

**KERNFORSCHUNGSZENTRUM  
KARLSRUHE**

November 1972

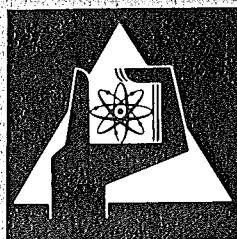
KFK 1741

Abteilung Dekontaminationsbetriebe

**Treatment and Final Disposal of Radioactive Wastes from  
Fuel Reprocessing in the Federal Republic of Germany  
– a Survey on Policy and R + D Work –**

H. Krause  
Gesellschaft für Kernforschung, Karlsruhe

R. Randl  
Bundesministerium für Bildung und Wissenschaft, Bonn



**GESELLSCHAFT  
FÜR  
KERNFORSCHUNG M.B.H.**

**KARLSRUHE**

Als Manuskript vervielfältigt

Für diesen Bericht behalten wir uns alle Rechte vor

GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H.  
KARLSRUHE

KERNFORSCHUNGSZENTRUM KARLSRUHE

KFK 1741

Abteilung Dekontaminationsbetriebe

TREATMENT AND FINAL DISPOSAL OF RADIOACTIVE WASTES FROM  
FUEL REPROCESSING IN THE FEDERAL REPUBLIC OF GERMANY  
- A SURVEY ON POLICY AND R + D WORK -

by

H. Krause

Gesellschaft für Kernforschung, Karlsruhe

R. Randl

Bundesministerium für Bildung und Wissenschaft, Bonn

Paper presented at the Symposium on the Management of  
Radioactive Wastes from Fuel Reprocessing, OECD-NEA and  
IAEA, Paris, November 27 - December 1, 1972



## Kurzfassung

Die vorliegende Arbeit gibt einen Überblick über den gegenwärtigen Stand sowie die Tendenzen und Ziele von Forschung und Entwicklung auf dem Sektor der Behandlung und Beseitigung der radioaktiven Abfälle aus der Wiederaufarbeitung bestrahlter Brennelemente in der Bundesrepublik Deutschland.

Hochaktive Spaltproduktlösungen (HLW) sollen nach einer zeitlich begrenzten Zwischenlagerung in Tanks verfestigt und die verfestigten Rückstände in einem Salzbergwerk gelagert werden.

Im Kernforschungszentrum Karlsruhe wird an der Verfestigung der HLW-Lösungen aus der Wiederaufarbeitung von LWR-Brennelementen gearbeitet. Ein kombiniertes Sprühkalzinier- und Glasschmelzverfahren ist bereits entwickelt und wird derzeit in einer halbtechnischen Anlage im inaktiven Bereich getestet. Der Probetrieb unter hochaktiven Bedingungen soll in der Mehrzweckversuchsanlage VERA erfolgen, die sich noch in Planung befindet und so konstruiert wird, daß sie auch zur Erprobung anderer Prozesse, z.B. des Thermitverfahrens, eingesetzt werden kann.

In der Kernforschungsanlage Jülich befaßt man sich mit der Wiederaufarbeitung von HTR-Brennelementen und der Verfestigung der hierbei entstehenden HLW-Lösungen. Sie sollen mittels eines Trommeltrockners in feste Produkte überführt, diese in Graphittiegeln mit den entsprechenden Zusätzen zu Glas geschmolzen werden.

Im Hahn-Meitner-Institut in Berlin wird Grundlagenforschung bezüglich der chemischen und physikalischen Eigenschaften von Gläsern betrieben.

Im Rahmen des Programms für die Endbeseitigung hochaktiver Abfälle in Salzformationen werden Berechnungen zur Wärmeverteilung im Salzgebirge vorgenommen, der Einfluß erhöhter Temperaturen auf die Gebirgsmechanik geprüft und die technischen

Einrichtungen für Transport und Beseitigung der Abfälle entwickelt. Die erste Versuchseinlagerung hochaktiver Gläser ist für 1976/77 vorgesehen.

Des Weiteren werden auch Verfahren für die Behandlung und Beseitigung mittel- und schwachaktiver Abfälle aus der Wiederaufarbeitung bestrahlter Brennelemente entwickelt. Schwerpunkte der Forschung sind hier der Einschluß von Konzentraten mit hohem Nitratgehalt in Bitumen oder andere organische Materialien, die chemische Zersetzung von  $\text{HNO}_3$  und von Nitraten, die Behandlung TBP-haltiger, erschöpfter Lösungsmittel und die Versenkung tritiumhaltiger Abwässer in isolierte Speichergesteine des tiefen Untergrunds.

### Abstract

In the Federal Republic of Germany the high-level wastes (HLW) will be solidified after a limited interim storage as liquids and the solidified residues will be disposed of in a salt mine. At the Karlsruhe Nuclear Research Center the solidification of HLW from the reprocessing of light water fuels has been studied. An inactive pilot plant is in operation. The corresponding hot cell facility (VERA) is under construction. Its particular layout allows also testing of advanced processes and products being developed now.

At the Jülich Nuclear Research Center R + D work is carried out on the reprocessing of HTR-fuels and the solidification of this particular HLW which is converted into a dry product by a drum-drier and then melted into glass in graphite crucibles.

Fundamental research on chemical and physical properties of glasses is performed at the Hahn-Meitner Institut Berlin.

In connection with the ultimate disposal of the solidified fission products in a salt mine, a computation program is under way on the predetermination of heat dissipation in salt. Furthermore, investigations into rock mechanics at elevated temperatures are carried out, and equipment for the disposal is being developed. First experimental disposal of high-level glasses is scheduled to start in 1976/77.

