### R&D on Intermediate Storage and Final Disposal of Spent HTR Fuel

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## Abstract

The back-end of the fuel cycle concept for spent High-Temperature Reactor fuel elements in Germany is based upon intermediate storage in shielded casks in a surface facility followed by direct disposal in a deep repository. Two storage facilities are in operation, whereas disposal in a salt dome repository is being designed. R&D results obtained so far support the chosen concept and underline the special safety features of the fuel elements, i.e. the coated particle fuel stabilised in a graphite matrix, which is extremely resistant against all conceivable attacks during storage and disposal.

## 1    Introduction

Based upon a former development at the Forschungszentrum Jülich (FZ Jülich, Research Centre Jülich), two high-temperature gas cooled and graphite moderated reactors (HTR) had been operated in Germany: a) the 15 MWe AVR reactor from 1967 until 1988 in Jülich, and b) the 300 MWe Thorium High Temperature Reactor (THTR 300) from 1985 until 1988 in Hamm-Uentrop. The status of their decommissioning has been reported in Session I of this conference.

Both reactors have in total produced about 1 Million of spent fuel elements during their operating time. The typical fuel element is a tennis-ball sized sphere from graphite, containing up to twenty thousand pinhead-sized fuel particles containing oxide or carbide fuel each. The particles are surrounded by a high-porosity buffer layer to limit the internal pressure from swelling and gas production, and coated with a high-density pyrocarbon layer (BISO) or with a combination of two pyrocarbon layers with a silicon carbide layer in between (TRISO) to retain radionuclides (see FIG. 1).

The spent fuel management concept for HTR in Germany is based upon intermediate storage followed by disposal in a deep rock salt repository without reprocessing.

Techniques for the intermediate dry storage in CASTOR-type transport/storage casks are available and practised with AVR fuel in Jülich as well as THTR fuel in Ahaus. Experiences are reported in Session II of this conference.

For disposal, emplacement in horizontal drifts using shielded casks, or in deep vertical boreholes using simple packaging were chosen to be the most promising concepts /1/.

This paper summarises the results obtained so far, as well as R&D still do be done to support intermediate storage and final disposal of spent HTR fuel. It supplements previous reports, e.g. /2/ through /13/.
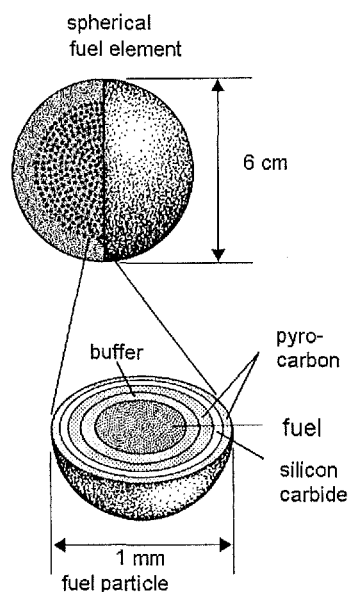


FIG. 1: Sectional view of a HTR fuel element

## 2    R&D work on intermediate storage

Principal goals of the activities on interim storage at FZ Jülich are to demonstrate the safety of dry storage and to provide data for the licensing of corresponding commercial storage facilities. Complementing the storage of AVR fuel, a research program was initiated to measure the release of gaseous radioactivity under realistic storage conditions. FIG. 2 shows the equipment in the hot cell of the AVR dry storage facility to measure the built up of the $^3H$, $^{85}Kr$ and $^{14}C(CO_2)$ concentration in the

gas atmosphere of a AVR dry storage can, filled with 950 fuel elements of the type GO[a] and connected to a gas sampling circuit. The storage temperature was 30 °C max. and the burn-up of the fuel 13-16 % FIMA. The specially designed dry storage can could be connected by remote handling to flexible tubes inside the hot cell. The build-up of the concentration was measured over several years. FIG. 3 - 6 show the trend of gaseous release.
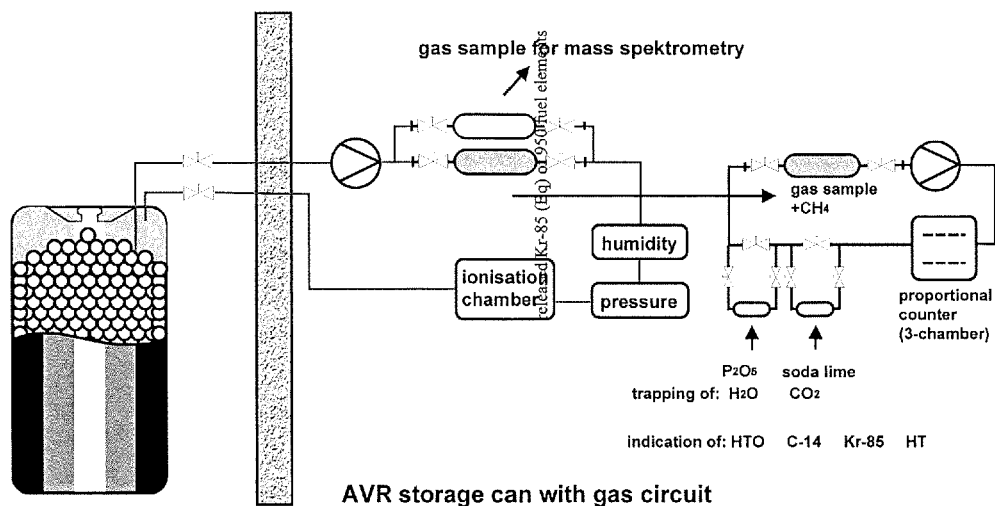


