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KUFA testing report

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KUFA Heating Test of HTR Pebble HFR K6/2

POST-IRRADIATION TESTING OF HTR-FUEL ELEMENTS UNDER ACCIDENT CONDITIONS

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ABSTRACT¹

In the framework of a Share Cost Action (SCA) of the European Commission, a European Project aimed for the development of High Temperature Reactor (HTR) technology has been approved. The project includes developments in the fields of reactor physics, fuel technology, safety, material needs and the feasibility of key components and systems.

A key point in the domain of fuel technology is the testing of the irradiation behaviour of new fuel types and their fabrication methods. In this context, the testing of irradiated fuels under loss-of-coolant accident conditions will be needed to assess the quality of new fuel concepts and fabrication methods. That means principally, the evaluation of the release behaviour of fission gases (Xe, Kr) and solid fission products (Cs, Sr, Ag, etc) under these conditions.

In the past, the so-called Cold Finger Apparatus (KÜFA) was developed in the Forschungszentrum Jülich (FzJ) to test HTR-fuel design and fabrication methods. Using this device, the fission product release from defected particles could be tested up to 1800 °C.

In the framework of the SCA/HTR-technology, an up-graded version of the KÜFA has been installed in the hot cells of the Institute for Transuranium Elements. In the present paper, a comprehensive description of the apparatus is presented, the calibration procedures described, and the future experimental programme discussed.

¹ Paper to be presented at the IAEA-Technical meeting on Gas Cooled Reactor fuels, 7th to 9th of June 2004 , Vienna, Austria.

INTRODUCTION

The opinion of nuclear power in most EU member states is suffering from the public concern about the acceptability of presently established nuclear technology, mainly due to safety issues, but also from the increasing difficulty of keeping economic competitiveness in a de-regulated market.

At present, light water reactors dominate worldwide nuclear energy production. Some advanced designs like the European Pressurised Water Reactor (EPR) meet the safety requirements, but are based on active safety systems which are costly and require more elaborate safety demonstration. On the other hand, the HTRs, being inherently safe, can meet all the requirements, reducing costs without affecting the safety.

Besides the safety, the HTRs offer the following advantages:

- Proliferation-resistant fuel type.
- Reduction of the radioactive waste burden.
- Ultra high burn-up potential ($> 200 \text{ GWd/t U}$).
- Well-known and proven technology (GCRs, AVR, Dragon, Fort Saint Vrain, etc.).
- Robustness of the fuel.
- Social acceptance through improved safety.
- Energy supply diversity (electricity, heat).
- Competitiveness (modular concept).

In the framework of a Share Cost Action (SCA) of the European Commission, a European Project aimed to the development of the High Temperature Reactor (HTR) technology has been approved.

In the domain of fuel technology is the testing of the irradiation behaviour of new fuel types and their fabrication methods. In this context, the post-irradiation testing of irradiated fuels under loss-of-coolant accident conditions will be needed to assess the quality of new fuel concepts and fabrication methods. That means principally, the evaluation of the release behaviour of fission gases (Xe, Kr) and solid fission products (Cs, Sr, Ag, etc) under these conditions.

In the past, the so-called Cold Finger Apparatus (KÜFA) was developed in the Forschungszentrum Jülich (FzJ) to test HTR-fuel design and fabrication methods. Using this device, the fission product released from defect particles could be tested up to $1800 \text{ °C}^{[1-3]}$.

In the context of the SCA/HTR-technology, an up-graded version of the KÜFA has been installed in the hot cells of the Institute for Transuranium Elements. In the present paper, a comprehensive description of the apparatus is presented, the calibration procedures described, and the future experimental programme discussed.

1. TEST BACKGROUND

The central aspect of the safety philosophy for a High Temperature Reactor (HTR) is the retention of fission products - particularly those of the iodine nuclides - in the fuel elements during operation and accidents. For this reason, the determination of the number of damaged particles constitutes the central objective of measuring the fission gas release in the reactor and in the extensive post-irradiation examinations under accident conditions. In modern production methodologies, the heavy metal contamination of fuel elements is kept very low. Consequently, solely the number of defective particles establishes fission gas or iodine release.

During a loss-of-coolant accident, the temperature in the core of a HTR will increase. The amount of this increase depends on the geometrical design of the reactor and the nature of the accident. For the extreme case of pressure loss in the core with the failure of all heat sinks, temperatures as high as 2000°C can be reached in a medium size HTR. On the other hand, for the case of small HTRs and the MODUL-concept in Germany, relatively low accident temperatures between 1400 and less than 1800 °C have been anticipated.

With the increase of the core temperature above normal, fission products may be released from the fuel elements into the primary circuit and, eventually, into the environment. For a realistic assessment of the fission product release, the conditions in the reactor core have to be simulated.

Fission Product	Half life	Relevance assessment
^{131}I	8 days	Greatest significance for design and licensing
$^{137}\text{Cs}/^{134}\text{Cs}$ ^{90}Sr	30 (2) years 29 years	Long term behaviour after extreme accidents and risk analysis
$^{110\text{m}}\text{Ag}$	253 days	Small inventory, short half-life. Important for maintenance
^{85}Kr ^{133}Xe	11 years 5 days	Not accident relevant. Particle defect indicator

Table I: *Relevant fission products to be measured*

The relevant fission products to be measured and their relevance in case of accident are given in Table I.

2. DESCRIPTION OF THE COLD-FINGER APPARATUS

2.1. General

The test requirements, arising from the necessity of evaluation of HTR-fuel elements for licensing purposes, led in the past to the development in FzJ of this highly specialised test equipment capable of coping with entire fuel elements (pebbles or compacts) as well as individual coated particles.

The basic function of this device is to heat the fuel elements up to the expected temperature in a dynamic He-atmosphere, and then to measure the fission product release. In the current up-graded version, designed for accident simulation tests of future HTRs, temperatures up to 2000 °C can be reached.

The fuel element is supported by three pins in the centre of a tantalum tube placed inside the furnace; helium flows through this tube from the bottom to the top (**Fig. 1**).

The tantalum tube and the fuel element are heated by an electrical resistance heater, which likewise consists of tantalum.

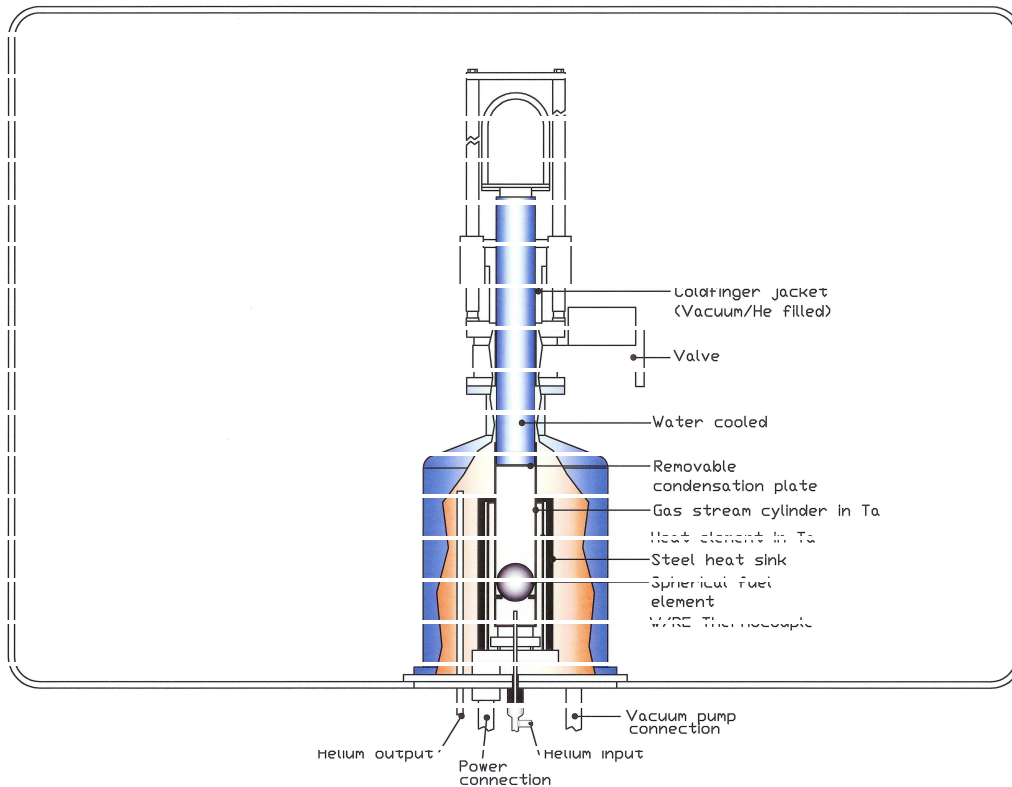


Figure 1: Cold-finger apparatus (KÜFA).

A W/Re-thermocouple, placed near to the specimen, measures the actual temperature during the tests. This thermocouple can be replaced if needed and serves, simultaneously, for the electronic regulation of the temperature of the furnace.

2.2 Measurement of the Fission Gases Release

The measurement of fission gases under accident conditions allows the detection of failed fuel elements. Through the analysis of the release curves, individual failed particles in the fuel element time can be detected as a function of temperature and heating. The two relevant radioactive isotopes ^{85}Kr and ^{133}Xe (see Table I) are relevant for this measurement. The release of fission gases also indicates the release of other fission products, like Iodine, which is difficult to measure directly but it is known to be released to the same extent as Krypton.

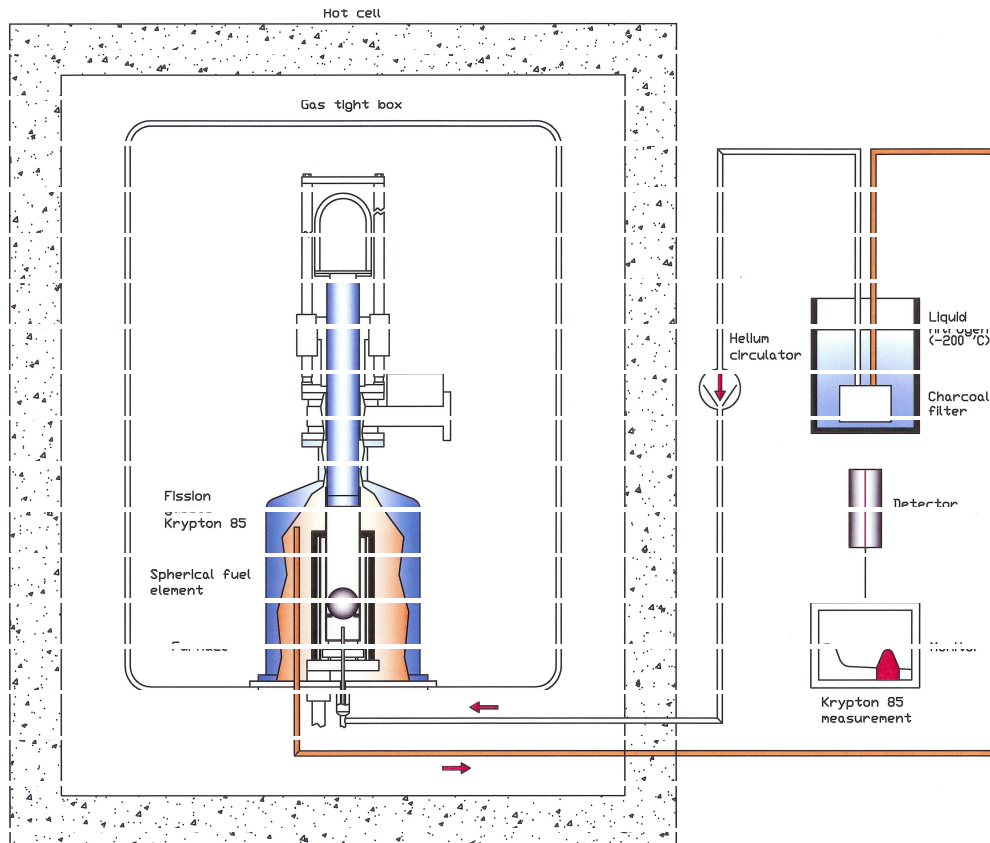


Figure 2: Scheme of the gaseous fission products release measurement.

The furnace is installed in an alpha-tight box in a hot cell, containing also the filters for the helium circuit (**Fig. 2**). Helium carries the fission gases into cold traps where ^{85}Kr and ^{133}Xe are retained.

The cold traps are installed outside the hot cell, and the helium is conducted back to the hot cell and released, in a controlled manner, through the ventilation system. The released fission products are adsorbed on an active charcoal filter at liquid nitrogen temperature. The activity in the measuring trap is then determined continuously by on-line gamma-spectrometry throughout the test. In principle, only the long-lived ^{85}Kr can be detected but, provided the cooling period of the fuel elements is less than 4 to 8 weeks, measurements of ^{133}Xe are possible as well.

The two cold traps are placed in a room beneath the hot cell. The second cold trap is solely meant to ascertain that all the ^{85}Kr -activity was retained in the first. As soon as activity is detected in the second cold trap, the first is changed.

2.3 Solid Fission Products Release

The determination of solid fission products is slightly more difficult than that of the chemically inert fission gases. At high temperatures they can get into the coolant gas by migration/diffusion and subsequent gaseous desorption, first, from the surface of the coated particles and, afterwards, from the surface of the fuel. On the other hand, such fission products are re-deposited by adsorption on cooler surfaces, and this deposition mechanism is exploited for trapping solid fission products in the cold finger test rig.

To detect the release of solid fission products, a water-cooled cold finger protrudes into the hot tantalum tube, at the end of which an easily replaceable condensation plate is held. The solid fission products released from the fuel element are deposited on this plate which has a temperature of less than 100 °C, typically 40 to 80°C depending on the testing temperature. This temperature has to be compared to the specimen temperature, which is in the range of 1600-2000 °C.

During the test, the cold finger can be removed from the furnace through an air-lock system (**Fig.3**) without needing to cool-down the specimen. In fact, the Helium circulation is maintained during the plate-changes, which assures the continuous monitoring of the specimen and the detection any coated particle failure.

After replacing the condensation plate, the cold finger is returned back to its position into the furnace. The plate is normally changed once or twice a day but, if necessary, this operation can be performed more frequently. The changing procedure lasts only few minutes and needs relatively easy manipulation.

The used plates are taken out of the hot cell to be measured by gamma-spectrometry in a low background laboratory to be measured. Measurement of gamma emitters such as ^{137}Cs , ^{134}Cs and ^{131}I is relatively simple, because their individual energy lines can identify them. Strontium 90, a beta emitter nuclide, has to be separated chemically from other fission products for measurement. Since strontium emits beta rays with the highest energy, the activity can also be estimated using a scintillation counter after a special calibration.

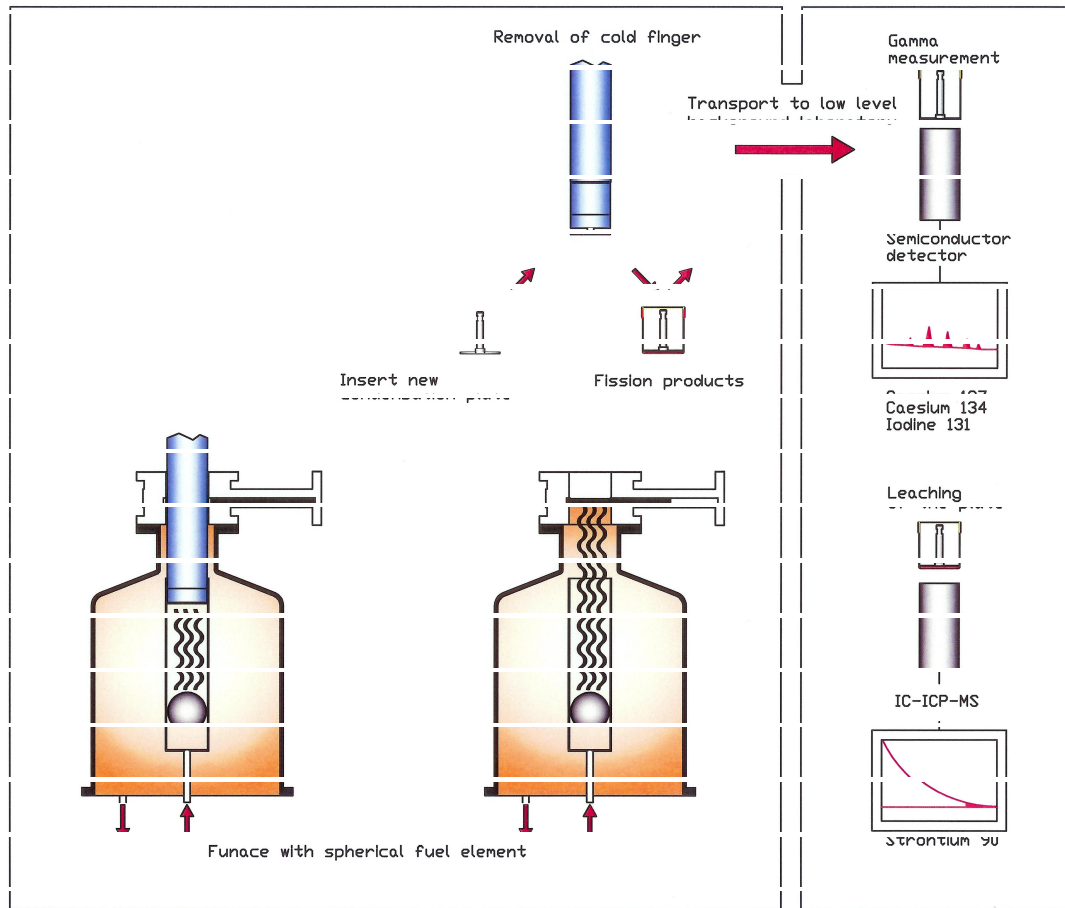


Fig. 3: Scheme of the solid fission products measurement

This procedure is relatively complicated and did not lead in the past to satisfactory results^[3]. Alternatively, the plates can be leached in nitric acid and the resulting solution later analysed using an Induced Couple Plasma Mass Spectrometer (ICP-MS).

