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DECONTAMINATION OF OPERATIONAL NUCLEAR POWER PLANTS

REPORT OF A TECHNICAL COMMITTEE MEETING
ON THE PROCEDURES FOR DECONTAMINATION
OF OPERATING NUCLEAR POWER PLANTS
AND HANDLING OF DECONTAMINATION WASTES
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
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FOREWORD

As the nuclear industry grew and its contribution to the welfare of man increased, a great deal of information became available on the various techniques in use for safe operation of nuclear power plants and managing radioactive waste.

Efficient maintenance is certainly an essential condition for safe operation of nuclear power reactors; however. maintenance work is also an important source of radiation to personnel. Radiation doses can be effectively reduced by implementation of the decontamination technology.

Decontamination processes as methods of reducing any ionising radiation from the surface of a contaminated article are widely used at the facilities of the nuclear fuel cycle for minimizing personnel exposure during subsequent operations and re-using equipment and components. Operation of the nuclear power plants must inevitably include decontamination procedures for the equipment, cooling and moderating media, surfaces of premises, etc.

In the second place, decontamination is also an important source of secondary wastes of all kinds and one must pay attention to the fact that efficient chemical decontaminants can create problems during subsequent treatment of effluents and/or conditioning of the wastes.

A Technical Committee Meeting on the Procedures for Decontamination of Operating Nuclear Power Plants and Handling of Decontamination Wastes consisting of experts from twelve Member States was convened at Mol, Belgium, from 23 to 27 April 1979 to review the status of decontamination methods for nuclear power plants in operation and to prepare a report on the subject.

The present report was drafted during the meeting and then revised by a consultant to the IAEA. It describes main mechanisms of contamination in nuclear power plants, various developed decontamination processes, planning the decontamination operation and available experience in decontamination of nuclear reactors, equipment and buildings.

It is hoped that the report will be of value not only to the organizations already engaged in these procedures but also to those who will enter the field later.

The Agency wishes to express its thanks to all participants who took part in the preparation of the report. The Agency wishes, in particular, to express its gratitude to N. Van de Voorde, of Belgo Nucleaire Centre d'Etudes de l'Energie, who served as Chairman of the meeting and who as a consultant to the Agency, contributed considerably to the editing of the material. The officers of the IAEA responsible for this work were V. Morozov, and his successor V. Tsyplenkov, of the Waste Management Section.

1. INTRODUCTION

In order to reduce the radiation fields around nuclear power plants, and, consequently, to limit the radiation exposure of and dose commitments to the operating and maintenance personnel, the contamination build-up should be kept to a minimum.

The most fruitful approach, from the point of view of economics and efficiency, is to tackle the problems of contamination and decontamination in the design and construction phases of the reactor. To do this, knowledge gained from the operation of existing power reactors should be used to make improvements in new designs. New structural materials with low corrosion rates or whose constituents are not activated by neutrons should also be used.

For older reactors, in most cases it is already too late to incorporate design changes without extensive and expensive modifications. For these plants, decontamination remains the most efficient way to reduce radiation fields.

The aim of this report is to deal with the different decontamination methods that may be applied to nuclear power plant circuits and equipment during operation. The factors that have to be considered in determining the type and the extent of the methods used are the engineering and the planning of the decontamination operation and the treatment of the resulting waste generated during the process are also discussed.

2. CONTAMINATION AND CONTROL OF CONTAMINATION IN NUCLEAR POWER PLANTS

Every type of nuclear reactor has its own mechanisms whereby contamination is built up at various locations in the circuits. An understanding of these mechanisms makes it possible to improve designs in order to slow down the build-up and make the removal of the contamination easier.

2.1 Mechanisms of Contamination

The accumulating radionuclides may originate from the fuel itself or, more often, from the products of the corrosion of the structural materials, which are circulated again and again through the neutron-generating core, and are activated and redeposited somewhere in the circuit. They then become a part of surface films which have varied structures.

2.1.1 Origin of Contaminants

2.1.1.1 Fission Products and Actinides

Such radionuclides can be produced through neutron irradiation of fissile or fertile materials. These materials may be present in minute amounts on the outer surface of the fuel elements. In normal cases this will represent a minor contribution to the total contamination of the system. The main source of fission products and actinides is the leaching of failed fuel elements. Radionuclides escaping through cladding failures normally account for 1 to 10% of the total surface contamination in water-cooled reactors. However, if the fuel failure rate has been high in the operating history of the reactor, the fission product contribution can be significantly higher. The main fission products and actinides occurring in reactor contamination deposits are listed in Table 2.1.1 together with their half-lifes.

2.1.1.2 Activation Products

Contaminants can be formed through the neutron activation of materials dissolved or suspended in the primary coolant of the reactor. The additives used for the chemical control of the coolant in LWR (B and Li) are not an important source of contamination (they give rise mainly to tritium under neutron activation). However, any impurities, which enter the coolant flow with these additives or with the make-up water, can contribute to the overall contamination. Other impurities can be released by the corrosion of core materials or out of core materials. These corrosion products become neutron activated in the core of the reactor.

Table 2.1.1

The Main Fission Products and Actinides occurring in Reactor Contamination Deposits

Fission Products	Half-life	Actinides	Half-life (years)
^{90}Sr	28.5 yr	^{234}U	247 000
^{95}Zr	65.5 d	^{236}U	23.9×10^{6}
^{99}Mo	66.2 h	^{238}U	4510×10^{6}
^{103}Ru	39.5 d	^{238}Pu	89
^{106}Ru	368 d	^{239}Pu	24 400
^{110m}Ag	255 d	^{240}Pu	6 760
^{134}Cs	2.07yr	^{241}Pu	14.6
^{137}Cs	30.2 yr	^{237}Np	2.13×10^{6}
^{141}Ce	32.5 d	^{241}Am	433
^{144}Ce	284 d	^{244}Cm	18.2

The main activation products occurring in reactor contamination deposits are listed in Table 2.1.2 together with their origin and characteristics.

In water-cooled reactors, 20-30% of the total contamination of the primary loop comes from the in-core structural materials.

Although information about other types of reactors remains scarce, it is thought, that in sodium-cooled reactors the distribution of contamination between fission and activation products would be about the same as in water-cooled reactors. In gas-cooled reactors the contamination would essentially consist of fission products and the levels would be rather low.

2.1.2 Transport and Redeposition of Contaminants

Contamination in water-cooled reactors is essentially governed by the behaviour of iron, chromium, nickel and cobalt atoms in the coolant. These elements exist in the circuit in the following forms:

- Dissolved ions, the concentration of which obeys the thermodynamical laws of equilibrium with the suspended or deposited solids as a function of the pH, the Redox potential, the temperature and pressure.
- Colloidal aggregates of various structures.
- Macroscopic aggregates suspended in the coolant flow, or loosely adhering to the walls of the system, or possibly deposited in deadlegs. These are usually called "crud".
- Films which adhere more or less firmly to the walls of the system. They usually have a well defined crystalline structure.

The coolant loop of an operating reactor is a dynamic system in which the various parameters (temperature, pressure, etc.) vary with space and time. Equilibrium state can be reached by continuous dissolution, transport and redeposition processes, with net growth of some solid phases and build-up of radioactivity. So, for example, films can be formed on core surfaces through local supersaturation due to bulk boiling (BWR) or local boiling (PWR). Dissolved ions from the corrosion of in-core or out-of-core structures can be exchanged between the solution and these films. Circulating crud can act as an intermediate in this step. Metallic atoms thus become fixed in the core for a time and are activated by the neutron flux. But, due to the intense local radiation field, colloidal particles are continuously produced from the films on the core surface. Their high surface-to-volume ratio makes them more soluble and the coolant becomes supersaturated compared to the larger crystals which form films out of the core. The activated nuclides will thus be used for the growth of these films. Other causes of radionuclide circulation can be temperature or pH gradients through the core or the heat exchanger. In any case, a number of mechanisms of this kind ensures the spread of the contamination throughout the whole coolant loop.

Table 2.1.2

The Main Activation Products occurring in Reactor Contamination Deposits

Origin	Reaction	Product	Half-life
^{59}Co	(n,γ)	^{60}Co	5.26 yr
^{58}Ni	(n, p)	^{58}Co	71.3 d
^{54}Fe	(n, p)	^{54}Mn	313 d
^{58}Fe	(n,γ)	^{59}Fe	44.6 d
^{50}Cr	(n,γ)	^{51}Cr	27.8 d
^{94}Zr	(n,γ)	^{96}Zr	65 d
^{64}Zn	(n,γ)	^{65}Zn	245 d
^{109}Ag	(n,γ)	^{110m}Ag	255 d
^{123}Sb	(n,γ)	^{124}Sb	60 d

The corrosion of ferrous metals in water practically always begins with the formation of rather soluble dihydroxides, but the further evolution of these products depends on the particular type of reactor. In PWRs and PHWRs, the coolant is kept in reducing conditions and the $Fe(OH)_2$ will be readily transformed in the sparingly soluble magnetite by the well-known Schikorr reaction:

$$7\ Fe(OH)_2 \rightleftharpoons 2\ Fe_3O_4 + Fe + 6\ H_2O + H_2.$$

Nickelferrites and chromites will form in the same way. These substances have lacunar spinel-type structures and the ions in them, especially the divalent ones, will easily exchange with those in the liquid, thus ensuring a fixation and release mechanism for Co^{2+} and Ni^{2+} in the region of high neutron flux although the oxide films on the core surfaces usually remain thin. The solubility of the whole oxide lattice is temperature-, pH and Redox-potential-dependent. (The pressure has small effect if any.)

In CANDU reactors, the pH is sufficiently high to ensure an increase in magnetite solubility with temperature, which works against the rapid growth of oxide films in the core. In PWRs, the pH is usually lower, but varies through the core due to boron consumption. The joint effect of temperature and pH gradients can cause cyclic deposition and redissolution of magnetite, thus increasing the net contamination rate.

In BWRs, the coolant is kept in oxidizing conditions and the $Fe(OH)_2$ will be oxidized in trivalent species such as Fe_2O_3, $Fe(OH)_3$ or FeOOH, all of which are very insoluble. Therefore, thick films will be formed, especially in the core where boiling occurs, but the largest part of them will adhere only loosely and remain for short times in the core. Experience has shown, that most of the contaminating activation products in BWRs does not arise from the corrosion of core structures, but basically from the corrosion of the feed water clean-up installation.

The transport of radionuclides, especially ^{58}Co and ^{60}Co, from the adherent part of the core films where they have been formed probably involves the dissolution of the oxides, transport in ionic or colloidal state and redeposition on growing epitaxial films with spinel-like structures on the stainless steel structures out of the core. Obviously, only those circuit parts where the water is liquid become contaminated.

In LMFBRs, the corrosion rate is governed by the oxygen content of the metallic coolant. The transport mechanisms would be similar to those in water-cooled reactors, but further experimental evidence is needed before reaching firm conclusions.

In gas-cooled reactors, the contamination is mainly due to fission products which are transported with graphite dust. Contamination levels are usually lower than in water-cooled reactors.

2.2 Control of Contamination

Contamination can be kept at low levels and made easily removable by adapting design criteria and operating procedures.

2.2.1 Design Criteria

During design of the reactor, special care will be taken to avoid construction materials with high corrosion rates and/or neutron absorption cross-sections. The half-lives and energy of emitted radiation of the activation products will also have to be considered.

As ^{60}Co usually gives rise to the most difficult contamination problems special attention will be paid to avoid the presence of cobalt in the primary coolant loop. Cobalt-bearing alloys such as stellites and other such surface-hardened metals have been prescribed and high-purity nickel will be chosen for the fabrication of the necessary stainless steels and high alloys.

In BWRs most of the contamination arises from the corrosion of the turbine and condensate systems, the structural materials for these parts will be chosen with great care. The use of brass in the condenser tubes is questionable since it produces ^{65}Zn and ^{110m}Ag.

In some reactors the use of antimony-containing antifriction alloys in pump bearings has lead to contamination problems with ^{124}Sb. Another parameter of importance in the design of nuclear facilities is surface conditioning. Porous materials should not be in contact with the primary coolant and the surfaces will be as smooth as possible, thus ensuring low corrosion rates and low probability for the crud adhering to the walls.

Some materials, especially stainless steels, grow protective oxide layers which adhere very strongly and are difficult to remove. If these films are formed during reactor operation, they will incorporate activation products (see 2.1.1.2) and strong chemicals will be needed to remove such contamination.

Junctions between dissimilar metals and alloys should be avoided as they increase the corrosion rate.

Finally, care should be given to ensure a good geometrical lay-out for the piping systems, heat exchangers and other process vessels.

Zones with reduced flow, in which deposits are easily trapped and difficult to remove, should be avoided, e.g., long horizontal pipes, abrupt changes in cross-section or direction, obstacles across flow, expansion devices, deadlegs, dilatation bends, etc. Provision has to be made for easy flushing and draining of unavoidable dead zones.

2.2.2 Operation procedures

The chemical quality of the coolant has to be controlled so as to minimize the corrosion rate and radionuclide transport. This can be done in several ways:

- Continuous bleeding of part of the coolant, treatment of the bleed as radioactive liquid waste and replacement with fresh, pure coolant.
- Bypassing of a part of the coolant flow through a purification installation. The treatment can be either physical (removal of crud by filtration), chemical (use of ion exchangers) or both.

Various combinations of filtration and ion exchange systems are used in practically all modern water-cooled power reactors.

In direct-cycle BWRs, both the reactor coolant and the turbine condensate have to be cleaned. The condensate is usually 100% treated; however, if the hot condensate is repumped directly from an intermediate pressure line, it cannot be treated by ion-exchange resins. Processes such as high temperature magnetic filtration are being developed at present but special care must still be given to the choice of turbine materials for BWRs operating with such cycles.

The coolant treatment rate has to be proportional to the steam flow rate (normally 1% to 2%) since most impurities come from the turbine and the condenser.

In PWRs, contamination theoretically occurs in the primary loop only. The chemical and volume control system (with the possible boron recycle control system) will be designed in relation to the total volume of the primary loop.

The coolant control systems have to keep the pH, Redox potential and impurity contents of the coolant at the specified levels. In PWRs the pH has to be adjusted to off-set variations in the boric acid content from the shim control. Radiolytic oxygen has to be removed. In BWRs it is usually sufficient to keep the impurity level low. Sometimes oxygen or hydrogen peroxide is added to get passivation of surfaces in the condensate system and thereby minimize corrosion.

Fuel failures have to be detected by monitoring noble gases, iodines, and ^{239}Np, and failed elements have to be replaced. Some reactor systems change fuel while at full power. In other types this can only be completed during a planned shutdown.

3. DECONTAMINATION PROCESSES

3.1 General

Successful radioactive decontamination of surfaces will remove all films and contamination, without excessive corrosion of the substrate. It is a clean-up process in which too strong an attack involving redeposition of loosed substances, must be avoided. For an absolute decontamination the upper metal layers will have to be carried away because contamination may have diffused through the oxide layers and may have reached the base material.

The film with the radionuclides can be attacked in several ways: either mechanically (flooding, brushing, abrasive cleaning) or chemically (using solvents, inhibited acids, etc.). A suitable process should be chosen according to the nature of the substrate and the contamination mechanism (see Chapters 2 and 4).

The efficiency of a decontamination operation can be assessed in several ways. One usually uses the decontamination factor (DF), defined as follows:

$$DF = \frac{\text{radiation intensity before decontamination}}{\text{radiation intensity after decontamination}}$$

The value of the DF depends upon the specific place chosen for the measurement to be performed and the measurement technique used. The value is not necessarily the same for all radionuclides. Therefore one can observe different DFs for different radionuclides. A DF reported in the literature is of value therefore only if all experimental procedures are described. It is the usual practice to give several values including extreme values and an average value (see 4.5 and 6.3 too).

3.2 Chemical Methods

The removal of films from metal surfaces depends on many variables among which are the temperature during contamination build-up, the age of the oxide layers and the nature of the surface. Films formed on carbon steel can usually therefore be dissolved in acids by single-step procedures; but layers grown on stainless-steel in PWRs are removed more easily by multiple-steps procedures.

3.2.1 Chemical Baths

This technique is mainly used on smaller items in centralized decontamination plants. The part to be decontaminated is immersed in a bath containing chemical reagents like citric acid, phosphoric acid, nitric acid, oxidizing or reducing chemicals, paint strippers or complex detergents.

Typical decontamination solutions include commercial cleaners, caustic permanganate, oxalic acid or phosphate base products, laundry detergents, and chloride base cleaners.

Such baths are often operated at elevated temperatures and the material to be decontaminated is moved in the liquid to improve the washing action. It must be remembered that once an immersion bath has been used, its contents are contaminated, so, care must be taken not to contaminate grossly the solution early in its life, otherwise it will not be suitable later for slightly contaminated articles. Furthermore, considerations should be given to the fact that this solution will eventually have to be treated as liquid radioactive waste, and that existing liquid waste treatment facilities should be capable of conditioning these wastes without problems. Finally, it should be noted that equipment which is not discarded as waste may need careful decontamination.

3.2.2 Equipment Rinsing

This method can also be used for contaminated chemical process loops or components of circuits carrying radioactive solutions. Loops and components are filled with the agent which is circulated until the required decontamination level is reached. Replacement and/or make of solutions is needed to avoid recontamination, thus close analytical follow-up is imperative.

On equipment with large vertical surfaces, the chemicals can be applied as a gel that, after a certain time, is removed and the surface cleaned by rinsing or spraying.

3.2.3 Chemical Reagents

Inhibited acids

The most effective decontaminants on carbon steel include 8-10% solutions of sodium hydrogen sulphate, orthophosphoric acid, sulfamic acid (NH_2SO_3H), citric acid or diammonia hydrogen citrate, and oxalic acid. Typically these solutions are used at 90°C with a contact time of 1 h. Caution must be exercised with H_3PO_4, citric acid and oxalic acid which form insoluble compounds with iron, to prevent redeposition.

Since the corrosion of metals in acid solutions occurs an inhibitor has to be added in an adequate concentration. The inhibitor has to block the dissolution of the bare metal but must not affect the reaction rate of the oxides. It has to be soluble and chemically stable at the operating temperature and the possible decomposition of reaction products must be harmless.

Of course, corrosion catalysts such as Cl^- and especially F^- are excluded. The most useful inhibitors are complex organic compounds which, when placed in an acidic medium, form ammonium, oxonium or sulfonium ions. A particularly efficient inhibitor appears to be phenylthiourea.

Electrochemical protection, either cathodic or anodic, is of little use in such complex systems as reactor loops.

Chelatants

Ethylenediaminetetracetic acid (EDTA) forms strong complexes with most metallic ions (it is a hexadentate ligand through the N atoms and the carboxyl groups). Thus, it can help to dissolve iron and other such metals from their oxides and to keep them in solution even at alkaline pH or in presence of anions which form insoluble salts with them.

EDTA and its complexes are stable up to 300°C. The acid and its sodium salts are used in many decontaminating mixtures. The user has however to bear in mind, that EDTA will tend to impair the insolubilization of the produced radwaste.

Multiple-step processes

Since single-step decontaminants are ineffective for removing adherent films from stainless steel, more complex procedures, including a preconditioning step, are required.

