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Advanced Reactor for Cogeneration of Heat & Electricity-R&D

Safety Testing report on HFR EU1 irradiated fuel

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Summary

This document is the final report dubbed deliverable D32.21, it contains the main results of interest achieved concerning the KüFa safety tests performed on the HFR-EU1/3 and HFR-EU1/4 pebbles. The Cold Finger Apparatus (KühlFinger-Apparatur ? KüFA) in operation at JRC-ITU is designed to experimentally scrutinize the effects of Depressurization and LOss of Forced Cooling (D-LOFC) accident scenarios on irradiated High Temperature Reactor (HTR) fuel pebbles. Up to 1600°C, the reference maximum temperature for these accidents, high-quality German HTR fuel pebbles have already demonstrated a small fission product release. This paper discusses and compares the releases obtained from KüFA-testing the pebbles HFR-EU1/3 and HFR-EU1/4, which were both irradiated in the High Flux Reactor (HFR) in Petten. We present the time-dependent fractional release of the volatile fission product Cs-137, Cs-134, Eu-154, Ag-110m as well as the fission gas Kr-85 for both pebbles. A description of the experimental setup and its accuracy is given. The data for the recently tested pebbles is discussed in the context of previous results

Approval

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Safety Testing report on HFR EU1 irradiated fuel (Deliverable 32.21)

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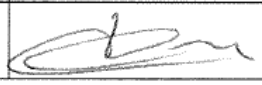
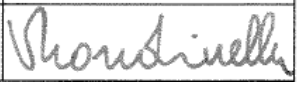
Signature				

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1 Introduction

The Cold Finger Apparatus (KühlFinger- Apparatur – KüFA) in operation at JRC-ITU is designed to experimentally scrutinize the effects of Depressurization and LOss of Forced Cooling (D-LOFC) accident scenarios on irradiated High Temperature Reactor (HTR) fuel pebbles. The Pebble Bed Modular Reactor type of fuel is composed of a graphite matrix containing TRi- ISOtropic (TRISO) coated particles. Up to 1600°C, the reference maximum temperature for these accidents, high-quality German HTR fuel pebbles have already demonstrated a low failure fraction of TRISO coated particles and a small fission product release.

The two most recently KüFA-tested pebbles HFR-EU1/3 and HFR-EU1/4 were irradiated in the High Flux Reactor (HFR) in Petten, Netherlands. Both fuel pebbles confirmed the low fission product release rate from coated particles under the temperature, burn-up and neutron fluence conditions adopted in the reactor during irradiation

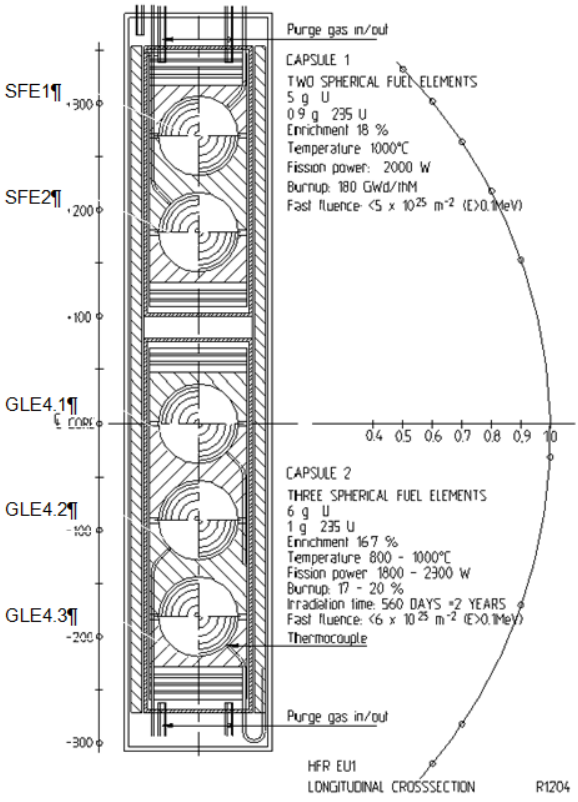
HFR-EU1 pebbles were manufactured by HOBEG (AVR GLE-4 type) and contain 9560 coated particles. It featured an initial enrichment of 16.76 wt% U-235 and was removed from irradiation in February 2010.The HFR –EU1 experiment was designed for testing spherical High Temperature Reactor (HTR) fuel pebbles for their potential for high temperature performance and very high burn up. These pebbles contain fissile material in the form of TRISO coated particles which are composed of UO_2 fuel kernels with successive isotropic layers of porous carbon, dense carbon, dense silicon carbide and dense carbon. Some 10,000 of these particles are embedded in a graphite matrix to form a sphere of 6 cm diameter.

This report presents the results from the KüFA tests performed on both pebbles and discusses the context of previous experiments.

2 Irradiation conditions

The HFR-EU1 irradiation experiment was performed during two European Framework Programs. The irradiation itself has been performed within RAPHAEL (ReActor for Process Heat And Electricity), a 4-year FP6 Integrated Project on Very High Temperature Reactors. The current nondestructive examination, performed by JRC-ITU, belongs to the FP7 project ARCHER (Advanced High-Temperature Reactor for Co-generation of Heat and Electricity R&D).

The HFR-EU1 experiment included 5 pebbles of 60 mm diameter respectively named HFREU1-1 to HFREU1- 5. HFR-EU1/1 and /2 are originating from INET, while pebbles HFR-EU1/3 to HFR-EU1/5 are German pebbles type GLE-4 (reload 21/2). The pebbles were embedded in a graphite matrix in the form of half-shells. The characteristics of the pebbles embedded in a graphite matrix in the form of half-shells are given in Figure 1.



Coated Particle	INET	AVR
Particle batch	V000802	HT-384-393
Kernel composition	UO_2	UO_2
Kernel diameter [µm]	490.3	502
Enrichment [U-235 wt.%]	17.08	16.76
Thickness of coatings [µm]:		
→ buffer	97.7	92
→ inner PyC	42.0	40
→ SiC	37.8	35
→ outer PyC	40.8	40
Particle diameter [µm]	926.9	916
Pebble		
Heavy metal loading [g/pebble]	5.02	6.0
U-235 content [g/pebble]	0.858	1.00 ± 1 %
Number of coated particles per pebble	8500	9560
Volume packing fraction [%]	5.0	6.2
Defective SiC layers [U/U_{tot}]	2.3×10^{-7}	7.8×10^{-6}
Matrix graphite grade	A3-3	A3-3
Matrix density [kg/m³]	1760	1750
Temperature at final heat treatment [°C]	N/A	1900

Figure 1 Conceptual sketch of HFR-EU1 and characteristic data of the pebbles

The irradiation initially planned to be performed for 22-24 cycles in core position H2 (Figure 2) started on 30 September 2006 in H2, i.e. after the conversion of the HFR core to low-enriched uranium (LEU). As it turned out that the heat developed by the fuel in H2 was about 20-30% lower than calculated, the experiment was moved to position H4 and lately to position F2 until the end of irradiation [1].