According to the experience gathered in the FzJ, use of the Cold Finger Test Rig allows very low fractional releases, down to 10^{-8} , to be measured^[3].

2.4. Typical tests

In **Fig. 4**, a typical heating curve is shown. The highest temperatures in the reactor core occur in depressurisation events with loss of all cooling systems. Depending on reactor design, this temperature can be as high as 1800 °C.

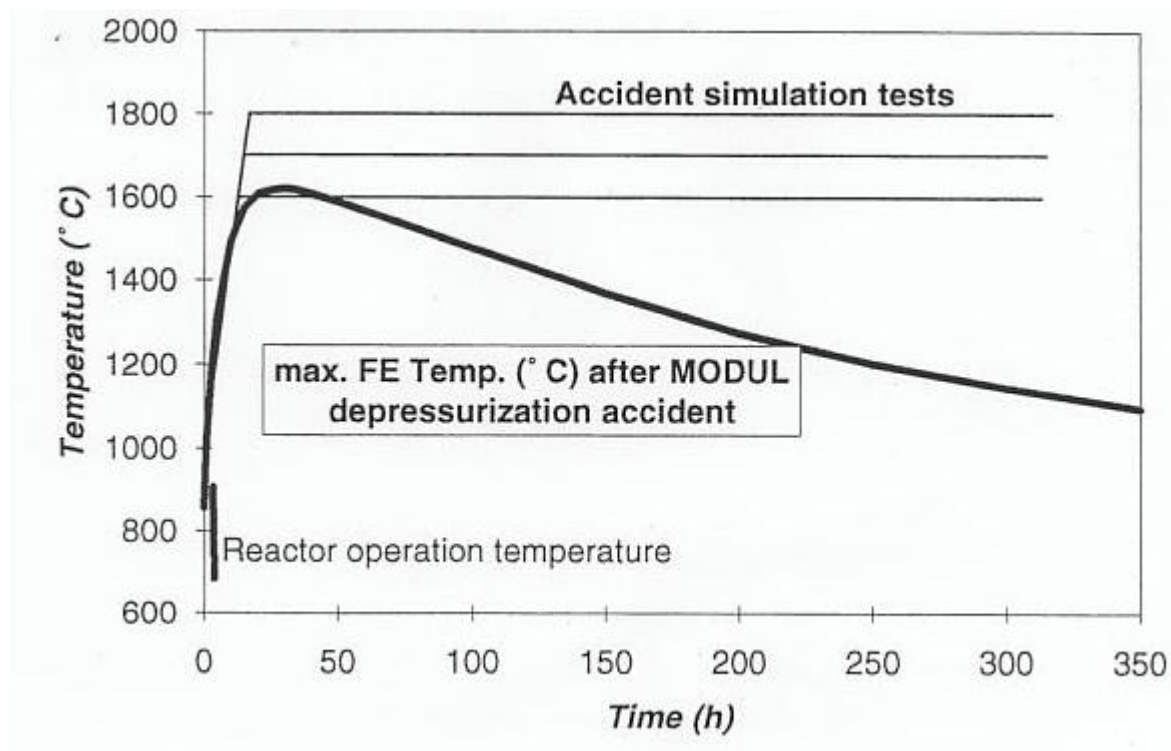


Fig. 4: Temperature evolution during a loss-of-coolant accident in a small HTR and in the heating tests.

3. SAMPLES TO BE ANALYSED

3.1. Already irradiated samples

Samples already irradiated in the Dido and AVR reactors in Jülich and in the HFR-Petten, have been transferred to the hot cell installation of the Institute for Transuranium Elements in Karlsruhe, to be tested using the KÜFA-installation. Irradiation details are given in **Table I**.

Sample No	Type	Fuel Element No	Enrichment [%]	End of Irrad.	Burnup [% FIMA]	Fluence [10^{25}m^{-2}]
1	HFR-K5	1	10.6	16.05.94	6.7	4.0
2		2	10.6		8.8	5.8
3		3	10.6		9.1	5.9
4		4	10.6		8.7	4.9
5	HFR/K6	1	10.6	04.05.93	7.2	3.2
6		2	10.6		9.3	4.6
7		3	10.6		9.7	4.8
8		4	10.6		9.2	4.5
9	FRJ2-K15	1	16.76	11.11.90	14.06	0.2
10		2	16.76		15.27	0.2
11		3	16.76		14.76	0.1
12	AVR-GLE 3	74/16	9.82	08.02.85	3.2	0.5
13		74/18	9.82		4.8	0.8
14		73/21	9.82	07.02.84	2.5	0.3
15		73/23	9.82		2.7	0.3

16	AVR-GLE 4	73/22	16.76	07.02.84	3.4	0.3
17		87/6	16.76	19.11.88	3.51	0.3
18		87/7	16.76		3.53	0.3
19		87/8	16.76		3.53	0.3
20		87/9	16.76		3.56	0.3
21		87/10	16.76		3.51	0.3

Table I: Fuel elements transferred from the FzJ to ITU-Karlsruhe

4.2 Samples to be irradiated in Petten

In the second part of the project, samples will be irradiated in the High Flux Reactor of Petten. To this goal, pebbles containing TRISO-particles fabricated by NUKEM and compacts from USA-production, as well as pebbles from Chinese production, will be irradiated up to a burnup of max. 20 % FIMA. In **Table II**, the main characteristics of these pebbles are presented.

Coated particles	
Kernel composition	UO ₂
Kernel diameter	500 µm
Enrichment	16.7 wt %
Coating thicknesses	
Buffer	92 µm
Inner PyC	38 µm
SiC	33 µm
Outer PyC	41 µm
Fuel Element	
Number of cp	9560
Volume packing fraction	6.2 %
Matrix density	1.75 g/cm ³

Table II: Pebbles from the old German production to be irradiated in HFR-Petten

CONCLUSIONS

Existing equipment, developed in the Fz-Jülich and designed to perform the PIE of HTR-fuel elements, was up-graded and modified for the installation in the hot cells of the ITU in the framework of a SCA of the European Commission. The cold testing of the device has been performed and the hot installation has been completed. After installation, several samples irradiated in the AVR-Reactor and Dido-reactor in Germany as well as in the HFR-Petten will be tested in the framework of the European Program. Additional samples will be irradiated in the Petten-reactor to ultra-high burn-up and subsequently tested in KÜFA-device. The first hot testing is foreseen for July 2004.

References

1. W. Schenk, "Nachbestrahlungsausheizverfahren für Kugelbrennelemente und andere Brennstoffproben", Jülich Report 1454, July 1977
2. W. Schenk, D. Pietzer and H. Nabielek, "Fission Product Release Profiles from Spherical HTR Fuel Elements at Accident Temperatures", Jülich Report 2234, September 1988

3. W. Schenk, R. Gontard and H. Nabielek, "Performance of HTR Fuel Samples under High-Irradiation and Accident Simulation Conditions, with Emphasis on Test Capsules HFR-P4 and SL-P1

POST-IRRADIATION TESTING OF HTR-FUEL ELEMENTS UNDER ACCIDENT CONDITIONS. PART 2

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ABSTRACT: This paper is part of a series of publications aimed to provide the scientific community with a complete description of the Cold Finger Device (in German KÜFA). The KÜFA-device has been designed to test under loss-of-coolant accident conditions, the quality of fuel elements based on coated particles for High Temperature Reactors (HTR). A general description of the equipment has been provided in a previous paper. In the present paper, the cooling system, the temperature control, the sweep gas system and the calibration procedures are described in detail.

0. INTRODUCTION

In the past, the so-called Cold Finger Apparatus (KÜFA) was developed in the Forschungszentrum Jülich (FzJ) to test the design and fabrication methods of High Temperature Reactor (HTR) fuel. Using this device, the fission product release from defect particles could be tested up to 1800 °C. In the framework of a European Commission program for HTR-technology development, an up-graded version of the KÜFA has been installed in the hot cells of the Institute for Transuranium Elements.

This paper is part of a series of publications aimed to provide the scientific community with a complete description of a equipment, the Cold Finger Device (in German KÜFA) projected to test - under lost of coolant accident conditions - the quality of fuel elements based on coated particles for High Temperature Reactors (HTR). In a previous paper the general description of the equipment and the background of the project have been provided^[1]. In the present paper, the furnace, the cooling system (in particular the cold finger), the temperature control, the sweep gas system and the calibration procedures are described in detail.

1. DESCRIPTION OF THE FURNACE

The furnace was designed by researchers at the Jülich Research Centre (FzJ) in co-operation with the company ALD-Hanau, Germany^[2-4]. It consists mainly of a floor plate and double-wall hood, both water-cooled. The furnace is fastened to the floor of the alpha-tight hot cell box via an O-ring. The hood can be open to unload and reload the fuel elements into the furnace, using a crane installed in the hot cell. The fuel elements can be placed, using a suction device connected to a vacuum cleaner, on a three-point support welded to the main tantalum gas pipe placed in the centre of the furnace.

A tantalum heater, carrying a maximum current of 2400 A at the maximum power of 7x10⁴ watts heats up the fuel element during the test. Two W/Re thermocouples are place near the samples for the determination of the exact temperature of the fuel elements during the test and to regulate the

temperature of the furnace.

The electronic temperature regulation system has been updated by the company GERO GmbH, Germany. The system is able to perform any heating curve up to 2000 °C.

2. COOLING SYSTEM

The cooling system (CS) is one of the most important constituent of the KÜFA-device. It has to work without human intervention during the whole test (up to 300 h). It consists (see **Fig 1**) of 6 different and independent closed cooling loops:

- The double wall hood of the furnace,
- The valve closing the furnace when the cold finger is taken out,
- The base-plate of the furnace attached to the floor of the alpha-box,
- The holders of the heating element (two), and
- The cold finger (CF).

The last cooling loop has a particular importance since it has to keep the replaceable plate at a temperature below 100°C during the test, facilitating the plate-out of solid fission products. To check the maximum temperature achieved by the plate on the CF, experiments were conducted at different furnace temperatures, up to 1800 °C. The results are gathered in Table I.

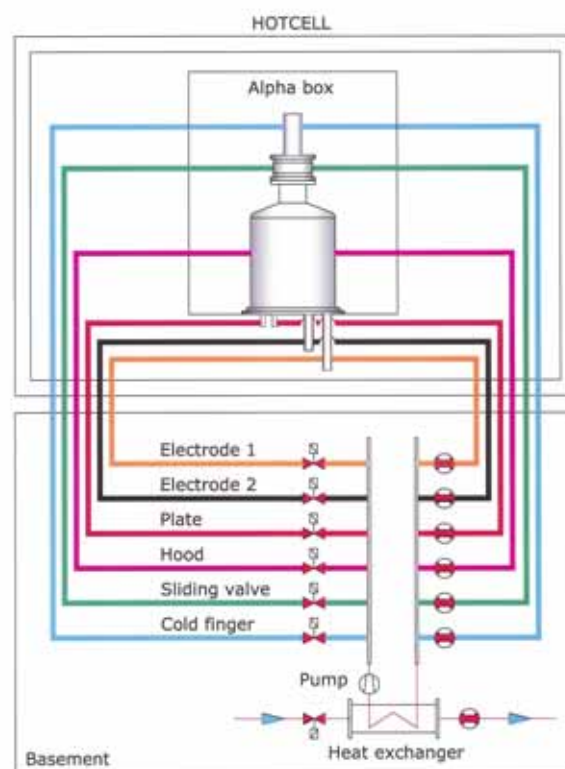


FIGURE 1: The cooling system of the KÜFA-device

To obtain these temperatures, the original design of the CF had to be modified. In the present version, the cooling water impinges directly onto the surface in contact with the replaceable plates, ensuring

the best cooling conditions.

TABLE I: Temperature of the cold plate as a function of the sample temperature

Temperature of the Fuel Element [°C]	Temperature at the Plate [°C]
1600	40
1700	45
1800	65

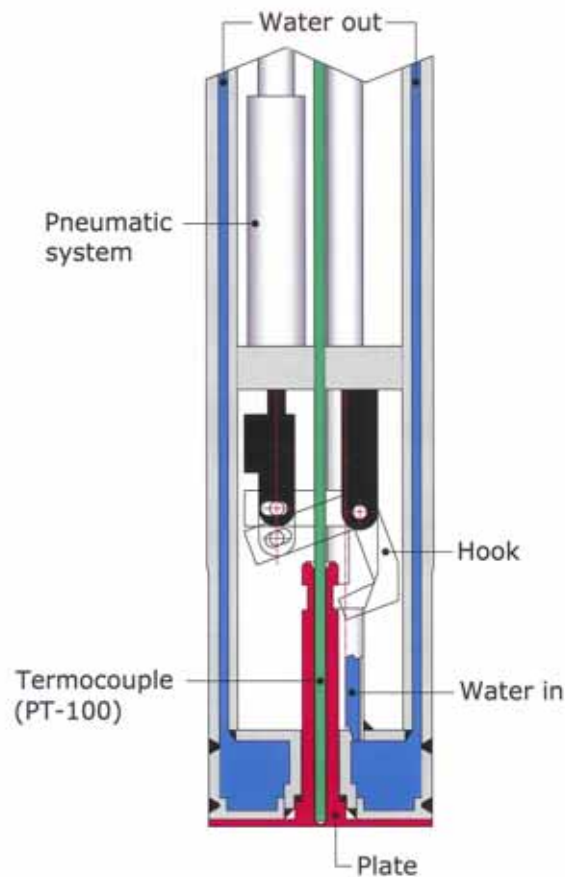


FIGURE 2: Cold-finger system

In the CS, each loop is controlled by individual valves, both at the water inlet and outlet. The flux and temperature can be controlled independently for each loop and, hence, the desired temperature at the different parts of the apparatus can be set. Whereas in the cold finger, the lower the temperature the better the results (better plate-out of the solid fission products) in the rest of the equipment higher temperatures are convenient. In particular, the temperature of the hood and that of the lower plate had to be kept at a slightly higher level to avoid the deposition of the solid fission products on the internal surfaces of the furnace rather than onto the surface of the plate at the cold finger.

A single pump (8 m³/h, at a max. pressure of 2 bar) for all the six loops, conveys the water through the different components of the device and, eventually, to a heat exchanger. There, heat is exchanged with an open secondary loop of running water, having an average temperature around 16 °C.

2.1 Safety features

The temperature of the six cooling loops is permanently controlled by a computer system. If the temperature exceeds a certain value, the heating is automatically interrupted. The temperature of the all cooling loops is recorded during the whole test.

3. DESCRIPTION OF THE COLD FINGER

The cold finger, shown in **Fig. 2**, has been designed to facilitate the plate out of solid fission products during the test. It protrudes into the furnace and is kept at a temperature below 100 °C by an independent cooling circuit as previously discussed. It can be taken out from the furnace, allowing the replacement of the removable plates during the test. During this operation, the sliding valve remains closed maintaining the gas circulation through the sample and allowing thereby the continuous monitoring of the fission gas.

3.1 Plate changing

Using an electric crane installed in the hot cell, the cold finger can be taken out of the furnace in order to remove the used condensation plate. The used plate falls into a holder after remote unlatching by a pneumatic system (see 4.2. below) and it can be, depending on its activity, locked out of the hot cell or measured on site by gamma spectrometry. Alternatively, the plates can be leached in nitric acid and the resulting solution analysed by Inductive Couple Plasma Mass Spectrometry (see ref.1).

Afterwards, the cold finger is reinserted into the furnace in an intermediate position. Then, the lock is evacuated and flooded with helium previous to the opening of the slide valve, which allows the insertion of the cold finger back into the furnace. After every change of the plate, the evacuation and He-flooding is done automatically three times before open the sliding valve. This ensures that the atmosphere is kept at the same purity level during the whole test.

3.2. Description of the plate fixation

The plate is fixed by a spring, holding a hook that pins the plate into position. The hook is connected to a pneumatic system that, when pressurised, opens the hook allowing the plate to be removed. A new plate can then be inserted and fixed by the spring to the cold finger by de-pressurisation of the pneumatic system.

3.3 Temperature control of the cold finger

A PT-100 thermometer is inserted in the plate, to measure the temperature near to the condensing surface (see **Fig. 1** and **Fig. 2**). This temperature is continuously recorded during the whole test. Upon failure of cooling, flow and temperature sensors in the cold finger cooling circuit allow the automatic shut off of the furnace.

According to the previous experience [2-4], most of the solid fission products released from a fuel element during a heating test are deposited on the plate of the cold finger. Only small quantities reach the traps foreseen to avoid any release of solid fission products to the exterior of the alpha containment.

