DEVELOPMENT OF A COLLIMATOR FOR GAMMA MEASUREMENT OF SPHERICAL FUEL ELEMENTS

M. Betti*, W.Kühnlein° and E.H.Toscano*

- *European Commision, Joint Research Centre, Institute for Transuranium Elements, Postfach 23 40, 76125 Karlsruhe, Germany*
  ° *Forschungsanlage Jülich GmbH, Jülich, Germany*

Abstract

In the hot cell laboratory of the Institute for Transuranium Elements, the burnup of spherical fuel elements for High Temperature Reactors, has to be determined through gamma-spectrometric measurements by measuring the ^{137}Cs activity created during irradiation. To this goal, a specially designed collimator will be positioned, in a hole pierced through the hot cell wall, in order to reduce the intensity of the gamma beam to a level compatible with the gamma detector electronics. The collimator has been designed on the basis of the pinhole camera principle, taking into account the special geometry of the fuel elements and allows the imaging of the whole active zone of the spherical fuel elements on the operational zone of the detector. In addition, to quantify the burnup, an accurate calibration of the detector is needed. The calibration will be performed using a solution of irradiated UO_2 (70 GWd/t[HM]), which has been carefully analysed by wet chemistry. To this aim, a graphite container, filled with a precisely determined amount of this solution, is placed in the centre of a graphite sphere having the same dimension as the fuel elements to be measured to take into account the gamma absorption of the graphite. This sphere will be used to calibrate the efficiency of the gamma detector. In the present paper the design of the collimator and the sample holder and the calibration procedure, including the wet chemistry analysis procedure, are described.

INTRODUCTION

In the framework of a Share Cost Actions (SCA) of the European Commission, a European Project of Development of High Temperature Reactor (HTR) technology has been approved. The project includes developments in the fields of reactor physics, fuel technology, safety, material needs and 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, the post-irradiation examination (PIE) of irradiated fuels is needed to assess the quality of new concepts. Among the PIE-methods the verification of the release behaviour of fission gases (Xe, Kr) and solid fission products (Cs, Sr, Ag, etc) under accident conditions is of paramount importance. In the past, the so-called Cold Finger Apparatus (KÜFA) was developed in the Forschungszentrum Jülich (FzJ) to test HTR-fuel design and fabrication methods. Using this device, the fission product release from fuel spheres can be tested up to 1800 °C. In the framework of the SCA/HTR-technology, an up-dated version of the KÜFA has been installed in the hot cells of the Institute for Transuranium Elements (ITU).

Aiming to complete the data basis of FzJ, several fuel elements – transferred from the hot cells in FzJ to ITU – will be tested with the new KÜFA-installation. Previous to the testing, the calculated burnup of the pebbles has to be checked, by means of a gamma measurement, determining the amount of ^{137}Cs present in the samples. In addition, the fuel elements will be measured before and after the KÜFA-test.To these goals, a special collimator and an ad hoc calibration procedure have been developed. In the present paper the design of the collimator and the method and the standard fabrication used for the calibration of the gamma measuring system are described.

1. Determination of the burn-up on the basis of the ^{137}Cs activity

The amount of ^{137}Cs present in a fuel element depends on the fissionable isotope (^{233}U, ^{235}U or ^{239}Pu) used to produce energy. In what follows, we will concentrate on the case of ^{235}U. Part of the amount produced disappears through radioactive decay or neutron capture. Considering all the phenomena that my occur during irradiation, the number of ^{137}Cs isotopes produced, $X_{Cs}(t)$, by the fission of ^{235}U is given by:

$$x_{Cs}(t) = \frac{N\gamma T_f^u \varnothing(t)}{\lambda + \left(T_a^{Cs} - T_a^U\right)\varnothing(t)}\left[1 - A(t)\frac{T_a^U}{T_f^U} - e^{-\lambda t - T_a^{Cs}\varnothing(t)t}\right]$$

where:

N: number of ^{235}U-atoms at the beginning of life
ø: neutron flux
λ: decay constant of ^{237}Cs
t: irradiation time
T_a^{Cs} :   absorption cross-section of ^{137}Cs
T_a^U :   absorption cross-section of ^{235}U
T_f^U :   fission cross-section of ^{235}U
γ: fission yield of ^{137}Cs

A(t):   burn-up ratio: number of fissioned ^{235}U / number of initial ^{235}U

Without considering the term between brackets, the amount of produced Cs atoms varies linearly with the burn-up. The term between brackets has to be considered only in the case of high burn-up. In fact, taking into consideration the half-life of ^{137}Cs and the relative small absorption cross-section, 0.1 barn, this term becomes relevant and cannot be neglected only at high burn-up.