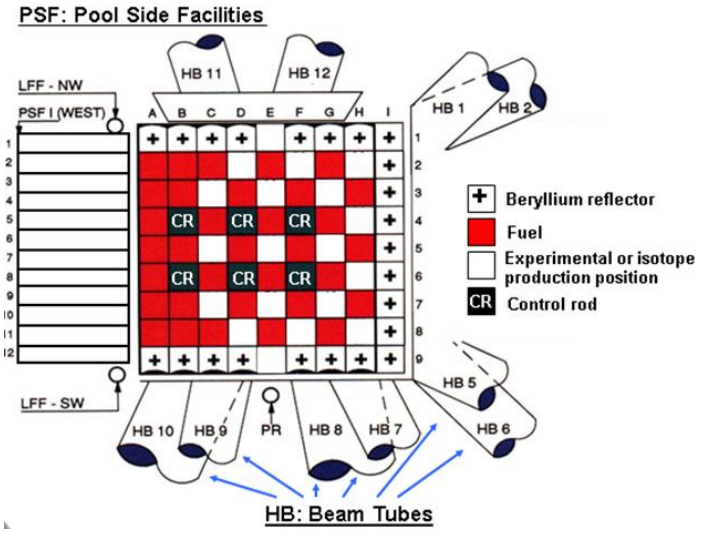


Figure 2: HFR standard core configuration

The irradiation test was performed in two campaigns, from 29 September 2006 to 24 February 2008 (12 reactor cycles of 28 days each) and continuing from 19 October 2009 to 19 February 2010 (4 reactor cycles), for a total of 445 efpd. The fluences provided cycle by cycle are given in Table 1.

Cycle	Core Position	efpd	Φ_1 [1)]	Φ_2 [1)]	Φ_3 [1)]	Φ_4 [1)]	thermal fluence	fast fluence (E> 0.1 MeV)
		[days]	Cycle averaged [10^{18} $m^{-2}s^{-1}$]				[10^{24} m^{-2}]	
2006-08	H2	24.2	0.409	0.612	0.602	0.697	1.46	1.63
2006-09	H2	26.9	0.412	0.615	0.603	0.691	1.60	1.82
2006-10	H2	28.6	0.443	0.645	0.632	0.730	1.80	3.05
2007-01	H4	27.9	0.695	0.927	0.873	0.866	2.09	3.05
2007-02	H4	28.9	0.720	0.974	0.915	0.878	2.19	3.30
2007-03	H4	27.9	0.710	0.963	0.907	0.878	2.11	3.14
2007-07	F2	27.5	0.778	1.078	1.049	1.106	2.62	3.39
2007-08	F2	28.7	0.790	1.097	1.069	1.139	2.83	3.61
2007-09	F2	28.0	0.786	1.089	1.060	1.118	2.70	3.49
2007-10	F2	27.8	0.762	1.054	1.024	1.072	2.58	3.36
2008-01	F2	27.8	0.746	1.035	1.010	1.066	2.56	3.29
2008-02	F2	28.8	0.795	1.103	1.077	1.149	2.85	3.63
2009-08	F2	24.8	0.770	1.088	1.060	1.101	2.36	3.06
2009-09	F2	31.4	0.778	1.099	1.070	1.111	3.011	3.91
2009-10	F2	24.3	0.766	1.082	1.054	1.087	2.284	2.98
2010-01	F2	31.7	0.757	1.070	1.043	1.086	2.971	3.84
total		444.88					38.02	49.51

Table 1: Measured fluences for HFR-EU1 cycle per cycle

During the irradiation campaign the fuel surface temperatures were kept constant at 900°C for the INET pebbles and 950°C for the AVR pebbles. The calculated maximum burn-ups were approximately 11% FIMA (INET) and 14% FIMA (AVR), which is significantly higher than the license limit for the HTR Modul (modular HTGR concept introduced the early 1980s with about 8% FIMA). The burn-up deduced from the gamma spectroscopy measurements performed on the two AVR pebbles HFR-EU1/3 and HFR-EU1/4, which were transported to JRC-ITU in February 2013, was in agreement with the calculated values as stated in Table 2.

Spherical Fuel Element	Origin	Burnup (% FIMA) (estimated)	Calculated B.U. % FIMA
HFR-EU1/5	Germany (GLE4/AVR)	13.47	
HFR-EU1/4		14.33	14,58
HFR-EU1/3		13.90	14,64 - 14,72

Table 2: Estimated Burn-up for each pebble

3 JRC-ITU KüFA setup and associated facilities

The KüFA is used to observe the effects of D-LOFC accident scenarios on irradiated HTR fuel pebbles and was originally designed by Schenk et al. at the Forschungszentrum Jülich. The KüFA facility was transferred to the ITU Hot Cells in the years 2000 and provided since then safety results concerning the latest HTR German fuel production (i.e. mainly HFR-K5, HFR-K6 qualification tests).
Both HFR-EU1/3 and HFR-EU1/4 pebbles were transported in ITU in February 2013 for safety tests and associated PIE examinations. The first HFR-EU1/3 pebble has been tested in May 2014 after some modifications concerning the original testing procedure. The test was performed using mainly a two plateaus profile (first plateau at 1600 ^{0}C and second at 1800 ^{0}C). The second pebble HFR-EU1/4 was tested in late 2014 with a ramp that is more comparable to the best estimate ramp encountered during a realistic accident.

3.1 KüFA setup

A sketch of the setup is depicted in figure 3. A pebble is heated by a furnace in He atmosphere for several hundred hours, simulating accident temperatures up to 1800°C. In the He gas line, an activated charcoal trap cooled by liquid N_2 collects the fission gas Kr-85 released from the fuel. It is gamma monitored with a NaI detector for on-line measurement of an accumulating release. Released volatile fission products condense on a water cooled stainless steel plate screwed onto a supporting structure, the "Cold Finger". It is positioned above the fuel element in the furnace and the temperature of the condensation plate is kept below 100°C at all times. By exchanging plates frequently during an experiment and analyzing the plate deposits by

means of High Purity Germanium (HPGe) gamma spectroscopy, a reconstruction of the release of gamma-active radio nuclides as a function of time and temperature is obtained [2].

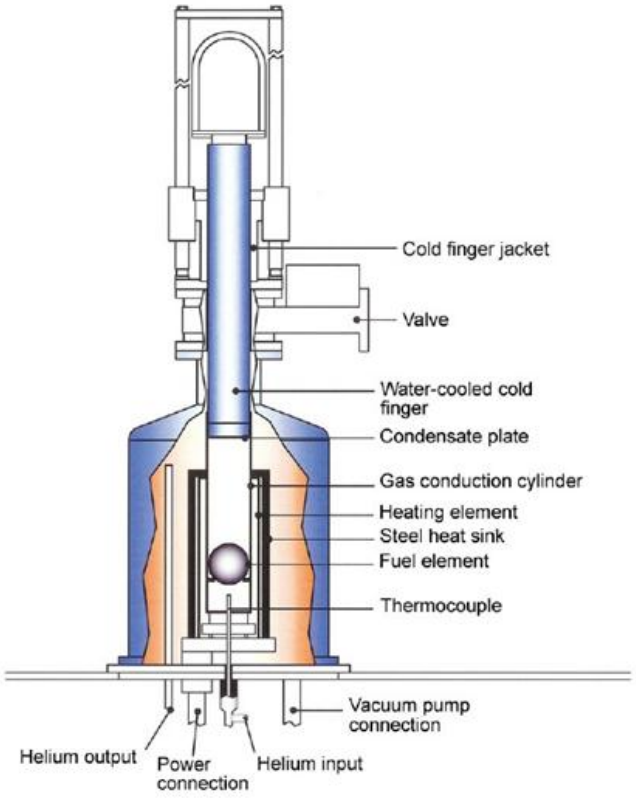


Fig. 3: KüFA facility in ITU

3.2 Cold plate measuring facilities

Considering the wide range of activity characterizing the removed cold plates (from a few microSv/h to up to 150 mSv/h), two different facilities were made available to allow the best dead time / detection limit compromise.

