DECONTAMINATION AND RECYCLING OF REACTOR COMPONENTS

M. Laser
Decontamination Department
Kernforschungsanlage Juelich GmbH
D 5170 Juelich/Fed. Rep. Germany

ABSTRACT

During the last 10 years about 5000 Mg of contaminated reactor components have been exchanged in the course of backfitting and repair work in the Federal Republic of Germany. The contamination is in the range between about 0.01 and 100 Sv/h. Furthermore three power reactor stations and one nuclear ship have been shut down. During the dismanteling again several thousand Mg of contaminated metallic components will be removed. Most of this material can be decontaminated and recycled by melting in a foundry. In the Federal Republic the limits for unrestricted release are surface contamination below 0.37 Bq/cm^2 and a specific activity of 3.7 Bq/g, assuming Co-60 and Cs-137 are the main contaminants. Legal problems arise because material with a specific activity up to 74 Bq/g can be handled without special license however the maximum specific activity for consumer goods is 0.37 Bq/g. In the decontamination shop of Kernforschungsanlage Juelich about 260 Mg of contaminated reactor components, have been decontaminated below the limits for unrestricted release in 1983 and 1984.

INTRODUCTION

During the last 10 years about 5000 Mg of contaminated reactor components have been exchanged in the course of backfitting and repair work in the Federal Republic of Germany. Furthermore 3 power reactors have been shut down. Some research reactors will be decommissioned in the near future. Finally the nuclear ship OTTO HAHN has been taken out of operation and all radioactive components have been removed and as far as possible decontaminated.

A very high percentage of the metallic material is contaminated and can be melted for reuse generally after decontamination. So the volume of radioactive waste is minimized and valuable metal is recycled. To prevent noticeable radiation exposures to the people special limits for contamination and activation of the material must be observed.

Experience has shown that about 85 % of the contaminated metal can be decontaminated in an economic way below the limits for unrestricted release.

LEGAL SITUATION

The German Atomic Law demands that removed components of a nuclear facility should be utilized in a harmless way or must be disposed of as radioactive waste if reuse is not possible or not acceptable from the economic point of view. But recycling is given priority to waste disposal[1].

However there are no definite regulations about conditions of unrestricted release of decontaminated metal. Therefore common regulations, e.g. on surface contamination, and regulations for unrestricted release of waste must be considered.

Principally there is an important difference between reuse of material and waste disposal.

- Waste resulting from handling radioactive isotopes in a license operation must be disposed of as radioactive waste unless another mode of disposal is allowed. A license for disposal can be given, if the activity is insignificant so that no special protection of life, health or objects is necessary [1]. The Federal Ministry of the Interior has recommended to give a license for conventional disposal, if the specific activity of the waste is lower than the 10^{-4} of the activity, which can be normally handled without special license referred to one gram of that waste[2]. The limit for license-free handling is given in the Radiation Protection Order[3]. Examples for the limits for conventional disposal are the following:

- H-3, U-nat	370	Bq/g = $1 \cdot 10^{-8}$	Ci/g
- C-14, Fe-55, Ni-63	37	Bq/g = $1 \cdot 10^{-9}$	Ci/g
- Co-60, Cs-137, Th-nat	3.7	Bq/g = $1 \cdot 10^{-10}$	Ci/g
- Ra-226, Pu-239	0.37	Bq/g = $1 \cdot 10^{-11}$	Ci/g

  Special conditions like the total amount of waste must be considered. Therefore deviations to higher or lower limits are possible.
- For the release of objects for use outside of controlled areas no special license is necessary, if the following limits of surface contamination
  0.037 Bq/cm^2 (= 10^{-6} Ci/cm^2) for high toxicity α-emitters
  0.37 Bq/cm^2 (10^{-5} Ci/cm^2) for other nuclides
  are not exceeded[3].

For the handling of material and objects outside of controlled areas furthermore following limits must be observed.

- For the licence free handling of radioactive material the specific activity must be lower than 74 Bq/g (= 2 nCi/g). (See Ref. 3.)

- For the production of consumer goods however the specific activity limit is 0.37 Bq/g (= 10 pCi/g) (See Ref. 4.)