This preconditioning step is done by using an alkaline solution of potassium permanganate (AP) which oxidizes the chromium in the protective oxide layer, making the film more porous and more easily attackable by the subsequent acid step.

Care has to be taken with respect to the following.

- That the AP solution does not corrode either the carbon or stainless steel, but is aggressive to Stellite and other cobalt alloys. Soluble cobalt ions can so be made available for activation in subsequent operation, generating ^{60}Co. Aluminium is attacked by alkaline solutions, too.
- Rinsing and neutralization between the alkaline and the acid step must be done very carefully since alkaline solutions entrapped in crevices can lead to the caustic embrittlement of the steel.

The most commonly used two-step processes for the chemical decontamination of reactors are given in Table 3.2.

Soft processes

The so-called "soft" processes include:

- The use of a modified Citrox process with low concentrations and continuous recycling through filters and ion exchangers (CAN-DECON process, see 5.3. for thorough description)

Table 3.2

Two-step Processes for the Chemical Decontamination of Reactors

Name of process	First step	Second step
APAC	Alkaline Permanganate	Ammonium Citrate
APACE	Alkaline Permanganate	Ammonium Citrate + EDTA
APOX	Alkaline Permanganate	Oxalic Acid
APCitrox	Alkaline Permanganate	Oxalic Acid + Citric Acid
APSul	Alkaline Permanganate	Sulfamic Acid
APBis	Alkaline Permanganate	Sodium Hydrogen Sulfate

- The addition of H_2O_2 just before shutdown of a PWR, or cycling the operating conditions. These methods will be thoroughly described in 5.2 where the possible advantages of these techniques versus the so-called "hard" conventional processes will be discussed.

Detergents and other additives

Greasy or oily contaminated deposits can be removed either by organic solvents (chlorinated hydrocarbons are to be avoided since remaining traces can release chloride ions in subsequent aqueous steps, thus enhancing corrosion rates) or by detergent-containing water, possibly with complexants or other chemicals.

Loose crud can be removed by water-containing surfactants. Redeposition of insoluble particulates can be prevented by increasing the viscosity of the solution by using methylcellulose or similar products.

3.3 Mechanical Methods

Mechanical methods can be sub-divided into non-destructive and destructive cleaning. Destructive cleaning of surfaces is generally very efficient in removing contaminants together with surface layers of the base material and is mainly applied in cases where disposal of waste is the objective. The question as to whether a technique is non-destructive or not is not only dependent on the technique used but widely depends on the item being cleaned and its future destination.

3.3.1 Manual Cleaning

This method using brushes and swabs with water and/or detergents or cleaning solvents is based upon the cleaning techniques used in the household. It applies mainly to complicated items of small to medium size.

3.3.2 Abrasive Blasting

Abrasive blasting is the treatment of a high velocity stream of abrasive material to a surface to remove the adhering films. The abrasive can be propelled either by:

- compressed air
- centrifugal force
- high pressure water or steam.

Abrasive blasting is suitable for the cleaning of large surfaces such as walls and floors, or the outside of tanks and other process equipment. It cannot be used where strong geometrical tolerances or surface smoothness are to be kept. It is very efficient for porous materials which can be decontaminated only by the mechanical removal of the surface. Ventilation problems can occur if contaminated particles are made airborne.

Abrasives used for decontamination are sand, metallic pellets, glass beads and hard synthetic powders such as carborundum, alumina, boron oxide or carbide, etc.

In dry blasting sand and compressed air are normally used. Mesh size has to be chosen in such a way as to achieve a compromise between cleaning speed and damage to the treated material, both of which increase with particle size. Airborne dust has to be carefully controlled; this is best achieved by using the so-called "vacuum blasting" technique, in which the nozzle is surrounded by a concentric cone connected to a vacuum pump.

Metallic abrasives, and also metal oxides, are less friable than sand and produce less dust. They are usually recycled. Equipment using centrifugal force can be used with such powders.

Wet blasting results in less damage to the equipment than dry blasting, but produces radwaste water and abrasive particles which adhere to the walls due to their wetness and have to be removed by brushing.

Much of the work with abrasives is done by direct contact and therefore is limited at low radiation levels. However, there is little experience with remote abrasive cleaning.

3.3.3 Machining, Grinding, Chipping

These methods are definitely destructive methods as material is removed from the surface (i.e. a few μm up to a few millimetres). The methods are typical for big components. The decontamination can be done either in a radioactive workshop or anywhere else in the installation. Even large equipment can be cleaned by this technique as demonstrated by the following examples:

- Removing superficial layers of the contaminated surface of a reactor vessel stud by machining;
- Grinding of the contaminated inside surface of a heavy piece of equipment (i.e. pump casing or piping).

It is obvious that these techniques can be directly applied only for equipment with contamination of low dose level. For higher dose level, long range application or complete remote operation has to be adopted.

Removal of surface contamination from walls, floors, ceilings, etc. may be an efficient means of reducing the volume of solid wastes. In this case, chipping, pneumatic hammering and scrubbing to a depth sufficient enough to remove the contamination can be applied. A powerful vacuum cleaning system is advised to collect the removed surface layers.

Special precautions must be taken where grinding is used not to contaminate other equipment and to collect all fines produced.

3.3.4 Jet Cleaning

Jet cleaning uses the decontaminating-solutions already studied in 3.2.1.3 ranging from soapy water to acidic solutions, but it adds vigorous mechanical action to the chemical attack. Often water alone can be used. Nozzles can be operated remotely and rotated in all directions, making it possible to clean large surfaces without requiring the operator with brushes and mops to come in close contact with the object. Of course, waste water is produced and has to be contained, but the volumes are small and, in the case of the interior of large vessels, certainly less than with the pure chemical methods for which the tank would have to be completely filled.

High pressure is necessary for efficient jet cleaning with low solution volumes. This can be provided through steam jet pumps (ejectors or injectors) or positive displacement pumps. Pressures up to 1000 kg/cm^2 are used.

Most of our present experience is with the use of steam ejectors. High pressure water jetting has proved to be a very efficient decontamination technique, both as far as removal of radioactive contamination and time needed are concerned.

3.3.5 Vacuum Techniques

Vacuum cleaning techniques are widely used in removing loose contaminations. Small model vacuum cleaners can be used, inside glove-boxes in order to remove dust and solid waste produced as a result of decontamination carried out before any dismantling of the box begins.

During grinding and pneumatic hammering of concrete surfaces or in similar type operations where particulate matter is being removed from the surface, a "vacuum heat" can be fitted to the tools or close to the area of operation to remove any contaminated fines as they are produced, thus preventing the spread of contamination.

Very powerful, commercial size household and industrial vacuum cleaner models can be used. A disposal drum can be positioned in the vacuum system between the actual machine and the end of the vacuum hosepipe so that any radioactive matter is deposited in the drum and does not enter the vacuum machine. Any exhaust gases can be discharged from the drum through a HEPA filter. The drum containing the removed fines can then be used as disposal container after appropriate conditioning of its content.

3.4 Advanced Methods

3.4.1 Ultrasonic Cleaning

Ultrasonic cleaning is based upon cavitation, i.e. the formation of vapor bubbles, in the antinodal lines of a pressure wave pattern travelling across a decontaminating bath, sometimes containing chemical additives, in which the objects to decontaminate are suspended. Cavitation provides very vigorous mechanical agitation when the antinodal lines cross the surface to be cleaned, since viscosity forces are locally destroyed by the rising bubbles.

The vibrations can be induced by piezoelectric crystals (such as $BaTiO_3$) or by magnetostrictive resonators. Frequencies in the range 18-50 kHz, usually in the neighbourhood of 20 kHz, are chosen. Other important parameters are vibration intensity, external pressure, vapor pressure, surface tension and viscosity. The last three are temperature dependent. For a given frequency, cavitation occurs only above a minimum vibration intensity which is related to all the other physical parameters.

Ultrasonic equipment has to be carefully designed and correctly used. The chemical decontamination process chosen has to be suitable for the job in question. The temperature has to be high enough to obtain a good chemical effect, but low enough to keep the transducers in working condition.

3.4.2 Electropolishing

Electropolishing is an electrochemical cleaning process in which the metallic object being decontaminated, is made anodic in a cell containing concentrated orthophosphoric acid as the electrolyte.

The anodic oxidation occurs more slowly in crevices and more quickly at peaks, resulting in a polishing effect which does not severely damage the surface. This process has been used at Hanford to decontaminate pieces of equipment and make it possible to recycle or dispose of their waste with less than 10 nCi/g of alpha-emitting radionuclides.

A limitation on the process is that the pieces must be small enough to be soaked in the cell.

3.4.3 Laser

This process, still in the development stage, aims at removing thermally the upper layers of contaminated metal or concrete surfaces. The energy would come from a coherent infrared beam. The vaporized material would be caught by an appropriate ventilation.

Laser decontamination would allow easy remote operation and is essentially developed for the treatment of surfaces in a high radiation environment, such as the reactor containment building.

However, a lot of work, including an economic evaluation, remains to be done in this field.

3.4.4 Other Techniques

Further techniques under development or proposed for investigation are:

Flame spalling - which is used in steel factories to remove oxide layers from steel plates and large bore tubes.

Plasma-cutting - is already often used for the volume reduction of equipment, even under water. It can also be applied to remove sticky surface layers by heat shocks.

Spark machining - a technique used for the manufacturing of precision tools by attacking the surface of an object with high voltage sparks.

All R & D activities obviously aim at the efficient elimination of surface contamination in sticky layers. Application of the procedures under development will largely depend on the complexity of the procedure itself, the actual problem for which it is foreseen, and the secondary wastes and the dose commitments it entails.

4. Planning the Decontamination Operation

The need for reactor decontamination should be understood at the very beginning of the design phase. Therefore, as the design of the various parts of the facility progresses according to the principles of 2.2.1 and as the ordinary operating procedures are being developed, one or more of the decontamination processes described in the previous chapter will be chosen after careful testing, and the material and personnel requirements will be evaluated and the decontamination procedure will be established.

4.1 General Considerations

Although decontamination considerations should be included in the design of each nuclear facility and all provisions should be made for trouble-free cleaning, the operation itself will be decided upon only after a careful comparative economic study of all alternatives available, including the possibility of having no decontamination at all.

The optimum decontamination process required for a specific purpose can only be chosen if the formation mechanism of the oxide layers on the structural material is known. The nature of these films and the way they incorporate radionuclides are also important, for example, it is easier to dissolve chemically pure magnetite formed on carbon steel rather than nickel- or cobalt containing analogous materials formed on stainless steel (see 2.1).

The next sections will outline the approach to be taken in order to plan the decontamination and the related operations successfully.

4.2 Selection of the Process, Testing and Evaluation

The successful development of satisfactory procedures and reagents for decontaminating large-scale equipment requires well-planned laboratory and pilot plant studies.

A few of the available processes will be selected for further examination after a literature survey is made of the decontamination efficiencies and corrosion rates of the structural materials of all the components of the primary loop, including such detailed items as pump seals, bearings, valve seats, etc. The rate of corrosion chosen for the reagent will include enhanced sensitivity of thermal treatment near welds or through galvanic coupling.

Experience in the decontamination of nuclear reactors still remains rather scarce, therefore, many figures will not be found in the literature, or their value will often be questionable. Materials will, therefore, have to be tested in the laboratory, by placing samples in beakers filled with the cleaning mixtures at various temperatures and concentrations, or by electrochemical measurements. Special attention should be given to passivation and reactivation phenomena through build-up and redissolution of protective layers.

However, the final choice should not be based solely on the literature and on laboratory results whose experimental conditions are usually different from those which occur in the real loop. Therefore, further testing has to be performed in engineered facilities which simulate as much as possible the circuit to be decontaminated. Whenever possible, components or critical parts of them will be tested under the conditions of the decontamination process.

Important variables to keep in mind in pilot-scale examinations as well as in the laboratory are:

- Surface-volume ratio (ratio of the surface area of the facility to be decontaminated to the volume of decontaminant).
- Surface preparation of the materials (preconditioning before contamination).
- Contact times of the different solutions.
- Rinsing times.
- Temperatures.
- Turbulence level (stirring and agitation in laboratory tests; flowrate and parallel or transverse arrangement of the samples in pilot-scale loops).
- Dissolved oxygen.

The reported results will be useful only if all operating parameters are carefully noted. Internationally agreed standard procedures should be sought which should allow interlaboratory comparisons and avoid costly duplications.

The final choice will always be a compromise between decontamination efficiency and corrosion rate. Eventually, an economic study (4.5) will make it possible to repair or replace heavily corroded components which cannot survive the decontamination.

4.3 Equipment and Personnel Requirements

Decontamination planning includes the evaluation of requirements for the equipment and personnel for the operation. The necessary equipment should be listed and at least the most frequently used equipment should be purchased and stored in a special decontamination centre.

Decontamination equipment includes:

- Chemical measurement and control devices, ranging from colorimetric pH-paper to sophisticated remotely-operated electronic instruments monitoring the pH, concentrations, Redox potential, electrical conductivity, etc.
- Radiological measurement and control devices.
- Clothing and protection equipment for personnel such as overalls, face masks, respirators, overshoes, and so on. Frogmen suits may be provided for work in a heavily contaminated atmosphere. Changing rooms should be available for the decontamination workers.
- Individual dosimeters (film, stylo, ...) for radiological monitoring of the personnel.
- Trucks, tanks, lifts, etc. for the transport of chemicals and pieces of equipment.
- Storage tanks and bins for chemicals. They will be designed according to the local conditions.
- Pipes, hoses, fittings, stirred vessels, pumps, weighing apparatus, etc. needed for the preparation of the various cleaning solutions and their injection in the system.
- Equipment for mechanical cleaning and other processes described in 3.2.2 and 3.2.3., if planned.
- Waste treatment plant, including storage tanks for spent solutions; installations for the evaporation or chemical precipitation and insolubilization of the concentrates and transport means to a provisional storage facility or a final disposal site.
- Mechanical tools allowing for repair and/or replacement of items failed during the decontamination. Spare parts will be available for those items which are likely to fail.

The whole operation will be supervised by a decontamination specialist with a strong technical and scientific background who will be aware of the most recent developments in the field. Authority and responsibility levels and distribution of work will be described in detail, and everybody will know what is expected from him.

Special emphasis will be put on the training of the personnel. This can be done by various audiovisual methods or, better by work on mock-ups. The workers would be taught not only the work itself but also the basics of radiological protection (exposure control and use of protective equipment). Insufficient training of personnel will result in greater activity releases to the environment and greater losses of valuable equipment.

4.4 Planning and Monitoring of the Operation

The decontamination planning will provide detailed operating and check-list procedures. These will include, for each step of the process chosen:

- Introduction of the reagent
- Heating of the solution to the decontamination temperature
- Valving and flow sequence
- Removal of spent solution with radionuclides
- Rinsing and flushing, with eventually backflushing of deadlegs

The procedures will indicate who has to do what job, how it has to be done, who controls it and how, and what has to be done in the case of an unexpected development in the sitution.

The whole operation will be preceded by measurements of radiation level and, if possible, of the oxide layer composition and thickness at well-defined sampling and control points.

During the operation, samples of the solution will be regularly taken at the sampling points and analyzed for reagent strength, crud concentration and composition, dissolved corrosion products, dissolved and suspended radionuclides. The on-line corrosion rate meters will be monitored to ensure corrosion remains within the acceptable range.

The planning will provide for different procedures according to the results of samplings and analyses. Unexpected events will have to be mastered as fast as possible.

After the operation, the radiation fields around the circuit should be measured again. The decontamination factor will be obtained from these measurements and those obtained prior to decontamination. Corrosion samples and, if possible, pipepieces, should be removed and examined for corrosion rate measurement and characterization of the oxides.

A report of the work will include the type of radioactivity (alpha or beta-gamma) personnel dose, instruments used for the measures, time and place of the measurement, state of the examined material and background for other sources. If any of these data is lacking, the information given will be difficult to compare with other work.

4.5 Economics of the Decontamination Operation

The decision to undertake a decontamination operation has to be taken after a careful cost-benefit analysis taking into account all financial, legal, radiological and environmental aspects of the problem. The extent to which the radionuclides have to be removed (total decontamination usually means disposal of the concerned object) will be determined on the same basis. Economics govern the choice of the decontamination process, too, as explained in 4.2.

The various costs and benefits involved in a decontamination operation are summarized in Table 4.5.

Table 4.5

Economics of Decontamination

	Costs	Benefits
Material and equipment	- Process equipment - Decontamination chemicals - Accelerated corrosion of plant equipment - Treatment, conditioning, transport, storage and disposal of generated wastes - Protection equipment for decontamination workers	- Direct maintenance may be substituted by remote maintenance - Protection equipment for plant workers may be made less heavy
Manpower	Decontamination workers specially trained	Plant staff may be reduced
Exposure of monitored people	- Exposure of decontamination workers to high radiation fields and contamination levels - Dose commitment and other risks associated with waste treatment, conditioning, transport, storage and disposal of the wastes	- Decrease in exposure of plant workers to radiation and contamination - Active inventory and radiation field around the installation reduced.

Obviously, the balance between these costs and benefits will be influenced by the regulations concerning maximum permissible irradiation levels and radionuclide intakes for the monitored workers and the general public, by the local wages and, last but not least, by the monetary cost assigned to the unit individual or collective dose. As international agreement about that factor is still lacking, it will be estimated before planning in the context of the local socio-political situation.

5. Decontamination of Reactors

5.1 Pressurized Water Reactors

The high-pressure - high-temperature operation of the primary loop of pressurized-water reactors, with slightly alkaline and reducing coolant, cause the formation of thin protective films which strongly adhere to the metallic walls. These films are crystalline, with a spinel-like structure, and consist essentially of magnetite on carbon steel and of nickelferrite on stainless-steel and high nickel alloys. Chromium, although present in minute amounts, strongly reduces the solubility of these compounds (see 2.1.1). These films incorporate or adsorb radioactive activation and fission products, which then diffuse through the oxide layer and in some cases can reach the first metal layer.

Mechanical cleaning methods, such as described in 3.3, are effective for removing loose or weakly adhering crud, but cannot strip away the tightly bound protective layers.

Single-step chemical cleaning using inhibited acids can remove magnetite on carbon steel but is unable to dissolve the chromium-containing passivated layers on other alloys.

So all practical methods for decontaminating stainless steel portions of PWR primary loops will include a pretreatment step with alkaline permanganate (AP) which oxidizes chromium to the Cr^{6+} level, making the film readily soluble in acids.

Use of oxalic acid in the dissolution step is questionable because heavy redeposition by precipitation of ferrous oxalate occurs after a few hours of contact. Ammonium citrate and phosphoric acid present the same drawback but to a lesser extent. It can be minimized through use of iron sequestrants such as EDTA.

Present experience in decontamination of PWRs (including pressure tube, light-water-cooled, graphite-moderated reactors such as Hanford N) is summarized in the following table and comments. (see Table 5.1.)

In addition to the so-called "hard" techniques discussed so far, there are "soft" techniques involving one of the following:

- Addition of H_2O_2 to the primary coolant just before the shutdown for refuelling. This increases cobalt solubility. The cobalt-charged coolant is treated by the normal chemical control system.
- Temperature, pressure or Redox potential cycling of the coolant, so as to loosen wall deposits. The suspended crud is removed by the filters in the normal chemical control system.
- Addition of chemicals in low concentration to the continuously recirculated coolant. This technique will be described more thoroughly when studying HWR decontamination. The operation at the port of shipment (see Table 5.1) was a step in that direction.

