract

base of the IAEA Research Contract No. 5762/RB. On the demonstration decontamination installations should be set up and of their performance for real contaminated samples from test nuclear facilities should be conducted. During this year, tow of electrodecontamination installations were set up and types some performance experiments were carried out with real contaminated samples coming from nuclear facilities. One is the batch electrodecontamination device, another is internal surface decontamination device for metal pipes. The former is used for small items, such as hand tools, valve core. It is comprised of oblique plane clarifier. DC supply unit and electrolyzer. ventilation system. During decontamination, iron hydroxide will be produced. The maximum yield is about 7 mg/cm².min (dry). Its precipitation rate is 0.13 mm/s. After clarifying, it can be the electrolyte without any visual effects reused as on parameters decontamination. The reference operation are as follows: electrolyte... 20% NaNO3, current density... 0.3 - 0.6 A/cm^2 , electrode separation...20 - 100 mm, circulation rate of electrolyte... 10 1/min, temperature... 20 - 55 °C. With those parameters, the surface contamination level can be reduced from 0.6 - 1240 Bq/cm² to lower than 0.08 Bq/cm² within 5 minutes.

The internal surface decontamination device is comprised rotation anode (article to be decontaminated), movable of cathode, circulation system of electrolyte and DC supply unit. The purpose of developing this device is to decontaminate the pipe contaminated with plutonium. in order to dispose them into shallow land repository after decontamination. The experiment result showed that under the given condition, 7 - 8 cm of pipe could be treated. after decontamination, the contamination level of internal surface could be reduced to background for fission product contaminated pipe taken from nuclear facilities. But it is needed to demonstrate this device with plutonium contaminated articles in situ. The reference operation parameters were as follows: electrolyte... 20% (wt) NaNO3, current density... 0.2- 0.6 A/cm^2 , circulation rate of electrolyte... 0.3 - 0.5 $1/cm^2$.min, electrode separation... < 5 mm, temperature... 20 - 80 °C.

1. Introduction

The contaminated articles produced during nuclear activities or decommissioning of nuclear facilities can be categories according their external shapes: divided into three pipe. In order to reduce the surface small article. plane and contamination level much as possible so that to make them as reusable or acceptable for shallow land disposal. simple and efficient decontamination devices are needed. Based on the results obtained under the IAEA Research Contract No. 5762/RB, simple decontamination devices were expected to meet those requirements.

According to the IAEA Research Contract No. 5762/R1/RB. tow types of devices were developed in the last year that will be used to decontaminate the small items and the internal surface of metal pipe. Necessary data and experiences will be gained with the decontamination experiments. The purpose of the experiments are to demonstrate the feasibility of those devices developed, and to provide designing base for practical devices.

2. Device Description

2.1 Batch Electrodecontamination Device This device is comprised of electrolyzer, oblique plane clarifier, ventilation system and DC supply unit (see Fig. 1) Electrolyzer... 500 X 500 X 700 mm Oblique plane clarifier... 500 X 500 X 700 mm Length of oblique plane... 400 mm Gap between the oblique planes... 25 mm Oblique angle... 60° DC supply unit... 200 A/0 - 24 V Ventilation casing... 700 X 700 mm

2.2 Internal Surface Electrodecontamination Device

This device is comprised of rotation anodic cathode (to be treated). horizontal movable cathode, circulation system of electrolyte and DC supply unit (see Fig. 2).



Fig. 1 Batch decontamination device diagram

1. electrolyzer, 2. Oblique plane clarifier 3. Precipitation pool



Fig. 2 Internal surface decontamination device

2.2.1 Anode

In order to simplify the sampling and weighing, the specific anode was designed (see Fig. 3). It is comprised of tow buck pipes made of carbon steel. It is 500 mm of length. On the internal surface of the anode, there are three annular notch, used for installation and fixation the testing samples. The testing samples could be carbon steel or stainless steel.

2.2.2 Movable Cathode

Cathode was made of brass. On the side surface of the front end, 120 holes with diameter 1 mm were distributed homogeneously. Electrolyte sprayed through them onto the internal surface of pipe. On the front and back of cathode, one and tow



Fig.3 Anode structure diagram

guide means were fixed, to keep the correct position of cathode and spray direction of electrolyte, also to keep from direct touching of cathode and anode. Anode was fixed with anode bar. It can move horizontally, carried by leading screw with designed speed.

2.2.3 Circulation System of Electrolyte

Electrolyte is pumped to anode from storage tank, then spraying onto internal surface of pipe. flow out from one end of the pipe to collection tray, coming back to oblique plane clarifier. Treated electrolyte stored in storage tank.

2.2.4 Basic mechanical Parameters of Testing Rotation speed of anode.... 30 r/min Movement speed of cathode.... 70 mm/min Cathode diameter.... 34.3 mm Cathode surface.... 30.66 cm² Anode I Øⁱⁿ 37 mm, electrode separation 2.7 mm Anode II Øⁱⁿ 43 mm, electrode separation 5.7 mm Anode III Øⁱⁿ 53 mm, electrode separation 10.7 mm

3. Experiment Results and Discussion

3.1 Experiment Results of Batch Electrodecontamination Device

3.1.1 Clarifying Efficiency of the Oblique Plane Clarifier

The size of iron hydroxide produced during so subside electrodecontamination process larger, is down quickly, test results showed that when the clarity was 70%, the subsidence rate of the minimum particle size was 0.13 mm/s. When current density was 0.6 A/cm^2 , electrode separation was 20 mm, the maximum yield of iron hydroxide was 7 mg/cm².min (dry). Water contents of the iron hydroxide subsided naturally was 81%. When the length of oblique gap between oblique plane was 400 mm, planes was 25 mm, oblique angle was 60°, flowrate of electrolyte was 10 1/min, the contents of iron hydroxide in the electrolyte treated would be lower than 15% of original. No visual effect was fond on electrodecontamination results, when treated electrolyte was reused.

3.1.2 Electrodecontamination Results

The reference operation parameters are as follows: Electrolyte... 20% (wt) NaNO3 Current density... $0.3 - 0.6 \text{ A/cm}^2$ Electrode separation... 20 - 100 mmCirculation rate of electrolyte... 10 l/minTemperature... 20 - 55 °CUnder those parameters, 125 pieces of hand tools a

Under those parameters, 125 pieces of hand tools and valve cores were decontaminated. The surface contamination level was reduced from 0.6 - 1240 Bq/cm² to lower than 0.08 Bq/cm² within 1 - 5 minutes, among them. considerable parts reached background level.

The experiment results showed that with the simple device, small contaminated items could be cleared sufficiently. It also showed that location of the items to be decontaminated should be arranged carefully upon their size and shape in the electrolyzer, to make the electronic field as homogeneously as possible.

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Otherwise some position of the item would be impacted seriously. If necessary, special cathode should be designed. For decommissioned items it need not do so, but when arrangement was unreasonably, more iron hydroxide would be produced.

Besides from those, the size and amount of the items will be limited by the size of electrolyzer and power of DC supply unit, so they must be designed according to the real items to be treated.

3.2 Experiment Results of Internal Surface Decontamination Device

3.2.1 Effects of Electrolyte Flowrate on Current Density

When anode I was used, the results were showed in table 1. For anode II with guide means of 40 mm and anode III with guide means of 50 mm, the results were showed in Fig.5.

Table	1.	Relation	of	Current	Density	with	Electrolyt	e F	lowrat	e
-------	----	----------	----	---------	---------	------	------------	-----	--------	---

flowrate		current densi	ty
(1/m1n)	4 V	6 V	8 V
6. 67 10. 00 13. 33 16. 67 20. 00 23. 33 26. 67 30. 00	0.042 0.042 0.055 0.055 0.055 0.055 0.055 0.055 0.055	0.292 0.292 0.292 0.292 0.292 0.292 0.292	0.583 0.583 0.583 0.583 0.583 0.583 0.583 0.583 0.583 0.583





1. Guide means 2. nut locking

The results indicated that when the gap between anode and cathode was narrow enough (for example for anode I the gap was 2.7 mm). electrolyte could fill up the gap completely, so current density was not affected by electrolyte flowrate. If the gap was wide, a proper couple of guide means should be adopted, to make current density a constant value with lower electrolyte flowrate The results also indicated that If the current density was constant, the metal removal rate would be a constant (see table 2)


Fig. 5 Relation of electrolyte flowrate with current density (Anode II, III)

Table 2. Metal Removal Rate Under Various Electrolyte Flowrate

flowrate (1/min)	voltage (V)	current density (A/cm²)	MRR (mg/cm ² .min)
6.67	8	0. 55	1.157
10.00	8	0.55	1.110
13.33	8	0.55	1.121

* electrode separation... 2.7 mm

3.2.2 Relation of Current density with Voltage

The relationships of current density with voltage for anode I and II were showed in Fig.6. The voltage could be estimated with voltage known for certain diameter of pipe. If the voltage needed closes to or exceeds the range of adjustable voltage, suitable cathode should be designed again.

3.2.3 Metal Removal Rate

Fig.7 showed the metal removal rate at various current II. density for anode I and The results indicated that metal removal rate depended on the electrode separation strongly at the same current density, comparison with batch electropolishing, the lower by 2 - 5 times. In order to obtain metal removal rate was higher DFs with lower current density, the electrode separation should be kept as near as possible. In general case, it should be less than 2.7 mm.



Fig. 6 Relation of current density with voltage



Fig.7 Relation of metal removal rate with current density

3.2.4 Surface Status

The surface was smooth and glossy after decontamination, it was much better than that of batch decontamination. It was mainly because that the gap between electrodes was much narrow than that used in batch cleaning, the anode rotated and cathode moved, that ensured the electrolysis uniformity.

3.2.5 Reference Operation Parameters

From the experiment results, following reference operation parameters were recommended.

Electrolyte... 20% (wt) NaNO3 Current density... $0.2 - 0.6 \text{ A/cm}^2$ Electrode separation... < 5 mm Flowrate of electrolyte... $0.3 - 0.5 \text{ l/cm}^2$.min

Temperature.... 20 - 80 °C

If there was greasy dirt on the surface, following step should be adopted:

Solution.... 20 g/l NaOH + 30 g/l Na₃PO₄ + 40 g/l Na₂CO₃ + 2 g/l Na₂SiO₃ Temperature... 80 - 90 $^{\circ}$ C Duration.... 30 - 50 min

3.2.6 Decontamination Results

To make the samples with fission product contaminated items from nuclear facilities, then fixed onto the annular notch of anode I. under the reference operation parameters to decontaminate them. The surface contamination level could reach background level, with cathode one way movement.

4. Conclusions

. Oblique plane clarifier was added into the electrolyte circulation systems of both batch and internal surface decontamination device. Iron hydroxide in electrolyte used could be cleaned to required level. That made electrolyte reusable and no visual effect on decontamination results.

. Batch decontamination device is suitable for small items. It is simple construction and practical. The demonstration test showed that under reference operation parameters, design requirement could be reached.

. Sufficient decontamination results could be obtained for fission products contaminated items with internal surface decontamination device. This device is developed for decontamination of plutonium contaminated pipe, to make it acceptable after decontamination for shallow land repository, so it is needed to demonstrate with real plutonium contaminated pipe in situ in the further.

ACTIVITY INVENTORY OF THE BIOLOGICAL SHIELDS OF THE FINNISH NUCLEAR REACTORS

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Abstract

As part of the decommissioning studies for the four Finnish nuclear reactors (two 465 MWe VVER-440 type PWRs at Loviisa and two 735 MWe BWRs at Olkiluoto) the activation products inventories of the biological shields of the reactors have been estimated. First, material data were updated and improved by analyzing samples taken from the bioshields. Then the radial irradiation flux distribution was calculated with the one-dimensional ANISN transport code using cross sections taken from the BUGLE-80 data library. Finally, the activation and the subsequent cooling of the bioshield materials were calculated with the wellknown ORIGEN-S program using its original LWR data libraries. Due to the different geometries (shielding) the radioactive inventory at the inner surface of the biological shield of a Loviisa unit will be about two orders of magnitude greater than that of an Olkiluoto (TVO) reactor. In the Loviisa bioshields H-3, Fe-55, Co-60 and Ni-63 are the most important radionuclides at short cooling times. In the long-term the activity will be dominated by Ca-41 and to a smaller extent C-14, Cl-36, Ar-39 (if it remains in the concrete) and Ni-59. In the bioshield concrete of the TVO reactors H-3 and Ar-39 are very dominant and there also Eu-152 is of relatively great importance. In the latter case the activation of the reinforcing bars must be taken into account, too. Of the nuclides mentioned above only Co-60 and Eu-152 are gamma-ray emitters. The calculations reported in this article were preliminary and approximate. There are many possibilities to improve the accuracy and reliability of activity inventory estimations, for instance the more precise definition material compositions and geometry, the use of more sophisticated computer codes and updated data libraries for activation flux calculations and the generation case-dependent activation cross section sets for final activity inventory calculations.

1. INTRODUCTION

At present, there are four nuclear power reactors in Finland. The Loviisa nuclear power plant consists of two VVER-440 type, 465 MWe PWRs. The units started commercial operation in 1977 and 1981, respectively. The plant is owned and operated by Imatran Voima Oy (IVO). The other two reactors are located at Olkiluoto. These 735 MWe BWRs of Swedish origin are owned and operated by Teollisuuden Voima Oy (TVO). Their commercial operation was started in 1979 and 1982, respectively.

The Finnish nuclear power companies have carried out decommissioning studies since the beginning of the 1980s. The companies presented their first decommissioning and dismantling plans to the safety authorities in 1987. In case of the Olkiluoto plant a more detailed plan was compiled in 1990. According to the present regulatory rules the Finnish power companies must update their decommissioning plans in every five years. The next versions must be presented to the authorities before the end of 1993.

The first comprehensive series of calculations for estimating the activity inventories of decommissioning waste of the Finnish nuclear reactors were carried out in 1988-1989 at the Nuclear Engineering Laboratory (YDI) of the Technical Research Centre of Finland (VTT)

[1,2]. The compositions of the bioshield materials were analyzed at the same time [3]. A few complementary studies have been performed later [4,5]. The activity inventories are taken into account in dose rate and radiation protection calculations and in safety assessments of the final disposal of the decommissioning waste, which are either already going on or will be started in the near future.

In this report, activity inventory calculations of the bioshield materials of the Finnish nuclear reactors are briefly described. At first, studies carried out for updating material data are discussed. Then, the methods used to calculate the irradiation flux and activity inventories are outlined. The main results are given and finally plans to improve the accuracy and reliability of activity inventory calculations are discussed.

2. UPDATING OF MATERIAL DATA

The main constituents of the fresh bioshield materials of the Finnish nuclear reactors have been reported quite well in pouring protocols and other documents. However, the concentrations of various trace elements were not measured during the construction. Therefore, as the first stage of this study the concentrations of fourteen elements (H, Li, B, N, Cl, K, Ca, Co, Ni, Cs, Ba, Sm, Eu, U) were measured using various analytical techniques as appropriate [3]. The composition of the bioshield material was considered to be uniform. The samples from the TVO reactors were taken from the upper, unirradiated part of the bioshield and the samples from Loviisa were made during the construction. Concrete samples were ground and homogenized and rebar samples were chipped before analysis.

The contents of Li and B in concrete samples and the contents of Li, B, K, and Ca in rebar samples were analyzed by atomic absorption spectrophotometry. Nitrogen contents in concrete were analyzed by Kjeldahl-method. The concentrations of hydrogen were derived from measurements of water content in concrete using thermogravimetric analysis.

Other elements were analyzed using instrumental neutron activation analysis. A computer program STOAV 84 [6] was used to calculate the results.

Table I shows the results of chemical analyses with error limits. When the content of element is below the detection limit of the analytical method, error limits have been omitted.

There were marked differences in the concentrations of H, Ni and K in the concrete samples from the two reactors. The material from Loviisa had a hydrogen content (15200 ppm), which is higher than that of the TVO sample (4600 ppm) by a factor of four; this was due to the presence of serpentine minerals with chemically bound water in the Loviisa samples.

When compared with other studies, e.g. [7], it has been concluded the concentrations of the most important elements in the bioshield materials of the Finnish nuclear reactors are now known from the point of the activation inventory calculations. The main uncertainty concerns the actual water content of the concretes, because the samples were not from the irradiated parts of bioshields.

3. CALCULATION SYSTEM

3.1. Neutron flux calculations

3.1.1. Programs and data libraries

Out-of-core flux distributions for the Loviisa and Olkiluoto reactors are normally calculated with the REPVICS program system, which was originally developed to estimate fast

	TVO(BWR)	IVO(PWR)	TVO (BWR)
	Concrete	Concrete	Rebar
Element	ppm	ppm	ppm
Н	4600 ± 230	15200 ± 2280	-
Li	23 ± 3	3.6 ± 0.4	< 0.5
В	2060 ± 103	2000 ± 100	240 ± 12
Ν	33 ± 3	33 ± 3	180
Cl	58	87	-
Κ	16900 ± 2800	< 400	< 0.5
Ca	75000 ± 9000	76000 ± 7000	30 ± 3
Co	22 ± 1	89 ± 3	98 ± 3
Ni	< 20	1800 ± 100	305 ± 10
Cs	4.2 ± 0.5	0.2 ± 0.05	< 0.5
Ba	930 ± 30	320 ± 40	< 40
Sm	4.0 ± 0.4	0.6 ± 0.1	< 0.08
Eu	0.74 ± 0.05	< 0.05	< 0.06
U	<u>15.6</u> ± 0.6	6.7 ± 0.9	< 2

neutron fluences in the reactor pressure vessels of the Loviisa reactors [8]. For decommissioning studies, only the one-dimensional branch of the package, the well-known ANISN code [9], has been utilized up to now. The cross sections have been taken from the BUGLE-80 library, which contains the coupled neutron and gamma constants for many nuclides and elements in 47 neutron and 20 photon groups [10]. Detailed descriptions of flux calculations for decommissioning studies can be found elsewhere [11,12].

The accuracy of the REPVICS code system has been checked against many measurements. One of the most interesting studies concerned samples scraped from the inner and outer surface of the pressure vessels of both Loviisa units. Some preliminary comparisons have also been made with data measured at the LR-0 zero power reactor in Řež (Czech Republic) [13]. All studies indicate that the calculation system is able to produce reliable results for fast fluxes and spectra up to at least the cavity outside the pressure vessel. The differences between calculated and measured values are less than 20%; in many cases the agreement is much better [14].

The suitability of the REPVICS code system to decommissioning calculations may, however, be doubtful. With the BUGLE-80 cross sections one may find it difficult to calculate accurately thermal absorption rates, which are of greatest importance in activity inventory calculations. The reason is that the BUGLE cross sections do not take into account upscattering of the thermal neutrons.

3.1.2. Geometry

The radial flux distributions were calculated in a one-dimensional cylinder geometry. In Tables IIa and IIb there are shown the geometric description of the Loviisa and Olkiluoto reactors used in the ANISN calculations. The volume between the components was assumed to be filled with water in the pressure vessel and with air outside the vessel.

3.1.3. Neutron source distributions

The activation of the reactor components and structures outside the active core is caused by neutron leakage, which may depend quite strongly on the core loading scheme. It may be difficult and also unreasonable to try to simulate actual irradiation conditions during the whole lifetime of a reactor. A better option is to choose a typical fuel cycle and to use its average neutron source distribution in irradiation flux calculations. This approach was applied in this study. However, the active cores of the Loviisa units were reduced in 1980-81 by replacing 36 fuel assemblies in peripheral core positions by shield assemblies made of steel. Afterwards, the fuel loading scheme was changed from the original Out-In-In scheme to a (very) low-leakage scheme. The effects of these two changes on the activation of the reactor components and structures in the radial direction were studied, too.

3.2. Activity inventory calculations

Radioactive inventory calculations were performed with the ORIGEN-S code of the SCALE-3 program package with its own data libraries [15,16]. The LWR data libraries with the three-group cross sections are intended for calculations in or near the reactor core. Their suitability for activation calculations in bioshield conditions may be somewhat doubtful. The accuracy of the ORIGEN-S flux weight factors condensed from the results of the ANISN calculations is not ascertained.

Component	nt Radius (cm)				
	Inner	Outer			
Core region	0.	137.0			
Shield elements					
Baffle					
Basket	150.5	154.0			
Barrel	155.5	161.5			
Cladding of the					
pressure vessel	177.1	178.0			
Pressure vessel	178.0	192.0			
Thermal shield	223.0	233.0			
Biological shield					
- steel plate	237.0	238.2			
- serpentinite concrete	238.2	305.8			
- steel plate	305.8	307.0			
- common concrete	307.0	590.0			

TABLE IIa. CYLINDRICAL RADIAL GEOMETRY IN THE ACTIVITY INVENTORY CALCULATIONS OF THE LOVIISA REACTORS [1].

Component	Radius (cm))
	Inner	Outer
Core region	0.	195.0
Moderator tank	214.5	217.0
Cladding of the		
pressure vessel	277.0	277.3
Pressure vessel	277.3	291.0
Thermal shield ¹		
- inner steel plate	313.9	313.95
- rockwool	314.0	331.0
- outer steel plate	333.6	334.0
Biological shield		
- zone A	343.0	350.0
- zone B ²	350.0	352.4
- zone C ³	352.4	355.6
- zone D	355.6	520.2
- zone E ³	520.2	523.4

TABLE IIb. CYLINDRICAL RADIAL GEOMETRY IN THE ACTIVITY INVENTORY CALCULATIONS OF THE TVO REACTORS [2].

¹ The thermal insulation was treated as air in the flux calculations ² Concrete: 86.92 vol%, Reinforcing bars: 13.08 vol%

³ Concrete: 77.14 vol%, Reinforcing bars: 22.86 vol%

The irradiation time was assumed to be 30 years (36 annual cycles with an average load factor of ca. 85%) for the Loviisa reactors and 36 years (40 annual cycles with an average load factor of 90%) for the TVO reactors. The choices were based on the design lifetimes of the reactors and their actual load factors.

4. MAIN RESULTS

Total and nuclide-wise activities at different points and regions of the bioshields of the Finnish nuclear reactors as a function of the cooling time were the main results of this study. Figures 1-2 are presented here as three typical examples of the results. They contain information on the activation products inventory at or near the inner surface of the bioshields.

Due to geometric (shielding) differences the specific activity at the inner surface of the bioshield of a Loviisa unit will be about two orders of magnitude greater than that of a TVO reactor. To some extent the nuclide-wise distributions are different, too. In the Loviisa bioshields H-3, Fe-55, Co-60 and Ni-63 are the most important radionuclides from the point of view of a prompt dismantling, which at present is the preferred option for the Loviisa reactors. In the long-term the activity will be determined by Ca-41 and to a smaller extent C-14, Cl-36, Ar-39 (if it remains in the concrete) and Ni-59. In the bioshield concrete of the



Figure 1. Inner surface activity in the bioshields (serpentine concrete) of the Loviisa reactors /5/.



Figure 2. Inner surface region activity in the bioshields of the Olkiluoto reactors; A) Concrete B) Reinforcing bars /5/.

TVO reactors H-3 and Ar-39 are very dominant in the successive time periods and there also Eu-152 is of relatively great importance. In this case the activation of the reinforcing bars must be taken into account. The specific activity of the bars shown in Fig. 2B corresponds to the average conditions in Zone B (see Table IIb). Co-60 is the most important nuclide also in case of the reinforcing bars.

Of the nuclides mentioned above only Co-60 and Eu-152 are gamma-ray emitters. These nuclides determine the external dose rate and radiation shielding requirements during the decommissioning. However, from the point of the safety of the final disposal also the beta-emitting nuclides can be important.

5. DISCUSSION

The radioactive inventories of the biological shields of the Finnish nuclear reactors have been estimated in a straightforward, but approximate way. The present knowledge on the compositions of the bioshield materials may be satisfactory mainly because of the measurements described in this report. A few checks against other studies indicate that no significant elements have been overlooked. However, the actual water content in the activated regions of the bioshields and the homogeneity of the materials are still uncertain.

The neutron flux calculations might be the weakest phase of our procedure, even if the methods applied were rather similar to those used in other countries [17-22]. To use a onedimensional code when trying to estimate neutron flux levels quite far from the source region is certainly not the best possible decision and the BUGLE cross sections may still worsen the situation. Our calculation system is, however, updated and improved continuously. Rapidly increasing processing speed of computers enables also the more accurate definition of input data (the geometry, the operation history etc.) and the application of more sophisticated methods (including even the Monte Carlo technique [23].

The original three-group cross sections of the ORIGEN-S code used in this study have been generated in the active core conditions, which may differ from those in the bioshields. A better option would be to utilize results of accurate neutron flux and spectrum calculations to create a case-specific cross sections for the most important activation reactions, the number of which is actually rather small [17,21].

In conclusion, even if our present calculation system may produce results, which are of the correct order of magnitude, a possibility to verify calculated activities against measured values would be of great importance. In the case of an operating reactor, however, such measurements are difficult. Therefore, well-defined benchmarks, against which one could verify his or her calculation system would be of great importance [17].

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DEVELOPMENT OF CUTTING TOOLS FOR THE DISMANTLING OF NUCLEAR FACILITIES

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Abstract

The purpose of this study, carried out by the Atomic Energy Commission - Nuclear Installations Dismantling Unit (CEA-UDIN), is to test cutting apparatus and tools suitable for use in dismantling operations on CEA sites and to do such development work as is necessary to make these apparatus and tools more efficient and better adapted to remote control operation.