In practice the licensing authorities also control the release of decontaminated material by special conditions to the operational license. In most

cases analogous to the license free handling of waste

- surface contamination must be lower than 0.037 Bq/cm^2 for high toxicity α-emitters and 0.37 Bq/cm^2 for other nuclides, and
- the specific activity must be lower than the 10^{-4}th of the license free activity per gram.

Furthermore in some cases the total weight of each charge is limited.

It can be shown that normally the surface contamination limit is most stringent. For example, a 1.25 mm thick steel plate contaminated with 0.37 Bq/cm^2 will contain 0.37 Bq/g after uniform distribution in the matrix by melting and therefore can be used for the production of consumer goods. Only thinner metal sheets will result in a higher specific activity.

At components with complicated shape like valves or tubes with a small diameter the surface contamination at the inner surface cannot be measured exactly. Therefore it is questionable if this material should be released for unrestricted melting. Up to the present time most of the licensing authorities did not allow the melting without a nuclear licence to the foundry.

If the specific activity is between 0.37 Bq/g and 74 Bq/g, formally the material may be handeled without a license, but the use for consumer goods is not allowed. In this case steel can be used for reinforced concrete, rolls etc. To guarantee this restricted use, the licensing authority must supervise the melting. If the above cited limits, especially the specific activity of 74 Bq/g, are exceeded, the melting furnace is a nuclear installation demanding a nuclear licence.

PUBLIC ACCEPTANCE

Recycling of decontaminated reactor components is a very sensitive object in view of the public acceptance. In the past most foundries were very choice of a completely nonradioactive scrap material. They fear that protests of the staff or of inhabitants could disturb the regular operation of the factory.

In one case the management of a foundry ordered an independent specialist to measure the decontaminated material, which officially was certified as nonradioactive material by the inspection authority.

In 1984 the "Greens" party put a question to the Federal Government in the German Parliament about the "uncontrolled melting of radioactive" scrap material from the power reactor station GUNDREMMINGEN. The government however could state that all material was decontaminated and officially released for unrestricted use and therefore the recycling was legal[4]. To be sure that a license for recycling of decontaminated metal holds against a possible suit, the licensing authorities will probably be very restrictive in the near future.

DECONTAMINATION OF REACTOR COMPONENTS

Arisings of Contaminated Components

The decontamination of reactor components in a technical scale came up in 1975, when in the nuclear power station WUERGASSEN (KWW) the steam drier was removed. Later on repair and backfitting measures became necessary especially at BWLs. Most of the removed material were tubes, vessels and condensors. Finally the installation of fuel element compact racks in the first half of the eighties rendered the old ones unnecessary. Therefore about 5000 Mg of radioactive scrap is waiting for decontamination or disposal.

Meanwhile also several power reactors have been shut down.

In 1974 the 100 MWe heavy water moderated CO_2 cooled pressure tube power station NIEDERAICHBACH has been shut down after only a few days operation. In 1981 the license for mothballing of the plant was given. Now the total destruction of this plant will start in 1985 with the removal of nonradioactive components. The work will be finished in the first half of the nineties with the restoration of the "green meadow". During and after this work about 1700 Mg radioactive material for decontamination, 1200 Mg radioactive waste and 130000 Mg nonradioactive waste have to be processed [5].

The 268 MWe BWR LINGEN(KWL) was in operation since 1968 when it was shut down for repairs and modification in 1977. However because of very stringent demands of the licensing authorities it was decided in 1979 to mothball the nuclear part of the power plant for 30 years. After this time a complete dismantling is envisaged. Instead of the nuclear part a gas turbine is planned to install.

