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Presented at the IAEA Technical Committee on Advanced Post-Irradiation Techniques for Water Reactor Fuel.
Dimitrovgrad (Russian Federation), May 14-18, 2001.

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GAMMA-SPECTROMETRIC DETERMINATION OF THE FISSION POWER OF FUEL RODS

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Abstract

The fission power constitutes essential information for the evaluation of the behaviour of fuel rods under irradiation. At SCK•CEN several methods are applied to determine this fission power: the in-pile thermal balance method and the out-of-pile γ-spectrometric, fluence-dosimetry and destructive-radiochemical methods.

The present paper discusses the γ-spectrometric determination of both burn-up and linear fission power. It relies on the determination of the concentration within the fuel of long-living (^{137}Cs – burn-up) or short-living fission products (^{140}Ba-La – linear fission power). Their measured activities are converted into absolute concentrations by calibrating the measurement system with a reference $^{152-154}$Eu source. The measurement methodology and data processing are described – the uncertainty on the final results is estimated to amount to ± 6 %.

Comparison with the other fission power determination methods reveals that the γ-spectrometric method is the best out-of-pile method next to the destructive-radiochemical method, with which excellent agreement within ± 2 % is observed.

1. INTRODUCTION

The fission power which fuel rods experience during their irradiation stage(s) is a basic vital quantity both to judge on the fuel rod behaviour and their specification in terms of safeguards. Experimental post-irradiation quantitative determination of this fission power is performed at the SCK•CEN hot laboratory LHMA (Laboratory for High and Medium level Activity) by high resolution γ-spectroscopy. This measurement technique has the advantage

- to be non-destructive, hence to be
  - reversible;
  - applicable at intermediate irradiation stages;
- to be fast;
- to generate at the same time
  - reliable quantitative data on total fuel rod fission power;
  - the fission power profile along the rod axis;
  - the axial profile of individual radionuclides, hence allowing to recognize fission product migration.

The present paper focuses on the methodology applied to determine both the rod burn-up (based on ^{137}Cs activity analysis) and the linear power (based on ^{140}Ba-La activity analysis). The accuracy of the method will be assessed on the basis of error analysis and through cross comparison of results obtained with other techniques.

2. GAMMA-SPECTROMETRY METHODOLOGY

The γ-spectrometric determination of the fission power relies on the sequential measurement, within a fixed collimated geometry, of the fuel rod under examination and a calibration source. From these measurements the absolute concentration of fission products inside the fuel, and hence the fission power, can be obtained.

2.1. Measurements

The fuel rod or calibration source is positioned in a fixed geometry in front of a collimator system. Both the fuel rod and the calibration source are extended volume sources - only a very small part is visible to the detector. The information present in the measured spectrum is

specific for a well-defined portion of the source, i.e. A_{ix} [counts·s^{-1}·mm^{-1}] is the net measured count rate for γ-peak i at position x within a cross section width Δx equal to the collimator system width (different collimators are available, 0.5, 1, 2 and 4 mm, in order to cope with different activity levels, which depend on fuel irradiation history and cooling time).
Multiple measurements (spectra) are required to obtain an accurate representation of the axial activity distribution along the source axis. The number of measurements and their position is chosen to yield a measured axial activity profile as close as possible to the actual activity profile. In regions where the activity gradients are low one measurement every ~ cm (each up to each third fuel pellet in case of fuel rods) is sufficient. Regions with large activity gradients are measured at intervals equal to the size of the collimator system. The presence and positions of gradients are determined by performing a gross-γ scanning prior to γ-spectroscopy. Typically ~ 20 measurements are performed on the calibration source, ~ 100 measurements on experimental fuel rods ~ 1 m in length and ~ 200 measurements on industrial rods ~ 4 m in length.

2.2. Fission indicator activity calculation

As a large number of spectra are measured, a high degree of automation of the data processing is implemented in order to take full advantage of the method.