The work, carried out over the last three years, can be split into two categories :

 comparative trials of five tools carried out in standardized inactive conditions : (hacksaw) recuperating saw, grinder, plasma torch, arc air and arc saw.

Comparisons have been made of performances and the production of secondary wastes (in mass and grain sizes). Improvements envisaged concern, mainly, the plasma torch and the arc saw.

* development of two tools for concrete cutting :

-a diamond saw used in the stage 3 dismantling of the AT1 installation at La Hague for the remote controlled cutting of a 200 mm thick reinforced concrete wall. The work was completed with good results.

-a shot blasting machine intended for the decontamination of plain or resin coated concrete walls and of stainless steel cladding of rooms in the AT1 installation. The active trials have proved positive.

I INTRODUCTION

The present report relates to the research agreement 5698/CF between the IAEA and the CEA-UDIN in the ambit of the second phase of the Research Programme on Decontamination and Dismantling of Nuclear Installations. The work, detailed below, has taken place over the last three years and concerns those cutting tools which have been found to be essential in dismantling operations.

In fact, these cutting tools enable us particularly to :

- * take samples.
- * cut up components so as to allow them to be removed from their original locations and to be packed into standard storage containers,
- * open up more or less inaccessible places,
- * decontaminate surfaces by mechanical ablation.

These operations have to be carried out so as to minimize occupational dose rates, in the minimum time (setting up, actual cutting, cleaning up, packing up, maintenance, etc.) and with the minimum production of technological and secondary wastes. The following criteria thus apply to the tools :

*	efficiency :	cutting speed, reliability, lifespan,
	*	

* waste products : quantity and grain size of secondary wastes,
 * ease of use : tool carrier interface constraints, suitability

The objective of our study is to test the tools which are usable on CEA dismantling sites and to develop them, as necessary, to correspond as closely as possible to the above criteria.

for remote control.

The first part of the present work programme involves comparative testing, in inactive conditions, of metal cutting equipment. The second part concerns developments carried out and results obtained in active conditions from two tools for concrete cutting.

II COMPARATIVE TRIALS OF METAL CUTTING TOOLS

II.1 Preamble

This work is part of the research programme on the decommissioning of nuclear installations, the costs of which are shared by the European Community - Action No. 3 "Dismantling Techniques : Contract CCE/CEA No. F12D0013F", on which the final report is due to appear in 1993.

The aim of these trials is to enable comparison of various cutting tools under standard working conditions. The comparison concerns the performance of the tools and the production of secondary wastes.

"Secondary wastes" are defined as the emissions produced by the cutting as such, that is the material removed from the object being cut and material resulting from wear of the cutting tool. This type of waste is, if only in its grain size, very different from other wastes.

The tools have been chosen on the criteria of :

-remote control	-their mass and bulk must be compatible with remote
	control,
-efficiency	-some are regularly used on dismantling sites, others
	are potentially usable,
-diversity	-to examine the various cutting methods : by removal
	of metal shavings, by abrasion and by metal melting.

Objects for test cutting have been selected to ensure that the trials are as representative as possible and consist of materials frequently used in nuclear installations, with a sufficiently large range of thicknesses and cutting lengths. The following five tools have been tested on mild steel and stainless steel (304 L) plates of 10 mm, 30 mm and 50 mm thickness and of standard dimensions 700 mm by 500 mm :

-hacksaw,	- plasma torch
-grinder,	- arc air
- arc saw	

II.2 Trial installation

The trials have been done in a stainless steel chamber, or cell, of 32 cubic metres (see figure 1), equipped with two observation ports, and a cutting bench motorized on the X and Y axes (see figure 2).

The ventilation apparatus is shown schematically in figure 3 and comprises a very high efficiency filter (HEPA filter) mounted at the cell inlet and a fan mounted downstream of a HEPA filter at the cell extraction, this second HEPA filter having an intermediate electrostatic prefilter. The airflow of 300 m³/h can be regulated by a control valve and is measured using a precalibrated diaphragm.



Figure 1 : Schematic view of the trial cell



Figure 2 : Schematic view of the cutting bench



Figure 3 : Schematic view of the ventilation and sampling system

II.3 Analytical techniques

Isokinetic samples are taken using glassfibre filters (filter yield more that 99,99 % for particles larger than 0.3 micrometre) of diameters 47 mm and 130 mm which are connected on both sides of the electrostatic filter. An inertial, diffusing spectrometer (SDI2001) is also used. This consists of an Andersen impactor associated with a diffuser comprising six columns containing various numbers of calibrated balls. This enables grain size distribution to be determined for particles between 0,075 and 15 micrometers.

In order to obtain samples according to established standards, the following procedure is followed : -purge of the cell at a rate of 1000 cubic metres per hour for 30 minutes prior to cutting. -ventilation turned off. -cutting begins. -ventilation turned on at 300 cubic metres per hour after the time needed to reach running conditions. -sampling commences, -end of sampling and cutting, -purge of cell at 1000 cubic metres per hour.

At least two complete trials (including the cutting and the subsequent weighings) were carried out for each test configuration, comprising :

-the actual cutting,

-measurement of grain size distribution,

-the following weighings:

-the plate before and after cutting (precision of balance : ± 1 g).

-tool wear (disc, blade, nozzle, electrode) before and after cutting

(precision ± 0.01 g),

-slag deposited (precision ± 0.01 g),

-attached particles sampled manually (precision $\pm 0,01$ g),

-particles deposited on specific test surfaces of the cell walls

(precision ± 0.01 mg), sampled by scraping.

-deposits on filters before and after cutting and after drying at 40 °C for at least ten hours (precision ± 0.01 mg)

-measurement of volumes removed from the filters (precision ± 0.5 litres), -measurement of the cutting time (precision ± 1 second).

II.4 Results of measurements

The trials have been carried out in 1992 and 1993.

Table no. 1 shows, for each configuration, the number of complete trials really done together with the mean length of the cut.

It should be noted that none of the samples was subject to any prior thermal or mechanical treatment.

Tables 2 to 9 give the mean values of the measurements made.

Cutting speed and mass of various types of wastes (see tables 2 to 7).

The first column sets out the general conditions of the trial.

The second column states the maximum cutting speed for each tool (for minimum production of attached slag).

The third column gives the total mass, relating to the section cut, of steel removed from the plate by the cut (kerf coefficient) : the mean width of the kerf in mm can be estimated by dividing these figures by the mass/volume of the steel.

The fourth column shows the mass lost by the tool (blade, disc, nozzle, electrode) related to the section cut (coefficient of tool wear). The mass of secondary solid waste related to the section cut is found, for each trial configuration, by adding the relevant lines in columns 3 and 4.

The four last columns give the mass, related to sections cut, for each type of secondary solid waste :

-slag deposited, -attached slag, -slag deposited on the chamber walls, -aerosols.

		stainless sto	eel		mild steel	
	10 mm	30 mm	50 mm	10 mm	30 mm	50 mm
reciprocating saw	Э/0.73 m	1 / <0.1 m		3 / 0.67 m	2 / 0.25 m	
grinder	3 / 0.43 m	3 / 0.33 m		4 / 0.35 m	1 / <0.1 m	
plasma torch	6 / 3.1 m	2 / 2.1 m	2 / 0.95 m	2 / 3.75 m	3 / 1.76 m	2 / 1.05 m
arc-air	4 / 0.7 m	2 / 0.15 m		2 / 0.4 m	2 / <0.05 m	
arc saw	2 / 0.6 m	1 / 0.3 m		2 / 0.9 m		

Table 1 - number of tests and average length cutfor each configuration

Text cont. on p. 66.

Table 2 - Stainless steel cutting data (thickness	10 mn	n)
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General conditions	cutting				san	nplings	
material: stainless steel thickness: 10 mm	speed:	kerf factor:	mean factor:		deposits		aerosols;
in air	(mm/mn)	removed mass cut section (kg / m2)	<u>iost mass</u> cut section (kg / m2)	sedimented: <u>collected mass</u> cut section (kg / m2)	attached: <u>collected mass</u> cut section (kg / m2)	on walls: <u>collected mass</u> cut section (kg / m2)	<u>collected mass</u> cut section (kg / m2)
tool:							
reciprocating saw	10	17,2	0,022	16,6	o	0_17	0,003
grinding	35,8	32,3	5,43	36,6	0,58	2, 1	1,17
plasma torch	591	25,2	0	14,7	11,2	0,12	Q, 5
arc-air	180	65,7	0,43	45,2	0	0,23	0,81
SIC SBW	87,8	78,4	7,53	61,3	0	0,95	1,31

Table 3 - Stainless steel cutting data (thickness 30 mm)

General conditions	cutting				san	nplings	
material: stainless steel thickness: 30 mm	speed:	kerf factor:	mean factor:		deposits		aerosols:
in sir	(mm/mn)	removed mass cut section (kg / m2)	<u>lost mass</u> cut section (kg / m2)	sedimented: <u>collected mass</u> cut section (kg / m2)	attached: <u>collected mass</u> cut section (kg / m2)	on walls: <u>collected mass</u> cut section (kg / m2)	<u>collected mass</u> cut section (kg / m2)
tool:							
reciprocating saw	0,56	20,3	0.022	17,7	ο	o	o
grinding	29,5	33,2	16.9	48,6	0,09	0,83	0,71
plasma torch	207	34.4	O	34,5	0	0,05	0,081
arc-air	86,2	87	3,43	91,1	0	0,2	0,48
arc saw	15,3	68,4	39,6	82,6	0	1,68	1,86

Table 4	- 1	Stainless	steel	cutting	data	(thickness	50	mm)	ļ
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General conditions		cutting		samplings				
material: stainless steel thickness: 50 mm	speed:	kerf factor:	mean factor:		deposits			
in air	(m m/ma)	<u>removed mess</u> cut section (kg / m2)	<u>lost mass</u> cut section (kg / m2)	sedimented: <u>collected mass</u> cut section (kg / m2)	attached: <u>collected mass</u> cut section (kg / m2)	on walls: <u>collected mass</u> cut section (kg / m2)	<u>collected mass</u> cut section (kg / m2)	
tool:								
reciprocating saw	1	1	1	/	1	1	1	
grinding	/	1	/	1	1	/	1	
plasms torch	115	38.4	0	39,6	0	0.02	0,11	
arc-air	/	/	/	1	1	1	1	
arc saw	/	/	1	1	/	/	1	

Table 5 - Mild steel cutting data (thickness 10 mm)

General conditions		cutting		samplings				
material: mild steel thickness: 10 mm	speed:	speed: kert factor: mean factor: deposits				aerosols:		
in air	(mm/mn)	removed mass cut section (kg / m2)	<u>lost mass</u> cut section (kg / m2)	sedimented: attached: <u>collected mass</u> <u>collected mass</u> <u>collected mass</u> cut section cut section (kg / m2) (kg / m2)		on walls: <u>collected mass</u> cut section (kg / m2)	<u>collected mas</u> s cut section (kg / m2)	
tool:								
reciprocating saw	11,6	17,1	0,007	16,9	ο	0,2	0,003	
grinding	36,1	36,7	2,9	32,9	1,03	1,05	1,56	
plasma torch	507	13	0	14,9	o	0,15	0,42	
arc-air	282	51	2,82	48,1	0	0,35	2,91	
AFC SAW	124	50,3	7,26	53,8	0	0,29	1,15	

Table) 6 -	Mild	steel	cutting	data	(thickness	30	mm)
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General conditions		cutting		samplings				
material: mild steel thickness: 30 mm	speed:	speed: kerf factor: mean factor: deposits		aerosols:				
in sir	(mm/mn)	<u>removed mass</u> cut section (kg / m2)	lost mass cut section (kg / m2)	sedImented: <u>collected mass</u> cut section (kg / m2)	attached: <u>collected mass</u> cut section (kg / m2)	on walls: <u>collected mass</u> cut section (kg / m2)	<u>collected mass</u> cut section (kg / m2)	
tool:								
reciprocating saw	1,95	19	0,014	19,2	o	O	0,004	
grinding	9,12	38	13,9	38,7	0	0,73	0,36	
plasma torch	314	17,6	o	12,2	3,53	0,13	0,44	
arc-air	76,8	82,6	4,21	84,1	0	0,46	2,02	
arc saw	1	1	1	1	1	1	1	

Table 7 - Mild steel cutting data (thickness 50 mm)

General conditions		cutting		samplings				
material: mild steel thickness: 50 mm	speed:	kerf factor:	mean factor:		aerosols:			
in air	(mm/mn)	<u>removed mass</u> cut section (kg / m2)	<u>iost mas</u> s cut section (kg / m2)	sedimented: <u>collected mass</u> cut section (kg / m2)	attached: <u>collected mass</u> cut section (kg / m2)	on walls: <u>collected mass</u> cut section (kg / m2)	collected mass cut section (kg / m2)	
tool:								
reciprocating saw	1	,	1	1	1	1	1	
grinding	1	/	1	1	/	1	. /	
plasma torch	136	25,2	0	21,4	5,81	0,09	O, 44	
arc-air	,	,	1	1	1	1	1	
arc saw	1	1	1	/	/	/	1	

•

thickness (mm)	tool	mini diam. (microm.)	mean diam. at peak (microm.) / cumulative mass	cumulative mass at 0,1 microm.	mean diam. at peak (microm.) / cumulative mass	cumulative mass at 1 microm.	mean diam. at peak (microm.) / cumulative mass	cumulative mass at 5 microm.	mean diam. at peak (microm.) / cumulative mass
	reciprocating saw								
	grinding	0,01	1	3%	02/8%	24%	1	61%	6 / 67%
10	plasma torch	0,05	0.1 / 16%	16%	1 / 71%	71%	1	90%	6 / 92%
	arc-air	0.03	01 / 15%	15%	1	34%	5 / 68%	68%	I
	arc saw	0,01	/	5%	08 / 46%	56%	/	95%	1
	reciprocating saw								
	grinding	0,05	01/7%	7%	/	19%	1	54%	8 / 76%
30	plasma torch	0,01	004 / 8%	29%	04 / 41%	64%	5 / 86%	86%	,
	arc-air								
	arc saw	0,01	/	5%	1 / 52%	52%	1	94%	1
	plasma torch	0.01	0 04 / 11%	34%	04 / 41%	64%	5 / 85%	85%	,
50	arc saw								

thickness (mm)	tool	mini diam.	mean diam. at peak (microm.) /	cumulative mass at 0, 1 microm	mean diam. at peak (microm.) /	cumulative mass a t 1 microm	mean diam. at peak (microm.) /	cumulative mass at 5 microm	mean diam. at peak (microm.) /
(1011)		(meron.)		microin.	Cottigiariae ulass	interon.	comorariae mass	interoin.	Cumulative mass
	reciprocating saw	0,1	/	0%	08 / 26%	35%	1	60%	6 / 72%
	grinding	0,05	0.1 / 4%	4%	1	12%	1	52%	8 / 69%
10	plasma torch	0,01	0.05 / 5%	9%	1 / 43%	43%	1	79%	6 / 84%
	arc-air	0,03	0.08 / 8%	10%	1	17%	4 / 58%	69%	1
	arc saw								
	reciprocating saw	0,3	,	0%	1 / 25%	25%	4 / 94%	100%	/
	grinding	0,03	0.08 / 10%	15%	1	24%	4 / 62%	74%	1
30	plasma torch	0,01	0.04 / 3%	7%	1 / 22%	22%	1	69%	6 / 79%
	arc-air								
	arc saw								
	plasma torch	0,01	0.04 / 6%	10%	1 / 32%	32%	1	78%	6 / 84%
50	ârc saw								

Table 9 - Aerosols size distribution (mild steel)

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The first two columns describe the trial configuration.

The third column gives the minimum diameter of the particles collected. The following columns give data in order of increasing aerosol sizes.

Columns 5,7 and 9 show percentages of mass of particles collected whose sizes are less than 0,1 micrometres, 1 micrometre and 5 micrometres respectively. These sizes have been defined in view of the spectra obtained (see the examples given in figures 4 to 8).

The other columns concern the peaks of grain size distributions : they give the mean diameter of the particles centred on each peak as well as the percentage of mass of particles whose size is less than this mean diameter.



Figure 4 - Aerosol size distribution Mild steel e = 10 mm thickness







II.5 Analyses

Figures 9 to 14 annexed give graphic representations of the variations in the principal parameters.

Cutting speed

Figures 9 and 10 represent the variations in the maximum cutting speed for each trial configuration, according to the thickness being cut.



Fig 9: feed rate for tools generating heat versus the thickness to be cut



Fig 10: feed rate for mechanical tools versus the thickness to be cut



Fig 11: kerf factor versus the thickness to be cut



Fig 12: Mean factor

In view of the large differences between these speeds and in order to keep the scales linear, these variations are shown in two graphics : the first relates to the thermal tools (the most rapid) and the second to the mechanical tools (the slowest).

Kerf width

Figure 11 shows, for each trial configuration, the variations in the kerf coefficient and the mean kerf width according to the thickness cut.







Fig 14: deposited and volatile particles from mild steel cutting versus the thickness

Tool wear

Figure 12 shows, for each trial configuration, the variations in the tool wear coefficient, according to the thickness being cut. Wear in the hacksaw blade and the plasma torch nozzle are too small to be shown.

Secondary wastes

Figures 13 and 14 show, for each trial configuration, a graphic representation of the total mass of secondary wastes (calculated by totalling columns 3 and 4 of tables 2 to 9) and their split into two categories corresponding to particles deposited and attached (columns 5 and 6 of tables 2 to 9) and to particles carried away (columns 7 and 8 of tables 2 to 9).

OBSERVATIONS CONCERNING CUTTING SPEEDS (fig. 9 and 10)

In conducting the trials we have sought the maximum cutting speeds resulting in the minimum of attached particles.

Of the tools tested, the thermal tools have cutting speeds greatly superior to those of the mechanical tools. Their power isquirements vary similarly.

The plasma torch is the most rapid tool; from 500 to 600 mm of cut per minute in 10 mm thick steel, while the hacksaw is the slowest; about 10 mm of cut per minute in 10 mm steel. The power requirement of the hacksaw is less than one tenth that of the plasma torch.

The type of steel being cut is not particularly important except with the grinder, where the cutting speed in 30 mm mild steel is one third of the speed in 30 mm stainless steel.

OBSERVATIONS ON THE KERF (fig. 11)

We can split the tools into two groups :

-the first group concerns the tools whose use has been well perfected :

- * the hacksaw gives a clean kerf which is regular and hardly affected by the thickness or type of steel being cut,
- * the grinder gives a regular kerf which is also hardly affected by the type and thickness of steel,

* the plasma torch in stainless steel makes a kerf twice as wide as that in mild steel and the kerf is also doubled in 50 mm thick steel compared to 10 mm steel.

-the second group comprises the thermal tools, where operation is less repetitive :

- * the arc air had to be used manually because of the very rapid electrode wear.
- * the arc saw had a simplified control apparatus which did not enable optimum adjustment of the electrical parameters, thus explaining why a falling curve was obtained.

OBSERVATIONS ON TOOL WEAR (fig. 12)

Since the wear on the hacksaw blade and on the nozzle of the plasma torch are very low, they may be taken as negligible. The trials have thus not provided an indication of the lifespans of these two tools.

The loss of mass from the disc of the arc saw increases from 7.5 kg when cutting one square metre of 10 mm thick stainless steel to 40 kg for one square metre of 30 mm stainless steel. In cutting the same materials the wear on the grinding disc increases from 5.5 to 17 kg/square metre and the wear on the electrode of the arc air from 0.5 to 3.5 kg/square metre.

OBSERVATIONS ON SECONDARY WASTES (fig. 13 and 14)

Figures 13 and 14 split the tools into two groups :

-the arc saw, the arc air, and the grinder which produce the most particles deposited and carried away,

-the plasma torch and the hacksaw which produce the least secondary wastes, the latter producing less than 1 % of aerosols.

Grain size distributions are bimodal for the hacksaw, the grinder and the arc air.

The arc saw has a monomodal distribution with about 50 % of the particles less than one micrometre in size.

Tables 8 and 9 show that only the plasma torch results in a trimodal distribution of aerosols, these aerosols being less for stainless steel than for mild steel. This may be explained by the vaporization of the stainless steel ingredients (chrome, etc.) and the greater oxidation of mild steel.
In the case of the plasma torch, a large disparity is observed between the peak values for 30 mm and 50 mm stainless steel. 0.04 micrometres, 0.4 micrometres and 5 micrometres and the peak values for 10 mm stainless steel 0.1 micrometre, 1 micrometre and 6 micrometres

This disparity may be explained by the large quantity of slag attached to the 10 mm stainless steel plate During the trials the optimum cutting conditions were not attained and the permissible maximum cutting speed could not be reached.

II.6 Conclusion

II.6.1 Applications for use

The plasma torch is the most rapid tool and shows the least tool wear. It is suitable for all metal thicknesses studied

The hacksaw is the slowest tool and shows little tool wear but is not suitable for thicknesses of more than 30 mm, particularly in stainless steel

The grinder is not well adapted to thicknesses over 30 mm, particularly in mild steel.

The arc air is very difficult to use under remote control since it is vital to begin the cut correctly by gradually increasing the cutting speed. During the cut it is then important to maintain a constant speed and to guide the blade correctly in the kert (tolerance of only a few tenths of a millimetre). It seems of little interest for cutting thicknesses of more than 30 mm

The arc saw is not suitable for remote control but can be used to cut thicknesses well in excess of 50 mm

II.6.2 Secondary wastes

The hacksaw produces the largest aerosols, minimum diameters being around one tenth of a micrometre (compared to a few hundredths of a micrometre for the four other tools) and the first peak being around 1 micrometre (compared to a few hundredths of a micrometre for the other four tools).

The arc air and the arc saw produce the greatest quantities of secondary wastes.

The grinder and the arc air produce the greatest quantities of wastes on the cell walls This is probably due to the streams of sparks which tend to project the particles towards the walls From this the plasma torch appears to be the tool with the highest performance. Its remote control requires the use of a motorized slave head on the electric arc to compensate for the manipulator arm's inability to achieve precise positioning.

The mechanical tools which function on the principle of cutting swarf off the metal also show advantages in that they do not involve great heating of the metal being cut (which 1° vital when samples are being taken for metallurgical analysis) and that at least 95 % of the metal removed from the kerf is in the form of easily collectible swarf. This last characteristic lessens the total operation time since cleaning time must be included in the total. Accordingly, these tools can, in certain cases, be preferable to the thermal tools even though their cutting speeds are less.

The arc saw deserves to be developed and tested in the conditions in which it is likely to show the best results, i.e. under water and in cutting very great thicknesses. This is an area where there are very few competing tools available and which is encountered particularly when dismantling power reactors.

III TRIALS AND DEVELOPMENT OF CONCRETE CUTTING TOOLS

III.1 General description

The two tools described below have been developed by the UDIN to satisfy its immediate needs. They were first used in the AT1 installation as part of its stage 3 dismantling according to the IAEA definition (Figure 15).

AT1 is an irradiated fuel reprocessing facility located in the La Hague complex in France.

Work in the high activity cells has been carried out remotely using the MA23 robot mounted on the ATENA carrier.

The following information has been obtained from the managers of the ATI project.

The diamond saw has been adapted from a commercially available tool in order to cut a hole, by remote control, in a reinforced concrete dividing wall between two chemical treatment cells. This was to enable the MA23 robot to gain access to the further cell.



The shot blaster has been developed to decontaminate, semi-automatically, the walls of cells amounting to about 3000 square metres of concrete and several hundred square metres of recovery flocr in stainless steel.

III.2 Diamond saw

III.2.1 Operating conditions and results.

The programme ran from November, 1990 to March, 1991.

The opening to be made measured 5.5 metres by 2 metres in a concrete wall 200 mm thick.

Cutting was carried out in air and dry (the air change rate of about 10 per hour was handled by the building's own ventilation).

In fact, any use of water was ruled out by owing to the risk of contamination of the uncovered concrete of the cell walls.

The blade cooling system, initially intended to be by water, was therefore replaced by a liquid nitrogen feed (flow of about 50 litres/hour). The tool is powered hydraulically (120 bars pressure, flow from 0 to 38 litres/minute), its dimensions are 880 x 430 x 210 mm and its weight about 10 kg. The blade has a diameter of 350 mm and is 4 mm thick, with a peripheral speed of 50 metres/second. Its speed of advance is about 2 mm/minute and its functioning temperature is around 10 °C.

The tool is fixed onto the polyarticulated arm of the ATENA carrier by a special support.

Cutting proceeded in good conditions. Constraints related to the removal of the cut blocks of concrete resulted in a total length of cut of 21 metres, which needed about 20 blades to complete. Some of these blades were replaced as part of preventive maintenance operations on the ATENA carrier.

Due to the conditions on site it was not possible to take samples of secondary wastes.

III.2.2 Conclusion

We can report that , although the need for remote control, gas cooling, etc. made for difficult operating conditions, this tool has proved to be particularly effective. The development needed is as follows :

-for the blade

*improvement in blade changing while under remote control, *research into larger diameters while conserving the remote control ability and the principle of mounting and motorization by eccentric rollers.

-for lubrication :

*search for the lubricant requiring the minimum flow,

-for secondary wastes :

*mass and grain size analyses.

III.3 Shot blaster

III.3.1 Operating conditions and results

This tool has been developed with the aim of :

-crust removal to a depth of several millimetres.

-brushing the treated surface,

-recuperating the contaminated rubble and particles.

