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Abstract

Four spherical high-temperature reactor fuel elements irradiated at the High Flux Reactor in Petten within the HFR-EU1bis irradiation campaign were transported to JRC-ITU for post-irradiation examination and accident testing. This technical note reports the accident tests performed on the first two fuel elements HFR-EU1bis/1 and HFR-EU1bis/3 with the Küfa device. The fuel elements were heated up to operational temperatures (1250°C) and then to accident temperatures (1600°C and 1700°C, respectively) for durations of several hundred hours to simulate depressurization and loss-of-forced-circulation accidents (D-LOFC). The overall low fractional release of Kr-85 and the absence of steps in the release curves suggest that no coated particles failed during the accident simulations. The fractional release of Cs-134 and Cs-137 was much higher than in previous experiments, suggesting increased diffusion through the coating layers. Finally, a mismatch of fractional cesium releases at 1250°C and above could be due to particle failure prior to the Küfa testing.

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1. Introduction

The performance of high-temperature reactors (HTRs) under accident conditions depends to a large extent on the quality of the fuel elements. Inside the graphite matrix of spherical fuel elements, fissile material is contained in the form of thousands of small spheres of fuel enclosed in several coating layers (≈ 1 mm diameter). Modern HTR fuel contains TRISO (tri-structural isotropic) coated particles (CPs) covered with a porous carbon buffer, an inner layer of pyrolytic carbon, a layer of ceramic silicon carbide and finally an outer pyrolytic carbon layer. Together, the coatings provide a barrier against the release of fission products both in normal operation and under off-normal conditions.

In order to provide robust data for reactor design and licensing, the performance of HTR fuel elements must be tested experimentally. For this purpose, irradiated fuel is subjected to conditions that simulate a depressurized loss of forced circulation (D-LOFC) accident (see, for instance, Reference [Haque *et al.* 2006]). In this reference scenario, all active heat sinks are removed and decay heat is carried away from the core only by means of natural heat transfer mechanisms (convection, conduction and radiation). Both single CPs and entire fuel elements can be studied. In the latter case, a large number of particles is effectively investigated at the same time, providing a result with high statistical relevance. Some 25 years ago, a method to expose a spherical HTR fuel element to a simulated D-LOFC accident and to simultaneously collect all released fission products was developed at Kernforschungsanlage Jülich (KFA). Today, an improved successor device is operational at the Institute for Transuranium Elements (ITU) in Karlsruhe.

The Küfa (an abbreviation of the German "Kühlfingeranlage") is an experimental apparatus in which spherical HTR fuel elements can be heated to up to 2000°C at ambient pressure in a helium atmosphere while the release of fission gases and volatile fission products is measured. The original device was taken into operation at KFA Jülich in 1984 [Schenk *et al.* 1988]. When the German HTR program was ended, the Küfa at KFA was decommissioned. In the framework of the European HTR-F project, parts of the Küfa as well as drawings and technical documents were transferred to ITU in 2001 [Toscano *et al.* 2004]. The new Küfa was taken into operation at ITU's Hot Cells unit in 2005 [Freis *et al.* 2010]. Since then, it has been used to perform simulated accident testing on eight spherical fuel elements. With the new device it is possible to simulate accident behavior up to 1800°C and to measure the fractional release of Cs-137 to a relative precision of about 10^{-7}. The ITU Küfa is currently the only device worldwide for the accident testing of irradiated HTR fuel.

In the irradiation experiment HFR-EU1bis, five HTR fuel elements of Nukem type GLE-4.2 were irradiated at the HFR in Petten, The Netherlands. The main pre-irradiation properties of the fuel elements are listed in Table 1. In this table and elsewhere in this document, standard uncertainty is given in parentheses next to the least significant digits to which it applies. The aim of the experiment was to investigate the potential for further improvements of HTR fuel technology based on TRISO coated particle fuel. In particular, the fuel element behavior under VHTR conditions at increased temperature as well as increased and accelerated burn-up and increased fast-neutron flux was studied [Fütterer *et al.* 2008]. After irradiation, four of the five fuel elements were transferred to ITU for Küfa accident testing and post-irradiation examination in the Hot Cells unit. One fuel element (HFR-EU1bis/2) remained in Petten and was examined in the hot cells of the Nuclear Research and Consultancy Group (NRG) [de Groot *et al.* 2008b]. This technical note reports the results of the first two Küfa heating tests, which were performed on fuel elements HFR-EU1bis/1 and HFR-EU1bis/3.