The "soft" techniques would avoid drawbacks such as:

- Production of large volumes of medium-level liquid wastes
- Handling of corrosive liquids

Table 5.1

Examples of PWR Decontamination

Reactor	Date of decontamination	Nature of contaminants	Parts decontaminated	Construction material	Process used	Duration	DF Achieved		
							Lowest	Highest	Mean
Shippingport (USA)	1964	Activation products	Primary loop	Stainless steel	Modified* APAC	15 d	∠ 1*	49	
Hanford N (USA)	1965	Activation products	Steam generators	Stainless steel	AP-diluted Sulfamic acid Concent. Sulfamic acid		1.6	10	
Hanford N (USA)		Activation products	Steam generators	Stainless steel	APAC				
			Rest of Primary loop	Carbon steel	Phosphoric acid				
SENA CHOOZ (France)		Activation products	Primary loop		APAC				
BR.3 MOL (Belgium)	1975	Activation products	Primary loop	Carbon steel	APAC	2 mo.	2.6	50	18.8
			Steam generator	Stainless steel			5.6	70	36
VVER-2 (GDR)	after 1969 (~ 10 times)	Activation products	Primary circuit with core	Carbon steel Stainless steel	AP Citrox APACE	35 h	2	100	51
			Cooling loop	Nichrome steel	APACE	24 h			
VVER-440 (GDR)		Activation products	Primary circuit (2-3 loops)	Carbon steel	APACE	60 h	2	100	51
			Steam generator	Stainless steel	APOX	35 h			
			Pressurizer		APACE	30 h			
Novovoronezhskaja (USSR)	1978	Activation products	Steam generator	Stainless steel Carbon steel	Alkaline Permanganate Oxalic and nitric acid	25 h			

* The APAC-process was run with rather diluted reagents (1 to 2%) at 120°C with 24 h contact time. The spent solutions were passed through mixed-bed ion exchangers and so deionized; then they were used as rinse liquors.
The personnel was carefully trained by a trial run;
The waste produced consisted of 50 m^3 active resin;
Active crud was trapped in deadlegs and other low-flow zones. Such hot spots were further shielded, cut away or backflushed when it was possible.

- Necessity of defuelling before decontamination
- Necessity of isolating corrosion components sensitive to corrosion by protective coatings, flushing pump seals and glands with deionized water or treating only part of the circuit.

5.2 Boiling Water Reactors

In boiling water reactors, contamination usually remains in the pressure vessel and the coolant purification loops. The vapor lines and the condensate system usually remain rather free from activity, but corrosion in these parts can contribute to the contamination in other parts of the loop (see 2.1.2). However, the turbine can become slightly contaminated with barium isotopes due to the entrainment of short-lived xenon by the steam.

The oxide films present in BWRs differ from those in PWRs (Fe_2O_3 may be present at some places) and a given process can be quite effective in a PWR but yield disappointingly low DFs in a BWR. However, AP Citrox would work for both reactor types.

Most decontamination experience available with BWRs relates to the treatment of components removed from the loop (see Chapter 6), sometimes after some mechanical decontamination.

The BWR at Gundremmingen/Donau, Federal Republic of Germany, has been decontaminated using the APAC-process.

The Dresden-1 BWR (USA) was being decontaminated at the end of 1979 following the NS-1 process developed for this purpose. The decontaminant is a proprietary mixture from Dow Chemical Company which has to be used at 120°C during 100 hours under a nitrogen blanket. Decontamination factors as high as 1000 were expected with quite low corrosion rates on both carbon steel and stainless steel. The operation would produce a liquid waste volume equal to 4 times the primary loop contents; this solution will be concentrated by evaporation. New large waste treatment facilities have been built at Dresden for that purpose.

5.3 Heavy-water Reactors

In heavy-water-moderated reactors, such as CANDUs, the Siemens PHWR or the British SGHWR, great importance has to be attached to the preservation of the isotopic quality of the moderator. This has a severe impact on the decontamination operations, since the heavy water may not be displaced by ordinary cleaning solutions and acids and organic reagents may not be used in large concentrations with the heavy water in the reactor if not wholly deuterated. That problem can be solved in two ways:

- the "hard" approach, in which the D_2O-moderator (and the coolant, if the reactor is D_2O-cooled) is removed and stored in a special tank, while the decontamination is done using one of the processes already studied for PWRs and BWRs.

- the "soft" approach, in which the shutdown core is left in its normal state, the decontamination being made by injections of small concentrations of chemicals into the coolant which is recirculated at high speed and continuously purified by filtration and ion-exchange (the normal coolant purification system is usually, but not always, sufficient).

Experience with the "hard" approach is summarized in the following table (Table 5.3) and comments:

Table 5.3

Examples of "Hard" HWR Decontamination

Reactor	Date of D_2O-Dumping	Nature of Contaminants	Parts Decontaminated	Construction Material	Process Used	Duration	DF Achieved		
							Lowest	Highest	Mean
Hanford PRTR (USA)	Begin Sept. * 1962	Actinides	Primary Loop	Stainless Steel	High-speed flushing	Several days	High for alpha 1 for beta-gamma		
					OPP + OPG	31 h	1	1	1
		Fission Products			2 x APOX + APACE	9 d	1.8	56.5	16
Hanford PRTR (USA)	End December 1965	Activated Stellite corrosion products	Primary Loop	Stainless Steel	AP Citrox	3 d	12	44	24
Winfrith SGHWR (UK)			Primary Loop		APAC				

* The Hanford Plutonium Recycle Test Reactor (PRTR) was a D_2O-moderated and cooled reactor with vertical pressure tubes.

The Hanford Plutonium Recycle Test Reactor (PRTR) was a D_2O-moderated and cooled reactor with vertical pressure tubes. The fuel was plutonium in various alloys and ceramic compounds. The 1962 decontamination followed a fuel cladding failure incident on August 21, 1962.

The mechanical step (coolant flushing) occurred with the D_2O after removal of the fuel elements; the heavy water was recirculated through filters and ion-exchange material. After the resins were exhausted, the D_2O was removed and the flushing procedure was repeated with demineralized H_2O. Then all loose debris from the cladding burst, which carried most of the alpha-contamination, were removed. The fission products were already strongly sorbed in the protective oxide layers on the walls and remained there.

The first two mechanical steps (oxalic acid-hydrogen peroxyde-peracetic acid followed by oxalic acid-hydrogen peroxide-gluconic acid) were intended to achieve the final removal of the actinides.

The last APACE step was intended to redissolve the ferrous oxalate precipitated in the previous steps. Its duration includes research time, since decontamination experience was totally lacking in 1962.

The "soft" approach has been especially used in Canada for the decontamination of CANDU-reactors (the heavy-water moderated, light-water cooled modification at Gentilly has been treated in a similar way). The so-called CAN-DECON process is a proprietary development of AECL and Ontario Hydro. The solution used contains citric acid, oxalic acid and EDTA which are added to the coolant so as to reach a final concentration of 0.1%. The solution is continuously recycled through filters and ion-exchangers which fix the radionuclides and regenerate the chemicals. Little liquid wastes are produced, if any; the activity becomes fixed on 10 to 20 m^3 of waste resins which can be solidified by the normal plant radioactive waste system or by a specialized mobile unit. Although the magnetite films cannot be removed, DFs from 5 to 15 are claimed.

The CAN-DECON process has been applied to the Chalk River NPD and the Gentilly-1 and Douglas Point power reactors.

A similar process with higher temperature and a greater concentration of chemicals could be applied for light-water moderated reactors. DFs of 50 would then be attainable.

5.4 Gas-cooled Reactors

The family of gas-cooled, graphite-moderated reactors includes: the Magnox reactors (metallic uranium, Magnox-clad fuel); the AGRs (UO_2, stainless-steel clad fuel) and the HTGRs (high-enriched UO_2 or ThO_2 fuel, with graphite coating). The first two types have been widely used in France and the U.K. (where AGRs are still being built). The HTGR is still in the development stage; prototype units have been built by the US (Peach Bottom Fort Saint-Vrain) by the OECD (the Dragon reactor at Winfrith, England) and by the Federal Republic of Germany (prototype AVR at Jülich and the demonstration power plant being built at Schmehausen-Ventrop).

Although gas-cooled reactors are among the oldest nuclear plants in the world, decontamination experience with them is still lacking. Problems likely to be encountered with the classical chemical processes are due to the limited strength and tightness of the primary circuit, which is not designed to handle liquids, and the usually very limited liquid waste handling capacity at GCR plants. Film chemistry also is expected to be different from that in water-reactors, due to the higher temperatures encountered.

Oxalic acid-hydrogen peroxide systems have been studied at ORNL and at Jülich.

Other studies, related to possible decontamination of the Peach Bottom HTGR, investigated detergents, EDTA and a modified APOX process. Alkaline solutions are efficient for removing iodine which is one of the main contaminants expected in gas-cooled reactors.

An air-cooled reactor at the Idaho NRTS was decontaminated by removing the active components and treating them by washing with soapy water and scrubbing with steel brushes; the treatment was repeated twice. After that, the contaminants, which were activation products embedded in a loosely adhering film of Cr, Al, Ni, Fe and Si oxides were no longer detectable.

Magnox fuel elements were decontaminated at Harwell in an ultrasonic bath containing detergents and EDTA.

5.5 Liquid-metal Cooled Reactors

Liquid-metal cooled reactors include essentially the fast-neutron breeder which usually is cooled with sodium (some early experiments used a Na-K alloy). Decontamination could possibly occur after shutdown and coolant dumping, but the following problems would occur:

- Sodium is activated with ^{24}Na, this isotope decays fairly rapidly with a half-life of 15 h.
- Water cannot be introduced into the system until the last traces of sodium have been removed. The water-sodium reaction is strong and releases explosive hydrogen gas.

Although some countries now seem well advanced in the development of LMFBRs, decontamination experience on them is still insufficient.

U.S. research in the field included:

- Steam cleaning; the resulting gas being passed through catalytic recombiners to avoid hydrogen buildup;
- Hot oil cleaning;
- Sodium dissolution in alcohols, such as ethanol or butanol.

Care has to be taken with flammable solvents and hydrogen produced, but the sodium alkoxide formed can be handled safely.

- Reaction of sodium with anhydrous ammonia.

Available practical experience includes:

- Decontamination of a heat exchanger of the Fermi Reactor (USA) using ethanol.
- The decontamination of fission-products bearing items of the Dounreay Fast Reactor (UK) using steam.
- The decontamination of the BR-5 reactor (USSR) after extended operation with defective fuel elements. After steam washing, the primary system was cleaned with $KMnO_4$, HNO_3, oxalic acid and finally water, and after several such cycles, a DF of at least 50 was reached.

6. DECONTAMINATION OF EQUIPMENT, BUILDINGS

6.1 General Considerations

In this chapter, "equipment" means any part of a nuclear facility except for reactors and small laboratory facilities. It includes pumps, valves, nozzles, pilot loop components, remote handling devices and large vessels, tanks and contact devices used in reprocessing plants, waste management complexes and other fuel cycle facilities. Pieces of equipment usually can be removed from the place where they operate and brought to a specialized decontamination centre.

"Buildings" include laboratories, hot cells, reactor halls and other rooms used for activities involving radioactive materials. The walls and floors of these constructions can become contaminated, either inside or outside, due to normal plant operation or incidental releases.

Decontamination decisions in these fields involve safety and economic considerations. The decision-maker always has to keep in mind that various decontamination levels, including no treatment at all, are possible, and he must reason with the future destination of the item to be decontaminated in mind: its reuse in a nuclear facility, release as waste in a controlled repository, or in an unrestricted dump, entombment of buildings including some physical protection, their demolition or release for unrestricted use; ground to be used as an industrial estate, as a residential estate, as agricultural fields, buildings to be condemned, and so on.

6.1.1 Safety

This will include classical safety (handling of heavy material, of corrosive chemicals, and so on) and radiological safety. This last item will consider:

- Dose commitment of decontamination workers.

  This is zero in case of no decontamination and increases as the pre-set residual level decreases.
- Dose commitment of waste treatment and disposal workers.

  This will depend on the volume and characteristics of the wastes to be generated, the options chosen for their management, the quality of equipment and the skill and training of decontamination and waste workers. Pieces of equipment which are not intended for reuse, but treated only to facilitate their subsequent management as wastes, have of course to be accounted for here.
- Dose commitment of future plant workers.

This will be maximum if no decontamination occurs, and zero in the hypothetical, practically unachievable case of total decontamination. This point has to be handled differently if the item or building is restored to its former use or to another function, bearing in mind radioactivity, or if it is transferred to another industry, in which the future workers have to be considered as the "general public".

- Dose commitment of the general public.
  This includes various factors such as:
- External exposure to radiation from contaminated objects, structures and soil;
- Contact exposure through fallout particulates or handling of objects, especially those contaminated with beta-emitting nuclides;
- Internal exposure through inhalation of particulates or gaseous nuclides such as ^{85}Kr or ^{131}I, or through ingestion of crops grown on contaminated ground.

The calculation of the dose to the general public will take into account dispersion of nuclides during the decontamination operation, residual levels after decontamination and exposure through the various stages of waste mnanagement.

The safety analysis will help to eliminate those options which would cause exposure to radiation workers or to individuals in the general public above that indicated in the ICRP recommendations.

6.1.2 Economics

The economic analysis will include:

- Decontamination costs:
  - Abrasives, chemicals and other goods used in the decontamination operation;
  - Depreciation of equipment and building of the decontaminaton centre, if any; and of large pieces of equipment used in the operation, such as trucks. scoops, remotely controlled robots,;
  - Corrosion and wear of the decontaminated items, if recycled;
  - Manpower used in the decontamination;
  - Collective dose commitment due to the decontamination operation.
- Waste management costs:
  - Goods, equipment, manpower and collective dose commitment from all steps of decontamination waste management including pretreatment, insolubilization, transportation, storage and disposal.
- Investment savings
  - Value of equipment and buildings which would become useless if not decontaminated, or of ground which would have to be condemned;

  - Increased efficiency in using equipment or buildings through better accessibility, increased safety, possibility of repairing or replacing internal parts, and so on.
- Manpower savings:
  - Increased utilization of manpower through lower radiation levels which make continuous work possible;
  - Increased efficiency of workers due to better psychological atmosphere through increased safety;
  - Decreased collective dose rate after decontamination.
- Waste management savings:
  - Lower requirements for insolubilization, storage and disposal if contamination or radiation levels are reduced (this, of course, depends upon local regulations) for items not to be recycled.

6.2 Decontamination Processes

6.2.1 Equipment Decontamination Centre

Each large nuclear facility would have a well-equipped decontaminaton centre, run by specially trained personnel. It would include:

- A reception cell in which the pieces brought in for decontamination would be checked, sorted and routed to the appropriate treatment line;
- An alpha-sealed room which workmen with suitable protective clothing and respiratory equipment may enter through airlocks;
- Beta-gamma shielded hot cells for treatment of items surrounded by strong radiation fields. Equipment in these cells will be remote-controlled.
- An area equipped for vehicle decontaminaton;
- Overhead cranes and other necessary heavy handling tools;
- Waste conditioning equipment;
- Changing rooms for pesonnel;
- Ventilation equipment and stack;
- Monitoring room for assessment of decontamination efficiency.

The processes used can be roughly classified as wet and dry ones. Wet process areas should be equipped with tray floors above sumps connected to appropriate liquid waste collection systems.

Dry processes include abrasive blasting, vacuum cleaning, brushing and other similar mechanical treatments. They usually generate a lot of contaminated dust and ventilation of these areas has to be designed with care.

Wet processes include water or steam jets, alone or with abrasives such as boron oxide, glass beads, sand and the like; treatment with detergents, complexants or other chemicals, either by soaking in vats or by spraying with nozzles. Rotating object holders or nozzles can be used to increase efficiency. Advanced wet processes used are electropolishing and ultrasonic cleaning.

Lead can be decontaminated by melting and slag removal. The geometrical layout of the decontamination centre will ensure, as far as possible, direct handling of the objects being treated, avoiding useless and sometimes cumbersome transfer operations. Objects with smearable contamination will be appropriately wrapped before entering the centre and, if necessary, after leaving it.

6.2.2 Decontamination of Buildings

Large surfaces can be decontaminated either:

- By flooding with detergent-containing water or steam:
  This procedure is not often used because of the large liquid waste volumes generated. This drawback can be alleviated to a certain extent by using high-pressure water- or steam-jets.
- By swabbing with decontamination solutions:
  After delineation of the contaminated zone, the area is covered with the minimum amount of decontaminant (water with detergents, if necessary, chelatants, gelifiants, or other chemicals) and swabbed with a suitable device (long handles allow "remote" operation).
- By mechanically removing the surface:
  This will be necessary for porous surfaces such as concrete or wood if they have not been protected by impermeable paints or coatings. In such cases contamination may penetrate deeply into the substrate. Removal can occur with pneumatic drills, hammers and chisels, vapor blast machines or vacuum blast machines. With the first two systems, the dust produced is not captured and the workmen have to wear protective clothing and respiratory equipment.
- By thermally removing the surface:
  This process using laser irradiation is still in the development stage.

An often useful alternative, especially in case of alpha-contamination is to fix the nuclides and shield their radiation by means of an appropriate coating. Permanent marks should then be used as a constant reminder of the underlying radioactivity.

6.3 Monitoring

In decontamination centres, provision will be made for a "cold" shielded area where the decontaminated items will be checked by appropriate counters and smear tests. According to the measured efficiency of the decontamination, the objects will be sent back for process reuse or released for reuse or for management as waste.

Routine monitoring will be made on releases through the stack and the liquid effluent lines. The efficiency of the various waste management steps will also be checked. Exposed workers will be repeatedly examined by health physicists and the doses received and internal contamination will be recorded.

In buildings, monitoring will be carried out either through the installed radiation detection system of the plant, or with portable measuring devices, possibly controlled remotely or mounted on a robot. Attention will be given to hot spot search since localized sources must be removed if efficient decontamination is to be achieved.

Outside the plant, monitoring can be done from vehicles, possibly shielded if radiation fields are strong, or from planes or, better, from helicopters. Aircraft surveys will be reliable only if both altitude and speed remain rather low.

In case of release of airborne activity (dust generation or leakage of radioactive gases), samples will be taken from the atmosphere and analyzed. In case of fall-out on land, samples have to be taken from ground, crops, drinking water, meat and milk. Food products have to be kept away from distribution markets if heavily contaminated (This was the case with ^{131}I contaminated milk after the 1957 Windscale fire.)

Decontamination workers have to undergo constant medical examination too, especially if their work generates much dust.

7. TREATMENT OF WASTE RESULTING FROM DECONTAMINATION PROCESSES.

7.1 General

A waste handling capability has to be developed everywhere decontamination operations are likely to occur. The spent cleaners and rinse solutions are radioactive wastes, the handling, storage and disposal of which should be made an integral part of the decontamination method and should be considered thoroughly during all phases of development and planning.

