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BURN-UP DETERMINATION OF HIGH TEMPERATURE REACTOR SPHERICAL FUEL ELEMENTS BY GAMMA SPECTROMETRY

Daniel FREIS, Ramil NASYROW and Enrique H. TOSCANO

European Commission, Joint Research Centre, Institute for Transuranium Elements,
Postfach 2340, Karlsruhe, Germany

ABSTRACT

The burn-up of nineteen High Temperature Reactor irradiated spherical fuel elements (SFE) has been determined by measuring the gamma activity, using an ad-hoc designed conical collimator. The fuel elements were irradiated (up to 10% FIMA) in different experiments conducted at the High Flux Reactor in Petten, the AVR experimental reactor in Juelich and in the FRJ2-Reactor in Juelich. The quantitative results obtained, based on the measurement of ¹³⁷Cs activity, are presented and compared with previous results obtained in the Forschungszentrum Juelich using a similar technique and with the results acquired from code calculations.

INTRODUCTION

The population of the EU share a concern about future nuclear power generation. The acceptance of currently established nuclear technology, mainly due to safety issues but also from the increasing difficulty of maintaining economic competitiveness in a deregulated market, is a main issue in the present energy policy of many countries. At present, light water reactors dominate the worldwide nuclear energy production. Some advanced designs, like the European Pressurised Water Reactor (EPR), meet the safety requirements but this concept is based on active safety systems, which are costly and require a more detailed safety demonstration. On the other hand, the High Temperature Reactors (HTRs), being inherently safe, can meet all the requirements, reducing the costs without affecting the safety.

In particular, the Very High Temperature Reactor (VHTR) constitutes - together with the Sodium Fast Reactors (SFR) - the nearest term of reference for the Generation IV Roadmap

reactor concepts. The today VHTR reference concept is a helium-cooled, graphite moderated, thermal neutron spectrum reactor with an outlet temperature of 1000°C or higher. The reactor will produce both electricity and hydrogen, using an intermediate heat exchanger to transfer the heat to a hydrogen production technology or a gas turbine.

In the framework of an Integrated Project (IP-RAPHAEL) from the European Commission, several tasks aimed at the development of VHTR technology have been undertaken. The project includes developments in the fields of reactor physics, fuel technology, safety, material needs and feasibility of key components and systems. In the domain of fuel technology a key point is represented by the testing of the irradiation behaviour of new type of fuels and their fabrication methods. In this context, irradiation experiments and the post-irradiation testing of irradiated fuels play a paramount role, and several techniques are being installed in different European laboratories to this purpose.

In particular, in the hot cell installation of the Institute for Transuranium Elements (ITU), the so-called Cold Finger Apparatus (KüFA), developed in the Forschungszentrum Juelich (FzJ) to test the fission product release of HTR-fuels has been installed as an up-graded version. In this context, 21 spherical fuel elements (SFE) from different irradiation experiments performed in the past in various testing reactors were transferred from the FzJ to ITU.

In addition, in the framework of the IP-RAPHAEL, the infrastructure necessary for the quantitative determination of

the burn-up of spherical fuel elements by gamma spectroscopy has been developed, with the twofold goal of testing SFEs before and after the KÜFA tests and to determine quantitatively the achieved burn-up during the irradiation experiments to be performed in the framework of IP-RAPHAEL.

The present paper presents the experimental details and the quantitative results of the burn-up determination obtained on nineteen SFE, coming from irradiation experiments conducted in the High Flux Reactor in Petten, the DIDO reactor and the AVR reactor in Juelich up to a maximum burn-up of 10% FIMA. The quantitative results obtained, based on the measurement of ^{137}Cs activity, are presented and compared with previous results obtained by the Forschungszentrum Juelich using a similar technique and with the results acquired from code calculations.

NOMENCLATURE

1. Theoretical Background

Several methods can be applied to determine the burn-up of irradiated fuel elements ^[1,2], but in the case of HTR fuel elements, the procedure had to meet certain requirements. The most important of these requirements is that the measurements should be done non-destructively since the fuel elements will be the object of further investigation in accident simulation tests. This request is fulfilled by using gamma spectrometry to establish the fission product inventory of a monitor isotope which can be related to burn-up. The monitor isotope should allow a simple and accurate determination of the burn-up, even though the exact irradiation history of the pebbles is not known. Last but not least, enough of the isotope used as burn-up monitor should be present in the spherical fuel elements to be measured in a reasonable short time with a low uncertainty.

All these requirements for the burn-up determination are fulfilled by using gamma spectrometry to establish the fission product inventory of ^{137}Cs as burn-up monitor. In fact, ^{137}Cs has a long half life time (30.09 years) as compared to the irradiation times, usually lasting between 3 years to a maximum of 6 years. Therefore, its decay plays only a minor role for long irradiation times and can be easily corrected using simple mathematics. In addition, the cross section of ^{137}Cs for neutron capture (n,γ) is very small with a value of about 0.25 barns and, thus, this neutron reaction is negligible. The generation of ^{137}Cs from ^{136}Xe through neutron capture and subsequently β -decay can be neglected likewise since the cross section for this neutron reaction of about 0.23 barns is also very small. Therefore, generation and decay of ^{137}Cs in the reactor is almost only determined by the fission of ^{235}U and ^{239}Pu and behaves linearly as a function of the burn-up^[1]. The fission yield of ^{137}Cs is different for each fissile heavy metal isotope. Nevertheless, the value of $6.33 \cdot 10^{-2}$ (which

considers also Pu-burning), can be adopted as a good approximation. Finally, another rationale to use ^{137}Cs as a burn-up monitor is that its short living daughter nuclide $^{137\text{m}}\text{Ba}$ has a very clear gamma peak at 661.61 keV, which can easily be identified by gamma spectrometry; due to the short half life time of $^{137\text{m}}\text{Ba}$, the activity of $^{137\text{m}}\text{Ba}$ and ^{137}Cs is proportional.

