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Experiences in the Management of Plutonium-Containing Solid-Wastes at the Nuclear Research Center Karlsruhe

W. Bähr, W. Hild, K. Scheffler

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Experiences in the Management of Plutonium-Containing Solid Wastes at the Nuclear Research Center Karlsruhe

by

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Kurzfassung

Plutonium-kontaminierte Festabfälle aus einer Brennelementproduktionsanlage, einer Wiederaufarbeitungsanlage und verschiedenen Forschungslaboratorien werden in den Dekontaminationsanlagen des Kernforschungszentrums Karlsruhe für die Beseitigung im Salzbergwerk Asse behandelt. Die Anlagen zur Behandlung dieser Abfälle sowie deren Konditionierung werden beschrieben. Ein Ausblick auf die zukünftige Entwicklung wird gegeben.

Abstract

Solid-plutonium-containing wastes from a fuel production plant, a reprocessing plant and several research laboratories are treated at the decontamination department of the Karlsruhe Nuclear Research Center for disposal in the Asse salt mine. Conditioning as well as future aspects in α-waste management are the subject of this Paper.

1. General Remarks

At all operations and processes of the plutonium fuel-cycle wastes are generated the treatment, solidification and disposal of which poses not only particular difficulties but also considerably high cost.

At the beginning of the "Plutonium Age" both the extremely high economic value of the plutonium and its pronounced political importance as fissile material, led to the development of rather costly and complicated techniques aiming at an as complete recuperation of plutonium as possible. The growing plutonium inventories, however, resulted in a steady decrease of the plutonium price thus drastically cutting down the efforts for the recuperation of Pu from wastes, expecially those procedures that aimed at the recuperation of mg-amounts of Pu from large waste volumes. It was thus mainly due to economic reasons, that the Pu-concentrations in the various waste streams were steadily increasing.

Experiments with small amounts of plutonium and other transuranium elements first started in the FRG in the early sixties. At the Nuclear Research Center of Karlsruhe, especially the Institutes for Radiochemistry and Hot Chemistry and later the European Institute for Transuranium Elements too started to handle those elements on a mg- to g-scale. Finally the Alkem Company erected a fuel-element production facility on the site, in which plutonium was handled on a kg-scale. Larger amounts of plutonium are furthermore handled in the first German reprocessing plant, WAK, since start-up of this facility in the early 70's.

Roughly 600 kg of Plutonium were handled in the above mentioned institutes and facilities since 1965 till mid 1974 both in solution and in solid form.

Up till now roughly 4700 drums of Pu-contaminated wastes with a total Pu-content of approximately 15 kg (i.e. some 3g/drum) were solidified. About 25% of these drums result from the solidification (concreting or bituminization) of evaporator-concentrates from low-and intermediate level effluents from reprocessing and fuel element fabrication. The other 75% result from the conditioning of solid wastes. All Pu-bearing wastes were exclusively conditioned into disposable products at the decontamination department of the Karlsruhe Nuclear Research Center. (1)