4. SWEEP GAS SYSTEM

In **Fig. 3**, the sweep gas system of the KÜFA device is depicted. During the experiment, the fuel element is swept from the bottom upward with helium (about 30 l/hour). The gas is continuously pumped into the system and, afterwards, put through filters into the hot cell installation exhaust.

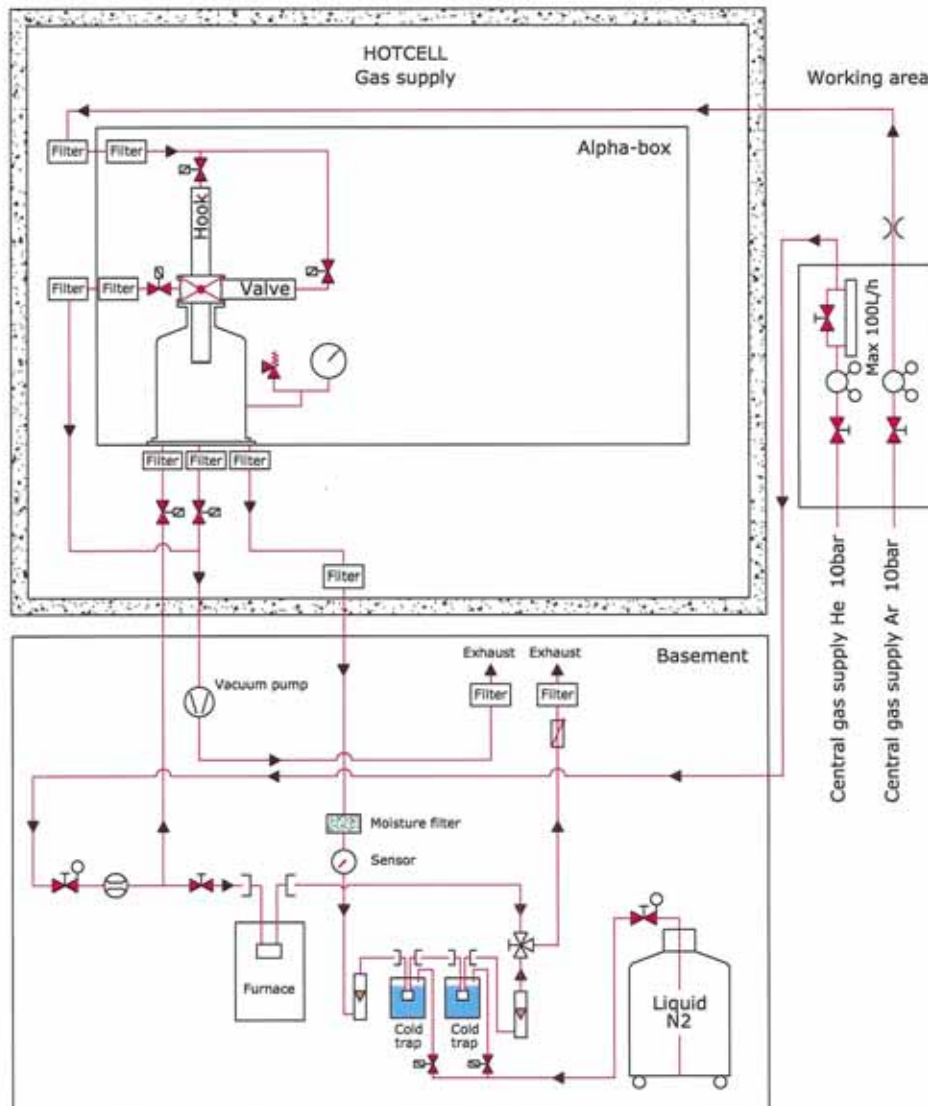


FIGURE 3: Sweep gas system

The gas is passed through cold traps (see **Fig. 4**) and the ^{85}Kr -isotope is continuously monitored as explained in ref. 1. The cold-traps/detectors system is located in the basement beneath the hot cell.

Absolute filters on every passage through the alpha-tight box ensure that no activity will be released to the environment. Moisture is trapped in ad-hoc filters and a detector continuously monitors the humidity level.

On the gas line a furnace can be inserted to perform the regeneration of the cold traps, by heating them up under flowing helium.

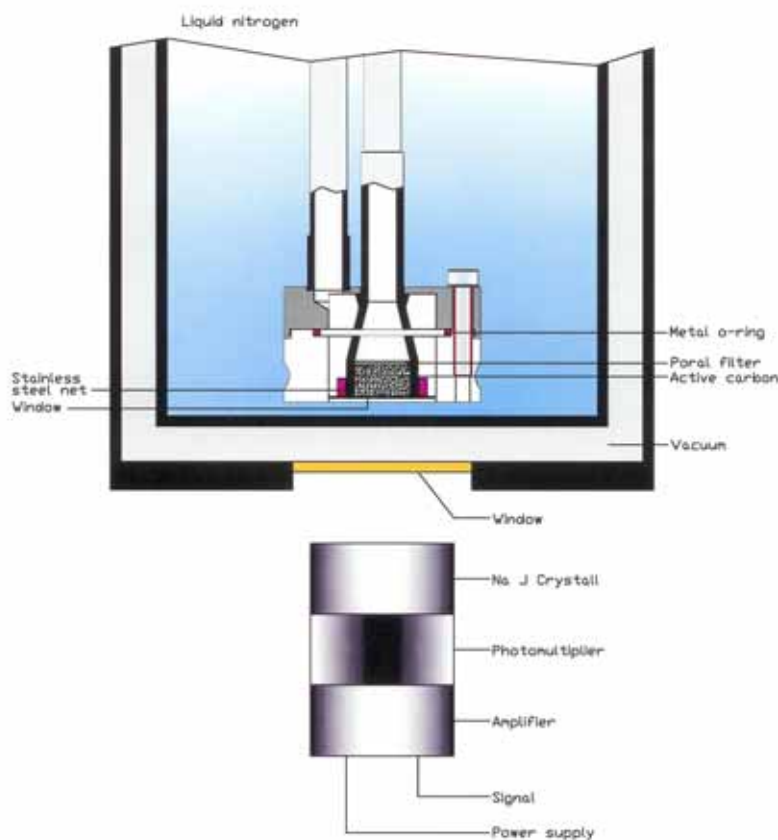


FIGURE 4: Cold-traps and detector system

5. SYSTEM CALIBRATION

In order to perform an accurate measurement of the release of the fission products, the different measuring systems have to be calibrated. In this respect, a special attention has to be devoted to the determination of the efficiency of the measuring devices.

Two measuring systems for both solid and gaseous fission products have to be considered. The first, is the sytem cold-trap/detector for the measurement of the fission gas release (^{85}Kr), using NaI-detectors. The second concerns with the plate-out efficiency of the solid fission products on the cold-finger (mainly ^{137}Cs and $^{110\text{m}}\text{Ag}$).

5.1 NaI-Detectors calibration

The gamma detectors have to be calibrated in order to allow a quantitative measurement that will permit the determination of the amount of failed particles, by comparing the amount of ^{85}Kr produced during irradiation (birth rate, given by calculation) with the actual release.

Since the expected activity is relatively low, the measuring system should be able to detect activities as low as 5×10^{-2} counts/s. A detailed description of the detectors and the sensibility attained will be

provided in the third part of this publication series.

The expected increase in activity caused by a broken particle is estimated by about 10 counts/s. The system should also allow the determination of the instant at which a particle fails. With this aim, the data-taking system records one experimental point per minute. During one minute the activity of the sample is accumulated and, at the end of the period, the total amount of counts can be added using special software developed by Perkin Elmer.

The calibration of the system and the determination of its efficiency can be done by ^{85}Kr -standards that are commercial available. With this aim, two different procedures were used:

- A certified standard (a known amount of ^{85}Kr enclosed in a glass-ampoule) was broken in stainless-steel container, having a known volume, previously pressurised with He. From this container, a known amount of ^{85}Kr was taken into a smaller volume and this put into the cold traps via a bypass.
- The second procedure is an on-site calibration: An ampoule, containing a certified amount of ^{85}Kr , is heated up to melting, liberating the whole amount of gas contained in it.

Both methods will be applied. Through the first method, an efficiency factor of 1×10^{-5} has been determined. The second will permit also the determination of the delay time for the gas to achieve the cold-traps/detectors system, located about 4 meters beneath the hot cell in which the KÜFA-device is located.

5.2 Determination of the efficiency of the cold finger

The fraction deposited on the plates of the cold finger must be known for the quantitative determination of the fission product activity released from a fuel sample. To this goal, a known amount of activity of the relevant solid fission products is placed in the furnace. Hence, this sample is heated up to the test temperature and, afterwards, the activity on the plates has to be determined by the available experimental methods for each isotope. This activity can be then be compared to the activity placed in the furnace at the beginning of the test, determining the efficiency of the cold finger.

In order to approximate to the release conditions in heating tests with spherical fuel elements, graphite spheres have been produced in the Research Centre Jülich, having the exact geometry of the fuel elements and provided with a central hole (see **Fig.5**). In this central hole, a special insert, also made of graphite, can be positioned. On this insert, a known amount of activity can be placed and a graphite-screw provides the final fixation.

For the initial activity, commercial standards can be used for the different relevant isotopes: ^{134}Cs , ^{137}Cs , ^{90}Sr and $^{110\text{m}}\text{Ag}$. Alternatively, a solution of fuel irradiated in commercial reactors may be use. This method has the advantage to represent the exact composition of irradiated materials, with the real isotopic relationships, and that the burnup can be determined, for instance, by the measurement of the amount of ^{148}Nd , contained in the solution.

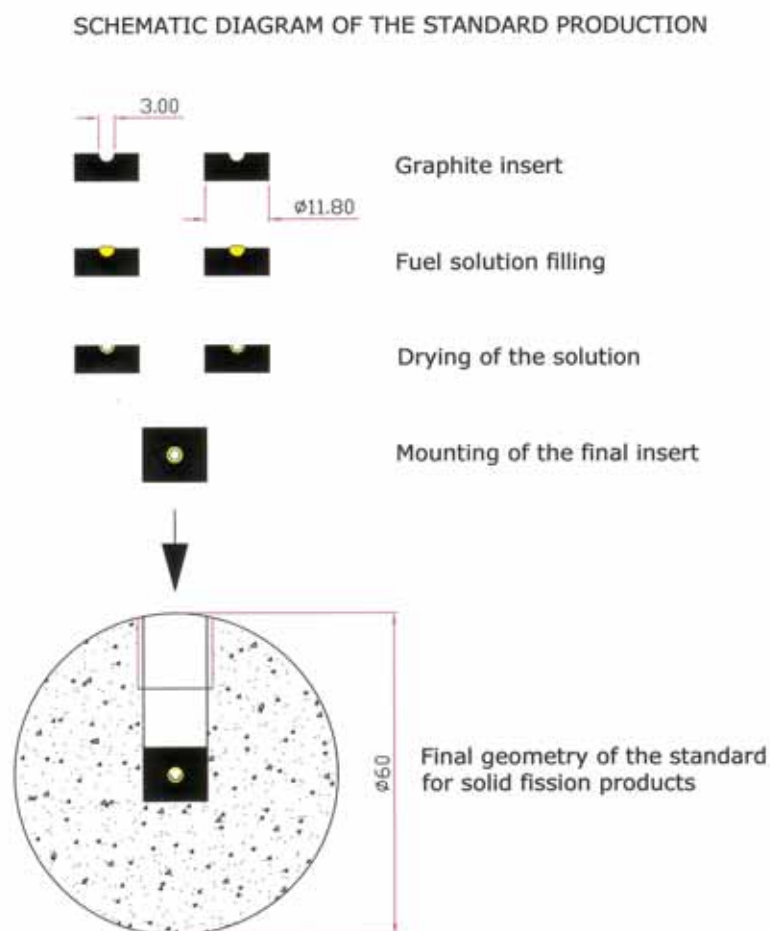


FIGURE 5: Efficiency of the cold plates

5.2.1. Standard preparation from fuel irradiated in commercial reactors

One pellet was cut from a fuel rod irradiated in a power reactor to a burnup of around 70 GWd/t[HM]. This sample was dissolved in a 6 molar HNO_3 solution at about 90°C. A small aliquot was transferred to a glove box for the separation process. The amount of the heavy metals, U and Pu, was determined using the Thermo-ionic Mass Spectrometry (TIMS)- Isotopic Dilution method.

The burnup of the sample was determined by measuring the amount of ^{148}Nd in the sample solution. The determination of the inventory of fission products was performed by Induced Couple Plasma Mass Spectrometry (ICP-MS) previous on-line chromatographic separation. The measurements were performed with an Elan 5000 ICP-MS (Perkin Elmer SCIEX) modified for installation in a glove box and coupled with a 4500I high-pressure chromatographic pump (Dionex). The experimental results will be reported in the third part of this publication series.

CONCLUSIONS

In the framework of a European Commission program for HTR-technology development, an up-graded version of the KÜFA has been installed in the hot cells of the Institute for Transuranium Elements. In the present paper, the cooling system, the temperature control, the sweep gas system and the calibration procedures of the KÜFA-device have been described in detail. The installation of the device is now being completed and the first experiments will be performed using spherical fuel elements irradiated in several reactors in Europe.

REFERENCES

- [1] H.Kostecka, J.Ejton, W.de Weerd and E.H.Toscano, "Post-Irradiation Testing of HTR-Fuel Elements under Accident Conditions. Part 1.
- [2] W. Schenk, "Nachbestrahlungsausheizverfahren für Kugelbrennelemente und andere Brennstoffproben", Jülich Report 1454, July 1977
- [3] W. Schenk, D. Pietzer and H. Nabielek, "Fission Product Release Profiles from Spherical HTR Fuel Elements at Accident Temperatures", Jülich Report 2234, September 1988
- [4] W. Schenk, R. Gontard and H. Nabielek, "Performance of HTR Fuel Samples under High-Irradiation and Accident Simulation Conditions, with Emphasis on Test Capsules HFR-P4 and SL-P1

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KüFA Heating Test of HTR Pebble AE-732100

Identification Nr.: AE-732100

Date: 04.10.2005

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1 Background

Objective of the experiment was the testing of the AVR (Allgemeiner Versuchs Reaktor) [Kugeler, Schulten 1989] fuel element (FE) with the identification number AE-732100. This FE is a GLE-3 type spherical fuel element with a burn up of 2,5 % FIMA. The heating test was performed in the KüFA device in Wing B, Hot Cell number 103. Beginning of the test was October the fourth 2005. The pebble was irradiated 21 years ago (end of irradiation was February 1984) in the AVR and therefore it was expected to release primary the long-lived fission products ^{137}Cs , ^{134}Cs , ^{85}Kr and ^{90}Sr . The parameters of the test (temperature and duration) were below the critical limits of the fuel element and so no failure of Coated Particles should occur and the fractional release of the fission products should be in a range of 10^{-6} to 10^{-5} for ^{137}Cs and 10^{-7} to 10^{-6} for ^{85}Kr and up to 10^{-5} for ^{90}Sr .

2 Fuel Element Data

2.1 General Data

Ident.Nr.: AE-732100

Type of sphere	GLE-3	Kernel composition	UO ₂
Type of graphite	Nukem A3-27	Enrichment	9,82% U-235
Particles in fuel element	16400	Kernel diameter	500 ±2 µm
Heavy metal weight	10 g	Kernel density	10,80 g/cm ³

Coating

Buffer layer thickness	93 µm ± 14%	Buffer PyC layer density	1,01 g/cm ³
Inner PyC layer thickness	38 µm ± 10%	Inner PyC layer density	1,86 g/cm ³
SiC layer thickness	35 µm ± 6%	SiC layer density	3,19 g/cm ³
Outer PyC layer thickness	40 µm ± 9%	Outer PyC layer density	1,89 g/cm ³

Irradiation history

Burn up	2,5% fima	Time of Irradiation	235 efpd
Thermal flux	$2 \cdot 10^{21} \text{ cm}^{-2}$	End of irradiation	Feb. 1984
Fast flux (>0,1 MeV)	$4 \cdot 10^{20} \text{ cm}^{-2}$	Irradiation temperature	ca. 700°C

Pebble inventory (est.) at beginning of the test

⁸⁵ Kr	$8,2 \cdot 10^8 \text{ Bq}$	¹³⁷ Cs	$1,8 \cdot 10^{10} \text{ Bq}$
⁹⁰ Sr	$1,6 \cdot 10^{10} \text{ Bq}$	¹³⁴ Cs	$5,0 \cdot 10^6 \text{ Bq}$
¹³¹ I	~ 0 Bq	^{110m} Ag	0,015 Bq

2.2 Burn up and Inventory calculation

The burn up was calculated with the computer program Inventar by W. Kühnlein [Duwe, Kühnlein, 1975]. The values were calculated for the end of irradiation.

The Inventar code was written in FORTRAN and works with the analytical solutions of the differential equations for the build up and burn up of the main heavy metals and main fission products in the AVR.