2.2.1. Spectrum analysis

The fully automated accurate γ-ray spectrum analysis program scans every spectrum for the occurrence of peaks and generates the location and net peak area of every peak. The net peak count rate is calculated as:

$$A_{ix} = \frac{N_{ix}}{t_{el}} \qquad (1)$$

with N_{ix} = net peak area of peak i at axial position x [counts·mm^{-1}]
Running the spectrum analysis program on a benchmark spectrum validates the peak identification, location and net peak area determination. This benchmark spectrum contains ~ 1000 γ-peaks with exact known position, width and area. For peak to background ratios > 1 the peak position is determined within ± 0.1 channel width and the net peak area within ± 2 %.

t_{el} = elapsed life time [s]
The loss of pulse correction is determined experimentally. A ^{152}Eu source, having a reasonable number of γ-lines in the energy range 100 keV - 1.4 MeV, is measured at several known distances from the detector in order to have activities ranging from very small values – typically a few 100 counts·s^{-1} – to the maximum possible – viz. ~ 20,000 counts·s^{-1}. The count rate errors due to loss of pulses are found to remain below 2 % at the highest total incoming count rate of 20,000 counts·s^{-1} (at this count rate the measurement system dead time is approximately 60 %). By using appropriate collimators, the count rate can be kept well below this value, thus giving rise to limited and accurately known loss of pulse corrections.
The elapsed lifetime is known to within ± 0.02 %.

A_{ix} = the net measured count rate for γ-peak i in the spectrum at axial position x [counts·s^{-1}·mm^{-1}]
with a global accuracy of ± 2 %

2.2.2. *Efficiency calibration*

The certified activity of the calibration source equals the total activity present inside the source. Hence, the efficiency of the measurement set-up is calculated from this total source activity, i.e. from the total area of the measured activity profile:

$$\varepsilon_i = \frac{1}{C_D P_\gamma T C} \times \frac{1}{2} \sum_n \left[A_{ix_{n+1}} + A_{ix_n} \right] \left[x_{n+1} - x_n \right] \qquad (2)$$

with C_D = decay correction

The decay correction corrects for isotope decay between reference date and measurement date. It depends on the half-life (known to within ± 1.2 % for ^{154}Eu and to within ± 0.3 % for ^{152}Eu)[1] and decay time (known very accurately). Hence the decay correction is done within ± 1 %.

P_γ = γ emission probability [γ·des^{-1}]

It relates the number of γ-photons to the number of atoms responsible for it. Values of the number of γ-photons emitted per desintegration are taken from nuclear data libraries [2],[3] and are known to within ± 1 %.

T = transparency correction

The transparency corrects for the attenuation of γ-photons emitted inside the source travelling in the direction of the detector. It is calculated by a code ST (Source Transparency) that determines the non-interaction escape probability for mono-energetic γ- or X-ray photons originating from arbitrary shaped geometric 2D objects. Total cross-sections for each compound are calculated on the basis of its composition and element cross-sections as taken from ref. [4]. Comparison of ST results with experimental measurements (by use of a calibrated source) proves the precision to be within ± 2 %.

C = calibration source certified value [des·s^{-1}]

For the Eu4 standard source the certified value amounts to 45.7 ± 0.5 (± 1 %) GBq ^{152}Eu and 6.46 ± 0.10 (± 1.5 %) GBq ^{154}Eu. [5]

ε_i = measurement efficiency at energy i [counts·γ^{-1}]

Taking into account the precision on the individual constituents, the overall accuracy amounts to ± 4.5 %.

The efficiency ε_i versus energy i is fitted by a polynomial on a log-log scale. The obtained polynomial parameters and their covariance matrix are used to calculate the efficiency and its precision at any energy value. This precision is found to range from 2 to 5 %, corresponding very well to the one estimated above from the precision on the individual constituents (± 4.5 %).