Taking into account the above mentioned particulars, an equation for the build-up and decay of ^{137}Cs during irradiation can be written as follows:

$$\frac{dN_{^{137}\text{Cs}}}{dt} = -\lambda_{^{137}\text{Cs}} \cdot N_{^{137}\text{Cs}} + \gamma_{^{137}\text{Cs}} \cdot \bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM} \quad \text{Eq. 1}$$

Where:

$\gamma_{^{137}\text{Cs}}$: fission yield, $\bar{\sigma}_f$: average fission cross section,

$\bar{\phi}_n$: neutron flux, $\lambda_{^{137}\text{Cs}}$: decay constant

N_{HM} : heavy metal atoms, $N_{^{137}\text{Cs}}$: ^{137}Cs atoms

Considering the following initial condition:

$$N_{^{137}\text{Cs}}(t_{irr.} = 0) = 0$$

Then, the solution of Equation 1 is:

$$N_{^{137}\text{Cs}}(t_{irr.}) = \frac{\gamma_{^{137}\text{Cs}} \cdot \bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM}}{\lambda_{^{137}\text{Cs}}} \cdot \left(1 - e^{-\lambda_{^{137}\text{Cs}} \cdot t_{irr.}} \right) \quad \text{Eq. 2}$$

The burn-up (BU), defined as the number of fissions per initial metal atoms (FIMA), can be determined by the simplifying assumption that the number of atoms available for fission is nearly constant during the irradiation. Hence, the following relation between the irradiation time ($t_{irr.}$) and the burn-up can be established:

$$BU = \frac{(\bar{\sigma}_f \cdot \bar{\phi}_n \cdot N_{HM}) \cdot t_{irr.}}{N_{HM}} \quad \text{Eq. 3}$$

Replacing of Equation 3 in Equation 2 leads to:

$$BU = N_{^{137}\text{Cs}}(t_{\text{irr.}}) \cdot \frac{t_{\text{irr.}} \cdot \lambda_{^{137}\text{Cs}}}{\gamma_{^{137}\text{Cs}} \cdot N_{\text{HM}}} \cdot \left(1 - e^{(-\lambda_{^{137}\text{Cs}} \cdot t_{\text{irr.}})}\right)^{-1}$$

$$\Leftrightarrow BU = \frac{N_{^{137}\text{Cs}}(t_{\text{irr.}})}{\gamma_{^{137}\text{Cs}} \cdot N_{\text{HM}}} \cdot C_{\text{corr}}(t_{\text{irr.}}) \quad \text{Eq. 4}$$

From the last equation, the burn-up is proportional to the amount of Cs, excepting for the correction term C_{corr} .

$$C_{\text{corr}} = t_{\text{irr.}} \cdot \lambda_{^{137}\text{Cs}} \cdot \left(1 - e^{(-\lambda_{^{137}\text{Cs}} \cdot t_{\text{irr.}})}\right)^{-1}$$

Due to the relatively long half-life of ^{137}Cs , this correction term should be taken into account only for long irradiation times (more than 5 years). As can be seen from Fig.1, for irradiation times of 3 years, the correction amounts to about 3 %.

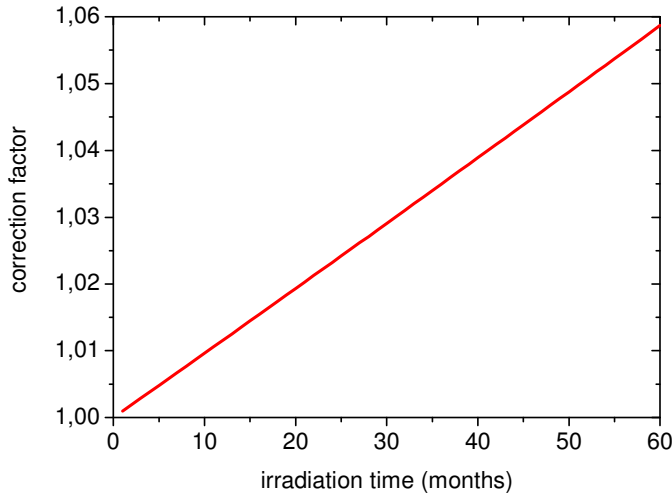


Figure 1: Correction factor for long time irradiation

Rearranging Eq. 4, the following relationship between the ^{137}Cs inventory and the burn-up can be easily deduced:

$$N_{^{137}\text{Cs}}(t_{\text{irr.}}) = BU \cdot \frac{\gamma_{^{137}\text{Cs}} \cdot N_{\text{HM}}}{t_{\text{irr.}} \cdot \lambda_{^{137}\text{Cs}}} \cdot \left(1 - e^{(-\lambda_{^{137}\text{Cs}} \cdot t_{\text{irr.}})}\right) \quad \text{Eq. 5}$$

Where constant power during irradiation has been assumed as the initial condition. In Fig. 2, the results from

Eq. 5 are compared with calculations using the programs Korigen^[3] from Forschungszentrum Karlsruhe (FzK) and Inventar^[4] from FzJ. The Korigen code needs burn-up dependent cross section libraries. Since these libraries are available for burn-ups up to 6% FIMA, the Korigen calculations were performed only up to this value. Nevertheless, calculations by means of the ORIGEN-code, using SCALE libraries, were performed up to higher burn-ups, showing similar results to those from Eq. 5^[5].