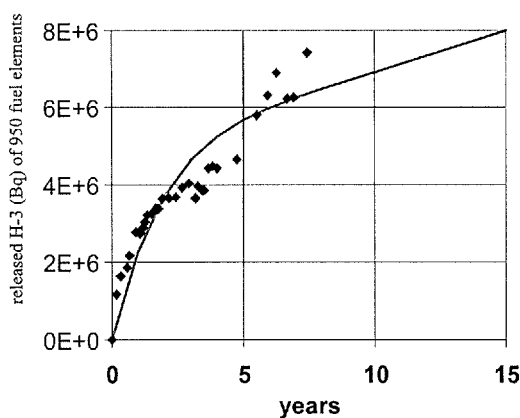FIG. 2: Release of gaseous activity during the storage of AVR fuel elements
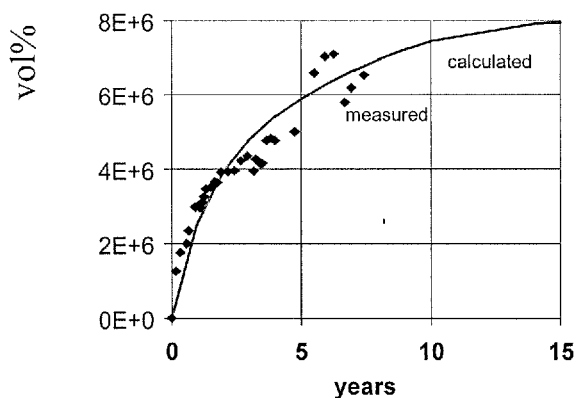


FIG. 3: Release of $^3H$
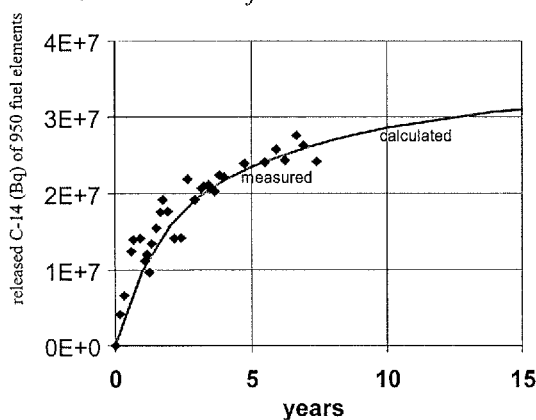


FIG. 4: Release of $^{85}Kr$
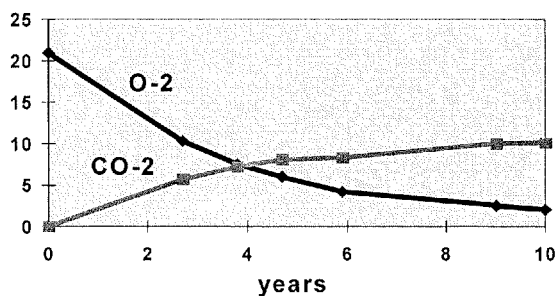


FIG. 5: Release of $^{14}C$



FIG. 6: Gas composition within a storage can

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[a] Compressed graphite matrix, oxide fuel
IAEA-TECDOC-1043 P.151-161

The $^3$H inventory of a fuel element is mainly generated in the graphite matrix by $^3$He and $^6$Li impurities. The release is controlled by absorption and desorption processes at the grain boundaries of the graphite and diffusion in the grains. The complete $^3$H inventory of a dry storage can amounts to about $2*10^{12}$ Bq.

The $^{14}$C inventory is mainly generated by (n-p)-processes of $^{14}$N impurities of the graphite. The release of $^{14}$C during storage is initiated by corrosion processes of the carbon by contact with air and gamma radiation. $CO_2$ will be generated. The inventory of 950 fuel elements amounts up to $7*10^9$ Bq. Only 1% can be released, until the oxygen content of a storage can is consumed.

The $^{85}$Kr inventory is generated by fission and amounts to $1*10^{13}$ Bq in a storage can. The release mainly depends on the number of defect particles. The specification of the fuel elements permits a defect rate of $3*10^{-4}$, that means a possible release of $3*10^9$ Bq. The extrapolation of the measurements shows a maximum amount of $1*10^7$ Bq.
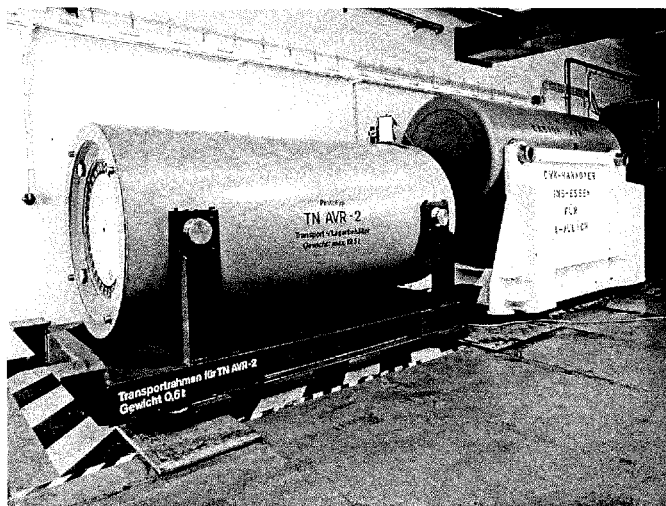


*FIG. 7: 2 prototype storage casks, each loaded with 1900 fuel elements (= 2 AVR-cans) burn-up: 12 - 16% fima*