Besides, investigations are carried out relative to the incorporation into bitumen and other organic matrices of high nitrate-bearing concentrates, the chemical decomposition of  $\text{HNO}_3$  and nitrates, the treatment of spent TBP-containing solvents, the disposal of tritium containing effluents into aquifers and the treatment of some other wastes generated in reprocessing plants.

## 1. INTRODUCTION

At the present time, water reactors with an installed total capacity of 1,600 MWe and a 15 MWe pebble-bed reactor (AVR) are operating in the Federal Republic of Germany. Under construction and ordered, respectively, are water reactors with 9,000 MWe in total, a 300 MWe THTR and a 300 MWe LMFBR [1, 2]. For the next decades a considerable increase in nuclear power generation is expected; a forecast till the year 2,000 is shown in Fig. 1.

Compared with nuclear power stations already working, the presently available reprocessing capacity is still very small. At the moment, only the 40 t/yr pilot plant WAK, in the neighbourhood of the Karlsruhe Nuclear Research Center (Kernforschungszentrum Karlsruhe, KFK) is in operation [3]. A large commercial reprocessing plant with a capacity of 1,500 t/yr is expected to start around 1982.

The conditions prevailing in the Federal Republic of Germany are not favourable neither for liquid storage of accumulated quantities of high-level wastes nor for discharge of radionuclides into the environment. Compared with the whole nuclear industry, reprocessing plants produce the largest amounts of radioactive wastes and by far the highest activities. Therefore, an extensive R + D program on management of these wastes is under way, covering mainly solidification of high-level fission product solutions (HLW) and the ultimate disposal of the resulting products but including also the treatment of particular low- and intermediate-level waste streams.



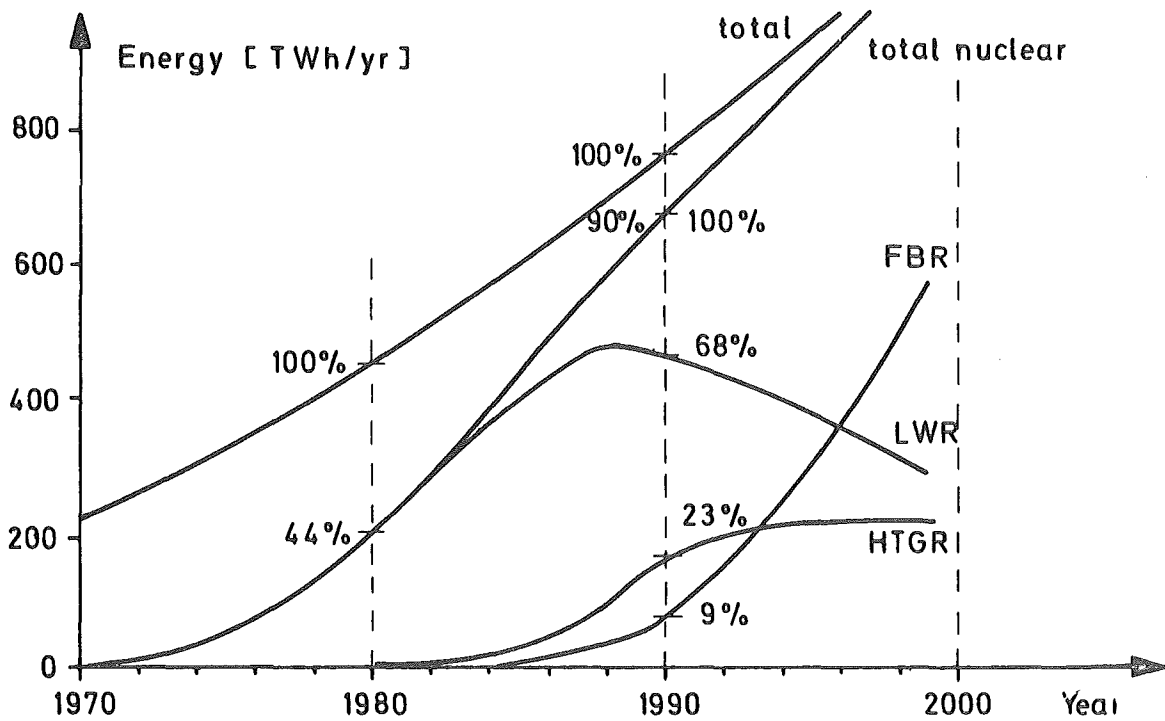
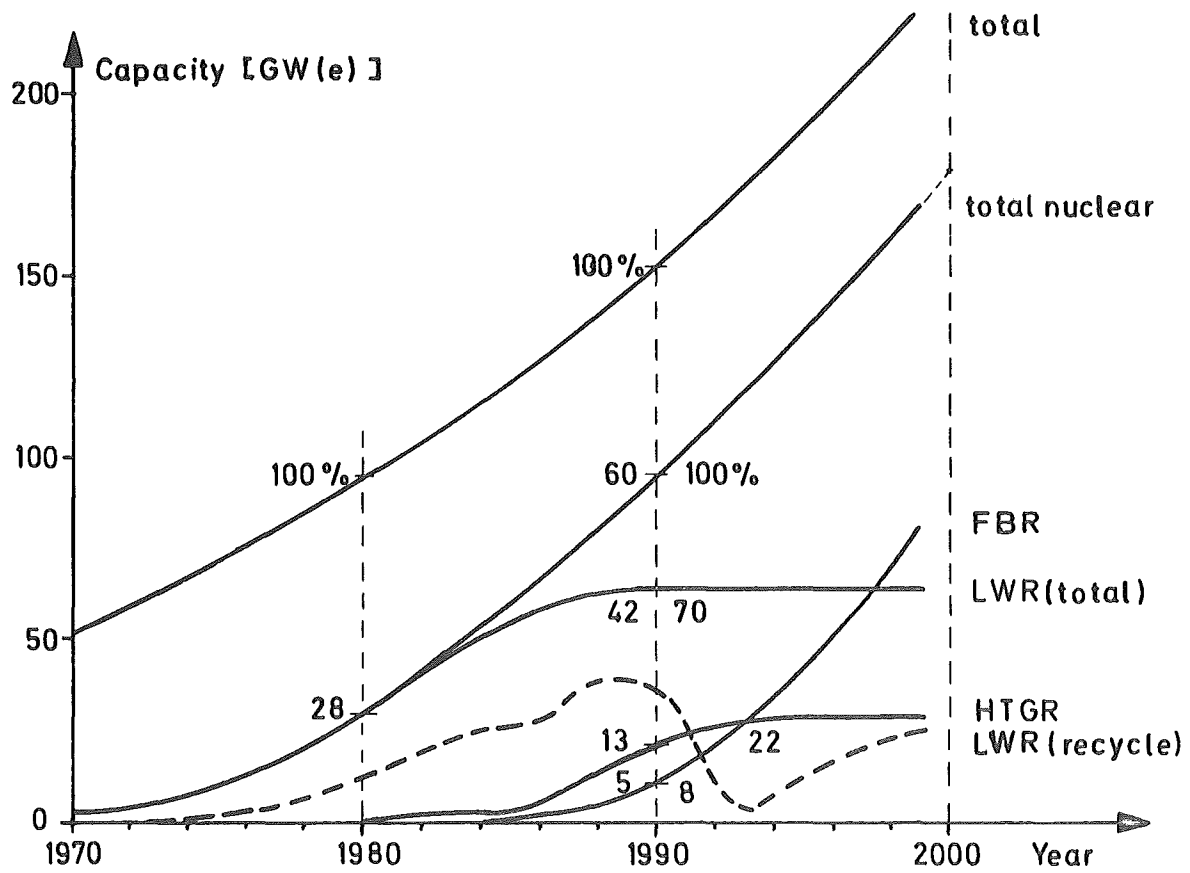