Decontamination wastes are usually too radioactive to be managed by dilution and dispersion. They will have to be concentrated to volumes suitable for long-term storage or disposal while the release of the remaining liquid, with only very low levels of activity will be permitted into a river, a lake or the sea. A lot of experience is presently available for the conditioning of radioactive wastes and the methods applied for decontamination wastes usually involve only minor modifications of standard processes.

Handling, controlling, monitoring, conditioning, transport, storage and disposal of decontamination wastes has to be done taking into account the chemical composition, the radiological level and the isotopic concentrations encountered. In all cases the safety of the waste workers and of the general public will be guaranteed in accordance with ICRP recommendations (ALARA exposure levels). Both material and health costs of decontamination wastes management, will have to be accounted for when doing the economic analysis leading to a solution to the decontamination problem.

7.2 Characterization of Decontamination Waste

Radioactive wastes from decontamination operations may differ from normal operation wastes, both in nature and quantity. Their chemical, physical and radiological characteristics will depend upon the mechanical and/or chemical methods adopted for decontamination and upon the object which is to be cleaned. A detailed characterization of the wastes to be produced, including the radionuclide inventory, should be a part of decontamination planning. Radioactive waste treatment facilities would be designed according to the quality and quantity of these wastes.

Most of them will be low level liquids and solids carrying the nuclides removed from the cleaned objects.

Solids will be protective clothing and tools used by the workers during the operation, contaminated pumps, pipes, hoses and the like, and filters used for treating the exhaust of ventilated areas. Levels usually will be quite low.

Liquids will consist of spent cleaning and rinse solutions containing abrasives, detergents, acids, oxidants, alkalis, complexing agents and dissolved or suspended corrosion products and radionuclides. Activity levels can range from low to medium, making shielding necessary in some cases.

Some constituents often found in decontamination wastes can cause trouble in treatment and conditioning. So abrasives will induce accelerated wear in equipment; detergents will cause foaming if attempts are made to evaporate the solution; strong reagents will severely corrode process vessels; complexants will impair the efficiency of precipitation/flocculation steps and the leaching resistance of the final product; some solutions, if stored together, would cause troublesome precipitations, or even explosive reactions, and some constituents will limit the choice of conditioning matrices.

7.3 Treatment, Conditioning, Storage and Disposal of Wastes

The contaminated solids produced in decontamination operations will be managed by incineration or compression, if possible, and then embedded in an appropriate matrix such as concrete or bitumen.

The liquids will be concentrated by evaporation or by filtration, preceded by a chemical precipitation and flocculation step.

Usually the solution will have to be pretreated:

- By gross filtration, to remove abrasive solids;
- By neutralization to reduce the corrosive power of acids, alkalis and oxidants;
- By some adapted chemical process aiming at the destruction of chelatants and/or detergents.

The whole process will have to be tested on laboratory and pilot scale with simulated solutions before attempting the real decontamination. The necessary apparatus will be sufficiently large and the structural materials will be chosen according to the expected composition.

Evaporation and chemical precipitation will produce a concentrate carrying most of the initial activity and a clear solution, for which controlled release into the environment will be allowed after a last purification step, such as ionic exchange or electrodialysis if local conditions make it necessary

The concentrate will be embedded in a suitable matrix such as concrete, bitumen or thermosetting resin, and conditioned in steel drums or concrete shieldings according to its radiation level.

Conditioned waste will then be stored in adequate structures, to wait for a disposal programme or for decay of short-lived radionuclides such as ^{60}Co. Disposal can be shallow land burial (as is done in the U.S.) deep ocean dumping (such as done by some European countries, according to the London Dumping Convention and the IAEA recommendations, under surveillance by the NEA) or in deep geological formations such as crystalline rocks, clays or salt.

Transport between production installations and storage or disposal sites will be carried out according to the appropriate regulations.

Although constraints, due to the presence of unusual chemicals, high amounts of undissolved solids and rather high activity levels, may pose difficulties in achieving the required treatment efficiency to meet the legal discharge limits; well-proven techniques are now available for the management of decontamination wastes. Experience gained in some countries is described in the appendices.

8. CONCLUSIONS

Decontamination processes used to decontaminate reactor primary circuits are sub divided into two groups. These are:

1) The "soft" decontamination techniques using low concentrations of chemicals with ion exchange regeneration;
2) The "hard" decontamination techniques using much higher concentrations of chemicals.

Both techniques have advantages and disadvantages. While the "soft" techniques have low corrosiveness, can be applied easily (provided the equipment is available) and quickly, can be used on the whole of the primary circuit, and the wastes are concentrated on filters and ion-exchange resins, DFs of only 2 to 6 are obtained.

On the other hand, the "hard" techniques give high DFs but the solutions are more corrosive, not always applicable to the whole circuit, and they produce large volumes of liquid waste which must be disposed of after treatment and solidification.

After completing a cost-benefit analysis of the various decontamination options, the operator must choose which option will produce the optimum decontamination. He should include in the analysis the cost of equipment needed for both techniques. For example, for the "soft" technique the coolant purification system will most likely need to be modified. Whereas, for the "hard" techniques, additional equipment will be needed to treat, reduce and solidify the large volumes of waste for disposal.

Both techniques have been used in power reactors. The "soft" techniques have been used successfully in the Canadian reactors. The "hard" techniques have been successfully applied in PWRs and BWRs. Although the soft techniques are showing promise for the decontamination of BWRs and PWRs, final process efficiency for these reactor types remains to be demonstrated.

Whichever method the operator decides to use, greater benefits and better optimization would result if up-to-date know-how were applied during the design phase of the reactor. The operator or utility may decide if it were cheaper and more effective to have portable equipment to decontaminate different reactors.

Future trends are undoubtedly aimed at more efficient decontamination, which means producing only a minimum in secondary radioactive wastes by application of highly selective techniques and/or processes using chemical compounds that can be either recycled after regeneration or completely destroyed leaving virtually no additional residues at all. Any step towards a reduction of waste arisings is a step in the right direction. Decontamination processes are most promising measures in that way.

THE SWEDICH PROGRAMME FOR RADIOACTIVITY CONTROL IN BWR-REACTORS

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The relatively low increase-rates of radiation from the pipings in the Swedish BWR-reactors have resulted in very low man-remdoses for the operation and maintenance personnel. The swedish radiation protection authority has still put up a new goal for the total man-remdose for powerproduction that can be hard to obtain. That is 0,2 man-rem per installed MW electric in a year and that is a figure that we hadn't been able to obtain in our first BWR-reactor in Oskarshamn.

As the goal is a mean-value for the whole swedish programme we started to look especially on the design of our new reactors to see if they could be even better and thus give us the possibility to stay under that level.
In this way the programme for activity control started up, and we soon found out that the new stations that had only partial condensate polishing not could be expected to give better results than the old ones.
This fact started up the discussions of a system-decontamination method and we decided to develop such a method on a joint swedish base where also the manufacturer Asea-Atom participated.
The research work for the decontamination method is beeing done at Studsvik (The swedish nuclear research centre) but must in a later phase be done at the power-reactors in real systems. The work with modeling the activity building-up process is predominantly done by the utilities in cooperation with Asea-Atom and that includes also a lot of special samplings, analyses and measurements done at the stations.

The strategy in using the decontamination method will depend on the results of the method itself but also on the prognose for past-decontamination operation that can come from the model.

Understandness of activity build-up

The understandness of the processes involved in the radioactivity build-up will come out from analyses of a big data-base. This data-base is now growing up from five different programmes

- normal operation chemistry control (collected from all swedish BWR reactors)

- integrated sampling techniques
- oxide characterization of real reactor surfaces
- fuel deposit examination
- MADAC (mobile analyser for determination of activity contamination)

From this data base we have already tried to get a good mathematical model describing the process of radioactivity build-up in our BWR-reactors. The model can be used as a prognose instrument and for testing different ways of controlling radioactivity build-up in the reactor systems.

What the modelling already has shown us is that the high temperature filtration of reacto-water through beds of magnetite that was utilized on our first swedish BWR stations can have a significant effect on the activity build-up of Co-60 in the systems. Beside the particle filtration they also utilized the isotopic exchange process that tends to equal that specific activity in the coolant to that in the filter media.
Problems with backflushing the filter and a subsequent increase in pressure-drop forced us to stop using the filters. Problems to describe and understand their effectiveness in contamination control have also contributed to the decision not to use high-temperature filters in the newer swedish BWR power stations.

These new findings from the model will once more start the discussion about high temperature filtration and especially about the possibilities to use the existing installations in our older reactors.

Development of a full system decontamination method

Due to the many advantages of a mild chemical decontamination method like the canadian can-decon method we decided early to see if a similar method could be developed for the swedish BWR-reactors.
After the first beacher-tests on typical BWR specimens we fixed the chemistry conditions for the process to be able to get along with corrosion tests for the different construction materials in our power-reactors.
The tests were done in a high temperature loop at Studsvik and included beside of a lot of corrosion test coupons also a contaminated piece of piping from the clean-up system in Oskarshamn.
To the loop was attached a special regeneration and clean-up loop that is necessary for this kind of decontamination process.
The loop could be operated under BWR conditions and this was done for a longer time before the first decontamination and also between each of the following IO decontamination cycles.
The decontamination results that could be followed in the contaminated piece of piping were satisfactory with DF $\sim$ 3.
The corrosion tests were also very satisfactory and after the ten decontaminnations no serious general or local corrosion attacks could be noticed on the test-coupons.

The components in the loop that also were examined in detail before tha decontaminations did not show any corrosion attacks of local character after the ten decontamination cycles.
The chemicals that were used at that time were a misture of oxatic and citric acid and EDTA in it's acid form. During the decontamination phase these chemicals were continuously regenerated through a cation exchange resin bed.

We think that in-pile decontamination could give more long-term effects and not give that rapid recontamination that can occur when a new protective layer is growing in contact with very contaminated water.

Therefore we had to do more exact examinations of fuel-decontamination to see what results that could give and to study eventual negative effects on the fuel integrity.

Fuel elements burned for about three years in one of our power reactors was decontaminated in a specially made loop in the hot-cell laboratory of Studsvik. The process showed no good decontamination effect on the fuel deposits and it was decided to find a more efficient chemistry for the decontamination process. Indications were showing that a more reducing condition should increase the efficiency of decontamination fuel surfaces and this is where we are today.

Beside of this we have also tested the procedures for the decontamination at a loop decontamination in the R2 test-reactor at Studsvik.

The main results of this decontamination can be summarized as follows :

- The time the reactor was needed for the decontamination procedure was less than 48 hours.

- The decontamination effect from the process was never expected due to the different types of oxides in the loop compared to the oxide-layers in a BWR. The loop had previously been operated under reducing PWR conditions.

- There is a problem to get a reactor loop completely free from residues of decontamination chemicals. Flushing of dead-legs could not be properly made in the loop and this is an important consideration that must be made before you go into a power reactor.
  The consequences of small amounts of residues don't seem to be too harmful. They will just give rise to a hort-time conductivity peak in the coolant during the heat-up of the system after a decontamination. The top of that peak can be in the order of magnitude 10-100 μS/cm.

The next part in the development programme will concentrate around the fuel decontamination. The most important questions are :
- can the decontamination influence the integrity of the fuel during following operation ?

- will the decontamination of the fuel decrease the rate of activity build up during following operations

- can the activity that will be released from the fuel during a decontamination be transported to the purification sustems in a safe way ?

DECONTAMINATION OF THE PRIMARY LOOP OF THE BR3 NUCLEAR POWER PLANT

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INTRODUCTION

The BR3 power station is a small PWR plant (net electrical output : 10.5 MW) located at the site and owned by the Nuclear Research Centre "CEN-SCK" in Mol.

Since the first criticality on Aug. 22, 1962, the reactor has operated with shim rods until Aug. 1964. After a period of modifications, the plant has operated from May 1966 to Nov. 1968 with D_2O-H_2O moderation (spectral shift).

From July 1969, the plant has operated continuously with chemical shim.

Until 1974, the plant was operated by several Belgian utility companies but is now operated by the "CEN-SCK".

The operation of the BR3 power plant has been devoted to two main goals, i.e. training of operating personnel and testing of fuel elements. From 1962 to 1974, the BR3 was used as a training centre by several Belgian utility companies. Most of these people who operated the plant successively are now working in the nuclear power plants of Chooz, Doel and Tihange.

More recently, the operation of the plant was aimed at the irradiation and testing of fuel elements in conditions representative of those encountered in current power reactors. Main contractors for the irradiation programme are Westinghouse, CEA (France), UKAEA and Belgonucléaire (Belgium).

The recent cores of the BR3 plant have been designed to introduce more and more plutonium fuel elements. The plant is now being operated (Core BR3/4A) with a 30 % plutonium core.

The two following cores (BR3/4B and 4C) will permit operation of the plant up to the end of 1980.

DECONTAMINATION OF THE PRIMARY SIDE OF THE BR3 NUCLEAR POWER PLANT

A chemical decontamination of the primary loop of the plant was performed in October 1975 after the operation of core BR3/3B. This operation was aimed at the following goals :

1. To permit the easy access to the steam generator in view of its inspection.

   The steam generator had not been inspected thoroughly since 1966 and it developed a leak - although very small - at the start of the core BR3/3B in July 1974

2. To reduce the radiation fields around the primary piping and equipment, to facilitate the repair and inspection tasks during defuellings.

The decontamination system used was a two-step process ; the first one by the application of an oxydiser to facilitate the final solubility of some cations, the second one by the application of a complexing solution to solubilise the cations in the form of complexes.

1. PRINCIPLE OF THE DECONTAMINATION

The first product employed (TURCO 4502) is a strong oxydiser ($KMnO_4$) in alcaline solution (KOH + NaOH). It has to be used with a concentration of 240 g/l at a minimum temperature of 95 °C.

The second product (TURCO 4521) is a complexant agent in acid solution (oxalic and citric acids) to be used with a concentration of 75 g/l and at a temperature range of 70 ... 80 °C.

For an optimum efficiency, the solutions have to be circulated and heated to the desired temperature simultaneously.

Further, the acid solution (TURCO 4521) can only be applied after complete elimination of the first one (TURCO 4502). The removal of the traces of the TURCO 4502 can only be carried out by rincings with hot demineralized water.

By virtue of their decontamination properties, these two solutions have a destructive, corrosive nature. This effect is certainly most pronounced for the TURCO 4502.

For this reason, after an inventory of all materials present in the primary loops, some components had to be removed and replaced by others or specially protected. The best examples of these actions are respectively the replacement of the reactor head by a temporary one for the decontamination and the protection of the two primary pumps by a back-flush with demineralized water.

2. *PREPARATION OF THE DECONTAMINATION*

The preparation works were aimed at the following goals :

- to realize the correct configuration of all hydraulic loops entering in the decontamination process
- fabrication and installation of new components and systems
- replacement or adaptation of the components whose structural materials were not compatible with the corrosive agents.

The main modifications or transformations to the existing systems of the plant are summarized hereafter :

1) Replacement of the reactor head

Some components of the control rods lifting mechanisms are chromium plated. As it was considered that it was virtually impossible to assure a reliable protection of these components, the reactor head is not included in the decontamination process and was replaced by a temporary one specially fabricated for the purpose.
This latter was fabricated for a maximum working pressure of 25 kg/cm^2 in the primary coolant and coated inside by stainless steel.

2) Protection of the primary pumps

The graphite bearings of the two "caned motor pumps" had to be protected against the corrosive action of the TURCO 4502.
The protection is realized by a back-flush of demineralized water entering the motor via the vent valve (see figure 2) with a nominal flow between 250 and 300 l/hr.
The system is comprised of mainly two injection pumps, each one capable of maintaining the nominal back-flush flow to the two primary pumps.
In case of loss of off-site power, they can be powered from the Diesel generator. Ultimately, an emergency tank, under N_2 pressure is foreseen and can maintain the back-flush flow for a limited time.

3) Charging pumps

The chrome plated pistons of the charging pumps have been replaced by conventional stainless steel ones.

4) Mixing tank

The decontaminating products - powders - are solubilized in a closed mixing tank vented to the ventilation system.
The feeding of the products is realized by an "Archimedes screw".

5) Evacuation of the decontaminating effluents from the pressure vessel.

The newly installed piping permit the direct evacuation of the effluents to the three High Level Liquid Waste storage tanks (HLLW).

6) Auxiliary steam connection to the main steam line

This connection makes possible to feed the secondary side of the steam generator with auxiliary steam and thus to accelerate the warming up of the primary loop.

7) Liquid waste treatment

   The system was installed in the waste pumps room and connected to the three High Level Liquid Storage tanks.

8) Replacement of joints and menbranes in the auxiliary loops involved in the decontamination.

9) Instrumentation

   All unneccessary instrumentation cells have been isolated.

Less attention was paid to the protection of the primary instrumentation cells (pressure, level) because most of them had to be replaced by new ones after the decontamination any way. However, it was guaranteed that these cells could furnish reliable indications during the process of decontamination.

New cells had to be installed to implement the instrumentation, the main ones being :

- Level and temperature cells for the Spray Storage tank. The readings of these cells are reported to the Operating Deck (Containment Building)
- Two new cells to record the level of the High Level Liquid storage tanks
- Pressure and level instrumentation cells for the SIBA tank (Emergency tank for the back-flush)
- Pressure and level instrumentation cells for the secondary side of the steam generator. The readings are reported to the Control Room.

3. *PROCEDURES FOR THE DECONTAMINATION*

The essential operations for the decontamination consist mainly to prepare, introduce, circulate, heat up and finally evacuate the chemical solutions. These operations are followed by rincings with demineralized water.

These diverse operations are describedin more detail in the following chapters. A schematic diagram of the loops involved is presented in fig. 1.

3.1. Mixing loop

The chemical solutions are prepared, homogenized and heated to the desired temperature in the Spray system consisting of :

- a mixing tank where the products are mixed with water
- the Spray Storage tank with a capacity of 12 m^3
- two circulating pumps
- a heat exchanger fed with auxiliary steam to warm up the solutions to the desired temperature (85 ... 90 °C).

Demineralized water is added directly to the Mixing Tank from the Charging system or to the Spray Storage tank from the Refuelling Water Storage tanks.

By virtue of the limited practical capacity of the Spray Storage tank (11m^3), the chemical solutions had to be prepared with an over concentration to take into account the dilution in the primary loops (± 16 m^3) with hot demineralyzed water.

3.2. Injection loop

The back flush system on the primary pumps (see figure 2) is first started. The chemical solutions are then injected directly, by the Spray pumps from the Spray Storage Tank to the empty and preheated (90 ... 95 °C) reactor vessel.

3.3. Filling up the primary loops

After injection of the chemical solutions, the Spray Storage tank was partially filled with demineralized water from the Refuelling Water Storage tanks.

By circulation over the Spray heat exchanger, this water is preheated and then injected in the high pressure loops.

The purification system (low pressure side) is filled up with preheated water from the Boric Acid tank.

3.4. Pressurisation of the high pressure loops

The pressurisation is maintained by a nitrogen blanket in the pressurizer to avoid any overpressure transients. The nominal working pressure is fixed at 20 kg/cm^2.

3.5. Heating up the primary loops

The chemical solutions are brought to the desired temperature by the primary pumps and by steam on the secondary side of the steam generator. The temperature is then stabilized by the Purification system.