In 1980 the owner of the nuclear power plant GUNDREMMINGEN (KRB) decided to decommission the 250 MWe BWR. The plant was in operation from 1966 to January 1977. It was shut down after damages resulting from a short circuit in the net. In 1983 the license for mothballing the plant was given. Contaminated equipment is to be decontaminated. For the decontaminated components a licence for unrestricted release should be proposed. About 2500 to 2800 Mg will be to decontaminate. According to estimations about 70 % of the material will be below the activity limits for unrestricted release. 25 % may have a residual contamination above 0.37 Bq/cm^2. Possibly it can be molten under controll. About 5 % may be radioactive waste, because it cannot be decontaminated under economic conditions. In a first test about 100 Mg of contaminated components with a contamination of about $6 \cdot 10^7$ Bq (= 1.6 mCi) have been decontaminated. 92 % of it can be released for unrestricted use. About 6 % are small pieces, which cannot be measured exactly because of their shapes. Nevertheless their activity is below the limits so that a controlled melting is proposed. About 2 % could not be decontaminated and will be disposed of as radioactive waste[6].

The nuclear ship OTTO HAHN was in operation from 1968 until 1979. In 1980 to 1982 the nuclear energy system was completely removed. The pressure vessel including shielding with a total weight of 480 Mg was transported to the nuclear research center Geesthacht. (GKSS) for material tests. All the other components were decontaminated or prepared for final disposal as radioactive waste. About 375 Mg of components could be released for unrestricted use. 305 Mg radioactive parts were packed into 400 litre drums or containers for intermediate storage. About 14000 m^2 surface of the ship were decontaminated. Meanwhile the ship is going to be used as a conventional container ship driven by a diesel engine [7].

Decontamination Procedures

Aims and procedures for the decontamination of removed components in the course of repair backfitting and decommissioning measures are very different from on-site decontamination of highly contaminated coolant system to reduce the radiation exposure of personell during inspection and repair work. The on-site decontamination of the coolant system is carried out with chemicals, which dissolve only the oxide layer. The underlaying metal must be protected against intergranular

stress corrosion. Dilute organic acids sometimes in combination with chelating agents are often used successfully. In PWRs an oxidation of chromium oxide or iron chromite resp. prior to the dissolution of the oxid layer is necessary. Decontamination factors between 5 and 20 are normally gained.

To the contrary decontamination of removed components with the aim of unrestricted use of the contaminated material can and often must be done with strong acids, which not only dissolve the oxide layer but also the underlaying metal to be sure that activity diffused into the metal matrix is removed. A careful treatment regarding surface state is not necessary. An important point of view in selecting a decontamination procedure is not only the removal of activity but also the treatment and final volume of the secondary waste. For example an agent may remove the activity very effectively, however it will also severely attack the equipment, which is used for rework or waste treatment. So a compromise is to be found between effective activity removal and protection of the equipment against attack to the agent especially against pitting and intergranular corrosion. An typical example is hydrochloric acid, which removes the activity in a short time but initiates pitting corrosion of the evaporator during secondary waste treatment.

A very simple method for the decontamination of stainless steel is pickling with a mixture of nitric and hydrofluoric acid (e.g. 15 % HNO_3/ 5 % HF). This mixture is effective up to about 40 g of iron per litre. The exhausted solution can be concentrated by evaporation. It attacks the steel uniformly. No pitting corrosion is to expect. In the waste treatment plant of the Nuclear Research Center Juelich (KFA) this solution is mixed with other waste water. If possible it is used for the neutralization of alkaline waste. The waste water is evaporated at a pH of about 3.5 to 4 up to a concentration of about 30 % solids. The concentrate is further evaporated to dryness with a rotating drum drier. Finally the dry product is immobilized with cement.

Carbon steel and cast iron are often decontaminated with organic acids. Depleted solutions may be mixed with other waste water and concentrated by evaporation. If a high amount of complexing organic acids or of other chelating agents are present an enhanced leachability of several radioacitve isotopes from cemented waste must be taken into account.

Hydrochloric acid has been used for the decontamination of the nuclear ship OTTO HAHN and partially in GUNDREMMINGEN. About the secondary waste treatment nothing has been published.

The application of ultrasonic waves may support or improve the decontamination especially of small pieces with complicated surface. Experiments at KFA have shown that stainless steel cladded transducers can also be used successfully in HNO_3/HF-mixtures.

Recently electropolishing in 70 % phosphoric acid has been proved to be a very suitable decontamination procedure [8]. About 64 Mg of the metallic material from the dismantling of the nuclear power station GUNDREMMINGEN have been decontaminated in this way below the activity limits. The secondary waste mass is reported to be lower than 2 % related to decontaminated steel.