Further measurements were performed at the two prototype transport and storage casks (FIG. 7). FIG. 8 and FIG. 9 shows the distribution of gamma- and neutron dose rate at the surface. Each of the casks was loaded with two dry storage cans, filled with 950 fuel elements of the type GO and GK[b] with high enriched fuel $(U,Th)O_2$ each. The burn-up of the fuel amounted to 13 - 16 % FIMA. The measurements were done after different decay times. One year after discharge (1990) the main part of the dose rate at the surface of the 30 cm thick steel wall of the cask was generated by $^{144}$Ce-$^{144}$Pr with the high gamma energy of 1,5 MeV. Fuel elements with lower burn-up (12%FIMA in the outer cans) produced due to the higher content of $^{235}$U more short-lived fission products like $^{144}$Ce. After several years of decay time, $^{137}$Cs is the dominating dose rate source caused by the higher burn-up (16% FIMA) of the inner positioned storage cans (FIG. 8).
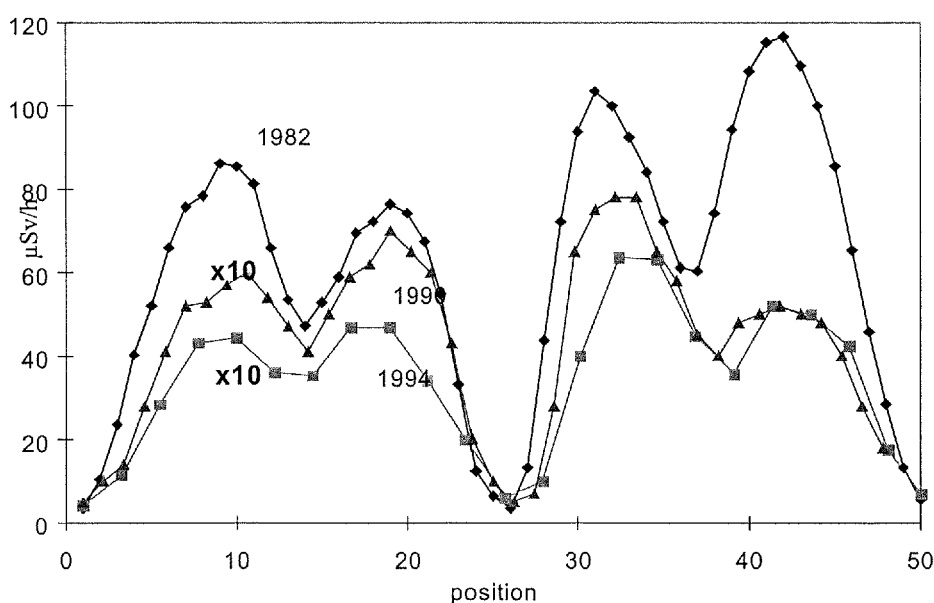


*FIG. 8: Gamma dose rate of AVR storage casks*

[b] compressed graphite matrix, carbide fuel
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FIG. 10: 50 to press to crush up to 9 spherical fuel elements (simulation of a mechanical pressure of 30 MPa)
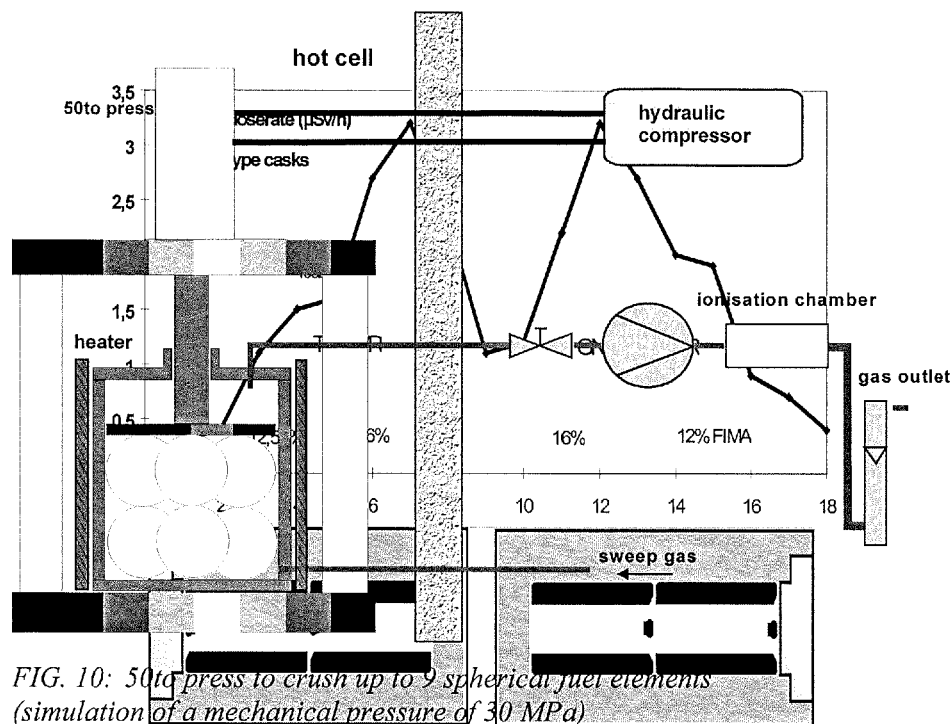
*FIG. 9: Neutron dose rate of AVR storage casks*

The neutron irradiation at the surface is essentially caused by the alpha decay of $^{238}$Pu. Due to the high portion of $^{232}$Th in the fuel (Th/U=5), a low concentration of $^{238}$Pu is breeded and accordingly a low level of neutron irradiation is measured (FIG. 9). The measurements were performed with the neutron REM counter of FAG. The distribution of the neutron dose rate shows very high values in the position of the two cans with the higher burn-up. This effect is caused by non-linear breeding of $^{238}$Pu with increasing burn-up.

## 3 R&D work on final disposal

The fuel elements of the HTR are proposed for final disposal without reprocessing. The aim of the disposal is the protection of individuals and the environment from the hazards of nuclear waste. Because of the long half-life of some radionuclides this protection must be guaranteed for time periods of hundreds or even thousands of years. One possibility to reach the protection aim is the disposal in a stable salt dome as geologic host formation. For this final disposal it must be distinguished between the normal disposal conditions and the hypothetical, but not absolutely impossible accident scenario of an water ingress into the repository.