- The low activity plates are transported to a low-background gamma spectroscopy laboratory. The HPGe detector setup in that laboratory is able to assess different activity levels when selecting an appropriate distance between sample and detector in a fixed geometry by using a standardized plastic structure. It is electrically cooled and housed in a shielding system to ensure a low background. The calibration for this setup was performed by using a certified mixed standard source containing Ba-133, Cs-137, Mn-54 and Zn-65 with a geometry representative to that of the KüFA condensation plates, but assuming a homogeneous distribution of fission products.

- The high activity plates are placed in a tailor made La Calhene container which provides a protection against both contamination and irradiation for the persons in charge of the measurements and allow some flexibility when dealing with the distance between the source and the detector. The HPGe detector used for this measurement was calibrated by using the ISOCS methodology and some validation tests were performed using some perturbation sources to ascertain that there is no loss of counting originating from a too high dead time during the measurements. The La Calhene system used is depicted in Fig. 4. This new setup allows some lower detection limit (with a reduced background when compared to the glove box measurements) and is free from any collimation device that would have introduced a bias in the measurement considering that a non-even deposit profile is present on the cold plate surface.

Fig. 4: La Calhene docking system for high activity cold plate samples

3.3 HTR pebble gamma spectrometry station

In order to perform a Fission Produts (FP) balance on the fuel pebble, the pebbles were gamma-counted on a dedicated gamma spectrometry station attached to the Cell 101. A dedicated pinhole system composed of several Denal pieces housed in a tailored made tube is used. The collimator penetrates through the concrete wall of about 1 m thickness and targets the pebble placed in a tin box that allows a reproducible positioning of the pebble. The tin box is mounted on a rotating table to take into account the 3D particles inhomogeneity within the pebble. A laser system assesses the positioning of the pinhole collimator with respect to the tin box (used as pebble support). An overview of the system is provided in Figure 5.

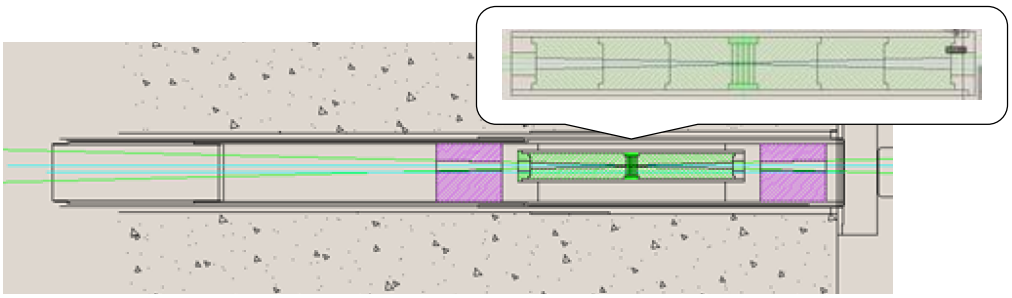


Figure 5: Pinhole collimator system

3.4 Chemical Cells in ITU

Considering that only gamma emitters could be investigated with the developed instrumentation, it was decided to analyze from time to time either the cold plates or the tantalum tube to investigate beta emitters of interest (Sr-90 for example). The preparation of the samples requires a leaching process (up to 60 ^{0}C in strong acid) that was performed in the Chemical cells in ITU. The liquid samples obtained were analyzed by ICP-MS to both cross check gamma measurements achieved and provide missing information on non-gamma emitters.

4 Preliminary considerations on the KüFA setup

This chapter will introduce a few parameters that should be discussed in order to properly assess the final release rates when dealing with the KüFA apparatus, mainly: cross contamination aspects, cold plate trapping efficiency and temperature assessment.

4.1 Cross contamination aspects

The cold plates once exposed to the KüFA atmosphere are enclosed in aluminium cans and have to be routed through the KüFA cell, the conveyor and finally the glove box for a final bag out. Even if a lot of effort has been deployed to limit the contamination (i.e. intensive use of tissue for the cans, removal of the cans prior to the bag out, decontamination of the different elements, cleaning of the gloves and operating tools, etc.), some cross contamination is still to be expected and may largely interfere with the measurements in case of low FP release during the KüFA test. In order to assess the cross contamination issue, the ratio between both Cs-137 and Cs-134 was judged satisfactory (Cs-134 having already heavily decayed in the different cells when compared to Cs-137). If some cross contamination is to happen, this ratio should present some chaotic behavior especially for low sample activities. As presented in the Fig 6, most of the low activity plates for the HFR-EU1/3 test up to number 17 present some cross contamination. The Cs-137 release presented during the 1600 ^{0}C phase is to be considered as an envelope value, considering that the trapped Cs-137 on the cold plate is to be swamped in the cross contamination originating from the contamination of the external plastic bags.

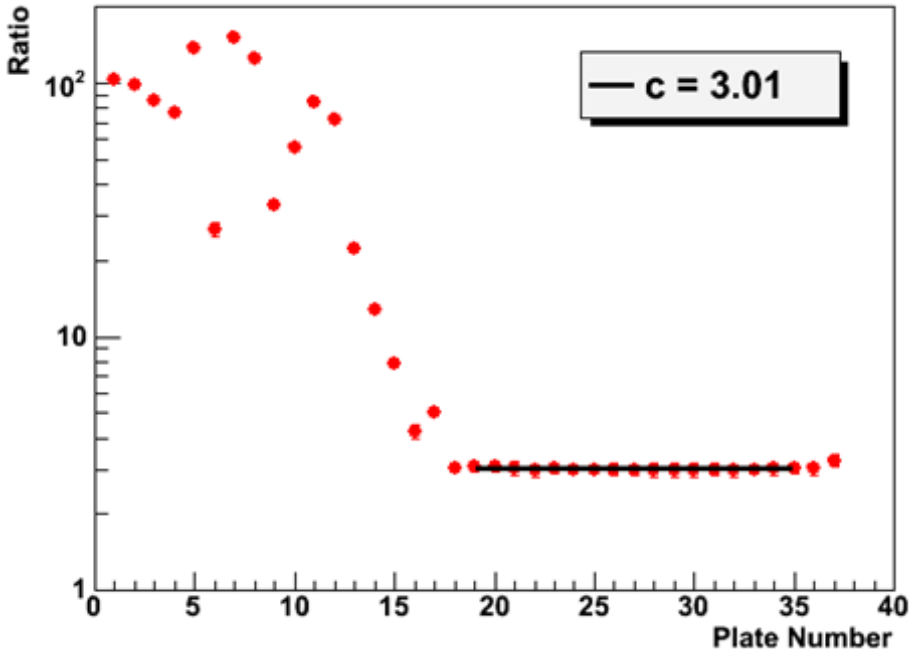
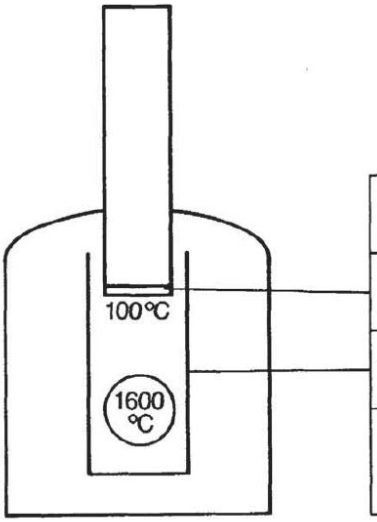


Fig. 6: 137Cs/134Cs ratio for the HFR-EU1/3 condensation plates

4.2 Cold plate efficiency aspects

The collection efficiency of the KüFA system is mainly derived from the original Schenk experiments where the efficiency were mainly said to be according to the following Fig 7.