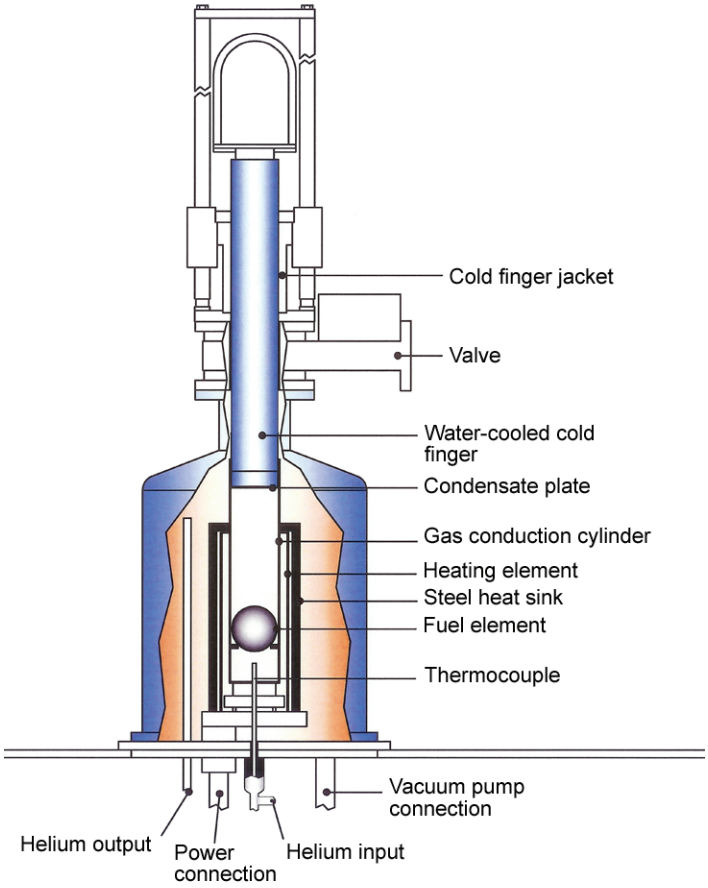

Figure 1: Schematic overview of the Küfa device at ITU

Table 1: General characteristics of the studied fuel elements (FEs)

Fuel element type:	GLE-4.2	Kernel composition:	UO_2
Graphite type:	A3-3	Enrichment (U-235):	16.67%
Number of coated part. in FE:	9560	Kernel diameter:	502.2(2.0) µm
Heavy metal mass (entire FE):	6 g	Kernel density:	10.86 g cm^{-3}
Free U fraction in the matrix:	7.8×10^{-6}		
Buffer layer thickness:	92(13) µm	Buffer layer density:	1.01 g cm^{-3}
Inner PyC layer thickness:	40.6(4.1) µm	Inner PyC layer density:	1.87 g cm^{-3}
SiC layer thickness:	35.9(2.2) µm	SiC layer density:	3.20 g cm^{-3}
Outer PyC layer thickness:	39.6(3.6) µm	Outer PyC layer density:	1.87 g cm^{-3}

2. Experimental setup

The experimental setup of the Küfa device has been described in detail in two recent articles [Toscano *et al.* 2004; Freis *et al.* 2010]. In the following paragraphs, its main features will be briefly summarized.

An overview sketch of the Küfa is shown in Figure 1. Its main components are a bell-shaped outer stainless-steel vessel, a gas conduction cylinder encompassed by a heating element, and a mobile cold finger. The heater is mounted in a water-cooled copper block and surrounded by heat shields. In order to minimize the absorption of fission products in the furnace, the Küfa was designed as a metallic resistive furnace. A high-current transformer installed behind the hot cell delivers the necessary current. All high-temperature parts of the Küfa consist of tantalum because this material is the least prone to carbidization at the contact point with the fuel element. The fuel element is placed in a three-point bearing about a third of the way up the central axis of the gas conduction cylinder. A thermocouple measures the temperature about 3 mm below the fuel element and enables the regulation of the furnace power. The furnace temperature as well as all operating processes and parameters are controlled centrally by a programmable logic controller and an associated computer program. All operating parameters are collected and recorded.

At the lower end of the cold finger, facing the fuel element, an exchangeable stainless-steel plate is placed. Thanks to efficient water cooling, the surface temperature of the plate itself is kept at less than 100°C even at maximum furnace temperatures. In this way, volatile fission products such as Cs-137 and Ag-110m plate out on the condensate plate with high efficiency. The cold finger can be pulled up through a system of locks containing a water-cooled gate valve, and the condensate plate can be exchanged semi-automatically using a telemanipulator. The used plates are then transported to a laboratory with low radiation background, where a gamma spectrum is recorded with a high-purity germanium detector to quantify the gamma-emitting isotopes [Freis *et al.* 2010]. During the experiment the device is flushed with helium. The helium enters the gas conduction cylinder below the fuel element, flows around it, and disperses in the bell. From there it is evacuated by a membrane pump and reaches an activated-carbon cold trap cooled with liquid nitrogen. Fission gases condense in the activated carbon and their activity can be measured by a sodium iodide gamma detector mounted below the cold trap [Stradal 1970]. The calibration procedures for the cold finger and the cold trap are described in detail in Reference [Freis *et al.* 2010]. The cold finger efficiency was previously found to be 70(16)%. Table 2 gives the calibration constants for the cold trap determined in the course of the experiments described in this report.