The use of the purification loop with the decontaminating solution was submitted to the following conditions :

- the temperature in any point must be higher than 55 °C to avoid any cristallisation of the chemical solutions. An upper limit for the temperature (65 °C) is set up by the charging pumps.
- an empty demineralyser vessel will always be used to filter out the solid particles in suspension
- one charging pump is in stand-by, isolated and filled up with demineralized water, the other circulates the chemical solutions
- a charging pump must be flushed with hot demineralized water as it has to be stopped.

3.6. Evacuation of the solutions

The high pressure loops are isolated from the purification system and first depressurized to 5 ... 6 kg/cm^2.
This nitrogen pressure then pushes the chemical solutions out of the reactor vessel via the Spray-out line to the High Level Liquid Waste storage tanks. The drainage of the cold legs is completed to a Discharge tank.

The charging pumps of the purification system are first flushed with hot demineralized water from the Boric Acid Tank and isolated.
It is then proceeded with the drainage of the chemical solutions to the High Level Liquid Waste storage tanks.

3.7. Rincings of the primary loops

Demineralized water preheated to 85 ... 90 °C in the Spray Storage tank is injected into the hot and empty pressure vessel and circulated for a couple of hours.
The procedures followed are identical to those described in § 3.2 to 3.6 above.

3.8. Storage of the active effluents

Three tanks (HLLW), 56 m^3 each, collect the highly active effluents from the decontamination.
The storage tank n° 1 receives the low active effluents from the rincings. These effluents can afterwards be transferred directly to the Waste Processing Dpt of the CEN-SCK after control of the activity level.
The storage tanks n° 2 and 3 receive respectively the high active and chemically loaded TURCO 4502 and TURCO 4521 solutions. These solutions are stored before treatment on site and ultimate transfer.

4. EXECUTION OF THE DECONTAMINATION

The actual decontamination was preceeded by series of tests on components and systems to prove their reliability.

The first phase of this testing period began on September 30, 1975 by the first filling up and pressurisation of the primary systems. As some difficulties were encountered with the leak tightness of hydraulic penetrations on the pressure vessel (Spray lines), the hydro-test of the primary system was delayed to October 4, 1975. The hydro-test was then carried out by maintaining a pressure of 37.5 kg/cm^2 at a temperature of 150 °C for one hour.

In a second phase, with the reactor still in nominal conditions (20 kg/cm^2 and 90 ... 95 °C) for most of the time, all auxiliary systems were thoroughly tested.

Some difficulties were reported with the two Spray pumps and the back-flush system :

1) Spray pumps

   The graphite rings of the mechanical seals had been replaced by synthetic material. Experience showed that a complete leaktightness could not be maintained at high temperature. So, the original graphite rings were again used but with a simple back-flush system of 2 ... 3 l/hr demineralized water for cooling and protection against corrosion by the TURCO 4502. New tests on each pump for about 20 hours at 85 ... 90 °C showed that the two pumps worked satisfactorily.

2) Back-flush system

   Some problems were experienced with the two hired injection pumps, but could afterwards be solved by a mechanical revision of the pumps.
   Instabilities of the back-flush flow were corrected with two small "surge-tanks" placed at the outlet of the injection pumps.

At the same time, all the proposed procedures for the actual decontamination were verified and carried out several times to familiarize the operators involved.

This test period ended on October 18 with a last complete simulation of all the operations.

4.1. *The operation with TURCO 4502*

The first phase of the decontamination process was initiated on October 20 with the solubilization of 3850 kg TURCO 4502. The eleven cubic meters of solutions (367 g/l concentration) were maintained at 90 °C until the injection into the primary system on October 21.

The solutions were circulated in the primary loops during 2.75 hr between 97 °C and 99 °C whereas the total time, between the injection and the complete evacuation was 6 hours.

The TURCO concentration ranged from 282 g/l after a half hour circulation to 226 g/l after 2.75 hours circulation.

The average activity of the solutions during the run amounted to 1.56 Ci/m^3, the isotopes Cr^{51} and Mn^{54} contributing to 70 % and 20 % respectively of the total activity (see tables 1 and 2).

Three successive rincings were carried out before the introduction of the TURCO 4521.
At the end of the last rincing, the remaining concentration of TURCO 4502 was reduced to 69 mg/l but the pH could not be lowered under ~ 10.5 (see table 3).

4.2. *The operation with TURCO 4521*

The second phase of the decontamination began on October 22 by the solubilization of 1200 kg TURCO 4521.
The eleven cubic meters of solutions (112 g/l), maintained at 80 °C, were injected into the primary loops on October 23 and circulated for 1.5 hours at 87 °C average temperature. The total time of the solutions in the primary system was 4 hours from the injection until the complete evacuation.

The TURCO concentration ranged from 86 g/l after a half hour circulation to 76 g/l at the moment of the evacuation.

The evolution of the Co^{60} activity was continuously monitored during the circulation of the products. As only a leveling-off of this activity could be observed, the circulation was limited to 1.5 hours. The mean activity of the solutions during the circulation amounted to 11,4 Ci/m^3, more than 90 % of which was represented by the Co^{60} (see table 2).

Five successive rincings of the primary loops were applied from October 23 to October 28. During the third rincing, the remaining concentration of TURCO 4521 could be reduced to 9 ppm (see table 3). The two last rincings were carried out at low temperature with a mixed-bed ion exchanger in service, so that the activity of the primary water could be reduced to about 8.10^{-3} % of the initial activity (see tables 2 and 4).

Two incidents occurred during these rincings. The first one occurred on October 24 when a joint on the Spray-out line (see figure 1) suddenly failed, with as consequence a rapid depressurisation of the primary loop. Fortunately, the liquids that leaked out were only slightly radioactive and the operations could be resumed a few hours later.
The second incident involved primary pump n° 1. This one was definitively switched off when abnormal noises, vibration levels and absorbed electric current were reported.

5. *RESULTS AND CONSEQUENCES OF THE DECONTAMINATION*

5.1. *Summary of the collected activities in the effluents*

Some two hundred and twenty cubic meters of liquid waste were generated during the decontamination process (see table 5). The most active of these solutions, the concentrated TURCO 4502 and 4521 solutions, were respectively stored in the High Level Liquid Waste storage tanks n° 2 and 3 whereas most of the effluents from the rincings were transferred to the Waste Processing Department.

The total activity of these effluents amounted to 232 Ci, of which more than 98 % for the corrosion products only (mainly Co^{60}).

The alpha activity of the effluents can be roughly deduced from the measurements carried out for the different phases of the decontamination. The results of these measurements are reported in Table 6.

5.2. *Chemical analyses of the decontaminating solutions*

Chemical analyses have been carried out on samples taken during the decontamination process and in the HLLW storage tanks. The main results of these analyses are summarized respectively in the tables 7 and 8.

5.3. *Radiation fields after the decontamination - Decontamination factors obtained*

- Some measurements of the radiation fields around the primary system before and after the decontamination are presented in Table 9.
  The Table 10 summarizes the obtained decontamination factors for different parts of the primary loops.

  As foreseen, the best decontamination factors were obtained where optimum temperature conditions could be maintained. The effect of temperature is clearly seen for the Purification loop, the worst results being found downstream the Regenerative Heat Exchanger.

  The low decontamination factor around the pressuriser is difficult to explain ; maybe the reason of this could be found in the low flow in this section of the primary loop.
- The radiation fields inside the Steam Generator after the decontamination are summarized in Table 11.

5.4. *Residual activities on the inner surfaces of the primary loops*

The residual activities on the inner surfaces of the primary loops were evaluated at different locations. Results for the Steam Generator are presented in Table 12.

5.5. *Damage to primary pump n° 1*

The lower radial and axial bearings including the graphite rings of this primary pump were found seriously damaged.
It has been proved that the back-flush system did not entirely prevent the infiltration of the decontaminating solutions into the motor.
Nevertheless is is most improbable that the decontaminating solutions were the only cause of the observed damages because the analysis of all the records proved that the first abnormal vibration levels, although slight, did happen before the introduction of the TURCO solutions. Moreover, the observation of the damaged components showed that these components were overheated but no

sign of corrosion was found. So it seems more likely that the damages were the result of an insufficient or deficient venting of the pump.

Primary pump n° 2 was also inspected after the decontamination. No damage was found, only the axial and radial clearances were measured and found to be just above tolerances. Again it could not be concluded that the slight abnormal wear on the graphite blocks and rings was the direct consequence of the decontamination process.

5.6. *Difficulties with the unloading of the reactor internals*

In December 75, it was proceeded with the unloading of the reactor internals. On this occasion it was observed that the two main upper internals, namely the Spray Box and the Instrumentation Basket (collar assembly) could not be uncoupled.
The reason for this situation could only be explained by a malfunctionning of the coupling connection between the collar and the Spray Box, namely the so-called "Corthals Joint". As no mechanical deformation of this joint could be identified, the only remaining cause was an internal friction due to TURCO deposits between the metallic surfaces. And indeed, the problem was resolved by the injection under high pressure of a cold solution of TURCO 4521 to the hydraulic seal of the "Corthals-joint".

6. *TREATMENT OF THE ACTIVE EFFLUENTS FROM THE DECONTAMINATION PROCESS*

As previously explained, the moderate active effluents - with a specific activity lower than $\leqslant 1$ Ci/m^3 - containing chemicals only in diluted form, were directly transferred to the Waste Processing Department of the CEN-SCK where they could be treated by conventional methods in existing facilities.

The same process could not be applied to the highly active and chemically loaded TURCO solutions.

The actual procedure applied consisted of thoroughly mixing definite quantities of the TURCO 4521 and TURCO 4502 with some added chemicals. By this process, a precipitation occurs and most of the active substances are recolted in the sediments.
Laboratory scale tests of this process showed a 98 % minimum efficiency.
After decantation, the liquid phase is recovered and processed by conventional methods.

Taking into account the different problems such as transport from BR3 to the Waste Processing, available place on site at the BR3 plant, time schedule to be respected and the foreseen activity of the effluents, the actual installation for the treatment was realized as follows :

1) On site at the BR3 plant

Two shielded mixing vessels of a 375 l/hr nominal treatment capacity - with all their associated auxiliary equipment such as piping, valves, pumps, instrumentation - are placed in the HLLW pump room.
The treated solutions are then transferred under vacuum from the mixing vessels to a shielded 750 l capacity container for transport to the Storage site of the Waste Department.

2) Storage site

A twenty cubic meters capacity storage vessel was installed and shielded with 50 cm thick concrete blocks.
The treated solutions transferred from the BR3 site are pumped into this vessel where the natural sedimentation is continued. Afterwards, the cleared liquid is pumped off and transferred for conventional treatment.

All of these equipments have been constructed so that they are completely demountable. The active sediments can indeed be flushed out of the vessel if another appropriate and definite solution is found for their storage.

7. *RADIATION EXPOSURE OF THE PERSONNEL*

The figures of the radiation exposure of the BR3 and foreign personnel during the decontamination period are presented hereafter. To get a correct idea of the impact of these figures, it should be remembered that the decontamination period followed directly two months defuelling and that the shut-down of the plant was continued up to mid-July 1976.

Radiation exposure (Rem)

Month	BR3 personnel (63 people in total)	Foreign personnel	Period
July & Aug. 75	44.7 58.4 13.7	21.8 25.9 4.8	Defuelling period
Sept. & Oct. 75	28.2 55.1 26.9	21.6 27.2 5.6	Decontamination
Nov. & Dec. 75	12.1 26.5 14.4	8.9 11.8 2.9	Shut-down (continued)

8. *CONCLUSION*

The experience at BR3 shows that very good results can be obtained by a attempt to decontaminate the whole primary side of a P.W.R. plant.
These results can be gained without any serious damage to the equipments. In our case, sixteen months after the decontamination, no damage has ever been attributed certainly to the decontamination process, if we except the dubious case of one primary pump.

However, the results of such a decontamination process have to be carefully weighted against other factors or problems and particularly the cost of such an operation, the exposition of the personnel to the radiations and last but not least the final evacuation and treatment of the large volumes of highly active effluents.

A decontamination is very expensive by the prices of the decontaminating products, the cost of the equipments and of the effluent treatment and also a long and tedious careful preparation in order to minimize the risks of the operation.

Finally, if it seems technically possible to extrapolate this experience to large production reactors, the problems associated with the storage, treatment of the large volumes of highly active effluents and the final evacuation of the residues are certainly of the most decisive.

FIGURES : 2
TABLES : 12

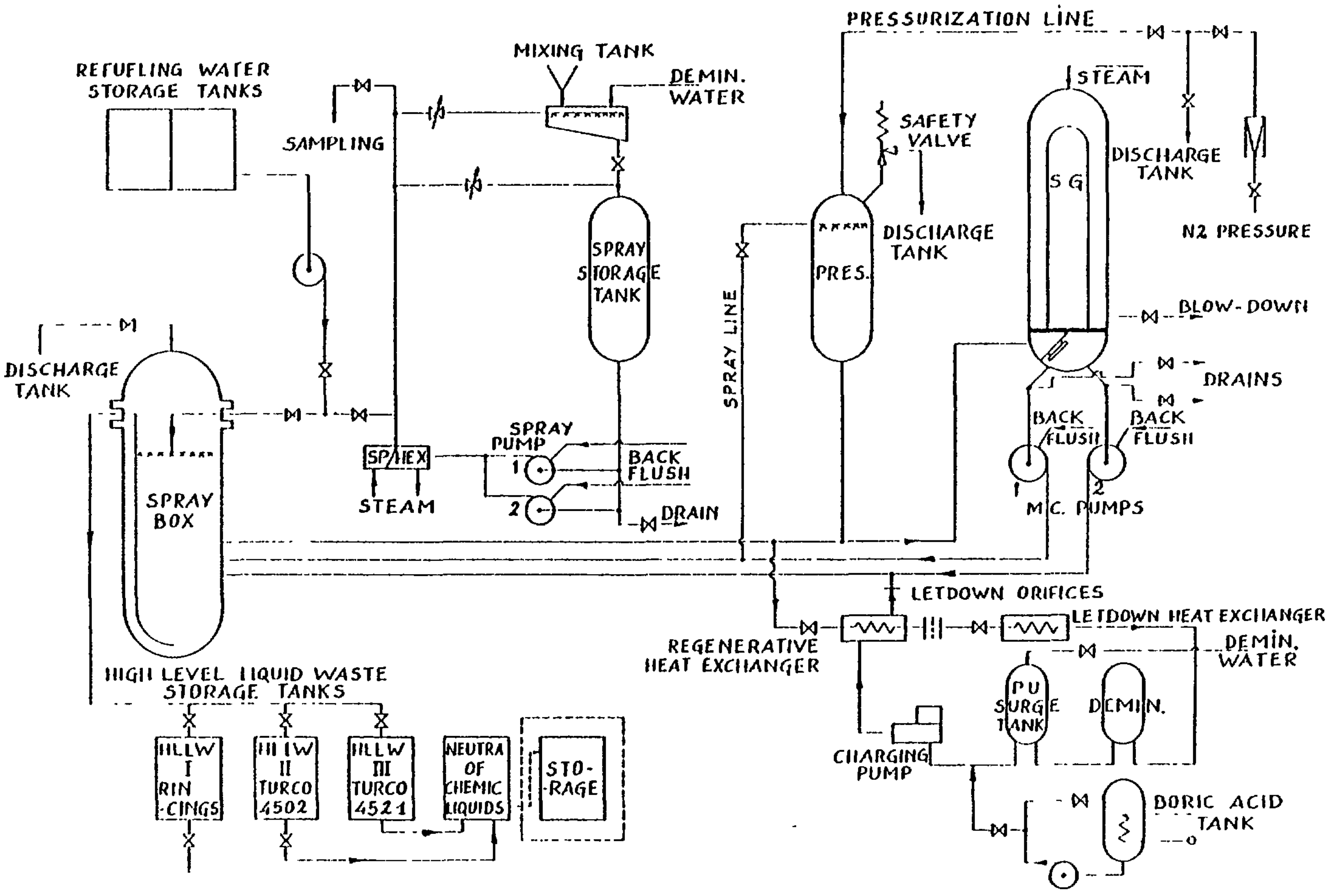

FIGURE 1: SCHEMATIC DIAGRAM OF THE DECONTAMINATION LOOPS

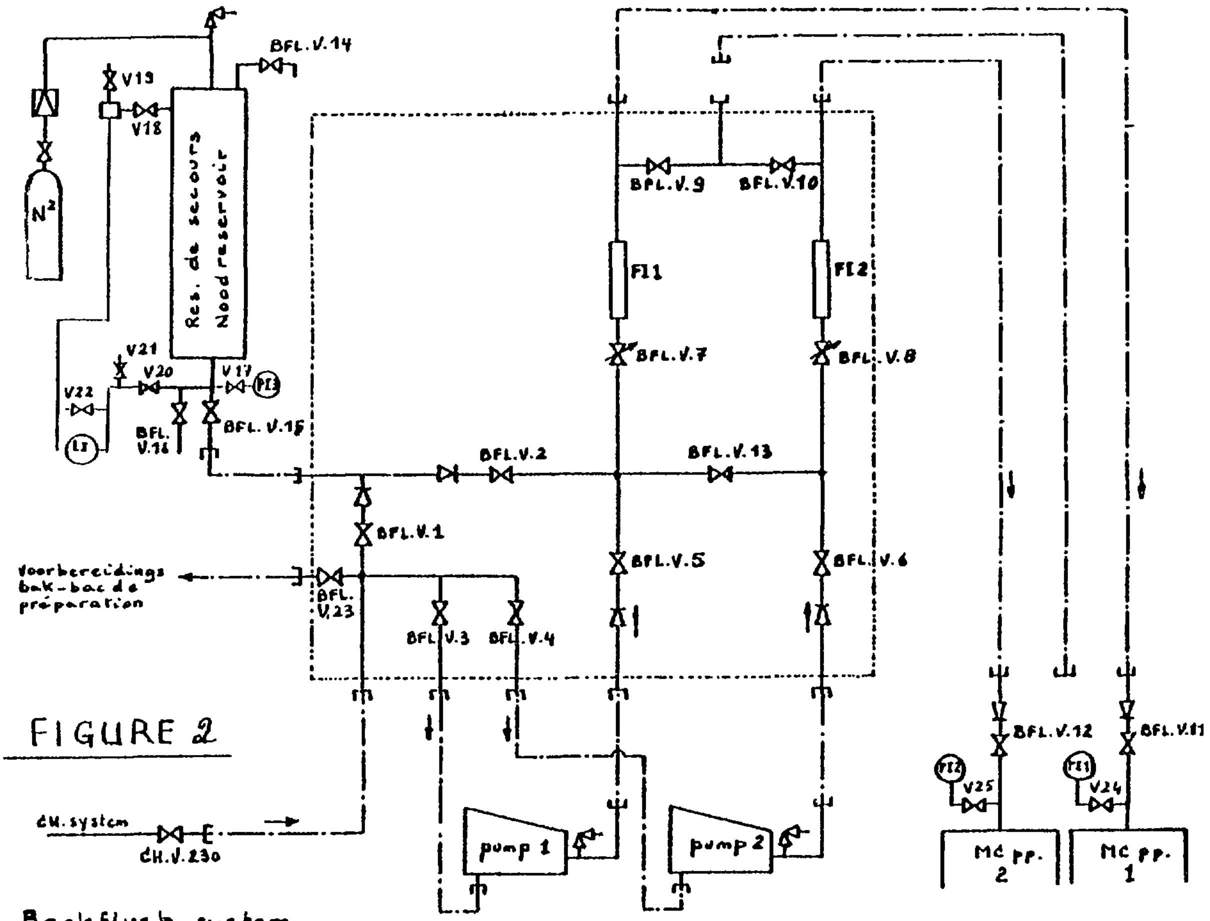

FIGURE 2

Backflush system.