The fission yield of ^{137}Cs, for ^{235}U fission, is of 6.2%, and 6.6% for ^{235}U and ^{239}Pu. Then, the burn-up can be calculated from:

$$A[\%FIMA] = \frac{X_{Cs} \cdot C \cdot 100}{\gamma \cdot X_s}$$

Where:

C: Correction factor for long irradiation times
X_S: Heavy metal concentration at the beginning of the irradiation.

2. **Discussion of the gamma measuring principle**

The normal gamma detectors are limited in the amount of gamma-photons per time unit that can be detected. For this reason, depending on the activity of the specimen elements to be measured, a collimator has to be usually placed between the samples and the gamma detector.
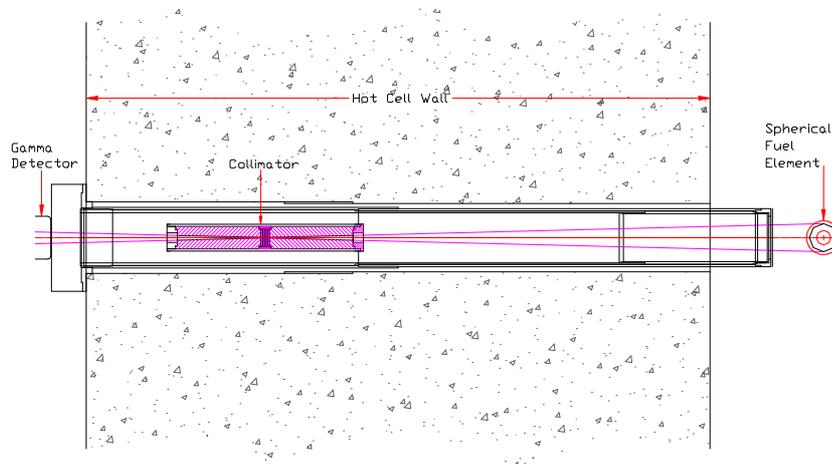

Fig. 1: Collimator based on the pinhole camera principle.

In **Fig. 1**, the measuring principle is shown. The design of the collimator is base on the pinhole camera principle in which all points of the object to be measured are recorded simultaneously by a stationary detector while the spherical fuel element is placed in a rotating table to take into account, during the measurement, for non-homogeneous distributions of the fuel in the active zone.
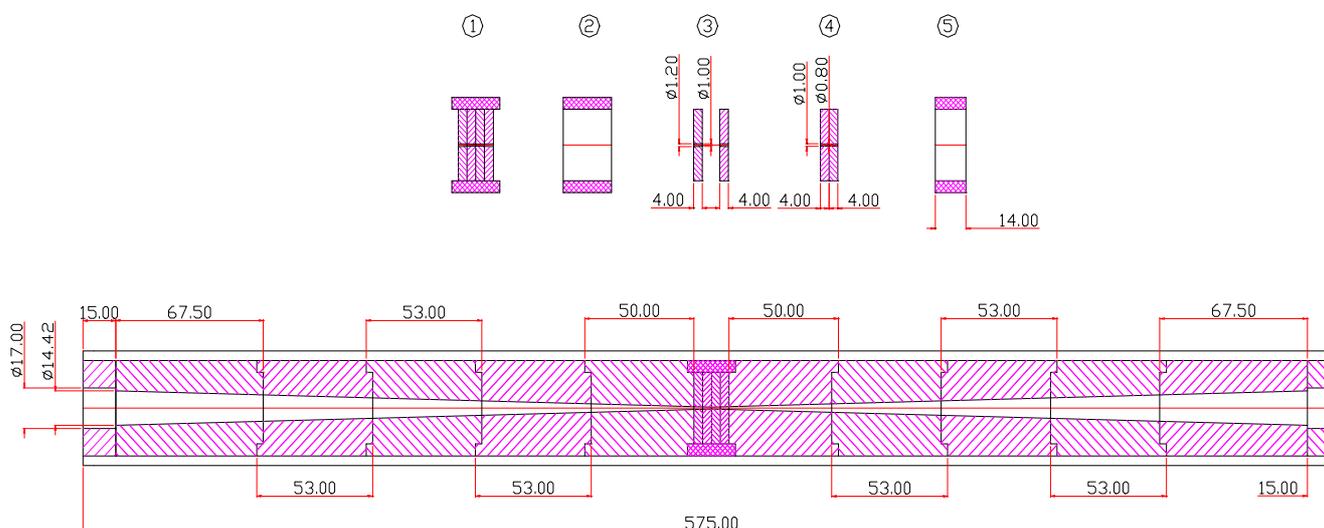
3. Description of the collimator