Amount of Plutonium Contaminated Solid Waste
in the FRG

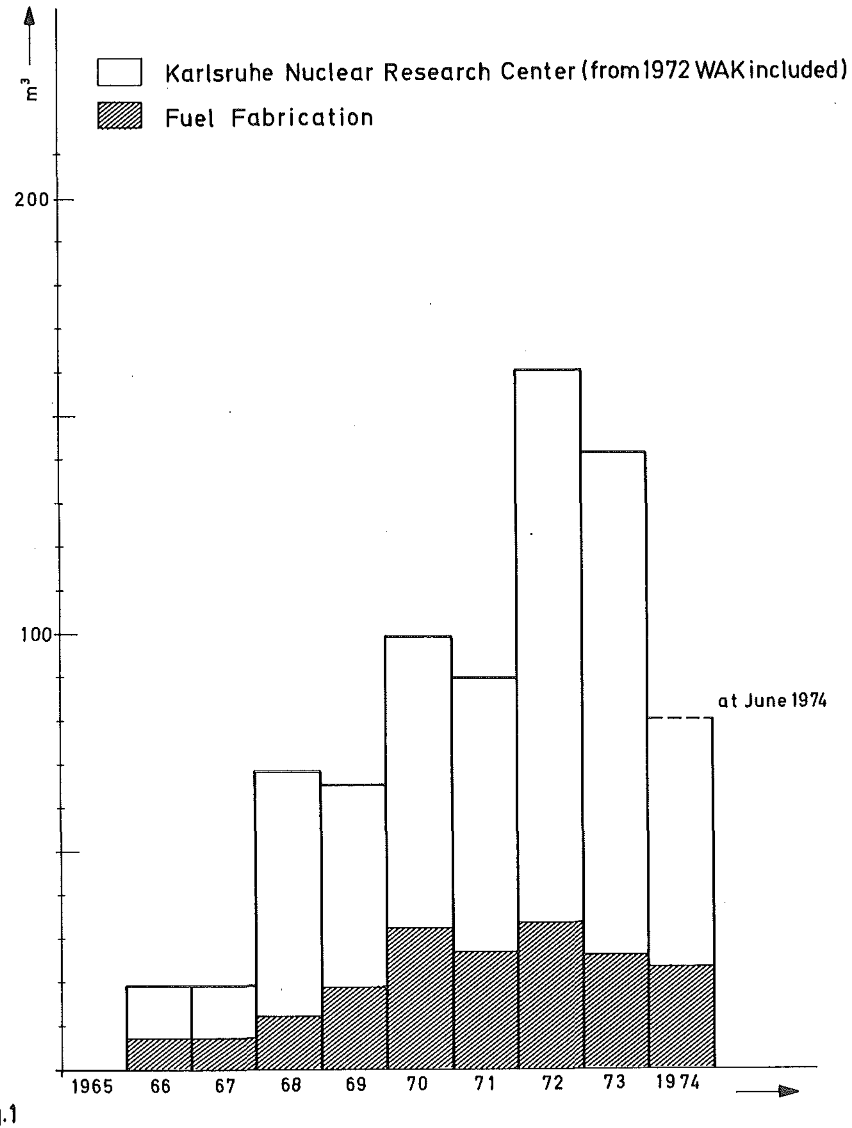


Fig.1

Fig. 1 gives a compilation of the total amounts of α-contaminated solid wastes generated at the FRG up till now. During the last 10 years a total of 730 m^3 of solid waste was generated.

The pronounced increase in the waste production since 1972 is on one hand due to the start-up of the reprocessing facility WAK at Karlsruhe. On the other hand it is due to start-up of a facility especially erected for treatment and conditioning into disposable form of bulky waste on large α-contaminated equipment like glove-boxes, process components etc. In the latter facility some 225 stainless-steel and plexiglas glove-boxes have been either decontaminated for reuse or treated as wastes till now. Prior to the start-up of this facility the contaminated equipment pending treatment was stored sealed in PVC-foil.

2. Actual practice

2.1. Segregation and collection of α-contaminated solid waste

The main quantities of α-contaminated solid waste, are generated within glove-boxes; only small amounts result from operation outside boxes, mainly during decontamination campaigns as for instance cellulose tissues, gloves etc., the contents of α-emitters of which is rather small. In the glove-boxes combustible waste like paper, gloves, plastic material, is collected either in plastic bags or in cylindrical cardboard containers which are removed from the boxes after being tightly sealed into plastic bags. All non-combustible waste like glass, metal or other spent material is always collected separately in cylindrical cardboard containers. After withdrawal from the boxes both waste categories are separately collected in 200 l-reinforced drums, which are transported to the decontamination department at Karlsruhe. Waste containing drums from waste producers outside the Nuclear Researcn Center Karlsruhe are transported to the treatment facilities in an additionary over-pack as shown in figure 2. This waste is almost exclusively shipped from the ALKEM-company, which since 1972 has transferred its fuel element production plant some 150 kilometers from the Center to Wolfgang. The decontamination department conditions and disposes these wastes on a contractory basis. For the transport of bulky material like stainless steel boxes or boxes with equipment for fuel element production special transport containers are available.