	Released portion	
	Cs, J, Ag	Sr
plate	0,7	0,2
Ta-tube	0,02	0,8
furnace	0,3	–

Fig. 7: Fission products distribution in the KüFA system

The efficiency with associated uncertainty for the four elements is:

ε_{Cs} = 0,70 +/- 0,13

ε_{Iodine} = 0,70 +/- 0,13

ε_{Ag} = 0,70 +/- 0,30

ε_{Sr} = 0,20 +/- 0,13

As first approach, the release fraction of each fission product was considered using the collecting efficiency of the Cs. All the following calculations were performed using a 72% value.

Nevertheless, a cross check calculation was performed at the end of each test in order to assess the global FP release by means of the already described gamma spectrometry station attached to the cell 101. During this validation process, it appears that the collecting efficiency on the cold plate was largely over estimated, most probably during the 1800 ^{0}C phase, leading to discrepancy in the FP balance up to a factor 3.

If the collecting efficiency was found to be in good agreement with the Ag-110m release this was most probably due to the early release of this FP during the safety test (i.e. 1600 ^{0}C phase). We consider that the final results should be presented with an estimation of the release that will be in accordance with the FP balance data. Considering at this stage no specific efficiency tests could solve the issue, it was decided to apply a coefficient factor from the beginning of the 1800 ^{0}C phase to all release data. These extrapolated data will be presented as dotted line in the presented results.

A campaign of efficiency tests with the following radiological source Ag-110m, Cs-137, Sr-85, Eu-152 will be performed to investigate this issue especially during the 1800 ^{0}C phase. Among the different parameters that may have influenced the efficiency, the following will be investigated:

- Increase in temperature of the cold plate during the 1800 ^{o}C phase (in the range 85 – 115 deg C)
- Amount of FP deposit already present on the cold plate that may prevent further collection
- Significant change in the flow prior to the cold finger bottleneck

4.3 Assessment of the temperature

The temperature in the KüFA furnace is measured by two Type C thermocouples. One of these also functions as temperature regulator and, during a heating test, provides a feedback to the control system to hold, increase or decrease the furnace heating power.

After replacing the inner Ta parts of the KüFA setup (the heating element, the heat shield and the inner tube holding the pebble) and the thermocouples measuring and regulating the furnace temperature, a possible temperature drift was assessed using a conventional melt wire technique. A graphite holder containing small graphite cups with Ta fillings for little wire pieces of dedicated pure metals with well-known melting points was designed and can be seen in figure 8. The Ta fillings were chosen to avoid the formation of eutectics between the pure metal wires and the graphite cups. The designed system was placed inside the KüFA instead of a pebble at the same location in the furnace.

Examples for metals with well-known melting points are Au, Cu, Fe, Ni and Pt. Fe and Ni are not suitable for a melt wire test with our setup since their respective phase diagrams with Ta show multiple possibilities for the formation of eutectics across a wide temperature range. To assess the temperature accuracy, Au, Cu and Pt were heated in the KüFA one at a time. For each run, the device heated up to a selected nominal temperature, which was kept constant for a duration of 4 h. After cooling down again, the melt wire was removed and visually examined. Au (melting point at 1064°C) did not melt at nominal temperatures of 1074°C and even 1094°C. Cu (melting point 1085°C) also did not melt at a nominal temperature of 1094°C, but did at 1133°C. Pt (melting point at 1768°C) formed an eutectic with Ta at a nominal temperature of 1770°C but did not actually melt, meaning that according to the Pt-Ta phase diagram the true temperature must be between 1725°C and 1768°C.

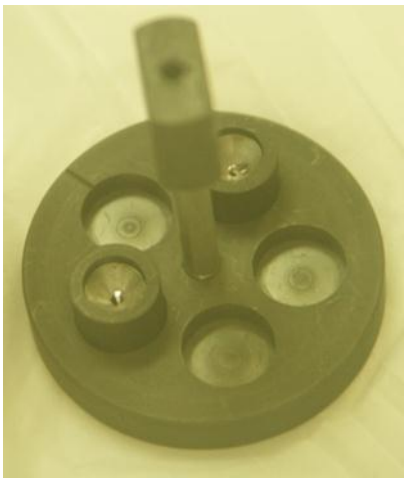


Fig. 8: Melt wire graphite holder containing 2 of 5 possible graphite cups with Ta fillings and pure metal wires

All results combined show that even for a brand new set of thermocouples and completely renewed inner KüFA furnace parts a drift of 30 - 45°C exists in such way that the true temperature of the experiment is lower than what the thermocouples read. While all plots in this paper show the nominal KüFA furnace temperature, the drift certainly has to be kept in mind when discussing the obtained fission product release data.

5 HFR-EU1/3 pebble safety test

5.1 Pebble inventory

HFR-EU1/3 was prepared for KüFA testing in May 2014. A previously carried out gamma measurement in the Cell 101 in March 2013 yielded the following activities:

$A_{Ag\text{-}110\,m}$ = (60 ± 20) MBq
$A_{Cs\text{-}134}$ = (44 .25 ± 2 .34) GBq
$A_{Cs\text{-}137}$ = (89 .01 ± 6 .63) GBq
$A_{Eu\text{-}154}$ = (3 .02 ± 0 .16) GBq

The 85Kr peak at 514 keV could again not be observed and a value for the end of irradiation was calculated from data given in [3] and propagated to the time of the KüFA test, yielding

$A_{Kr\text{-}85}$ = (7 .11 ± 0 .02) GBq.

5.2 Temperature transients achieved

The HFR-EU1/3 KüFA test started on 7 May and ended on 13 June 2014. The initial temperature schedule was:

- heated to 1050°C (similar to irradiation temperature) and held for 30 min.
- heated to 1250°C with the same rate and held at that temperature for 90 min.

After the aforementioned mini temperature plateau, the test entered the accident phase of the experiment for the pebble:

- heating to 1600°C with a 47°C/h ramp and holding at this temperature for 300 h.
- Subsequently, heating to 1800°C continuously (at the same rate) and holding for 200 h.
- The final cool down to 30°C took place at a rate of 75°C/h.

The heating sequence needed to be modified due to unforeseen events, making the total duration of this heating test approximately 744 h instead of the originally planned 564 h.