Table 2: Cold trap calibration parameters for the Küfa heating tests of pebbles HFR-EU1bis/1 and HFR-EU1bis/3

Date and time	Aliquot activity (Bq)	Gamma count rate (s^{-1})	Calibration factor ($Bq^{-1}s^{-1}$)
2008-03-31 13:46	86 583	9.6131	1.11×10^{-4}
2008-07-23 12:00	82 108	11.067	1.35×10^{-4}
2008-08-11 09:43	79 853	16.275	2.04×10^{-4}

3. Irradiation campaign HFR-EU1bis

During the HFR-EU1bis irradiation, the five fuel elements were irradiated for 10 cycles and a total of 249 equivalent full power days (efpd). The central temperature of the fuel elements was set to 1250°C during the burn-up cycles, except for a brief temperature excursion due to an operator error involving the gas mixing system [de Groot *et al.* 2008a]. The target burn-up was 15.4% FIMA; however, only 11.07% FIMA was actually achieved in the fuel element with the highest burn-up. The acceleration factor (defined as the reduction in time required to reach the target neutron flux, as compared with irradiation in an HTR reactor [Petti *et al.* 2003]) was supposed to be 7.5, whereas a value of about 5.4 was finally reached in the element with the highest burn-up.

All five fuel elements were stacked in a single irradiation rig. During the irradiation experiment, gas samples from the rig were taken about once per week and analysed by gamma spectroscopy in order to measure the fission gas release and detect potential fractured particles.

Fuel element HFR-EU1bis/1 was placed in one of the upper irradiation positions and therefore exhibited a somewhat lower burn-up of 9.34% FIMA and a lower fast fluence of 2.41×10^{21} cm^{-2} as compared with the fuel elements irradiated in the central positions. Fuel element HFR-EU1bis/3 was irradiated in a central irradiation position. It reached a burn-up of 11.07% FIMA and received a fast neutron fluence of 2.86×10^{21} cm^{-2}. Together with pebble HFR-EU1bis/4, this fuel element had the highest burn-up of all HFR-EU1bis fuel elements. Tables 3 and 4 summarize the parameters of the irradiation campaign and the irradiation history of the two fuel elements [Fütterer *et al.* 2006; de Groot *et al.* 2008b].

Table 3: Parameters of the HFR-EU1bis irradiation campaign

Beginning of irradiation:	2004-09-09	End of irradiation:	2005-10-18	
Irradiation duration:	249 efpd	Irradiation temperature:	≈ 1250°C	
Fractional fission gas release by end of irradiation (*R*/*B*)*				
Kr-85m	Kr-87	Kr-88	Xe-135	Xe-133
3.8×10^{-6}	2.8×10^{-6}	3.6×10^{-6}	3.6×10^{-6}	8.0×10^{-6}

* release (*R*) to birth (*B*) ratio of the nuclide

Table 4: Irradiation history of fuel elements HFR-EU1bis/1 and HFR-EU1bis/3

Identification number	Burn-up (% FIMA)	Thermal fluence* ($E < 0.683$ eV) (cm^{-2})	Fast fluence* ($E > 0.1$ MeV) (cm^{-2})
HFR-EU1bis/1	9.34	1.34×10^{21}	2.41×10^{21}
HFR-EU1bis/3	11.07	1.59×10^{21}	2.86×10^{21}

* normalized to the measured burn-up

4. Küfa heating experiments

4.1 HFR-EU1bis/1

The heating test of fuel element HFR-EU1bis/1 began on 2008-03-28. Due to the short cooling time since the end of irradiation (less than three years), the shorter-lived fission products with a half-life around one year were still present (see Table 5). Therefore, it was possible to also measure the release of Ag-110m in addition to the comparatively long-lived cesium isotopes. While Ag-110m is not particularly relevant to accident scenarios, it is of the utmost interest for reactor operation at increased temperatures due to its high volatility. This is particularly important for future HTR variants with direct cycles where gas turbine contamination can be substantial.

Table 5: Fission product inventory of HFR-EU1bis/1 prior to heating experiment (in Bq)

Cs-137*	Sr-90**	Cs-134*	Ru-106*	Kr-85**	I-131**	Xe-133**	Ag-110m*
6.13×10^{10}	5.46×10^{10}	2.81×10^{10}	4.60×10^{10}	6.48×10^{9}	≈ 0	≈ 0	2.79×10^{7}

* measured; ** calculated using the INVENTAR code [Duwe & Kühnlein 1975]

Therefore an especially long phase (200 h) of simulated operation at 1250°C was chosen, with the aim of determining the equilibrium release of Ag-110m during operation and the resulting contamination of the primary circuit. The cesium contamination of the graphite matrix and thus potential particle failure during irradiation was also assessed in this way. After the simulated operation, an accident simulation up to 1600°C (corresponding to the nominal peak fuel temperature obtained from thermal-hydraulic calculations [Haque *et al.* 2006]) was conducted during 200 h. This phase was followed by an additional heating to 1700°C during 150 h. In-between each of the heating phases the fuel element was cooled to room temperature. All temperature values for the heating program of pebble HFR-EU1bis/1 are given in Table 6.

The results of the condensate plate activity measurements are given in Table 7. A graph of the cumulated releases of all measured fission products is shown in Figure 2.

The Kr-85 equilibrium fractional release during the simulated operation at 1250°C amounted to 1.4×10^{-6}. During the second phase at 1600°C a fractional release value of 1.0×10^{-5} was measured, whereas during the third phase at 1700°C the release increased to 2.4×10^{-5}. This corresponds to the inventory of about 0.2 particles.