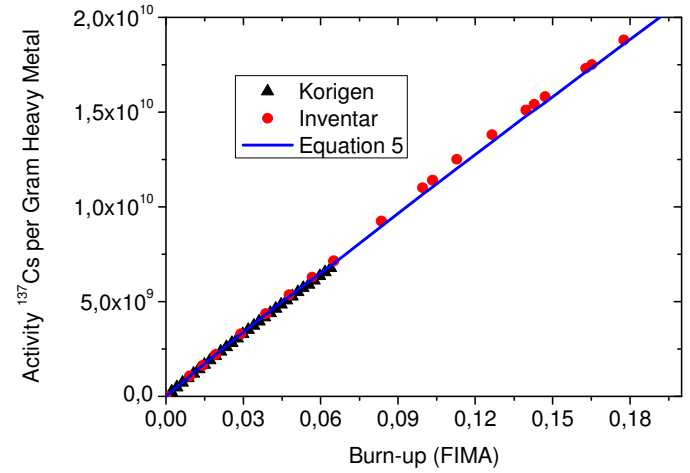


Figure 2: ^{137}Cs inventory calculated with different methods

In conclusion, the values obtained using Eq. 5 are in good agreement with established calculation programs and, hence, the errors coming from the approximations adopted can be considered as negligible. Consequently, if the ^{137}Cs inventory is determined with sufficient accuracy, Eq. 2 can be used for the determination of the burn-up of the spherical fuel elements. In what follows, the experimental setup for the determination of the ^{137}Cs inventory by gamma spectrometry will be described in detail.

2. Experimental

2.1. Setup in the Hot Cell

The measurement was performed in a hot cell, with a collimator placed in a hole drilled through the wall between the spherical sample and the detector. To average any inhomogeneity in the fuel distribution of a single spherical fuel element, the specimen was rotated (typically, at 30 rpm) during the entire measurement, using a revolving table.

The measuring system consists of a pure germanium detector and a signal amplifier. The impulses provided by the Ge counter are analysed simultaneously by a full featured 16K channel integrated Multi-channel Analyser (MCA) based on digital signal processing techniques (DSA-1000 from Canberra). The analysis of the peaks was performed and displayed by the software InterWinner®.

2.2. Description of the Collimator

Gamma detectors are generally limited in the amount of gamma-photons per unit time that can be detected without saturation. For this reason, depending on the activity of the specimen to be measured, a collimator has usually to be placed between the samples and the gamma detector. If quantitative measurements are envisaged, the characteristics of the collimator have to be adapted to the specific geometry of the samples to be measured. In the present case, the spherical fuel element geometry calls for a conical geometry of the collimator in order to be able to detect the whole active zone of this type of samples.

Hence, to be able to perform quantitative measurements of spherical fuel elements, a conical collimator, based on the pinhole camera principle, was developed. It consists of a cylinder, made of the tungsten alloy Denal® D175 (density 17.5 g cm⁻³), in which two cones were drilled in both sides (as illustrated in Fig. 3), having an aperture angle that takes into account the distance between the central orifice and the active zone of the detector on one side, and the spherical fuel element on the other. The pinhole camera principle ensures that the whole fuel zone of the pebble is seen by the detector without loss of information. In addition, the absorption of the gamma radiation in the collimator remains constant for the whole spherical geometry.

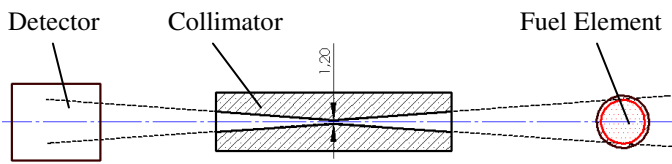


Figure 3: Illustration of the Collimator Principle

The dimensions of the collimator can be established by assessing the maximum and minimum ¹³⁷Cs activity expected from a fuel element and the detection and saturation limits of a gamma detector. The maximum can be estimated to be about 10¹² Bq, which corresponds to a very high burn-up of about 87 % FIMA. Assuming this maximum activity, the gamma ray intensity of the pebble would be about 8.4 · 10¹¹ quanta per second. Since the saturation limit of current detectors lies in the range of 10⁵ impulses per second, then, the total absorption factor (k_s) has to be below 10⁻⁷. On the other hand, the system should be able to detect an activity as low as possible. A reasonable minimum value for the rate of impulses is about 10 imp/second. This corresponds to a minimum activity of about 10⁹ Bq or 8.4·10⁸ emitted gamma quanta per second (which corresponds to a very low burn up of about 0.1 % FIMA). Then, the absorption factor has to be higher than 10⁻⁸.

$$10^{-8} \leq k_s \leq 10^{-7}$$

Considering that the half value width, dk, of the tungsten alloy for a gamma energy of 661.61 keV, amounts to 4.1 mm and the mass attenuation coefficient, μ, amounts to 0.169 mm⁻¹, then the geometry of the collimator can now be calculated by assuming an effective aperture surface. This effective surface can be estimated to be composed of the surface of the aperture and a virtual border surface (A_{border}). The free surface of the hole amounts, in our particular case, to about 0.5 mm².