SPALTPRODUKTINVENTAR						
3	NUKLID	3	ATOMZAHL	3	AKTIVITAET	3
3		3		3	ZERF/SEK	3
3		3		3	MIKROCURIE	3
3	KR 85	3	1.63E+18	3	3.30E+09	3
3		3		3	8.92E+04	3
3	SR 89	3	8.36E+18	3	1.29E+12	3
3		3		3	3.48E+07	3
3	SR 90	3	3.45E+19	3	2.70E+10	3
3		3		3	7.30E+05	3
3	ZR 95	3	1.41E+19	3	1.74E+12	3
3		3		3	4.70E+07	3
3	RU 103	3	5.23E+18	3	1.06E+12	3
3		3		3	2.87E+07	3
3	RU 106	3	4.74E+18	3	1.04E+11	3
3		3		3	2.81E+06	3
3	AG 110M	3	1.65E+15	3	5.22E+07	3
3		3		3	1.41E+03	3
3	I 131	3	9.49E+17	3	9.46E+11	3
3		3		3	2.56E+07	3
3	XE 133	3	1.32E+18	3	2.00E+12	3
3		3		3	5.41E+07	3
3	CS 134	3	6.63E+17	3	7.10E+09	3
3		3		3	1.92E+05	3
3	CS 137	3	4.01E+19	3	2.94E+10	3
3		3		3	7.94E+05	3
3	BA 140	3	3.03E+18	3	1.90E+12	3
3		3		3	5.14E+07	3
3	CE 141	3	7.71E+18	3	1.90E+12	3
3		3		3	5.13E+07	3
3	CE 144	3	2.67E+19	3	7.52E+11	3
3		3		3	2.03E+07	3

3	ABBRAND			3
3	ABBRAND	U	233	3
3				3
3	ABBRAND	U	235	3
3				3
3	ABBRAND	PU	239	3
3				3
3	ABBRAND	PU	241	3
3				3
3	GESAMTABBRAND			3

Table 2-1: Burn up calculation for AE-732100 with Inventar

3 Calibration and Test Preparation

3.1 Cold Trap Calibration:

Before the test the cold traps were cooled down with liquid nitrogen and a 15 h Background measurement was performed.

The calibration of the first cold trap was performed after the heating test with two aliquots of ^{85}Kr which were brought in through a bypass.

3.1.1 Calibration 1

Date and time	06.10.2005 / 12:00
Activity in sample	77.891 Bq
Impulses per second	9,1267
Calibration factor	$1,17 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$

Table 3-1: cold trap calibration 1

3.1.2 Calibration 2

Date and time	06.10.2005 / 12:00
Activity in sample	76.099 Bq
Impulses per second	9,487
Calibration factor	$1,25 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$

Table 3-2: cold trap calibration 2

The average calibration factor amounts to $1,21 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$.

3.2 Test Preparations

The test preparations were performed as described in the working instruction for the KüFA device.

1. A new and not contaminated tantalum tube was installed in the furnace.
2. A new condensate plate was installed at the cooling finger.
3. A clean cold trap was included in the helium circuit and cooled down with liquid nitrogen. A second new trap was prepared and inserted in the circuit behind the first one.
4. The pebble was brought in the device.
5. The ^{85}Kr scanning for both traps was prepared and started.
6. The heating program was written and started.
7. After the test, two aliquots of ^{85}Kr were inserted into the first cold trap through a bypass to calibrate the trap.

4 Test procedure

The goal of the heating of pebble AE-732100 was the commissioning of the KüFA under remote controlled conditions. The heating program was divided into two steps. First heating up to 1600°C and cooling down to room temperature and then heating up to 1800°C. The furnace was held at each temperature for 5 hours. (see Figure 4-1) Before the first heating step the pebble was held at 300°C for 20 hours.

As can be seen from table 4-1 at the end of each heating phase (after 1600°C and 1800°C) the condensate plate was changed.

Time (h)	Endurance (h)	Temperature (°C)	Gain (°C/h)	Plate
0		20		6
0,2	0,2	300	1400	6
20,2	20	300	0	6
22,2	2	1600	650	6
27,2	5	1600	0	6 - 2
43,2	16	20	-98,75	2
44,2	1	1800	1780	2
49,2	5	1800	0	2
65,2	16	20	-111,25	2

Table 4-1: heating program

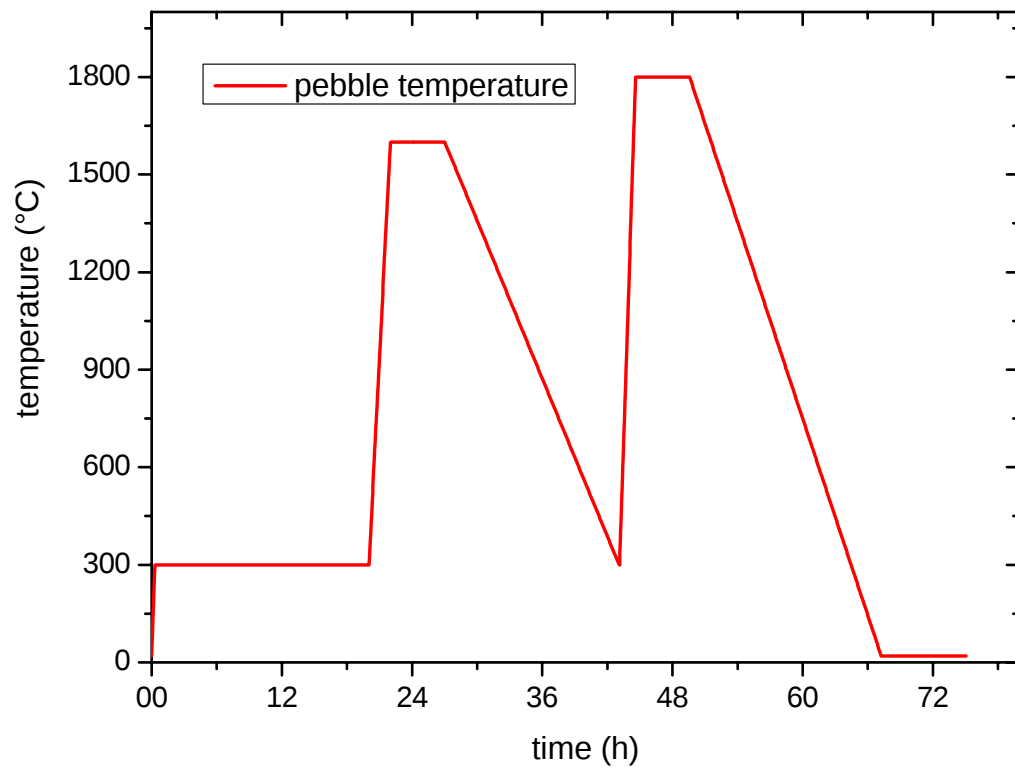


Figure 4-1: heating program

5 Results

5.1 Fission gas (^{85}Kr) release

The ^{85}Kr release during the test was about 12.000 Bq which is equivalent to a fractional release of $5,9 \cdot 10^{-6}$ of the inventory. The activity increase in figure 5-1 at 54 hours respectively 70 hours is due to the calibrations after the test. For this reason the fractional release curve was interrupted at 54 h.

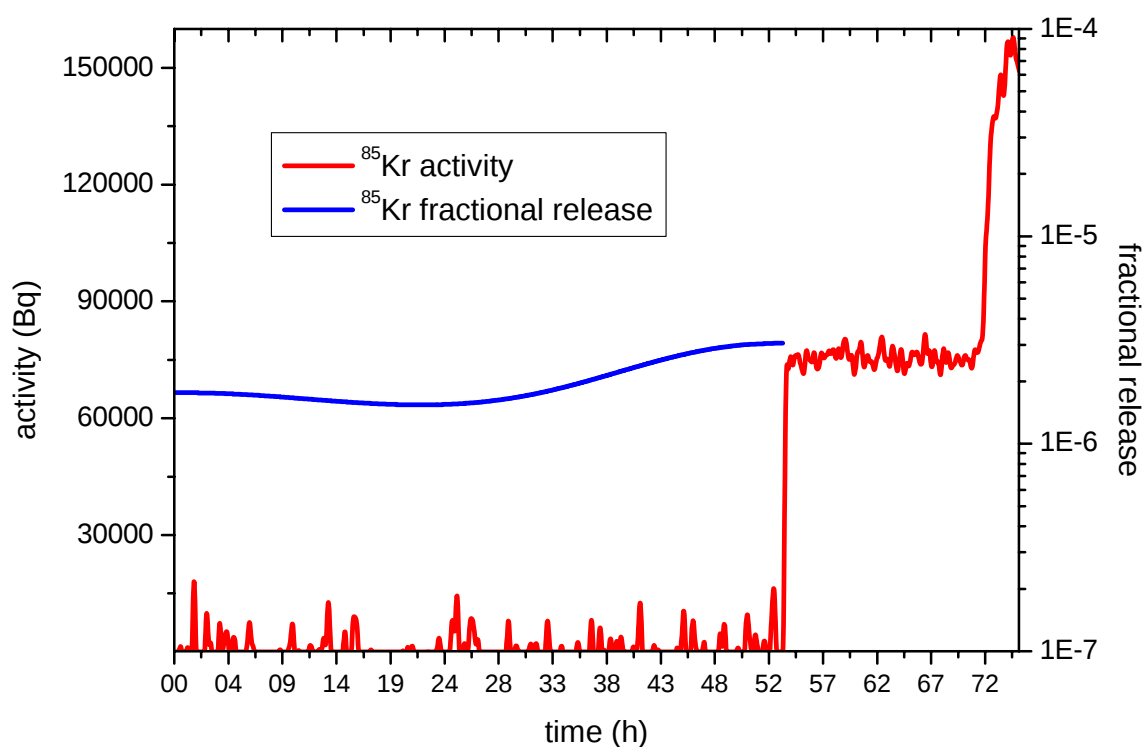


Figure 5-1: ^{85}Kr release in Becquerel and fractional release

In the second cold trap no increase of activity could be recognized.

5.2 Release of solid fission products

5.2.1 Gamma spectrometry of the condensate plates

Subject: Gamma-spectrometry for project number 22835. (KüFA, AE-732100, see detailed information in [Carbol 2006])

Objective of the gamma measurements were the two condensate plates of the KüFA device in Wing B, Hot Cell 103. These plates were used in the heating and fission product release experiment of a burned up spherical fuel element (Identification Nr.: AE-732100) from the AVR reactor. [Kugeler, Schulten 1989] Since the pebble was irradiated 20 years ago (end of irr. Febr. 1984) it was expected to release primary the long-lived fission products, for example ^{137}Cs and ^{90}Sr . The parameters of the test (temperature and duration) were below the critical limits of the fuel element and therefore the gamma measurements should show a relative small activity of about 10^{-6} to 10^{-5} fractional release of ^{137}Cs (20.000 to 180.000 Bq ^{137}Cs) and up to 10^{-5} fractional release of ^{90}Sr (160.000 Bq ^{90}Sr).

Nuclide	Plate 6	Plate 2	Reference Plate (5)
$^{110\text{m}}\text{Ag}$	2.4 ± 0.4	-	-
^{134}Cs	$2.6 \cdot 10^2 \pm 8$	$1.5 \cdot 10^2 \pm 5$	0.42 ± 0.02
^{137}Cs	$9.1 \cdot 10^4 \pm 2.6 \cdot 10^2$	$6.7 \cdot 10^4 \pm 2 \cdot 10^2$	3.7 ± 0.2
^{90}Sr	No value	No value	No value
^{60}Co	7 ± 0.5	54 ± 0.3	-

Table 5-1: γ -measurement results of the condensate plates

The results of the gamma measurements of the plates are finally corrected through the efficiency factor of the cooling finger for Cesium (70%) respectively Strontium (20%).

^{90}Sr will be measured in the second phase of the project by leaching of the condensate plates and the subsequently measurement by IC-ICP-MS.

Due to contamination during the transfer of the plates to the measuring place, the results of ^{134}Cs are influenced by leftovers from PWR and BWR-fuels.

5.2.2 Release of ^{137}Cs

As shown in Figure 5-2 the release of ^{137}Cs was approximately in the expected range of 10^{-6} to 10^{-5} fractional release. The values are shown in released activity in Becquerel and in fractional release.

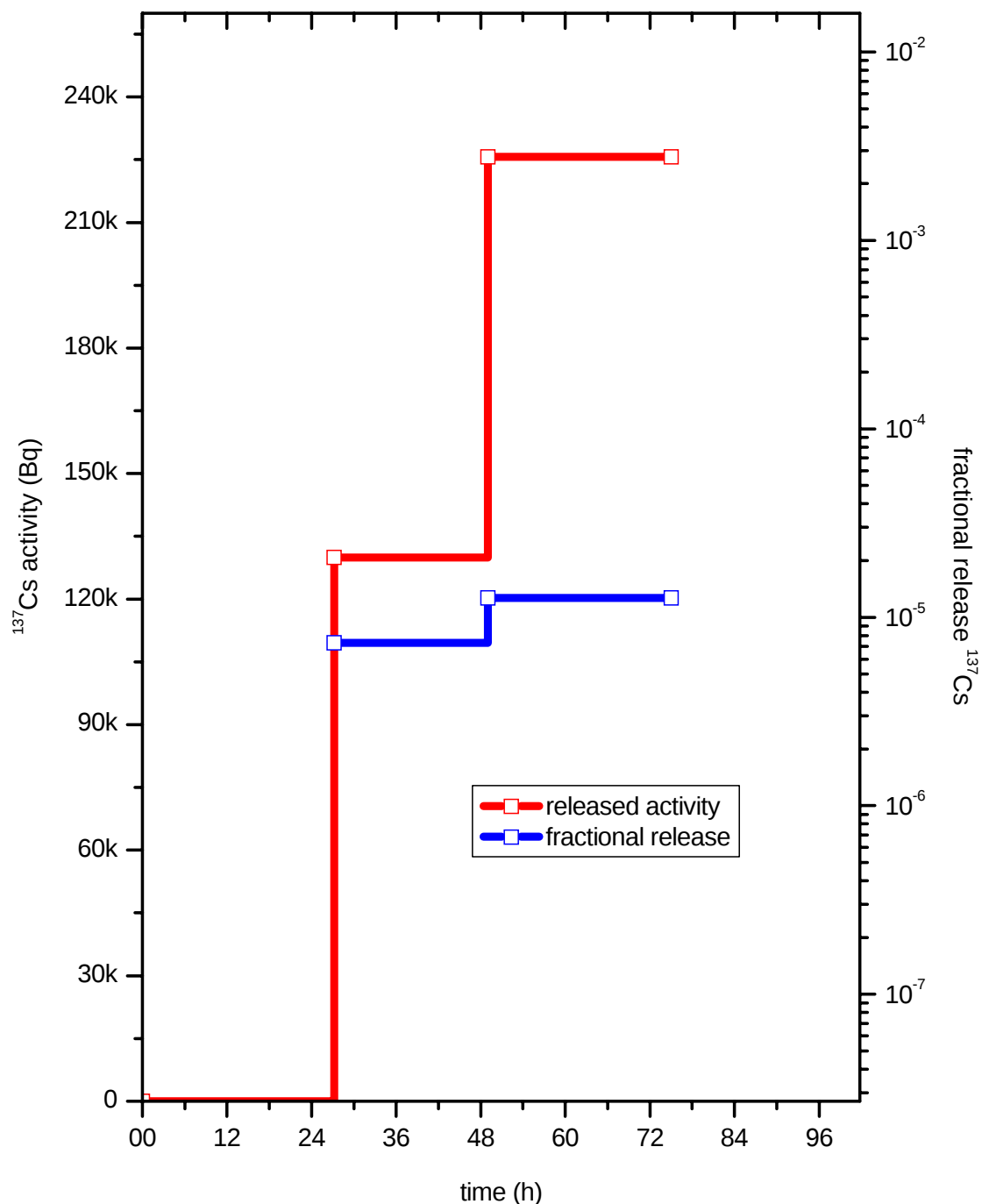


Figure 5-2: ^{137}Cs release of AE-732100

6 Appendix

6.1 Results of γ -spectrometry

The complete results of the γ -spectrometry [Carbol 2006] are given in the following tables.

Radionuclide	Activity (Bq/plate)		
	AE732100-2	AE732100-5	AE732100-6
^{54}Mn	9.2 ± 0.3	b.d.l.	27 ± 1
^{60}Co	54 ± 0.3	b.d.l.	7 ± 0.4
$^{110\text{m}}\text{Ag}$	b.d.l.	b.d.l.	2.4 ± 0.4
^{125}Sb	12 ± 2	b.d.l.	53 ± 4
^{134}Cs	$1.5 \cdot 10^2 \pm 0.05 \cdot 10^2$	0.42 ± 0.02	$2.6 \cdot 10^2 \pm 0.08 \cdot 10^2$
^{137}Cs	$6.7 \cdot 10^4 \pm 0.2 \cdot 10^4$	3.7 ± 0.2	$9.1 \cdot 10^4 \pm 0.26 \cdot 10^4$
^{152}Eu	6.4 ± 0.8	b.d.l.	b.d.l.
^{154}Eu	15 ± 0.6	0.18 ± 0.03	9.3 ± 1.1
^{155}Eu	b.d.l.	b.d.l.	b.d.l.

b.d.l. below detection limit

Table 6-1: Results of the measurement of condensation plates placed on a distance of 15 cm from the HPGe detector, given with the associated combined uncertainty ($k=1$).