Already during the first 200 h of simulated operation at 1250°C, a marked fractional release of Cs-137 and Cs-134 on the order of about 7×10^{-5} was found. During the second phase, the accident simulation at 1600°C, the fractional release of Cs increased to about 1.2×10^{-3}. During the third phase at 1700°C the release of Cs surprisingly increased less strongly than expected, so that finally a cumulated fractional release of 4.3×10^{-3} was obtained. The good agreement of the measured release fractions of Cs-137 and Cs-134 confirms that they are in fact due to activity originating from the fuel element and not due to contamination of the condensate plates in the cell or during transport.

Table 6: Heating program for pebble HFR-EU1bis/1

Step no.	Time (hh:mm)	Duration (min)	Temp. (°C)	Plate no.	Step no.	Time (hh:mm)	Duration (min)	Temp. (°C)	Plate no.
1	00:00	30	20–300	86	26	330:00	360	1600	101, 102
2	00:30	300	300–300	86	27	336:00	1080	1600	102, 103
3	06:30	180	300–1250	86	28	354:00	1440	1600	103, 104
4	09:30	630	1250	86	29	378:00	1440	1600	104, 105
5	20:00	1440	1250	86, 87	30	402:00	1560	1600	105, 106
6	44:00	1380	1250	87, 88	31	428:00	690	1600	106, 107
7	67:00	1440	1250	88, 89	32	439:30	930	1600–20	107
8	91:00	1440	1250	89, 90	33	455:00	60	20	108
9	115:00	1500	1250	90, 91	34	456:00	30	20–300	108
10	140:00	1580	1250	91, 92	35	456:30	180	300	108
11	166:30	2580	1250	92, 94	36	459:30	120	300–1250	108
12	209:30	300	1250–20	94	37	461:30	600	1250	108
13	215:30	60	20	95	38	471:30	90	1250–1321	108
14	216:30	30	20–300	95	39	473:00	360	1321–1605	108, 109
15	217:00	180	300	95	40	479:00	120	1605–1700	109, 110
16	220:00	120	300–1250	95	41	481:00	930	1700	110
17	222:00	600	1250	95	42	496:30	420	1700	110, 111
18	232:00	90	1250–1320	95	43	503:30	1140	1700	111, 112
19	233:30	360	1320–1600	95, 96	44	522:30	1380	1700	112, 113
20	239:30	210	1600	96	45	545:30	1620	1700	113, 114
21	243:00	900	1600	96, 97	46	572:30	1440	1700	114, 115
22	258:00	300	1600	97, 98	47	596:30	1440	1700	115, 116
23	263:00	1200	1600	98, 99	48	620:30	630	1700	116, 117
24	283:00	1440	1600	99, 100	49	631:00	900	1700–20	117
25	307:00	1380	1600	100, 101		646:00			

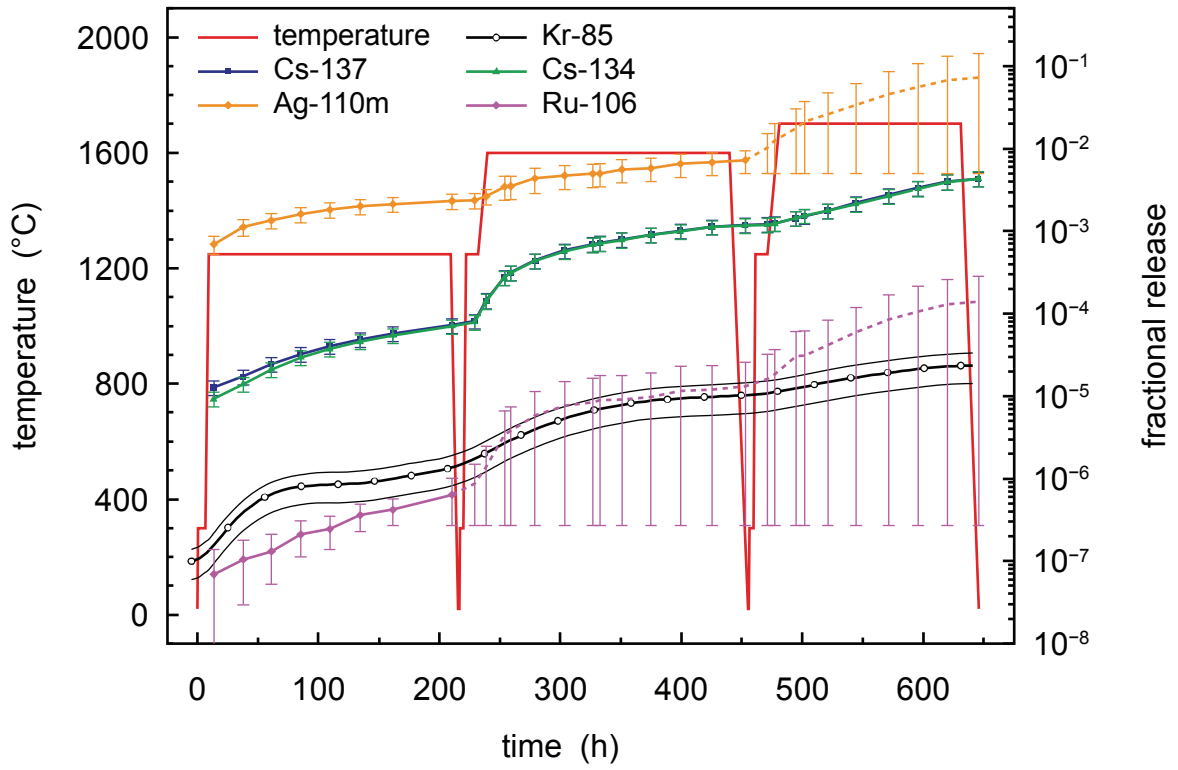

Figure 2: Temperature profile and fractional fission product release for HFR-EU1bis/1