The virtual border surface is now calculated using the half value width and the geometry of the collimator. If it is assumed that, due to the relatively long distance between the source and the collimator the radiation arrives parallel, then, the wall thickness (x), which has to be traversed by the gamma rays, can be estimated as a function of the radius (r):

$$x = \frac{2 \cdot (r - r_{hole})}{\tan\left(\frac{\alpha}{2}\right)}$$

Where α is the aperture angle of the collimator. Then:

$$I(x) = I_0 \cdot e^{-\mu \cdot x}$$

The virtual border surface can now be determined by integration of the attenuation factor over the whole cross section surface of the collimator.

$$A_{border} = \int_{r_{hole}}^{r_a} 2 \cdot \pi \cdot r \cdot e^{-\left(\frac{2 \cdot \mu \cdot (r - r_{hole})}{\tan\left(\frac{\alpha}{2}\right)}\right)} dr$$

With the specified values the virtual border surface amounts to about 0.22 mm². The effective aperture surface amounts, accordingly, to ~ 0.7 mm².

In summary, for the particular geometry in our hot cell, the collimator had to have a length of 346 mm. The cone has an aperture angle of 2.86° and the minimum aperture at the centre of the collimator amounts to 0.8 mm. The outer diameter was selected to be 48 mm. This geometry guarantees that the inner fuel zone (about 50 mm) of the fuel element is totally covered in the measurement.

The total attenuation factor of the collimator can now be written as the ratio between the gamma intensity of the whole pebble and the intensity which passes through by the

collimator. For the geometry of our collimator, this factor was experimentally determined to amount to $5.94 \cdot 10^{-8}$.

2.3. Calibration of the Gamma Detector

Gamma detectors have to be calibrated for a particular geometry to establish the efficiency of the system in order to be able to obtain quantitative results. This can be performed using adequate calibration standards. The ideal situation for an efficiency calibration is to have a calibration standard identical to the sample to be measured, keeping all the geometrical factors constant. In our case, a graphite spherical sample containing a known amount of radioactive material, distributed in the matrix as in the spherical fuel elements would be the most approximate situation but, regrettably, this configuration is difficult to accomplish. A relatively easy to perform arrangement is a point source in the centre of a graphite sphere and, then, to obtain the quantitative calibration of the gamma measuring system, perform the necessary corrections for the respective gamma radiation self-absorption, both in the standard and the samples.

2.3.1. Description of the standard

In order to manufacture the type of standard previously discussed, a hole (12 mm in diameter) was drilled up to the centre of a graphite sphere, having the same diameter (60 mm) as the spherical fuel elements to be measured. Then a graphite insert with a central hole was fabricated and filled with a known amount of a solution (of which the amount of ^{137}Cs had been accurately determined), coming from the dissolution of a fuel pellet taken from a PWR-reactor fuel rod. The solution was allowed to dry and a second layer of fuel solution was filled in, repeating these steps several times until the desired ^{137}Cs activity (about $2 \cdot 10^9$ Bq) in the insert was achieved. Subsequently, the insert was fixed with resin and mounted into the graphite sphere. Finally, the insert was fixed by screwing a graphite pin, as shown in Fig. 4.

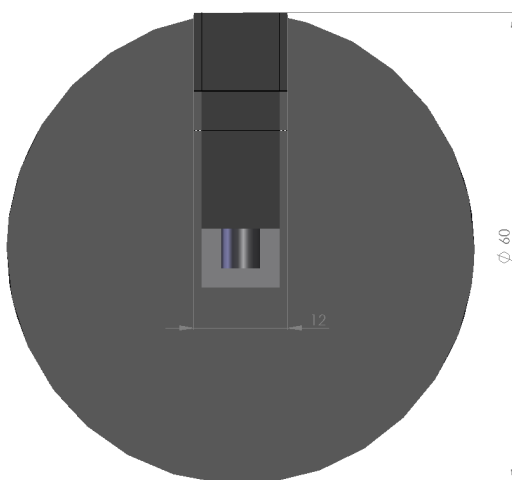


Figure 4: Standard Sphere

2.3.2. Inventory in the standard

The fuel solution filled in the standard sphere was obtained by dissolving a fuel pellet from a PWR-fuel rod, burn-up ~10.4 % FIMA. The fission product inventory, including ^{137}Cs , of this solution was determined by gamma spectrometry^[9] (see Table I) whereas the burn-up was established by determining the ^{148}Nd -content by IC-ICP-MS^[2].

Radionuclide	A (Bq/g solution)
^{60}Co	3.5 ± 1.7
^{106}Rh	$1.27 \cdot 10^3 \pm 0.07 \cdot 10^3$
^{125}Sb	$1.07 \cdot 10^3 \pm 0.05 \cdot 10^3$
^{134}Cs	$4.61 \cdot 10^4 \pm 0.05 \cdot 10^4$
^{137}Cs	$3.16 \cdot 10^5 \pm 0.06 \cdot 10^5$
^{144}Ce	$1.06 \cdot 10^3 \pm 0.05 \cdot 10^3$
^{144}Pr	$1.37 \cdot 10^3 \pm 0.25 \cdot 10^3$
^{154}Eu	$1.01 \cdot 10^4 \pm 0.01 \cdot 10^4$
^{155}Eu	$4.25 \cdot 10^3 \pm 0.05 \cdot 10^3$
^{241}Am	$3.15 \cdot 10^3 \pm 0.05 \cdot 10^3$

Table I: Results of the gamma measurement of the diluted fuel solution

The ^{137}Cs final activity in the standard ($A_{137\text{-Cs, st}}$) was established by multiplying the dilution factor (f_{dil}) by the measured activity per gram of the dilution ($A_{137\text{-Cs, dil}}$) and the total mass of the original solution brought in into the sphere (m_{st}).