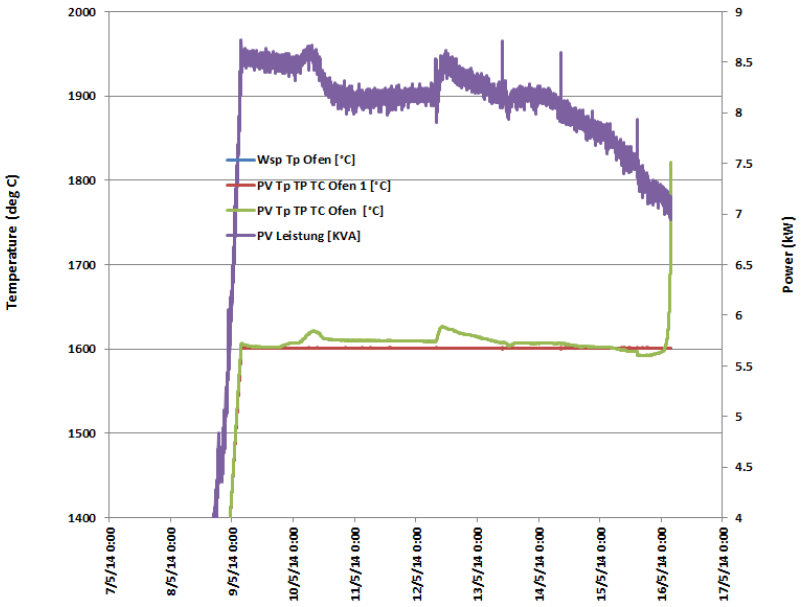


Figure 9: Failure of the TCs during the temperature transients for the HFR-EU1/3 KüFA test

About 120h after the start of the heating sequence a notable degradation of the two furnace thermocouples began. The regulating thermocouple read an increasingly too high temperature, causing the control system to continuously lower the furnace heating power by more than 1 kW (figure 9) and, thus, significantly reduced the actual true temperature. The second thermocouple began to malfunction in the same way but more severely. About 210 h into the experiment it read temperatures above 1820°C, causing the control system to initiate an emergency shutdown of the KüFA setup. Both thermocouples were renewed and the HFR-EU1/3 KüFA test was resumed on 21 May 2014 by directly heating from 30 to 1600°C at a rate of 47°C/h and conservatively prolonging the 1600°C plateau. Throughout the rest of the experiment the setup was stable with respect to the furnace temperature and figure 18 shows that during the 1600°C and 1800°C accident plateaus the respective furnace heating powers were 8.7 and 12.7 kW in accordance some previous tests. The reference test could be summarized as depicted in Fig 10.

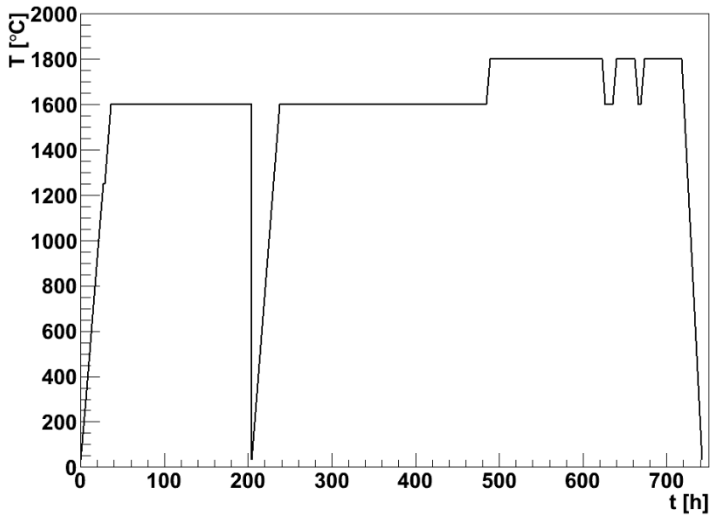


Figure 10: Temperature transients for the HFR-EU1/3 KüFA test

During the 1800°C phase the KüFA was operated in a remarkably hot weekend; this corresponded to a slightly less efficient water cooling supply. This was enough to accelerate the heat up of the cold finger to critical temperatures, which was already observed in the 1800°C segment of the HFR-K5/3 KüFA test. To avoid another emergency shutdown of the system, which happens automatically when the cold finger temperature exceeds 115°C, condensation plates were exchanged more frequently. In two cases this was not possible because of laboratory safety reasons and instead the furnace temperature was reduced to 1600°C again for a few hours (two spikes to be seen at the end of the 1800 ^{o}C plateau in Fig 7.).The power consumption of the KüFA is provided in Fig. 11.

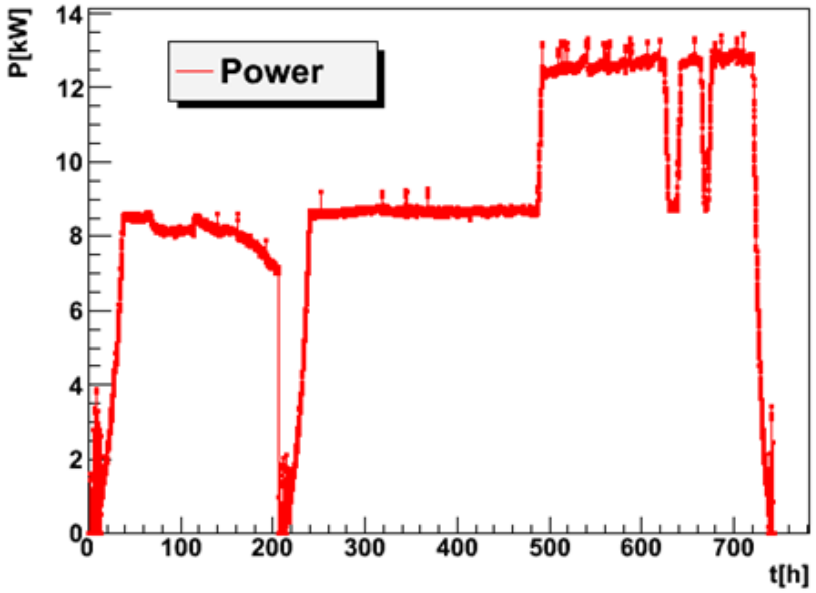


Figure 11: Power consumption during the HFR-EU1/3 KüFA test

5.3 Cs results

During the initial 3 mini plateaus at 300°C, 1050°C and 1250°C no significant releases were observed for Cs.
The high contamination level of Cs-137 (i.e. Cs-137 release swamped in the Cs-137 contamination from the hot cell) built up to a release of 10^{-5} as shown in Fig 12. At the same time the Cs-134 release stayed below

10^{-6}, providing more correct information about the pebble's behaviour due to the significantly lower cross contamination level.
During the 1800°C phase of the HFR-EU1/3 KüFA test, the Cs-134 release rapidly increased by several orders of magnitude until it reached (0.25 ± 0.055) at the end of the experiment as depicted in Fig 13. In this phase the pebble release dominated the measured level of Cs-137, leading to consistent release curves for Cs-137 and Cs-134. The correct total release values for Cs-134 and Cs-137 obtained by direct gamma counting are also consistent and amount to 0.693 ± 0.024 and 0.688 ± 0.034 respectively.