2.2.3. *Fission indicator activity calculation*

Knowing the efficiency ε_i of the measurement system, the activity of a specific isotope (^{137}Cs as long life BU indicator or ^{140}Ba-La as a short life linear power indicator) can be calculated from the net measured count rate of its γ-peak i as obtained from the spectrum analysis:

$$\alpha_i = \frac{1}{C_D P_\gamma T \varepsilon_i} \times \frac{1}{2} \sum_n \left[A_{ix_{n+1}} + A_{ix_n} \right] \left[x_{n+1} - x_n \right] \qquad (3)$$

with C_D = decay correction

With the half-life of ^{137}Cs known to within ± 0.7 % and of ^{140}Ba-La to within ± 0.4 % (ref. [1]), the precision of the decay correction amounts to ± 1 %. As reference date the end of irradiation (EOI) date is taken.

P_γ = γ emission probability [γ·des^{-1}]
As for $^{152\text{-}154}$Eu within the calibration source, the number of γ-photons emitted per desintegration for ^{137}Cs and ^{140}Ba-La is known to within ± 1 %.

T = transparency correction
The same ST code is used to calculate the attenuation of γ-photons emitted by ^{137}Cs inside the fuel rod on their way to the detector. In addition to the intrinsic precision of the ST code (± 2 %), the calculation of the source transparency for fuel samples might suffer from a non-uniform radial ^{137}Cs distribution. Depending on the irradiation and especially the temperature history of the fuel rod, the radial BU is non-uniform (viz. higher burn-up at pellet rim) and ^{137}Cs might have migrated (towards the pellet periphery). In order to estimate the error introduced when assuming a flat radial activity distribution, the energy dependence of the source transparency for a typical fuel rod has been calculated for different radial activity distributions. For the ^{137}Cs (daughter ^{137m}Ba) γ-peak at 661.6 keV, the difference of the transparency for a flat radial distribution and the extreme maximum transparency case with all activity concentrated in an annular ring of 0.5 mm width at the pellet surface amounts to 7 %.

ε_i = measurement efficiency at energy i [counts·γ^{-1}]
with accuracy ± 4.5 % - eq. (2)

α_i = total activity within a fuel rod of a specific isotope, measured on the basis of γ-peak i, at reference date [Bq]
accuracy ± 5.7 % in case of uniform radial activity distribution

2.3. *Burn-up calculation*

Foregoing activity α_i still has to be corrected for the decay of the concerned radioactive nuclide during the irradiation period.
As the time-integrated total amount of fissions having occurred during the total irradiation period is the measure for the burn-up of a fuel rod, the irradiation-history-corrected activity in case of urn-up determination is calculated as:

$$A_{i_{BU}} = \alpha_i \times \frac{\lambda_i \times \sum_j P_j t_j}{\sum_j P_j \times (1 - e^{-\lambda_i t_j}) \times e^{-\lambda_i t_{r_j}}} \qquad (4)$$

with P_j = relative measure of fuel rod power during irradiation time t_j
The thermal reactor power [MW_{th}] is mostly used as representative basis for relative power history assessment.

t_j = irradiation time at constant power P_j [s]

λ_i = decay constant for nuclide i [s^{-1}]

t_{r_j} = elapsed time between end of irradiation time t_j and reference date (EOI) [s]

$A_{i_{BU}}$ = total equivalent activity of a specific fission product (e.g. ^{137}Cs as long-living BU indicator) within a fuel rod resulting from the time-integrated fissions within the fuel [Bq]
The correction factor is a measure of the ratio between the total amount of fission product formed (numerator = equivalent perfect integrator as if no decay had happened during the irradiation period) and the amount present at the reference date (EOI) as due to the formation/decay during the irradiation period (denominator).

The irradiation history correction is invariant with respect to absolute power scaling. In order to estimate the influence of the uncertainties in relative fuel rod power assessment, the relative

range of irradiation history corrections per unit of relative power variation has been calculated. The precision on the irradiation history correction is found to range, for a ± 10 % variation on the relative power levels, from ± 0.1 % to ± 5 % depending on the isotope half life. By selecting appropriate isotopes accurate irradiation history corrections well below ± 1 % can be obtained, hence giving rise to an accuracy of $A_{i_{BU}}$ within ± 5.8 %.