Radionuclide	Activity ratio of sample to detector distance, 15 cm/ 0 cm		
	AE732100-2	AE732100-5	AE732100-6
^{54}Mn	0.99 ± 0.04		0.98 ± 0.06
^{60}Co	0.84 ± 0.005		0.46 ± 0.03
$^{110\text{m}}\text{Ag}$			0.47 ± 0.12
^{125}Sb	0.86 ± 0.17		0.32 ± 0.09
^{134}Cs	0.75 ± 0.02	0.43 ± 0.03	0.70 ± 0.02
^{137}Cs	1.00 ± 0.03	0.47 ± 0.02	0.92 ± 0.03
^{152}Eu	2.0 ± 0.33		
^{154}Eu	0.91 ± 0.04		0.68 ± 0.1
^{155}Eu			

Table 6-2: Ratio of the activities obtained for two different sample to detector distances, together with the associated combined uncertainty ($k=1$).

ITU-project number is 22835 and the internal project no. HCT/004/05.

7 References

[Carbol 2006]

CARBOL, P.

Report: JRC-ITU-TN-2006/

JRC - Institute for Transuranium Elements Karlsruhe, 2006

[Duwe, Kühnlein, 1975]

DUWE, R. ; KÜHNLEIN, W.

Berechnung des Spaltproduktinventars in Brennelementen mit U 235, Th 232 und U 238 als Schwermetalleinsatz

Institut für Reaktorwerkstoffe Kernforschungsanlage Jülich GmbH, 1975

[IAEA 1997]

Fuel Performance and Fission Product Behaviour in Gas Cooled Reactors

IAEA-TECDOC-978, Wien 1997

[Nabielek, Pitzer, Schenk 1986]

NABIELEK, H. ; PITZER, D. ; SCHENK, W.

Spaltproduktfreisetzungsvorlauf von Kugelbrennelementen bei Störfall-temperaturen

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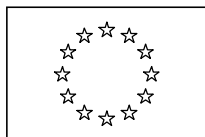
Institut für Reaktorwerkstoffe Kernforschungsanlage Jülich GmbH, 1986

[Kugeler, Schulten 1989]

KUGELER, K. ; SCHULTEN, R.

Hochtemperaturreaktortechnik HTR

Berlin; Heidelberg; New York; London; Paris; Tokyo; Hong Kong: Springer, 1989



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Institute for Transuranium Elements
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Title

KüFA Heating Test of a HTR Fuel Element

Fuel Element Identification No.: AVR 74/18
Internal identification No.: AE-741800

Author(s)

D. Freis and E. H. Toscano

Report Nr.:

JRC-ITU-TPW-2006/XX

Classification:

Commercial-in-confidence

Type of report:

Technical Report

	Name	Date	Signature
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approved by the head of unit	Dr. J.P. Glatz		
released by the director	Prof. T. Fanghänel		

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KüFA Heating Test of HTR Pebble AE-741800

Identification Nr.: AE-741800

Date: 17.01.2006

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1 Objective

Objective of the experiment was the testing of the AVR (Arbeitsgemeinschaft Versuchs Reaktor) Jülich fuel element: AVR 74/18 with the internal identification number AE-741800. It is a GLE-3 type spherical fuel element with a nominal burn up of 4,8 % fima. The heating test was performed in the KüFA device in Wing B, Hot Cell number 103. Beginning of the test was January the 18th 2006. The pebble was irradiated 21 years ago (end of irr. is Febr. 1985) and therefore it was expected to release primary the long-lived fission products, ^{137}Cs , ^{134}Cs , ^{85}Kr and ^{90}Sr . The parameters of the test (temperature and duration) were below the critical limits of the fuel element and so no failure of Coated Particles was expected and the fractional release of the fission products should be in a range of 10^{-6} to 10^{-5} for ^{137}Cs and 10^{-7} to 10^{-6} for ^{85}Kr and up to 10^{-5} for ^{90}Sr .

2 Fuel Element Data

2.1 General Data

Ident.Nr.: AE-741800

Type of sphere	GLE-3	Kernel composition	UO ₂
Type of graphite	Nukem A3-27	Enrichment	9,82% U-235
Particles in fuel element	16400	Kernel diameter	500 ± 2 µm
Heavy metal weight	10 g	Kernel density	10,80 g/cm ³

Coating

Buffer layer thickness	93 µm ± 14%	Buffer PyC layer density	1,01 g/cm ³
Inner PyC layer thickness	38 µm ± 10%	Inner PyC layer density	1,86 g/cm ³
SiC layer thickness	35 µm ± 6%	SiC layer density	3,19 g/cm ³
Outer PyC layer thickness	40 µm ± 9%	Outer PyC layer density	1,89 g/cm ³

Irradiation history

Burn up	4,80% fima	Time of Irradiation	480 efpd
Therm. flux	4,15 · 10 ²¹ cm ²	End of irradiation	Feb. 1985
Fast flux (>0,1 MeV)	8 · 10 ²⁰ cm ²	Irradiation temperature	ca. 700°C

Pebble inventory (est.) at beginning of the test (17.01.2006)

⁸⁵ Kr	1,45 · 10 ⁹ Bq	¹³⁷ Cs	3,4 · 10 ¹⁰ Bq
⁹⁰ Sr	2,89 · 10 ¹⁰ Bq	¹³⁴ Cs	1,77 · 10 ⁷ Bq
¹³¹ I	~ 0 Bq	^{110m} Ag	0,079 Bq

2.2 Burn up and Inventory calculation

The burnup was calculated using the computer program Inventar by W. Kühnlein [Ref. 2]. The values were calculated for the end of irradiation.

The Inventar code is written in FORTRAN and is based on the analytical solutions of the differential equations for the build up and burning of the main heavy metals and fission products in the AVR. For this reason, the values reported in Table 2-1 should be considered as an approximation.

SPALTPRODUKTINVENTAR AE-741800						
3	NUKLID	3	ATOMZAHL	3	AKTIVITAET	3
3		3		3	Bq	3
3		3		3	MIKROCURIE	3
3	KR 85	3	2.89E+18	3	5.87E+09	3
3	SR 89	3	6.81E+18	3	1.05E+12	3
3	SR 90	3	6.22E+19	3	4.86E+10	3
3	ZR 95	3	1.30E+19	3	1.60E+12	3
3	RU 103	3	5.08E+18	3	1.03E+12	3
3	RU 106	3	1.04E+19	3	2.27E+11	3
3	AG 110M	3	8.49E+15	3	2.69E+08	3
3	I 131	3	8.33E+17	3	8.31E+11	3
3	XE 133	3	1.12E+18	3	1.71E+12	3
3	CS 134	3	2.35E+18	3	2.51E+10	3
3	CS 137	3	7.66E+19	3	5.60E+10	3
3	BA 140	3	2.56E+18	3	1.60E+12	3
3	CE 141	3	6.48E+18	3	1.60E+12	3
3	CE 144	3	3.76E+19	3	1.06E+12	3

3	ABBRAND			3
3	ABBRAND	U	233	3
3			.00 % FIMA	3
3	ABBRAND	U	235	3
3			3.90 % FIMA	3
3	ABBRAND	PU	239	3
3			.85 % FIMA	3
3	ABBRAND	PU	241	3
3			.07 % FIMA	3
3	GESAMTABBRAND			3
3			4.81 % FIMA	3

Table 2-1: Burn up calculation for AE-741800 with Inventar

3 Calibration and Test Preparation

3.1 Cold Trap Calibration:

Before the test the cold traps were cooled down with liquid nitrogen and a 15 h Background measurement for the gamma detectors was performed.

The calibration of the first cold trap was performed before, during and after the first phase of the heating test with three aliquots of ^{85}Kr which were brought in through a bypass in the piping leading to the hot cell.

The aliquots were taken from a container, where a certificated standard (about 5×10^5 Bq) was broken, under a known He-pressure, in a known volume from which, using another container, the aliquots were taken. The volumes of the two containers were certified.

Volume of container 1: 3931.37 ml
Volume of container 2: 84.455 ml

The aliquot was certified to have an activity of 5,493,000 Bq at 20.09.2002. The correction for the calibration was made using the ratios of the container volumes and under consideration of the decay law.

$$A = A_{cf} \cdot \frac{V_2}{V_2 + V_1} \cdot \left(\frac{V_1}{V_2 + V_1} \right)^{n-1} \cdot e^{(-\lambda \cdot t_{cf})}$$

A_{cf} = activity of certified standard , t_{cf} = time since certification

V_2 and V_1 = volumes of containers

n = number of aliquot taken from container 1

3.1.1 Calibration 1

Date and time	17.01.2006 / 12:00
Activity in sample	73.080 Bq
Impulses per second	9,858
Calibration factor	$1,35 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$

Table 3-1: cold trap calibration 1

3.1.2 Calibration 2

Date and time	30.01.2006
Activity in sample	71.745 Bq
Impulses per second	8,487
Calibration factor	$1,18 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$

Table 3-2: cold trap calibration 2

The average calibration factor amounts to $1,27 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$.

3.2 Test of the gas circuit

After the test also a test of the whole circuit was done with an aliquot of ^{85}Kr of 70.155 Bq which was inserted in the gas pipe before the furnace. No losses of ^{85}Kr were recognized.

The theoretical activity increase in the cold trap can be described with the following equations. A_B is the activity in the sample container with the Volume (V_B). It is surveyed with the nominal gas stream of Helium ($\dot{V}_{He}$).

Sample container:

$$\frac{dA_B}{dt} = -\frac{\dot{V}_{He}}{V_B} \cdot A_B \quad \text{Equation 3-1}$$

Initial Condition:

$$A_B(t=0) = A_0$$

Solution:

$$A_B(t) = A_0 \cdot e^{\left(-\frac{\dot{V}_{He}}{V_B} \cdot t\right)} \quad \text{Equation 3-2}$$

Furnace:

$$\frac{dA_G}{dt} = -\frac{\dot{V}_{He,G}}{V_G} \cdot A_G - \frac{dA_B}{dt} \quad \text{Equation 3-3}$$

Initial Condition:

$$A_G(t=0) = 0$$

Solution:

$$A_G(t) = \frac{A_0}{\left(\frac{\dot{V}_{He,G}}{\dot{V}_{He}} \cdot \frac{V_B}{V_G}\right) - 1} \cdot \left(e^{-\frac{\dot{V}_{He}}{V_B} \cdot t} - e^{-\frac{\dot{V}_{He,G}}{V_G} \cdot t} \right) \quad \text{Equation 3-4}$$

Cold Trap:

$$\frac{dA_T}{dt} = \frac{\dot{V}_{He,G}}{V_G} A_G \quad \text{Equation 3-5}$$

Initial condition:

$$A_T(t=0) = 0$$

Solution:

$$A_T(t) = A_0 \cdot \left(1 - \left(\frac{V_G \cdot \dot{V}_{He} \cdot e^{\left(-\frac{\dot{V}_{He,G}}{V_G} \cdot t \right)} + \frac{V_B \cdot \dot{V}_{He,G} \cdot e^{\left(-\frac{\dot{V}_{He}}{V_B} \cdot t \right)}}{V_G \cdot \dot{V}_{He} - V_B \cdot \dot{V}_{He,G}} + \frac{V_B \cdot \dot{V}_{He,G} \cdot e^{\left(-\frac{\dot{V}_{He}}{V_B} \cdot t \right)}}{V_B \cdot \dot{V}_{He,G} - V_G \cdot \dot{V}_{He}} \right) \right) \quad \text{Equation 3-6}$$

The circulating gas is heated in the furnace and subsequently expanded. This caused a higher flow than the nominal flow of 30 l/h.

Activity of the aliquot:	70.155 Bq	Volume of the sample container:	85 cm ³
Furnace temperature:	ca. 1200°C	Volume of the furnace:	28.600 cm ³

Nominal Helium flow ($\dot{V}_{He}$): 30 l/h

Estimated Helium flow through oven ($\dot{V}_{He,G}$): 2 – 3 times $\dot{V}_{He}$

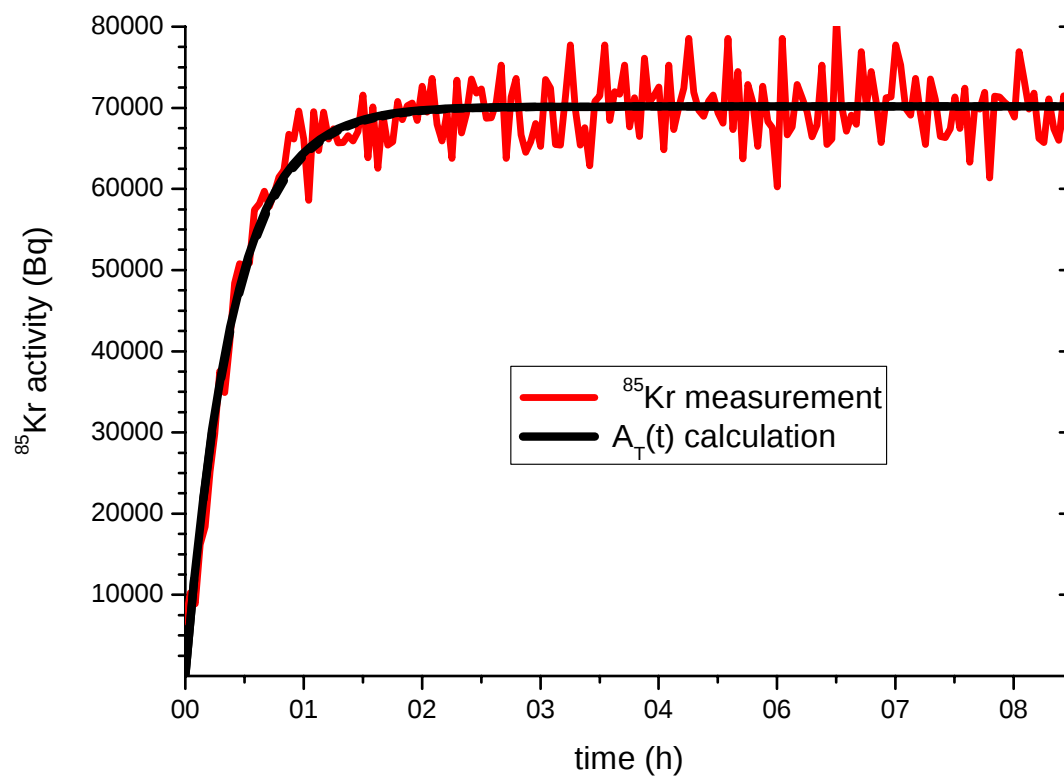


Figure 3-1: Calibration curve after test

3.3 Calibration of the cold finger

The calibration of the cold finger was performed using non-radioactive Cs. A known amount of a solution containing a known amount of Cs was placed in a Ti-plate and allowed to dry. After that, the Ti-plate was placed in the furnace and heated to different temperatures. Afterwards, the cold finger condensate plates were leached in a 2 M nitric acid solution and the leachates analysed by ICP-MS. The overall efficiency of the cold-finger for Cs was evaluated to be around 70% as was determined in previous calibrations performed in Fz-Jülich. More work is under way, using certified standards, to determine the efficiency at different temperatures.

3.4 Test Preparations

The test preparations were performed as described in the working instruction for the KüFA device.

1. A new and not contaminated tantalum tube was installed in the furnace.
2. A new condensate plate was installed at the cooling finger.
3. A clean cold trap was included in the helium circuit and cooled down with liquid nitrogen. A second new trap was prepared and inserted in the circuit behind the first one.
4. The pebble was brought in into the device.
5. The ^{85}Kr scanning for both traps was prepared and started.
6. The heating program was written and started.
7. Before and after the test, an aliquot of ^{85}Kr was inserted into the first cold trap through a bypass for calibration.

4 Test procedure

The heating experiment AE-741800 was the second preliminary test of the KüFA device under hot conditions. The test program was divided into three steps. The first was heating up to 1000°C for 10 h to consolidate the same equilibrium conditions as in operation. Then followed an accident simulation up to 1600°C and after 100 h the pebble was cooled down to room temperature. Finally the pebble was heated up to 1800°C for 100 h.

Before the first heating step the pebble was held at 300°C for 10 hours to clean the system of possible impurities.

The condensate plates were changed after 50 h and 100 h at 1600°C and daily at 1800°C.