### 3.1 Behaviour under normal disposal conditions

Under normal disposal conditions the repository will be dry. The rock salt will become plastic under the pressure of the overlaying strata and will thus creep into cavities not completely filled with waste. This will result in a high mechanical load on the fuel elements which may be crushed up if no countermeasures are taken. Appropriate countermeasures like e.g. grouting with cement have been developed and successfully tested /3/.To investigate the stability of the fuel elements a remote press with a maximum pressing power of about 30 MPa was installed in the hot cell facility "GHZ" of the FZ Jülich. The equipment consisted of a cylindrical steel vessel and the pressure piston. The vessel was swept with an inert gas and connected to a gas measuring circuit (see FIG. 10).

The release of $^{85}$Kr is an indication for the particle failure, because this fission gas is closed up in the coated particles. This $^{85}$Kr release can easily be monitored by means of an ionisation chamber. The fuel elements are crushed and densified until the cylindrical space of the pressure vessel is completely filled. The tests showed that particle failure depends on the coating design and the burn-up. However, the failure fraction observed remained below 1% (see FIG. 11) /4/. Coated particle failures may largely influence the long-term safety of a repository as far as the containment of long-lived radionuclides is concerned.

## 3.2 Behaviour under accidental conditions under the aspect of long-term safety

No radionuclide release can take place in a dry repository. Only if the radioactive waste gets into contact with the ground water radionuclides can be transported to the environment. Salt domes are geologically stable formations, which have been sealed from the ground water for more than $10^6$ years. Therefore these formations are considered as ideal for final disposal.

However, in different accident scenarios it is assumed, that ground water may penetrate into the storage field through little crevices in the anhydride layers, which may be part of the salt dome. This water will form saturated, high corrosive salt brines and after corrosion of the storage casks the brines will interact with the fuel elements. A large number of experiments to study the behaviour of HTR fuel elements in such salt brines were performed at the FZ Jülich starting in the late 70th /5, 6, 7/. A short review of the obtained results is summarised in this chapter.

Different types of AVR fuel elements[c] were exposed to Q-brine[d] /8/at temperatures between 50 and 90°C and pressures up to 13 MPa. The fractional release of $^{137}$Cs is shown in FIG. 12. The release from fuel elements with BISO particles is less than $10^{-4}$, related to the inventory of one coated particle respectively the inventory generated by the U-contamination of the matrix graphite. After 200-300 days the release rate decreases to a stable equilibrium controlled by the diffusion of $^{137}$Cs from the pyrocarbon kernel into the matrix. Calculations based on a diffusion/adsorption model with the GETTER code match the experimental results very well. For the long time calculations the release of $^{137}$Cs can be described as a instantaneous release of about 20% of the matrix inventory, which is related to the amount of kernels with a defect in the coating, and a slow release of the remaining $^{137}$Cs in the graphite particles over a time period of several 100 years /4, 6, 9/.
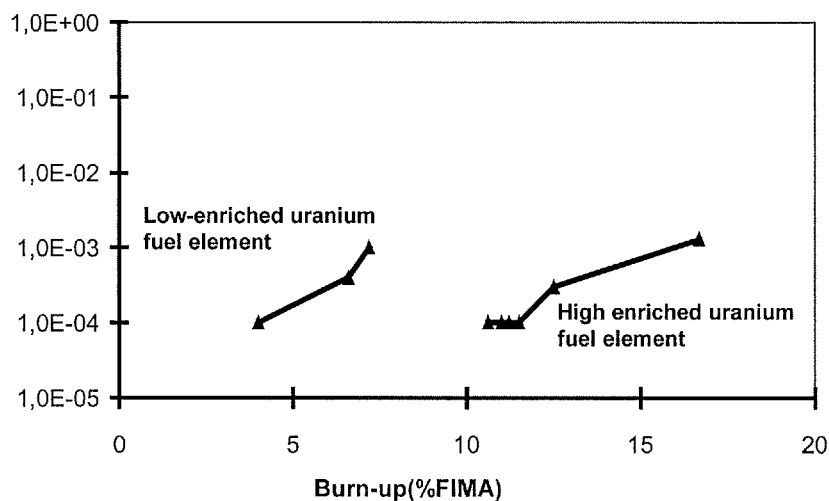


*FIG. 11: Fractional failure of coated particles after crushing the matrix*
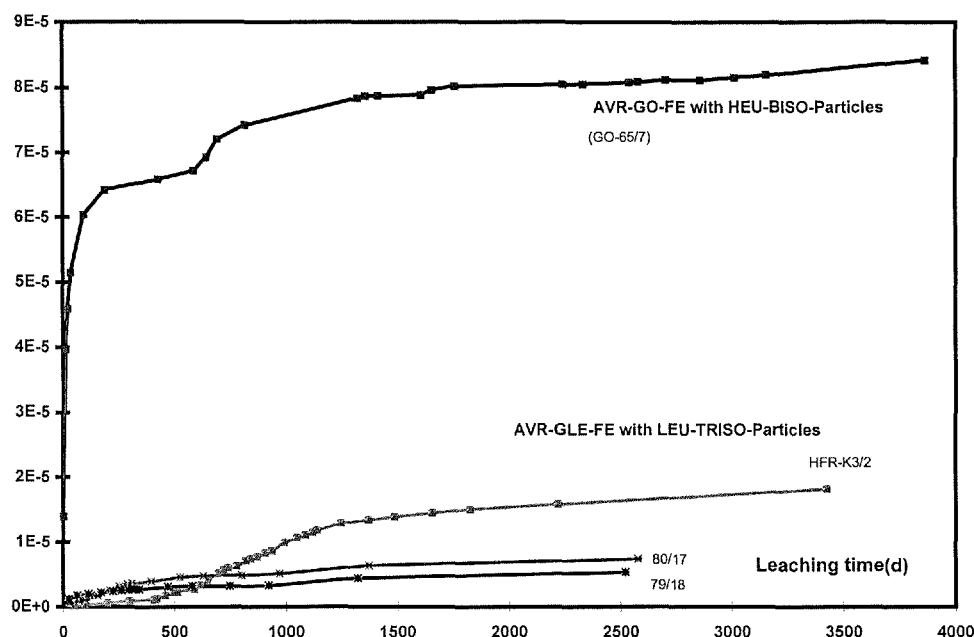
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*FIG. 12: Fractional release of $^{137}Cs$ inventory in Q-Brine at 90 °C/13 MPa*

The comparable low fractional release of the fuel elements with TRISO-coated particles is caused by a corresponding lower U-contamination of the graphite matrix during fuel manufacturing, and therefore the long time behaviour of the fuel elements with TRISO particles is even more convenient.