In order to calculate the burn-up from the above activity $A_{i_{BU}}$, still two fuel specific factors need to be calculated:

- the average yield of fission product atoms is calculated as a weighed average over all contributing fissile targets:

$$Y_i = \frac{\sum_j n_j \sigma_j Y_{i_j}}{\sum_j n_j \sigma_j} \tag{5}$$

with
- n_j = fraction of atoms of fissile target j as obtained from the initial fuel composition – mostly known with an accuracy better than ± 1 %
- σ_j = thermal fission cross-section for fissile target j [b] as obtained from ref. [2], accuracy ± 0.5 %
- Y_{i_j} = yield of fission product i for fissile target j [at/fission] as obtained from ref. [2], accuracy ± 0.5 %
- Y_i = average yield for fission product i [at/fission] within ± 1 %

- the average energy release per fission is also calculated as a weighed average over all contributing fissile targets:

$$E_f = \frac{\sum_j n_j \sigma_j E_j}{\sum_j n_j \sigma_j} \tag{6}$$

with
- n_j = fraction of atoms of fissile target j as obtained from the initial fuel composition – mostly known with an accuracy better than ± 1 %
- σ_j = thermal fission cross-section for fissile target j [b] as obtained from ref. [2] , accuracy ± 0.5 %
- E_j = the total energy released per fission for fissile target j [MeV/fission] as obtained from ref. [2] , accuracy ± 0.5 %
- E_f = average energy release per fission [MeV/fission] within ± 1 %

The burn-up of the fuel rod (i = ^{137}Cs) is then obtained as:

$$BU_r = \frac{A_{i_{BU}}}{\lambda_i} \times \frac{1.854 \times 10^{-24} \times E_f}{Y_i} \times \frac{1}{W_{HM_r}} \tag{7}$$

with
- $A_{i_{BU}}$ = total equivalent activity of a specific fission product (e.g. ^{137}Cs as long-living BU indicator) within a fuel rod resulting from the time-integrated fissions within the fuel – eq. (4) [Bq]

λ_i = decay constant for nuclide i [s^{-1}]
known within ± 0.7 % (^{137}Cs)

E_f = average energy release per fission – eq. (6) [MeV/fission]
with 1.854×10^{-24} being the conversion from MeV to MWd

Y_i = average yield for fission product i – eq. (5) [at/fission]

W_{HM_r} = heavy metal weight within the fuel rod [t]
as obtained form the fuel rod specifications – mostly known with an accuracy better than ± 1 %

BU_r = fuel rod burn-up [$MWd \cdot t_{HM}^{-1}$]
with a global accuracy of ± 6 %

2.4. *Linear power calculation*

As the fission rate is the measure for the linear power level of a fuel rod, the irradiation-history-corrected activity in case of linear power determination is calculated as:

$$A_{i_{LP}} = \alpha_i \times \frac{P_{ref}}{\sum_j F_d(F_a(P_j, t_j), t_{r_j})} \tag{8}$$

with P_j = relative measure of fuel rod power during irradiation time t_j
The thermal reactor power [MW_{th}] is mostly used as representative basis for relative power history assessment.

P_{ref} = reference power level
The power level P_j [MW_{th}] most prevailing during the irradiation history is mostly taken as reference

t_j = irradiation time at constant power P_j [s]

t_{r_j} = elapsed time between end of irradiation time t_j and reference date (EOI) [s]

$F_a(P_j, t_j)$ calculation of fission product activity build-up after irradiation at power P_j during a time t_j
If a precursor isotope exists that has any effect on the time evolution of the treated isotope, the more complicated mother-daughter formalism is used.

$F_d(F_a, t_{r_j})$ decay correction
If a precursor isotope exists that has any effect on the time evolution of the treated isotope, the more complicated mother-daughter formalism is used.