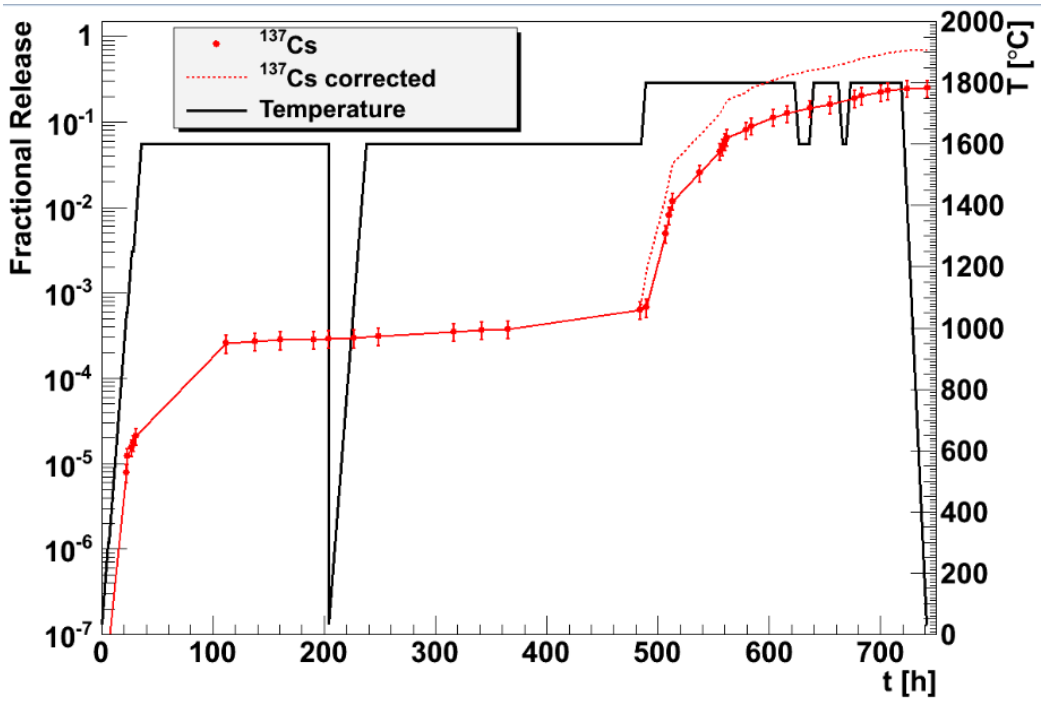


Figure 12: Cumulative Cs-137 release

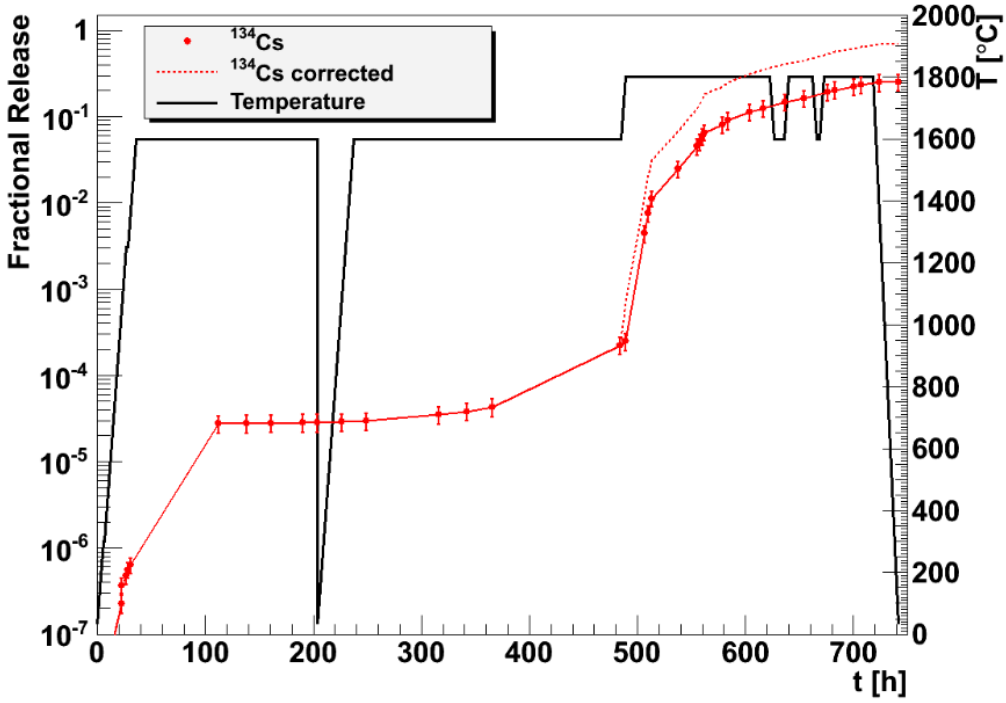


Figure 13: Cumulative Cs-134 release

5.4 Ag results

The Ag-110m release depicted in Fig. 14 was significant from the beginning of the heating sequence. Even at low temperature (i.e. below 1000°C), some Ag-110m release was detected that is mostly originating from the silver trapped in the graphite during the irradiation phase. It would have been of interest to proceed to further investigation concerning the graphite cups of the FHFR-EU1 irradiation. Unfortunately, these gamma measurements techniques performed with a high hot cells background did not allow silver to be detected. Below 500°C, the cumulative release was still below 10^{-5}, but after the 1250°C plateau it already grew to 5×10^{-3}. At 1450°C on the heating ramp to 1600°C the Ag-110m release amounted to 2×10^{-2} even before the reference accident temperature was reached.

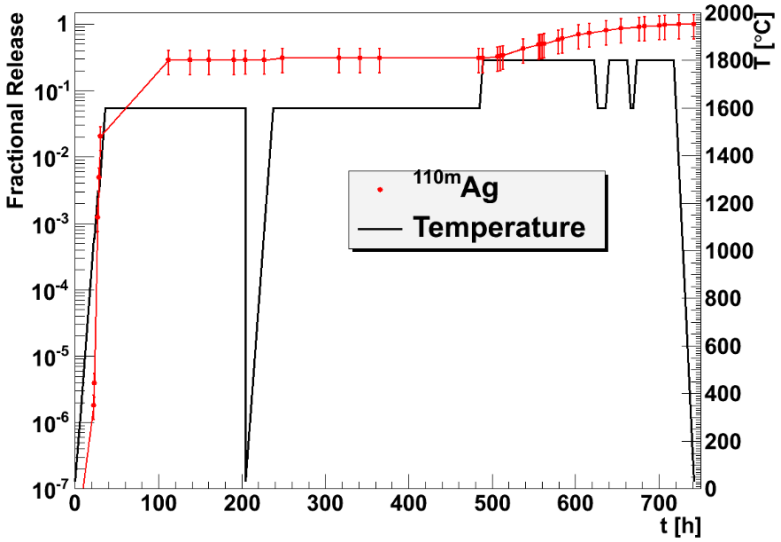


Figure 14: Cumulative Ag-110m release

Throughout the 1800°C segment of the heating test Ag-110m was again released at high rates, resulting in a total release of almost the complete pebble inventory 0.99 ± 0.39.