2.3.3. Determination of the calibration factor Standard

The calibration factor for the quantitative determination of the burn-up on basis of the gamma measurement is the ratio between the counted impulses per unit time and the known ^{137}Cs activity in the standard. In general, the calibration factor has to be corrected considering a geometrical term (which takes into account the distance as well as the geometry of the detector, the standard and the sample), an attenuation factor for the collimator, a factor for the energy dependent efficiency of the detector, a factor for the probability of gamma emission at the given energy and, most importantly, a correction factor, which considers the differences in geometry and self absorption between the sample and the standard. In our case, given the standard described in § 2.3.1., all these factors are constant, except for the correction factor for the differences in the geometry and self-absorption between standard and fuel elements. Hence, the correction factor can be expressed as one proportional constant, c_{pc} , which takes into account the differences in self-absorption in the standard as compared to the fuel element.

If it is supposed that the fuel zone of the spherical fuel element is homogenous and is assumed to be composed of infinitely point sources of radiation, then the calculation of the total attenuation in the fuel element can be performed by integration over the total volume:

$$f_{pebble} = \frac{1}{a \cdot V_p} \cdot \int_{V_p} f_{self}(P) \cdot f_{dist}(P) \cdot f_{layer}(P) \cdot a \cdot dV$$

In Table II, the correction factors for the different fuel element designs are gathered. The correction factors have been calculated considering the different absorption coefficients for the fuel zone and the differences in geometry between sample and standard. The detailed calculations can be found in the Appendix A.

Type	μ_B (mm ⁻¹)	r_i (mm)	r_a (mm)	f_{pebble}	f_{corr}
HFR-K5	0.0147	25	30	$5.839 \cdot 10^{-8}$	1.0158
HFR-K6	0.0147	25	30	$5.839 \cdot 10^{-8}$	1.0158
GLE 3	0.0148	25	30	$5.829 \cdot 10^{-8}$	1.0141
GLE 4 (4-2)	0.0141	25	30	$5.907 \cdot 10^{-8}$	1.0276

Table II: Correction factors for several types of fuel element

The calculation of the correction factors was performed using analytical mathematics and is, therefore, exact except for the small error from neglecting the angle of incidence, as discussed in Appendix A. Hence, it can be expected that the error made through the correction factor is below 0.05 % and, thus, negligible, especially when considering that the inaccuracy in the spheres geometry and the statistical error of the gamma measurement can be significant higher.

3. Specimens

3.1. Description of the Spherical Fuel Elements

The irradiated fuel elements measured were of various types and had particular irradiation histories in three different test reactors. Some of them were irradiated in the experimental reactor AVR Juelich, some in the FRJ-2 Juelich and the remainder in the High Flux Reactor (HFR) in Petten. The cooling time to the date of measurement varied from 12 to 21 years (see Table III).

All spherical fuel elements consist of a graphite matrix and have an outer diameter of 60 mm. The fuel is dispersed in many coated particles, which are placed in an inner fuel zone

of 50 mm in diameter. The graphite matrix has a density of 1.75 g/cm³.

Sphere	Type of FE	End of Irr.
HFR-K6/1	GLE-4 similar	May-93
HFR-K6/2	GLE-4 similar	May-93
HFR-K6/3	GLE-4 similar	May-93
HFR-K6/4	GLE-4 similar	May-93
HFR-K5/1	GLE-4 similar	May-94
HFR-K5/2	GLE-4 similar	May-94
HFR-K5/3	GLE-4 similar	May-94
HFR-K5/4	GLE-4 similar	May-94
FRJ2-KA2/1	GLE-4-2	May-90
FRJ2-KA2/2	GLE-4-2	May-90
FRJ2-KA2/3	GLE-4-2	May-90
AVR-74/16	GLE-3	Feb-85
AVR-74/18	GLE-3	Feb-85
AVR-73/22	GLE-4	Feb-84
AVR-87/06	GLE-4	Nov-88
AVR-87/07	GLE-4	Nov-88
AVR-87/08	GLE-4	Nov-88
AVR-87/09	GLE-4	Nov-88
AVR-87/10	GLE-4	Nov-88

Table III: Tested Fuel Elements

All fuel elements contain modern TRISO type coated particles (two layers of PyC, one embedded SiC layer and one buffer layer) with low enriched uranium-oxide (UO₂). The difference between GLE-3 type elements and GLE-4 type lies in the heavy metal loading and enrichment. GLE-3 spheres have a total loading of 10 g uranium with an enrichment of 10% ²³⁵U. The fuel is dispersed in 16,400 coated particles, having an outer diameter of 920 μm. GLE-4 spheres have a total loading of 6 g uranium with 16.7% enrichment and the fuel is dispersed in 10,000 coated particles. The fuel elements from the irradiation tests HFR-K5 and HFR-K6 were manufactured under similar conditions than the GLE-4 type spheres. They contain 14,600 coated particles with a total uranium loading of 9.44 g and an enrichment of 10.6% ²³⁵U.

3.2. Irradiation Experiments

3.2.1. HFR-K5 Experiment

The HFR-K5 irradiation experiment was performed on four fuel spheres, having a design for the operation in the 200 MWth Modular High Temperature Reactor (HTR-MODUL) of Siemens/KWU.

The main irradiation data is given in table IV^[6].