Fig.:1 Distribution of Power Plant Capacity and Energy Production in the FRG

This paper will give a survey on the German activities in the mentioned fields, dealing very briefly with topics which are presented as separate papers at this conference and in some more detail with topics which are not discussed here.

## 2. SOLIDIFICATION OF HIGH-LEVEL FISSION PRODUCT SOLUTIONS

### 2.1 HLW from the Reprocessing of Fuels from Water-Cooled Reactors

At the WAK-plant  $UO_2$ -fuels from boiling and pressurized water reactors with stainless steel- or zircaloy-cladding are reprocessed. A Purex process with chop-leach head end has been selected. Emphasis was put to the production of HLW with minimum salt content by application of uranium(IV) or gaseous compounds ( $NO_2$ ) or electrolytical reduction in place of ferrosulfamate and  $NaNO_2$  usually applied as redox reagents.

#### 2.1.1 Laboratory Experiments

At the KFK laboratory experiments have been carried out to develop a suitable glass for the incorporation of the HLW produced at WAK. This glass should present a relatively low melting point and a low corrosivity, thus facilitating somewhat the difficulties with the melting equipment. A low leach rate was desired but not obligatory because storage of the glass blocks in a salt deposit is anticipated and the virtual safety will be guaranteed by this type of ultimate disposal.

Under these aspects, a borosilicate-glass has finally been chosen. Besides the development of its optimum composition, studies have been performed on the glass properties, on the volatility of particular radionuclides, on the diffusion of radionuclides in the glass and from the

glass into rock salt, on the resistance to leaching, and so on.

In order to verify the results by experiments with original high-level wastes, a bench scale unit in a hot cell has been constructed which allows the production of glass blocks of roughly 500 ml volume and a total activity up till 20,000 Ci by a pot process. In this installation the studies mentioned above can be repeated on real high-level glasses and supplemented by investigations into the long-term behaviour of the glasses, their recrystallization, the energy storage, possible reactions between glass and salt, etc. Moreover, the efficiency of different types of off-gas cleaning will be tested [4].

#### 2.1.2 Incorporation of HLW by a Thermite Process

Although the properties of the borosilicate glass developed and intended for use in the first solidification campaigns of HLW are considered to be quite suitable, they are, however, not yet fully ideal. Therefore, as an alternative, another solidification method for HLW has been investigated at KFK. It is based on thermite processes to incorporate HLW into ceramic matrices. As a first step, denitration and spray calcination can be applied to transform the HLW into a dry calcined product which, afterwards, can be mixed with aluminum powder and an appropriate oxidant (e.g.  $\text{MnO}_2$ ), and ignited. By the reaction



temperatures well above  $2,000^\circ \text{C}$  are attained so that the mixture melts and forms, after cooling, a ceramic slag together with a metallic phase. The following composition

leads to a product of good quality:

|                  |        |
|------------------|--------|
| FP oxides        | 25 o/o |
| SiO <sub>2</sub> | 15 o/o |
| MnO <sub>2</sub> | 40 o/o |
| Al               | 20 o/o |

The high temperature opens a wide field of possible compositions so that products of better quality than the usual glasses can be obtained. The off-gas treatment is a major problem but could be solved to a large extent by proper selection of the reagents and layout of the off-gas system. It has been possible also to carry out the process continuously [4, 5].

Besides the good quality of the final products a further advantage of the thermite process is its high reaction velocity leading to very small units even for high throughputs.

### 2.1.3 Fundamental Investigations

In addition to the laboratory experiments performed at KFK concentrating, above all, on the development of a suitable glass, fundamental investigations are being made at the Hahn-Meitner-Institut (HMI) Berlin. Here, among others the process of devitrification is studied in detail; to enumerate some of the activities, the basic investigations of the kinetics of the crystallization process, the size of the crystallites and the chemical composition of the various phases should be quoted. Some crystalline phases have already been identified and characterized.