For radionuclides other than cesium the leaching rates are presumed to be lower because of their lower mobility. The following experiment has proved this for the long-term relevant nuclides $^{99}Tc$ and $^{237}Np$ /10/. A bore in a graphite sphere had been filled with 400 µl of a radionuclide solution. Then the hole was sealed with a screw and the sphere were immersed in Q-brine or water at 90°C and a pressure of 15MPa. Table 1 shows the breakthrough time of different nuclides. Although no breakthrough had occurred for technetium after 400 d, a permeability of technetium can not be ruled out with regard to the long time scale of a final repository; moreover the breakthrough can be forced by a cyclic change of the pressure from 100 kPa to 15 MPa.

*Table 1: Breakthrough time of different radionuclides*

| Nuclide | Leachant | | |
|---|---|---|---|
| | Water (d) | Q-Brine (d) | Q-Brine pressure cycled (d) |
| $^{99}Tc$ | 90 | > 400 | < 4 |
| $^{137}Cs$ | 20 | 20 | 10 |
| $^{237}Np$ | 150 | 150 | 30 |

The radionuclide release depends not only on the diffusion of the nuclides through the graphite matrix. The basic process is the dissolution of the radionuclides in the fuel kernels, which differs for the fuel matrices $UO_2$ or $(Th,U)O_2$. A release from intact coated particles didn't occur, but the fuel elements contain between $10^{-4}$ and $10^{-5}$ defect particles from the production process. This rate increases by a factor of approximately ten for highly irradiated material. Therefore, the source term for radionuclide release is mainly influenced by the number of broken coated particles. To investigate the behaviour of the irradiated fuel, coated particles were collected from irradiated, electrochemically disintegrated fuel elements. The particles were carefully point-loaded until the coating cracked. The single kernels were leached with Q-brine in air at 20°C and at 90°C, respectively, and 100kPa or 13MPa, respectively. The following Fig. 13 and FIG. 14 show the release rates of different radionuclides from the two fuel matrices /7/.
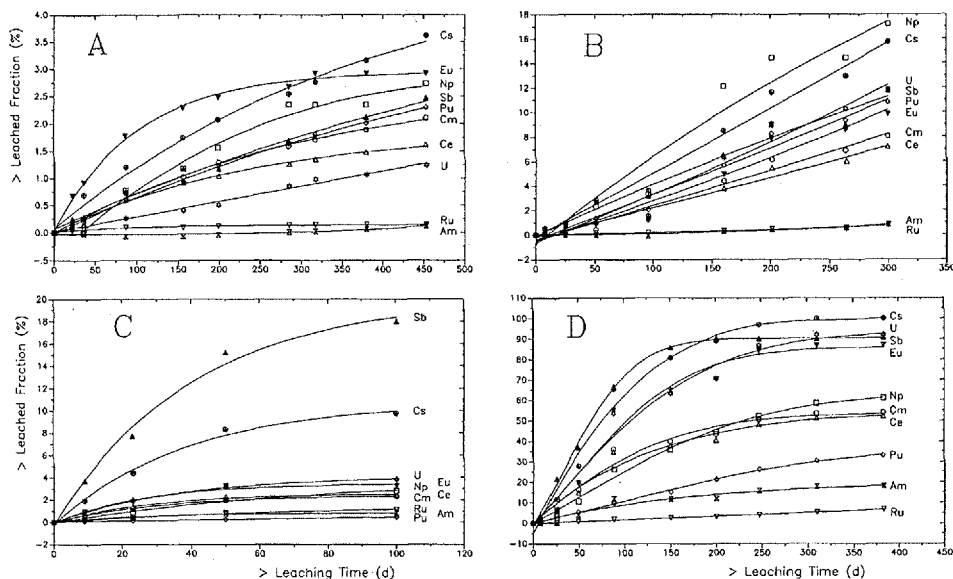
FIG. 13: Radionuclide leaching from UO₂ kernel with Q-Brine
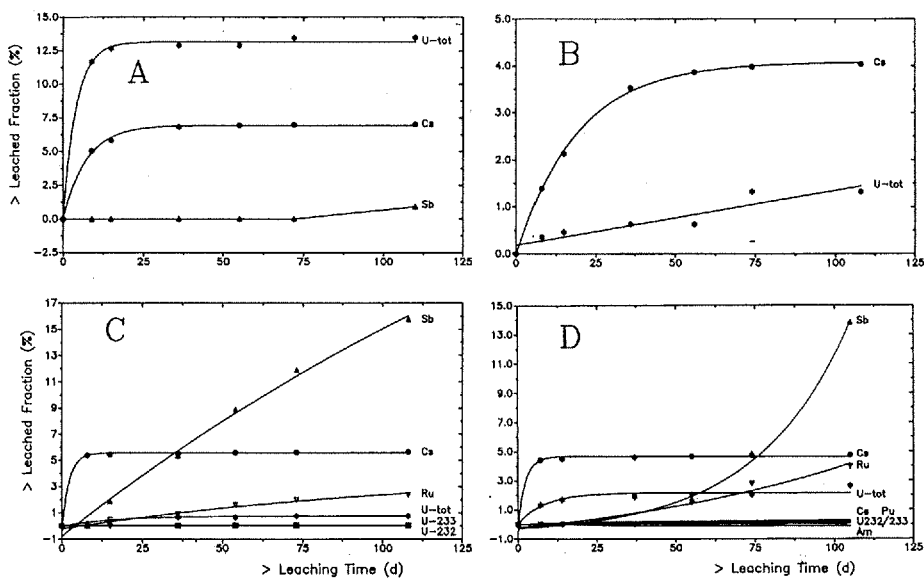A: 20°C/100 kPa   B: 20°C/13 MPa   C: 90°C/100 kPa   D: 90°C/13 MPa