Parameter	Fuel element designation			
	HFR-K5/1	HFR-K5/2	HFR-K5/3	HFR-K5/4
Burn-up [%FIMA]	6.7	8.8	9.1	8.7
Neutron flux $10^{25} \text{ m}^{-2} [> 0.1 \text{ MeV}]$	4.0	5.8	5.9	4.9
Max central temperature ^(a) [$^{\circ}\text{C}$]	1 020	1 070	1 010	1 030
Max power output ^(b) [kW/FE]	2.21	3.03	3.19	3.06

Table IV: HFR-K5 Test Element Specification and Irradiation Data

a) Calculated from the measured surface temperature and FE power output.

b) Fission Power and γ –heat

3.2.2. HFR-K6 Experiment

As for the HFR-K5 experiment, the spherical fuel elements HFR-K6 were designed for the planned reactor HTR-MODUL.

The main irradiation data is given in the table V^[6].

Parameter	Fuel element designation			
	HFR-K6/1	HFR-K6/2	HFR-K6/3	HFR-K6/4
Burn-up (% FIMA)	7.2	9.3	9.7	9.2
Neutron flux $10^{25} \text{ m}^{-2} [> 0.1 \text{ MeV}]$	3.2	4.6	4.8	4.5
Max central temperature ^(a)	1 090	1 130	1 140	1 130
Max power output ^(b)	1.82	2.51	2.70	2.48

Table V: HFR-K6 Test Element Specification and Irradiation Data

a) Calculated from the measured surface temperature and FE power output.

b) Fission Power and γ –heat

3.2.3. FRJ2-KA2

The FRJ2-KA 2 irradiation tests were performed on GLE-4-2 type fuel spheres. They are identical to type GLE-4 with a different type of matrix graphite (NUKEM A3-3 instead of A3-27). The irradiation was performed up to a maximum burn-up of 2.02 %FIMA. The test was intended to determine the uranium contamination of the matrix during fabrication^[7]. The spheres have been measured by gamma spectrometry before in the KFA Juelich^[8] and, thus, their inventory (given in Table VI) is well known.

FRJ2-KA 2	Burnup (FIMA)	Fission product inventory (Bq)		
		^{131}I	^{133}Xe	^{137}Cs
1	1.84	1.09E+12	2.86E+12	1.28E+10
2	2.00	1.18E+12	3.07E+12	1.38E+10
3	2.02	1.19E+12	3.11E+12	1.40E+10

Table VI: Fission product inventory in FRJ2-KA 2 Fuel Elements (reference date May 1990)

The main irradiation data is given in the table VII^[8]. Since the capsules were not instrumented, the values for the temperature could only be calculated.

Parameter	Fuel element designation		
	FRJ2-KA 2/1	FRJ2-KA 2/2	FRJ2-KA 2/3
Burn-up (% FIMA)	1.84	2.00	2.02
Neutron flux $10^{25} \text{ m}^{-2} [> 0.1 \text{ MeV}]$	< 0.2	< 0.2	< 0.2
Max centre temperature ^(a)	550	550	550
Max power output ^(b)	1.56	1.70	1.72

Table VII: FRJ2-KA2 Test Element Specification and Irradiation Data

a) Calculated

b) Fission Power and γ –heat

3.2.4. AVR Irradiation

The spheres which were irradiated in the AVR are of GLE-3 respectively GLE-4 type. Since the exact route through the reactor is not known, no exact irradiation history could be calculated. The best estimation is given in Table VIII.

Parameter	Fuel element designation			
	AVR 74/16	AVR 74/18	AVR 73/22	AVR 87/06
Burn-up (% FIMA)	3.13	4.62	3.20	2.88
Neutron flux $10^{25} \text{ m}^{-2} [> 0.1 \text{ MeV}]$	0.5	0.8	0.3	0.3
Max centre temperature ^(a)	1100	1100	1100	1100
Max power output ^(a,b)	2.5	2.5	2.5	2.5

Parameter	Fuel element designation			
	AVR 74/16	AVR 74/18	AVR 73/22	AVR 87/06
Burn-up (% FIMA)	3.13	4.62	3.20	2.88
Neutron flux $10^{25} \text{ m}^{-2} [> 0.1 \text{ MeV}]$	0.5	0.8	0.3	0.3
Max centre temperature ^(a)	1100	1100	1100	1100
Max power output ^(a,b)	2.5	2.5	2.5	2.5

Table VIII: AVR Spheres Specification and Irradiation Data

a) Estimated

b) Fission Power and γ –heat

4. Results

The results of the measurement of the ^{137}Cs activity and the calculated burn-up, are shown in Table IX. The results are based on a 30 min measurement, considered to be enough to obtain reliable quantitative results. To confirm this assumption, in three cases extended measurements were performed and the measurements of 4 pebbles were repeated (in bold in Table 9). As can be seen from Table 9, the reproducibility of the measurements was better than 0.2% FIMA.