-recycling the shot.

-filtering the motor air.

Figure 16 shows the ventilation/filtration system.

The blasting pistol weighs about 5 kg, is equipped with a brush and will be adapted for remote control : grip adapted to the carrier and a remote control apparatus (see Figure 17).

On-site work is programmed for 1993. Trials in active conditions were done during the summer of 1992 on a wall of one of the chemical treatment cells, the pistol being handled manually.



- 1 Dust cleaner
- 2 Collector
- 3 Vibration sifter
- 4 Under pressure tank 5 Frame
- 6 Air valve
- 7 Evacuation cap 8 Filter 9 Dust receiver

- A Fan
- Filter В
- С Decantation drum
- D Vacuum pump E Handle
- F Pistol

FIGURE 16 SHOT BLASTING DIAGRAM



FIGURE 17: BLASTING PISTOL

45 square metres of resin coated concrete were treated, 32 kg of concrete was recuperated in containers and 4 kg of steel shot from the initial 100 kg was used up.

The depth per pass was from 3 to 4 mm for a treatment rate of about 4 square metres per hour, involving pistol movement of about 3 metres per minute.

Total initial activity was from 100 to 700 Bq/square centimetre in alpha emitters and the non fixed contamination was from 100 to 220 Bq/square centimetre in alpha emitters and trom 30 to 100 Bq/square centimetre in beta-gamma emitters (locally 1200 Bq/square centimetre)

Total residual activity after treatment is from 0,2 to 0,6 Bq/square centimetre in alpha emitters (locally 14 Bq/square centimetre), while the non fixed activity is too low for measurement

III.3.2 Conclusion

The decontamination factors achieved with the shot blasting tool have been very high

T tals done in active conditions on metallic objects (mild and stainless steels) have also shown positive results.

Since the surfaces to be treated are very large it has been decided to decontaminate them in a semi-automatic fashion. This presupposes on one hand the availability of carriers suitable for all configurations : horizontal or sloping floors and wells. vertical or stepped walls, and on the other hand, the creation of and interface between the tool and the carrier.

In order to operate the pistol in the AT1 cells by remote control UDIN is studying two configurations

-the pistol to be handled by means of the MA23 mounted on the ATENA carrier if the pistol/carrier interface involves a simple grip. In this case the flexibility and following ability of the MA23 will enable the cutting parameters to be satisfied, particularly concerning the orientation of the nozzle in difficult places and the force applied to brushing the surface,

-the pistol to be fixed on a special carrier in the event of the above functions having to be achieved using a minimum of actuators and captors.

IV. CEA-UDIN "TOOLING" PROGRAMME

For the future, the UDIN "tooling" programme foresees

-continuation of shot blasting trials particularly in order to specify the characteristics of the shot to be used according to the material for treatment and the carrying out of decontamination work with either a special carrier to be built or the MA23 (or both) and linked to the equipment and software of the Computer Assisted Remote Control.

-development of the arc saw.

-trials in active conditions concerning the decontamination and cutting of large diameter pipes using explosives.

-gathering of results obtained into a data base,

-study into the use of lasers for culting from a distance

ELECTROCHEMICAL DECONTAMINATION FOR THE MAIN PIPELINE OF THE PRIMARY CIRCUIT BY MOVABLE CATHODE

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Abstract

Contamination of the primary circuit of water-cooled reactors is essentially governed by the behaviour of iron, chromium, nickel and cobalt isotopes in the coolant. Since cations can easily be built into the metallic surface, the decontamination can only be carried out by the removal of the upper layer of the items containing the above mentioned radioactive isotopes.

An effective decontamination procedure removes the most part of contamination without excessive corrosion of the substrate.

Electrochemical decontamination is the reverse procedure of electroplating. The upper oxidelayers containing the contaminants can be removed from the metal surface using a direct electric current between the workpiece and a cathode in an electrolyte. The method can be applied by immersing the workpiece to be decontaminated in an electrolyte tank as an anode.

During the last years a special electrochemical procedure was developed for the decontamination of large or inmobil equipments using movable cathodes.

Three electrochemical, remotely operated decontamination equipments have been developed in Hungary for the Paks NPP.

Considering the good decontamination results achieved by the above mentioned equipments the development of the electrochemical method has been extended for the decontamination of the main pipeline.

Experimental methods

- Development of a weak acidic electrolyte on the base of laboratory equipments
- Design of a crawler as a carrier of testing and decontamination devices (decontamination head, isotope container, TV-cameras, etc).
- Construction and testing of the crawler and its accessories

Results obtained

Using an electrolyte containing oxalic and citric acid (pH = 3.5) the achievable decontamination factor was relatively high (20-500) and the corrosion rate was low.

- The crawler was tested in horizontal, vertical and apple pipes under inactive circumstances.

On the base of the tests some modifications will be carried out.

1. INTRODUCTION

In Hungary nuclear energy makes a major contribution (40 %) to the overall power generation, and the future development of the power production may be based on nuclear energy. At the present time the development of nuclear power is doubtful in Hungary, but negotiations are going on about the construction of further nuclear power units.

The existing Paks Nuclear Power Station has been in operation since the end of the year 1982. It has 4 reactors of 440 MWe each.

Radioactive contamination occurs in every area involved in the use of radioactive materials. Systematic and efficient maintenance is an essential condition for the safe operation of nuclear power plants, however maintenance and supervision works are dangereous because of radiation hazard of personnel. Radiation doses can be reduced by decontamination of the primary circuit. On metallic surfaces decontamination can only be achieved by the removal of the upper layer of the surface containing the contaminants.

2. ELECTROCHEMICAL DECONTAMINATION IN HUNGARY

Electrochemical decontamination by movable cathode has been used since the beginning of the operation of the first reactor unit. Electrochemical decontamination is the reverse of electroplating. It can be applied by immersing the items to be decontaminated in an electrolyte tank as an anode

of an electrolitic cell.

Because of the big volume of radioactive effluents generated during the immersion method, an in-situ decontamination procedure was developed to decontaminate immobile or large equipments. The first electrochemical equipment using movable cathode was developed for the decontamination of the control rods (Fig.1.), but the movable cathode was handled by a worker. High (200-400) DFs were achieved by this procedure, but when the inital dose rate was high, personnel's exposure was high as well.

During the last years three electrochemical, remotely operated decontamination equipments were developed in our Institute for the Paks NPP. These are for the decontamination of:

The main circulating pump case (Fig. 2.).
The collectors of the steam generator, (Fig.3.)
The main gate valve case of the primary circuit (Fig.4.)

The obtained DF-values were favourable (20-500), and after several subsequent decontamination the increasing of recontamination has not been observed because of the smooth surface prodeuced by the electropolishing effect.



Fig. 1. Electrochemical decontamination by manhandling



Fig. 2.: Electrochemical decontamination for the main circulating pump case



Fig.3: Electrochemical decontamination for the collector of the steam generator



Fig.4: Electrochemical decontamination for the case of the main gate valve

The advantages of the remotely operated electrochemical decontamination procedure are as follows:

- high decontamination factor (20-500)
- short application time
- produces smooth surfaces which reduces recontamination
- low collective dose
- low volume of liquide radioactive wastes

Considering the obtained results with the electrochemical decontamination technology, the development of the procedure has been extended for the decontamination of the main pipeline of the primary circuit. The development is carried out in two steps:

- Development of appropriate electrolyte
- Design and construction of the remotely operated equipment

3. DESIGN CRITERIA FOR THE DECONTAMINATION EQUIPMENT

3.1. Electrochemical criteria

The electrochemical decontamination procedures developed so far use strong acides - $pH \sim l$ - as electrolytes, which would cause corrosion on the metal surface if they can not be removed in a short time. Therefore the following criteria were established.

- pH of the electrolyte be close to neutral
- High decontamination factor (> 100)
- Low corrosion effect
- Short contact time

3.2. Mechanical engineering criteria

- The crawler must be equal to running along the main pipe (diameter: 594 mm) of the primary circuit from the reactor vessel to the gate valve and from the steam generator to the gate valve.
- Go round the extended proof sticks (max: 60 mm) by a passive way.
- Ensure manual pulling back.
- Detect and indicate its position on the control desk.
- Carry a visual observation device.
- Run through the whole piping including the vertical and horizontal sections and the angle pipes.
- Run through the gate valves.
- The TV-camera and the isotope container must (for radiography) always be in the axle of the pipe.
- Dead weight:

Crawler unit: 50 kg Container: 50 kg

- The tractive power: max 100 kg
- Pneumatic drive: step by step.
- Electric remote control by a computer, forwardreverse moving with a fixed pace.

- The equipment fixes itself to the pipe when operating trouble occurs.
- The crawler operates max 40[°]C, under dry condition of the pipe.
- The axial turning is acceptable.

4. LABORATORY EXPERIMENTS OF ELECTROLYTES

The electrochemical decontamination technologies developed so far use strong acides - $pH \sim 1$ - as electrolytes, which can cause corrosion on the metal surface. Since the spent electrolyte can not be removed immediately from the bottom of the main pipeline therefore we tested not only the achievable DF, but also the corrosion effect of different electrolytes.

The stainless steel samples for the decontamination experiments were treated in a rocker-type reactor under the following conditions:

> Temperature: 295 ^OC Pressure: 12 MPa Duration: 500 h Composition of the liquid: H₃BO₃: 8 g/l pH: 7,8 (with N₂H₄ and NH₄OH) ⁵⁸Co: 50 kBq/cm³

The stainless steel samples for the corrosion experiments were neutron activated in a research reactor.

We tested the different compositon of oxalic-, citric-, phosphoric-, sulfuric and tartaric acid and their ammonium salts, in the pH-range of - 0,5 to 7.

Sodium-sulfide as an alternative (alkaline) electrolyte was also tested.

The experiments were carried out under the following conditions:

Current density: 50 A/dm² Temperature: 85-90⁰C Distance between the electrodes: 5 mm the surface to be decontaminated: 16 cm²

Figure 5. shows the decontamination factor (DF) and the corrosion rate using 50 g/l to phosphoric acid and its ammonium salts versus pH.





Figure 7. shows the DF and the corrosion rate using 50 g/l tartaric acid and its ammonium salts versus pH.

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Figure 7. DF and corrosion rate using 50 g/l tartaric acid and its ammonium salts. The duration was 5,60 and 120 s for the DF tests. ---- represents the corrosion rate ______ shows the values pH, where the ratios between the DF and corrosion rate are favourable.

Figure 8. shows the DF and the corrosion rate using 50 g/l citric acid and its ammonium salts versus pH.



Figure 8. DF and corrosion rate using 50 g/l citric acid and its ammonium salts. The duration was 5, 60 and 120 s for the DF tests. --- represents the corrosion rate. ---- shows the values pH, where the ratio between the DF and corrosion rate are favou**v**able.

Figure 9. shows the DF and the corrosion rate using sulfuric acid and its ammonium salts versus pH.



Figure 9. DF and corrosion rate using 50 g/l sulfuric acid and its ammonium salts. The duration was 5, 60 and 120 s for the DF tests. --- represents the corrosion rate. ---- shows the values of pH, where the ratios between the DF and corrosion rate are favouyable.

When comparing decontamination factors and corrosion rates, the compositions of oxalic-and citric acid were the best, resulting in high decontamination factor and low corrosion rate. Since the electric conductivity of oxalic and citric acid is relatively low, the current density is not high enough. This composition can be used for the immersion method, but not for in situ technology using movable cathod. In this case a small amount 5 g/l of sulfuric acid is to be added to increase the current density. When using Sodium-sulfide the decontamination factor was sufficient (DF = 100-500) and corrosion rate was low enough, $(\sim 1.10^{-3} \text{ mg.min}^{-1} \cdot \text{cm}^2)$ but we don't suggest it, for other aspects (waste management, H₂S-formation).