FIG. 14: Radionuclide leaching from (Th,U)O₂ kernel with Q-Brine
A: 20°C/100 kPa        B: 20°C/13 MPa    C: 90°C/100 kPa    D: 90°C/13 MPa

The high leaching rates from the $UO_2$ fuel cannot be explained by the metal oxide's solubility in water only. The main is very probably the combination of weakening of the $UO_2$ during irradiation by fission product build-up, mainly at the grain boundaries, and the very aggressive salt brines. In contrast to the $UO_2$ the $ThO_2$ withstands such an attack /7/.

In conclusion the amount of leachable radionuclides is largely dependant upon the uranium contamination of the fuel element during manufacturing and the failure of the particle coating. Therefore the corrosion of the coating is a central problem for the long-term safety. The corrosive attack of the coating by Q-brine was investigated by the following experiment /4/. The top of an unirradiated particle was cut off and then fixed with the intersection towards an irradiated particle in order to produce a typical radiation field. The space between these particles was filled with Q-brine (FIG. 15). After a leaching time of about 2 years the state of the coatings at the intersection has been investigated by optical and electron microscopy. No indication of a corrosive attack could be detected, as it is shown in FIG. 16. However, with respect to the time scale of final disposal this result does not rule out any corrosive process. Therefore further investigations are necessary to describe the long-term behaviour of the coating under disposal relevant conditions.
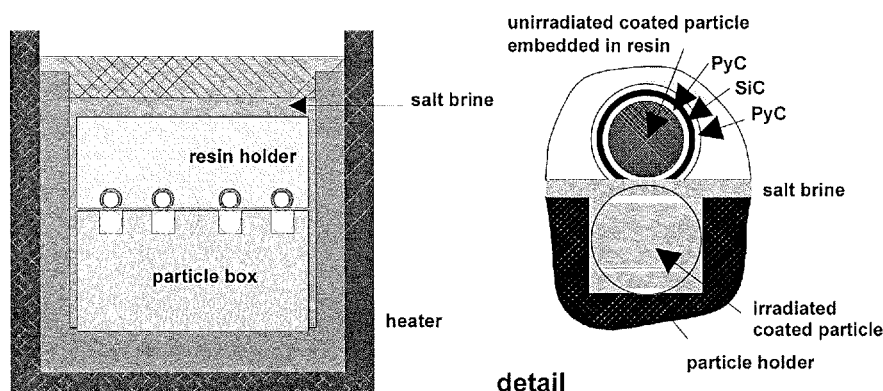


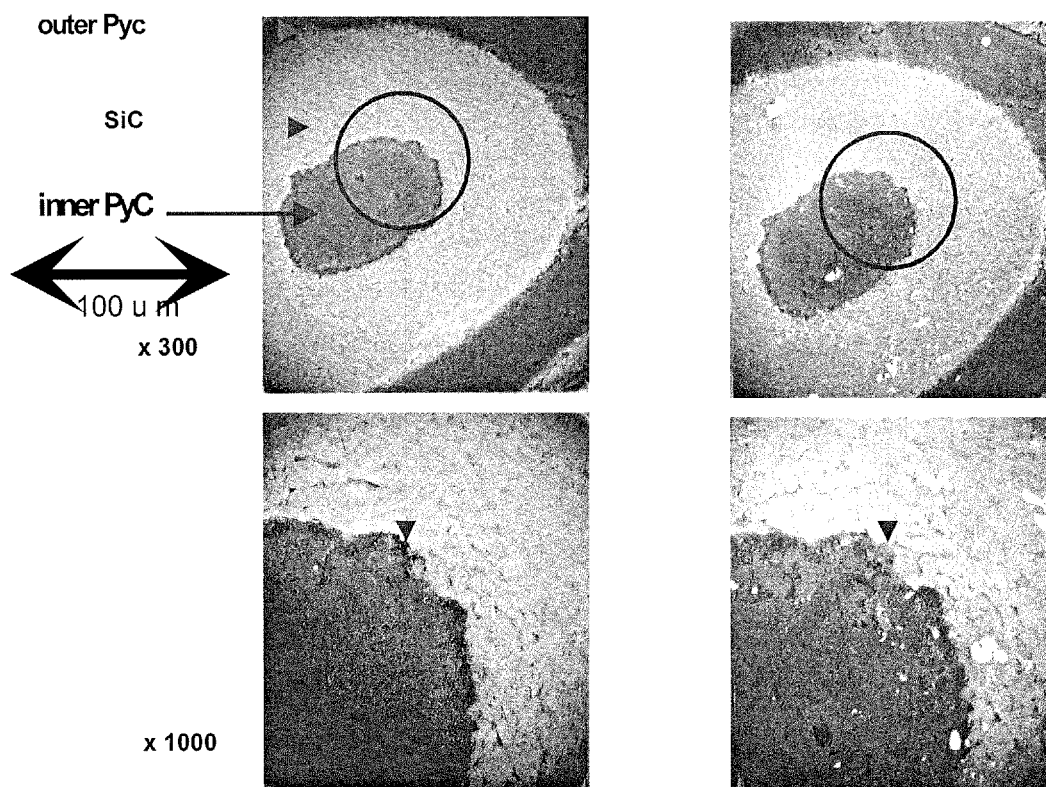FIG. 15:    Leaching of a polished particle with realistic radiation effects



FIG. 16: SEM micrographs of a polised TRISO particle at the beginning (left) of leaching and after 2 years (right).