Fundamental investigations into the leaching process are under way, too. Here, mainly the leaching mechanisms and kinetics are studied. One of the results is, that leaching can be described by a  $\sqrt{t}$  formula that can be improved significantly by adding a positive linear term. Apparently the leach process is not exclusively controlled by diffusion. Another process seems to be dissolution [6].

#### 2.1.4 Development of a Technical Solidification Process

Besides the laboratory experiments some work is done at KFK on development of an appropriate technical process for the solidification of HLW. From the beginning, a process was favoured which could be operated continuously and could be scaled up to high throughputs without requiring a multiplicity of parallel units. On this basis, a combination of a spray calciner with an induction heated furnace was finally chosen. In this process the HLW is first denitrated by reaction with formic acid. The resulting solution, in which the nitric acid has been decomposed completely and part of the nitrates converted into oxides or other compounds, is fed into the spray calciner. There it is transformed into a dry powder, whereby another part of the fission product nitrates is calcined into oxides. The calcined powder is separated by sintered metal filters and drops into an induction-heated melter together with glass-forming additives. From there the melt flows into the final storage containers. The calciner is heated with steam which is injected inside the calciner and recycled after filtration and reheating.

For about two years the process has been tested in an inactive pilot plant with simulated HLW. Results obtained so far are very promising. Difficulties encountered at the beginning with the spray nozzle, the electrical steam heaters, the filter system and some other parts of the installation were solved in the meantime.

The principle applied with the internal heating is considered to be very advantageous. As the wall temperatures are relatively low, the risks of product crust formation and of corrosion are greatly reduced. The main advantage, however, seems to be the fact that contrary to units with wall heating (heat transmission by radiation) internal heating allows a scale-up to large throughputs [4c, 7].

A hot cell installation (VERA) is being constructed and will be available for demonstration of the process towards the end of 1976. Its capacity will be 20 litres HLW/h, thus allowing the solidification of more than the quantities of HLW generated at WAK. Thus, the operation of the VERA-plant should provide the experience necessary for the design and operation of a solidification plant for the large reprocessing plant anticipated for the eighties. Furthermore, the VERA-plant should allow to test new processes on a pilot scale and with real high-level wastes, as e.g., the thermite process mentioned above. To accomplish the different tasks, VERA is being conceived as a multi-purpose installation characterized by mounting on racks the process components.

## 2.2 Fission Products from HTR

In relation with its high-temperature gas-cooled reactor development program, Kernforschungsanlage Jülich (KFA) is

engaged with the thorium fuel cycle. The most advanced procedure for reprocessing this type of fuel is characterized by the following steps: Combustion of the graphite matrix, dissolution of the resulting oxides, separation and purification of the fissionable material by TBP-extraction similar to the Purex-process. However, there is a marked difference relative to usual Purex-wastes due to the high fluoride and aluminum nitrate content.

At KFA, it is planned to store the HLW for a period of about 5 years. After this, it should be denitrated by reaction with formaldehyde and then concentrated by evaporation. After mixing with glass-forming additives ( $\text{SiO}_2$ ,  $\text{CaO}$ ,  $\text{Na}_2\text{CO}_3$ ,  $\text{B}_2\text{O}_3$  etc.), the mixture is fed onto a drum-drier where the dry product is scraped off by an alumina blade and transported into a graphite crucible where it is fused by induction heating [8].

Alternatively, a phosphate glass process (PHOTHO) is developed at KFA by the Gelsenberg company for the solidification of thorium-bearing waste [9].



### 3. FINAL DISPOSAL OF SOLIDIFIED FISSION PRODUCTS IN A SALT DEPOSIT

In the Federal Republic of Germany, the disposal of radioactive wastes in rock salt formations is considered to be one of the most promising, safest and best established methods. Therefore the abandoned salt mine Asse II near Brunswick was acquired by Gesellschaft für Strahlen- und Umweltforschung (GSF) München. After repair of some above ground and underground installations, about 4,000 m<sup>3</sup> of solid and solidified, respectively, low-level radioactive wastes have been disposed of in the old mined-out rooms since 1967 in the framework of an experimental program. The low-level waste drums are simply stacked one above the other. Medium-level wastes have been stored since summer 1972 by lowering them from shielding casks into a special room [10-12].

Due to the release of decay heat, the solidified high-level wastes cannot be stored in large piles. They have to be placed in single bore-holes located at sufficient distance from each other. Only in this way the appropriate dissipation of the decay heat through the salt is guaranteed. As in salt domes or saddles like the Asse structure the base surface is small compared to the total volume, deep bore-holes are necessary if optimum utilization of the total volume of the salt formation is desired. However, the depth of the bore-holes is limited by heat dissipation requirements.

To investigate these problems, a large computer program is being carried out at the Technical University of Aachen.

The maximum temperatures to be expected in the glass cylinders and in the salt at different distances have been calculated as a function of fission product content and age, diameter of glass blocks, depth of the bore-holes, distance of the bore-holes from each other and some other parameters. Some of the results are given in a separate paper read at this conference [13]. The calculations have shown that the disposal of solidified HLW is principally possible within allowable temperature limits for the glass and the salt.

The results of the calculations have been checked by GSF in field experiments with electrical heaters. Good agreement was attained. Calculations and experiments will be continued to establish the optimum storage conditions. Besides these experiments, the plastic deformation of the salt in the mine at elevated temperatures, and some other problems have been investigated.