The composition of the proposed electrolyte is the following:

```
oxalic-acid: 5-10 g/l
citric-acid: 40-50 g/l
sulfuric acid: 5 g/l
pH = 3,5 (with ammonium hidroxid)
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The figures 10. and 11. shaw the DF and corrosion rate versus pH of two diffent electrolytes. If the electrolyte contains sulfuric acid both DF and corrosion rate increase.



, ...



Fig. 11: Corrosion rate versus p_H

The expected decontamination factor: 200-400The estimated corrosion rate: $0,8-1,2.10^{-1}$ mg.min⁻¹.cm⁻² (at 50 A/dm² current density, 5-10 s contact time)

The feed rate of the electrolyte: 10-20 1/h

After the decontamination process the treated surface can be controlled by the TV-camera and if the quality of the surface is not appropriate, the cleaning process can be repeated. After the decontamination the inner surface of the pipe must be cleaned to metal.

5. The decontamination head (Fig.12.)

A decontamination head with moving cathods is mounted up on the crawler. This head consists of two moving cathodes, which works perpendicular to the pipe axis. The moving cathodes are in 180 degrees from each other.



Fig.12 : The decontamination head

The decontamination head consists of not only the moving cathodes, but a TV-camera for visual testing as well.

The moving cathodes are pressed on the inner surface of the pipe by pneumatic cylinders. The two cathodes are guided by an electromotor in a slowly rotating ring and the decontamination of a band is carried out by turning of this ring.

There is a TV-camera in the ring between the two moving cathodes. The picture of the inner surface can be observed by a monitor on the control desk to detect the possible failors.

A feed pump provides the accurate dosage of the electrolyte through a flexible tube for the moving cathodes. The feed rate does not depend on the hidraulic resistance because the pump is a piston-type one. The feed pump and the direct current supply is located at the pipe-end. A water-jet pump sucks up the used electrolyte through a flexible tube from the bottom of the pipe to reduce the corrosion effect.

The used electrolyte can be recirculated or pumped into the active sewage.

6. THE CRAWLER (Figure 13.)

The crawler consists of two essential parts:

- The units for the visual quality control and the decontamination.



Fig. 13: The crawler.

-Two identical units for conveyance of the control and decontamination units. These units are duplicate, to ensure safe operation.

The control and decontamination units are mounted between the conveyance units.

Tasks of the front unit: to carry the control units and the decontamination head. Tasks of the rear unit: to carry the control units, the decontamination head, the electric cabels and the steel wire for pulling back.



Fig.14.: The crawler in the pipe

The crawler is moved by stands step by step. There are two different types of stands. The fixing stands with roller ensure the positioning of the unit in the pipe axis.

The moving of the crawler is performed by the walking stands step by step. There are 3 walking stands on the front of the unit and 3 walking stands on its rear. The unit moves in the following way:

- a) The front walking stands are in a fixed position pressed to the pipe wall. The walking stands on the rear are in a pull-back position
- b) The unit makes a caterpillar (peristaltic) movement. The rear of the unit runs forward by a pneumatic cylinder. The unit huddles up.
- c) The front walking stands go in a pull-back position. The rear walking stands press in a fix position to the wall and the unit finishes its caterpillar movement, the front of the unit goes foreward by the pneumatic cylinder, and so on.

7. TESTING OF THE CRAWLER

The crawler was tested under inactive circumstances in horizontal, vertical and angle pipes. The results of the test are the followings:

- The going round of the extended proof sticks is unsafe and active sensors must be used in the future.
- The rubber tires on the roller of the fixing stands wear quickly. These must be changed for a better quality ones.
- The position of the decontamination head is unsuitable because it get caught in the angle pipe wall.
- The dead weight of the crawler is too high (about 100 kg). This weight can be reduced using aluminium and plastic material instead of stainless steel.
- An additional movable TV-camera is needed looking back and aside.
- The giving up of one conveyance unit is to be considered.

8. SUMMARY

A crawler was consructed for decontamination and material testing of the main pipe of the primary circuit of VVER nuclear power plant.

Electrochemical decontamination procedure and a movable head was developed.

The crawler was tested under inactive circumstances and on the base of these tests some modification would be carried out.

DECOMMISSIONING AND DECONTAMINATION STUDIES FOR NUCLEAR FACILITIES

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Abstract

Envisaging the diverse needs of the nuclear industry with respect to decontamination and decommissioning, an organised Research & Development programme was undertaken in India. Under the IAEA Co-ordinated Research Programme, studies were initiated on different areas of Decontamination and Decommissioning. These studies comprised of chemical and electrochemical processes for decontamination. This report summarises the results obtained during different studies carried out in these areas. Laboratory studies on dissolution of magnetite/nickel ferrite powders and oxide films deposited on metal specimens resulted in identifying suitable dilute chemical formulations based on EDTA, oxalic acid, citric acid, ascorbic acid, etc. Studies on the behaviour of ion exchange resins indicated that it is possible to effectively remove the activity with simultaneous regeneration of the formulation chemicals by the cation exchange resins and complete elimination of all the organic acids on mixed bed resins. Study on compatibility of the formulations with the different materials of construction is also reported. Electrochemical parameters have been established for electropolishing of stainless steel components in an electrobath. Creation of a decontamination data base and its usefulness is also reported.

1. INTRODUCTION

In India, nuclear power stations of the Pressurised Heavy Water Reactor type are in operation in different parts of the country. Two boiling water reactors are in operation at Tarapur. Some of these reactors have completed more than a decade of operating life. Need has been felt to decontaminate the reactor systems and components in order to reduce radiation exposure to operating personnel. A Research and Development programme was therefore undertaken to develop dilute chemical formulations suitable for decontamination of our reactor systems.

Data exists on the decontamination of radiochemical plants using different chemical formulations. Efforts were made to co-relate the data and carry out various parametric studies.

Decontamination of equipment and components is routinely carried out in the Decontamination Centre at BARC, Trombay. Studies were conducted on electropolishing of metallic components as a decontamination technique to optimise various electrochemical parameters.

2. **OBJECTIVES**

The following are the major objectives of our Research and Development work under the Coordinated Research Programme.

- Development of suitable chemical formulations for in-situ decontamination of reactor systems.
- Studies on electrochemical methods of decontamination of metallic components.
- Creation of a Data Base on plant scale decontamination campaigns.

3. **RESULTS AND DISCUSSIONS**

3.1 Development of dilute chemical formulations

A number of candidate formulations were investigated depending on the characteristics of the contamination layers in the reactor systems.

Carbon steel, monel-400, zircalloy-2 and stainless steel form the major materials of construction in the PHWRs. Out of the total surface area of these construction materials exposed to the PHT system about 94% can be attributed to carbon steel, monel-400 and zircalloy-2, and only about 6% to stainless steel. The major chemical constituents of the oxide film are magnetite and nickel ferrite. Activated corrosion products Co-60, Mn-54, etc., and fission products Cs-137, Ce-144, Ru-103, Ru-106 are the contaminants in the oxide film. Stainless steel is the chief construction material for Boiling Water Reactor system and the major contaminant film consists of chromium rich ferrites.

3.1.1 Dissolution of oxide powders

Dissolution of the oxide deposits on reactor system surfaces is the key for removal of radioactivity.

Magnetite and nickel ferrite powders were synthetically prepared in the laboratory in order to study their dissolution characteristics in various dilute chemical formulations. The formulations chosen should have adequate dissolution capacity without having any adverse impact on base metals. The chemical decontamination formulations studied include suitable combinations of an organic acid, a reducing agent and a complexing agent. The dissolution process involves reaction of the oxide ion with H + to form water and simultaneous complexation of the metal ions released from oxide lattice. Use of reducing agent for the reduction of Fe(III) to Fe(II) destabilises the lattice structure and enhances the dissolution process.

The following three dilute formulations were found to be promising.

- 1. EDTA, Citric Acid and Ascorbic Acid.
- 2. Picolinic Acid and Ascorbic Acid.
- 3. EDTA, Ctric Acid and Oxalic Acid.

The total concentration of the reagents in each of these formulations was less than 1 gm per litre.

Detailed dissolution study of magnetite powder was carried out using these formulations. Typical results showing the dissolution capacity of the above formulations is presented in Table I. It is evident that the EDTA based formulations (1) and (3) are kinetically superior to the formulation containing picolinic acid. The kinetic studies indicated that the dissolution follows the cubic rate law. It was also observed that the conditions for dissolution of magnetite by these dilute chemical formulations are also favourable to the dissolution of nickel ferrite.

3.1.2 Dissolution of oxide layers on metal specimens

Oxide films deposited coupons of carbon steel, monel-400 and stainless steel were prepared by autoclaving under simulated PHT system conditions. Good oxide coated films could be obtained on carbon steel coupons whereas in the case of monel and stainless steel coupons surface tarnishing with discontinuous oxide film growth was observed. These coupons were subjected to defilming studies in batch mode as well as recirculation mode. Laboratory scale glass loop set up and a pilot scale loop installation were used for dynamic studies under recirculation mode. A schematic diagram of the laboratory scale glass loop and the pilot scale loop are shown in fig. 1(a) and 1(b).

Formulation	Composition of formulations			pН	Time for 100% dissolution at:	
	Chemical A	Chemical B	Chemical C		50°C	90°C
1	EDTA	Ascorbic acid	Citric acid	3.0	37 mts.	30 mts.
2	Picolinic acid	Ascorbic acid		3.2	12 hrs	5.5 hrs
3	EDTA	Oxalic acid	Citric acid	2.3	37 mts.	25 mts.

Table I: Data for dissolution of magnetite in different decont. formulations

Monel and carbon steel coupons could be effectively decontaminated at 50°C with the above formulations. However formulation (2) containing picolinic acid was found to react slowly. Increase in temperature up to 90°C improved dissolution rates in all the cases, but with the formulation (2) base metal attack was noticed.

These formulations did not have noticeable effect on stainless steel surfaces. An oxidative pretreatment with alkaline permanganate followed by an acid dissolution step could however remove the oxide layer fully without base metal attack.

3.1.3 Decontamination studies

The carbon steel and monel-400 coupons exposed to PHT system of PHWR contained both activated corrosion products and fission products. Bench scale and dynamic loop decontamination runs indicated that decontamination factor varying from to 5 to 50 could be achieved using the above dilute chemical formulations.

3.1.4 Base metal corrosion behaviour

Base metal corrosion behaviour in the above formulations was studied at 50°C and 90°C by weight loss method for carbon steel, monel-400, zircalloy-2 and stainless steel (304 and 403) using rectangular specimens. Corrosion on stainless steel (304 and 403), monel and zircalloy-2 was found to be negligible. In the formulations containing picolinic acid, carbon steel exhibited higher corrosion rates than EDTA based formulations.

3.1.5 Activity pick up and behaviour of ion exchange resins

The adsorption by ion exchange resins of metallic ions removed by dissolution of magnetite layer of the coupon specimens in the decontamination formulations was studied in the glass loop experiments. Inactive metal ions of iron, nickel and activated corrosion products Co-60, Co-58, Mn-54 and fission products Cs-137, Ce-141, Ce-144, Pr-144 were also effectively removed by the cation exchange resins. Ru-103, Sb-125 were taken up by anion exchange resins. Ru-106, Zr-95 and Nb-95 were taken up by both the ion exchange resins.

It was observed that the complexants are adsorbed on cation exchange resins in varying degrees. Studies on adsorption of EDTA on cation exchange resins as a function of pH is shown in fig. 2. It was observed that the maximum loading of about 11.5 mM per litre of resin occurs at a pH of around 1.7.

Feasibility of complete removal of the formulation chemicals at the end of the decontamination cycle was studied. It was possible to adsorb all organic acids and the remaining traces of metallic ions on the mixed bed resins.



Fig. 1(a). Laboratory scale glass loop for decontamination studies.



Fig. 1(b). Flow diagram for pilot plant loop.



Fig. 2. Variation of EDTA adsorption on cation IX as a function of pH.

3.2 Electro polishing studies

With a view to effectively and economically decontaminate equipment and components for reuse and for decommissioning, studies were initiated on the development of electrochemical techniques, using an electrobath. The electrochemical parameters such as current, voltage, time of contact, electrolyte composition and metal removal rate were studied for stainless steel components. Stainless steel specimens could be effectively electropolished to a bright smooth finish with a contact time of about 3-5 minutes using a mixture of 40% phosphoric acid and 20% sulphuric acid as electrolyte with a current density of 400-500 mA/sq.cm. Studies on active specimens indicated that irrespective of the surface conditions, stainless steel surfaces could be effectively decontaminated with decontamination factors ranging from 100-500. Use of organic acids as electrolytes was explored for the decontamination of carbon steel surfaces. Depending upon the nature of contamination decontamination factors ranging from 2 to 20 could be achieved.

3.3 Database on decontamination

Numerous data obtained during the plant scale decontamination campaigns in the radiochemical plants were utilised to create a DATA BASE on decontamination. The same was used for statistical analysis to have a correlation of various parameters affecting the decontamination process such as decontamination frequency, reagent combinations, incremental DF, surface to volume ratio etc. The DATA BASE was useful to assess the performance of various decontamination campaigns. Similar trends were observed for most of the cell equipments. Fig. 3 shows the effects of decontamination frequency on the decontamination factor achieved under a specific set of conditions.



Fig. 3. Effect of decontamination frequency on the decontamination factor.

4. CONCLUSIONS

The studies carried out under IAEA Co-ordinated Research Programme were useful in identifying suitable chemical formulations for decontamination of Indian PHWRs. The data generated during the study is beneficial for decontamination application of power reactor systems.

An electrochemical bath has been installed for electropolishing of miscellaneous components used in the nuclear facilities.

The Data Base structure has been suitably refined to include gross decontamination data obtained from nuclear facilities.

5. FUTURE PLANS

As an on-going programme, studies on development of chemical formulations will be pursued further for selection of optimum decontamination processes for nuclear facilities.

Studies on process qualification with the candidate formulations will be undertaken for specific reactor applications.

Electrobrushes/movable electrodes are being developed for specific decontamination applications.

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PROJECT MANAGEMENT AND GENERAL PLANNING OF DECOMMISSIONING ACTIVITIES FOR THE GARIGLIANO AND LATINA NPPs

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Abstract

This paper is a follow-up of a previous one, on the same subject, dated May 1991.

The Italian strategy for decommissioning NPPs and the present situation of Garigliano (BWR 160 MWe) and Latina (Magnox, 210 MWe) plants in relation to licensing and operational issues are reported. Some information about Caorso (BWR, 860 MWe) and Trino (PWR, 270 MWe) plants is also included.

The lack of specific national regulations and the absence of a final repository for radioactive waste severely affects the planning of decommissioning of Italian NPPs. Nonetheless a general strategy to carry out the decommissioning has been established and is based on two main points: 1- the plants will be brought in a relatively short time to a "passive safe enclosure" state (in Italian "Custodia Protettiva Passiva" CPP); 2- the final dismantling and the subsequent possible un-restricted release of the site will be delayed for several years. The CPP is meant to be an inherently safe state, thus ensuring personnel and general public radiological and conventional protection without the necessity of any action by operating personnel, only periodical inspections being sufficient to the purpose.

Garigliano and Latina plants have been granted operation licences whose aim is to bring them in line with the CPP state, up to now however only a few operations have been authorized. The reactors have been de-fueled and the irradiated fuel has been shipped off-site. Conditioning of operational waste is being performed while the operations to further confine radioactive materials have still to be authorized. For Trino and Caorso facilities a general plan for decommissioning activities must still be officially approved.

1 DECOMMISSIONING STRATEGY

The strategy adopted by Enel for decommissioning its NPPs, and accepted by the Italian control authorities with the issue of the new operation licences for Latina and Garigliano plants, can be summarized as follows:

- within 10 years of the final shut-down the plant will be put in the CPP state;
- for a relatively long period of time, presently estimated as a few decades, the plant will remain in the CPP state;

- the plant will then be dismantled and the site released without restrictions, if wished.

This strategy, delaying the final dismantling and final unrestricted site release, is similar to that chosen in other countries in the world.

The main activities to be performed to bring the plant in line with a CPP state are the following:

- de-fueling of the reactor and off-site shipment of irradiated and fresh fuel;
- treatment and conditioning of operational waste;
- confinement and containment of the residual radioactivity in a limited number of buildings, with partial dismantling of contaminated systems and components; the systems, components and materials containing the radioactivity have to be protected to reduce their deterioration in time.

While the general criteria to be followed in the design of the CPP state are the following:

- the radiological impact on the site during the CPP period has to be negligible both in normal and accidental condition;
- the safety of the plant during the CPP should be ensured without the need for active systems, while the passive systems should be designed so that they require minimum maintenance;
- the operations foreseen during the CPP period should only be those associated with the periodical inspections;
- the actions carried out to reach the CPP state should neither impair nor complicate the future dismantling operations.

Important steps to take within the boundaries of the chosen strategy are:

- the operative definition, for each plant, of the CPP state and the consequent outline of the actions to be performed;
- the minimum number of years for which the final dismantling is delayed.

The CPP state implies more than what is foreseen by phase 1 of the reference strategy stated by IAEA but, for several reasons that apply specifically to our country, it cannot be a complete IAEA phase 2 condition.

The conditioning of several thousand tons of contaminated material, needed to reach stage 2 of the IAEA strategy, cannot be done because in our country there is still not a final disposal site for radioactive waste and, as a consequence, there is a lack of boundary conditions for the actual engineering of the conditioning itself. Moreover there is no indication about the "de minimis" levels for releasing or recycling materials from dismantling.

2 LATINA

Latina NPP was equipped with a single reactor, of the gas-graphite Magnox type, with a design electrical output of 210 MW. After operating from 1963 to 1986 the plant was taken out of service by a governmental decision in 1987.

In April 1991 Enel was granted a new operation licence authorizing the activities aimed at decommissioning the Latina plant. The licence required that, by April 1992 (one year from the issue of the licence), Enel submit to the control authority, for approval, a general design illustrating the actions necessary to bring the plant in line with the CPP state. The design had to include the inventory of the radioactive materials present on the site and the general handling criteria of the radioactive waste including those resulting from fuel reprocessing.

As required, the design was presented in due time by Enel and is now being discussed. A brief summary of the radioactivity inventory results and of the solutions proposed to put the plant in the CPP state is given in the following paragraphs.

2.1 RADIOACTIVITY INVENTORY

2.1.1 Activated materials

The total weight of the activated steel is about 2000 ton. Five years from the final shut-down the quantity of steel having a specific radioactivity greater than 10^5 Bq/g is about 1080 ton, 120 ton of which have a specific radioactivity greater than 10^7 Bq/g. The total weight of the activated graphite is about 2100 ton whose specific activity, at 5 years from shut-down, has been evaluated as about $3x10^5$ Bq/g. The activated portion of the biological shield, assuming as "radioactive" the concrete when it has at least 0,1 Bq/g of induced activity, is about 11000 ton.

2.1.2 Contaminated materials

The total weight of the contaminated steel is about 5200 ton, while the concrete from the scarification of contaminated surfaces has been evaluated as about 1000 ton. The main contaminated components are: the boilers (3400 ton total), the primary circuits (1000 ton); the blowers (180 ton) etc..

2.2 PROPOSED SOLUTIONS TO PUT THE PLANT IN CPP CONDITION

The NSS of Magnox reactors does not have a global container, at Latina, moreover, the steam generators (boilers) are outside the reactor building. Therefore it is not always possible to ensure a double containment for the radioactivity. However the components that will not have a double containment are those not heavily contaminated and it has been calculated that if any barrier fails the impact on the environment will be negligible.
2.2.1 Reactor Vessel

The reactor vessel will be isolated from adjacent systems. The primary circuits ducts and all the instrumental penetrations will be cut and sealed with welded plates outside the biological shield. The stand-pipes will be sealed at their free ends. No action is anticipated on the internal components of the reactor vessel and on the graphite in particular. The biological shield will also be sealed to work as a second container for the major part of the residual radioactivity.

2.2.2 Primary circuits

The ducts of the primary circuits, about 40 m for each of the 6 circuits, will be removed and cut in several pieces each of which will be sealed with solid plates at the ends and stored inside the 6 dead spaces below the blowers. Due to their considerable volume, the pieces of duct could be used to contain contaminated materials.

2.2.3 Charge and discharge machines

The 2 C/D machines are being dismantled. The external parts, which are only very slightly contaminated will be decontaminated and, if possible, released. The internal components are kept inside the machine pressure vessel which is sealed with bolted plates and stored on site. One machine has already been dismantled, the other one will be dismantled by the end of 1993.

2.2.4 Boilers

The thermal insulation outside the boilers has already been removed. A small fraction of it, near the man-holes, was found to be slightly contaminated, and has been separated from the rest, which should be handled as conventional waste. The boilers will be kept in their present place, a reinforcement will be added at their base to improve the resistance to seismic stresses. All penetrations and connections with other components will be severed and sealed, the external surfaces will be protected against corrosion.

2.2.5 Fuel pit and fuel discharge route

The fuel pit will be decontaminated, drained, painted to fix the residual contamination and used as a deposit for contaminated materials. The irradiated fuel discharge route will be almost completely dismantled and the resulting contaminated materials will be stored on site.

2.2.6 Operational waste

The amount of operational waste of the Latina plant is relatively small and it will be conditioned according to the Technical Guides issued by the Italian control authority. A particular problem is presented by the conditioning of the Magnox "splitters" cut from the irradiated fuel before its shipment. On the one hand the conditioning with cement does not seem to comply entirely with the specifications stated by Technical Guide 26 on waste management. On the other hand no final answer has still been reached on the suitability of the dissolution process.

The sludges have been chemically and radio-chemically characterized. Due to the presence of alfa emitting nuclides, 21 GBq in total, and to the small quantity (about 20 m^3) they will be dried and put in suitable containers to be stored on the site.

2.2.7 Waste from fuel reprocessing

The irradiated fuel from Latina has been entirely reprocessed by BNFL. In the future, according to international agreements, the "glasses" will return to Italy and be taken to an interim storage facility.

3 GARIGLIANO

Garigliano NPP was equipped with a single reactor, of the BWR dual-cycle type, with an electrical output of 160 MW. After operating from 1964 to 1978 the plant was taken out of service by an Enel decision in 1982.

In September 1985 Enel was granted an operation licence to allow the actions needed to put the plant in the CPP state, within 10 years. Due to past delays, the achievement of the CPP state is not foreseen until the end of 1996, provided the Italian control authority approves the designs submitted by Enel.

The main data on the radioactivity inventory have been already given in a previous paper (1991).

3.1 REACTOR BUILDING (SPHERE)

In August 1991 Enel submitted a Detailed Design Report to bring the sphere in line with the CPP state. The goals of the design have already been described in a previous paper (1991); the proposed solutions and the main activities already performed will now be summarized.

3.1.1 Main internal systems

The vessel and the primary circuit have been roughly decontaminated and drained after replacing the vessel cover and the upper shield blocks. In-vessel components were replaced inside the vessel and properly fastened.

The fuel pool and the reactor channel are being roughly decontaminated. They will be drained and covered with a light metal structure sealed on the pool upper edges to constitute a barrier against the spreading of contamination. All circuits will be drained and insulated. The valves connecting various circuits will be kept closed, the connection with external equipment will be severed.

3.1.2 Containment sphere

The structure and components arrangement inside the sphere allows, in a relatively easy way, the division of the internal volume in two separate areas: area 1 and area 2.

Area 2, the most internal one, contains, within concrete structures, the most activated and/or contaminated systems and components. This area shall not be decontaminated and will be confined and made inaccessible in normal circumstances during the CPP period.

Area 1 is the "hollow space" between area 2 and the spherical container. This area will be decontaminated where necessary to bring the contamination level below 4 Bq/cm^2 . The few internally contaminated components which are in the area will be sealed with adequate care (e.g. sealing of valve stems etc.).

The separation of area 2 from area 1 will be completed closing and sealing all entrances and sealing interconnection pipes (as e.g. floor drainage pipes). A controlled intercommunication between the two areas is foreseen, placing a few filtering devices which will allow the passage of air while preventing the migration of contamination.

All mechanical penetrations of the sphere will be severed and sealed inside and outside the sphere, electrical penetration will be disconnected. The access holes will be sealed from the inside, only the normal staff entrance will remain operable.

A polyurethane foam has been qualified as a sealing material. The qualification process included density, traction, compression, fire resistance, adhesion and permeability tests before and after ageing.

The container has already been fully inspected and found to be in good condition, its external surface has been sanded and re-painted

3.1.3 Ventilation system

A new ventilation system will be built. The normal operation of the new system will be passive, the air flux being activated by the daily internal temperature variation (the container is metallic). The ventilation air, after filtration and de-humidification, will enter area 1, then area 2 and, after filtration, will be released outside. Special check valves, with extremely low pressure drop, have been designed, tested and qualified for the purpose. The overall air flow has been calculated as 600/700 Nm³ a day in summer and 300/400 Nm³ a day in winter, and ensures 4 complete air changes each year. An active extraction fan will also be available.

The mathematical model used to simulate the air flow through the sphere has been validated with experimental measurements. Four measurement campaigns (17-21

August 1987 and 16-20 March, 26-30 July and 7-13 December 1988) have been carried out; during the campaigns the containment was isolated and the following parameters recorded: differential (external vs internal) pressure, external abs pressure, external air temperature, external humidity, wind velocity and direction, the sun's irradiation and the container's temperature. The radioactive release associated with the ventilation air has been evaluated as very very low (less than 100 Bq per year). Several deteriorated situations were also examined (check valve failure, loss of seals integrity etc) and the results showed that the proposed solution is safe.

3.1.4 Control system

During the first two years of the CPP period a set of parameters will be checked; these include: internal air humidity, differential pressure (external vs internal), water level in the collecting tanks for the condensation, air contamination of area 1, filters and outlet air radioactivity. Should the design parameters be confirmed, less binding controls will be defined for the following years.

3.2 INTERMEDIATE LEVEL WASTE

3.2.1 Solid waste

Solid intermediate level waste consists of activated material discharged in the past from the core (fuel element channels, in core instrumentation tubes, control rods etc.) and stored in a pit outside the reactor building. The total weight of the activated material is about 3.8 ton, the volume is 1.4 m^3 and the total calculated radioactivity is about 750 TBq (1992).

The waste will be conditioned with cement inside baritic concrete containers. The containers have the shape of an octagonal prism, their overall length is 4.6 m, the maximum external diameter is 2.1 m and the final weight of each container after the waste conditioning will be about 50 ton.

The containers and the conditioning process have been fully qualified to meet the requirements of Technical Guide 26 and to ensure the transportability of conditioned waste according to IAEA Safety Guides; in particular appropriate tests were made on the cement cast inside the containers, on the obtained cement+steel matrix, on the containers and on a model (scale 1 to 2) of the final conditioned waste.