Table 1

IDENTIFICATION OF THE SAMPLES

	FIRST PHASE (TURCO 4502)		
	1st sample	2nd sample	Last sample
Preparation of products in S.S.T.	A/01	A/02	A/A
Products in the primary loop	A/A/01	A/A/02	A/A/A
First rincing	A/B/01	A/B/02	A/B/A
Second rincing	A/C/01	A/C/02	A/C/A
Third rincing	AD/01	A/D/02	A/D/A

	SECOND PHASE (TURCO 4521)		
	1st sample	2nd sample	Last sample
Preparation of products in S.S.T.	B/01	B/02	B/A
Products in the primary loop	B/A/01	B/A/02	B/A/A
First rincing	B/B/01	B/B/02	B/B/A
Second rincing	B/C/01	B/C/02	B/C/A
Third rincing	B/D/01	B/D/02	B/D/A

Table 2

RADIOCHEMICAL ANALYSES OF THE PRIMARY COOLANT

(All activities in μCi/cc)

Isotope	A/A/A	A/B/A	A/C/A	A/D/A
Co^{60}	2,4 E-02	2,4 E-02	3,2 E-03	1,2 E-03
Mn^{54}	3,0 E-01	6,9 E-03	5,9 E-04	7,5 E-05
Co^{58}		8,7 E-04	1,0 E-04	1,8 E-05
Cr^{51}	9,8 E-01	3,7 E-02	1,6 E-02	1,1 E-03
Fe^{59}		5,0 E-04	6,7 E-05	
Ce^{141}			4,8 E-05	
Ce^{144}		4,7 E-04	1,8 E-04	
Ru^{103}	3,4 E-03	2,3 E-04	1,4 E-04	1,6 E-04
Ru^{106}	≤ 2,1 E-02	≤ 1,6 E-03	≤ 8,0 E-04	3,3 E-04
Ag^{110m}	1,9 E-02	8,4 E-04	4,6 E-04	1,1 E-05
Zr^{95}		1,3 E-04		3,3 E-05
Nb^{95}		4,8 E-04		1,5 E-05
Cs^{137}			≤ 5,0 E-05	1,4 E-05
Sb^{124}	2,4 E-02	1,7 E-03	1,4 E-03	1,4 E-04
Sb^{125}	5,1 E-03	7,6 E-04	5,3 E-04	9,0 E-05

Isotope	B/A/A	B/B/A	B/C/05	B/D/03	B/E/16	B/F/05
Co^{60}	9,9 E-00	3,0 E-01	4,3 E-01	2,4 E-02	2,3 E-02	1,5 E-02
Mn^{54}	4,2 E-01	5,9 E-02	6,6 E-02	3,0 E-03	2,5 E-03	1,6 E-03
Co^{58}	4,2 E-01	3,5 E-02	4,5 E-02	2,0 E-03	1,1 E-03	7,2 E-04
Cr^{51}	2,2 E-02	1,3 E-01	4,9 E-02	1,7 E-03	3,6 E-03	7,6 E-04
Fe^{59}	2,6 E-02	1,0 E-02	1,1 E-02			
Ce^{141}						
Ce^{144}	8,0 E-02	3,9 E-03			3,7 E-04	
Ru^{103}			7,1 E-04	2,0 E-04	2,4 E-04	1,2 E-04
Ru^{106}					2,2 E-03	
Ag^{110m}					2,4 E-03	3,0 E-04
Zr^{95}		1,4 E-03	3,4 E-03			
Nb^{95}	2,0 E-02	2,6 E-03	2,0 E-03			
Cs^{137}						
Sb^{124}	1,4 E-02	9,0 E-06	1,6 E-02	9,1 E-04	1,4 E-03	1,4 E-03
Sb^{125}			2,3 E-02	1,7 E-03	2,2 E-04	4,8 E-04

Table 3

PRODUCTS CONCENTRATION IN PRIMARY LOOP

Sample	Concentration g/l	pH	
A/A/A	226		TURCO 4502
AB/A	6,9		
A/C/A	0,65	11,40	
A/D/A	$6{,}9\ 10^{-2}$	10,45	
B/A/A	75,5		TURCO 4521
B/B/A	3,2	4,8	
B/C/A	0,097	7,7	
B/D/A	0,009		

Table 4

CHEMICAL ANALYSES OF PRIMARY COOLANT DURING THE LAST RINCINGS AFTER APPLICATION OF TURCO 4521

Sample	Date	Hour	Li (ppm)	K (ppm)	Na (ppm)	B (ppm)	pH
B/D/A	24.10.75	22.10	0,1	1	0,35		7,7
B/E/01	25.10.75	11.00	-	6	0,45		9
B/E/03	25.10.75	13.00	0,05	13	0,48		8,6
B/E/07	25.10.75	22.10	1,8	1	0,06		8,4
B/E/11	26.10.75	15.30	2,6	5	0,26	94	7,8
B/E/16	27.10.75	12.00	70	5	0,30	515	8,5
B/F/01	27.10.75	17.45	0,35	10	0,6		8,3
B/F/02	27.10.75	20.30	1,7	7	0,38		7,7
B/F/04	28.10.75	01.45	3,5	2	0,25	673	6,9
B/F/05	28.10.75	05.45	4,4	1	1,1	665	7,0

TABLE 5

SUMMARY OF THE ACTIVITIES COLLECTED IN THE EFFLUENTS

Nature of effluents	Volume (m^3)	Activity (Ci)
1st phase (Turco 4502)	50*	19,46
2nd phase (Turco 4521)	30**	191,5
Two rincings of first phase	31	1,46
Third rincing of the 1st phase + (partially) 1st rincing of the 2nd phase	29	16,22
Rincings of the 2nd phase	80	3,8
Total	220	232,44

* after dilution

** with 5 m^3 first rincing and after dilution

Isotope	Activity (Ci)	% of total activity
Co^{60}	186,5	80,24
Cr^{51}	21,64	9,31
Mn^{54}	12,64	5,44
Co^{58}	8,54	3,67
Fe^{59}	0,053	0,023
Total	229,37	98,68

TABLE 6

ALPHA ACTIVITIES OBSERVED IN THE PRIMARY COOLANT

	A/A/A	A/B/A	A/C/A	A/D/A
Total (dpm/cc)	4360	180	43	8,3
Plutonium (dpm/cc)	2760	81	22	1,3

	B/A/A	B/B/A	B/C/05	B/D/03	B/E/16	B/F/05
Total (dpm/cc)	2420	364	36	1,1	8,1	1,0
Plutonium (dpm/cc)	2270	190	13,6	0,5	7,2	0,6

TABLE 7

MAIN CHEMICAL SUBSTANCES MEASURED DURING THE DECONTAMINATION

	Sample	SiO_2 mg/l	Fe mg/l	Ni mg/l	Cr mg/l	Mn mg/l
TURCO 4502	A12 384g/l Turco	-	-	-	2980	20.000
	A/A/A	97	70	-	2300	16.000
	A/B/A	0,1	1,8	-	95	440
	A/C/A	0,025	0,37	-	-	-
	A/D/A	-	-	1,1	-	-
TURCO 4521	B/A/A	640	460	86	6	137
	B/B/A	55	55	7	1,3	10
	B/C/A	0,5	5	3	-	1,3
	B/D/03	0,025	0,24	0,3	0,017	0,17
	B/E/15	1,19	0,058	-	-	1,8
	B/F/04	0,28	0,01	-	0,85	5

TABLE 8

CHEMICAL ANALYSES OF THE EFFLUENTS

		SiO_2 (kg)	Fe (kg)	Ni (kg)
Quantities injected in the primary loop	Turco 4502	0,058	-	0,04
	Turco 4521	0,002	0,142	0
	Total A	0,060	0,142	0,04
Effluents	1st phase	0,05	0,06	0
	2d phase	18,64	5,95	1,32
	Total B	18,69	6,01	1,32
Balance	B - A	+18,63	+5,868	+1,28

TABLE 9

RADIATION FIELDS ON HIGH PRESSURE LOOPS AND PURIFICATION LOOP

	Location	Before decontamination - mR/h	After decontamination - mR/h
HIGH PRESSURE LOOPS	Cold leg n° 2	540	18
	Cold leg n° 1	460	13
	Hot leg	1100	78
	Primary pump 1	280	60
	Primary pump 2	290	62
	Steam generator tube bundle		
	- upper level	660 650	12 24
	- mean level	500 700	8 10
	- lower level	800 760	46 10
	Steam generator inlet & outlet chambers	220 940 380	10 100 68
	Steam generator manhole	420	28
	Pressurizer		
	- upper level	56	32
	- mean level	60	24
	- lower level	150	44

TABLE 9 (continued)

RADIATION FIELDS ON HIGH PRESSURE LOOPS AND PURIFICATION LOOP

	Location	Before decontamination - mR/h	After decontamination - mR/h
High pressure loop	Pipings purification loop	640	24
		760	45
	Regenerative heat exchanger	580 (inlet)	100
		950	360
		190 (outlet)	60
	Purification pipings after RHex	420	210
Low pressure loop (purification)	Pipings	200	65
	Letdown heat exchanger		
	- inlet	190	85
	- outlet	240	70
	Surge tank	320	50

Table 10

DECONTAMINATION FACTOR

	Number of measurements	Mean	Max	Min
Primary pipings	20	18,8	50	2,6
Steam generator	10	36	70	5,6
Pressurizer	9	2,35	4,1	0,7
Purification	50	6,7	27	0,6
Primary pumps				
Pump body	4	5,6	6,5	4,7
Motor stator	4	3,6	5,1	2,8

Table 11

OBSERVATIONS MADE IN THE PRIMARY SIDE OF THE STEAM GENERATOR

RADIATION

	γ radiation mRem/h	(β+γ) radiation mRad/h		
		Min.	Mean	Max.
Hot-leg	30 to 50	72	80	139
Cold-leg	20 to 40	46	68	91

Table 12

RESIDUAL ACTIVITIES ON SURFACES (SMEAR SAMPLES)

ISOTOPE	% OF TOTAL REACTIVITY
Co^{60}	76,35
Mn^{54}	4,49
Co^{58}	2,76
Cr^{51}	-
Fe^{59}	1,08
Ru^{106}	7,21
Ru^{103}	0,90
Ag^{110m}	2,34
Ce^{144}	2,71
Sb^{124}	0,43
Sb^{125}	0,54
Nb^{95}	1,12
Zr^{95}	0,90
Cs^{137}	0,73
Total	7.10^{-4} μCi/cm^2

DECONTAMINATION OF TECHNOLOGICAL SYSTEMS IN GERMAN DEMOCRATIC REPUBLIC NUCLEAR POWER PLANTS EQUIPPED WITH WWER-TYPE REACTORS*

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1. Introduction

Due to activity accumulation, the dose rate level of all material surfaces in contact with the moderator/primary coolant at operating temperature increases following an e-function during the entire operating time of a water-moderated and water-cooled reactor facility. At the present state of the art, this phenomenon can be influenced by degrees by means of structural and technological measures and material choice and water regime but it cannot be prevented.

In the nuclear power plants now in operation, dose rates limiting the access to facilities shut down for inspection or maintenance have already been reached after one reactor campaign. At the time of in-service inspections legally prescribed in many countries today or of necessary repair work due to wear - as a rule after a number of operating years - there is a radiation situation which has severe consequences for technology, duration and organization of respective work.

With a number of methodical variants of preparatory measures for such work, decontamination has gained considerable importance but its potential has not yet been comprehensively used in NPP practice. The latter applies particularly to such decontamination methods which cover the entire primary circuit and technologically separable partial systems or non-dismountable large equipment. This is favoured by the fact that

- the material expenditure for implementing such measures is of course high but evidently often overestimated;
- practical experience with the effects on materials is still small and also overshadowed by few negative examples;
- the requirements of decontaminating technological systems have been insufficiently considered so far in NPP projects.

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* This paper was prepared after the Technical Committee Meeting on request by the Secretariat.

It is therefore the aim of this paper to discuss experience gained in decontaminating technological systems of NPP units in the GDR and the conclusions to be drawn.

2. Decontamination as an integral part of maintenance strategy

According to the principles upheld in the socialist countries the protection of the health of workers takes priority in the conditioning of working processes. So, for working conditions during inspection and maintenance periods in nuclear power plants, the necessity is derived to include measures of minimizing external radiation exposure and of maximally preventing incorporation risks in the planning of maintenance work.

Further, in deciding on the means suited to this effect (shieldings, auxiliary devices, decontamination a.o.) and considering the respective total extent of inspection and maintenance work

- the creation of optimal technical conditions for successive work, e.g. the testability of the surface state, the accessibility for unpredictable measures of treating defect points, is aimed at;
- the economic expenditure is optimized, the effects of the measures taken on the duration of outage time, that is on the "scheduled path" of the maintenance process, being the centre of attention.

With increasing extent of scheduled work, with increasing number of working points in the considered system and particularly with increasing extent of potential successive inspection work to be allowed for, the decontamination of entire technological systems will gain in competitiveness.

3. Chemical decontamination methods for technological systems

In a development process of several years the decontamination methods characterized in Table 1 have been tested. Now they are available, at sufficient reliability, for preparing, in terms of radiation-protection engineering, repetitive large-scale revisions or have already been introduced into routine application.

3.1. Decontamination of the primary circuit of the WWER-2 type

So far, the reactor facility of the WWER-2 type has been decontaminated ten times immediately after a campaign; the core has always been involved in the process. While, in the first years, a modified "AP-Citrox" method /1/ was the active principle, later a transition to an "AP-CE" method was developed with a mixture of citric acid and ethylenediaminetetraacetic acid (EDTA) being used as a pickling agent. It is characteristic that

- the agents used are nearly stoichiometrically related to the oxide amounts to be dissolved and contain only a slight excess. Assessed by chemical expenditure, this method lies already at the limit of so-called soft decontaminations /2/; there is no "refreshing" of the circulating solution by means of cation exchange;
- the transition from oxidation bath to pickling occurs without water exchange or interim flushing, which influences the economy of the process very positively;
- the chemical and activity drag-out from the primary circuit is managed without additional storage volumes, since, simultaneously with the flushing process, high-performance evaporator facilities process part of the decontamination solutions and make water circulation possible;
- the process is operatively controlled by temperature-time allowances along with continuous registration of values for redox potential (E_H), pH, electrolytic conductivity ($\varkappa$) and pulse rate (Σ);
- the decontamination of the primary circuit directly follows, in organization, a scheduled shutdown of the unit and does not call for the installation of additional technological equipment.

Experience with this method is reported in greater detail in /3/. We consider the finding essential that the structural materials of the core (nichrome steel, ZrNb1 and ZrNb2.5, UO_2) are sufficiently resistant to the mixture. Neither fuel rod integrity nor core function in the successive campaign are negatively influenced by using this method. In this connection some results shall be reported below which were obtained as a "by-product" of a large-scale experiment made in cooperation with the USSR to study the fission product leakage caused by defects of fuel element cladding.

In this experiment there were experimental assemblies in the WWER-2 core for three successive reactor campaigns, which contained artificial leaks (holes of 1 mm diameter) in three neighbouring fuel elements each. According to the usual classification of cladding leaks the case "contact of fuel - coolant" was thus simulated.

At the end of the respective campaign, these assemblies were included in primary circuit decontamination by the AP-CE method.

The results obtained show that

- due to primary circuit decontaminations fission product leakage in the successive campaign is not sharply increased;
- by decontamination measures the fission product and fuel amounts leaked out are largely removed.

The Figure attached shows an underwater photo of the leak in the experimental assembly in operation for the longest time, after three reactor campaigns and three primary circuit decontaminations, which gives a visual impression of the state of fuel elements.

3.2. Decontamination of separate cooling loops of the WWER-2 type

If revision work is concentrated on one cooling loop of the WWER-2 type, this can be separately decontaminated beforehand, since, due to design, there is a possibility of isolating the loops and a junction between the cold and the hot line. Then the following system parts are cleaned:

3 steam generators, main circulating pump, sections
of the main coolant loop, 2 standby isolating
valves.

Mixture and method parameters principally correspond to the AP-CE method described under 2.1. Depending on the required degree of cleaning, this can be simplified to a one-step process, or the technological stages of

oxidation - draining - pickling - draining - flushing - draining

follow each other in series.

For many scheduled outages, the decontamination of a system section is of course more advantageous, from economic aspects, than the maximum variant according to 2.1. Separate loop decontaminations were performed to prepare, e.g., repair work at steam generators /4/.

3.3. Decontamination of the primary circuit of the WWER-440 type

For periodic inspections of the facility and material state following an integral testing, volumetric test methods (ultrasonics, radiography) and methods for surface crack testing (electromagnetic, magnetic, dye penetrant testing, visual inspections) are used /5/. For the latter special demands are made concerning the surface purity of the concrete location of inspection; for all this work complex radiation protection measures are imperative. Therefore, in-service inspections of WWER-440 reactor pressure vessels are made by meane of a testing container which represents an alternative solution compared with underwater testing with respect to defect detection, personnel employment and possibility of treating defect points. As an additional radiation protection measure, chemical decontamination of large parts of the primary circuit is the first stage.

Depending on the concrete revision programme, primary circuit decontamination covers the following components:

- Reactor pressure vessel
- Reactor head
- Pressurizer
- 2 ... 6 complete coolant loops (the organizational and economic optimum lies at a decontamination extent of three coolant loops).

Here the "AP-CE" mixture comes into contact with a group of materials which, in some power plant units, also contains low-alloy carbon steels. Occurring corrosion effects are described in Table 2. The results of so far three in-service inspections of decontaminated WWER-440 units /5/ as well as extensive laboratory test series and results of companion samples confirm the integrity of structural materials.

3.4. Decontamination of other non-dismountable components of the WWER-440 type

The decontamination of one of the six horizontal steam generators of the WWER-440 reactor facility is performed after installing some technological auxiliary equipment /6/ which permits

- the isolation of the steam generator collectors from the main coolant loop,
- the filling of the steam generator tubes with decontamination solutions,
- a forced circulation of the decontamination solutions through the steam generator tubes,
- the draining of the used decontamination solutions for special water treatment,

Similar technologies are known from the nuclear power plants Novo Voronesh /7/, Kola /8/ and Koslodui /9/.

Technical auxiliaries have also been developed for the decontamination of pressurizers, permitting temporary isolation of this aggregate, filling with decontamination solution and draining. In applying the respective technology, boric acid of sufficient concentration is always added to the decontamination solutions and flushing waters to ensure the subcriticality of the core if the decontamination solution unintentionally penetrates into the reactor.

Other decontamination technologies and auxiliaries have been developed and utilized for

- casings of main circulating pumps
- casings of main isolating valves
- ion exchange filters.