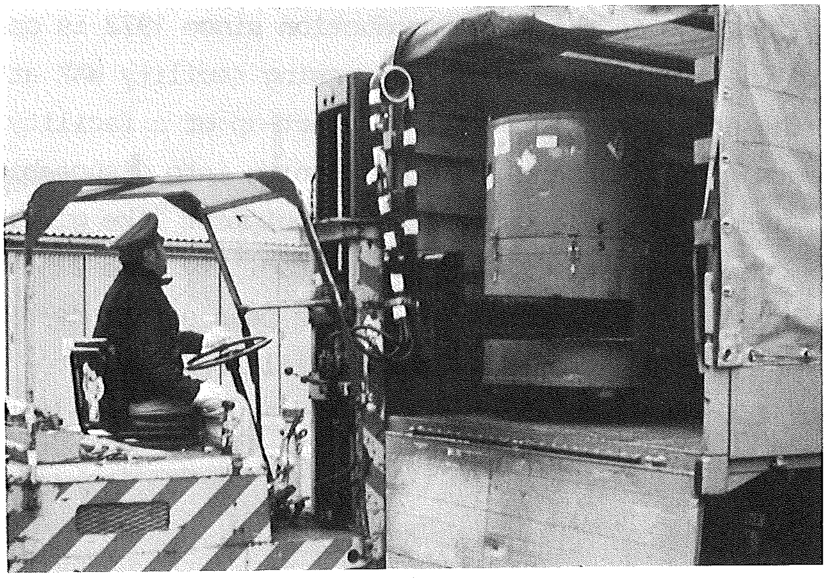


Fig.2

2.2. Determination of plutonium in solid waste.

Both neutron and γ-counting as well as decay-heat measurement can be applied for quantitative Pu-determination. According to the experience in the FRG counting of fission neutrons as well as calorimetric determinations can only be applied for relatively high Pu-contents in the g/l-range. This is why these methods are only used for wastes that pass through a plutonium recovery treatment. (2)

For real wastes the plutonium determination is nowadays performed by γ-spectrometry of the 50 keV- and 380 keV-lines of Pu-239 with a single-channel-analyzer equipped with sodium iodide detector.

The sample i.e. the 200 l-drum containing the waste is placed on a turntable that rotates at 2 rev/min. A threaded spindle moves the γ-detector up and down the rotating drum. Geometrical errors of the system are thus almost completely compensated (fig. 3). The absorption coefficient of the sample can be determined by comparing the intensity of the 50 keV and the 380 keV lines: fission products, AM-241 and uranium in considerable concentration disturb the measurements.

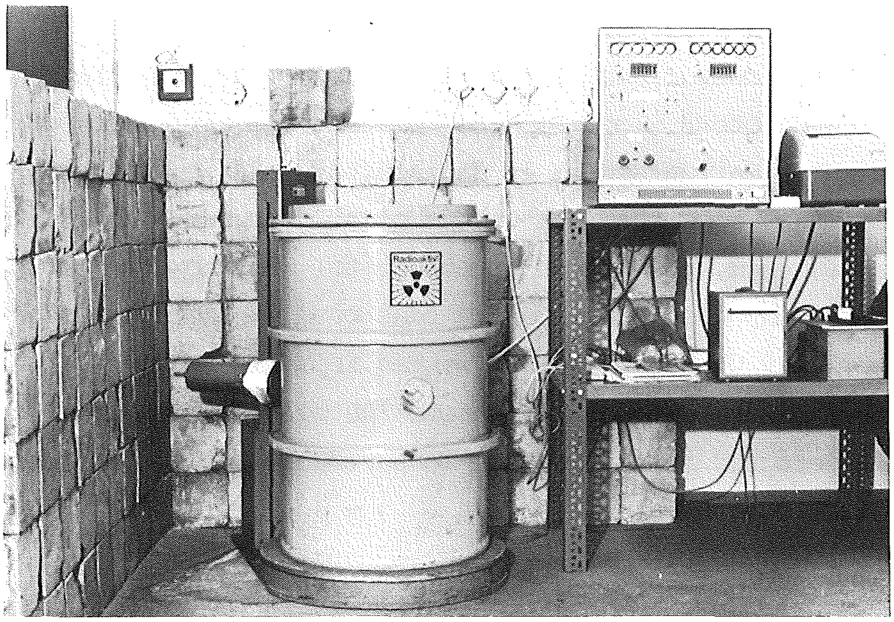


Fig. 3

For "pure" Pu-wastes, however, rather good results are obtained with a detection limit in the order of 2mg Pu per 200 l waste drum.

2.3. Treatment and disposal of α-contaminated solid wastes

In principle no technically demonstrated treatment procedures are available for α-contaminated combustible and non combustible wastes up till now. Treatment techniques applied in the early days never left the experimental scale and were almost completely dropped.

As a consequence of both the ideal disposal possibilities in the FRG and economic considerations the rather small quantities of α-contaminated solid wastes were disposed of without considerable volume reduction.

Up till now all wastes with Pu-contents smaller than 10 mg Pu per 200 l drum were treated together with the large amounts of other radioactive wastes at the Nuclear Research Center Karlsruhe.