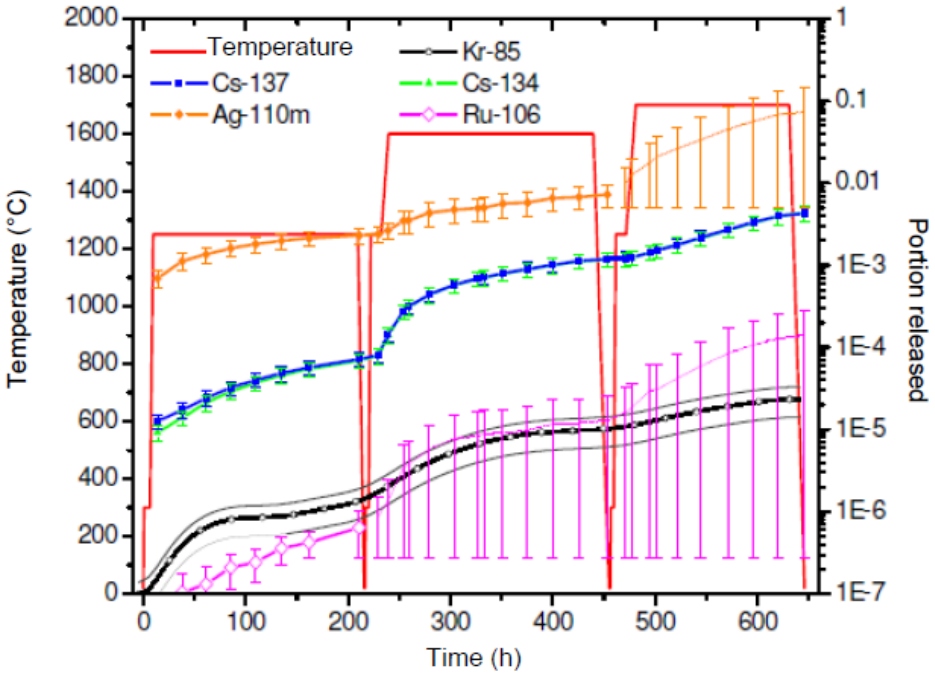


Figure 15: Cumulative Ag-110m release from HFR-EU1bis 1 pebble

When compared with the HFR-EU1bis pebbles (Fig.15 [2]), the amount of Ag-110m at the end of the 1600^{0}C phase was relatively high: about 23% release; for FR-EU1bis 1 the corresponding release was below 1% and did not increase significantly thereafter.

5.5 Eu results

After about 430 h at a nominal temperature of 1600°C (in fact less due to the thermocouple failure) and the subsequent heating ramp to 1800°C, slightly more than 10-4 of the Eu-154 inventory was released (Fig. 16). During the 1800 ^{0}C plateau, the Eu-154 release gradually increased to (6 ± 2) × 10^{-4}.

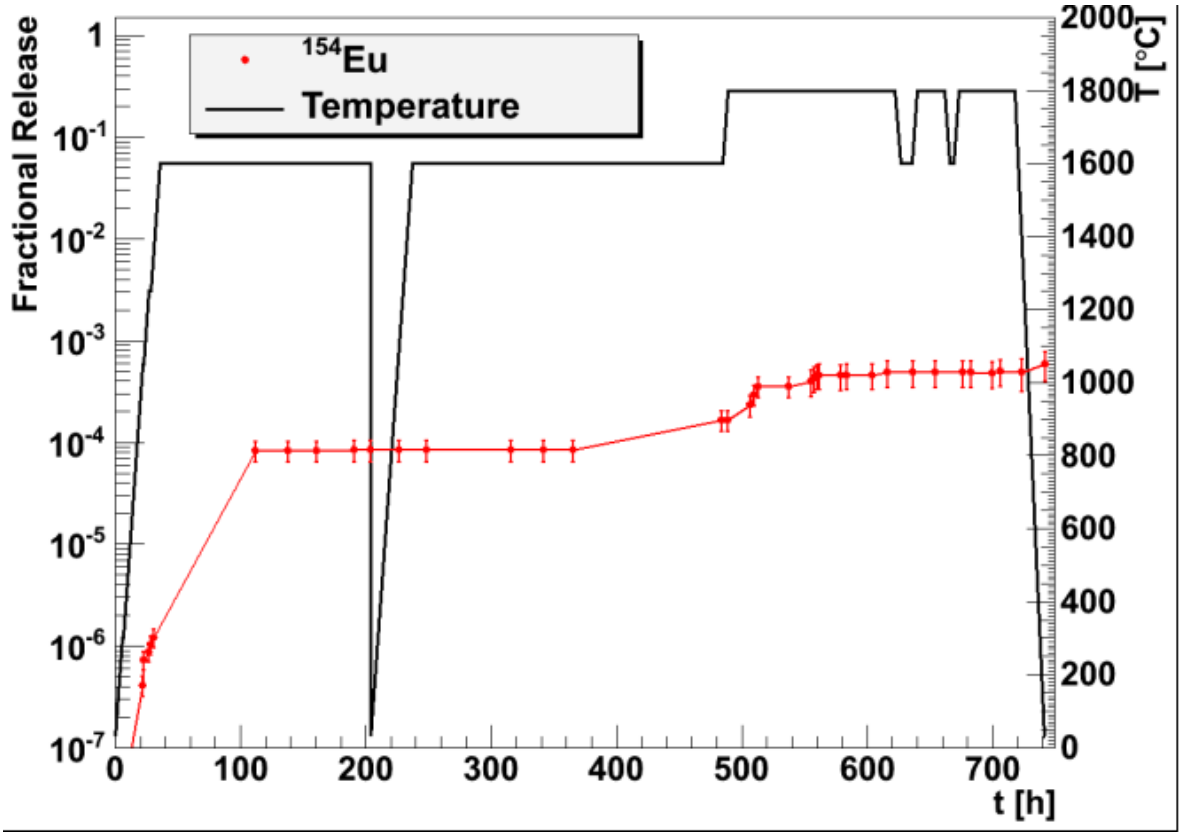


Figure 16: Cumulative Eu-154 release

The HFR-EU1/3 test is one of the few test performed in ITU that provided a measurement above the detection limit threshold. It can be partly explained by the new measurement device that has been developed for high activity plates. The Eu release is quite measured routinely for US fuel qualification.

5.6 Kr results

Before the 1800 ^{0}C plateau, Kr-85 release stayed below the detection limit in the range of 10^{-6} as depicted in Fig 17.