An additional problem of final disposal is the production of hydrogen by either radiolysis of water or corrosion of metals. This hydrogen may increase the release of radionuclides to the environment by pressing contaminated brine out of the repository. A set of experiments concerning the formation of hydrogen by radiolysis has recently been finished /11, 12, 13/.

Different irradiated fuel elements were exposed to Q-brine under argon or air atmosphere in a spherical autoclave (FIG. 17). The gap between autoclave wall and fuel element had a thickness of 1 or 2 mm, respectively. The experiments were performed at 22 or 55°C brine temperature. A gas plenum with a pressure gauge for continuos measuring was located above the autoclave. Gas samples were taken with an attachable gas sampling tube to analyse the gas composition by gas chromatography and radio gas chromatography.



FIG. 17: Spherical autoclave

These experiments had the additional aim to determine the release of $^{14}C$ in gaseous form of $^{14}CO_2$ and solved in the brine. The $^{14}C$ is mainly formed by an (n,p) reaction of the nitrogen impurities in the cooling gas helium. The fresh $^{14}C$ is absorbed at the graphite matrix and due to the high temperature mounted into the crystal structure of the graphite. This radionuclide is important for the long-term safety because of its long half-life together with its different chemical behaviour in comparison to the other, mostly cationic radionuclides. Moreover, it acts as an indicator for the corrosion of the graphite matrix. Therefore brine in- and outlet were attached to the autoclave. The brine was replaced in the same time intervals as the gas samples had been taken.

The following diagram (FIG. 18) shows the pressure build-up for the different experimental conditions.
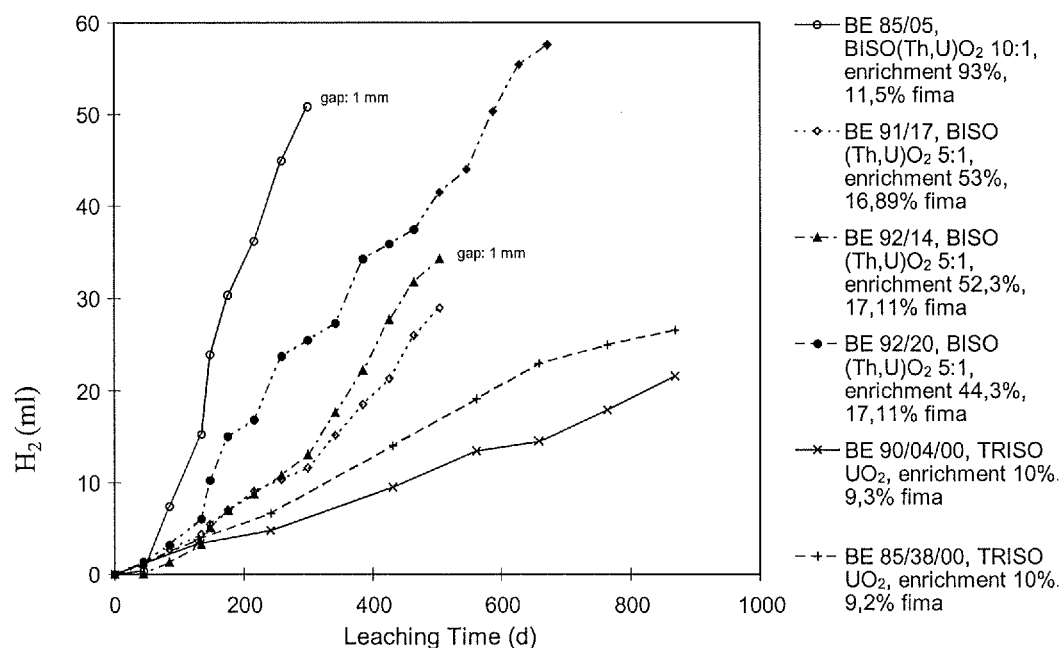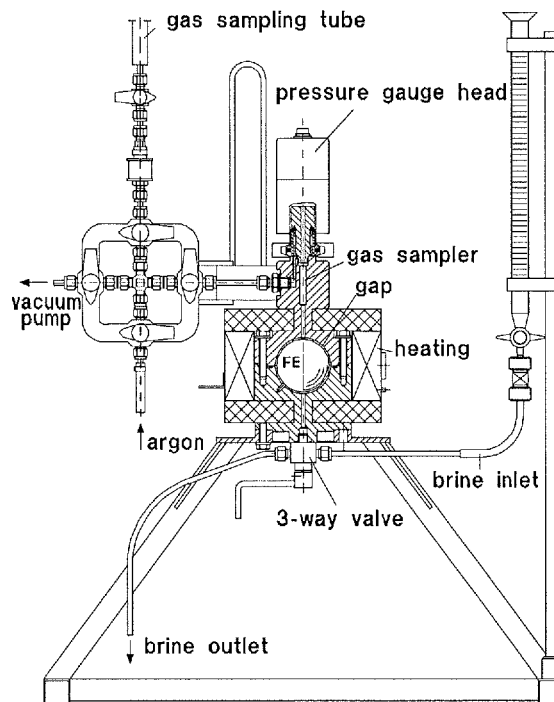


FIG. 18: Hydrogen build-up

Gas-chromatographic analyses proved, that the raising pressure was caused by hydrogen. Helium and tritiated hydrogen could be detected in trace amounts only, $^{85}$Kr and $^{14}$C were never found in the gas plenum.

The formation of hydrogen was not significantly influenced by the thickness of the brine layer. This led to the theory that the water radiolysis mainly depends on degradation on the surface and in the pores of the graphite matrix by α- and β-radiation rather than γ-radiolysis in the gap. The next step will now be the development of a model to prove this theory by calculation.