The application of decontamination pastes consisting of inert material like aluminium oxide or titan dioxide mixed with mineral acids are used very successfully. The contaminated component can be coated with the paste by spraying easily. The advantage is a lower secondary waste volume and no higher costs, especially if the pastes used can be prepared in own production. Beside the chemical treatment also mechanical treatment, rinsing with water and removal of paints are usual in decontamination of low contaminated components.

Decontamination Facilities in the Federal Republic of Germany

Since many years the two big German Nuclear Research Centers, KFA Juelich and KfK Karlsruhe, operate efficient decontamination shops. Primary contaminated components and equipment from their own institutes is cleaned for reuse. With the beginning of the extensive repair and backfitting measures they also accepted orders so far they have available capacity.

In most cases service companies like the working group Kraftanlagen Heidelberg/Nukem or Gesellschaft für Nuklear Service GNS remove components in situ and ordered the decontamination, for instance in Juelich or Karlsruhe. Recently Kraftwerk Union KWU has also installed an own decontamination shop. On-site decontamination work was done by Gg. Noell (nuclear ship OTTO HAHN and decommissioning of the nuclear power station NIEDERAICHBACH) as well as other small service companies. Decontamination of the first 100 Mg charge of the shut down power reactor GUNDREMMINGEN is mainly done by its own staff.

Decontamination of Reactor Components in Juelich

At the present time about 600 Mg contaminated components of power reactors from repair and backfitting measures are under contract in Juelich. It is mainly

- a steam drier from the BWR WORGASSEN(60 Mg),
- condenser tubes (360 Mg),
- turbine housings, and
- fuel element racks.

The surface dose rate of the steam drier is up to about 0.1 Sv/h (= 10 rem/h), that of the condenser tubes, racks and turbine housings between about 0.01 mSv/h (1 mrem/h) up to about 1 mSv/h (= 100 mrem/h). The components were transported to Juelich in steel containers, concrete containers or shielding casks.The maximum allowable weight is 5 Mg with respect to the load capacity of the installed crane. The most problematic component was the steam drier because of the relative high dose rate and the complicated internal structure. First we had to decide if the heavy parts should be cut into smaller pieces by remote handling with following decontamination or if it would be more advantageous to predecontaminate large parts and than to cut them by direct operation. It soon became clear that the latter alternative was much more time and costs saving [9].

For the decontamination of large parts a 10 m^3 pickling bath is available. Parts up to 3 m length, 2.50 width and 2 m height can be treated here.

The 3 to 5 Mg pieces were submerged in the acid for some days until the dose rate was reduced to about 200 mR/h. Than the pieces were rinsed and cut into smaller pieces by a plasma torch, so that all the inner surfaces were open to further decontamination. The final treatment occurs by pickling, treatment with decontamination paste and mechanical treatment. Areas with unremovable contamination, especially welding seams are cut away. About 90 % of the material could be cleaned below the activity limits for unrestricted release. In a similar way the fuel element racks were

treated with acid. After rinsing all screws were removed and the disassembled components were dipped again into the pickling acid. In most cases a simple mechanical treatment with sponges or brushes was added.

The outside contaminated condenser tubes were cut into 2.2 m long pieces, and closed by pinching. They were loosely packed into a rotating drum. After addition of the decontamination agent the drum was rotated for half an hour. Already after a few minutes the surface layer was removed.

MEASUREMENT OF SURFACE CONTAMINATION

Plain surfaces are measured with commercially available proportional counters. However curved surfaces need special measuring devices. Such were develpoed in cooperation with the electronics work shop of the health physics department of the KFA [10].

These new contamination detectors consist of input amplifiers, resistant to overdriving with discriminators, special α/β windows of 100 cm^2 and a unit weight of 1 mg/cm^2, and are equipped with electric cable connections with counting gas passage.

For inside measurements of pipes cylindrical probes with diameters of 15 and 40 mm are available. Pipes with a diameter of 90 mm can be measured with a flat detector, which is 200 mm long and 50 mm broad. For the outside measurements a detector block has been developed consisting of four or more flat detectors cylindrically arranged around the pipe.