All waste handling and loading operations will be carried out under water. To do this the lateral walls of the pit will be elevated with carbon steel plates. A guard wall will be built all around the elevation plates and filled with clean water kept at a higher level to ensure against contaminated water leakage. On one lateral side of the pool the elevation plates will have an opening where the containers will be joined using inflatable seals. The waste will be retrieved and transferred into steel baskets using special manipulators. A water purification system will be used for the treatment of the pit water. A shed with an appropriate ventilation system will ensure the static and dynamic containment during the operations.

The main steps of the retrieval and conditioning process are the following:

- pit water purification and mud removal;
- positioning of the container and inflation of the relative seals;
- positioning of the steel basket on the free portion of the pit floor;
- filling to the maximum level of the guard wall, verification of tightness and subsequent filling of the internal pit up to the operational level;
- transfer of the waste inside the steel basket;
- lifting and insertion of the basket inside the container in a horizontal position;
- closing of the container with the relative lid;
- casting of cement inside the container;
- closing of the container inlet and outlet openings for water and cement;
- lowering of the water to the starting level and decontamination (washing) of the lateral pit surfaces;
- deflation of the container-pit seals and container removal.

The shed and the systems needed for the conditioning process are now under construction, while the retrieval and conditioning operations should start in September 1993 and be finished within the year.

The retrieval and conditioning activities will require 5 full-time operators for about 4 months, the forecast total dose commitment is 0.061 Man-Sv. Six containers are needed instead of 10, the previously estimated number of containers has been reviewed to take account of 60 Co decay; the containers will be temporarily stored on the site.

3.2.2 Resins, sludges and concentrates

During plant operation the following ILW were produced:

 ion-exchange granular resins from demineralizers of the cycle water 	163 m ³
- filtering sludges (fossil sands and powder resins) from rad-waste and fuel pool filters	39 m ³
- evaporation concentrates, made of a saturated solution of sodium sulphate	60 m ³
- decon. solutions (Turco Decon 4521, 6% weight)	2 m ³

The radio-chemical characterization is summarized in table I.

Table	I
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Waste	Spec. activity solution KBq/g	Spec. activity solid phase MBq/g	137Cs content %			
Resins	***	0.1 / 0.6	65 / 90			
Sludges	6 / 170	0.6	23 / 100			
Concentrates and decon. solutions	2.4 / 7.2		95			

The waste is now contained in different metal tanks inside an underground building. They will be stirred, retrieved, transferred to a conditioning machine and conditioned with cement inside metal drums. The stirring and retrieval operations will be performed with special equipment consisting of a pumping set bound to a telescopic arm whose base is fastened to the ceiling of the building. The solidification machine will be the MOWA designed by Nukem. A shed with an appropriate ventilation system will ensure the static and dynamic containment during the operations.

The conditioning process has been fully qualified to meet the requirements of Technical Guide 26 and to ensure the transportability of the conditioned waste according to IAEA Safety Guides; in particular appropriate tests were made on the cement+waste matrix and on the final conditioned waste; concrete shields will be added to the drums obtained from granular resins and sludges to meet IAEA requirements for transportation.

The chosen solidification formulae are shown in table II.

The production of 1240 drums of 400 litres is forecast, the total dose commitment has been evaluated as 0.086 Man-Sv.

The definition of contracts for the performance of this activity is under way.

Type of waste	Waste Kg	Cement Kg	Water/cement ratio	Dry waste in the matrix [%]	
Resins	301	443	0,49	11,5	
Sludges	281	463	0,49	7,5	
Concentrates and decon. solutions	276	560	0,4	7,3	

Table I	l
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3.3 OFF-GAS STACK

One of the operations needed to to bring the plant in line with the CPP state is the total demolition of the off-gas stack. The stack, in fact, is near the reactor building, the maintenance operations to keep it in good condition would be expensive and it would serve no useful purpose during the CPP period.

The stack is about 92 m high, with a base diameter of 4.8 m, its above-ground weight is about 900 ton. The total estimated radioactivity is 67 MBq and is concentrated in the first few millimeters of the internal surface.

A preliminary design report for the demolition of the stack has been submitted to the control authority. According to this design the internal surface will be scarified for 1 cm with a machine operated by remote control. The stack will then be demolished in one piece cutting the concrete at its base with another machine operated by remote control. The internal scarification should allow the release of the remaining concrete without radiological constraints.

A detailed design will be started as soon as the control authority gives us a preliminary positive opinion.

4 CAORSO AND TRINO

Caorso NPP was equipped with a single reactor, of the BWR Mk II type, with an electrical output of 860 MW. The plant has operated from 1981 to 1986 (4 cycles) producing about 28 TWh. Trino NPP was equipped with a single reactor, of the PWR type, it has operated from 1965 to 1987 producing about 24 TWh. Both plants were taken out of service by governmental decision in 1990.

A general decommissioning plan for both facilities, submitted in 1991, has still not been officially approved. Meanwhile the activities for the release of the new operation licences are under way; the technical documentation which will back-up the request will also comprise, as required by the control authorities, the full radioactive inventory and a general design of the operations needed to bring the plants in line with the CPP state.

Meanwhile a few activities are being performed under the existent licence. Trino reactor has been de-fueled and the irradiated fuel is being shipped to reprocessing facilities, the shipments should be ended within 1994. A Detailed Design Report has been submitted to the control authorities for resin conditioning (ILW).

De-fueling of Caorso reactor has still not been authorized. A Detailed Design Report has been submitted to carry out a few modifications needed for off-site shipment of irradiated fuel.

DECOMMISSIONING PROJECT MANAGEMENT: THE JAPAN POWER DEMONSTRATION REACTOR DECOMMISSIONING PROGRAM

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Abstract

The Japan power demonstration reactor (JPDR) decommissioning program is in progress of developing new technology for reactor decommissioning and collecting various data on project management and performance of developed dismantling devices. The experience and the data obtained from the JPDR decommissioning program are expected to contribute to future decommissioning of commercial nuclear power plants.

1. Introduction

Since the first generation of electricity by the Japan Power Demonstration Reactor (JPDR) in 1963, the utilization of nuclear power has been increasing in Japan. As of the end of September, 1992, 41 nuclear power plants are operating with 33,279 MW electricity generation, and the nuclear power shares almost 27% of nation's electric supply.⁽¹⁾ However, some of these nuclear power plants are expected to terminate their duty life in the near future.

In consideration of future decommissioning of aged nuclear power plants, the longterm program issued 1981 by the Japan Atomic Energy Commission (JAEC) pointed out that wide ranging technology for future decommissioning of commercial nuclear power plants should be developed and demonstrated through the JPDR decommissioning program in Japan Atomic Energy Research Institute (JAERI)⁽²⁾.

The JPDR decommissioning program⁽³⁾ was initiated in 1981 under contract with the Science and Technology Agency (STA) in Japan. The JPDR decommissioning program consists of two major phases; Phase 1 began in 1981, aiming at developing the technology necessary for reactor decommissioning. Phase 2 began in 1986, actual dismantling of the JPDR to reach green field condition, that is, stage 3 in IAEA definition, using the technology developed in Phase 1. So far the dismantling activities are in progress successfully, and various data on the dismantling activities have been collected and accumulated in the decommissioning database. These data are analyzed for future decommissioning of commercial nuclear power plants.

This paper describes the dismantling activities, the management systems, the data analysis and the lessons learned relating to the JPDR decommissioning program.

2. JPDR Decommissioning Program

2.1 JPDR

The JPDR is a BWR-type (45 MWt) demonstration reactor. It started to generate electricity for the first time in Japan in October 1963. In 1972 the power was increased to 90 MWt for enhancement of neutron irradiation capability. The JPDR was shut down in March, 1976 due to several problems such as cracking on the nozzle of in-core monitor tubes, the failure of control rod drive mechanism and other complications. Table I shows the major specifications, operation history and radioactive inventory of the JPDR.

The residual radioactive inventory in the JPDR is estimated to be approximately 130 TBq as of April 1988. Almost all radioactive inventory (99.9%) remains in the reactor internals, the reactor pressure vessel(RPV) and the biological shield concrete.

2.2 Development of Decommissioning Technology

The technology necessary for reactor decommissioning was developed covering eight areas in Phase 1 of the JPDR decommissioning program. Table II shows items of the research and development program. In particular, considerable efforts were made to develop remotely operated cutting tools to minimize radiation exposure to workers in dismantling highly activated components⁽⁴⁾⁻⁽⁶⁾. For example, underwater plasma arc and underwater arc saw cutting systems were developed to be applied to dismantling of reactor internals and the RPV, respectively. These were designed not only to dismantle the JPDR, but to take into consideration the applicability to commercial nuclear power plants as well.

2.3 JPDR Decommissioning Plan

Actual dismantling of the JPDR (Phase 2) began December 1986 using the developed techniques. The objectives of dismantling the JPDR are:

- (1) to demonstrate the techniques developed in Phase 1,
- (2) to obtain experience on dismantling activities, and
- (3) to establish decommissioning database,

for future decommissioning of commercial nuclear power plants.

The primary considerations for the dismantling work are safety of workers and prevention of release of radioactive materials. A local ventilation system and underwater

Specifications	
Type of reactor	BWR
Thermal power	90MWt (45MWt initially)
Pressure vessel material inner diameter height thickness Biological shield material thickness Reactor enclosure inner diameter height	ASTM-A302-56GrB 2.1 m 8.1 m 7 cm reinforced concrete 1.5 to 3 m 15 m 38 m
Operation history Operation time Output of electricity	17,000 hours 1.4 × 10 ⁶ KWH
Radioactive inventory	131 TBq (as of April, 1988)
Reactor enclosure inner diameter height Operation history Operation time Output of electricity Radioactive inventory	15 m 38 m 17,000 hours 1.4 × 10 ⁶ KWH 131 TBq (as of April, 1988)

Table I Major specifications, operation history and radioactive inventory of the JPDR

Items	Techniques			
Radioactive inventory estimation	Calculation technique Measurement technique			
Measurement of radioactivity inside pipe with non-destructive technique	Mini-detector Scanning device control system			
Reactor disassembly and demolition	Plasma arc Arc saw Rotary disk knife Shaped explosive Abrasive-water jet Diamond sawing and coring machine Controlled blasting technique			
Decontamination for rector decommissioning	System decontamination Concrete surface removal Decontamination of disassembled components			
Radioactive waste treatment transportation and storage	Containers for dismantling waste Volume reduction of metal waste Cement solidifaction of concrete waste Fixation of surface contamination			
Radiation control	Remote high dose-rate measuring instrument Dust monitor Contamination inspection apparatus Waste package contamination and dose rate monitor			
Remote handling	Underwater manipulator system			
Decommissioning system engineering	Computer code and data base			

dismantling machines operated by remote handling are used to minimize radiation exposure to workers, while building walls are used as a confinement boundary for the release of radioactive materials. The dismantling schedule made in advance shows that the critical-path is strongly related to dismantling highly radioactive components such as reactor internals and the RPV. All buildings will be demolished and the site will be landscaped by 1995.

2.4 Dismantling Activities

(1) Machinery around the reactor

On the first stage of the JPDR dismantling activities, the machinery around the reactor in the reactor building was removed to provide necessary space for installation and operation of the large remote operated cutting tools to dismantle the JPDR reactor itself.

For example, the equipment in the forced circulation system was dismantled so that the space could be used for a packaging area. The equipment was cut with hand tools such as an oxygen torch and a band-saw, because the radiation dose rate at the working area was low enough for safety of workers.

(2) Reactor internals

The reactor internals were removed by the underwater plasma arc cutting system. The plasma torch was operated in the most cases by a mast type manipulator having four degrees of freedom. Otherwise, the master-slave robotic manipulator was used for handling the plasma torch to demonstrate and verify its newly developed robot technology. First, each reactor internal was removed from the RPV wall, the cut pieces were then transferred underwater to the spent fuel storage pool through the canal. These pieces were cut into smaller segments suitable for packaging using another underwater plasma arc cutting system installed in the spent fuel storage pool. Figure 1 displays the schematic diagram of the underwater plasma arc cutting systems. During the cutting operations, aerosols and gases were exhausted by local ventilation systems, which consist of a green house on the service floor and air curtain made by air blowing/exhausting system above the water surface.

(3) Pipes connected to the reactor pressure vessel

After removing the reactor internals, the pipes connected to the RPV were dismantled using shaped explosives, rotary disk knifes and conventional cutting tools. First, the concrete blocks in the pipe penetrations of the biological shield concrete were dismantled to prepare for removing the pipes. The shaped explosives were used under both in-air and underwater conditions. To dismantle the water level monitoring system pipe, a transporter of the explosives was used as shown in Fig.2. For other pipes, the explosive was positioned manually using a charging stick from outside the biological shield concrete.



Fig.1 Schematic diagram of the underwater plasma arc cutting systems



Fig.2 Schematic diagram of the shaped explosive cutting systems

Two rotary disk knife cutting systems were fabricated, one for cutting the 12-inch pipe of the forced circulation system and the other for cutting the 4-inch pipe of the reactor feedwater system. The cuttings were successfully performed except for an elbow of the forced circulation system. The actual inside diameter of the elbow was smaller than that shown in the "as-built" drawings. Because the design of the knife was based on the diameter shown on the drawing, the knife cutting head could not be inserted into the elbow. Finally the elbow was cut by conventional tools.

(4) Reactor pressure vessel

The RPV was dismantled using the underwater arc saw cutting system, after removing the pipes connected to it. Before installing the underwater arc saw cutting system, a temporary cylindrical water tank was installed in the space between the RPV and the biological shield concrete. The tank was filled with water for cutting the RPV underwater. The top flange portion of the RPV body was cut vertically into 9 pieces. The other parts were cut into 8 horizontal pieces and 9 vertical pieces. Figure 3 shows the underwater arc saw cutting systems together with the cut off lines of the RPV. A specially developed device was used to hold and lift the cut-off pieces. The pieces were washed by a water jet above the water surface before being put into containers. No significant radioactivity in the working environment was observed during the dismantling activities.



a. underwater arc saw cutting system



Fig.3 Schematic diagram of the underwater arc saw cutting systems together with the cut off lines of the RPV

(5) Biological shield concrete

Following the dismantling of the RPV, the activated portion of the biological shield concrete was dismantled using both the diamond sawing/coring and the abrasive water jet cutting techniques. Figure 4 shows schematic diagram of the biological shield concrete, indicating radioactivity levels and the area classification. First the diamond sawing/coring, next the abrasive water jet cutting were applied to cut the upper and lower half of the activated biological shield concrete, respectively. The dust generated during cutting was exhausted by a temporary dust collector, and the slurry produced by the water jet cutting was treated by the waste processing systems.

The controlled blasting technique has been applied to the rest, which is only slightly activated. The layer of 40 cm width from the inner surface of the biological shield concrete was dismantled in the first stage of this work. The wastes were put into containers for storage. The outer layer of the biological shield concrete will be dismantled by both the controlled blasting and conventional tools. The concrete and reinforcing steel bars in the outer layer will be disposed in the test shallow land burial place in JAERI's site.

Table III lists the major milestones of the JPDR dismantling activities. In parallel with dismantling the biological shield concrete, decontamination of the building inner surface has been started since the end of 1992. Various techniques such as steel blasting, and scabbler are applied to the decontamination work. After confirming that there is no



Fig.4 Schematic diagram of the JPDR biological shield concrete, showing radioactivity levels and the area classifications

Components	Techniques	Activity period	Remarks		
Equipment around reactor	Conventional tools	Dec. 1986 - Oct. 1987	Continued with next phase		
Equipment in dump-condenser bld.	Conventional tools	Dec. 1986 - Dec. 1987			
Reactor Internals (I)	Plasma arc & robotic manipulator	Aug. 1987 - Mar. 1988	Three reactor internals		
Equipment in spent fuel storage bld.	Conventional tools	May 1988 - Aug. 1988			
Reactor Internals (II)	Plasma arc & mast-type manipulator	Nov. 1988 - Jan. 1990			
Pipes connected to RPV	Shaped explosives, disk cutter, etc.	Aug. 1989 - Mar. 1990			
RPV	Arc saw & mast-type manipulator	Sep. 1989 - Oct. 1990			
Equipment in turbine bld.	Conventional tools	Apr. 1990 - Mar. 1992			
Biological shield (I)	Diamond sawing/coring	Sep. 1990 - Mar. 1991	Highly radioactive part (upper)		
Biological shield (II)	Abrasive water jet	Apr. 1991 - Oct. 1991	Highly radioactive part (lower)		
Equipment in rad-waste bld.	Conventional tools	Jun. 1991 - Mar. 1992			
Biological shield (III)	Controlled blasting	Feb. 1992 - Sep. 1992	Slightly radioactive part (continued)		

radioactivity, all building structures will be demolished, and the site will then be renovated and landscaped.

3. Management For Dismantling Activities

3.1 Safety Assurance

At the beginning of the JPDR dismantling activities, the Japan Nuclear Safety Commission (JNSC) published the report "Philosophy of Safety Assurance during Reactor Dismantlement – Dismantling JPDR"⁽⁶⁾. This report states guidelines to ensure safety of the JPDR dismantling activities. The enforcement plan of the JPDR dismantling was made on the basis of this report, and it was submitted to the Japanese STA in July 1986. After JNSC's authorization of this plan, the JPDR dismantling started. The key principles of the enforcement plan are to ensure the safety of the workers and to prevent radioactive materials from being released.

The JPDR dismantling activities have been done according to the key principles. In addition, the Quality Assurance (QA) committee was organized by the experts in JAERI. The main tasks of the QA committee are to check up the enforcement plan of each dismantling activity, and to review the report to be submitted to STA. Especially, when applying the new technology to the dismantling activities, the activity plan was examined in detail by the QA committee with regard to the safety of workers and efficient work procedures.

3.2 Organization

The JPDR decommissioning program has been carried out basically by the for divisions and one laboratory :

- JPDR administration division,
- Decommissioning program management division,
- Reactor decommissioning operation division,
- Decommissioning waste management division,
- Decommissioning technology laboratory, and
- Radiation control devision.

The actual dismantling activities are managed by the staffs in the rector decommissioning operation division. Before implementing the dismantling activities, the report describing details of each dismantling activity is prepared by the staffs in both the decommissioning program management and reactor decommissioning operation divisions. The report is submitted to STA for authorization of the work plan in each step of the dismantling activities. The report contains work specifications and schedules, characteristics of the components to dismantled, weight of waste arisings and so on. After accomplishing each process of the dismantling activities, the results of the work is also reported to STA, and examined for compliance with the plan submitted before. The decommissioning waste management division and the decommissioning technology laboratory are mainly in charge of waste management and data collection and analysis, respectively.

3.3 Radiation Protection Control

Radiation protection control is one of most important considerations in the program. Dose equivalent rate and radioactive concentration are measured daily and reported by health physicists. Each work plan is made on the basis of this measurement and work areas are classified into various categories. For example, radioactive contamination is classified into three groups: C-1(less than 4 Bq/cm²), C-2(4 to 40 Bq/cm²) and C-3(more than 40 Bq/cm²). Work conditions, such as respiratory protection and personal protective clothing, are then decided according to this categorization. Data on radiological safety

such as dose equivalent rate, radioactive contamination and radioactive nuclide concentration in air are measured periodically in the work areas. The category is changed as dismantling activities are accomplished and the method of dismantling components is sometimes reformed under considerations of safety of workers and efficient work accomplishment. Radiation exposure to workers is also measured daily using alarm pocket dosimeters, as will be described later.

3.4 Waste Management

At the early stage of planning the dismantling activities, characteristics of decommissioning wastes were evaluated based on calculations and measurement. On the estimations, more than 4000 samples were taken to evaluate the amount of radioactive contamination in the whole JPDR facility. The total amount of 30,000 tons of solid wastes including approximately 4000 tons of low level radioactive wastes were expected to be produced in dismantling the JPDR. In the actual dismantling work, dismantled components were classified into four levels (or more detailed levels in some cases), on the basis of on-site measurement of radioactivity. The components are put into 200 liter drums, steel containers $(1m^3 \text{ or } 3m^3)$, or shielded containers according to their radiation levels. The amount of wastes estimated is verified through the comparison with the actual data associated with each radiation level.

In addition to the solid wastes, gaseous and liquid wastes are also produced from the dismantling activities. Gaseous wastes are filtered and exhausted through the stack after confirming that the radiation level is less than $3x10^{-6}$ Bq/cm³. Liquid wastes are treated by the water treatment systems, which were used during the operation of the JPDR. After confirming that its radioactivity is less than $4x10^{-1}$ Bq/cm³, it is diluted with water to be one hundredth in radioactivity, then discharged to the Pacific ocean.

4. Data Collection And Analysis

4.1 Data Collection And Retrieval Systems

Information about the JPDR dismantling activities has been collected and accumulated in the decommissioning database. This database is to be used for :

- (1) managing on-going JPDR dismantling activities,
- (2) verifying the developed Code Systems for Management of Reactor Decommissioning (COSMARD)⁽⁷⁾, and
- (3) planning future decommissioning of commercial nuclear power reactors.

The data have been collected on data collection and retrieval systems, that use the JAERI mainframe (FACOM-M780) and minicomputers. In addition, information about machine performance and operability are also collected when newly developed decommissioning techniques are applied to the dismantling activities.

Figure 5 shows concept of the data collection and retrieval systems. The JPDR dismantling data are basically grouped into three categories: radiation control data, dismantling operations data, and waste management data. The radiation control data, including working hours and external dose of workers, are collected using magnetic identification cards and pocket dosimeters. When passing through the gate of radiation control area, workers are required to keep records of entering/exiting time and external dose by inserting each work's magnetic card and pocket dosimeter into a magnetic reader connected to a minicomputer. The other data have been collected in the form of description made by worker's supervisors. The descriptions are submitted daily to the data collection staff in the systems engineering group. The other data such as machine performance and unexpected occurrence have been stored as a form of documentation.



Fig.5 Concept of the data collection and retrieval systems in the JPDR decommissioning program

4.2 Use of Data for The Project Management

The data collected during the dismantling activities are used for managing the JPDR dismantling work.

As for the radiation control data, cumulative dose and other information, for example, date of medical examinations are checked for compliance with regulations. When collective dose itemized by a work permission number exceeds a designated value, the reason is to be analyzed. The analyzed results are used to modify the plan of other dismantling activities if necessary. With regard to waste management data, dismantled components are basically managed by linking to containers. Each container is identified by a designation number which, in turn, can provide the following information: source and quantity of waste, date of transportation and storage, radioactivity level, processing method, etc. Figure 6 shows histogram of major management data collected in the dismantling activities along with the major activities and major project milestones so far. As described before, the data on project management such as waste generation, manpower expenditure and radiation exposure to workers are analyzed and reported as a periodical documentation. Figure 7 also shows the numbers of containers with different types used by the end of March, 1992.

4.3 Data Analysis

(1) Manual dismantling work

Among the information about the JPDR dismantling work, labor hours expended in doing dismantling work are useful so that it will be applicable to evaluated unit factors and productivity loss factors in estimating labor hours.

Figure 8 shows the relationships between the weight of dismantled components and labor hours needed in dismantling the components such as cables, pumps and piping. The unit factors evaluated are shown to be different for different components types. In addition, it was found by analyzing dismantling activities that the unit factors are changed depending on the dismantling procedure. For example, it was initially assumed that



Fig.6 Histogram of major management data collected in the dismantling activities along with the major activities and project milestones



Fig.7 Number of containers used in the JPDR dismantling activities (as of the end of March, 1992)



Fig.8 Relationships between the weight of dismantled components and labor hours needed in dismantling cables, pumps and piping

pumps would be removed by cutting them into small pieces. However, in the actual dismantling activities, small pumps were removed in one piece. The manpower expenditure was found to be less than the estimate. In the same manner, various unit factors were verified to be practicable in accordance with component types.

From the data analysis, it was also found that worker productivity is reduced when personnel respiratory protection, personnel protective clothing, scaffolding and so on are used. The more manpower was expended in a congested area as shown in the figure. Although it was assumed that productivity would be reduced due to respiratory protection and protective clothing in congested areas, about twice the labor hours were expended compared to other areas.

(2) Remote dismantling work

The data collected on various dismantling activities using remotely operated cutting machines were also analyzed to characterize the dismantling activities. Figure 9 shows the data on manpower expenditure, collective dose of workers and waste generation in dismantling reactor internals, the pipes connected to the RPV, the RPV and highly activated portion of the biological shield concrete. The work was done with newly developed remote cutting machines. The work efficiency, that is, the ratio of manpower expenditure of the dismantling activities to the weight of the dismantled components was calculated to be in the range of 2000–500 man-hours per ton. This was quite large compared with that of hands-on dismantling activities. The efficiency of hands-on work was approximately 80 man-hours per ton on average in the reactor building. In spite of the low work efficiency, collective dose of workers was low enough to meet safety requirements. The remotely controlled dismantling systems including the local ventilation was, therefore, proved to be effective to minimize radiation exposure to workers.

5. Lessons Learned

More than 20 decommissioning projects are underway around the world⁽⁶⁾. The JPDR decommissioning program is rather forward among them except the Shippingport decommissioning project⁽⁷⁾, and it has a unique feature to develop various kinds of dismantling technologies and to dismantle the facility by segmenting them into small pieces for packaging. Various useful lessons were drawn from the actual dismantling work. The description of the lessons learned is given in terms of three major categories as follows.

- (1) dismantling techniques
 - The dismantling techniques developed in the JPDR decommissioning program were proved to be useful for dismantling radioactive components, especially underwater plasma arc and underwater arc saw cutting techniques were effective to prevent contamination being released as well as to minimize radiation exposure to workers.
 - Remote cutting techniques used in the dismantling work have to be improved to be more simple and multi purpose for use in future.

(2) dismantling work

- The dismantling work has been well managed to minimize the radiation exposure to workers. This is due to precise estimation of the radioactive inventory and daily measurement of the dose rate at work areas.
- The dismantling work has been done with no serious problems as scheduled. Detailed planning of work activities was attributed to the success of the dismantling work. The quality assurance program has played an important role in checkup of worker's safety.



- Fig.9 Management data collected from the dismantling activities in the reactor building
- (3) waste management
 - All wastes have been stored in waste storage facility in JAERI's site. However, it must be necessary to consider minimizing the volume of the radioactive waste by various methods such as melting and super compaction.
 - Adequate treatment of secondary products such as slurry and abrasives will be necessary to reduce waste arisings in dismantling biological shield concrete.

6. Concluding Remarks

The JPDR decommissioning program was commenced in April 1981 to reach a green field condition. In the early stage of this program, various decommissioning technology was developed not only to dismantle the JPDR but to demonstrate these for

future decommissioning of commercial power plants as well. Following the technology development, the actual dismantling activities started in December 1986.