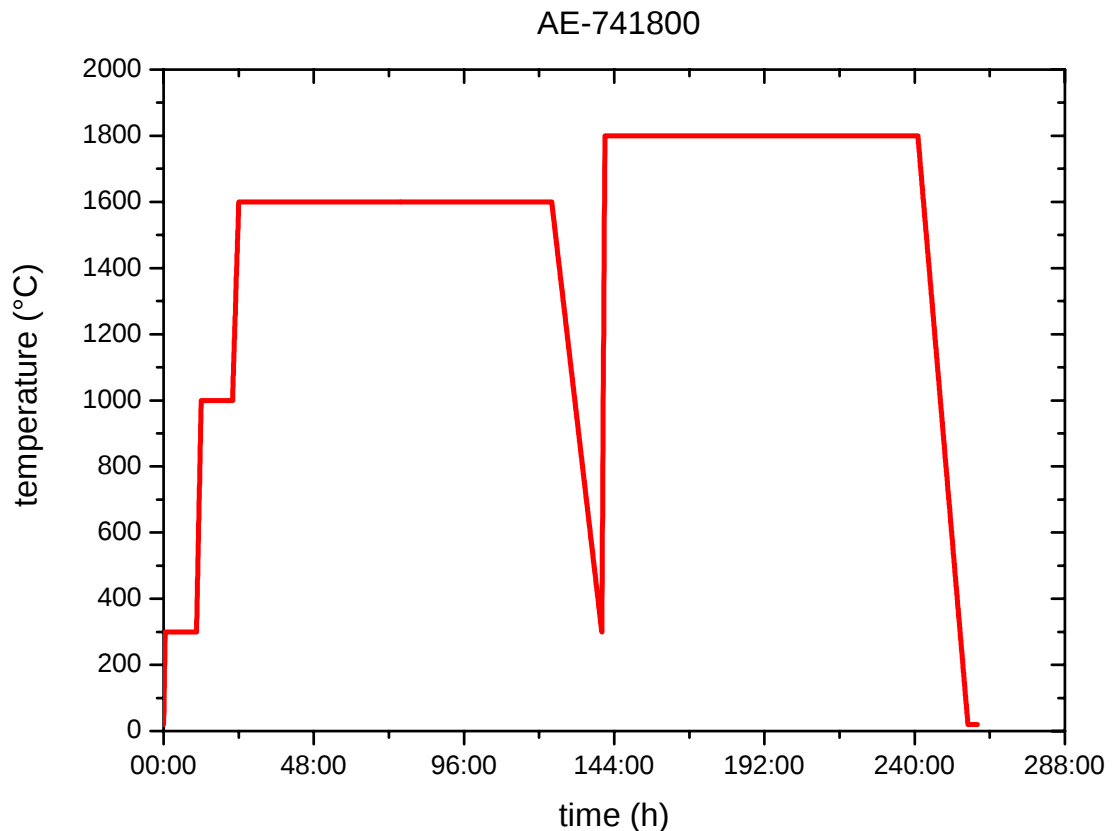


Figure 4-1: heating program AE-741800

Time (hh:mm)	Endurance (min)	Temperature (°C)	Gain (°C/h)	Plate
00:00		20		3
	30		560	
00:30		300		3
	600		0	
10:30		300		3
	89		797.75	
11:59		1000		3
	564		0	
21:25		1000		3 - 4
	34		0	
21:59		1000		4
	120		300	
23:59		1600		4
	71		0	
25:10		1600		4 - 7
	60		0	
26:10		1600		7
	120		-790*	
28:10		20		7
	-		-	
-		-		7
	-		-	
28:10		20		7 - 10
	17		988.24	
28:27		300		10
	60		0	
29:27		300		10
	89		471.91	
30:56		1000		10
	120		300	
32:56		1600		10
	2924		0	
81:40		1600		10 - 9
	3516		0	
140:16		1600		9
	444		-86.96	
147:40		935		9 - 11
	453		-86.96	
155:13		300		11
	60		0	
156:13		300		11
	90		1000	
157:43		1800		11

Table 4-1: heating program AE-741800

* averaged value, the cooling in the marked intervals was uncontrolled and is therefore described by an exponential function

Time (hh:mm)	Endurance (min)	Temperature (°C)	Gain (°C/h)	Plate
157:43		1800		11
	837		0	
171:40		1800		11 - 12
	1440		0	
195:40		1800		12 - 13
	1440		0	
219:40		1800		13 - 14
	395		0	
226:15		1800		14
	85		-401.65*	
227:40		1231		14
	30		1138	
228:10		1800		14
	930		0	
243:40		1800		14 - 15
	870		0	
258:10		1800		15
	960		-111.25	
274:10		20		15

Table 4-2: heating program AE-741800

* averaged value, the cooling in the marked intervals was uncontrolled and is therefore described by an exponential function

At 28:10 h and at 226:15 h an alert occurred and the apparatus was shut down automatically by the control system. In the first case the plate temperature was, due to graphite deposition, higher than the allowed value. The test was paused until this problem was solved. In the second case the alert was caused by a bubble in the cooling system. The system could be restarted within 85 minutes after the alert.

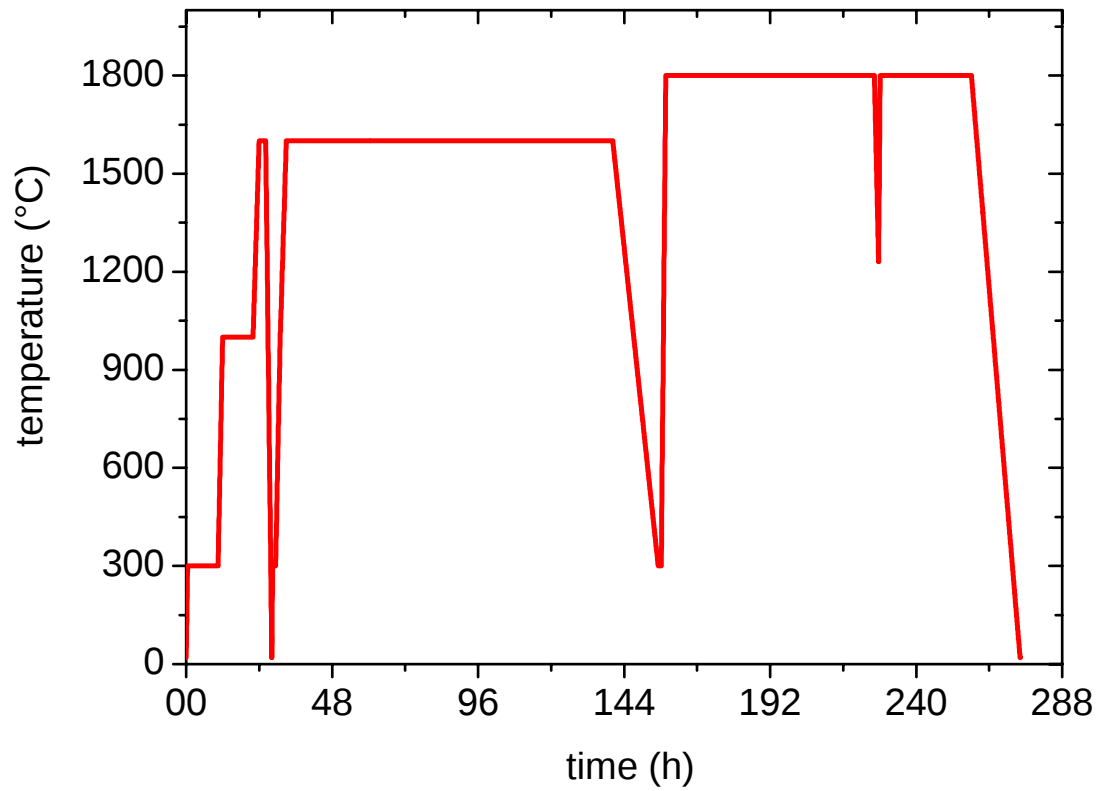


Figure 4-2: Temperature Gradient of AE-741800

5 Results

5.1 Fission gas (^{85}Kr) release

The total ^{85}Kr release during the test was about 8,500 Bq which is equivalent to a fractional release of $6.05 \cdot 10^{-6}$ of the inventory.

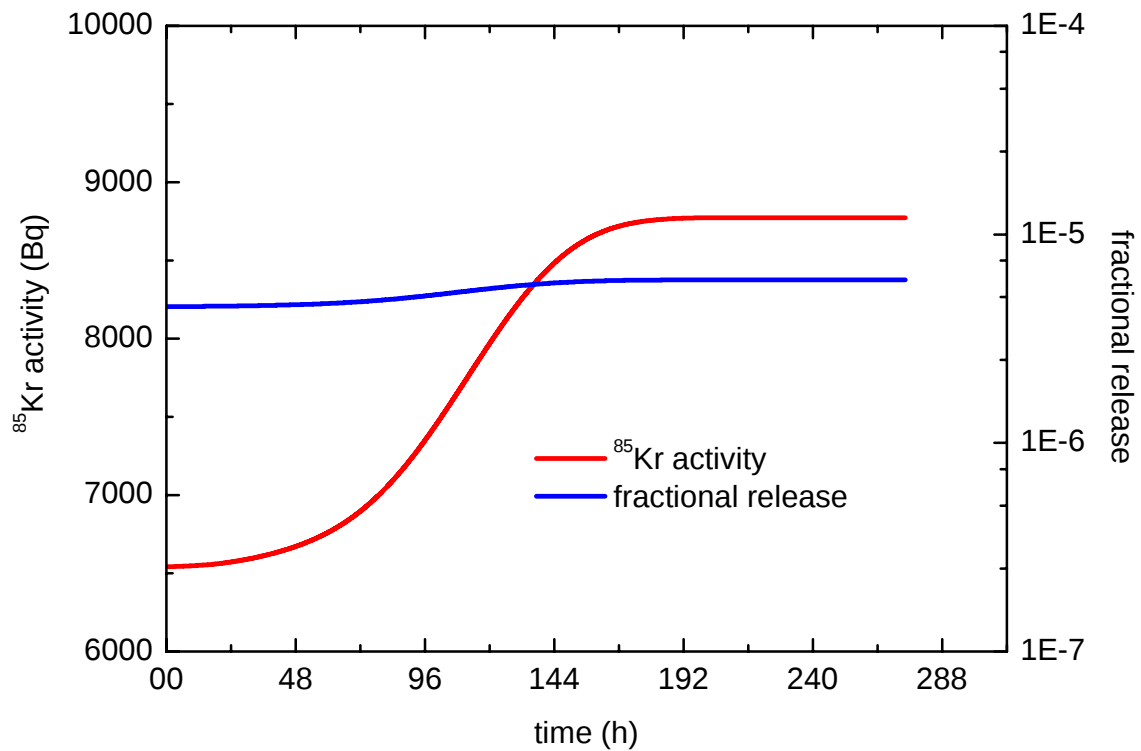


Figure 5-1: ^{85}Kr release in Bq and fractional release

In the second cold trap no increase of activity was detected, corroborating that all the whole activity released was retained in the first cold trap.

5.2 Release of solid fission products

5.2.1 Gamma spectrometry of the condensate plates

Subject: Gamma-spectrometry for project number . (KüFA, AE-741800, see detailed information in [Carbol 2006])

Objective of the gamma measurements were two condensate plates of the KüFA device in Wing B, Hot Cell 103. These plates were used in the heating and fission product release experiment of a burned up spherical fuel element (Identification Nr.: AE-741800) from the AVR reactor. [Kugeler, Schulten 1989] Since the pebble was irradiated 21 years ago (end of irr. Febr. 1985) it was expected to release primary the long-lived fission products, ^{137}Cs , ^{134}Cs , ^{85}Kr and ^{90}Sr . The parameters of the test (temperature and duration) were below the critical limits of the fuel element and therefore the gamma measurements should show a relative small activity of about 10^{-6} to 10^{-5} fractional release of ^{137}Cs (20.000 to 180.000 Bq for ^{137}Cs) and up to 10^{-5} fractional release of ^{90}Sr (160.000 Bq for ^{90}Sr).

Plate	^{137}Cs activity (Bq)	Uncertainty (Bq)	measurement date
3	7.698E+03	2.531E+02	04.08.06
4	1.267E+04	1.661E+02	03.08.06
7	6.679E+03	2.237E+02	04.08.06
10	8.235E+04	1.555E+03	02.08.06
9	2.871E+04	2.500E+02	03.08.06
11	1.036E+04	2.233E+02	04.08.06
12	7.162E+03	2.424E+02	03.08.06
13	5.803E+03	2.113E+02	03.08.06
14	1.140E+04	2.172E+02	02.08.06
15	1.874E+04	3.018E+02	04.08.06

Table 5-1: Results of the measurement of condensation plates placed on a distance of 15 cm from a 3x3 NaJ detector, reported with the associated combined uncertainty.

The results of the gamma measurements of the plates are finally corrected through the efficiency factor of the cooling finger for Caesium (70%) respectively Strontium (20%).

^{90}Sr will be measured in the second phase of the project by leaching of the condensate plates and the subsequently measurement by IC-ICP-MS.

5.2.2 Release of ^{137}Cs

The ^{137}Cs release is shown in Fig. 5-4, where the absolute activity and the fractional release are plotted as a function of time. The absolute values of the gamma measurements are gathered in Table 5-1.

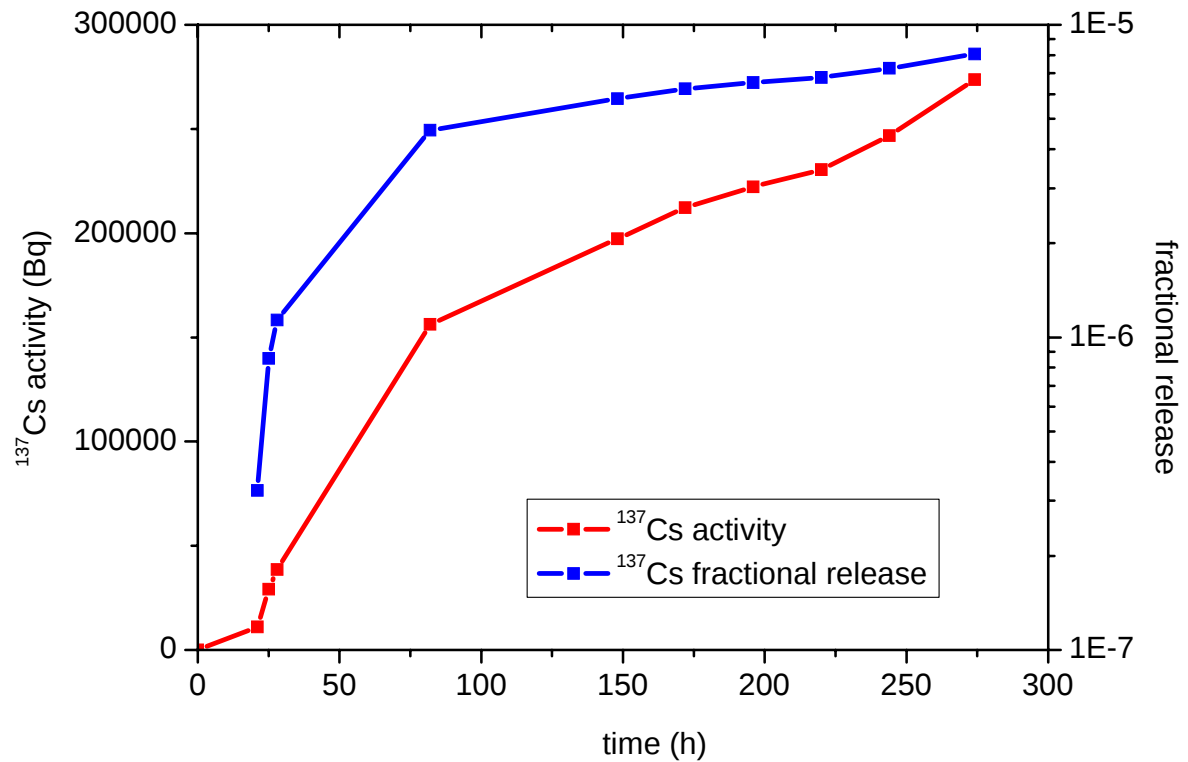


Figure 5-2: ^{137}Cs release of AC-741800

5.3 Summary of the results

All the results of the present test are summarised in Fig. 5-5, where the fractional for ^{85}Kr and ^{137}Cs are depicted as a function of the test time together with the temperature profile in the different phases of the test.

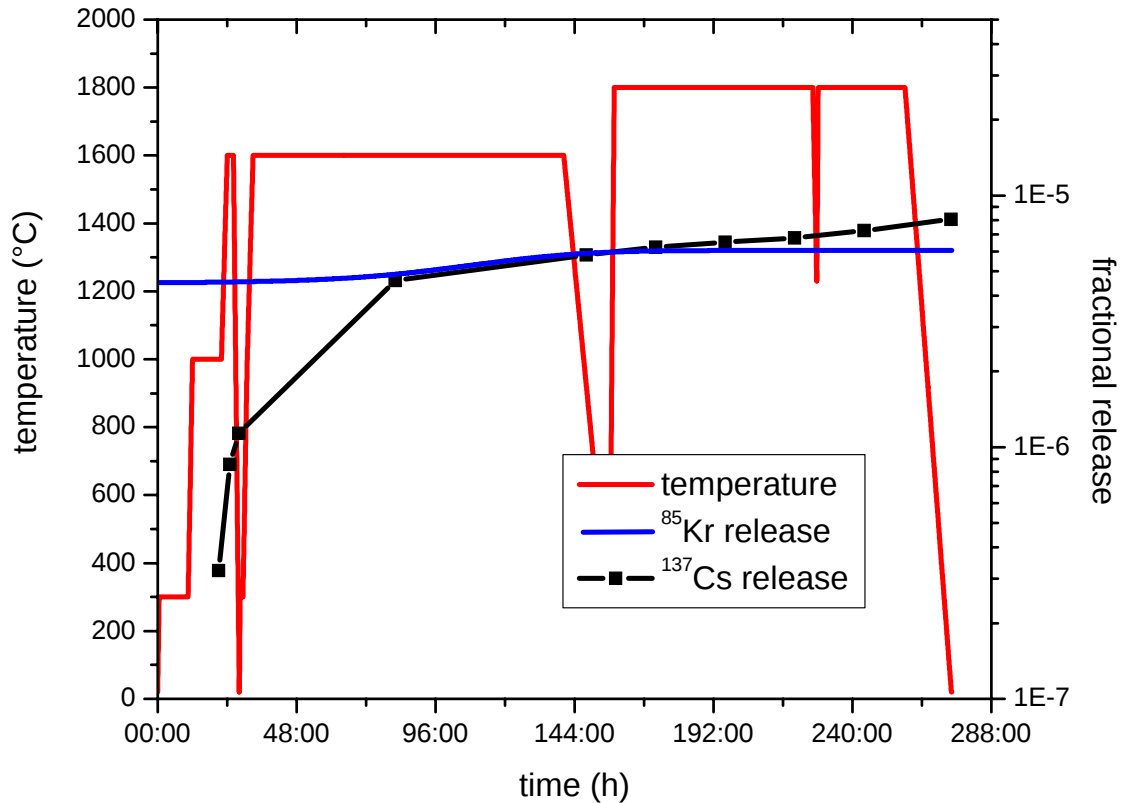


Figure 5-3: fission product release of AVR 74/18

Conclusions

The fuel element AVR 74/18 (AE-741800) was tested in the KÜFA-installation in Hot Cell 103, Wing B. In the first phase of the test, comprising 100 h at 1600°C, almost no release of the gaseous fission products was measured, whereas the Cs-release started to be measurable from the first plates and increased with increasing testing temperatures.