Combustible waste is since a couple of years incinerated in the incincerator plant of the waste treatment department. The plant operates with a dry offgas purification system, that passes the flue gases through a series of ceramic filter candles. (1)

The furnace has, however, not specially been designed for the combustion of Pu- or other α-contaminated wastes. The efficiency of the off-gas purification system is satisfactory as far as the Pu-decontamination is concerned, the other plant components as for instance the furnace, the filter chambers etc, are, however, not α-tight. It has happened occasionally that room contaminations occurred, especially when combustible wastes from the reprocessing station were incinerated containing some 100 mg of Plutonium that could not be detected in the control-measurements due to pronounced fission product concentrations.

Due to the experience gained with the incineration of combustible wastes of low Pu-contents a Pu-concentration limit $\leq$ 10 mg Pu/200 l drum was set up above which combustible wastes may not be incinerated in the existing plant. The incinerator ashes are collected in drums where they are solidified by means of cement-grout.

All combustible wastes with Pu-concentrations higher than 10 mg Pu/drum are up till now conditioned by a special treatment into disposable forms without volume reduction: This is performed by inserting the 200 l reinforced drum containing the α-contaminated waste into a 400 l sheet-metal drum and filling the voids with concrete. (fig. 4). This treatment procedure is certainly rather expensive but for the moment being a very simple and extremely safe method. A safe final disposal in the salt mine Asse is guaranteed by the additionary containment of the complete 7.5 cm thick concrete layer between the two metal drums. This treatment procedure has been adapted not only for the above mentioned safety aspects related with perpetual disposal but also for the following practical reasons: Unpacking of the expensive 200 l reinforced drum serving as transport container too into cheaper sheet-metal drums, is connected with the risk of contaminations.

Non-combustible wastes are treated in the same way: Wastes containing $\leq$ 10 mg Pu/200 l reinforced drum are baled together with other radioactive wastes in a baling press. The compressed waste residues are packed into a basket that is inserted into a 250 l sheet metal drum. (1)

Packaging of α-Contaminated Solid Waste for Disposal

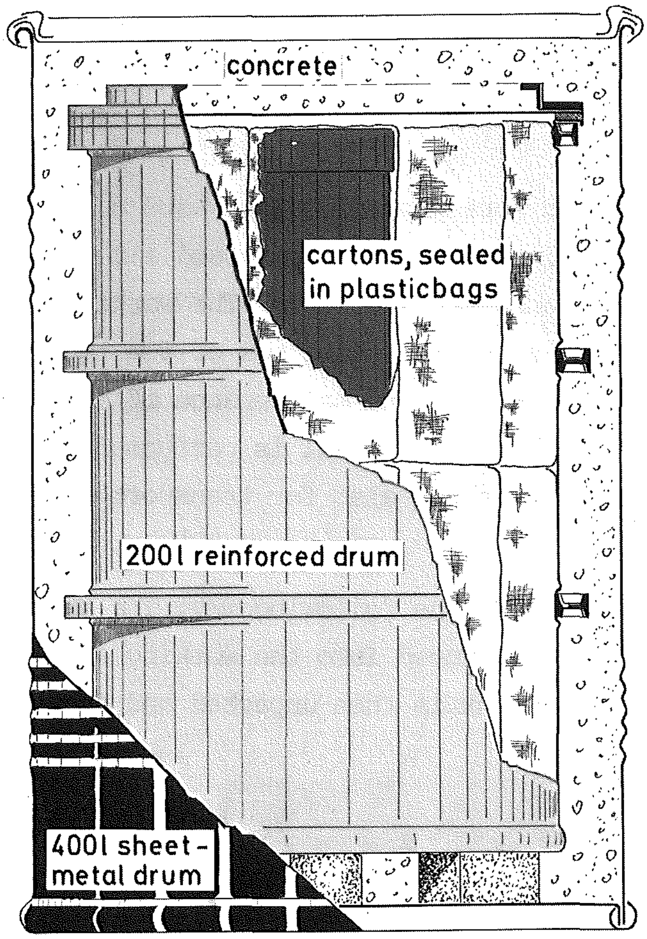


Fig. 4

The voids between the basket and the inner drum surface (some cm thick) are completely filled with concrete grout. Non combustible wastes with Pu contents of more than 10 mg Pu/drum are treated in the same way as described for the combustible wastes in this contamination range.