Ag-110m was already strongly released during the first heating phase, with a fractional release up to 2.3×10^{-3}. During the 1600°C phase this value increased to 7.2×10^{-3}. It was however not possible to reliably measure the release during the third phase at 1700°C. Due to the strong release of Cs-137 and Cs-134 it was not possible to bag out the condensate plates and they instead had to be measured through the window of the glove box connected to the hot cells. Because of the high background in that location the low gamma activity of Ag-110m could not be detected. In the graph the cumulated detection limits and the corresponding uncertainties are plotted as conservative limits.

It was possible to measure the release of Ru-106 only during the first phase, up to a fractional release of 6.4×10^{-7}. For the second and third phases, the detection limits and corresponding error bars were again used, as for Ag-110m.

Table 7: Results of the condensate plate activity measurements (in Bq) for HFR-EU1bis/1

Plate no.	Date	Cs-137	Cs-134	Ag-110m	Ru-106
86	2008-04-10	$5.45(17)\times10^{5}$	$1.807(57)\times10^{5}$	$1.31(18)\times10^{4}$	$< 2.24\times10^{3}$
87	2008-04-10	$1.971(62)\times10^{5}$	$9.29(29)\times10^{5}$	$8.1(1.1)\times10^{3}$	$1.12(56)\times10^{3}$
88	2008-04-09	$2.995(94)\times10^{5}$	$1.406(44)\times10^{5}$	$4.32(65)\times10^{3}$	$8.5(6.3)\times10^{2}$
89	2008-04-10	$3.45(11)\times10^{5}$	$1.613(51)\times10^{5}$	$4.80(73)\times10^{3}$	$2.6(1.5)\times10^{3}$
90	2008-04-10	$3.29(10)\times10^{5}$	$1.542(48)\times10^{5}$	$3.88(60)\times10^{3}$	$1.05(98)\times10^{3}$
91	2008-04-10	$3.65(12)\times10^{5}$	$1.690(53)\times10^{5}$	$3.36(55)\times10^{3}$	$3.8(2.0)\times10^{3}$
92	2008-04-09	$3.73(12)\times10^{5}$	$1.734(55)\times10^{5}$	$2.60(45)\times10^{3}$	$1.9(2.0)\times10^{3}$
94	2008-04-10	$6.36(20)\times10^{5}$	$2.996(96)\times10^{5}$	$3.73(73)\times10^{3}$	$< 2.00\times10^{3}$
95	2008-05-05	$3.95(16)\times10^{5}$	$1.792(80)\times10^{5}$	$8.4(6.9)\times10^{2}$	$< 7.70\times10^{3}$
96	2008-05-05	$2.70(10)\times10^{6}$	$1.230(50)\times10^{6}$	$4.9(2.9)\times10^{3}$	$< 1.61\times10^{4}$
97	2008-05-05	$5.61(22)\times10^{6}$	$2.56(11)\times10^{6}$	$< 1.46\times10^{4}$	$< 6.70\times10^{4}$
98	2008-05-06	$1.659(62)\times10^{6}$	$7.57(31)\times10^{5}$	$6.9(8.3)\times10^{2}$	$< 1.09\times10^{4}$
99	2008-05-05	$5.42(21)\times10^{6}$	$2.47(10)\times10^{6}$	$< 1.44\times10^{4}$	$< 6.55\times10^{4}$
100	2008-05-05	$5.95(22)\times10^{6}$	$2.68(11)\times10^{6}$	$6.9(4.9)\times10^{3}$	$< 5.95\times10^{4}$
101	2008-05-06	$4.81(18)\times10^{6}$	$2.165(88)\times10^{6}$	$3.6(3.0)\times10^{3}$	$< 2.24\times10^{4}$
102	2008-05-05	$1.065(43)\times10^{6}$	$4.79(21)\times10^{5}$	$1.5(1.5)\times10^{3}$	$< 1.95\times10^{4}$
103	2008-05-05	$3.37(13)\times10^{6}$	$1.573(68)\times10^{6}$	$1.0(1.2)\times10^{4}$	$< 7.05\times10^{2}$
104	2008-05-06	$4.74(18)\times10^{6}$	$2.123(86)\times10^{6}$	$3.5(3.1)\times10^{3}$	$< 2.54\times10^{4}$
105	2008-05-05	$4.35(17)\times10^{6}$	$1.962(84)\times10^{6}$	$< 1.32\times10^{4}$	$< 5.88\times10^{4}$
106	2008-05-06	$5.30(19)\times10^{6}$	$2.366(93)\times10^{6}$	$3.9(3.0)\times10^{3}$	$< 1.11\times10^{4}$
107	2008-05-05	$2.57(10)\times10^{6}$	$1.150(49)\times10^{6}$	$< 7.86\times10^{3}$	$< 3.54\times10^{4}$
108	2008-05-20	$1.08(19)\times10^{6}$	$4.61(84)\times10^{5}$	—	—
109	2008-05-20	$1.11(17)\times10^{6}$	$5.92(94)\times10^{5}$	—	—
110	2008-05-20	$8.15(88)\times10^{6}$	$4.09(60)\times10^{6}$	—	—
111	2008-05-20	$4.07(45)\times10^{6}$	$1.98(29)\times10^{6}$	—	—
112	2008-05-20	$9.8(1.0)\times10^{6}$	$4.76(71)\times10^{6}$	—	—
113	2008-05-20	$1.82(19)\times10^{7}$	$6.77(74)\times10^{6}$	—	—
114	2008-05-20	$2.36(25)\times10^{7}$	$1.08(16)\times10^{7}$	—	—
115	2008-05-20	$2.61(28)\times10^{7}$	$1.232(97)\times10^{7}$	—	—
116	2008-05-20	$2.67(28)\times10^{7}$	$1.30(20)\times10^{7}$	—	—
117	2008-05-20	$1.59(17)\times10^{7}$	$7.6(1.2)\times10^{6}$	—	—