In the second phase of the test, comprising 100 h at 1800°C, a slight release of fission gas in the range of 10^{-5} fractional release was measured. Cs increased further.

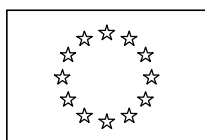
The observed behaviour is either due to increasing release to enhanced transport through the coating layers of many particles or is the consequence of damage induced in the coatings of a few particles. No pressure vessel failure was observed.

The present is the first revision of this report. A further revision will be issued when:

- a. More accurate inventory calculations will be provided by Fz-Jülich in the framework of the RAPHAEL-IP
- b. A better calibration of the cold finger will be performed using certified standards
- c. Further results for ^{90}Sr and ^{109}Ag will be available from the leaching of the plates and subsequently analysis by ICP-MS of the leachates

References

1. Duwe, Kühnlein, 1975
DUWE, R. ; KÜHNLEIN, W.
Berechnung des Spaltproduktinventars in Brennelementen mit U 235, Th 232 und U 238 als Schwermetalleinsatz
Institut für Reaktorwerkstoffe Kernforschungsanlage Jülich GmbH, 1975
2. IAEA 1997]
Fuel Performance and Fission Product Behaviour in Gas Cooled Reactors
IAEA-TECDOC-978, Wien 1997
3. Nabielek, Pitzer, Schenk 1986
NABIELEK, H. ; PITZER, D. ; SCHENK, W.
Spaltproduktfreisetzungsvorlauf von Kugelbrennelementen bei Störfall-
temperaturen
Als Manuskript gedruckt
Institut für Reaktorwerkstoffe Kernforschungsanlage Jülich GmbH, 1986
4. Kugeler, Schulten 1989]
KUGELER, K. ; SCHULTEN, R.
Hochtemperaturreaktortechnik HTR
Berlin; Heidelberg; New York; London; Paris; Tokyo; Hong Kong: Springer, 1989
5. D. Freis
PhD-Thesis, in preparation



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Date: 11.09.2006
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Title

KüFA Heating Test of a HTR Fuel Element

Fuel Element Identification No.: HFR K6/2
Internal identification No.: AC-065600

Author(s)

D. Freis and E. H. Toscano

Report Nr.:

JRC-ITU-TPW-2006/XX

Classification:

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Technical Report

	Name	Date	Signature
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released by the director	Prof. T. Fanghänel		

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KüFA Heating Test of HTR Pebble HFR K6/2

Identification Nr.: HFR K6/2

Date: 15.07.2006

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1 Background

Objective of the experiment was the spherical fuel element HFR K6/2 with the internal identification number AC-065600. The FE is a GLE-4 similar type spherical fuel element which was specially designed for the irradiation in the High Flux reactor Petten (HFR) to test the fuel elements for the planned HTR-MODUL. It was irradiated up to a burn-up of ca. 9.7 % FIMA.

The heating test was performed in the KüFA device in Wing B, Hot Cell number 103. Beginning of the test was July the 15th 2006 at 15:30. Since the FE was irradiated in 1993 (end of irradiation was May 1993) it was expected to release primary the long-lived fission products ^{137}Cs , ^{134}Cs , ^{85}Kr and ^{90}Sr .

The test parameters (temperature and duration) were selected to be at the beginning below the critical limits of the fuel element. Consequently, no failure of Coated Particles (CPs) should occur during the first part of the test and the fractional release (FR) of the fission products should be in a range of 10^{-6} to 10^{-5} for ^{137}Cs and 10^{-7} to 10^{-6} for ^{85}Kr , whereas for ^{90}Sr up to 10^{-5} FR was expected.

2 Fuel Element Data

2.1 General Data

Ident.Nr.: HFR K6/2

Type of sphere	GLE-4 similar	Kernel composition	UO ₂
Type of graphite	Nukem A3-27	Enrichment	10.6% U-235
Particles in fuel element	14,600	Kernel diameter	500 ±2 µm
Heavy metal weight	9.44 g	Kernel density	10,80 g/cm ³

Coating

Buffer layer thickness	93 µm ± 14%	Buffer PyC layer density	1,01 g/cm ³
Inner PyC layer thickness	38 µm ± 10%	Inner PyC layer density	1,86 g/cm ³
SiC layer thickness	35 µm ± 6%	SiC layer density	3,19 g/cm ³
Outer PyC layer thickness	40 µm ± 9%	Outer PyC layer density	1,89 g/cm ³

Irradiation history

Burn up	9.3% fima	Time of Irradiation	633 efpd (26 cycles)
Thermal flux	$2.5 \cdot 10^{21} \text{ cm}^{-2}$	End of irradiation	May 1993
Fast flux (>0,1 MeV)	$4.8 \cdot 10^{21} \text{ cm}^{-2}$	Irradiation temperature	ca. 1140°C

Pebble inventory (est.) at beginning of the test (06.07.2006)

⁸⁵ Kr	$4.29 \cdot 10^9 \text{ Bq}$	¹³⁷ Cs	$7.81 \cdot 10^{10} \text{ Bq}$
⁹⁰ Sr	$6.29 \cdot 10^{10} \text{ Bq}$	¹³⁴ Cs	$1.47 \cdot 10^9 \text{ Bq}$
¹³¹ I	~ 0 Bq	^{110m} Ag	~ 0 Bq

2.2 Burn up and Inventory calculation

The burnup was calculated using the computer program Inventar by W. Kühnlein [Ref. 2]. The values were calculated for the end of irradiation.

The Inventar code is written in FORTRAN and is based on the analytical solutions of the differential equations for the build up and burning of the main heavy metals and fission products in the AVR. For this reason, the values reported in Table 2-1 should be considered as an approximation.

SPALTPRODUKTINVENTAR						
NUKLID	ATOMZAHL	AKTIVITAET		ZERF/SEK	MIKROCURIE	
KR 85	4.93E+18	1.00E+10	2.70E+05			
SR 89	4.19E+18	6.45E+11	1.74E+07			
SR 90	1.10E+20	8.64E+10	2.34E+06			
ZR 95	9.97E+18	1.23E+12	3.31E+07			
RU 103	4.83E+18	9.81E+11	2.65E+07			
RU 106	1.81E+19	3.97E+11	1.07E+07			
AG 110M	5.13E+16	1.63E+09	4.39E+04			
I 131	8.69E+17	8.66E+11	2.34E+07			
XE 133	1.20E+18	1.83E+12	4.94E+07			
CS 134	1.14E+19	1.22E+11	3.30E+06			
CS 137	1.44E+20	1.06E+11	2.86E+06			
BA 140	2.19E+18	1.37E+12	3.71E+07			
CE 141	4.77E+18	1.17E+12	3.17E+07			
CE 144	3.74E+19	1.05E+12	2.85E+07			

ABBRAND				
ABBRAND	U	233	.00 % FIMA	
ABBRAND	U	235	7.49 % FIMA	
ABBRAND	PU	239	2.02 % FIMA	
ABBRAND	PU	241	.51 % FIMA	
GESAMTABBRAND		10.02 % FIMA		

Table 2-1: Burn up calculation for HFR K6/2 using the code Inventar

3 Calibration and Test Preparation

3.1 Cold Trap Calibration:

Before the test the cold traps were cooled down with liquid nitrogen and a 15 h Background measurement for the gamma detectors was performed.

The calibration of the first cold trap was performed before, during and after the first phase of the heating test with three aliquots of ^{85}Kr which were brought in through a bypass in the piping leading to the hot cell.

The aliquots were taken from a container, where a certificated standard (about 5×10^5 Bq) was broken, under a known He-pressure, in a known volume from which, using another container, the aliquots were taken. The volumes of the two containers were certified.

Volume of container 1: 3931.37 ml
Volume of container 2: 84.455 ml

The aliquot was certified to have an activity of 5,493,000 Bq at 20.09.2002. The correction for the calibration was made using the ratios of the container volumes and under consideration of the decay law.

$$A = A_{cf} \cdot \frac{V_2}{V_2 + V_1} \cdot \left(\frac{V_1}{V_2 + V_1} \right)^{n-1} \cdot e^{(-\lambda \cdot t_{cf})}$$

A_{cf} = activity of certified standard , t_{cf} = time since certification

V_2 and V_1 = volumes of containers

n = number of aliquot taken from container 1

3.1.1 Calibration 1

Date and time	06.07.2006 / 17:00
Activity in sample	55,800 Bq
Impulses per second	6.75
Calibration factor	$1.21 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$

Table 3-1: cold trap calibration 1

3.1.2 Calibration 2

Date and time	10.07.2006 / 11:37
Activity in sample	53,600 Bq
Impulses per second	6.27
Calibration factor	$1.17 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$

Table 3-2: cold trap calibration 2

The average calibration factor amounts to $1.19 \cdot 10^{-4} \text{ Bq}^{-1} \text{ s}$.

3.2 Calibration of the cold finger

The calibration of the cold finger was performed using non-radioactive Cs. A known amount of a solution containing a known amount of Cs was placed in a Ti-plate and allowed to dry. After that, the Ti-plate was placed in the furnace and heated to different temperatures. Afterwards, the cold finger condensate plates were leached in a 2 M nitric acid solution and the leachates analysed by ICP-MS. The overall efficiency of the cold-finger for Cs was evaluated to be around 70% as was determined in previous calibrations performed in Fz-Jülich. More work is under way, using certified standards, to determine the efficiency at different temperatures.

3.3 Test Preparations

The test preparations were performed as described in the working instruction for the KüFA device.

1. A new and not contaminated tantalum tube was installed in the furnace.
2. A new condensate plate was installed at the cooling finger.
3. A clean cold trap was included in the helium circuit and cooled down with liquid nitrogen. A second new trap was prepared and inserted in the circuit behind the first one.
4. The pebble was brought in into the device.
5. The ^{85}Kr scanning for both traps was prepared and started.
6. The heating program was written and started.
7. Before, during and after the test, three aliquots of ^{85}Kr were inserted into the circuit through a bypass to calibrate the cold trap.

4 Test procedure

The heating of pebble HFR K6/2 was the fourth test of the KüFA under remote controlled conditions. The heating program was divided into three steps and went over 400 h. The first step was heating up to 1600°C for 100 h and cooling down to room temperature. Then followed a phase 1700°C for 100 h and finally the pebble was heated up to 1800°C for 100 h.

In table 4.1 the details of the heating programs are given together with the plate changing period. Figures 4.1 and Figure 4.2 show schematically the heating schedules.

Due to problems with the cooling system, in one case, and with the software in the second case, the test had to be interrupted for some hours. The interruption caused an abrupt cooling of the FE (in about 1 hour) in both cases. The two periods are marked (*) in Table 4-1.

Time (hh:mm)	Step Duration (min)	Temperature (°C)	Heating rate or dwells (°C/h)	Plate No.
00:00		20		50
	30		560	
00:30		300		50
	600		0	
05:30		300		50
	90		500	
07:00		1050		50
	560		0	
16:00		1050		50
	60		46	
17:00		1096		50 - 51
	360		46	
23:00		1372		51 - 52
	300		46	
28:00		1600		52
	1020		0	
45:00		1600		52
	60		-1580*	
46:00		20		52 - 53
	30		560	
46:30		300		53
	90		500	
48:00		1050		53
	840		0	
62:00		1050		53
	60		550	
63:00		1600		53 - 54
	1440		0	
87:00		1600		54 - 55
	180		0	
90:00		1600		55

Table 4-1: heating program HFR K6/2

Time (hh:mm)	Step Duration (min)	Temperature (°C)	Heating rate or dwells (°C/h)	Plate No
90:00		1600		55
	60		-1580*	
91:00		20		55 - 56
	30		560	
91:30		300		56
	90		866.67	
93:00		1600		56
	1200		0	
113:00		1600		56 - 57
	1500		0	
138:00		1600		57 - 58
	780		0	
151:00		1600		58
	240		50	
155:00		1800		58 - 59
	1620		0	
182:00		1800		59 - 60
	360		0	
188:00		1800		60 - 61
	960		0	
204:00		1800		61 - 62
	1620		0	
231:00		1800		62 - 63
	1260		0	
252:00		1800		63 - 64
	480		0	
260:00		1800		64 - 66
	960		0	
276:00		1800		66 - 67
	1440		0	
300:00		1800		67 - 68
	420		0	
307:00		1800		68 - 69
	960		0	
323:00		1800		69 - 70
	540		0	
332:00		1800		70 - 71
	900		0	
347:00		1800		71 - 72
	480		0	
355:00		1800		72
	960		-11.25	
371:00		20		72

Table 4-1: heating program HFR K6/2

* uncontrolled cooling

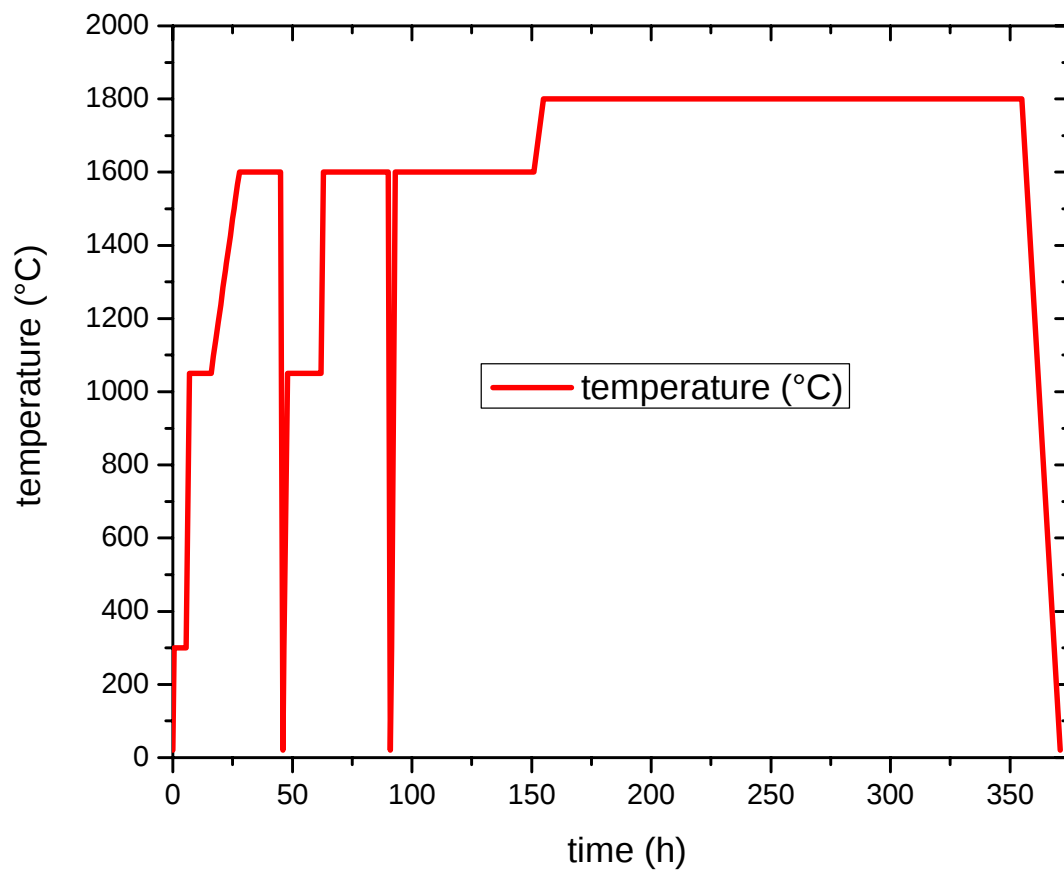


Figure 4-1: heating program for HFR K6/2

5 Results

5.1 Fission gas (^{85}Kr) release

The ^{85}Kr release during the first phase of the test was about 4.4×10^4 Bq which is equivalent to a fractional release of 10^{-5} of the inventory, as calculated using the Inventar-code (see Fig. 5-1).