More complicated and still more expensive techniques are applied for the treatment and disposal of large and bulky equipment like glove boxes and special process equipment that asks for exhaustive dismantling prior to the conditioning into disposable form. A special α-tight working area of 80 m^2 is available for these operations. Equipment up till 6 m length and a weight of 3 tons can be handled. The bulky waste items are introduced to the working area via a lock. The operators enter the working area via a special frogman suit lock and are always supervised from an outside control room through large windows. In total 4 operators can be in action at one time in this working area. A special stainless-steel immersion basin for the decontamination of glove boxes mainly is installed at the floor of the working area. (3)

For the dismantling and scrapping of bulky waste numerous tools like saws, plate-shearing mashines, compressed air operated cutting disks, and an oxygen lance are available. The working area is connected via a door with a special glove box of 6 m length, which is mainly utilized for the repair and maintenance of α-contaminated masterslaves. Introduction and removal is performed by the use of a special container that serves also for transport of the equipment.

Glove boxes are scrapped in the following way: The boxes, sealed in plastic foils, are introduced into the working area via a lock by means of a crane. The box is then unpacked and opened (fig. 5).

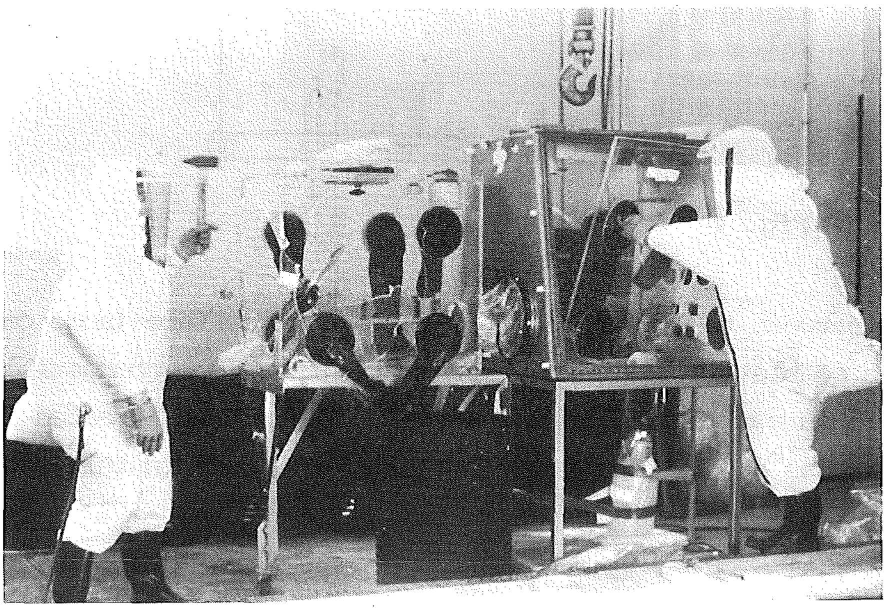


Fig. 5

The installations in the boxes which are normally not recovered are either directly or after further cutting into pieces packed into 175 l sheet-metal drums. Emptied boxes which are not decontaminated for reuse are also cut into pieces and packed into the mentioned drums (fig. 6).