The JPDR decommissioning program have been progressing successfully without any serious problems. Adequate project management has been applied to the program for safety assurance, radiation protection and waste management. Various cutting tools with remote operation developed in the early state of the program have been used successfully in dismantling the highly activated components such as the reactor internals, the RPV and the biological shield concrete. These techniques were proved to be useful to minimize the radiation exposure to workers. In particular, underwater cutting tools such as the underwater plasma arc and the arc saw cutting system were very effective to prevent contamination being released as well as minimize the radiation exposure to workers.

In the dismantling activities, various data on project management such as manpower expenditure, radiation exposure to workers and waste arisings are being collected continually and these are stored in the decommissioning database. The database will contribute to planning future decommissioning of commercial nuclear power plants.

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STUDY ON THE CYCLIC VOLTAMMETRIC BEHAVIOUR OF V(III) AND V(II) IN THE LOMI DECONTAMINATION SYSTEM

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Abstract

A fundamental study was conducted to analyze the mechanism of decontamination, as a part of the efforts to develop regenerative LOMI decontamination process. Most of the chemical reactions between picolinate and metal ions such as V, Fe, Ni and Co in the LOMI decontamination system have been known fast chelation reactions so that the chemical reactions can be represented with thermodynamic equilibrium.

The chemical species in the LOMI decontamination system above pH 5 has not been well investigated. Reduction mechanism of the V (III) - picolinic acid system based on the results obtained from cyclic voltammetry with various scan rates was investigated.

1. INTRODUCTION

Most of the chemical reactions between picolinate and metal ions such as V, Fe, Ni, and Co in the LOMI decontamination system have been known fast chelating reactions so that the chemical reactions can be represented with thermodynamic equilibrium.

The electrochemical properties of the V^{3+}/V^{2+} -NTA (nitrilotriacetic acid) system have been studied [1]. From the $E_{1/2}$ value and Nernst equation, Meier et al. evaluated the formation constant for V(II)NTA(H₂0)⁻. Kato et al. [2] studied the polarographic behaviour of V(III)-HEDTA (Nhydroxyethylethylenediaminetriacetic acid). They proposed the electrode mechanisms for V(III)-HEDTA and calculated the equilibrium constants by a curve fitting method. Jung et al. [3] studied reduction mechanism of VO⁺²-DTPA (diethylenetriaminepentaacetic acid) at a mercury electrode in aqueous solution. They proposed that reduction reactions of the VO²⁺-DTPA system occurred through an ECE reaction sequence.

The chemical species in the LOMI decontamination system above pH 5 has not been well investigated. In this study, the reduction mechanism of the V(III) - picolinic acid system based on the results obtained from cyclic voltammetry with various scan rates was investigated as a part of the efforts to develop the regenerative LOMI decontamination process.

2. EXPERIMENTAL

Vanadium chloride (VCl₃) was used as a source of V(III). Picolinic acid was used as a ligand. Sodium perchlorate monohydrate (NaClO₄. H₂O) was used as a supporting electrolyte. NaOH and HClO₄ were used to control the hydrogen ion concentration.

Cyclic voltammogram was taken with an EG & G PARC Model 273 potentiostat and Model 270 software. Hanging mercury drop electrode was used as a working electrode. The surface area of a mercury drop was maintained at 2.2 ± 0.05 mm². SSCE (sodium chloride saturated calomel electrode) was used as a reference electrode. Platinum electrode was used as a counter electrode. All measurements were carried out at $20\pm0.1^{\circ}$ C and under highly pure nitrogen atmosphere.

3. RESULTS AND DISCUSSION

A set of cyclic voltammogram of an aquous solution of V(III)- picolinate at constant hydrogen ion concentration was recorded by varying the scan rate.

Fig.1 shows a typical cyclic voltammogram of this system. There are 4 reduction peaks (I_c , II_c , II_c , IV) and 2 oxidation peaks (I_a , II_a). IV seems to be related to the reduction of hydrogen ion, I_c and II_c have their own oxidation couples (I_a , II_a).



POTENTIAL (V vs SSCE)

Fig.1. A Typical Cyclic Voltammogram of V³⁺-Picolinate System. [V³⁺] = 3 mM, [Pic. Acid] = 18 mM, μ = 0.5 M NaClO₄, pH = 5.5, HMDE = 2 2 mm², Scanning Rate = 10 V/s.

Effects of scan rate on the cyclic voltammogram are shown in Fig.2. When the scan rate is relatively low (1 V/sec), III_c is not shown. As the scan rate increases, III_c becomes to appears. In case of peak II, II_c moves to negative direction and II_a moves to positive direction with increasing the scan rate. As shown in Fig. 2(d), III_c begins to appear at -1.1V(vs.SSCE). This peak grows continuously as the scan rate varies from 20 to 100 V/sec. The appearance of III_c changes relative intensity between peak current of the first curve $[i_{p,l(a)}]$ and that of the second curve $[i_{p,l(b)}]$.



Fig 2 Cyclic Voltammograms of V³⁺-Picolinate System at Various Scanning Rates $[V^{3+}]=3 \text{ mM}$ [Pic Acid]=18 mM $\mu=0.5$ M Mclo4, pH=5.5 HMDE=2.2 mm² Scanning Rate (a) 1 V/c (b) 2 V/s, (c) 5 V/s, (d)10 V/s, (e) 20 V/s, (f) 30 V/s, (g) 50 V/s (h) 100 V/s

Where lilc does not appear, $i_{P,I}(A) > i_{P,I}(B)$

Where IIIc appears, $i_{p,I}(A) < i_{p,I}(B)$

Fig. 3 is a plot of $i_{P,a,1(A)}/i_{P,c,1(A)}$ against the square root of scan rate. At lower scan rate region, the ratio of peak current is near unity. But, deviation from unity increases steeply as the scan rate increases. Fig.4 is a plot of peak potential and half wave potential against the square root of scan rate. The difference between II_c and II_a, [$\Delta Ep(11)$], increases with increasing the scan rate. But, the scan rate does not affect $\Delta E(1)$. The value of $\Delta E(1)$ is around 60mV.

Cyclic voltammograms of an aquous solution of V(III)- picolinate were obtained from the variation of scan rate and hydrogen ion concentration. Fig. 5 is a plot of half wave potential value for electrode reactions I,II against the hydrogen ion concentration. Redox couple of peak II becomes to appear above pH 3.5, and $E_{1/2}(II)$ decreases by the increase of pH.

Reactions at the electrode are divided into reversible, quasi reversible and irreversible by the existence of redox couple in the cyclic voltammogram. As shown in Figs.1 and 4, electrode reaction I is reversible and number of electron engaged



Square Root of Scanning Rate (V/sec)1/2

Fig. 3. The Peak Current Ratio(i_{P.a}(1)/i_{P.c}(1)) vs. Scanning Rate for Peak I of V³⁺-Picolinate_System: [V³⁺]=3 mM, [Pic. Acid]=18mM, μ=0.5 M NaClO₄, pH=5.5, HNDE=2.2 mm²



Fig. 4. Effect of Scanning Rate on Half Wave Potential($E_{1/2}$) and Peak Potential(E_p) of V³⁺-Picolinate System: [V³⁺]=3 mM, [Pic, Acid]=18 mM, μ =0.5 M NaClO₄, pH=5.5, HMDE=2.2 mm².



Fig.5. Effect of pH on Half Wave Potential($E_{1/2}$) for Peak I and II of V³⁺-Picolinate System: [V³⁺]=3 mM, [Picolinic Acid]=18 mM. μ =0.5 M NaClO₄, HNDE=2.2 mm²

in this reaction is one. The reaction II is quasi reversible because $\Delta E_p(II)$ increased by the increase of scan rate. But, III_c had no oxidation counterpart, the reaction occurred at this point is regarded as irreversible(Fig.1). The reaction product at III_c affects the reaction I because the appearance of IIIc affected the relative intensity of the peak current of the first curve[i_P,I(A)] and second curve[i_P,I(B)]. This means that the product at III_c will be changed to the species which is reacted at I_a.

Jones and Colvin[4] reported that standard reduction potential of V^{3+} to V^{2+} is -0.255V(vs. SHE). Therefore, the reduction peak of free V^{3+} to V^{2+} does not appear in this experimental region. Chemical species and their dissociation constants in V(II)/V(III)-picolinate system at 4<pH<5 were observed by Smee[5]. He reported that complexes in solution were as followings:

V(III) complex: $V(pic)_1^{2+}$, $V(pic)_2^{+}$, $V(pic)_3$, $V(OH)(pic)_2$. V(II) complex : $V(pic)_1^{+}$, $V(pic)_2$, $V(pic)_3^{-}$

Considering the dissociation constants, main chemical species in this system are V(pic)₃, V(OH)(pic)₂, V(pic)₃⁻. Since redox couple of I appeared at pH > 2.0 and half wave potential($E_{1/2}$) for reaction I is almost same in the pH range from 3 to 5.5(Fig.5), the electrode reaction I is as follow(pH=5.5):

Peak I, $V(III)(pic)_3 + e^- - V(II)(pic)_3$

Above reaction is well coincides with the result investigated by Smee. For a reversible reaction, the relationship between peak current and scan rate is described by the next equation.

$$i_p = 2.69 \times 10^5 n^{3/2} \Lambda D^{1/2} C_0 v^{1/2}$$

where i_p is the peak current(ampere), n is the number of electrons per species oxidized or reduced, A is an area of electrode(cm²), D is the diffusion coeffecient (cm²/sec), C₀ is the concentration(mol/cm³) and v is the scan rate(V/sec).

The diffusion coefficient calculated from the above equation for peak I was in the range of $(6\pm0.5) \times 10^{-6} \text{ cm}^2/\text{sec}$.

Because redox couple of II appeared at pH>3.5, the reacted chemical species is assumed to be $V(OH)(pic)_2$. From the investigation of the pH effect on $E_{1/2}(II)$, hydrogen ion is thought to be involved in the reaction II(Fig.5). II_a coincided with I_a at higher scan rate(above 30 V/sec). This means that the product at II_c will be changed to the species which is reacted at $I_a(Fig.2)$. However, II_c has its own reduction counterpart at lower scan rate. From the above considerations, next reactions are suggested at II and in solution(pH=5.5).

Peak II, $V(III)(pic)_2(OH) + H^* + e^- \longrightarrow V(II)(pic)_2 + H_2O$

Chemical reaction, V(II)(pic)₂ + Pic⁻ ---- V(II)(pic)₃

A fact that Fe(III)-EDTA(ethylenediaminetetraacetic acid) and analogous complexes form a dimer in addition to a hydroxo-complex in weak acid solution is well known [6,7]. The presence of a hydroxo-complex in weak acid solution was also considered in the V(III)-picolinate system because formation of dimer was supposed. Although there are few reports on the equilibrium constant between monomer and dimer, electrode reaction at peak III_c reveals the existence of dimer(not shown in Fig. 5, but only reduction peak appeared in the pH range above 5.0 and scan rate above 20 V/sec).

On the basis of this assumption and the fact that appearance of III_c affects the relative intensity of the peak current of first curve $[i_{P,I(A)}]$ and second curve $[i_{P,I(B)}]$, next reaction may happen at III_c and in solution(pH= 5.5).

Peak IIIc, 1/2 [V(III)(pic)₂(OH)]₂ + H⁺ + e⁻ ----> V(II)(pic)₂ + H₂O

Chemical reaction, $V(II)(pic)_2 + Pic^- \longrightarrow V(II)(pic)_3$

Conclusion

Three types of reduction reactions happened in V^{3+} -picolinate system(pH = 5.5). Reaction I is reversible, reaction II is quasi reversible and reaction III_c is is irreversible. The reaction at III_c causes the reduction of V^{3+} as totally irreversible.

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DECOMMISSIONING OF NUCLEAR INSTALLATIONS IN THE RUSSIAN FEDERATION AND NEWLY INDEPENDENT STATES OF THE FORMER USSR

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Abstract

The results of NPP decommissioning in 1989 - 1992 have been summed up. Particular emphasis is laid on pre-decommissioning stage, the stage of all finally shutdown NPP units in CIS.

Special attention is given to feasibility of VVER units, recycling of NPP metals and working out specifications for residual activity concentration content in NPP materials, thus making their unrestricted use possible, to the approaches to designing of bioshielding for the next generation NPP and to the choice of their structural materials.

Fundamental and applied research conducted in the CIS over the last four years is reviewed and it is stressed that fundamental research can change the approach to the solution of the problem for different nuclear plants types.

The recommendations based on the results of the investigations are given and the general directions for research and practical work including those conducted jointly with international community are outlined.

1. INTRODUCTION

In three CIS countries seven NPP units (NPPU) have been finally shutdown (Table 1)'. The operation of a number reactors in Russia producing plutonium for military purposes has also been stopped [2].

Unfortunately, because of the privity imposed on military enterprises, the experience gained in recent years by the CIS experts in particular is not widely known to the scientists and engineers, wich may sometimes result in the duplication of the basic research work [2]. We hope that the establishment of the International Nuclear Industrial Forum, Moscow [3] will make it possible to solve this problem and to concentrate greater intellectual efforts and material resources on decommissioning.

It should be noted that all finally shutdown NPPU in the CIS are at "pre-decommissioning" stage, i.e. intermediate between "operation" and "decommissioning" (Table 1, [4,5]), that is why special attention was also given to improving the NPP Concept over the period concerning.

^{&#}x27; The fourth unit of Chernobyl NPP is not considered in this report as a "special point" among the shutdown NPP units, in spite of the fact that it is also a decommissioning problem [1].

TABLE	1
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Name	Reactor type, power(MWe)	Date of commissioning	Date of shutdown	Status (as of 1 inuary 1993)
RUSSIAN FEDERATI	LON			
Beloyarsk 1 Beloyarsk 2	LWGR, 100 LWGR, 200	1964 1968	1981 1990	Preparatory stage; buil ngs and equipment are partially isolated
Novovoronezh 1 Novovoronezh 2	PWR, 210 PWR, 365	1964 1969	1984 1990	Preparatory stage; fea bility studies on engineerinrg options and radiological inspection completed
UKRAINE Chernobyl 4	LWGR, 1000	1984	1986	Preparatory stage; uni is isolated by a protective envelope
ARMENIA				
Armenia 1 Armenia 2	PWR, 420 PWR, 420	1976 1979	1989 1989	Preparatory stage; fea ibility stydies on engineering options id radiological inspection completed

Current status of finally shutdown NPP in CIS

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2. PREPARATION TO DECOMMISSIONING FOR NPP WITH VVER REACTORS

Pre-decommissioning is structurally similar to commissioning stage of NPP life cycle (Fig. 1), though it may cause certain linguistic misunderstandings. The decommissioning Concept adopted earlier in the USSR and summarized in [6,7] differs from the strategies followed elsewhere [8] as it gives special attention to the stage concerned '. It should be noted, however, that this stage is concerned by the experts of many countries, e.g. in Finland [9], either as a separate one or as a part of "operation" and "decommissioning" stages.

Preparation to decommissioning includes:

- complete engineering survey of the actual state of buildings, structures and NPP equipment including engineering, radiological measurements, specifying radioactive waste estimates etc

- removal of fissile materials from the unit or the NPP (i.e. of nuclear fuel at different stages of its use and its "deposits")

- draining of the NPP systems or units containing liquid radioactive waste and its treatment

- decontamination of the systems and equipment, particularly those necessary for carring out further work or for utilization

- disposal of operation radioactive waste if the NPP is to be shutdown

- analysing the possibilities and preparation of standard equipment and NPP systems for use at the two final stage of life cycle

- designing and manufacturing of supplementary equipment and systems to be used at these stages.

Though pre-decommissioning activities at the first construction stage of Beloyarsk NPP with LWGR reactors was started earlier than at others CIS NPP, it was slowed down quite considerably after partial isolation of its units.

Remotely-controlled: complex for reactor core and housing dismantling designed for this purpose [10] will not only cost hundreds of millions roubles but will also take a long time for its manifactoring and development which in present economic situation makes the possibility of its creation doubtful.

The work at Beloyarsk NPP with the choice of proper technological processes, methods and ways should continue, however, both for attaining NPP's nuclear and radiological safety and for gaining experience in decommissioning a NPP with RBMK.

Preliminary work has also been started at plutonium production shutdown reactors in Russia requiring, within the framework of domestic strategy, the development of remotely-controlled tools.

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^{&#}x27; Another essential difference is including the NPP life extension direction into the general decommissioning Concept. But being the subject of another IAEA programme this direction will not be considered here.

Construction	Commissioning			Operati	on		Pre deco	eparation to ommissioning	De	commissioning
11	I	I //	I	f	I	I	I	11	I	
A	В	C	D	E	F	G	Н		I	K

A - B -- Construction B - C -- Commissioning C - H -- Operation -- Beginning of data selection for NPP closure D D - E -- Expert feasibility studies Е -- Establishing of criteria and NPP closure options selection E - F -- Analysis of the options -- Selection of the option F F - G -- Development of the project in accordance with the chosen option G - H -- Project updating Н -- Final shutdown H - I -- Preparatory stage to decommissioning or life extension I - K -- Decommissioning

FIG.1. NPP life cycle with its last stages

On the other hand, successful complete engineering survey of Armenian ' and the first construction stage of Novovoronezh NPP gave valuable information on radiation and strength characteristics of VVER NPPs. This extensive information is summarized in the papers and reports [10 - 15].

With the corresponding economic estimates the complete engineering surveys form the basis for feasibility (their structure being given in Appendix 1), the choice of directions and options of unit decommissioning being based on them (Table 1).

The problem of nuclear waste management at the end of NPPU life cycle is particularly difficult for any country. These difficulties were aggravated by the desintegration of the USSR into independent states. Thus, it took about two years to remove nuclear fuel from Armenian Republic, but the removal of other radioactive waste from independent Armenia seems to create an insurmountable problem.

3. RECYCLING OF NPP MATERIALS

Not only does recycling of NPP materials facilitate the waste burial problem but it partially offsets the expenditures on NPP pre-decommissioning and decommissioning due to the reuse of the valuable materials.

Over the period concerned the work was carried out along the two lines.

3.1. Remelting of low level activity metals

Low level activity metals remelting installations, i.e. induction furnaces, were designed independently in Russia and Slovakia with financial backing of the Joint Fund, their testing carried out colloborated at Beloyarsk and Bohunice NPP.

Up to 95 per cent of low level materials were proved to be returned into commercial use without further control. Complete technological process of metal recycling includes:

- decontamination with resulting low level scrap, liquid and solid radioactive waste

- radioactive metal melting producing metal ingots with activity lower than that of original material, accomplished by radioactive gas and higher level slug formation

- ingot remelting to produce cleaner metal with radioactive gas and higher level slug formation.

The first and the third stages of the process are not always necessary. One of them or both can be avoided: if the initial material activity is low melting process alone will be sufficient for obtaining ingots with activity corresponding to temporary specifications adopted in the former USSR.

Testing of remelting metal furnace at Beloyarsk NPP, its development and practical implementation in recent years proved the effectiveness of the process, and the delivery of improved furnaces to a number of NPPs in Russia is being scheduled.

^{&#}x27; It should be noted that the acute energy shortage problem in Armenia made its government take the decision to start up Armenia NPP again, though the reasons for its shutdown have not been eliminated [4].
3.2. Specifications for unrestricted material release

A new level of nuclear installations material use can be achieved after specifying their residual activity concentration limits.

This work was also conducted jointly by Russian and Slovak experts. It is based on minimal individual dose value (10 μ Sv) and collective dose equivalent (1.0 man·Sv) for the sources and practice that may be exempted from regulatory control according to IAEA and ICRP recommendations and on the most stringent scenarios used for such materials [16].

Russian experts have chosen the following among the most stringent scenarios.

For external exposure:

- a person lives in a building with recycled reinforcement of concrete in its walls, ceilings and floors

- a person is inside a massive vehicle
- a person works in a production area with machines made of recycled NPP metal or deals with NPP metal blanks
- a person wears a recycled metal bracelets or watch.

For internal exposure:

- a person drinks water out of a tank made of NPP concrete or metal.

The choice of scenarios was made by Russian and Slovak experts independently and we believe the quantitative agreement of the results of the calculations for similar (but not identical) scenarios to prove their quality.

To consider the most stringent conditions we assumed in our calculations that a person spends 24 hours in his house for many years, that a worker in the production area is in contact with the machines and blanks 8 hours a day, that a person drinks water out of the same tank all his life etc.

Due to such stringent scenarios and the criteria adopted, the values of activity concentration limits obtained are less than those of the experts of other countries (Table 2).

These calculations were used for compiling the draft standard "Nuclear Power Plant Unit Closure. Residual Radioactivity Grounds of Recycle Materials for Unrestricted Release".

4. FACILITATION OF NPP DECOMMISSIONING AT DESIGN STAGE

In the former USSR and in Russian Federation this work was conducted in the three main directions.

4.1. Supplements to regulating documents

The section concerning NPP decommissioning or closure were introduced into the adopted technical codes for NPP safety and sanitary rules.

This issue is considered in greater detail in the second document [17], with a special chapter devoted to this problem.

It lists the main requirements to the work to be carried out after the NPP final shutdown, which is necessary to make their decommissioning easier, at design stage in particular, as well as the requirements to designing and engineering to facilitate the work at the last stages of an NPP life cycle (Fig.1).

4.2. New approaches to NPP design

Many unique concepts of increased safety reactors appeared in Russia in recent years but their development, however, did not always take decommissioning problem into account.

_					
Nuclide	Metal	Struct. mater1al	Nuclide	Metal	Struct. material
Mn-54	3.0	4.5(-2)'	Ce-144	4.0	_
Co-57	8.3	-	U -238	2.0	4.0
Co-60	1.0	1.0(-2)	Np-237	1.0	0.1
N1-59	1.2(+3)	1.1(+4)	Pu-238	2.0	0.2
N1-63	7.4(+2)	-	Pu-239	0.1	0.15
Zn-65	4.0		Pu-240	0.1	0.15
Se-75	8.0	1.7(-1)	Pu-241	0.5	0.15
Sr-90	1.4	-	Pu-242	0.1	0.15
Nb-94	1.0	2.5(-2)	Pu-244	0.1	0.15
Nb-95	3.0	5.2(-2)	Am-241	0.1	0.15
Tc-99	3.2	5.2(-2)	Cm-244	2.0	0.30
Ru-106	5.0(+1)	-	Unknown c	0- 3.0	-
Ag-108m	4.0	-	mposition	of	
Ag-110m	4.0	1.0(-1)	d-nuclid	es ''	
Sb-125	9.0	-	Ca-41	-	3.3(+2)
Cs-134	2.0	-	Sm-151		6.6(+2)
Cs-135	1.3(+3)		Eu-152	. .	1.6
Cs-137	4.0	7.4(_2)	Eu-154	-	2.5(+2)
Ce-141	1.0(+2)	1.2	Eu-155	-	2.4(+2)
	/ = /	2			

Mass activity concentration limits for unrestricted exemption of materials from regulatory control, Bq/g

 $4.5(-2) = 4.5 10^{-1}$

'' For NPP materials only

The new requirements [17], however, imply new approaches to designing of next generation NPPs [18,19].

It was shown earlier [20] that the concrete amount of shielding of the NPPs constructed according to one of the Soviet concepts is several thousands tons more that that is practically necessary.

Therefore the suggested option of a monolithic-block shielding [19,20] while retaining its strength and protective capabilities will make it possible to reduce the volume of reinforced concrete to be buried and at the same time to reduce the collective exposure dose of the personnel carrying dismantling out as the time for shielding, first of all bioshielding, disassambling is considerably reduced, and the generation of high level activity dust and gases are eliminated.

4.3. Choice of NPP structural materials

Structural materials, both samples from shutdown NPPU and candidate materials were studied very intensively during this period. The investigations were carried out by means of irradiations at a research reactor. It was shown that a number of widely used materials, e.g. serpentin, bring about the collective dose increase both for operating personnel and for those doing dismantling work.

Large-scale research of structural material deposits has been and being performed, and the most promising ones from the standpoint of exposure and radioactive waste decrease are seleted [11-14, 21 - 23]. Data bank of such sites and materials as well as investigations of other materials suitable for next generation NPP construction are being made.

5. RESEARCH AND DEVELOPMENT IN 1989 - 1992

In addition to the research and development work described above other investigations [24] including fundamental research [25] were conducted in the CIS and primarily in Russia.

Thus in ref. 25 it is demonstrated that the inclusion of multistep (x,n) reactions leads to the generation of long-lived Al isotope, which after dozens of years will be responsible for fast reactor coolant activity and will not allow to do without remotely-controlled equipment during dismantling, as was believed earlier when only single-step reactions were considered. It cannot be stated a priori that in the course of fundamental research similar reactions and other problems essential to decommissioning will be discovered.

Among the applied researches hard chemical and electropolishing metal decontamination and radioactive waste treatment and disposal are worth mentioning.

It was disposal and burial of enormous quantities of radioactive waste that became the starting point to the development alternative operation waste management Concept with the main emphasis on technologies that make it possible to separate their non-radioactive components and to decrease significantly the quantity of solidified radioactive waste [26].

Alongside with improving approaches, methods, technologies and means of NPP closure, specifications aimed at solving this problem were also developed ([17], Table 3).

DOCUMENT	STATUS		
1. NPPU Closure. Terms and	Completed by final		
Definitions 2. NPPU Decommissioning.	unauthorized draft Idem		
Basic Guidelines			
3. Requirements to NPP Project Measures for Facilitation of Decommissioning	Idem		
4. NPPU Decommissioning. Framework Programme	Idem		
5. NPPU Closure. Nuclear Safety Requirements	Idem		
6. NPPU Closure. Radiological Safety Requirements	Idem		
7. NPPU Closure. Residual Radioactivity Grounds of Recycle Materials for Unrestricted Release	Completed by draft		
8. Guidelines "Common Fund for Decommissioning of NPP"	Authorized Guidelines		

TABLE 3

Regulatory and Engineering Codes and Specifications

Here Common Fund for Decommissioning of Nuclear Power Plant and Norms for Accumulating the Fund are of special interest. As in other countries [27], its foundation was given a high priority, as being necessary for solving practical decommissioning tasks including interim storages and repository construction as well as for continuing research work.

The economic crisis and high inflation rate, however, have made the functioning of rouble funds difficult. And the Common Fund for Decommissioning met this fate, too.

6. CONCLUSIONS

In spite of economic problems in Russia and other countries of the former USSR the results obtained in solving decommissioning problem over the last four years make it possible to draw the folloving conclusions:

* special attention should be given to pre-decommissioning stage without which the final stage of nuclear plant life cycle would be impossible to carry out

* special role of controlled and unrestricted nuclear installations material use should be emphasized on the basis of their correctly specified residual activity concentration limits

* as the lack of local and regional storages and repositories slows down both NPP and other nuclear installations decommissioning, their construction should be made economically attractive for both investors and local population of corresponding regions, and should be made necessary for nuclear facilities

* solutions of some particular problems should be found as quickly as possible: production of low level metal waste remelting furnaces; making the best of a retired large-scale equipment and units; on-site operation radioactive waste treatment and their subsequent storage with considerable decrease of their volume etc.

7. FUTURE WORK

1. A particular VVER standard unit decommissioning project should be carried out, probably, with the aid of international community. This work should be also useful for a number of easteuropean countries.

2. Research and development and engineering work should be continued especially that along the most promising directions

3. In this more open world international community should make all possible efforts to develop common recommendations and standards on controlled and, especially, unrestricted nuclear installations material use.

4. It seems appropriate to begin the work at consolidating the results obtained, including creting of international data banks for materials for future generation NPP and other nuclear installations.

5. It would also be reasonable to unify the international experience in decommissioning finally shutdown plutonium production reactors as well as retired nuclear powered submarine and other large-scale installations.

APPENDIX 1

Decommissioning feasibility

of Novovoronezh NPP construction stage I

Volume I

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- 4.5. Radioactive waste storages
- 5. Radiological situation in the main buildings, inside the equipment and on site
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- 9. Unit I decommissioning expenditures
- 10. Comparison of NPPU I decommissioning options
- 11. Discussioning of the feasibility results
- 12. Conclusions Appendices. Local cost estimates

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DECONTAMINATION FOR THE DECOMMISSIONING OF NUCLEAR POWER PLANTS

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Abstract

Decontamination during decommissioning of nuclear facility is used to minimize radiation exposure of personnel and public, to recover of metals and to reduction of radwaste amount to be disposed of. For obtaining of maximal benefits decontamination processes should be optimized. Results and activities related to development and optimization of chemical and electrochemical decontamination of metals from NPP A-1 are summarized in the paper.

1.Introduction

Decontamination of metallic materials and equipments during decommissioning of nuclear power plants /NPP/ may be used to minimize radiation exposure of the decommissioning staff and public, to reduce volume of radioactive waste for final disposal and thus reducing costs for disposal as well as recover of metals as non radioactive scrap. In order to to obtain these benefits, not only all particular issues of decontamination process but all related processes with their costs and benefits have to be accounted for at selection and development of appropriate methods and their application, too. Great attention is being addressed therefore to evaluation and optimization of all the issues, having influence on the overall costs and benefits of decontamination methods.

2.Objectives of the work

Main objectives of research work within the Contract with the IAEA were:

-optimization of carbon steel chemical decontamination by solvents based on formic acid and simultaneous action of ultrasound for materials from the NPP A-1. -Laboratory experiments for preliminary selection of electrolytes (except H_3PO_4) and current flows (anodic, cathodic and alternating) for electrochemical decontamination of steels for unrestricted release. Verification of the process under pilot plant condition.

- -Design, manufacturing and testing of appropriate equipment for pilot plant verification of the chemical decontamination combined with ultrasound and the electrochemical decontamination.
- -Elaboration of methodology for evaluation if it is actually worth to decontaminate the facility and metals during NPP decommissioning.

3. Results and discussion

3.1. Choice and characterization of samples for experiments Samples were taken from different parts of the secondary and auxiliary circuits of the NPP A-1 and from materials placed in reactor (so called chains of testimony specimens).

Layers of corrosion products on the samples were characterized by Moessbauer spectroscopy, X-ray diffraction and gamma spectrometric analysis.

From the results of corrosion layer analyses we can suggest that the contaminated corrosion layers have different chemical and phase composition on the surfaces of different systems. Components found in the corrosion layers were: compact, mechanically firm magnetite with different degree of non-stoichiometry and substituted Fe ions (by Cu, Zn, Mn), hematite, goethite, maghemite, hydrooxides and other Fe²⁺ and Fe³⁺ compounds. Presence of FeC₂O₄.2H₂O was proved by X-ray analysis.