The technical installations for transport and storage are being developed by KFK. The loading system will be similar to that employed for the storage of intermediate-level wastes. The glass blocks (volume about 25 litres, diameter 20 cm) will be transported through the shaft into the mine in shielding casks, the weight of which will be about 10 tons being the maximal load of the shaft. In the storage area, situated at a depth of 775 m the shielding casks will be positioned on the radiation protection slide of the bore-hole. After the bottom of the shielding container and the slide of the bore-hole have been opened, the glass cylinder can be lowered. Once the bore-hole is filled about 5 m below ground level, the remaining volume will be filled up with crushed salt. Since at the relatively high storage-temperatures enhanced plastic flow of

the salt is occurring, the glass cylinders will be tightly imbedded in the salt after a short period of time.

The first experimental storage of real high-level waste is anticipated for 1976/77. During a period of several years the consequences of this operation can be observed and compared with the results of the calculations and the inactive experiments thus leading to the definition of optimum conditions for a safe storage. This program is considered to provide sufficient supporting data before large amounts of solidified HLW will have to be disposed of towards the mid eighties [13].

#### 4. TREATMENT OF MISCELLANEOUS WASTES ARISING IN REPROCESSING PLANTS

##### 4.1 Incorporation into Bitumen of Effluents with High Nitrate Contents

As all low- to intermediate-level liquid effluents generated at KFK are evaporated and the experience with this practice has been very satisfactory, the waste streams arising at WAK have been treated by the same method. During the first year of operation of the WAK-plant the increase of the discharge into the river Rhine amounted to only 100 mCi of B/ $\gamma$ - and 18 mCi of  $\alpha$ -emitters. However, in reprocessing plants many of the low- and intermediate-level effluents contain large amounts of nitrates which finally are to be found in the evaporator concentrates. These concentrates at KFK are incorporated into bitumen since this process has proven to be advantageous from many points of view (e.g. volume reduction, resistance to leaching, and even costs in the case of large throughputs). As a consequence, some investigations are carried out to study the problems arising by the bituminization of concentrates with high nitrate contents [4, 14].

##### 4.1.1 Combustion Tests

The burning points of several types of bitumen and of 70 mixtures of bitumen with salts (up to 60 wt.o/o of nitrates) have been determined. It turned out that the burning points of bitumen products with high nitrate contents were lower by about 100<sup>o</sup> C compared to pure bitumen but that normally no fire hazard exists. Besides the labora-

tory studies, tests with large batches (60 - 220 kg,  $\text{NaNO}_3$ -content 40 - 60 o/o) in open and closed containers were performed. The combustion was smooth at the beginning and became more violent later; however, in no case any explosions ever occurred.

For a quantitative and quick oxidation of bitumen a sodium nitrate content of about 87 wt.o/o would be necessary. In our bituminizing installation the maximum salt content attainable is about 60 o/o. A comparison of the reaction energy of these bitumen/sodium nitrate-mixtures with those of several propellants and explosives shows clearly that mixtures of 40 o/o bitumen with 60 o/o sodium nitrate should not be classified into the category of explosives.

Furthermore, the resistance of bitumen/sodium nitrate-mixtures to mechanical or thermal shocks and to shock waves of explosives was studied. The highest load (4 times higher than a shock caused by a drop on its edge of a 200-litre cask filled with the bitumen product from a height of 40 m) was generated by shock waves. In none of the experiments an explosion occurred [15].

#### 4.1.2 Leach Tests

The leaching behaviour of bitumen samples of different compositions simulating actual wastes (salt content approximately 40 wt.o/o) was investigated over a period of one year. The samples showed an average leaching rate of  $5 \times 10^{-4} \text{ g/cm}^2 \cdot \text{d}$ . The lowest value was  $1 \times 10^{-5} \text{ g/cm}^2 \cdot \text{d}$ , the highest  $2 \times 10^{-3} \text{ g/cm}^2 \cdot \text{d}$ . The experiments, however, have shown that very often the leaching of bitumen/salt

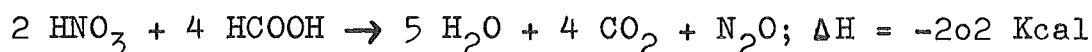
mixtures is not proportional to  $t^{0.5}$ . For one and the same sample different values were found for the power of  $t$  in different leaching time intervals. It turned out that products containing coarser crystals were leached more easily than products with finer crystals. Some samples began to swell while kept in distilled water, especially those which contained soda. When such samples (38.5 o/o soda) were stored in 3 o/o NaCl solution, they started floating in the leaching solution within half a year. Products of high salt content coated by a layer of 5 mm pure bitumen and products of low salt content ( $\leq 1$  wt.o/o) are up till now completely resistant to leaching (period of observation 2 1/2 years).

#### 4.1.3 Irradiation Tests

Samples of distilled and blown bitumen and bitumen/nitrate-mixtures were irradiated with 10 MeV electrons and  $\gamma$ -rays (fuel pond) up to  $5 \times 10^8$  rad. In mixtures of blown bitumen with 50 o/o  $\text{NaNO}_3$  the softening point rised by  $45^\circ$  C, in mixtures of distilled bitumen with 50 o/o  $\text{NaNO}_3$  by  $7^\circ$  C. After the irradiation both products showed a porosity of about 30 o/o and an equivalent decrease of density. Irradiation of samples of bitumen and bitumen/nitrate-mixtures enclosed in ampoules (300 Torr argon) to  $10^8$  rad led to the formation of some radiolysis gases, mainly hydrogen. Calculations show that a bitumen product with a specific activity of 1 Ci/litre (0.5 years old FP) will receive a total dose of about  $2.5 \times 10^7$  rad. From the above mentioned experiments it results that this causes an accumulated hydrogen formation of 18 litres in a 175-litre drum. Calculations show that the storage of such drums in a room (25 o/o of volume filled) would lead to a hydrogen content of 3 o/o in the air, assuming complete release and no ventilation.