4.2 HFR-EU1bis/3

The heating experiment on pebble HFR-EU1bis/3 began on 2008-07-23. The fission product inventory of Pebble 3 prior to the heating test is shown in Table 8. The Küfa experiment was performed in two steps. As in the case of HFR-EU1bis/1, the fuel element was heated at the irradiation temperature for a fairly long period of 100 h in order to obtain information on the equilibrium release during operation. Subsequently a classical accident simulation up to 1600°C during 200 h was performed. Between the two heating steps, the fuel element was cooled to room temperature. In order to preserve the fuel condition for other post-irradiation examinations, a further heating to higher temperatures was not performed in this case. All temperature values for the heating program of pebble HFR-EU1bis/3 are given in Table 9.

Table 8: Fission product inventory of HFR-EU1bis/3 prior to heating experiment (in Bq)

Cs-137*	Sr-90**	Cs-134*	Ru-106*	Kr-85**	I-131**	Xe-133**	Ag-110m*
7.22×10^{10}	6.30×10^{10}	3.50×10^{10}	4.89×10^{10}	7.37×10^{9}	≈ 0	≈ 0	3.73×10^{7}

*measured, ** calculated using the INVENTAR code [Duwe & Kühnlein 1975]

Table 9: Heating program for pebble HFR-EU1bis/3

Step no.	Time (hh:mm)	Duration (min)	Temp. (°C)	Plate no.	Step no.	Time (hh:mm)	Duration (min)	Temp. (°C)	Plate no.
1	00:00	60	20	120	15	114:30	210	1250–1412	124
2	01:00	30	20–300	120	16	118:00	240	1412–1600	124, 125
3	01:30	180	300	120	17	122:00	120	1600	125
4	04:30	120	300–1250	120	18	124:00	1380	1600	125, 126
5	06:30	10	1250	120	19	147:00	1380	1600	126, 127
6	06:40	1310	1250	120, 121	20	170:00	180	1600	127, 128
7	28:30	1440	1250	121, 122	21	173:00	1440	1600	128, 129
8	52:30	2610	1250	122, 123	22	197:00	1380	1600	129, 130
9	96:00	120	1250–20	123	23	220:00	1620	1600	130, 131
10	98:00	60	20	124	24	247:00	1500	1600	131, 132
11	99:00	30	20–300	124	25	272:00	1470	1600	132, 133
12	99:30	180	300	124	26	296:30	1530	1600	133, 134
13	102:30	120	300–1250	124	27	322:00	120	1600–20	134, 135
14	104:30	600	1250	124		324:00			