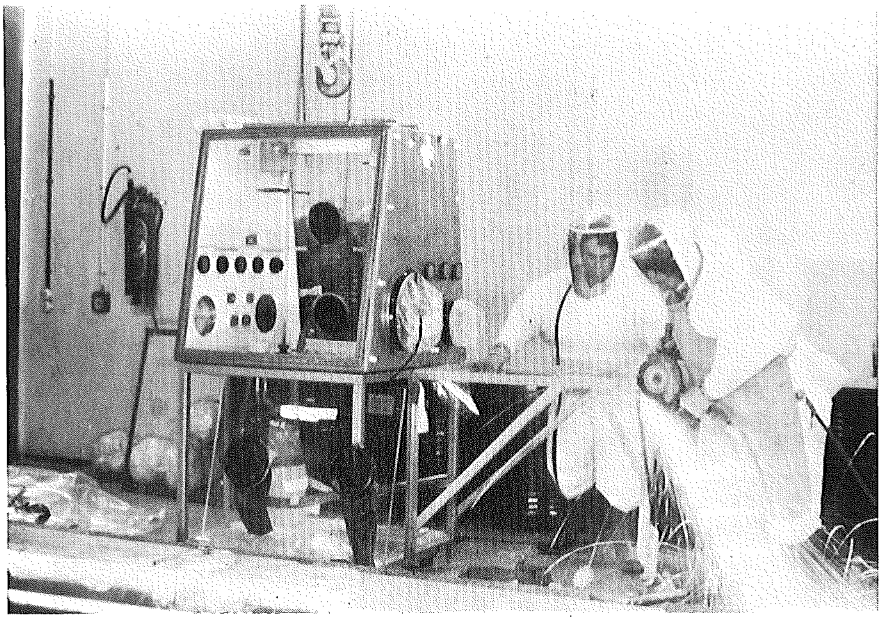


Fig. 6

After filling a certain number of drums with scraps, concrete grout is pumped into the drums via a special pipeline entering the cell from outside. After setting of the concrete the 175 l sheet-metal drums are in the lock inserted into 200 l reinforced drums which are wrapped into plastic foils to avoid external contamination. After closing the 200 l reinforced drum, the plastic foil is separated and the drum withdrawn from the lock.

These dismantling and packaging techniques have been very successfully applied since 3 years.

Another possibility for the treatment of contaminated glove-boxes is an effective decontamination aiming at a reutilization of the boxes. For economic reasons only stainless-steel boxes are suitable for this alternative. The glove boxes are completely emptied and afterwards treated with a special pickling paste for stainless steel and rinsed in the immersion basin (fig. 7). By further treatment with different decontamination solutions the boxes are predecontaminated to an extend that allows the transfer of the boxes to a large β/γ-decontamination facility where the final decontamination is performed. According to the requirements the glove boxes can be decontaminated until MPC-values. A very successful method for effective fine decontamination is the electro-polishing procedure, leading to excellent results for stainless steel boxes.

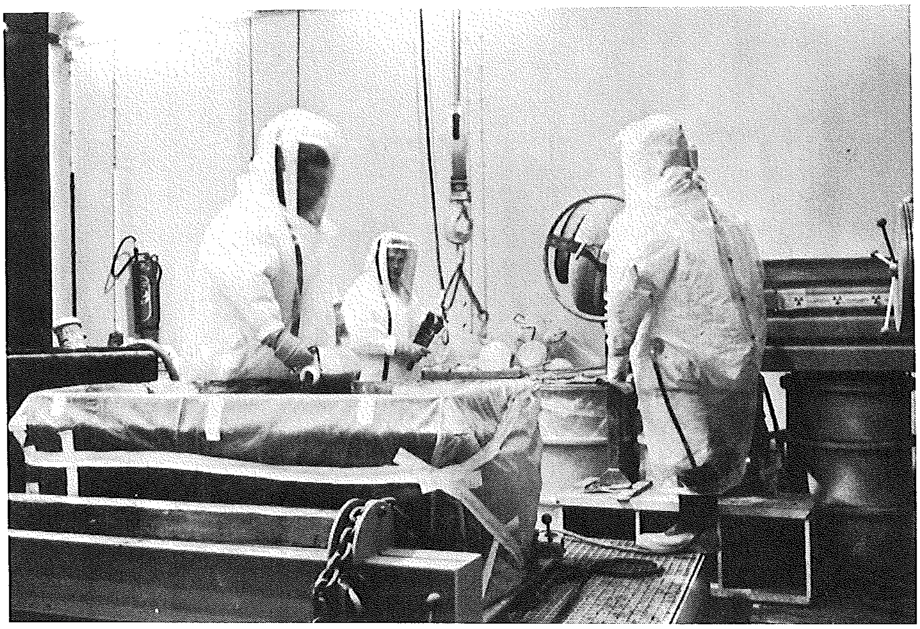


Fig. 7

2.4. Final Disposal of Pu-contaminated wastes

It is generally agreed upon that geological formations like salt deposits offer for the moment being the best solution for the disposal of radioactive waste. Due to their geological history salt formations act as a natural containment that guarantees complete isolation of the radioactive waste from the biocycle. This is an essential requirement for the safe disposal of radioactive waste. (4)

Treatment of these wastes had to meet the acceptance conditions established in 1971 for the disposal of radioactive wastes in the salt mine Asse, the general repository for radioactive wastes in the FRG, allowing a maximum Pu-239 content of 8g per 200 liter drum of solid waste. The contents of other nuclides from the transuranium elements per 200 liter of solidified waste is limited to a total of 0,5 Ci, Plutonium included. (5)

In the FRG the salt mine Asse II has been selected as the central repository for all radioactive wastes generated. It is thus, that the drum containing the conditioned α-wastes are disposed of at the Asse mine, too.