#### 4.2 Decomposition of Nitric Acid and Nitrates

The large amounts of waste nitric acid arising in reprocessing plants lead to large amounts of radioactive residues if evaporation is applied and preceded by neutralisation. For this reason, a process was worked out at the KFK for the decomposition of nitric acid and nitrates by reaction with formic acid at boiling temperature. As is shown by the formula



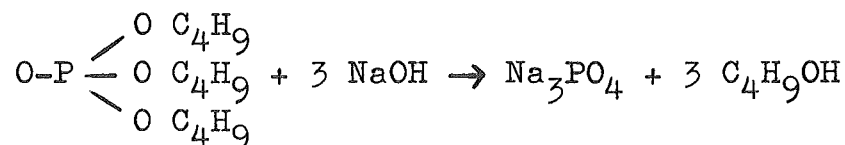
besides water, all reaction products are gaseous and can be discharged through the stack after mechanical filtration. Only toward the end of the process a few percent of nitrous fumes are formed. The reaction can be carried out both batchwise and continuously. The aqueous phase can be evaporated practically without production of residues.

Experiments have shown that the reaction described can be used successfully for removal of higher nitrogen oxides from off-gases, too. A further application can be found in the HLW solidification process. There the thermal decomposition of the nitrates at high temperatures can be replaced by the chemical decomposition. As a result, the volatility of ruthenium which in some processes can increase to as much as 70 o/o could be lowered to much less than 1 o/o [4, 16].

As the reaction between formic acid and nitric acid is stoichiometric, it can be used to adjust every desired pH in different processes (e.g. separation of individual nuclides [17], lowering of nitric acid content in HLW before tank storage, etc.) without any addition of salts.

### 4.3 Treatment of Spent Solvent

Another waste arising in reprocessing plants are tributylphosphate (TBP)-containing spent solvents. To treat them, several processes have been developed at KFK [4, 18]. The first is the saponification of TBP by reaction with NaOH following the formula



If concentrated NaOH is applied, the reaction time is less than 7 hours. After saponification, two liquid phases are formed. The lower phase contains water, the soluble salts and almost all of the radionuclides and can be incorporated into bitumen. Both the upper phase, the diluent, and the distillate consisting of butyl alcohol are nearly inactive; the alcohol can be burnt, the diluent can be burnt, too, or after appropriate decontamination recycled. The flow-sheet of the process is shown in Fig. 2.

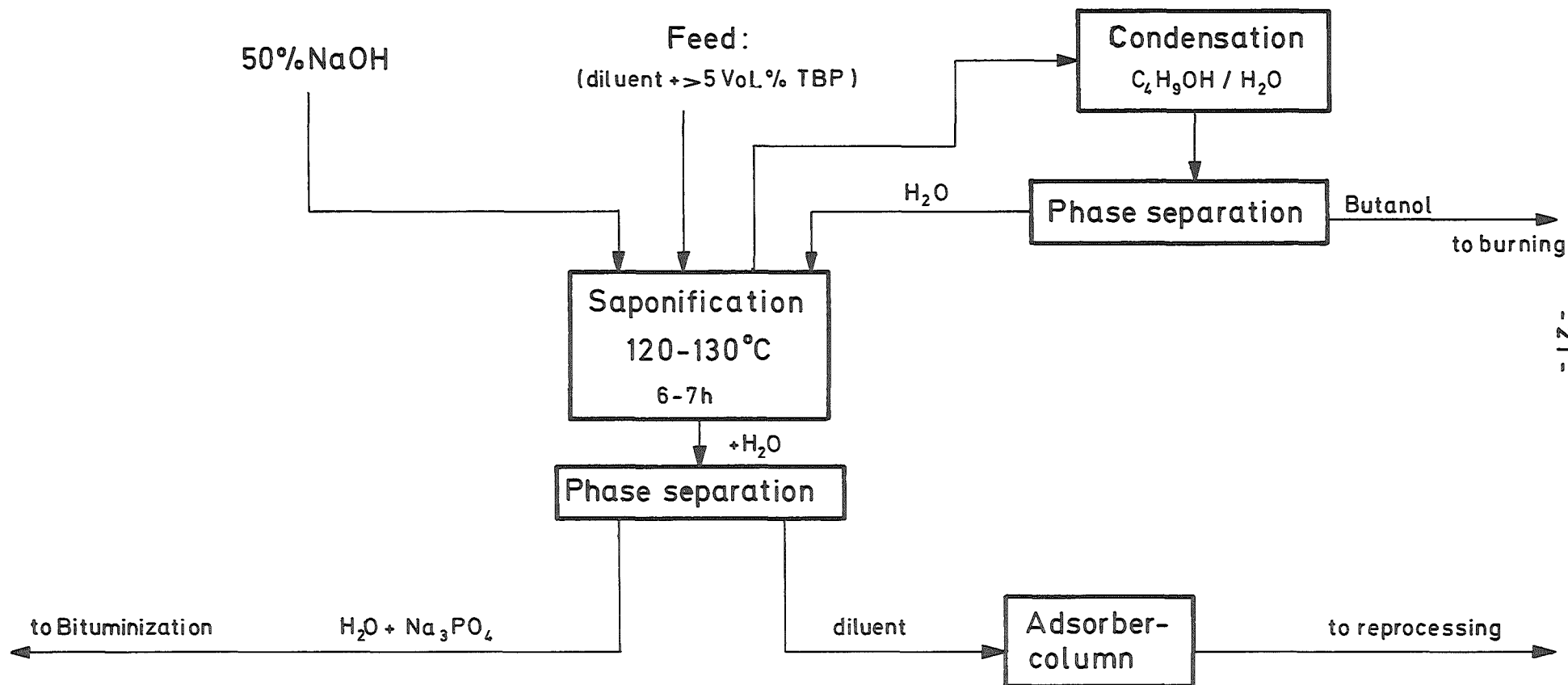
Another method worked out at KFK is the extraction of TBP with concentrated phosphoric acid, which leads to the formation of TBP.  $2 \text{ H}_3\text{PO}_4$ . This adduct settles as a heavy phase which after separation from the diluent can be decomposed by addition of water. Two phases are formed again and separated. The lower phase, i.e. diluted phosphoric acid, contains the bulk of the radionuclides and can be treated as aqueous waste. The upper phase, TBP, still contains some radionuclides and can be solidified by mixing with granulated PVC or other waste plastics. At ambient temperature, the formation of solid blocks in 200-litre drums takes a few days, at higher temperatures a few hours. The diluent, separated within the first step, still contains