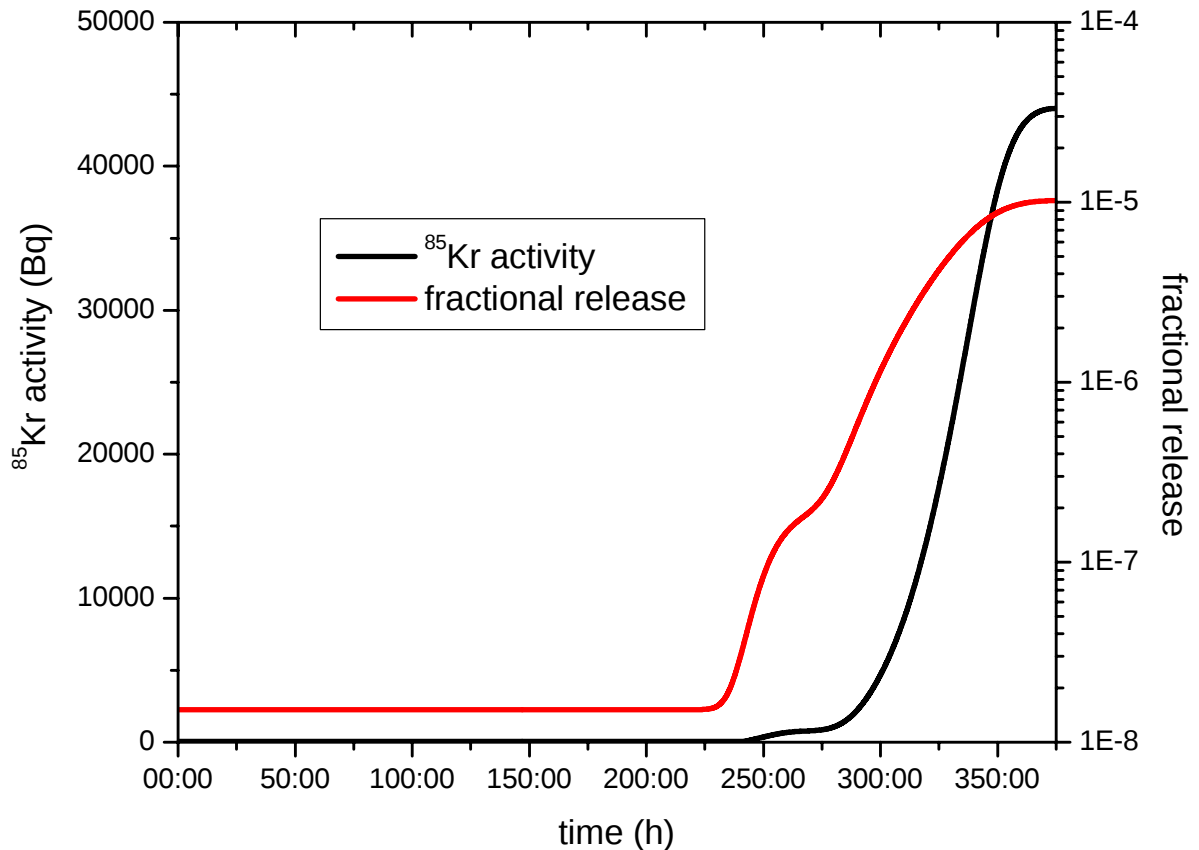


Figure 5-1: ^{85}Kr release in Bq and fractional release

In the second cold trap no increase of activity was detected, corroborating that all the whole activity released was retained in the first cold trap.

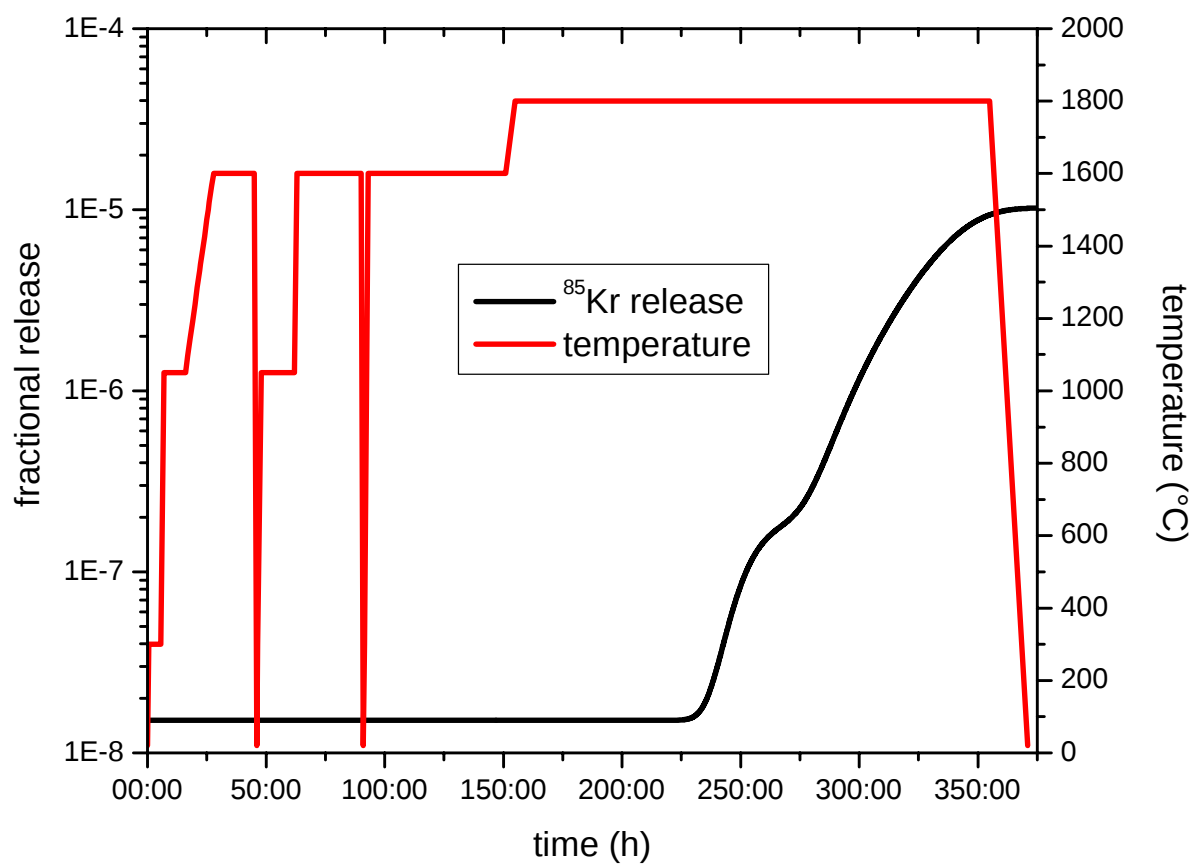


Figure 5-2: ^{85}Kr release and temperature gradient

5.2 Release of solid fission products

5.2.1 Gamma spectrometry of the condensate plates

Subject: Gamma-spectrometry for project number. (KüFA, HFR K6/2)

Objective of the gamma measurements were condensate plates of the KüFA device in Wing B, Hot Cell 103. These plates were used in the heating and fission product release experiment of a burned up spherical fuel element (Identification Nr.: HFR K6/2) from the HFR reactor Petten. Since the pebble was irradiated 13 years ago (end of irr. May 1993) it was expected to release primarily the long-lived fission products, for example ^{137}Cs and ^{90}Sr . The parameters of the test (temperature and duration) were beyond the nominal critical limits of the fuel element and therefore the gamma measurements were expected to show an activity of up to 10^{-1} fractional release of ^{137}Cs (ca. 10^{10} Bq ^{137}Cs) and up to 10^{-1} fractional release of ^{90}Sr (ca. 10^{10} Bq of ^{90}Sr).

The results of the gamma measurements of the plates are finally corrected through the efficiency factor of the cooling finger for Caesium (70%) respectively Strontium (20%).

5.2.2 Release of ^{137}Cs

The release of ^{137}Cs is shown in Fig. 5-3 and displayed in activity in Becquerel and in fractional release.

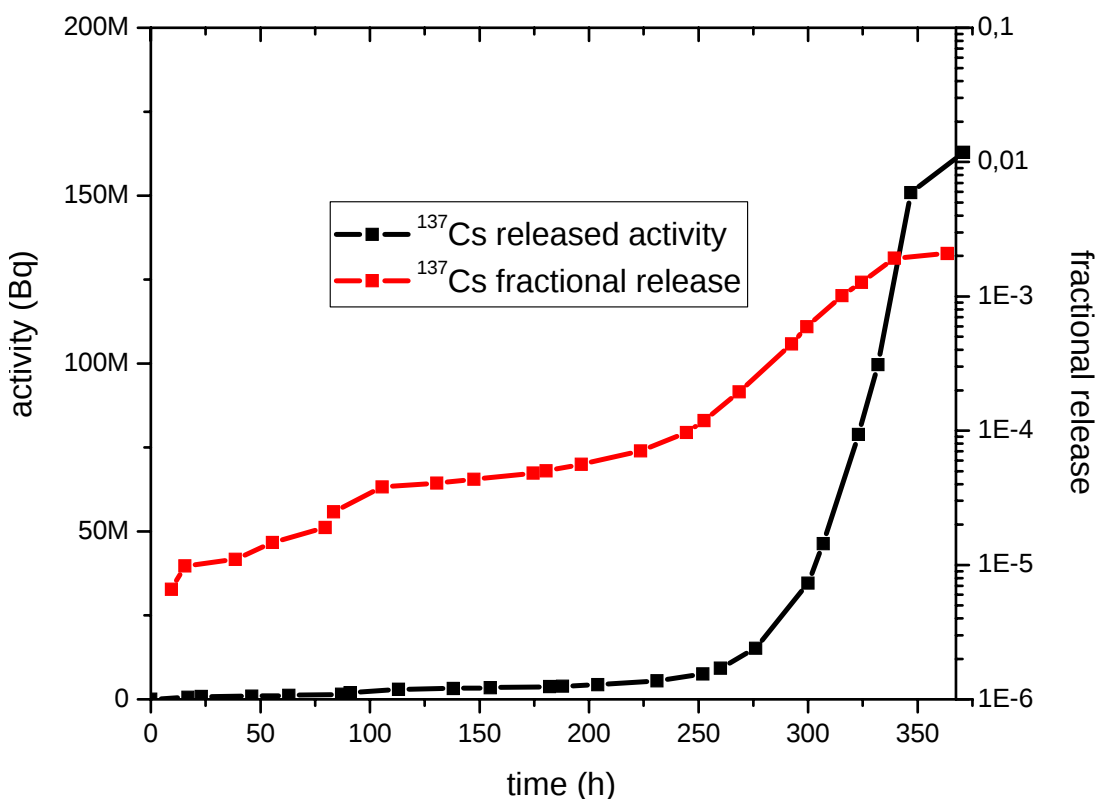


Figure 5-3: ^{137}Cs release of HFR K6/2

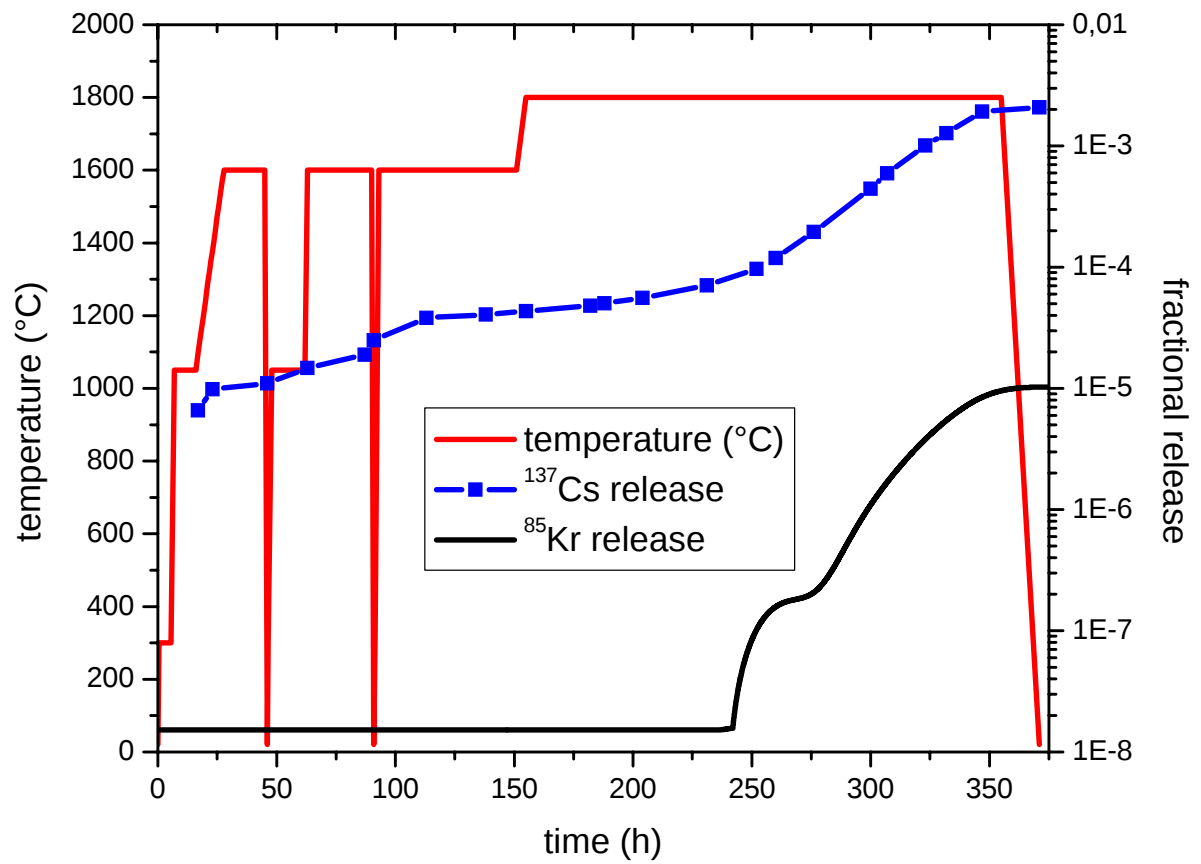


Figure 5-4: fission product release of HFR K6/2

5.3 Results of γ -spectrometry of the cold plates

The results of the γ -spectrometry of the cold plates are given in the following tables. Plate 65 was not used for the test and is, therefore, missing in table and diagram.

Plate	^{137}Cs activity (Bq)	Uncertainty (Bq)	measurement date
50	3.608E+05	6.816E+03	17.08.06
51	1.765E+05	3.916E+03	17.08.06
52	6.585E+04	1.584E+03	17.08.06
53	1.995E+05	4.708E+03	17.08.06
54	2.329E+05	5.291E+03	17.08.06
55	3.261E+05	6.146E+03	17.08.06
56	7.210E+05	9.977E+03	17.08.06
57	1,490E+05	3,198E+03	17.08.06
58	1.424E+05	3.244E+03	17.08.06
59	2.608E+05	5.869E+03	17.08.06
60	1.181E+05	2.397E+03	17.08.06
61	3.082E+05	5.408E+03	17.08.06
62	7.925E+05	1.193E+04	17.08.06
63	1.456E+06	2.049E+04	17.08.06
64	1.196E+06	1.723E+04	17.08.06
66	4.143E+06	1.589E+05	08.09.06
67	1.353E+07	4.192E+05	08.09.06
68	8.239E+06	2.791E+05	08.09.06
69	2.278E+07	7.388E+05	08.09.06
70	1.454E+07	4.754E+05	08.09.06
71	3.586E+07	1.105E+06	08.09.06
72	8.408E+06	2.895E+05	08.09.06

Table 5-1: Results of the measurement of condensation plates placed on a distance of 30 cm from a 3x3 NaJ detector, given with the associated combined uncertainty.

Conclusions

The fuel element HFR K6/2 (AC-065600) was tested in the KÜFA-installation in Hot Cell 103, Wing B. In the test, comprising 100 h at 1600°C and 200 h at 1800°C, a low release of the gaseous fission products was measured, whereas the Cs-release started to be measurable from the first plates and increased with increasing testing temperatures.

The observed behaviour is either due to increasing release to enhanced transport through the coating layers of many particles or is the consequence of damage

induced in the coatings of a few particles. No pressure vessel failure was observed.

The present is the first revision of this report. A further revision will be issued when:

- a. More accurate inventory calculations will be provided by Fz-Jülich in the framework of the RAPHAEL-IP
- b. A better calibration of the cold finger will be performed using certified standards
- c. Further results for ^{90}Sr and ^{109}Ag will be available from the leaching of the plates and subsequently analysis by ICP-MS of the leachates

References

1. Duwe, Kühnlein, 1975
DUWE, R. ; KÜHNLEIN, W.
Berechnung des Spaltproduktinventars in Brennelementen mit U 235, Th 232 und U 238 als Schwermetalleinsatz
Institut für Reaktorwerkstoffe Kernforschungsanlage Jülich GmbH, 1975
2. IAEA 1997]
Fuel Performance and Fission Product Behaviour in Gas Cooled Reactors
IAEA-TECDOC-978, Wien 1997
3. Nabielek, Pitzer, Schenk 1986
NABIELEK, H. ; PITZER, D. ; SCHENK, W.
Spaltproduktfreisetzungsvorlauf von Kugelbrennelementen bei Störfall-temperaturen
Als Manuskript gedruckt
Institut für Reaktorwerkstoffe Kernforschungsanlage Jülich GmbH, 1986
4. Kugeler, Schulten 1989]
KUGELER, K. ; SCHULTEN, R.
Hochtemperaturreaktortechnik HTR
Berlin; Heidelberg; New York; London; Paris; Tokyo; Hong Kong: Springer, 1989
5. D. Freis
PhD-Thesis, in preparation