Fuel Element	^{137}Cs (Bq)	BU-calc. (Eq. 5)	BU-code (Korigen)	BU-FzJ (Inventar)
HFR-K6/1	8.36E+10	7.69%	8.10%	7.20%
HFR-K6/2	1.05E+11	9.71%	10.34%	9.30%
HFR-K6/3	1.00E+11	9.25%	9.82%	9.70%
HFR-K6/3	1.06E+11	9.80%	10.44%	9.70%
HFR-K6/4	9.45E+10	8.70%	9.21%	9.20%

Fuel Element	^{137}Cs (Bq)	BU-calc. (Eq. 5)	BU-code (Korigen)	BU-FzJ (Inventar)
HFR-K5/1	7.53E+10	6.93%	7.26%	6.70%
HFR-K5/2	9.47E+10	8.72%	9.24%	8.80%
HFR-K5/2	9.57E+10	8.81%	9.34%	8.80%
HFR-K5/3	1.04E+11	9.55%	10.16%	9.10%
HFR-K5/4	1.01E+11	9.26%	9.84%	8.70%
FRJ2-KA2/1	1.24E+10	1.77%	1.82%	1.84%
FRJ2-KA2/2	1.37E+10	1.96%	2.02%	2.00%
FRJ2KA2/2	1.27E+10	1.81%	1.87%	2.00%
FRJ2-KA2/3	1.33E+10	1.91%	1.97%	2.02%
AVR-74/16	3.69E+10	3.17%	3.29%	3.20%
AVR-74/16	3.58E+10	3.07%	3.19%	3.20%
AVR-74/18	5.44E+10	4.67%	4.90%	4.80%
AVR-73/22	2.26E+10	3.24%	3.36%	3.40%
AVR-73/22	2.24E+10	3.20%	3.33%	3.40%
AVR-87/06	2.01E+10	2.88%	2.98%	3.51%
AVR-87/07	2.17E+10	3.10%	3.22%	3.53%
AVR-87/07	2.16E+10	3.09%	3.21%	3.53%
AVR-87/08	2.14E+10	3.07%	3.18%	3.53%
AVR-87/08	2.22E+10	3.17%	3.29%	3.53%
AVR-87/09	2.17E+10	3.10%	3.22%	3.56%
AVR-87/10	2.09E+10	3.00%	3.11%	3.51%

Table IX: ^{137}Cs Inventory and Burn-up of the Fuel Elements (repeated measurements in bold)

Conclusion

A method for the quantitative determination of the burn-up of HTR spherical fuel elements has been developed and successfully tested on 19 irradiated HTR-spherical fuel elements. The procedure is based on the measurement of the ^{137}Cs gamma activity, using a conical collimator, which exploits the pinhole camera principle to determine the activity of a single fuel element. The results were compared and found in good agreement with previous results obtained in FzJ and code calculations. Additional work is underway to ascertain the accuracy of the measurements performed: From one of the spherical fuel elements, a representative sample will be drilled, pulverised and, subsequently dissolved to perform quantitative burnup determination on the basis of the mass spectrometric determination of ^{148}Nd .

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ANNEX A

CALCULATION OF THE GAMMA ABSORPTION IN THE FUEL ELEMENT

The gamma rays are already absorbed in the interior of the spherical fuel elements in an amount that depends on material and geometry of the pebble. This phenomenon is known as self-absorption and occurs in the fuel as well as in the graphite matrix. The spherical fuel elements consist of an inner graphite matrix containing the fuel coated particles, generally 50 mm in diameter, which is surrounded by an outer pure graphite layer, about 5 mm in thickness.

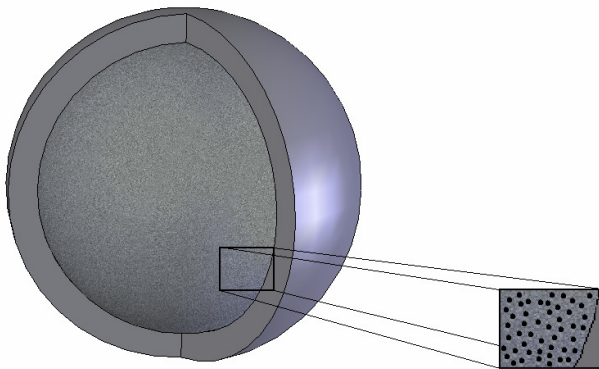


Figure A-1a: Spherical Fuel Element for HTR

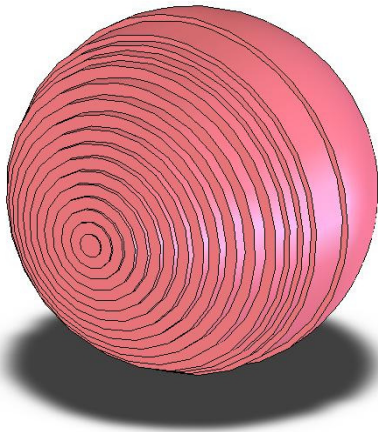


Figure A-1b: Decomposition of a Sphere in Layers 3D

As schematically shown in Fig. A-1a, several thousand multilayered coated particles (from 6000 to 30000, depending on the design) are imbedded in the graphite matrix. The diameter of these coated particles is about 1 mm so the inner fuel zone can be viewed, in good approximation, as an almost

homogenous fuel/graphite mixed area. Hence, the ^{137}Cs activity per unit volume, a , amounts to:

$$a = \frac{3 \cdot A_{\text{Cs-137}}}{4 \cdot \pi \cdot r_i^3}$$

Where r_i is the radius of the fuel zone and $A_{\text{Cs-137}}$ is the total activity in the pebble.

To calculate the absorption factor the total integral over the volume has to be solved. Due to the fact that this is not possible using simple mathematical methods, the fuel zone was divided in many differential concentrically arranged cylinders, as shown in Fig. A-1b. Hence, the attenuation factor for each cylinder can now be calculated through integration over their length and, finally, the total attenuation factor, f_{pebble} , can be calculated by integration over all cylinders:

$$f_{\text{pebble}} = \frac{3}{4\pi r_i^3 a}$$

$$\int_0^{\frac{1}{2}L(y)} \int_{\frac{1}{2}L(y)}^{\frac{1}{2}L(y)} 2\pi \cdot a \cdot y \cdot f_{\text{self}}(x, y) \cdot f_{\text{dist}}(x, y) \cdot f_{\text{layer}}(x, y) \cdot dx \cdot dy$$

Where the variable y represents the particular radius of a concentric cylinder.