For the outside measurement of condenser tubes - in the present case with a diameter of 24 mm - an automatically working device has been constructed. The detector consists of 4 flat detectors, which are shielded with 5 cm of lead. The system has a background of 4.3 Ips and an efficiency of 43 % for Sr-90/Y-90. The condenser tubes are moved on cone-shaped rolls through the detector block and are sorted out to contaminated tubes and tubes for unrestricted release.

UNRESTRICTED RELEASE

For the unrestricted release of the decontaminated scrap an extensive documentation has to be established. Each part, which is ready for release is specified and the following data are tabulated: customer, charge number, individual number, type of material, geometry, surface area, mass, measuring instrument, efficiency of the instrument, measured data, calculated surface contamination and specific activity.

The total surface of all parts is measured. In most cases also samples are analyzed in the laboratory. Later on 10 % of the material is measured by an independent health physics team of the research centre. Finally random samples were controlled by an independent expert. Than the supervising authority agrees with the unrestricted release. By this multistage release procedure a maximum of safety can be guaranteed. Now the customer can deliver the released material to a foundry.

In 1983 and 1984 about 260 Mg of decontaminated reactor components have been released for recycling. However by the technical improvements in the decontamination of the condenser tubes and by automatization of the tube measurement the throughput is increased to about 10 Mg condenser tubes per month beside the decontamination of other components.

CONCLUSION

The decontamination of reactor components below release limits has been proven to be possible in technical scale at a acceptable costs. About 90 % of the decontaminated material has been released. The melting in a conventional foundry does not cause any problem.

ACKNOWLEDGEMENT

I would like to acknowledge the kind assistance of my collegues W. Schulz and F. Rosenbaum.

REFERENCES

1. GESETZ über die friedliche Verwendung der Kernenergie und den Schutz gegen ihre Gefahren (Atomgesetz AtG), BGBl I Nr. 131 p. 3053 (1976).

2. AUSLEGUNG des § 4 Abs.4 Satz 1 Nr. 2e StrlSchV, GMBl 1979, p. 631.

3. VERORDNUNG über den Schutz vor Schäden durch ionisierende Strahlen (Strahlenschutzverordnung - StrlSchV), BGBl I Nr. 12 p. 2905 (1976).

4. ANTWORT der Bundesregierung auf eine kleine Anfrage des Abgeordneten Sauermilch und der Fraktion DIE GRÜNEN, Bundestagsdrucksache 10/2050 (1984).

5. KERNFORSCHUNGSZENTRUM KARLSRUHE GmbH, "Demontage und Beseitigung Anlage Niederaichbach, Kurzbeschreibung", (1984).

6. E. EDER, J. KOLLERBAUER, and L. SCHÄFFLER, "Abbau, Dekontamination und Freigabeverfahren zur schadlosen Wiederverwertung von Anlagenteilen aus dem Kernkraftwerk Gundremmingen, Block A", Bericht des Bayerischen Landesamtes für Umweltschutz, (1984).

7. K. D. HENNING, "Strahlenschutzaspekte bei der Stillegung des Kernenergieschiffes OTTO HAHN", Bericht der 17. Jahrestagung des Fachverbandes für Strahlenschutz e.V., FS-83-32-T, p. 617, (1983).

8. N. EICKELPASCH, und M. LASCH, "Neuere Erfahrungen zur elektrochemischen Dekontamination", VGB Kraftwerkstechnik Heft 3, (1984).

9. M. LASER, F. ROSENBAUM und W. SCHULZ, "Dekontamination und Beseitigung von ausgebauten Reaktorkomponenten", Bericht der 17. Jahrestagung des Fachverbandes für Strahlenschutz e.V., FS-83-32-T, p. 617, (1983).

10. M. KELLER, "Alpha-Beta-Kontaminationsdetektoren insbesondere für Freigabemessungen von Anlagenteilen aus kerntechnischen Anlagen", Bericht der 17. Jahrestagung des Fachverbandes für Strahlenschutz e.V., FS-83-32-T, p. 519, (1983).