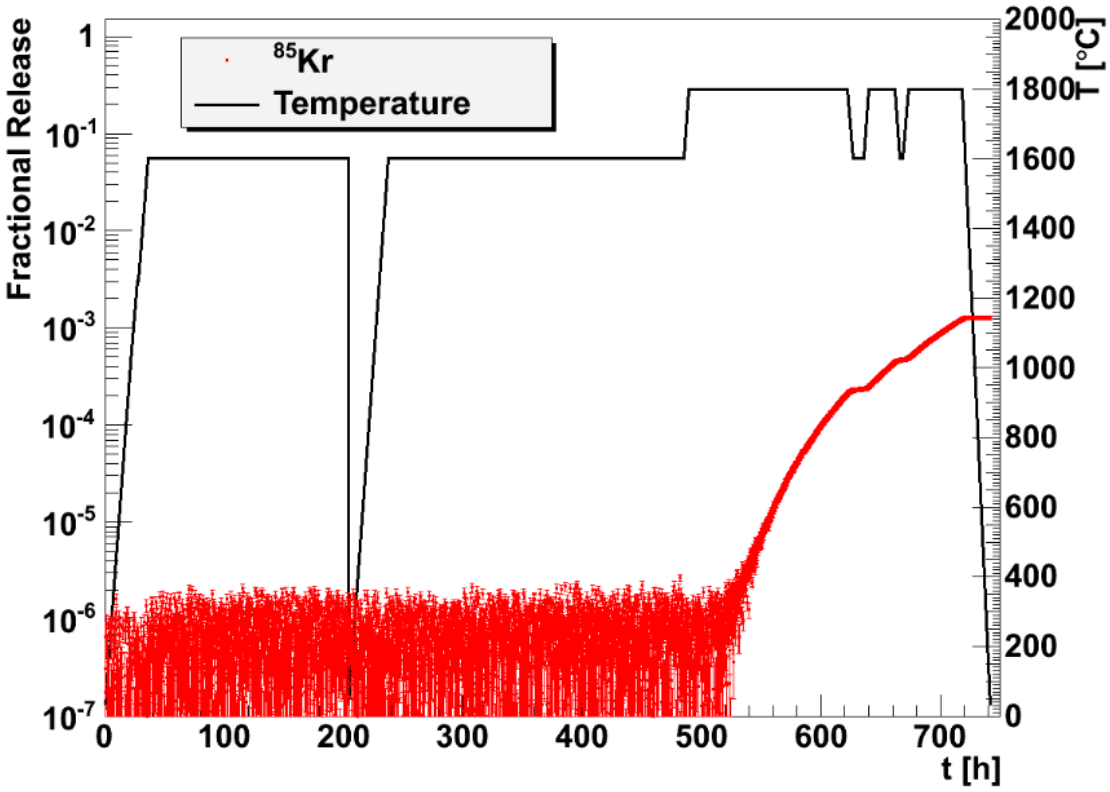


Figure 17: Cumulative Kr-85 release

During the 1800 °C phase, Kr-85 became detectable, being delayed 40 - 50 h with respect to the Cs and yielding a final value of (1.263 ± 0.006) × 10-3. The Kr-85 release stops at once when cooling the pebble down to 1600°C again. For HFR-EU1/3, the final Kr-85 release corresponds to the total release of the fission products from 12.1 broken coated particles (i.e. SiC layer and PyC layers).

6 HFR-EU1/4 pebble safety test

6.1 Pebble inventory

A gamma measurement carried out in the Cell 101 in February 2014 yielded the activities (calculated back to the End Of Irradiation time) :

$$A_{Ag\text{-}110\,m} = (1.12 \pm 0.14)\ GBq$$
$$A_{Cs\text{-}134} = (126 \pm 4.4)\ GBq$$
$$A_{Cs\text{-}137} = (94.4 \pm 2.73)\ GBq$$
$$A_{Eu\text{-}154} = (3.66 \pm 0.28)\ GBq$$

6.2 Temperature transients achieved

The HFR-EU1/4 KüFA test was initiated in January 2015. The KüFA test was to follow the HTR-Modul temperature transient that is the reference for the LOFC accident at FZJ. The expected temperature transient is provided in Fig. 18 and is compared to the transient performed on the HFR-EU1/3 pebble.

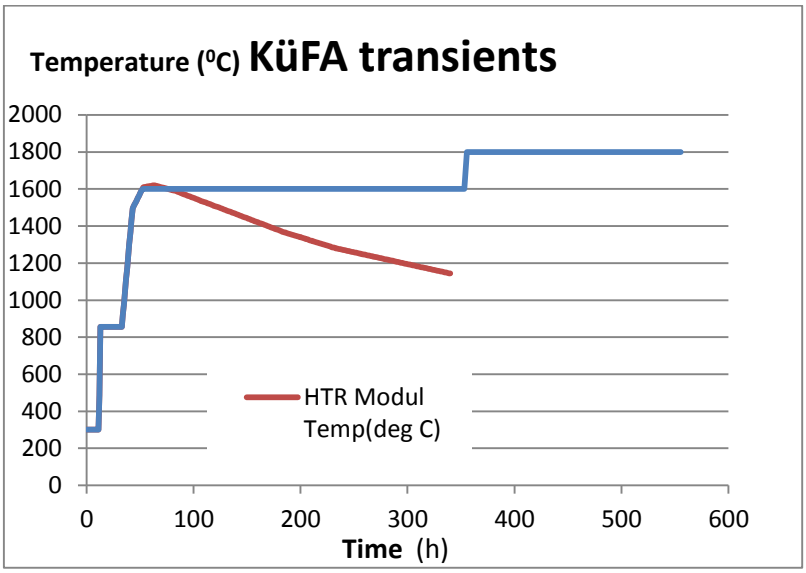


Figure 18: HTR-Modul temperature transient

Prior to the KüFA test, some preliminary tests were performed to assess the residual contamination level of the KüFA system after the decontamination phase that was initiated following the HFR-EU1/3 high release rate test. Hence, at the end of the HFR-EU1/3 test a large part of the volatile fission products that did not interact with the KüFA cold plate were collected in the cold finger penetration tube at the level of the valve isolating the cold finger from the KüFA system. The measurements performed at this level by the mean of irradiation devices indicated a dose rate that exceeded 1 Sv/h and prevented the thermocouple replacement at the bottom of the KüFA system to take place. In order to tackle this issue a tailor made decontamination system was successfully implemented in the hot cell that led to a decrease of the dose rate. (decontamination factor about 20)

The mock-up used to develop the decontamination procedure is presented in Fig. 19. The spraying system developed induced inside the KüFA bell and other internal parts some fission products deposits. During the bake out phase of the new tantalum parts, some deposits were detected on the blank cold plate that led to a significant residual dose rate (in the range of 1 mSv/h). We suspect some revolatilization phenomena to have taken place during the heating plateau at 1600 ^{0}C.

Considering that some further decontamination process is to be pursued to lower the internal contribution of the contaminated parts, the HFR-EU1/4 test will not be performed within the ARCHER timetable.

Figure 19: Plexiglas mock-up for the decontamination device

7 Conclusion

HFR-EU1/3 retained all fission products except Ag-110m well up to the beginning of the 1800°C accident phase. Slightly more than 10^{-4} of the Eu-154 and slightly less than 3×10^{-4} of the Cs-134 inventory were released up to this point of the experiment while Kr-85 could not be seen at all. At 1450°C on the heating ramp to 1600°C already 2×10^{-2} of the Ag-110m inventory was released even before the reference accident temperature was reached. Throughout the 1800°C phase of the HFR-EU1/3 KüFA test, the entire Ag-110m inventory of the pebble was released and also the Cs release increased to about 70 %, significantly exceeding all previous heating tests carried out at JRC-ITU. This was expected to happen due to the high burn-up (14 % FIMA) of HFR-EU1/3. Furthermore, a total fraction of $(6 \pm 2) \times 10^{-4}$ Eu-154 was released and, after 40 - 50 h into the 1800°C accident phase, the release of Kr-85 became significant and amounted to $(1.263 \pm 0.006) \times 10^{-3}$ at the end of the campaign, corresponding to 12.1 coated particle inventories.

The releases obtained from KüFA testing HFR-EU1/3 were significantly higher than the ones found after previous heating tests with fuel pebbles from the HFR-K6 and the HFR-EU1bis irradiation campaigns. (Fig. 15) Possible reasons for this are an overestimation of the KüFA furnace temperature in the past exceeding 30 - 45°C, an overestimated cold finger efficiency η and, of course, the different burn-up history for each pebble. Overall it can be stated that, apart from the high Ag-110m release from HFR-EU1/3, the tested fuel retains fission products remarkably well at a nominal accident reference temperature of 1600°C. However, for temperatures towards 1800°C the retention capability decreases significantly.

Due to some re-volatilization issue inside the KüFA bell, the HFR-EU1/4 experiment initially planned in late December 2014 will not be performed before the end of the ARCHER programme. This test will be considered once the internal contamination level of some parts will have been removed.

References

[1] M. Laurie, A.Marmier, M. A. Fütterer; “Results of the HFR-EU1 Fuel Irradiation of INET and AVR Pebbles in the HFR Petten”; Proceedings of HTR 2010; Paper 050; 2010

[2] D. Freis, Störfallsimulationen und Nachbestrahlungsuntersuchungen an kugelförmigen Brennelementen für Hochtemperaturreaktoren, PhD Thesis, 2010.D.

[3] HFR-EU1 depletion calculations - ARCHER Project Deliverable D32.31