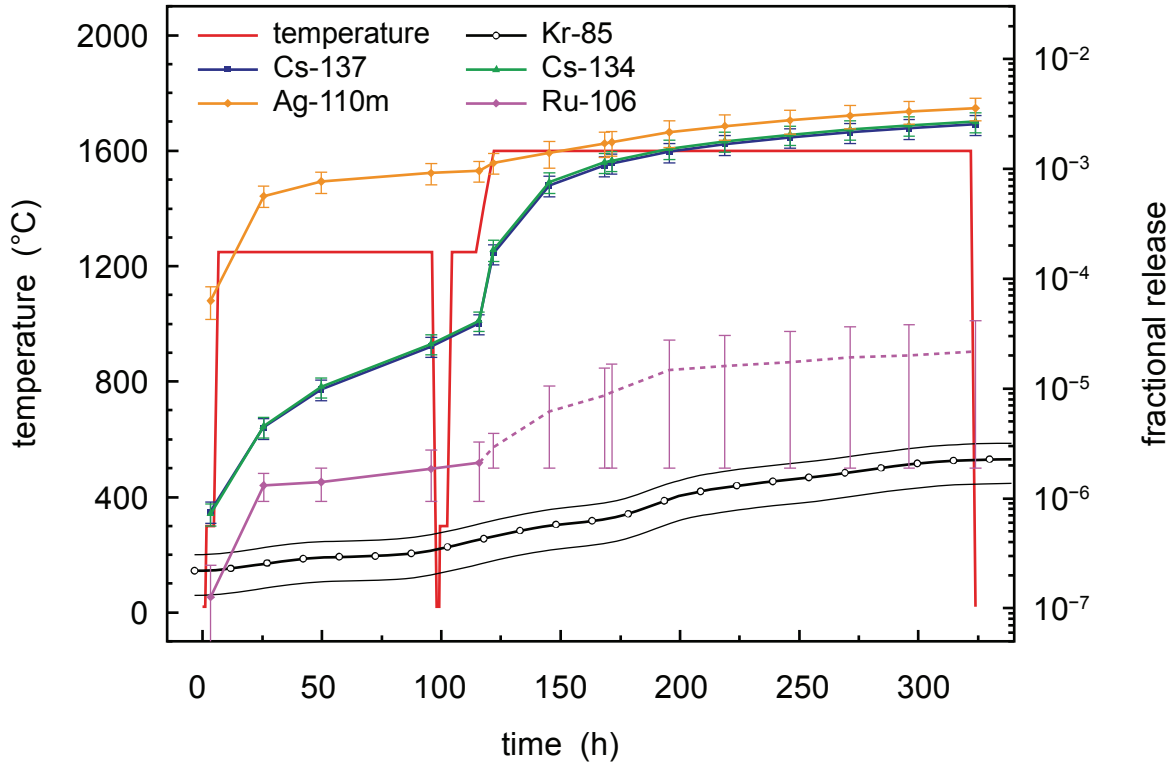

Figure 3: Temperature profile and fractional fission product release for HFR-EU1bis/3

The results of the condensate plate activity measurements are given in Table 10. A graph of the cumulated releases of all measured fission products is shown in Figure 3.

During the first heating phase, the simulated operation, only minute amounts of Kr-85 near the detection threshold of the measurement device were released. During the subsequent accident simulation, the fractional release increased from 3.4×10^{-7} to 2.3×10^{-6}. Apparently no particle fracture occurred during the entire experiment, indicating that the observed release originated exclusively in the graphite matrix.

The fractional release of Cs-137 and Cs-134 amounted to about 4.0×10^{-5} already during the simulated operation. During the accident simulation, it gradually increased by a factor of about 50 to 2.5×10^{-3}.

The fractional release of Ag-110m was considerable and amounted to 9.6×10^{-4} already during the first heating phase. It further increased to 3.6×10^{-3} during the accident simulation. While the Cs release increased visibly at the beginning of the 1600°C heating phase, the increase of the Ag-110m release was relatively weak (only by a factor of about 3).

A release of Ru-106 was observed only during the simulated operation phase. The cumulated fractional release during this phase was 2.1×10^{-6}. During the 200-h heating to 1600°C the activity deposited on the condensate plates was below the detection threshold. For this reason, the release values during this phase are conservatively approximated by the respective detection thresholds and assigned uncertainties of the same magnitude.

Table 10: Results of the condensate plate activity measurements (in Bq) for HFR-EU1bis/3

Plate no.	Date	Cs-137	Cs-134	Ag-110m	Ru-106
120	2008-08-20	$3.83(40)\times10^{4}$	$1.77(19)\times10^{4}$	$1.52(40)\times10^{3}$	$< 4.10\times10^{3}$
121	2008-08-25	$1.83(17)\times10^{5}$	$9.26(85)\times10^{4}$	$1.20(13)\times10^{4}$	$3.81(76)\times10^{4}$
122	2008-08-20	$2.75(25)\times10^{5}$	$1.43(13)\times10^{5}$	$4.74(48)\times10^{3}$	$< 3.04\times10^{3}$
123	2008-08-20	$7.19(66)\times10^{5}$	$3.70(34)\times10^{5}$	$3.66(66)\times10^{3}$	$< 1.46\times10^{4}$
124	2008-08-28	$7.44(68)\times10^{5}$	$3.89(35)\times10^{5}$	$9.7(2.8)\times10^{2}$	$< 8.15\times10^{3}$
125	2008-08-28	$6.60(60)\times10^{6}$	$3.47(31)\times10^{6}$	$4.09(91)\times10^{3}$	$2.6(2.6)\times10^{4}$
126	2008-08-20	$2.70(24)\times10^{7}$	$1.40(13)\times10^{7}$	$6.2(5.9)\times10^{3}$	$< 1.06\times10^{5}$
127	2008-08-25	$1.85(17)\times10^{7}$	$9.52(86)\times10^{6}$	$7.7(2.7)\times10^{3}$	$< 8.02\times10^{4}$
128	2008-08-20	$2.46(22)\times10^{6}$	$1.27(12)\times10^{6}$	$9.5(5.7)\times10^{2}$	$< 2.03\times10^{4}$
129	2008-08-20	$1.51(14)\times10^{7}$	$7.85(71)\times10^{6}$	$9.6(6.4)\times10^{3}$	$< 1.76\times10^{5}$
130	2008-08-28	$1.27(11)\times10^{7}$	$6.48(59)\times10^{6}$	$7.4(1.5)\times10^{3}$	$< 4.45\times10^{4}$
131	2008-08-25	$1.30(12)\times10^{7}$	$6.57(60)\times10^{6}$	$7.0(1.6)\times10^{3}$	$< 4.67\times10^{4}$
132	2008-08-28	$1.077(98)\times10^{7}$	$5.47(50)\times10^{6}$	$6.6(1.7)\times10^{3}$	$< 5.34\times10^{4}$
133	2008-08-25	$9.74(88)\times10^{6}$	$4.91(45)\times10^{6}$	$6.88(99)\times10^{3}$	$< 2.53\times10^{4}$
134	2008-08-28	$9.92(90)\times10^{6}$	$5.02(46)\times10^{6}$	$5.6(1.5)\times10^{3}$	$< 5.24\times10^{4}$