Material pieces of carbon and stainless steel pipes were used in the pilot experiments. Pipes were/were not cut longitudinally and segmented by oxyacetylene torch to the pieces of 8-26 kg mass with contaminated area of $0.2 - 0.5 \text{ m}^2$. Activity was measured manually on more places (marked A-D) of the materials, on the same areas before and after experiments, by beta gamma contaminationsmonitor Berthold LB 122. 3.2. Development of chemical decontamination

3.2.1 Laboratory experiments

Composition of the solution based on formic acid [1,2] was optimized for chemical decontamination.

Experimental results of synthetic powder Fe_3O_4 dissolution rates in formic, oxalic and citric acids, EDTA and their mixtures depending on agent concentrations, pH, etc. were investigated and compared. Comparison of these results with stabilised chemical equilibriums in solutions and distribution diagrams of particular complexes enabled optimization of decontamination solution composition for decontamination objectives during decommissioning.

Optimal decontamination solution composed of: formic acidcomplexing agent (EDTA, EDTANa₂, EDTANa₄)-corrosion inhibitor FEDTA) (solution further called provided sufficient pre-dismantling decontamination efficiency for and in combination with ultrasonic even for post-dismantling decontamination of carbon steel samples from various NPP A-1 systems.

Results of FEDTA solution (2.2%) action on samples from NPP A-1 turbocompressors under static conditions are given for ilustration in Tab.1.

Sample No.	DF				
	after 24 hrs action	after 2 hrs finishing			
1 2	153 76	306 193			
3	27	102			

Tab.1 Decontamination efficiency of FEDTA solution (2.2% wt)at 20° C on turbocompressor material

The samples were gradually decontaminated in the same solution and then finished in new solution of the same composition. Ratio of decontamination solution volume to contaminated area corresponded to decontamination capacity of solution required for gradual pre-dismantling decontamination of all 6 turbocompressor loops in the same solution. Similar results were obtained for samples from the explosive mixture gas (D_2+O_2) combustion system, steam generators and testimony specimens.

Samples from tab.1 after "model pre-dismantling decontamination" were then gradually decontaminated by "post-dismantling" method in FEDTA solution (2.6%) under simultaneous action of ultrasound. Results are in Tab.2.

Tab.2 Decontamination of turbocompressor samples in FEDTA solution (2.6%) at 35^oC under simultaneous action of ultrasound. Duration 30 minutes.

Sample No.	DF	rest contam.[Bq/cm ²]
1	9	<0.37
2	15	<0.37
3	16	<0.37
4	20	<0.37

Results showed possibility and approprietness of this process for carbon steel decontamination in FEDTA solution with simultaneous action of ultrasound.

3.2.2 Pilot plant experiments

For pilot plant experiments an equipment was designed and manufactured. The equipment cosists of stainless steel tank of $1.5m^3$ volume and enables circulation of solution by means of a pump on the by-pass, heating of solution by heating elements and removal of steam by suction frame connected to a ventilation system. There are at the bottom of the tank three ultrasonic (US) emitters (dimension 400x400x100mm) with effective into working media 3x1KV. Decontamination can output be carried out by putting particular pieces of material directly into the tank or by putting material into a basket (made of iron bars) that is put into the bath.

Results of some experiments in FEDTA solution (3,6wt%) and simultaneous action of ultrasound for different times of decontamination (0.5, 1, 1.5 hours) and for various configuration of the samples toward US emitters are in Tab.3.

Pipes (samples No 1,12,13) and pieces of longitudinally cut pipes (samples No 2-11) were decontaminated so that corro-

sion layer on samples No 2-8 was faced directly to US emitters. Samples were laid horizontally 10 cm above US-emitters.

sample	A _s [Bq/cm ²]						
No	t=0	t=0,5h	t=1h	t=1,5h	notice		
1	5	1,5		0,23	pipe		
2	2	clean			corrosion la-		
3	5-50	7(5%)*	clean		US emitter		
4	3-5	1(5%)*	clean		"		
5	0,8-2,5	clean			"		
6	0,3-1,0	clean			н		
7	1	clean			11		
8	0,8	clean			11		
9	4	2	clean		corrosion la-		
11	2-6	0,5-1	clean		to US emitter		
12	5			clean	pipe		
13	1200		0,3-1,5	0,2			

Tab.3 Decontamination of carbon steel materials cut from secondary circuit of NPP A-1. FEDTA conc.=3,6wt%, $T=30^{\circ}C$, simultaneous action of ultrasound.

clean-residual activity of materials is<0,1Bq/cm²

* -share of material area with given contamination. The rest is clean

Results of the experiments proved that decontamination was efficient in various configuration of samples toward US-emitter at which intensity of US-field was on the surface of the materials in the range of 0,4-0,5 V/cm². Under these condition activity of materials 10-1200 Bq/cm² was reduced to a rest activity lower than 0,37 Bq/cm² within 0,5-1,5hour. Experiments with decontamination of carbon steels in FEDTA solution was also carried out in an operational NPP A-1 equipment of 2,4x1,67x1,36m size, in which 8 US-emitters, 1KW output each were installed. The emitters are laid out on 2 opposite (longer) walls. Results of these experiments were similar as the results under pilot plant conditions.

Total amount of material taken into experiments was 1036 kg. From this material 84% had been decontaminated to activity lower than $0,37 \text{ Bq/cm}^2$. About 30-40 kg of radioactive salts were produced during decontamination of 1000 kg of carbon steel and 1000-3000 kg of carbon steel was possible to decontaminate by 1m^3 of FEDTA, conc. 40g/1.

3.3 Development of electrochemical decontamination

3.3.1 Laboratory experiments

Various inorganic, organic and organic sulphoacids have been tested together with additives for property modifications of electrolytes at screening tests for selection of suitable electrolyte components.

This effort resulted in electrolyte based on citric and sulphuric acids with addition of $\rm NH_4NO_3$ for suppression of cathode gases formation.

Laboratory experiment results from decontamination of stainless and carbon steel samples from different NPP A-1 equipments using this electrolyte and current density $+5A/dm^2$ had proved that under these conditions is 10-25 min. electrolysis sufficient to decontaminate steels for free release.

Using of citric acid in concentrations 50-100g/l led to stability of dissolved metal cations with their summary concentration up to 18g/l in a wide pH interval (up to pH=11) without formation of sedimenting sludges.

Addition of sulphuric acid 10-20g/1 increased conductivity of electrolyte and presence of $S0_4^{-2}$ anion supported metal dissolving.

3.3.2 Pilot plant experiments

Electrolyte No 3 was verified under pilot plant conditions for decontamination of metallic materials from different NPP A-1 equipment after their predismantling decontamination. Schema of designed and for the pilot plant experiments fabricated equipment is on the Fig.1. Normal function of this equipment is evident from description on the Fig.1.

Carbon steel pipes used by experiments were cut from the secondary circuit. They were chemically decontaminated but rest activity, presented mainly on the spot of corrosion layer rests, was higher than that for free release. Stainless steel pipes were cut from CO_2 purification system that had been decontaminated before dismantling. Geometric characteristics of the materials and parameters of the experiments are shown in the tab.4.

Experiments were carried out in decontamination steps. Each step consists of electrolysis under given parameters for time t (by carbon steels is t=5min, by stainless steels is t=3min), rinsing the sample with water and short wipe with brush (mechanical cleaning). Results of experiments are for stainles steel in the tab 5.



Fig. 1. Equipment for electrochemical decontamination.

No	1[cm]	d[cm]	S[cm ²]	t(+/-)[s]	I[A]	U[V]	geom
1C	28	28	1407	+	120	9,5))12cm
2C	34	15,5	83	+	40	3	11
3C	45	15,5	1095	-	80	4	10
6C	27	28		+300/300	+65/-80	+4/-8	tt
8C	33	28	1451	-	125	8	"
11C	37	15	871	+	75	6	11
12C	28	28	1231	30/60	+105/-105	+7/-7	11
15	34	7,5	400	+	35	5	
25	42	7,5	494	+	40	6	∽ ^{7cm}
35	42	7,5	494	-	40	6	11
45	42	7,5	494	+	40	5	"
75	42	7,5	494	+	40	5	"
85	44	8	352	60/30	+30/-30	+4/-2	⊖ 3cm

Tab 4 Geometric characteristics of samples and parameters of experiments

XC-sample No of carbon steel XS-sample No of stainless steel l-sample's length d-pipe's diameter S-sample's area I-current density U-voltage t-duration of electrolysis

Sar	nple		A _s [Bq/cm ²]				
No	area	t=0'	3'/m	6'/m	9'/m	12'/m	
15	A	20,2		+ 3,6/	+ 0,9/0,2		
25	A B	12,4 10,4	+ 5,5/2,1 4,1/0,5	+ 1,4/0,3 0,4/0,1	+ 0,2/b 0,1/b		
35	A B	6,5 6,5	- 3,0/1,4 2,4/1,0	- 1,1/0,9 0,9/0,6	- 0,7/0,7 0,6/0,5	+0,7/0,3 0,5/0,1	
4 S	A B	27,6 23,7	+14,8/1,0 9,9/0,6	+ 0,5/b 0,5/b			
75	A B	18,8 17,8		+ 6,5/0,5 11,4/0,6	+ /0,2 /0,2	+ /b /b	
85	A B	10,4 12,4	+- 5,5/1,1 1,0/1,0	+- 1,0/0,5 1,0/0,5	+- 0,2/b 0,2/b		

Tab 5 Decontamination of stainless steel in the electrolyte No 3.

3'-time of electrolysis 3min m -mechanical cleaning (soft) +- current mode A_s-average activity of the area b -background

The results of electrochemical decontamination of carbon steel proved that this type of decontamination can be effectively used to clean for free release chemically unsuccessfully decontaminated materials. Only small differences in time required for complete decontamination of material were observed by different polarisation of material. Efficiency of the process may be improved by short anodic polarization before the decontamination is completed. Combination of electrochemical decontamination and mechanical effect improves the efficiency of the process.

3.4 Methodology for cost-benefit analysis for decontamination

Methodology for determining if decontamination of metals during NPP decommissioning is cost beneficial or not only based on comparison of costs and benefits be can incurred from performing of planned (required) work with and decontamination. If this comparison without is to be have to be representative all costs and benefits included for all succeding activities from starting one up to conditioning and final disposal of waste arisings during all in line performed activities. Reason for this this is that decontamination can have influence on amount and type of and secondary radwaste arisings, applicable primary dismantling and handling equipment, duration of activities, claim on personnel and so on the costs. If this could not have been done the results so obtained and decision based on them would be distorted.

Following main quantities and issues are to be taken into account by comparison of processes carried out with/without decontamination:

- exposure of personnel during dismantling, succeeded material handling, conditioning, transport and final waste disposal
- 2) waste volume for final disposal
- 3) costs for conditioning, handling of primary and secondary waste from all related activities inclusive transport and disposal of conditioned waste
- 4) cost for recovered valuable secondary raw material
 (scrap)

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- 5) intangible benefits
- 6) cost of equipment for dismantling, handling and other activities requested to perform all necessary work
- 7) personnel costs
- 8) constraints and limits (insufficient disposal volume for some kind of waste, limit imposed by regulatory body on personnel exposure by dismantling etc.)

If these quantities are to be compared they have to be given in the same units (money) what can be a certain problem for some items.

From methodological point of view the comparison of quantities should be made in the following order:

- Division of the working process into simple activities succeeding each other. All activities should be taken into account by which decontamination has influence on the costs/benefits
- 2) Estimation of costs/benefits for each included activity.
- 3) Assessment of constraints and limits.
- 4) Comparison of results for processes with/without decontamination.

It can be thougt from the abovementioned that comparison is not complicated, what is not truth. There is no simple formula that allows a user to quickly and easily determine if a decontamination will be cost beneficial for a particular process. Each element for this comparison can be estimated on past experience only for a particular case and conditions. Results for the same process can vary in different countries and among various utilities in the same country.

Conclusions

Decontamination of carbon steels in FEDTA solution of conc. about 40 g/l under simultaneous action of ultrasound with sufficient field intensity (>0,3 V/cm²) is successful for pipes decontamination to activity lower than 0,37 Bq/cm² in 0,5-2 hour by temperature 30-35 ^{O}C .

Electrochemical decontamination of carbon steel in the electrolyte based on citric and sulphuric acids with additives

can be effectively used for cleaning of chemically unsuccessfully decontaminated materials for free release.

In further work are to be improved handling of materials before and during decontamination and measurement of material's activity.

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REMOVAL, TRANSPORT AND DISPOSAL OF REACTOR PRESSURE VESSELS IN ONE PIECE FROM SWEDISH NUCLEAR POWER PLANTS — THE SKB STUDY

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Abstract

As part of the Swedish waste management program, SKB carried out the first decommissioning study in 1979 with the objective to describe and report the technique and costs for dismantling of Swedish nuclear power plants. This study was revised in 1986, It was stated in the studies that the nuclear power plants could be dismantled in a safe way with regard to health physics considerations and that all the necessary equipment was considered commercially available as they were normally used during maintenance and refurnishing in the plants. Excepted was the special equipment which has to be used for handling and cutting of reactor internals, the reactor vessel and parts of the biological concrete shielding containing neutron induced radioactivity.

The "RPV (Reactor Pressure Vessel) - One piece study" which is part of the IAEA decontamination and decommissioning programme evaluates the technical possibilities and the costs for dismantling, transportation and disposal of the Swedish reactor pressure vessels in one piece. The study concludes that it is technically possible to take care of the RPV:s as whole units. Because of the high amount of induced activity in the reactor internals these are to be handled and disposed separately. The PWR:s and the older BWR:s with external main circulation pumps are somewhat more difficult with respect to dismantling works than the late generation BWR:s with internal pumps. However the "One-piece" methodology proves to be very cost-effective compared to cutting of reactor pressure vessels in small pieces.

1. INTRODUCTION

Work on decommissioning in Sweden is at present in a planning stage. Several aspects have to be considered in this planning. They include:

- the characteristics of the reactor systems as built, such as their material composition, volumes and shapes, their radioactive properties and their accessibility for disassembly,
- the requirements set by the waste handling system on size, weight and radioactivity source strength of the dismembered parts,
- the facilities for waste transport and disposal,

- the costs and
- the timing of the decommissioning

The planning must comply with the regulatory requirements stated in law or by the Government through its supervisory authorities. Such planning was started in 1976 and has continued and evolved since then. Whereas the size and lifetime of the Swedish nuclear programme has been a highly controversial issue in the political debate, decommissioning as such has not been much debated.

Decommissioning in Sweden will involve twelve nuclear power plants located at four different sites (fig. 1). Nine are of the boiling light water reactor type designed and delivered by the ABB-Atom company and three are of pressurized light water reactor type delivered by Westinghouse (Table I).



Figure 1 Swedish nuclear power sites

Unit	Owner	Туре	Net electric rating MW	Start of routine operation
Barsebäck 1	South of Sweden	BWR	600	1975
Barsebäck 2	Power Board	BWR	600	1977
Forsmark 1 Forsmark 2 Forsmark 3	Forsmark Power Group	BWR BWR BWR	967 968 1105	1981 1981 1985
Oskarshamn 1	OKG AB	BWR	440	1972
Oskarshamn 2		BWR	595	1975
Oskarshamn 3		BWR	1166	1985
Ringhals 1	Vattenfall AB	BWR	795	1976
Ringhals 2		PWR	850	1975
Ringhals 3		PWR	915	1981
Ringhals 4		PWR	915	1983

Table I. Swedish nuclear power plants in operation

Following a referendum in 1980 on the future of nuclear power in Sweden the Swedish parliament decided that these reactors shall be finally shut down at the end of 2010 at the latest.

The Swedish nuclear waste management system in operation and in planning under the SKB responsibility is shown in figure 2.

2. OBJECTIVE OF THE "ONE-PIECE RPV STUDY"

As part of the Swedish planning for decommissioning of the twelve nuclear power plants the methodology, costs and safety was studied and reported by SKB in 1979 and 1986. The studies dealt with disintegration of the reactor pressure vessels and showed that the decommissioning of the commercial nuclear power plants can be performed in a safe way with conventional technique. Only the handling and cutting of the internal core components, the reactor vessel and parts of the biological shield would require special arrangements and equipment.

The disintegration of the RPV:s however showed to be a very manpower consuming part of the decommissioning work which also resulted in significant accumulation of radiation doses.

The idea of taking care of the RPV:s as whole units became interesting and positive experiences was gained from the decommissioning of the Shippingport reactor.

The study with the objective to evaluate the technical conditions and possibilities of dismantling, lifting, transportation and disposal of the full-size RPV:s in one piece was started in 1989. Also a cost calculation, time schedule and the safety aspects was to be studied.





The study has so far been reported in two stages. The first stage is to be considered a conceptual study and has described a course suggestion regarding methodology for lifting and transportation of the whole RPV:s. The reverse installation principle was proposed for the BWR RPV:s and the Ringhals 2 steam generator replacement principle was suggested for the PWR RPV:s.

In stage two of the study a thorough investigation was performed concentrating on the Forsmark 1 BWR unit. Main differences compared to Forsmark 1 was described in principle for the other Swedish RPV:s. A complementary study has lately been performed for Ringhals 1 (BWR) and Ringhals 3 (PWR) mainly concentrating on the heavy weight lifting.

The third stage of the study is a preliminary safety analyses for the disposal of the whole RPV:s. The third stage has not yet been decided.

3. ANALYSES AND RESULTS

3.1 General scope

Stage two of the "One piece" - RPV study, the Forsmark 1 study was finalized and reported in August 1991. The study covered the following items:

- Principles for RPV dismantling and transportation
- Requirements on radiation shielding based on radioactivity inventory
- Radiation shielding remedies
- Dismantling of the Forsmark 1 RPV (Internal parts, thermal insulation, functional requirements during dismantling, lifting and transportation technique, civil works and ground examination, release of RPV, attachment of radiation shielding, transportation, intermediate storage etc.)
- Other Swedish RPV:s
- Final disposal
- Radiation exposures
- Cost estimation
- Time schedule

3.2 Radionuclide inventory and dose rate levels

A very fundamental and interesting question has been whether the Swedish RPV:s could be handled, transported and disposed with the highly irradiated core components, the internals, left inside. As base for necessary calculations the estimated inventory of relevant radionuclides has been used. Typical figures on radioactive in-

ventory are shown in table II. The table shows the amount of induced activity after 40 years operation and 5 years decay. The amounts of radioactivity content are given in GBq:s.

	PW	/R	BW	R
Radionuclide	Internals	RPV	Internals	RPV
Fe 55	10 ⁸	$4 \cdot 10^{4}$	$5 \cdot 10^{7}$	1 · 10 ⁴
Co 60	$2 \cdot 10^7$	$5 \cdot 10^3$	$4 \cdot 10^{6}$	1 · 10 ³
Ni 63	2 · 10 ⁷	3 · 10 ³	6 · 10 ⁶	1 · 10 ³
Total	$2 \cdot 10^8$		1.	10 ⁸

Table II. Radioactivity inventory in a Swedish PWR and BWR reactor, GBq

It shall be noted that the induced activity in the RPV is concentrated to the core area which will be object to radiation shielding during transportation and disposal. The table also tells that the main amount of induced activity is to be found in the internal core components as for example the moderator tank.

A major concern for the study has been to evaluate whether the RPV can be handled with the internals still in it. The results of the dose rate calculations showed a dose rate level of 2,5 mSv/h at 2 metres distance in the core area of an empty RPV. Leaving the moderator tank inside would result in a corresponding dose rate increase by a factor of 30. The dose rates and the decay rate are shown in figure 3.

As a criteria in the study there has been stated that the handling and transport of the RPV shall be performed according to the IAEA transportation regulations which allows a maximum dose rate of 0.1 mSv/h at 2 metres. This means a radiation shield has to be attached to the RPV around the core area. Figure 4 shows the dose rate levels at 2 metres as function of steel shield thickness and the minimum steel shielding necessary to obtain dose rate levels beneath the IAEA-limit.

The required steel thickness of 80 - 90 millimetres results in a shield weight of about 90 tonnes. With the internal core components included a shield weight of 300 - 400 tonnes is required resulting in a total weight of the lift to be in the area of 1500 tonnes. With respect to safety, lifting- and transportation technique and load capacity this alternative so far has been judged unrealistic. Checking of original structural strength calculations has shown that loads up to 1000 tonnes is acceptable with proposed dismantling and lifting methodology. As a consequence the continuation of the "One-piece"-study only deals with the RPV excluding the internals.

Except for the radiation shielding around the core area also the following remedies have to be taken.



Figure 3 Forsmark 1 RPV dose rates as function of decay time without radiation shielding



Figure 4 Forsmark 1 RPV dose rates at 2 m as function of shield thickness

- The nozzles on the RPV have to be tightened with shielding plugs to prevent radiation beams from inside the RPV and to prevent airborne contamination outside the RPV.
- The operation of the ventilation systems has to be thoroughly prepared to meet the requirements according to the different stages of the preparation and dismantling work. It seems to be no problem to manage this with minor modifications in the existing systems.
- Cleaning and decontamination is very important when dismantling a nuclear power station to reduce the amount of radioactive matter to be disposed. For the RPV however we predict a minimum of decontamination except for necessary measures in connection to nozzle cutting.

The total activity inventory in an empty PWR RPV is about five times higher than for a typical BWR regarding the dominating nuclide Co 60. The specific activity in the core area which has to be used as dimensional for designing the radiation shield, is in the range of ten times the BWR figures. The shielding of the core area in a PWR RPV requires a steel thickness of about 130 millimetres.

3.3 Dismantling and lifting technique

The principles for dismantling and transportation of some typical Swedish RPV:s will be described and shown in the following.

Figure 5 shows the Forsmark 1 RPV which is the main object of the study. The Forsmark 1 RPV represents one of the newest and largest RPV:s and is equipped with internal main circulation pumps. The lifting weight is about 865 tonnes including the radiation shield.

The first stage in the dismantling procedure is the removal of the internals from the RPV. This work can be started as soon as the spent fuel and the control rods has been removed. The internals will be transferred to the fuel pools and cut into pieces before transportation to the intermediate storage for spent fuel and internals. The moderator tank rack which is welded to the RPV bottom walls will be left in the RPV. The weight of the rack is 16 tonnes. Total weight of the internals is 140 tonnes.

Internals with lower activity levels for example moisture and steam separators will be temporary stored in the fuel pools but they will be the first components to be disintegrated and transported away.

Depending on radioactivity levels the disintegrated internals will be transported either in core material transport casks to CLAB for intermediate storage or transferred to shielded waste transportation containers for final disposal in SFR (Final repository for operational and decommissioning waste).

To get access to areas for support purposes and cutting of nozzles some insulation has to be removed in an early stage. Parts of the insulation contains asbestos material which require very stringent handling from labour protection point of view. The insulation also contains radioactivity, both induced into the material as well as surface



Figure 5 Forsmark 1 - Lifting and transportation principle

contamination. One of the first steps in the insulation work will be on the primary pipe penetrations between the biological shield and the RPV nozzles. Also the thermal insulation at the control rod nozzles and instrumentation nozzles can be dismantled in an early stage.

The need for systems and functions during the dismantling period has been investigated. Some of the systems can be switched off very early and the last function to be taken away is probably the reactor hall crane and other lifting devices.

The nozzle cutting technique will be more or less conventional but in some cases it has to be remote controlled. This technique has been tested on a feed water nozzle in Forsmark during a normal outage without problems.

The lifting and transportation technique will be a reverse installation technique. It is commercially available through special contractors and the detailed performance will be defined at the time of dismantling.

Structural strength calculations have to be reviewed for the actual building constructions and some reinforcement or other temporary remedies could be necessary. We assume that buildings affected by the RPV lifting and transportation may be object to structural testing and approved by authority before the lifting. A temporary housing of the RPV might be a solution to overcome problems regarding the transport from radiation point of view, planning problems regarding time for decommissioning of the plant versus commissioning of final repository. The availability to the reactor site for other industrial activities might also be of influence.

The transportation to SFR area can be made by a multi-wheeled vehicle. The transport will be classified as "Special arrangement" according to the IAEA regulations. Of course there will be a lot of special checks and requirements as to for example ground examinations, maximum load, maximum speed, minimum curve radias etc.

The dismantling of the Ringhals 1 RPV which together with the Ringhals 3 RPV (PWR) has been studied and reported lately within the Swedish decommissioning programme, contains two additional problems compared to the Forsmark 1 RPV. An additional amount of work concerns cutting of the twelve main circulation pipes fitted to RPV nozzles and releasing of the RPV support skirt. The support skirt is welded to the spherical tank bottom, bolted and injected with concrete to the bottom of the RPV cavity where the accessibility is bad.

The proposed lifting and transportation of the Ringhals 1 RPV is shown in figure 6. The lifting principle is based on a steel beam construction with climbing jack devices for the vertical lifting and for lowering onto the transportation vehicle. The horizontal movement is performed by means of rails. The radiation shield is attached to the RPV during or at the end of the vertical lifting out of the RPV cavity.



Figure 6 Ringhals 1 - Lifting and transportation principle

The dismantling procedure for the Ringhals 3 RPV of PWR type contains one major problem which is the cutting of the six pipes or nozzles for the main circulation loops. The nozzles also have the function of support for the whole RPV weight and they are injected in concrete in special foundations. There are two possibilities regarding the dismantling; the cutting can be done either in the nozzles themselves close to the RPV wall or in the piping at about one metre distance from the RPV wall.

The proposed lifting and transportation of the Ringhals 3 RPV is shown in figure 7. The total weight is about 500 tonnes including the radiation shield. As already mentioned the lifting and transportation technique is based on the same principle that has been successfully used for the steam generator replacement in Ringhals 2. However, because of the heavy load of the RPV a special lifting device has to be arranged at the top of the containment building. The lifting device and the beam construction can be fitted to and used also in the other PWR:s.

3.4 Storage and disposal

In part 1 of the Study it was established that the final storage for reactor wastes, SFR1, and its later enlargement for decommissioning wastes might be suitable for the RPV:s without internals providing the transports down to the storage area could



Figure 7 Ringhals 3 - Lifting and transportation principle

be safely arranged. The total amount of activity in this storage will not be very much higher with the reactor vessels included. It was also established that the activity of the internals is much too high for storage in SFR1.

The transports on ground have to be performed by means of special vehicles who may have to be developed both for on-site transports, RPV handling on-board a RO/RO ship and for transport to final disposal position. All the reactor sites in Sweden are located at the coastline and have special designed harbours for spent fuel casks and reactor waste casks shipment. The presently used ship is probably not suitable for the RPV:s from dimensional point of view. Alternatives must thus be investigated.

For the underground transportation to the final disposal position in SFR a special vehicle with hydraulic transmission and suspension suitable for safe transportation and release of RPV has been suggested.

The RPV:s will be stored in a convenient accessible area in the SFR and it is proposed that the central tunnel is suitable for the purpose of final disposal. The RPV:s will be as tightly as possible packed together and placed on a concrete foundation. As a final measure walls might be built around the RPV:s and the space filled with sand or other material.

A temporary storage will be arranged at the SFR area before their final disposal as they will be the last components to be disposed in the repository.

3.5 Accumulated mandoses

Estimation of accumulated mandoses has been done for the dismantling and removal of the Forsmark 1 RPV.

Table III shows the distribution and total mandoses.

Activity	Accumulated mandoses man Sv
Removal of thermal insulation material	0.8
Release of RPV	1.8
Lifting preparations	0.1
Attachment of radiation shield, RPV lid and lifting onto transportation vehicle	0.3
Total	3.0

Table III. Estimated mandoses for Forsmark 1 RPV removal

The figures are to be considered as rough estimations to be used for planning and budget purposes and they mainly serves as indication to where extra measures must be taken from radiation protection point of view.

Because of the additional works expected for dismantling of older BWR:s as Ringhals 1, the radiation exposures also are expected to be somewhat higher than in the Forsmark 1 case. This is also expected for the Ringhals 3 RPV because of the higher amount of radioactive inventory.

3.6 Time schedule

The dismantling and removal of the Forsmark 1 RPV is estimated to be performed during a six month period with a manpower consumation of about 6000 manhours.

For BWR:s with external circulation pumps and for PWR:s the total manpower consumation is expected to be somewhat higher, but has not been object to further calculations.

3.7 Costs

The costs have been estimated for Forsmark 1 and the total cost and its distribution is shown in table IV.

Activity	Cost (MSEK)	
Insulation removal Release of RPV Internal lifting preparations Outer lifting preparations Transport opening Lifting and transportation	2.0 2.5 1.4 2.2 0.9 5.6	
Total	1.2	

Table IV. Costs for removal and transport of Forsmark 1 RPV

The cost estimations have an uncertainty of about 25 percent.

The corresponding cost for Ringhals 1 and Ringhals 3 is estimated to be about 20 MSEK. The costs for handling the RPV:s as whole units are considered to be one fourth of the disintegration method cost.

4. CONCLUSION