Fig.2: Design of the collimator.

In Fig. 2 the design of the collimator is shown. The material used to perform the collimator was a W-alloy, having a density of 19.5 g/cm^3. The solid angle was calculated taking into account the distances between the fuel element and the active zone of the gamma detector.
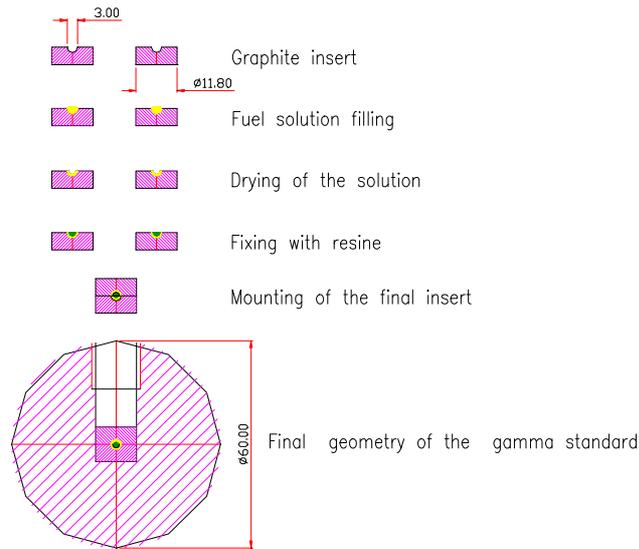
In the central part, a minimum orifice of 0.5 mm in diameter is foreseen. The collimator design allows for a variation of this diameter to permit the measurement of less active fuel elements.

4. Calibration procedure

About 1 pellet was cut from a fuel rod irradiated in a power reactor to a burnup of around 70 GWd/t[HM]. This sample was dissolved in a 6 molar solution at about 90°C. A small aliquot was transferred to a glove box for the HPLC separation process. The amount of the heavy metals, U and Pu, was determined using the TIMS-method.

The burnup of the sample was determined by the measuring the amount of ^{148}Nd in the sample. The determination of this isotope as well as the ^{137}Cs, and complete inventory of fission products was performed by ICP-MS previous on-line chromatographic separation. The sample solution (dissolved spent fuels) after convenient spiking and dilution are loaded on the loop of the injection valve of the chromatographic system. After injection in the separation column, the use of 0.1M oxalic acid/0.19M LiOH as eluent allows the separation of Nd from U and Pu and from the other fission products. The measurements were performed with an Elan 5000 Inductively Coupled Plasma Mass Spectrometer (Perkin Elmer SCIEX) modified for installation in a glove-box and coupled with a 4500l high-pressure chromatographic pump (Dionex) was used. The guard column and the analytical

SCHEMATIC DIAGRAM OF THE STANDAR PRODUCTION

3.00

Graphite insert

ø11.80

Fuel solution filling

Drying of the solution

Fixing with resine

Mounting of the final insert

ø60.00

Final geometry of the gamma standard

column were placed inside the glove-box. The effluent from the chromatographic column was directly connected to the nebuliser of the mass spectrometer. The injection volume was typically 250µl [3-7].

CONCLUSIONS

A method for the determination of the burnup on the basis of gamma measurements of HTR-spherical fuel elements has been described. To this goal, a collimator, based on the pinhole camera principle, has been installed in hole drilled in the hot cell wall. A calibration procedure, based on a well-characterised solution of a fuel irradiated in a power reactor has been discussed. On the basis of these measurements the burnup can be evaluated using standard equations.

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