Chemical reaction:



Process:



**Fig. 2 Saponification of Tributylphosphate (TBP) in hydrocarbon diluent**  
( Block diagram )

some radionuclides, mainly complex ruthenium compounds. It can be purified very effectively by passing it through a column with suitable adsorbents. Decontamination factors of  $10^6$  have been achieved, thus allowing reuse of the solvent. The flowsheet of this procedure is shown in Fig. 3.

#### 4.4 Disposal of Tritium Containing Effluents by Injection into Aquifers

The processes known and applied today for the treatment of radioactive effluents do not involve the separation of tritium. This could only be done by fractionated distillation, but even if this expensive process would be applied, the problem of disposal of the enriched tritium fraction would still have to be solved. As the discharge into a river of all the tritium arising in a large reprocessing plant would certainly raise local problems, the disposal of tritium into an aquifer is studied at KFK. There, some isolated oil lenses are to be found in the very neighbourhood of the Center in depths between 900 and 1,300 meters, some of which are already exhausted. One of the latter is actually being prepared for the injection of tritium containing effluents from the WAK-plant. The first injection is expected to start within the next months [19].

As this type of disposal does not lead to any contamination of the environment, it is one of the favoured methods for the future. Geologic formations which are suitable in principle for aquifer storage are to be found in several parts of the Federal Republic of Germany.



#### 4.5 The Removal of Noble Gases and Iodine from Off-Gases

The discharge of the noble gases to the atmosphere does not lead to serious world-wide problems in the next future; however, under unfavourable circumstances it could result in increased dose rates in the vicinity of large reprocessing plants. For this reason, some investigations into the separation of noble gases are being made in the Federal Republic of Germany.

A very promising method to remove krypton, xenon and argon besides the other contaminating fission products from the burner and dissolver off-gases produced in the reprocessing of HTR-fuel elements has been developed at KFA. The processes applied are based on liquefaction of the oxidised gas streams by compression and cooling, respectively, and subsequent rectification combined with adequate filter and absorber systems. The decontamination factors for the noble gases as well as for tritium, iodine and cesium are quite satisfactory [20].

A lot of work has been carried out at KFK to separate iodine from reactor off-gases by adsorption on charcoal, zeolites and other special inorganic adsorbers [21]. Recently, the investigations into the latter have been extended to the off-gas treatment in reprocessing plants [22].

## CONCLUSION AND OUTLOOK

WAK, the first small reprocessing plant in the Federal Republic of Germany has been operating for one year. Its radioactive effluents have been treated without difficulties by the Decontamination Department of the Karlsruhe Nuclear Research Center within the approved waste management system characterized by the following main features. The treatment procedures for low- and intermediate-level liquid effluents consist mainly in evaporation and allow to keep at very low levels the discharges into the environment. Conditioning of liquid concentrates and slurries as well as all types of low- to intermediate-level solid wastes results in high volume reductions and final products suitable for ultimate disposal.

For the intended future solidification of HLW a process is being developed which can be scaled up to large throughputs and leads to a chemically stable and sufficiently insoluble glass product.

Disposal of low- and intermediate-level solid and solidified wastes in the Asse salt mine is being practised; the preparations for the first experimental storage of the high-level glasses scheduled for 1977 are in progress.

On the basis of these activities it should be possible to handle safely also the large amounts of reprocessing wastes anticipated for the eighties. Nevertheless, R + D work will be continued to further improve the above mentioned processes and adapt them to large throughputs as well as to complete recent developments. In the last field we should cite, e.g., the thermite process for solidification

of HLW, the removal of radioactive noble gases and iodine from off-gases, and the disposal of tritium. Special emphasis will be put on studies related to the treatment and disposal of  $\alpha$ -bearing wastes. In case that THTR reactors become more important in the Federal Republic of Germany the investigations into the solidification of their HLW have to be intensified. With respect to the anticipated large reprocessing plant an optimization study on its overall waste management is under way, comprising all aspects from waste production to ultimate disposal. In this context, its siting in the immediate neighbourhood of a salt deposit suitable for disposal of the radioactive residues is considered. Such a location would offer many advantages, e.g., considerable savings in waste transport cost and certainly simplifications in the treatment of some types of waste.

In addition to the above mentioned activities, a program is performed at KFK which is only indirectly connected with the waste management from reprocessing plants, but which might, however, have a certain influence on it. It relates to the separation of actinides from HLW in view of their later application as energy or neutron sources [16, 17]. Furthermore, investigations will be executed on the utilization of solidified HLW as a radiation source in the inactive sewage treatment. First experiments have demonstrated that an irradiation unit of approximately 2 MCi  $^{137}\text{Cs}$  would be sufficient to sterilize 1,000 m<sup>3</sup> per hour of sewage or sludge [23]. This sewage volume corresponds to that of a community with about 250,000 inhabitants. A rough calculation shows that in the mid eighties enough vitrified HLW would be available in the Federal Republic of Germany to sterilize all the sewages produced. As it was found that for irradiation doses of about  $5 \times 10^5$  rad the

chlorine splits off from polychlorinated phenols in aqueous medium [24], irradiation could be also applied for partial decomposition of biologically non degradable organic compounds in the wastes of some chemical, e.g., biocide industries. Should future results in this domain continue to be promising, a great field could be opened for the beneficial application of solidified HLW whose cooling time could be extended without additional cost, thus reducing the problems connected with the heat dissipation in ultimate disposal.