The Study shows that the method of dismantling, transport and disposal of the Forsmark 1 RPV as a whole unit is realistic and cost effective compared to disintegration of RPV. It has been shown likely that also the other RPV:s in Sweden can be handled in the same way.

To confirm and establish the ideas concerning the methods for dismantling and heavy lifting a more detailed study has to be done in collaboration with specialist contractors.

The dose rate calculations have shown that 5 years after shut-down it seems not appropriate to perform the necessary operations with the high level radioactive internal core components left in the RPV.

The safety assessments with respect to final disposal in SFR has to be further investigated.

The time needed to dismantle and take care of a whole RPV is estimated to be about 6 months. The radiation exposure is estimated to be about 3 manSv accumulated dose.

The cost is estimated to be in the range of 16 - 20 MSEK.

5. FUTURE WORK

The future work on the "One-piece"-study will be done in connection to and in accordance with the over all planning of the shut-down and decommissioning of nuclear power in Sweden.

As a next stage it is assumed to once more perform a review of the RPV disintegration alternative mainly with respect to cost analyses before the "One-piece"-alternative is adapted as the Swedish methodologi in the decommissioning planning.

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PWR OXIDE CHARACTERISATION/SURFACE FINISH IMPROVEMENT AND DEVELOPMENT OF DECONTAMINATION PROCESSES FOR GAS AND WATER COOLED REACTORS

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Abstract

Status of nuclear power and decontamination studies in the UK is described. Commercial operation of Sizewell Unit B is planned to begin in early 1994. The Nuclear Electric strategy for the development of nuclear power is to pursue the twin Sizewell Unit C station as the next step. The Berkeley Magnox station has been permanently shutdown. At Berkeley, there are eight boilers on each reactor. Because of the massive size and volume of Magnox boilers, decontamination studies focused on tests with cheap and simple reagents; formic acid was chosen due to the results of these studies.

Laboratory scale decontamination tests have been carried out on contaminated Magnox boiler fin specimens using formic acid alone and in combination with formaldehyde. With a 1.5% solution of formic acid alone at 23°C, decontamination was 90% complete after 48 hours, further decontamination to give an overall DF of 17 took place when the temperature was raised to 50°C. Tests have also been carried out with Windscale AGR specimens in 5% formic acid at 60°C. On sections of the superheater, DFs of >100 were achieved for the dominant ⁶⁰Co and ¹³⁴Cs/¹³⁷Cs isotopes. The reagent was, however, less effective on steels with a higher chromium content such as the Refrasil insulation package.

Experimental work by Nuclear Electric on water reactor decontamination techniques has actually ceased. The LOMI and NP/POD processes which were developed at Berkeley have been licensed for a large number of partial circuit decontaminations, particularly in the USA.

A number of techniques for improvement of surface finish are being studied, including electropolishing, mechanical polishing, pre-oxidation and application of surface coatings. Experiments have confirmed that electropolished surfaces show radioactivity uptake a factor of four to five lower than on mechanical ground or machined surfaces.

1. Status of Nuclear Power in the UK

Progress with Sizewell B. The programme is only about a month behind schedule but it is still anticipated that fuel load will meet the planned date of November 1993 with commercial operation in early 94. A number of major hold points have been successfully met, on or before schedule, eg all primary circuit pipework was installed ahead of schedule and the main simulator was commissioned in November 1992, thus meeting the requirement for it to be in operation 12 months before fuel load.

'1994' Review of Nuclear Power in the UK. The formal position regarding the future of nuclear power in the UK is that it will be subject to a review in 1994. However, this debate has to some extent been brought forward by the investigations into the future market for coal in the UK. The Nuclear Electric strategy for the development of nuclear power is to pursue the twin unit Sizewell C station as the next step.

Magnox and AGR Operating Status. Berkeley (and Hunterston A in Scotland) have been shutdown permanently. All the fuel has been removed from Berkeley to Sellafield for reprocessing. Nuclear Electric has been udertaking Long Term Safety Reviews (LTSRs) to support continued operation of its seven remaining Magnox stations, as a result Magnox lifetimes are being extended but most Magnox stations are likely to close within the next ten years. Hence, to maintain the nuclear contribution to electricity generation at 20-25%, new capacity such as Sizewell C will be required.

2. Decontamination Studies

2.1 Gas Reactors

As part of a study to decided the fate of the boilers on decommissioned Magnox stations, Nuclear Electric has looked into decontamination as a possible option (Ref. 1). At Berkeley, there are eight boilers on each reactor. these represent fairly massive mild steel structures, each weighing approximately 350 tonnes with an internal volume of several hundred cubic metres. These boilers comprise tube-type heat exchangers, which operate with the water/steam circuit inside the tubes and the CO₂ primary coolant outside. The boilers are contaminated with radioactive material transported in the CO_2 gas flow from the core, with the main isotopes of interest in terms of radiation fields being 60 Co and 137 Cs. Because of the massive size and volume of Magnox boilers, decontamination studies by Nuclear Electric focused on tests with cheap and simple reagents and resulting from this work formic acid was chosen as the optimum reagent.

At BTC laboratory scale decontamination tests have been carried out on contaminated Magnox reactor boiler fin specimens using formic acid alone and in combination with formaldehyde. With a 1.5% solution of formic acid alone at 23°C, decontamination was 90% complete after 48 hours, further decontamination to give an overall DF of 17 took place when the temperature was raised to 50°C. Tests have also been carried out with Windscale AGR specimens in 5% formic acid at 60°C. On sections of the superheater, DFs of >100 were achieved for the dominant 60 Co and 134 Cs/ 137 Cs isotopes. The reagent was, however, less effective on steels with a higher chromium content such as the Refrasil insulation package.

A number of methods for treating the effluent from boiler decontaminations have being investigated including neutralisation plus precipitation, oxidation with hydrogen peroxide, catalytic formate destruction and electrolysis. This work has indicated that catalytic destruction of formate at its present stage of development is unlikely to
be successful. Poisoning of the palladium catalyst occurs in both pure formic acid and in the presence of iron with rates being more rapid in the latter case. The most promising technique appeared to be electrolysis. This could be manipulated to produce either ferric floc or iron metal with the formic acid being oxidised to carbon dioxide.

The decontamination studies have not progressed further over the last three to four years because decontamination option for application is not considered an to decommissioned Nuclear Electric reactors. Radiation fields on the boilers are already low and our current decommissioning strategy (see Section 5) involves doing no major work on the active circuits for a considerable period of time.

There are still plans to decontaminate the boilers on the Windscale AGR using a vapour phase technique employing nitric acid as the reagent but the work has been held up by lack of funds. Current plans envisage total dismantling of the Windscale AGR to a green field site.

2.2 Water Reactors

Experimental work by Nuclear Electric on water reactor decontamination techniques has effectively ceased. The LOMI and NP/POD processes which were developed at Berkeley have been licensed to various commercial companies and have been used for a large number of partial circuit decontaminations, particularly in the USA. The AP/LOMI has been validated process for full circuit decontaminations on BWRs and is in the process of being PWRs. the validated for USA full In circuit decontamination studies are being coordinated by EPRI and we maintain a close interest in this work. It is planned to carry out a full circuit decontamination of Indian Point 2 in 1995.

Nuclear Electric continues to review developments on PWR decontamination techniques, particularly on waste treatment procedures. We have a close interest in the development work BRADTEC has done in this area. This company was formed by one of the developers of the LOMI process. They have been carrying out extensive research into electrochemical ion exchange techniques to reduce the amount of ion exchange resin utilised in cleaning up the decontamination solutions. In this promising technique which is termed ELOMIX (Ref. 2), the radioactive isotopes such as cobalt are plated out on a metal rod, together with other metal ions released during the decontamination. The decontamination chemicals are also regenerated for further This technique is now at the pilot plant stage. use.

Nuclear Electric has carried out an extensive review of PWR component decontamination practices as part of our specification of the Sizewell B decontamination facility. This facility will incorporate chemical dissolution tanks, electrochemical dissolution facilities and water jetting/grit blasting facilities. A review of the application of decontamination prior to dismantling Sizewell B has been carried out by Nuclear Electric (Ref. 3). This fulfils a PPG (PWR Project Group) requirement to review chemical decontamination options for the decommissioning of Sizewell B and to define a suitable strategy for the application of decontamination. It was reasoned that the reference method for Sizewell B should maximise the decontamination factor on stainless steel rather than Inconel. This is because work in the vicinity of stainless steel pipework and components will represent the biggest source of radiation dose to workers. The reference method chosen is the POD (PWR Oxidative Decontamination) process used in a multi-cycle mode with repetitive applications of NP (nitric acid/permanganate) and oxalic acid. The report provides estimates of the total quantity of oxide on circuit surfaces at the end of 30 EFPY (effective full power years) of operation. τt concludes that to treat the decontamination reagent required to dissolve the oxide and the released metal ions from the oxide will require 20.1 m^3 of strong cation resin and 9.1 m^3 of weak base anion resin giving a total resin volume of 29.2 m³. The report recognises that by the time Sizewell B is due for decommissioning, other reagents and more volume efficient waste treatment methods will be available. It is also probable that the overall strategy may be reviewed in the light of international experience with the decommisssioning of PWRs.

3. Surface Finish Improvement to Reduce Activity Uptake

Reduction of radiation fields by treatment of surfaces is an active research area at the present time. A number of techniques are being studied, including electropolishing, mechanical polishing, pre-oxidation and application of surface coatings. Also, deliberate addition of cations (eg zinc) in order to modify surface oxidise to reduce activity uptake is being considered.

3.1 Ringhals Experiments

Studies of the effectiveness of electropolishing and mechanical polishing in reducing activity uptake have been carried out on the Ringhals 4 reactor in Sweden. Coupons with various surface finishes were attached to the manway insert in the channel head bowl during 1987/88 and 1989/90. The results from the first experiment are summarised in Ref. 4. These experiments have confirmed that electropolished surfaces show radioactivity uptake a factor of four to five lower than on mechanically ground or machined surfaces. It is reasoned that grinding produces a highly damaged surface with potential for considerable activity uptake. However, surprisingly, strip clad stainless steel left in the as-clad condition and as-cast stainless steel both showed relatively low levels of activity uptake, even though these surfaces were extremely rough. This effect is thought to be due to the formation of a protective oxide during the manufacturing process when the surfaces are exposed to high temperatures. In the

second Ringhals experiment various electropolishing techniques and operating parameters were compared. There was little difference between the processes studied and the procedures were reasonably tolerant of the conditions.

3.2 Doel Experiments

A joint LABORELEC-EBES/Nuclear Electric/EPRI study of surface pre-conditioning and coatings to reduce activity uptake on surfaces has been undertaken on the Doel 2 reactor in Belgium with coupons being exposed in the hot and cold legs of a steam generator channel head. This study commenced at the beginning of Cycle 12 in 1986; coupons have been exposed in each subsequent Cycle up to Cycle 18 in 1992/93. Results from these experiments are reported in Refs. 5,6 and 7. The most dramatic improvement in activity uptake occurred with chromium plated coupons. These showed very low levels of activity uptake, in all cases over an order of magnitude lower for the main ⁵⁸Co and ⁶⁰Co isotopes than on as received and electropolished The addition of a passivation stage after the coupons. chromium plating treatment produced no further benefit, in fact it had a detrimental effect by increasing activity uptake by up to a factor of two. Electropolishing did not have a marked effect on activity uptake. There was only a factor of two reduction in activity uptake on the CF8M coupons compared with the as received coupon, and with the 309L coupons the electropolishing treatment slightly increased the levels of activity uptake. However, the latter slightly anomalous result may have been due to an insufficient electropolishing treatment.

4. Radioactivity Source Control

Nuclear Electric has been monitoring the development of radiation fields on KWU/Siemens PWRs in Germany. The most recent reactors of the KONVOI type have eliminated Stellite from the reactor pressure vessel and almost all other areas of the primary circuit. The only appreciable area remaining is in the CRDMs (control rod drive mechanisms). This has resulted in dramatic reductions in radiation fields and man-Sv arisings (Ref. 8). Channel head radiation fields average less than 10 mSv per hour, compared with levels of 50-100 mSv per hour on reactors which have not undergone Stellite replacement.

The radiation fields on the KONVOI reactors are now dominated by ⁵⁸Co rather than ⁶⁰Co, the latter has been reduced to very low levels as a result of the Stellite replacement programme. This has implications for decommissioning since the total inventory of ⁶⁰Co at the end of reactor life will be reduced by at least an order of magnitude.

The success of the Stellite reduction programme can be judged by comparing the figures given in Table 1. The personnel doses on the KONVOI plants are in the range 100-300 man-mSv, whereas on the Stellited plant doses are from five to ten times higher.

Plant	Year Commissioned	1986	1987	1988	1989	1990	1991
Stellite Plant							
Grafenrheinfeld	1982	2335	1594	1453	1949	2971	1462
Grohnde	1984	365	708	504	758	619	813
Trillo	1988	-	-	132	475	786	1679
Stellite Reduced Plant							
Philippsburg-2	1984	316	374	422	336	250	305
Brokdorf	1986	5	23	164	282	273	328
Isar-2*	1988	-	-	31	149	90	162
Emsland*	1988	-	-	10	146	149	128
Neckarwestheim-2*	1989	-	-	0	193	294	262

TABLE 1. PERSONNEL DOSES AT KWU PWRs (man-mSv)

All stations have annual refuelling shutdowns. 10 man-mSv = 1 man-Rem

* Konvoi plants.

5. Decommissioning Strategy

Nuclear Electric has been reappraising its decommissioning strategy for its eight Magnox and five AGR stations. This study has been directed towards reducing its decommissioning cost liabilities. As discussed in Ref. 9 the various options and combinations of options have been studied by carrying out a detailed "Best Practicable Environmental Option" analysis using standard decision As a result of this study a analysis techniques. decommissioning strategy termed "Deferred Safestore" has been proposed. This can be summarized as follows: stage 1 (defuelling) at 0 to 3 years after shutdown; prepare for surveillance period at 4 to 6 years; surveillance period at 6 to 35 years; construct safestore at 35 to 39 years; second surveillance period 40 to 135 years; stage 3 dismantling/site clearance at about 135 years and taking 10 alternative option called vears. An in-situ decommissioning has been considered at stage 3. This involves building a stable mound over the buildings and in effect burying them. Whatever final option is chosen the Deferred Safestore Strategy produces significant cost savings. Nuclear Electric is proposing that it is adopted for decommissioning its 13 gas-cooled stations and that financial provisions should be made on that basis. These provisions will be made sufficient to allow for the more expensive option of eventual complete dismantling if the in-situ option is not chosen by future generations or is not possible on all stations.

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THE STATUS OF THE EXPERIMENTAL BOILING WATER REACTOR (EBWR) DECONTAMINATION AND DECOMMISSIONING PROJECT

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Abstract

The present status of the EBWR (Experimental Boiling Water Reactor) D&D project is reported. This project consists of four major phases. Phase I started in 1986 as preparatory work for D&D and was completed by 1988. Phase II was the dismantling of EBWR components except the reactor vessel complex during 1988 to 1989. Phase III is the dismantling of the reactor vessel complex during 1989 to 1994. Finally, in Phase IV, decontamination, cleanup and project closeout will be conducted during 1994 to 1996. The projects schedule was impacted by several key factors including:

- limitation of available D&D labourers;
- unexpected delays in construction of facilities for Phase III D&D activities;
- implementation of an extensive D&D labourer training programme;
- waste shipments to DOE Hanford Facility for disposal instead of to DOE INEL Facility.

The above identified concerns have now been resolved. Physical dismantling has resumed. The reactor vessel will be segmented by the abrasive water jet cutting in consideration of cost effectiveness and safety. This work will be conducted during this year.

1. Introduction

The Argonne National Laboratory Experimental Boiling Water Reactor (EBWR) (Figure 1) was a fully operational prototype facility built to demonstrate an integrated power plant using a direct cycle boiling water reactor as a heat source.⁽¹⁾ Initial operation began in 1956 as a 20 MW(t), 5 MW(e) design. During certain experiments it was actually operated as high as 70 MW(t) through 1959. Due to the success of these high power experiments, EBWR was upgraded for operation up to 100 MW(t) and became operational again in 1962. EBWR was next used in the Plutonium Recycle Program, studying the utilization of plutonium as a fuel in light water thermal systems. The plant was shutdown, defueled and placed in dry lay-up in 1967. The D&D of EBWR began in January of 1986.



FIG 1 The EBWR facility

2. Facility Description

Containment Building

The containment building is a circular, domed structure made of steel plates welded together that originally formed a gas-tight envelope around the power plant. It rises 63 feet (19 meters) above, and extends 56 feet (17 meters) below, ground level and has an inside diameter of approximately 80 feet (24 meters). Below ground level, the steel shell is 5/8-inch thick (1.6 centimeters); above ground level it is 3/8-inch thick (0.95 centimeters). The interior of the steel shell is lined with a two-foot (0.61 meters) thickness of reinforced concrete below the main floor level. Above the main floor, to a height of 25 feet (7.6 meters), there is a onefoot (0.30 meters) thick concrete lining. At the 26 foot (7.9 meters) height, a one-foot thick (0.30 meters) concrete ceiling slab faced with a 3/8-inch (0.95 centimeters) thick steel plate completes the concrete envelope surrounding the power plant inside the containment shell.

The building contains a main floor and three lower floors on which all the power plant equipment is located.

Reactor Pressure Vessel and Internals

The reactor pressure vessel is contained within a shielded cell which extends from the main floor downward approximately 25 feet (7.6 meters) to the region of the pump floor. The vessel is made of carbon steel, clad with stainless steel on those surfaces which were in contact with reactor water or stream. It is approximately 7 feet 5 inches (2.3 meters) in outside diameter, 24 feet 8 inches (7.5 meters) in length, and has a nominal wall thickness of 2 1/2 inches (6.3 centimeters). Nine control rod drive tubes and four forced circulation inlet pipe stubs extend downward from the bottom of the vessel, penetrating the cell's bottom shielding plug. Two 12-inch (30 centimeters) diameter forced circulation outlet pipes also extend from the pressure vessel through the cell's 4-foot (1.2 meters) thick bottom shield. The vessel is closed by a forged steel coverplate approximately 9 inches (23 centimeters) thick which is retained by the reactor vessel studbolts.

The outside of the pressure vessel is covered by a layer of thermal insulation consisting of a stainless steel wool which is held in place by stainless steel bands and wire mesh. A dead air space separates the steel wool from the inner surface of the cavity steel cylinder, approximately 8 1/2 feet (2.6 meters) in diameter and made of 3/4-inch (1.9 centimeters) thick plate, which constitutes the inner boundary of the cell. Lead bricks are stacked against the outside of the cylinder to provide a gamma-radiation shield. Shield cooling coils made of copper tubing are fastened to the steel cylinder beneath the lead.

With the termination of the EBWR research program in 1967, the reactor was shut down, the fuel was removed and sent offsite for processing, and the EBWR Facility was placed in dry lay-up. This deactivated condition was achieved by draining all liquids from the reactor systems, flushing them thoroughly, and then leaving all valves in their open position. Also, certain flanges were removed and in some cases holes were drilled at system low points. The fuel storage pit was drained, flushed, and decontaminated.

3. EBWR Decommissioning Program Objectives

The EBWR D&D Project is directed toward the following objectives:

- The removal of all radioactive materials associated with the EBWR Facility from the ANL-East site.
- The decontamination and clean-up of the EBWR Facility to allow its unrestricted reuse.

3.1 Phase I, Preparations

Phase I consisted of establishing the project management group, planning the project, developing budgets and schedules and devising the data management/reporting systems. Facility services were obtained, tools and equipment assembled and a work group obtained. The first major work effort was to remove the asbestos from the EBWR facility. This was accomplished in about eight months. Significant preparation of areas for asbestos removal was required due to each level or area presenting a different piping configuration from previous work areas. In some cases, temporary flooring had to be erected or suspended under piping to allow workers access to the areas of removal. Originally, design of the piping systems provided ease of plant operation and maintenance; and, obviously, removal of the asbestos at the time of plant D&D was not considered. To remove about 500 cubic feet (14 cubic meters) of asbestos from an area, about 80 percent of the time was spent setting up equipment and the work area and only about 20 percent of the time was involved in actually physically removing the asbestos. A total of about 2,600 cubic feet (74 cubic meters) of asbestos was removed from the EBWR Facility during the Phase I D&D portion of this project. Of this total, only about 35 cubic feet (1 cubic meter) was radioactively contaminated and required disposal as radioactive waste. Phase I of the D&D was completed in FY 1988.

3.2 Phase II, Primary & Secondary System Components Removal

Phase II consisted of removal of the turbine generator system, the condenser and circulating water systems and all steam piping. Reactor systems were removed and size reduced for packaging using conventional powered port-a-band saws, air saws, plasma arc torches and oxy-acetylene torches. Personnel radiation exposures were easy to manage due to the 21 year-long decay period during safe storage of the facility.

During these activities, there was an effort made to decontaminate some reactor piping systems. Much of this equipment was wiped down using wet wipes to remove some minor amounts of loose contamination. There had been some pipe system decontamination done at the time of plant shutdown in 1967. Pipe scale samples showed minimal loose or transferable contamination. Therefore, a large scale decontamination effort was not felt to be cost effective.

Decontamination of larger equipment items, such as the condenser and turbine units, was very effective. These decontamination efforts released about 90 percent of these components to the on-site scrap metal recycler. The Spent Fuel Pool was also cleaned out, drained and repainted. A thorough general cleanup of the EBWR Building (Shell) was conducted. This Phase completed in late 1989.

3.3 Phase III-A, Reactor Vessel Internal Components Removal

Preparations for this Phase began in late 1989. To support this part of the project several new systems were designed and constructed. These consisted of a Waste Filtration and Transfer System to provide the capability of filling and maintaining level in the spent fuel pool and providing filtration capabilities for the various future cutting operations. Also installed was a HEPA system near the reactor vessel head area to filter out airborne radioactivity during the cutting operations that can not be performed under water.

Other Phase III-A preparations included the construction and installation of a mockup of the reactor internals, the installation of work tables and platforms, installation of an observation enclosure, staging of cutting equipment and special tools and conducting dry-runs of component removal using the mock-up. Unexpected schedule delays as a result of DOE Tiger Team findings have prolonged the period required to start Phase III-A D&D. The Phase III-A activities expected to begin in April, 1993 involve the removal and size reduction of the reactor vessel internals (Figure 2), including the nine control rods, core shroud, core plate holdown assembly, steam duct, poison ring, feed water rings and shock shields. Completion of Phase III-A is planned for September, 1993.



FIG. 2. EBWR cutaway view.

The first dismantling work to be performed in this phase entails the removal, size reduction and packaging of the reactor vessel internals for disposal. The majority of this segmenting and packaging work will be performed in the water-filled spent fuel pool. Tools used for the size reduction will include: underwater plasma arc cutting, abrasive cut-off wheel, guillotine saw, and other tools attached to an X-Y mast with a telescoping arm or via long-handled poles. Components will be positioned on an aluminum worktable in the fuel pool. The worktable can be adjusted to allow work to be performed on individual components at water depths ranging from 5-15 feet (1.5-4.6 meters) underwater. The internal components will be transferred to the pool, size reduced, and the material packaged for disposal in an appropriate type of waste container. Close interactions with disposal site personnel and ANL-East Waste Management staff will determine the best waste packaging container for each component to assure minimization of waste volumes.

3.4 Phase III-B, Reactor Vessel Removal

Phase III-B will consist of the removal and size reduction of the thermal shield, control rod bottom guides, bottom shield plug, control thimbles, miscellaneous nozzles and finally, the reactor vessel itself. The design of the "A" frame and hoist required to lift and support the 38 ton reactor vessel during segmenting operations is in progress.⁽²⁾

The vessel will be segmented using a Flow International abrasive water jet system.⁽³⁾ The system consists of an electric powered 75 horsepower intensifier unit, two scarifier units and one abrasive cutting lance. The system also includes a wall tracking unit, sand hopper, bag filter and a demister/HEPA filter unit. The cutting technique can cut a variety of materials and has been used extensively by a wide range of industries from food processors (without abrasives) to defense contractors (with and without the abrasive). The technique has been used to cut concrete, aluminum, carbon steel and stainless steel within the nuclear facility decommissioning community. In addition, a relatively new, surplus unit was available from the Oak Ridge National Laboratory D&D Program. Cold testing of the unit has been performed using 1" thick carbon steel and results were satisfactory. Typical abrasives used are garnet, aluminum oxide and iron fillings, with a particle size ranging from 40 to 120 mesh. An abrasive water jet--to our knowledge--has never been used to segment a reactor vessel. The unit operates at a pressure of 35,000 psi (241 MPa). The cutting rate for segmenting the EBWR reactor vessel will be about 1 inch/minute (2.5 centimeters/min) and an approximate total cutting time of 120 hours for the entire reactor vessel removal effort. An overhead "A" frame hoist will be used to lift the vessel up and out of the reactor cavity. Four circumferential cuts will be made on the main vessel body, and these rings further size reduced for shipment. The bottom head and upper flange will be shipped separately. The cutting head will be mounted to a motorized carrier to make the cuts through the reactor vessel. (Figure 3)

Conservatively, we have estimated that 600 linear feet (183 meters) of cutting will be required, and this work will consume about 7,000 pounds (3,175 kilograms) of garnet abrasive and about 3,000 gallons (11,400 liters) of water. The resulting secondary waste generated will be approximately 62 cubic feet after treatment.



FIG. 3. Abrasive water jet cutting system.

3.5 Phase IV, Removal of Reactor Cavity Liner and Shield

Phase IV will consist of the removal of the reactor cavity liner and shield items, decontamination of the cavity and fuel pool and a general decontamination and clean-up of the EBWR shell. This work will consist of removal of the lead brick shielding and removal of the carbon steel cavity liner. The majority of this material should be able to be disposed of as clean scrap. Some of this material will require disposal as radioactive waste, which is most likely for those materials near the former core region where the highest neutron fluences were present. Materials behind the upper half of the reactor vessel are very likely to exhibit no induced radioactivity and should be able to be recycled.

Activated concrete removal will be the last large scale portion of the D&D effort to be performed at EBWR. Currently, the projected removal method for the activated concrete entails use of the abrasive water jet and removal of only those areas of the bio-shield with activation above the unrestricted use levels.

3.6 Phase V, Project Closeout

Phase V will provide the final radiological characterization and Facility Certification. Preparation of the final D&D report and placement of D&D records in the site record repository will take place in late FY 1996.

3.7 Disposal of Project Waste Materials

Materials that have no detectable radiation levels or contamination levels are released to a commercial disposal firm for disposition as scrap or placement in a landfill. Since there are no <u>de minimis</u> levels authorized, any items with above background radiation levels or contamination levels is considered radioactive and is packaged and shipped to the DOE-Westinghouse Hanford site for disposal by shallow land burial. Shipments are made by dedicated tractor-trailer. The packages are usually steel drums or M-3A bins. The bins are constructed from 12 gauge steel, with angle iron reinforcement and measure 4'x5'x6' high.

A total of 54 bins were shipped for disposal through 1992. These bins accounted for 5,150 cubic feet and 7.9 E-2 curies of radioactivity.

A total of 2,915 cubic feet of nonradioactive waste was buried at the ANL landfill. A total of 2,574 cubic feet of abestos was also disposed of from the facility D&D work.

3. Future Work

The disassembly and cutting of various reactor components will be performed using a variety of cutting tools and methods. The experiences and results obtained will be the subject of future publications. This knowledge will prove useful to future D&D work at other DOE sites as well as at Argonne National Laboratory.

The development of special tools, component specific shielded transfer containers and ALARA concepts utilized will also be documented for future reference at DOE sites.

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