5. Summary and conclusions

The fuel element HFR-EU1bis/1 was heated to a maximum temperature of 1700°C in three phases, whereas fuel pebble HFR-EU1bis/3 was heated up to 1600°C in two heating phases. Between each heating phase the fuel elements were cooled to room temperature. During the first phases of the Küfa experiments, the temperature was set to 1250°C, as during the irradiation. This was mainly done to obtain information on the release of Ag-110m at the increased irradiation temperature of the HFR-EU1bis experiment. The cumulated fractional releases of gaseous and volatile fission products are summarized in Tables 11 and 12.

The release curve of Kr-85 didn't show any obvious jumps either at 1250°C or during the accident simulations. The cumulated fractional release of Kr-85 was 2.4×10^{-5} for HFR-EU1bis/1 and 2.3×10^{-6} for HFR-EU1bis/3. The overall low release of Kr-85 indicates that no particles failed during the experiment and merely represents the expected release from free uranium contamination in the graphite matrix.

A notable observation is the relatively high release of cesium. The cumulated fractional release of both investigated isotopes reached 4.3×10^{-3} for HFR-EU1bis/1 and 2.7×10^{-3} for HFR-EU1bis/3, indicating increased diffusion through intact particle layers or external contamination of the graphite matrix.

Table 11: Summary of the results of the Küfa heating test with pebble HFR-EU1bis/1

Heating temperature (°C)	Heating duration (h)	Fractional release				
		Kr-85	Cs-137	Cs-134	Ag-110m	Ru-106
1250	210	1.4×10^{-6}	7.2×10^{-5}	7.0×10^{-5}	2.3×10^{-3}	6.4×10^{-7}
1600	200	1.0×10^{-5}	1.2×10^{-3}	1.2×10^{-3}	7.2×10^{-3}	—
1700	150	2.4×10^{-5}	4.3×10^{-3}	4.3×10^{-3}	—	—

Table 12: Summary of the results of the Küfa heating test with pebble HFR-EU1bis/3

Heating temperature (°C)	Heating duration (h)	Fractional release				
		Kr-85	Cs-137	Cs-134	Ag-110m	Ru-106
1250	100	3.4×10^{-7}	3.9×10^{-5}	4.1×10^{-5}	9.6×10^{-4}	2.1×10^{-6}
1600	200	2.3×10^{-6}	2.5×10^{-3}	2.7×10^{-3}	3.6×10^{-3}	—

In HFR-EU1bis/1, the leveling-off of the release curve for Cs-137 and Cs-134 during the third phase was not expected from the behavior during the second phase. Possibly the higher temperature during the second phase, following the simulated operation, led to an increased release of the cesium present in the outer particle layers and in the graphite matrix, such that these "reservoirs" were depleted and diffusion from the inner particle layers through the SiC layer could not replenish them quickly enough. The good agreement between the releases of Cs-137 and Cs-134 indicates that the measured activity of the condensate plates is indeed due to release from the fuel element and not due to external contamination.

The fractional release of Ag-110m already amounted to 2.3×10^{-3} for HFR-EU1bis/1 and 9.6×10^{-4} for HFR-EU1bis/3 during the simulated operation phases. The subsequent release during the accident simulation was lower than expected.

To determine the origin of the relatively high initial release of cesium and silver at 1250°C, a detailed analysis based on a mechanical failure model as well as a diffusion simulation was carried out. It indicated a likely contamination of the graphite matrix during the irradiation campaign. This may have been caused by an increased irradiation temperature causing an enhanced diffusion through the intact coatings of the particle, especially the SiC layer. It is also conceivable that an external contamination of the graphite matrix occurred during the irradiation, possibly from failed particles in one of the fuel elements irradiated as part of experiment HFR-EU1bis. During one of the irradiation cycles, the fuel element capsule was subjected to an unexpectedly high temperature excursion. This may have led to the failure of a number of coated particles, whose inventory was then released into the graphite matrix.

The contamination of the graphite matrix was probably less important for the cesium release from the HFR-EU1bis fuel elements under accident conditions, since the observed releases were comparatively well reproduced by the applied diffusion model. They were, however, consistently about two to three orders of magnitude higher than those observed from the HFR-K6 pebbles [Freis *et al.* 2010]. This is presumed to be due to differences in batch production or irradiation conditions; in this respect no qualitative difference between the fuel elements HFR-EU1bis/1 and HFR-EU1bis/3 was observed.

Overall, our results point to the conclusion that the irradiation temperature of TRISO fuel elements should remain below 1250°C to prevent fission product release at a level which is unacceptable for inherently safe HTR concepts.

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