4. Summary

The state of development reached in the GDR in methods for decontaminating technological systems as well as their importance for radiation protection during inspection and maintenance periods can be characterized as follows:

- In the nuclear power plants of the GDR, chemical decontamination of systems or system sections hold a definite position in maintenance strategy. They are economically effective particularly for large-scale revisions, since they facilitate and accelerate work. As a rule, they can reduce outages (increase in availability).
  Present experience, however, requires a limitation in the frequency of chemical decontamination within the designed service life of a system.

- The above-described decontamination technologies considerably reduce the dose rate level in the equipment treated and yield decontamination factors (DF) up to about 100 ($P_{\gamma + \beta}$, measured at the internal side of opened system). The observed scattering range reaching down to DF 2 is induced by numerous factors, e.g. flow conditions in the system and conditions of build-up of the crude layers. Existing residual contamination is mainly due to a thin mostly invisible deposited film, so that, if necessary, the decontamination effect can be further improved in a simple way (wiping off). In this way it could even be attained in special cases to reconstruct components of the primary circuit in specialized machine-building enterprises under conventional conditions.

- Large-scale decontamination has decisively contributed to the fact that the collective dose per unit and per operating year could so far be kept at a low level. Even in years when there was a considerable extent of inspection work in the controlled areas of the units, as a rule the yearly dose lies below the average values of US nuclear power plants equipped with pressurized-water reactors (cp. Table 3) /10/. So the experience gained with the WWER-2 type is that depending on the preceding method of primary circuit decontamination the collective dose for yearly reloading and revision outage (GI) reaches the following values:

Methods of primary circuit decontamination	Collective GI dose / mSv /
AP-Citrox	1650 (880 ... 2280)
AP-CE	430 (380 ... 660)

The range of values in parentheses is caused by the yearly changing extent of work and many other details, the higher effectiveness of the AP-CE mixture, however, can be taken for granted.

For WWER-440 facilities experience has shown that the first comprehensive in-service inspection of a unit will induce an application of 1100 ... 1500 mSv, if, for instance, primary circuit decontamination and test container are used as radiation protection measures.

5. References

/ 1/ R. Winkler et al.; Kernenergie 12 (1969) H. 11, S. 341 "Die Dekontamination des Primärkreislaufes im Kernkraftwerk Rheinsberg"

/ 2/ G.R. Choppin et al.; EPRI-NP-1033 (1979) "Literature review of dilute chemical decontaminations processes for water-cooled nuclear reactors"

/ 3/ K. Oertel et al.; Kernenergie 24 (1981) H. 5, in preparation "Dekontamination des kompletten Primärkreislaufes von Kernkraftwerken mit Druckwasserreaktoren"

/ 4 / R. Winkler, W. Brune; Kernenergie 19 (1976) H. 4, S. 101 "10 Jahre Kernkraftwerk Rheinsberg"

/ 5 / D. Pastor et al.; IAEA-CN-39/46 (1980) "Experience in In-Service Inspections of Reactor Pressure Vessels"

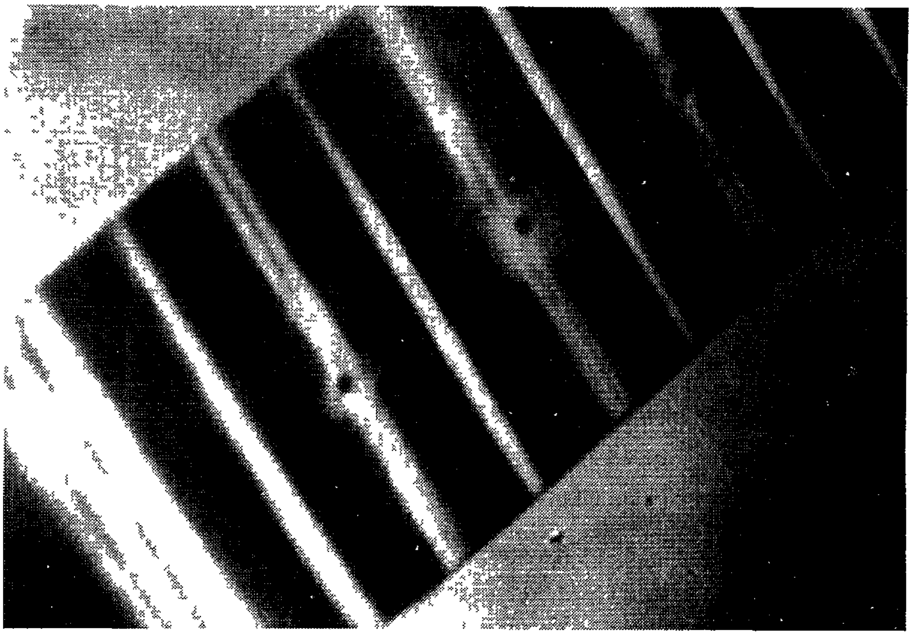
/ 6/ D. Legler et al.; DDR-WP 108843 (20.11.1976)
"Verfahren und Anordnung zur Dekontamination von im Primärkreislauf von Druckwasserreaktoren liegenden Dampferzeugern"

/ 7/ L.I. Golubjew; Atommaja Energija Bd. 44 (1978) H. 5, S. 438
"Dezaktivatsia parogeneratorov Novovoronezhskoi AES"

/ 8/ Ju.W. Balaban-Irmenin et al.; Cottbus, DDR, 20.-23.11.1979
- Sbornik naucno-techniceskich dokladow -
"Sovescanie specialistow po podgotovke trebovanij dlja proektirovanija tipowogo ucastka dezaktiwacii AES s serijnymi reaktorami"

/ 9/ G. Gentschev; loc. cit., S. 156

/10/ T.D. Murphy; Atomkernenergie 30 (1977) 4, S. 293
"An overview of the activated corrosion product reduction programme of US power reactors"



Underwater photo of a working assembly with 3 defined leaks after 3 campaigns and after 3 decontaminations of the WWER-2 type

Table 1: Characteristic parameters of the chemical decontamination methods used

Reactor type	Decontamination object	Activity removed /GBq/			Contamination duration [a]	Mixture	Process time [h]	Waste [m^3]
		Σ	Co-58	Co-60				
WWER-2	Primary circuit with core	3.700 - 39.600	100 - 2.520	20 - 5.280	1 - 2	AP-CE1)	35	15
WWER-2	Cooling loop	740 - 1.110	20 - 110	10 - 190	1 - 3	AP-CE1)	24	3
WWER-440	Primary circuit (2-3 loops) without core	22.220 - 40.700	2.500 - 5.700	1.070 - 1.620	4 - 5	AP-CE	60	20
WWER-440	Steam generator	740 - 1.110	20 - 320	10 - 150	1 - 3	AP-Ox	35	10
WWER-440	Pressurizer	40	3	2	1 - 3	AP-CE	30	15

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1) earlier AP-Citrox /1/

Table 2: Corrosion of structural materials at the AP-CE decontamination method

Material	Corrosion loss [µm/decontamination]
Carbon steel 48TC, 22K, Cm20	71
Chrome steel 3X18, 2X17N2	2
Al-Fe bronze	1.5
Austenitic nichrome steel	0.05
ZrNb1, ZrNb2.5	0.1

Table 3: Average values of occupational radiation exposure in US nuclear power plants equipped with PWR facilities according to /10/

Calendar year	Number of pressurized-water reactors	Yearly dose [mSv/unit]
1969	4	1650
1970	5	5990
1971	6	3400
1972	8	4630
1973	12	7720
1974	18	2640
1975	26	3090
1976x)	31	4550

x) data incomplete

EXPERIENCES WITH DECONTAMINATION METHODS AT THE NUCLEAR POWER PLANT MÜHLEBERG, SWITZERLAND

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Introduction

There are various methods available for the on-site decontamination of removable equipment in Switzerland:

- a shot peening glass beads method, using a mixture of demineralized water and glass beads within a closed box

- high pressure water jet method

- wet cleaning methods, using washing basins of different dimensions, detergent and brushes.

The manuel cleaning is limited to a few cases, where the special form or the dimensions of the equipment do not allow the application of the glass bead method.
Examples of the first two methods are discussed here. These methods have been in use at the nuclear power plant Mühleberg, which is a General Electric Mk I boiling water reactor of 306 MWe (net) power and which has been in commercial operation since October 1972.

Decontamination by Shot-Peening with Glass Beads

Shot peening decontamination is a preferred method for the following reasons:

- wide applicability for contaminated and inactive materials
- quick effectiveness of cleaning
- non abrasive
- low quantities of secondary waste
- no danger of incorporation

The method can be used either to remove loose contamination and reduce the dose rate to safe manrem during maintenance and repair, or to achieve such low contamination levels that the material can be disposed of as inactive.

The requirements for sucessful treatment are

- extended planar surfaces
- angles of impact greater than 45°
- no precision parts with complicated forms
- limitations on weight and dimensions

Description of the shot-peening installation

The main part of this installation consists of a closed box with windows. Inside this box the contaminated object lays on a turnable disc. The mixture of glass beads and water is injected through a pistol type nozzle (s. Fig. 1), collected by a sump and recirculated by a pump. The operator holds this nozzle in his hand, protected by rubber gloves and directs the outcoming mixture of water and glass onto the rotating object inside the box.

Surveillance during the whole decontamination process is guaranteed by the windows. The pressure of the water/glass mixture is about 60 kg/cm^2. The dimensions of the box, in this case approx. 1,2 m x 1,2 m x 1,5 m (wide x depth x height), limit the maximum size of treatable objects. The maximum weight of one piece is limited to 1 t.

Experiences with the shot peening glass beads installation

This installation has been in operation since 1975. During this time about 1500 parts with a total weight of approx. 6 t have been sucessfully treated. The annual working hours are of the order of 500, half of this time is used up during the shut down period. The efficiency can be summarised as follow:

reduction of dose rate:	100 - 500	times for plane surfaces
	10 - 20	for more complicated forms

Example 1: plane surfaces

Fuel storage racks showed an average dose rate of 700 mR/h due to surface contamination. After cleaning twice with high pressure water at 600 kg/cm^2, a remaining dose rate between 30 - 50 mR/h was measured. After additional cleaning by the shot peening glass bead method (several steps) the dose rate was 20 - 30 μR/h. The remaining contamination was detected on a piece with a surface of 6 cm^2 and gave a result of $6{,}3 \cdot 10^{-3}$ μCi/cm^2.

Example 2: complicated form

The impeller of the reactorwater clean-up pump, which is a complicated form, showed a dose rate of 12 R/h after removing from the casing. This dose rate has been reduced to a level of about 0,7 R/h by cleaning with glass beads.
Other pieces have been sucessfully treated, for example: nuts and bolts, valves, parts of pumps, flanges, parts of control rod drives and trubine inlet valves.
The sucessfull application of this method to decontaminate pieces to below active levels depends on the established limits in each country. In such cases where these limits cannot be reached a more abrasive treatement is possible by using sand instead of glass beads.

Treatment of decontamination wastes from shot peening

The annual production of glass beads is in the order of 200 - 300 l and can easily be solidified with cement after separation from the contaminated process water in the coarse filter (s. Fig. 1).This processwater gives together with flushing water, a volume of 50 - 80 m^3/a and is sent to the radwaste system, where it is treated by powdex resin.

Doses to personnel

The accumulated doses to the personnel for decontamination work with this shot peening glass beads installation is approx. 1,5 manrem/a. Without this treatment, before maintenance or repair, the doses to personnel would be at least 10 times higher.

Decontamination by high pressure water jet

Where large surfaces need to be decontaminated high pressure water jets have been found to be efficient and easy to use. For example, the Mühleberg Boiling Water Reactor, a GE/Mark I, has a suppression chamber in the form of a torus (4000 m^3) which is partially filled with approximately 2'000 m^3 of water.

The protective painting on the inner surface of this torus consists of a Zincsilicate-type, which has partly dissolved
A cleaning of the whole surface by high pressure water jet where it was in contact with the suppresion pool water, was necessary to reduce the dose rate before repainting. The radioactivity before decontamination was measured as:

50 - 100	standard values*	below water level
5 - 10	standard values	above water level

The dose rate within this tours, which was caused by 75 % Co-60 and 25 % Cs-137, was between 5-10 mR/h before decontamination.
A high water pressure jet was used with a pressure of 600 kg/cm^2 and at ambient temperature. During 12 days a total amount of approx. 300 m^3 water (250 m^3 demineralized water + 50 m^3 tap water) was sprayed at the surfaces. The final contamination was measured as follow:

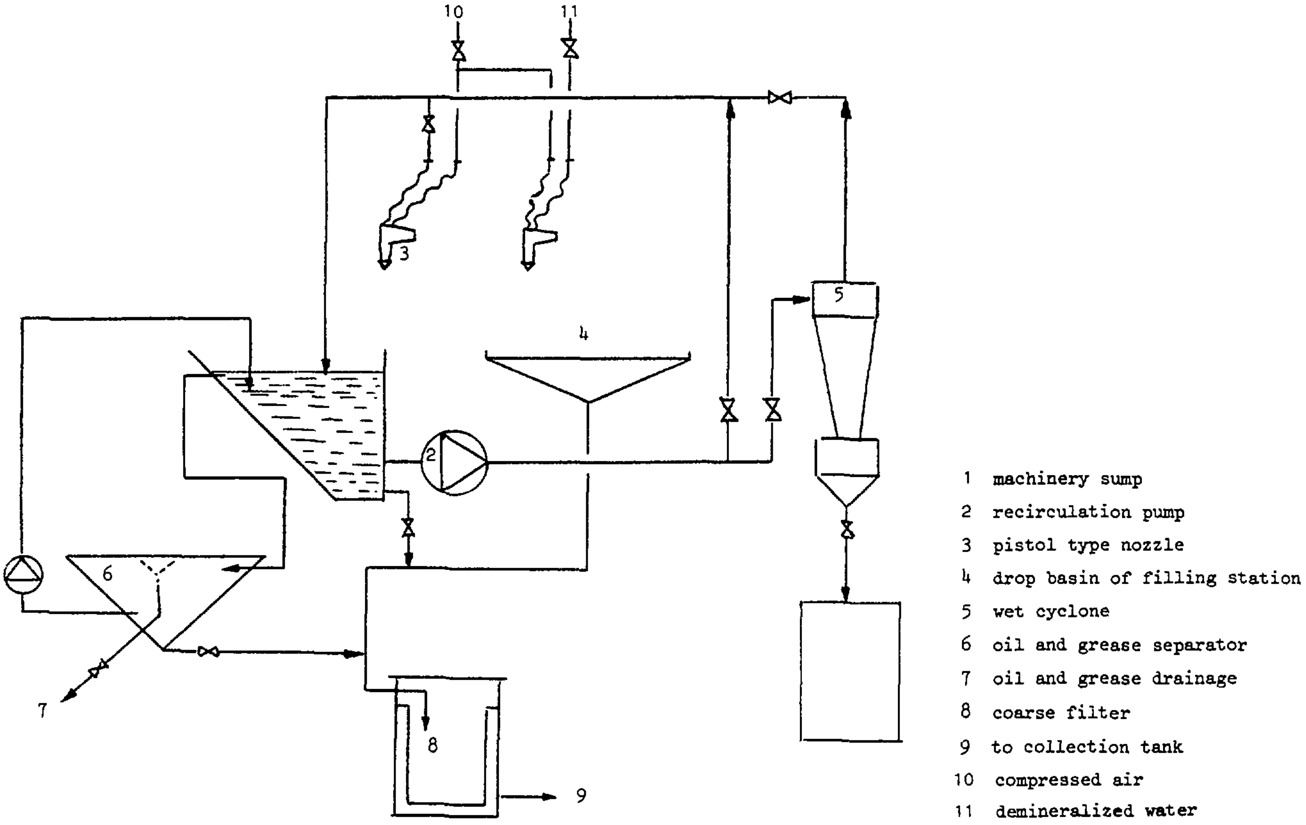
1 - 3 standard values* above and below water level

giving a dose rate of 2 mR/h. This value is only a guide to the effectivness of decontamination, because part of this dose rate comes from other pipes and pumps close to the suppresion chamber.
A new protective painting (tar-epoxy-type) was applied with a minimal thickness of 0,45 mm. 991 man-hours were required for decontamination and repainting and a total man-rem of 5,9 was accumulated.

* s. page 7

FIGURE 1

Basic flow diagramm of a shot peening glass bead installation



Conclusions

Shot peening with glass beads is the preferred decontamination method in Switzerland for most removable contaminated equipment. The application of this method has given good resultats, not only for reducing dose rates of contaminated equipment before repair or maintenance, but also for decontaminating metal wastes below or near active levels. Similar installations are available in all other nuclear power plans in Switzerland.

The high pressure water jet is the preferred decontamination method for large surfaces in nonremovable components or can be applied as pretreatment to the shot peening method.

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* 1 standard value is equal to 10^{-2} µCi/100 cm^2 according to the Swiss Regulation on radiation protection, dated 30 june 1976.

AN OVERVIEW OF THE CANADIAN DECONTAMINATION PROGRAM

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The main thrust of the Canadian program for the decontamination of power reactor systems and components started in 1970. It has been coordinated by the Decontamination Task Force which is comprised of members from the Public Utilities and AECL, the federal agency. The Task Force reports to the Activity Transport Working Party (ATWP). This latter group is also comprised of members from the Utilities and AECL.

The objectives of the ATWP were to develop an understanding of radiation field growth around nuclear heat transport systems and use this understanding to make improvements in operating reactors and future reactors. The objectives of the Decontamination Task Force are:

(1) to understand decontamination processes;

(2) to develop decontamination procedures;

(3) to feed back information to reactor designers.

Throughout the work, AECL staff have led and directed the efforts of both the ATWP and Decontamination Task Force. While in the early years, the main contributors were from AECL and Ontario Hydro, the program has been, and remains, the national program. The present work groups comprise personnel from AECL, Ontario Hydro, Hydro Quebec, New Brunswick Power Company, and the consultants, London Nuclear Decontamination Limited.

1. Decontamination Experience

Hundreds of man-years have been expended to develop the techniques we now use in CANDU heavy water reactors. This has involved extensive development and testing of reagents for decontamination purposes, examination of the effects of the decontaminating reagents on all system components, and this work culminated in the successful application of the CAN-DECON* process in the power reactors:

NPD
Gentilly-1
Douglas Point

This is a process in which:

- the reactor is shut down, but is neither defuelled nor drained;
- a small amount of a mixture of weak acids and chelating agents, the decontamination reagent, is added directly to the heat transport fluid, typically to give a concentration of 0.1 wt %;
- the heat transport fluid is circulated through the reactor, its components, and the heat transport fluid cleanup system;

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*CAN-DECON - CANDU Decontamination

- the reagent attacks the contaminated oxide film and "crud" deposits, and releases to the heat transport fluid both particulate and dissolved material;
- sub-micron filters are used to remove the particulate material;
- cation-exchange resin is used to remove dissolved metal ions from the heat transport fluid and to regenerate the reagent;
- the process is continued as long as contaminants are being removed or until the allowed time has elapsed;
- a mixed bed of cation and anion exchange resins is used to remove the reagent and any remaining dissolved metals.

Other types of decontaminations carried out in Ontario Hydro's stations are given in Table 1.

2. Future Plans

The future plans of the Decontamination Task Force are:

(a) Decontaminate the complete heat transport circuit of Douglas Point NGS. This will be completed either in September 1979 or September 1980.