$^{14}$C was not found in the gas phase but dissolved in the brine. However, the main part of the dissolved $^{14}$C occurs not as inorganic compound. The organically bound $^{14}$C is considered as $C_1$-compounds, but this could not be proved because of the low amount. Fig. 20 and Fig. 19 show the fractional release of inorganic and organic $^{14}$C for the different fuel element types.

However the total release is very low and the leaching rate decreases to nearly zero with time. Due to the rather short half-life of about 5000 years, $^{14}$C may therefore be no major problem for long-term safety of a well-designed deep repository.
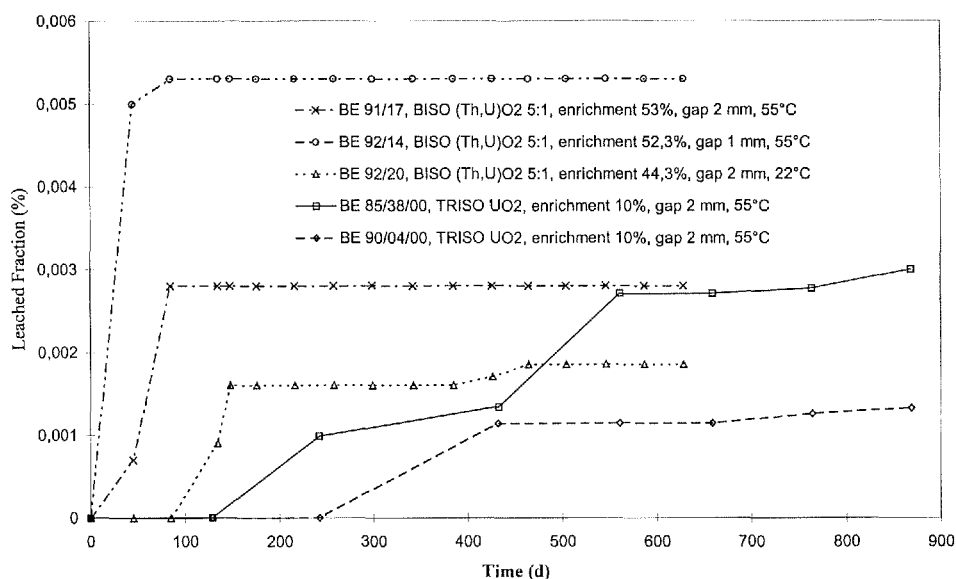


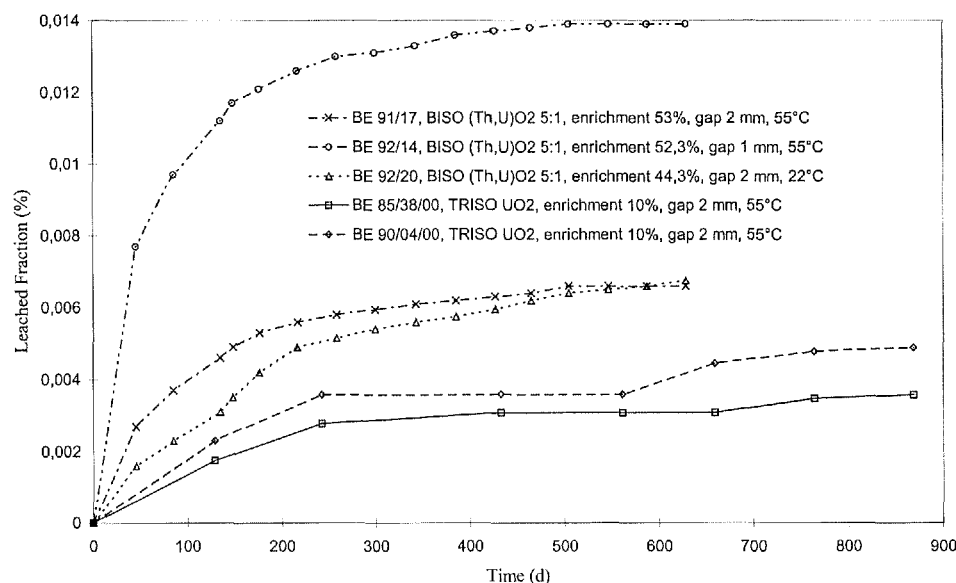Fig. 20: Fractional release of inorganic $^{14}$C



Fig. 19: Fractional release of organic $^{14}$C

# 4    Conclusions

The back-end of the fuel cycle concept for spent High-Temperature Reactor fuel elements in Germany is based upon intermediate dry storage in shielded casks in a surface facility followed by direct disposal in a deep salt repository. Depending on the disposal technique a simple conditioning may be carried out prior to disposal.

Long-term experiments have proven, that HTR fuel elements can safely be stored in dry casks. Only trace amounts of volatile or aerosol-bound radionuclides were found to be released during storage, which represent no risk for the public or the environment. Two facilities to store spent HTR fuel in dry CASTOR-type casks are being operated in Jülich and Ahaus.

Disposal concepts assume the emplacement of spent HTR fuel elements in thick-walled casks in horizontal drifts, or in thin-walled containers in boreholes. In both cases, the ceramic fuel element itself represents the main technical barrier against the long-term release of radionuclides, if the waste disposed off comes into contact with water at all. Leaching experiments have proven that only extremely low amounts of radionuclides are released from the graphite matrix. The release from the coated particles is extremely low and results mainly from defect coatings. Defects in the coating are known to be very low and depend upon type of coating and burn-up. Hence, the fuel elements are well-suited for disposal in a salt repository.

However, there are some questions left: How stable is the particle coating against mechanical and chemical impacts in the long-term run? Does a changing pressure during disposal have a major influence on the release of radionuclides from the graphite matrix? These questions should be further investigated in order to optimise the concept in terms of safety and economics.

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