In conclusion, it can be stated that despite the small extent of the actual German reprocessing capacities, the waste management policy established has already been conceived to cope with the requirements of the future. The work done as well as the intended further developments justify the expectation that even the wastes of large reprocessing plants could be safely treated and disposed of at minimum release of radionuclides into the environment. On this basis full acceptance by the public could be anticipated.

REFERENCES

- [ 1] KEMPKEN, M., Verzeichnis der Kernkraftwerke der Welt, Atomwirtschaft 16 8/9 (1971) 487.
- [ 2] Stromerzeugung aus Kernkraftwerken - Nuclear electricity generation - Jun. 1972, Atomwirtschaft 17 9/10 (1972) A 300.
- [ 3] EITZ, A.W., RAMDOHR, H., SCHÜLLER, W., Die Wiederaufarbeitungsanlage Karlsruhe, Atomwirtschaft 15 2 (1970) 74.
- [ 4] Jahresberichte der Abteilung Dekontaminationsbetriebe
  - a) 1968, KFK-1030 (1970)
  - b) 1969, KFK-1346 (1971)
  - c) 1970, KFK-1500 (1972).
- [ 5] RUDOLPH, G. et al., Lab-scale R + D on fission product solidification by vitrification- and thermite-processes, Paper No. 24, this Conference.
- [ 6] HEIMERL, W. et al., Studies on the behaviour of radioactive waste glasses, Paper No. 20, this Conference.
- [ 7] GUBER, W. et al., Pilot plant experience on high-level waste solidification and design of the engineering prototype "VERA", Paper No. 19, this Conference.
- [ 8] HALASZOVICH, St. et al., Interim storage and solidification for THOREX-type fission product solutions, Paper No. 26, this Conference.
- [ 9] LAHR, H., Verfestigung hochradioaktiver Abfälle in Phosphatgläsern, NUCLEX 72, Nuclear Industries Fair, Basel 1972, Technical Meeting No. 9/19.
- [10] Studiengruppe für Tieflagerung, Jahresberichte 1964 / 1965 / 1966, KFK-357 (1965), KFK-481 (1966), KFK-663 (1967);  
Endlagerung radioaktiver Rückstände, Jahresberichte 1968 / 1969 / 1970 / 1971, KFK-1138/GSF T-25 (1969), GSF T-27/KFK-1212 (1970), KFK-1454/GSF T-34 (1971), GSF T-42/KFK-1585 (1972).



- [11] KRAUSE, H. et al., "Experimental storage of radioactive wastes in the Asse II salt mine", Disposal of Radioactive Wastes into the Ground(Proc. Symp. Vienna 1967), STI/PUB/156 (1967) 519.
- [12] ALBRECHT, E. et al., "Disposal of radioactive wastes by storage in a salt mine in the Federal Republic of Germany", Management of Low- and Intermediate-Level Radioactive Wastes (Proc. Symp. Aix-en-Provence 1970), STI/PUB/264 (1970) 753.
- [13] KÜHN, K. et al., Disposal of solidified high-level radioactive wastes in the Asse salt mine, Paper No. 35, this Conference.
- [14] KLUGER, W., KRAUSE, H., NENTWICH, O., Fixing of radioactive residues in bitumen, KFK-1037 (1969).
- [15] BACKOF, E., DIEPOLD, W., Untersuchungen über die thermische und mechanische Empfindlichkeit von Bitumen/Sauerstoffsalzgemischen, Institut für Chemie der Treib- und Explosivstoffe (ICT) Berghausen/Karlsruhe, Techn. Mitt. Nr. 9/70 (1970).
- [16] Projekt Actiniden, 1. Halbjahresbericht 1971 / 2. Halbjahresbericht 1971 / 1. Halbjahresbericht 1972, KFK-1456 (1971), KFK-1544 (1972), KFK-1656 (1972).
- [17] KOCH, G. et al., Recovery of transplutonium elements from fuel reprocessing high-level waste solutions, Paper No. 43, this Conference.
- [18] DROBNIK, S. et al., "Behandlung und Beseitigung radioaktiver Lösungsmittelabfälle aus der Wiederaufarbeitung bestrahlter Kernbrennstoffe", Reaktortagung, Hamburg 1972, Deutsches Atomforum e.V., Kerntechnische Gesellschaft im Deutschen Atomforum e.V. (1972) 264.
- [19] HILLD, W. et al., Deep-well disposal of tritium containing liquid effluents, Paper No. 39, this Conference.
- [20] LASER, M. et al., Off-gas treatment and krypton disposal in HTGR-fuel element reprocessing, Paper No. 3, this Conference.

- [21] WILHELM, J.G., Trapping of fission product iodine with silver impregnated molecular sieves, KFK-1065 (1969).
- [22] WILHELM, J.G., SCHÜTTELKOPF, H., An inorganic adsorber material for off-gas cleaning in fuel reprocessing plants, Paper presented at the 12th AEC Air Cleaning Conference, Oak Ridge, Tenn., Aug. 1972 (to be published).
- [23] SCHEFFLER, K., personal communication (1972).
- [24] GILBERT, E., personal communication (1972).