(b) Be ready to decontaminate the complete heat transport circuits at Pickering. Ready by spring 1980.

(c) Design, then construct portable cleaning equipment for the decontamination of components, such as:

- heat exchangers
- pump impellers
- single steam generators
- steam generator heads prior to inspection

(d) Assist the consultant, London Nuclear Decontamination Limited, in their efforts to apply the CAN-DECON process and Canadian expertise to other reactor types.

The remainder of the program is highlighted in the following tables:

Table 2	-	The Testing Program Required to Qualify a Decontamination
Table 2(a)	-	Component Evaluation
Table 3	-	The Cycling Decontamination Techniques
Table 3(a)	-	Their Advantages/Disadvantages
Table 4	-	The CAN-DECON Process
Table 4(a)	-	The advantages of CAN-DECON
Table 4(b)	-	Its use in power reactors
Table 4(c)	-	Cost of CAN-DECON versus hard decontamination techniques
Table 4(d)	-	Effectiveness of CAN-DECON at Douglas Point NGS.

Table 1

DECONTAMINATION EXPERIENCE

in Ontario Hydro

Station	Component	Technique
Type: Area		
Pickering	Fuelling Machine	Coating
Pickering	Fuel Transfer Elevator	Mechanical
Type: Component		
NPD	Fuelling Machines	Chemical
NPD	Heat Exchanger	Chemical
Douglas Point	Bleed Cooler	Chemical, Mechanical
Douglas Point	Main Pumps	Chemical
Douglas Point	Pump Bowl	Chemical
Pickering	Fuelling Machine	Chemical
Pickering	Main Pumps	Chemical
Bruce	Bleed Cooler	Chemical
Type: Whole System		
NPD	Heat Transport	Can-Decon
Douglas Point	Heat Transport	Redox
Douglas Point	Heat Transport	Can-Decon

Table 2

TESTING PROGRAM REQUIREMENTS

Selection of Reagents should include study of effects on decontamination factor* and materials corrosion of:

- Reagent Concentration
- Flow
- Temperature
- Components Evaluation
- Fuel Failure

*Decontamination Factor (DF) = $\frac{\text{radiation field before decontamination}}{\text{radiation field after decontamination}}$

Table 2(a)

COMPONENT EVALUATION

Valves
  - packing

Grayloc

Gaskets

Pump Components

CANDU
  - End closures
  - Rolled joints

Plus all components in the heat transport circuit

Table 3

CYCLING TECHNIQUES

Temperature - lowering and raising the coolant temperature

Hydraulic - equipment manipulations that change the water velocity and pressure

Redox - changing from reducing conditions with excess hydrogen to oxidizing conditions by the addition of oxygen and back again

These techniques can be used by the reactor operator during any shutdown period.

Table 3(a)

CYCLING TECHNIQUES

Advantages	Disadvantages
Applicable in D_2O	Decontaminates Monel and Carbon Steel, provided Fe_3O_4 is not stabilized by $Ni(Fe_2O_4)$
No downgrading	
30 hours downtime	
Less than 1 rem	
Solid waste only	
No corrosion	

Table 4

CAN-DECON

In the present decontamination procedure, about 0.1% of the mixed organic acids are added to the coolant. The acids dissolve the oxide films and the embedded radionuclides from the metal surfaces in the heat transport system. The chelated metals are then transported by the coolant to ion-exchange resins in the purification system where the metals are removed and the organic acids are regenerated for further decontamination. The crud produced is removed by filters. The process of using dilute reagents and regeneration by ion exchange is called the Can-Decon process.

Table 4(a)

CAN-DECON

The advantages of using dilute reagents and the regenerative process are:

- The system need never be drained; as a consequence there is little or no downgrading of the coolant.
- Very low concentrations of decontaminants are used (about 1 g/kg) which reduces downgrading.
- Because the decontaminants are regenerated, the process can be continued as long as activity is still being removed.
- All the wastes are concentrated on ion-exchange resins, thus greatly simplifying disposal.
- Because the reagents are dilute, corrosion is slight.

Table 4(b)

CAN-DECON

The feasibility of the Can-Decon process has been successfuly demonstrated during the decontamination of:

- the whole heat transport system at Gentilly-1,
- the whole heat transport system at NPD,
- the bleed system at Douglas Point,
- the whole heat transport system at Douglas Point.

Table 4(c)

CAN-DECON

Costs for Power Reactors

	DPNGS (200 MWe)	PNGS (540 MWe)
Equipment	75k$	900k$
O&M	165k$	350k$
TOTAL	240k$	1 250k$

Disposal Cost

	DPNGS	PNGS
Can-Decon	80k$	125k$ *
Citrox	500k$	1 500k$ **

* Cost of disposal for CAN-DECON included in O&M (Operation and Maintenance) cost above

** Cost for disposal of volume of waste
- Does not include equipment for solidification, or equipment to do a Citrox (hard) decontamination

Table 4(d)

Effects of CAN-Decon at Douglas Point NGS

Radiation Field Reductions

Location	Before Decontamination (mR/h)	After Decontamination (mR/h)	Average DF
Outside Feeder Cabinet Doors	2 000 - 3 000	300 - 500	6.2
East Reactor Vault	1 900 - 7 900	650 - 850	4.5
West Reactor Vault	1 600 - 5 800	420 - 970	4.3
Lower Primary Pump Area (Affected by Fields from Monel Boilers)	800 - 4 000	500 - 1 400	1.8

Bibliography

1. "Decontamination of CANDU Primary Coolant Systems," P.J. Pettit, AECL-5113, 1974.

2. "Techniques to Reduce Radiation Fields," B. Montford, AECL-5523, 1976.

3. "Activity Transport in CANDU's," M. Tomlinson, Editor, AECL-5113, 1974.

4. "Decontamination by Cycling Techniques at the Douglas Point Nuclear Generating Station," B. Montford, Proc., American Power Conference, 1973.

5. "Decontamination of a Fuelling Machine Contaminated by Irradiated Fuel Debris," C.S. Lacy, Nace, Corrosion, 1978.

6. "Decontamination Experience at CANDU-PHW Reactors," C.S. Lacy, W.B. Stewart, A.B. Mitchell, BNES Conference, Bournemouth, England, October 1977.

7. "CAN-DECON, A Nuclear Reactor Decontamination Process," London Nuclear Decontamination Limited, Niagara Falls, Ontario, Canada, L2E 6V9.

CONSIDERATIONS ON THE DECONTAMINATION OF A REPROCESSING PLANT

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1. GENERAL REMARKS

As for nuclear power plants, decontamination of a reprocessing plant is an essential means for the safe operation of such a plant, too.

Partial or total decontamination of plant circuits, equipment or operational areas is executed in view of reducing the radiation level and thus the dose commitment for the operation personnel. This is both true for the operational period and the shut-down and decommissioning phase. Efficient operation of a plant depends on a smooth maintenance, that in turn asks for appropriate decontamination methods. It is thus, that also the decommissioning stage of a plant can be considered as the final part of the operation of the reprocessing plant where principally the same techniques are applied that have been utilized during the plant operation.

In addition to the needs of decontamination for plant maintenance, decontamination of plant circuits is sometimes required, too, for the restoration of the chemical and radiochemical specifications in the final uranium- and plutonium products, serving as basic material for the fabrication of new fuel elements.

The postulation that the best decontamination procedure is to avoid contamination is obviously also true for a reprocessing plant. Appropriate measures should already be taken at the design stage and operational conditions should aim at minimizing the risks for and the levels of contamination.

Decontamination techniques have to consider not only the nature of the contamination but also the compatibility of the technique with the postulations for the safe operation of the plant be it either in view of resistance to corrosion, or rate of recontamination or purity of the final products. Apart from this, the aspect of the waste resulting from decontamination plays an important role, too, both as far as its volume and its chemical nature are concerned. Existing waste treatment facilities should be capable of taking care of the particular decontamination wastes, too.

2. CONTAMINATION

Contaminations encountered in a reprocessing plant differ fundamentally from those found in power reactors. This is both true for the radionuclides and for the chemical nature of the contaminants.

In reprocessing plants the contaminating nuclides are essentially fission products together with uranium and plutonium, whereas activation products do practically not play any role at all.

Contamination is characterized by the chemical nature of the process streams circulated in the plant. These are mainly :

- nitrates of the dissolved fuel, i.e. uranium, plutonium and fission products contained in nitric acid,

- organic solvents like dodecane, containing tributylephospate as extractant for the extraction of uranium and plutonium, and

- alcaline solutions like sodiumhydroxide or sodiumcarbonate for off-gas and solvent purification.

Due to the chemical nature and the rather low process temperatures prevailing in a reprocessing plant (tmax. ~ 100°C), the basic physicochemical processes leading to contamination are mainly adsorption, ion-exchange and precipitation.

The particular "crud" formation found in power reactors does not exist in reprocessing plants. There is, however, a similar formation of solid residues, that could be compaired with this "crud" especially as far as its radiation level and chemical resistance are concerned. This rather annoying solids originate mainly from the chemical and radiolytical degradation of the extractant and the solvent, forming phosphate-rich heavy phases saturated both with plutonium and particular fission products as zirconium, ruthenium, strontium and cesium. These heavy phases are both insoluble in solvent and aqueous solutions, they easily adhere and stick to surfaces and often are responsible for the blockage of narrow pipe lines.

Apart from this "crud" resulting from solvent degradation, undissolved fuel powder, containing metal dust from the fuel element hulls (originating during chopping the fuel) and some of the original crud deposits on the surface of the fuel elements, originates in the head-end part of a fuel reprocessing plant and tends to form another kind of contaminating "crud" in the subsequent extraction steps.

Selection of appropriate plant operation conditions, as for instance an efficient filtration of the extraction feed to separate the latter solids and an as efficacious as possible elimination of the organic degradation products are the best remedy against excessive contamination by these "cruds".

In addition to the interior contamination of process equipment just mentioned, surface contamination in process cells, intervention and working areas can happen due to leaking equipment, spills and contamination transfer. Again prudent operation of the plant, and extremely careful execution of all works bearing a potential contamination risk, are the best countermeasure against excessive contamination.

3. DECONTAMINATION

As already stated, decontamination procedures should aim at an optimum reduction of radiation levels without deviating the successive operational safety and without creating additional secondary problems for instance in the waste management.

One of the essential presuppositions that have to be met in decontamination is to avoid any excessive general and/or local corrosion. Any depassivation of the construction material has in this context to be overcome by a special passivation prior to starting again with the normal operation. Compatibility of the construction material with the decontamination procedure should be optimal, i.e. the material behaviour should exactly be known. Protection against corrosion is not only essential for the operational phase of the plant, it should be maintained during the decontamination, preceeding decommissioning, too, particularily in view of avoiding secondary contamination of process cells resulting from leaking corroded equipment.

It follows from this postulation, that mild decontamination chemicals and techniques should preferentially be applied. As far as internal process equipment surfaces are concerned decontamination solutionsshould be equal or similar to those utilized during the normal operational process : changes in concentration, consecutive changes in chemistry (acidic, caustic-attack, and oxidizing-, reducing action), intensive agitation of the liquids to assure efficient renewal of liquid films at the surface, increased temperature and prolonged residence time are parameters that play an important role when executing decontamination. It is obvious that each procedure has to take care of the chemical nature of the contamination and has to be adopted according to the needs.

External contaminations should be eliminated as fast as possible after occurrence. Here the commonly known techniques of washing and rinsing, assisted by application of high pressure and/or decontamination chemicals and mechanical brushing are successfully applied.

Again consideration of the chemical nature of the contaminant and the characteristics of the contaminated surfaces will help in selecting or adopting an appropriate procedure.

Whenever decontamination does not lead to the wanted substantial reduction in contamination and radiation, more corrosive chemicals may exceptionally be applied under extremely controlled conditions and followed by additional protective measures.
Finally, if the aim cannot be achieved, dismantling and substition of the contaminated equipment might be the last solution. Resistant external surface contaminations can be immobilized and/or shielded by application of protective paints or other surface layers.

Apart from the in situ decontamination of process equipment, preferentially grouped into separate loops, decontamination of small equipment, like pumps, motors, etc... is executed after dismounting in special decontamination shops utilizing different chemical baths, ultrasonic techniques and all kinds of spraying and blasting procedures.

3.1 Decontamination experience at Eurochemic

After the termination of its reprocessing activities, treating more than 200 tons of irradiated fuel with 90 million curies of fission products, the Eurochemic reprocessing plant was decontaminated in view of allowing either

- the refurbishing and adaptation of the plant for a new start-up in the future, or

- the complete decommissioning of the plant.

To meet this aim, mild decontamination techniques had to be applied and use was made of the experience gained in equipment decontamination during 8 years plant operation.

From the rather detailed description of the various works executed at Eurochemic during the first decontamination and partial dismantling campaigns in [1, 2] only some of the results are mentioned hereafter.

In a first rinsing operation - aimed at a recuperation of fissile material - utilizing mainly diluted and concentrated nitric acid containing sometimes reducing additives and applying a down-stream routing (high-active to low active process equipment), 1300 g plutonium and 250 Kg uranium were recovered.

In the second step, chemical decontamination was applied by alternating chemical attack (acid-caustic, oxidizing-reducing), routing upstream (low active to high active process equipment) solutions like diluted and exceptionally concentrated nitric acid and sodium hydroxide, 0.1 molar potassium permanganate in 5 or 8 molar nitric acid, 0.1 molar oxalic acid 0.5 molar nitric acid, 0.5 molar hydrazinenitrate, 10 wt-% sodiumtartrate- and 10 wt-% sodiumcitrate-solutions. As already indicated, good agitation and whenever possible heating of the solutions was applied. A detailed analytical programme allowed a realistic follow-up of the rinsing and decontamination operations.

In total 370 m^3 rinsing and decontamination solutions were produced containing 140 000 Ci of fission products, 390 Kg uranium and roughly 3900 g plutonium. Part of the plutonium and uranium were recuperated and the totality of the solutions was concentrated by evaporation in the medium level waste evaporator of Eurochemic into about 70 m^3 liquid waste concentrate that were pumped into the tanks of the medium level waste tank farm awaiting solidification in the bituminization plant.

At the end of these rinsing and decontamination operations all process cells were accessible. Dose rates of up to several 10 million mrad/h came down to average dose rates of 10 to 100 mrad/h.

Equipment showing still hot spots was afterwards decontaminated by high pressure water jetting through existing spray ramps and by introducing special spray nozzles.

Water pressure utilized varied between 200 and 400 bar.
Whenever good access to the contaminated surface was possible, decontamination factors of up to 1000 were obtained. The procedure is particularly effective in eliminating contaminations of the above "crud" type.

Blocked tubes and vessel connections were opened and decontaminated by high pressure water jetting or other means of pressurizing, too.

A very small amount of equipment that could not be sufficiently decontaminated or that would notbe reused in case of a new start-up was dismounted, cut up and conditioned as waste.

Decontamination of external equipment surfaces, process cells and intervention areas was executed by application of :

- high pressure water jets (250 - 400 bar)
- intermediate pressure water jets ($\leq$ 80 bar) allowing addition of decontamination chemicals and heating of the solution, and

- mechanical decontamination by brushing with various decontamination agents.

In general the high pressure water jetting technique has given full satisfaction both with respect to efficiency and waste produced.
At a jetting rate of 15 to 25 m^3/h and a specific water consumption of 55 to 60 l/m^2 surface decontaminations to MPC levels have been achieved. The procedure has at the same time the advantage that it leads to practically salt free waste solutions that can easily be concentrated into negligible amounts of liquid waste concentrates.

Very efficient decontamination could also be obtained by spraying a 0.1 % citric acid solution at pressures between 60 and 100 bar on contaminated surfaces.

Whenever both procedures fail, or leave residual spots of resisting contamination, mechanical techniques have successfully been applied.

Of particular interest to power reactor stations is in this case the fact, that all the water ponds of Eurochemic, utilized for the reception, intermediate storage and the mechanical pretreatment of irradiated fuel and representing a total volume of about 2500 m^3, have been emptied and decontaminated.

It is last not least worthwile to mention, that the total analytical process control laboratory has been decontaminated. All analytical boxes of a shielded, an unshielded and an α-box chain have been dismantled, decontaminated and conserved for an eventual reuse.

4. WASTE TREATMENT

As already mentioned selection of the decontamination technique should consider existing waste treatment facilities and waste conditioning procedures. This is not only true for volume arisings but also for the chemical nature of the produced wastes.
Some of the chemicals contained in the waste might for instance interfere with the normal waste treatment procedure by either spoiling the decontamination factors of a chemical precipitation process or by exothermic decomposition during a bituminization process, or by retarding the hardening of a concreting process. Although appropriate chemical pretreatment procedures can be conceived to overcome these difficulties, care must be taken with respect to the possibility of achieving such pretreatment in an existing waste facility.
In some cases it might be more advantageous to select another chemical compound for the decontamination.

The waste treatment facilities of Eurochemic, that have to take care of the liquid wastes resulting from decontamination and that are rather representative for reprocessing plants, consist essentially of an evaporator, a tank form for the intermediate storage of the medium level waste concentrates produced in the evaporator and a bituminization plant, where the waste concentrates are solidified by incorporating the solid residues into bitumen which is afterwards stored in a subsurface engineered storage bunker.

The bituminization facility is equipped with a chemical pretreatment station, where for instance neutralization, precipitation and ammonia boil-out can be achieved.
In order to avoid any deviation from specified operation conditions and final product characteristics, detailed analytical investigations are performed during the waste solidification campaigns.

In addition to the chemical pretreatment prior to solidification, direct measures can be and have been applied during the concentration step in the evaporator, by for instance appropriate waste partitioning in order to neutralize excessive chemical activity through neutralization, oxidation, reduction or decomplexation.

5. CONCLUSIONS

Although fundamental differences prevail in the radiochemical and chemical nature of contaminations in a power reactor and a reprocessing plant, the fundamental approach towards decontamination follows essentially the same lines for both plants :

- the best decontamination is to avoid contamination,
- first measures against contamination should already start at the design stage of the plant, comprising for instance, care for smooth surfaces, avoid dead zones, limit welds as much as possible in down stream parts, foresee possibilities for internal and external water jets, provide appropriate surface coatings, etc...,
- select appropriate operation conditions to reduce formation of contaminating cruds (filtration, elimination of solvent residues and degradation products, keep solutions agitated, rinse systems from time to time, avoid heels, etc...),
- adopt your decontamination procedure to the needs of your facility with respect to :
    - corrosion behaviour,
    - dose commitments,
    - operational safety and
    - compatibility with the waste treatment facilities.

Efficient decontamination procedures have been developed and applied in various reprocessing plants in different countries. Experience gained at Eurochemic can in this respect be considered as a valuable contribution that certainly can be of help for the operation of nuclear power reactors, too.

6. REFERENCES

[1] J. Broothaerts, E. Detilleux, L. Geens, W. Hild, "Experience gained with the Decontamination of a Shut-Down Reprocessing Plant", IAEA-SM-234/39, November 1979.

[2] J. Broothaerts et al, "Industrial Experience gained in the Decontamination of Process Cells, the Dismantling of Process Equipment and the Conditioning of Special Solid Wastes in a Shut-Down Reprocessing Plant", IAEA-SM-234/40, November 1979.

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