3. Future aspects of the treatment and disposal of α-contaminated waste in the FRG

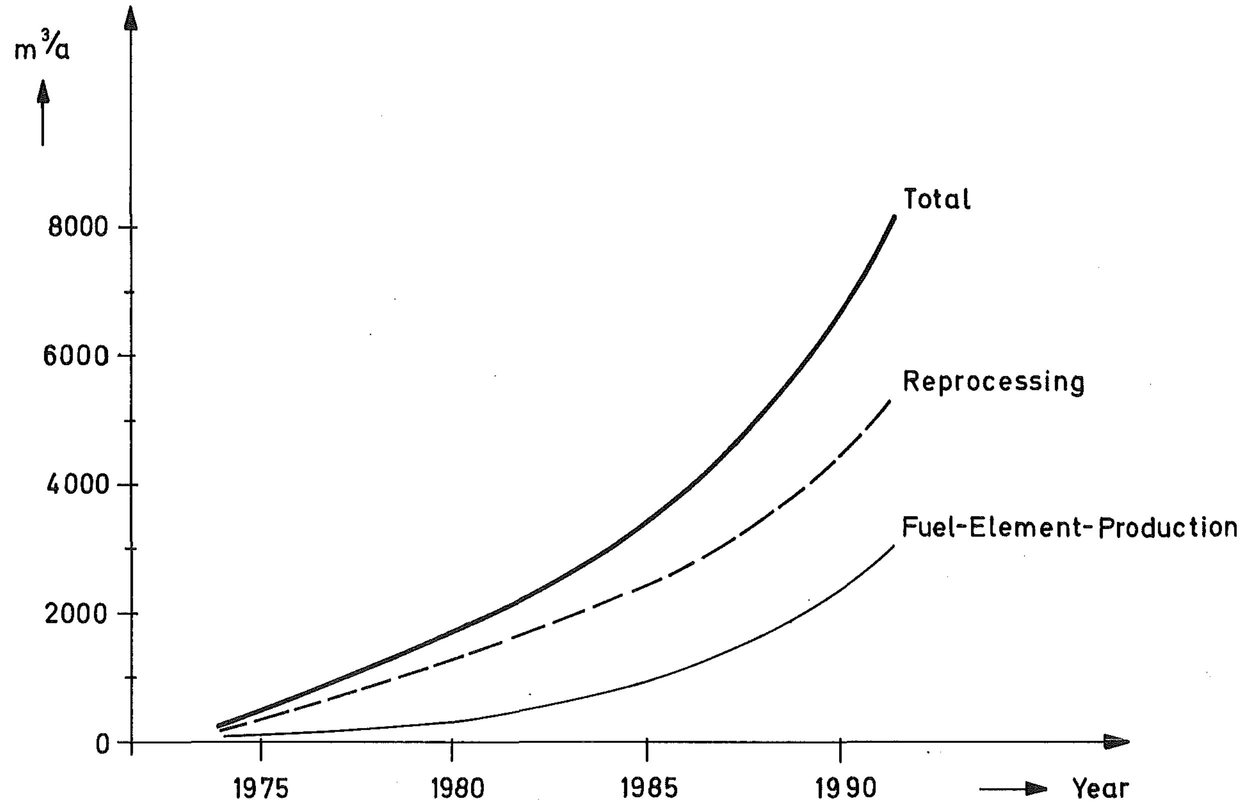
The experience collected during treatment and disposal of the Pu-contaminated wastes that have been generated in the FRG up till now, show that the procedures actually applied represent a satisfactory solution to the problem. With the expected expansion of the nuclear energy during the next decades, however, a drastic increase in the generation of radioactive wastes is inevitable.

The rising utilization of Plutonium in thermal power reactors will certainly lead to a higher production of Pu-contaminated wastes too. Even if advanced technologies for fuel-element production and reprocessing of irradiated fuel are taken into account, the production rate of solid Pu-and α-contaminated waste amounting 1970 to 100 m^3/year can be estimated to rise more than 1000 m^3/year in 1980 and to almost 10000 m^3/year in 1990 (fig. 8). At the same time Pu-losses to the solid waste will most probably increase too.(6) It should be mentioned, that the above made estimations are based on the assumption that the actual treatment and disposal techniques for Pu-contaminated waste continue to be applied. These relations and the fact that the rather long half-lives of the α-emitters in question ask for an extremely high degree of safety for long-term storage demonstrates clearly that the actually applied methods have to be thoroughly reconsidered.