Using this approximation, errors arise from the distance between the radiation source in the pebble and the detector by neglecting the aperture angle of the collimator. The error amounts to less than 0.01% for a point source and to less than 0.03%, for a whole pebble.

Using polar coordinates for simplification, the attenuation factor for the fuel zone can be calculated with the attenuation coefficient, μ_B , at 661.61 keV.

$$f_{\text{self}} = \int_{\frac{L(\varphi)}{2}}^{\frac{L(\varphi)}{2}} e^{-\mu_B \cdot (0,5 \cdot L(\varphi) - x)} dx$$

Where μ_B can be written as a combined value for uranium dioxide and graphite:

$$\begin{aligned}\mu_B &= \mu_G v_G + \mu_{UO_2} v_{UO_2} \\ &= \mu_G \frac{\rho_B}{\rho_G} w_G + \mu_{UO_2} \frac{\rho_B}{\rho_{UO_2}} w_{UO_2}\end{aligned}$$

μ_G, μ_{UO_2} : attenuation coefficients

ρ_{UO_2}, ρ_G : densities

v_G, v_{UO_2} : volume fractions

w_G, w_{UO_2} : mass fractions

$L(\varphi)$ is the length of the differential cylinder element:

$$L(\varphi) = 2 \cdot r_i \cdot \cos(\varphi)$$

The influence of the graphite layer has also to be included in the total attenuation factor:

$$f_{layer} = e^{(-\mu_G \cdot s(\varphi))}$$

The variable $s(\varphi)$ indicates the length of the graphite layer, which has to be traversed by the radiation. From simple geometric considerations:

$$s(\varphi) = \sqrt{r_a^2 - r_i^2 \cdot \sin^2(\varphi)} - r_i \cdot \cos(\varphi)$$

Finally, the total integral for the attenuation factor is:

$$\begin{aligned}f_{pebble} &= \int_0^{\pi/2} \int_{-r_i \cos(\varphi)}^{r_i \cos(\varphi)} \left(\frac{3\pi \cdot a \cdot r_i^2 \cdot \sin(\varphi) \cdot \cos(\varphi)}{2\pi \cdot a \cdot r_i^3} \right) \\ &\cdot e^{(-\mu_G (\sqrt{r_a^2 - r_i^2 \cdot \sin^2(\varphi)} - r_i \cdot \cos(\varphi)))} \\ &\cdot \left(\frac{e^{(-\mu_B \cdot (r_i \cdot \cos(\varphi) - x))}}{4\pi \cdot (f_{BE,K} - x)^2} \right) dx d\varphi\end{aligned}$$

The solution of this integral could be performed analytically with the program Maple[®].

In addition to the correction factors for the fuel elements, the analogous correction factor for the calibration standard described in Section 2.3.1 has to be calculated. In this case, only the absorption in graphite has to be considered. Hence:

$$f_{st} = \frac{e^{-\mu_G \cdot s}}{4 \cdot \pi \cdot (f_{FE,C})^2}$$

μ_G = mass attenuation factor

$f_{FE,C}$ = distance between FE and collimator

s = distance from the inner to the outer surface of the FE

Now, the final correction factor can be obtained by dividing the attenuation factor for the fuel element by the same factor for the standard:

$$f_{corr} = \frac{f_{pebble}}{f_{st}}$$

In the diagram shown in Fig. A-2, the relation between the heavy metal loading of the spheres and the correction factor is given. The values are valid for gamma emission at 661.61 keV and a fuel zone radius of 25 mm. As can be seen, all correction factors are near one. The reason for that can be found in the similar geometry and material of the pebbles and the used standard.

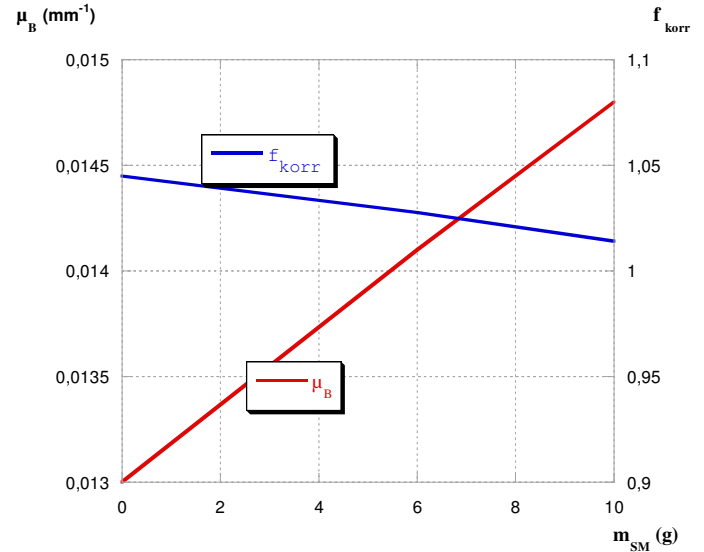


Figure A-2: Correction Factor as Function of Heavy Metal Loading

The ¹³⁷Cs activity can now be obtained from the experimental data, by means of the following equation:

$$A_{Cs-137} = \frac{I_m}{f_{corr} \cdot k_{PK} \cdot t_m}$$

k_{PK} = proportional constant, f_{corr} = correction factor

t_m = measurement time, I_m = counted impulses