In this connection special emphasis has to be devoted to the definition of a realistic concentration limit that allows a clear classification of α-bearing and α-free radioactive waste.

The special situation asks for immediate extensive research and development activities in the field of the overall α-waste-management. The efforts have to be focused on the following two main lines: On the one hand improvement of the various process steps in the nuclear fuel cycle aiming at a maximum recovery of fissile material and a drastic reduction of α-bearing wastes. On the other hand development of treatment procedures that take care of the anyhow arising α-bearing wastes in such a way that two waste streams are obtained: one concentrate containing essentially all the α-emitters in an as small as possible volume and a second stream that is virtually α-free.

Plutonium-Containing Solid Waste and Plutonium-Losses to Solid Waste in Fuel-Element-Production-and Reprocessing-Plants



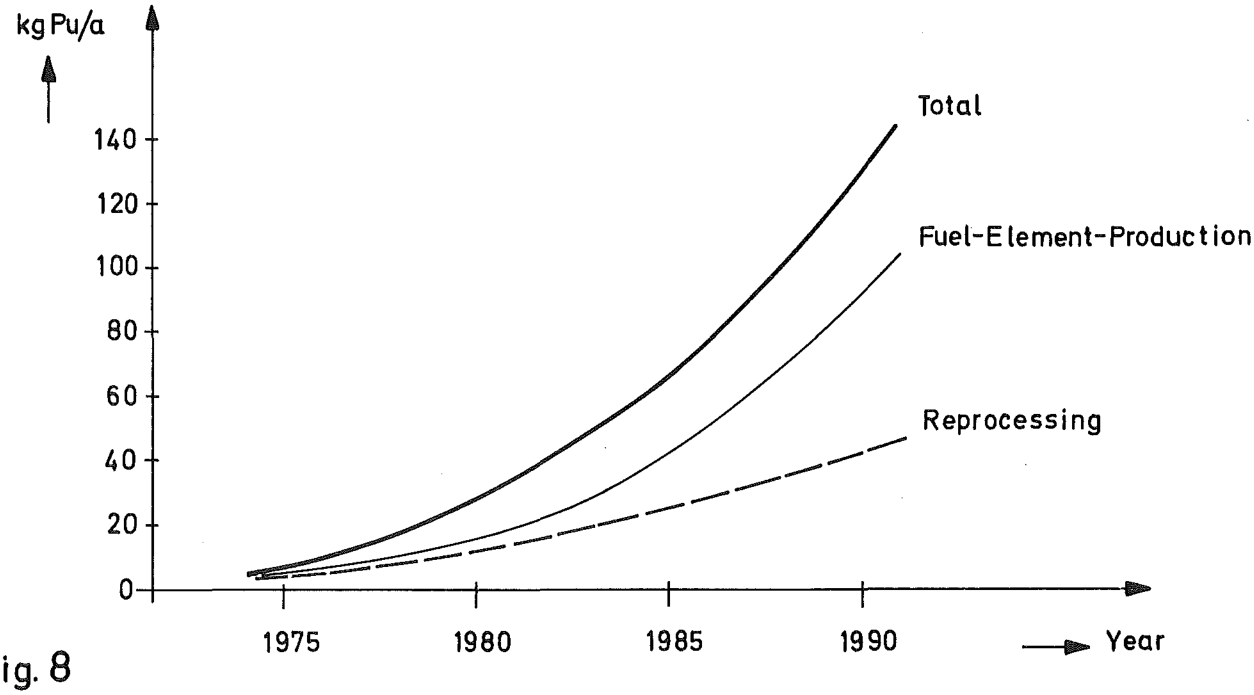


Fig. 8

The concentrate can either be passed to a Pu-recovery step or directly be solidified in such a way that safe long-term storage- even in retrievable form - is possible.

In this context various research programs are actually defined and executed according to the priorities selected. For the treatment of combustible wastes for instance, dry and wet combustion procedures will be investigated and demonstrated on a pilot-scale. A further main effort will be devoted to the selective separation of α-emitters from the large volumes of low and intermediate level liquid effluents or their concentrates in order to reduce the α-emitter-concentration in the solidified residues of this waste category as low as possible. At the same time emphasis will be given to the development of new solidification products for α-emitters that meet the extremely strong safety requirements for long-term disposal.

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