$A_{i_{LP}}$ = total equivalent saturation activity at the reference power of a specific fission product (e.g. ^{140}Ba-La as short-life linear power level indicator) within a fuel rod [Bq]
The correction factor converts the activity of a fission power indicator present at the reference date (EOI) taking into account the power history induced build-up/decay (denominator) to the saturation activity corresponding to the chosen reference power (numerator).

As in the case of the burn-up calculation, the irradiation history correction is invariant with respect to absolute power scaling and the accuracy of $A_{i_{LP}}$ amounts to ± 5.8 %.

The linear power level of the fuel rod (i = ^{140}Ba-La) is then obtained as:

$$LP_r = A_{i_{LP}} \times \frac{1.602 \times 10^{-13} \times E_f}{Y_i} \times \frac{1}{L_f} \tag{9}$$

with $A_{i_{LP}}$ = total equivalent saturation activity at the reference power of a specific fission product (e.g. ^{140}Ba-La as short-life linear power level indicator) within a fuel rod – eq. (8) [Bq]

E_f = average energy release per fission – eq. (6) [MeV/fission]
with 1.602×10^{-13} being the conversion from MeV to J [W·s]

Y_i = average yield for fission product i – eq. (5) [at/fission]

L_f = length of the fuel stack witching the fuel rod [cm]
as obtained form the fuel rod specifications and/or the gross-γ scan, with an accuracy < ± 0.1 %

LP_r = linear power level of the fuel rod at reactor power P_{ref} [W·cm^{-1}]
with a global accuracy of ± 6 %

3. RESULTS AND COMPARISON WITH OTHER METHODS

At the Belgian high flux materials testing reactor BR2 (Belgian Reactor 2), several methods are applied to determine the fission power: the thermal balance method, the fluence-dosimetry method, the destructive radiochemical method and the γ-spectrometry method.[6],[7]
The various methods are compared for two typical irradiations in BR2:

- the irradiation of a fuel bundle of 9 rods within the CALLISTO loop (Capability of LWR Irradiation in Steady State and Transient Operation Conditions – a closed loop within BR2 in which PWR conditions of water chemistry, pressure and temperature are simulated);[8]
- the irradiation in the PWC-CCD device (Pressurized Water Capsule – Cycling and Control Device) - an instrumented irradiation rig filled with stagnant water, which can contain a single fuel rod to be tested under steady-state and transient conditions.[9]

3.1. Other methods for fission power determination

3.1.1. The thermal balance method

The thermal balance method relies on the measurement of the total heat produced during irradiation. Both irradiation devices are equipped with appropriate instrumentation:

- differential Cr-Al thermocouples to measure the temperature gradient between the outlet and the inlet of the cooling water circuit of the device;
- a diaphragm connected to an accurate differential manometer to measure the coolant flow rate in the irradiation device.

The uncertainty of fuel rod power determination by the thermal balance method at BR2 amounts to ± 5 %.[6]

3.1.2. The fluence-dosimetry method

In this method, the fuel rod is irradiated together with fluence dosimeters, located as close as possible to the rod. Two types of dosimeters are used: Co (with response in the thermal and epithermal neutron energy range) and Fe (with response in the fast neutron energy range). The irradiation device is modelled in a multigroup neutron calculation. The calculated dosimeter response, at the dosimeter location, is then normalized to the measured dosimeter response, which normalizes the whole calculated neutron flux chart. The calculated fission power in the fuel rod is thus obtained in absolute units.

3.1.3. The destructive radiochemical method

The radiochemical method involves fuel dissolution, chemical separation steps, and radiometric (α- and γ-spectrometry) and TIMS (Thermal Ionisation Mass Spectrometry) measurement of selected fission indicators (Nd, Ce and Cs isotopes) and U, Pu and transuranium isotopes (Am, Cm).[10] The procedure using ^{148}Nd as fission indicator is widely

accepted as the reference method for burn-up measurements, since it has been qualified as ASTM E321-69. The destructive radiochemical measurement method allows the determination of the fuel rod burn-up (i.e. of the time-integrated fission power) to within ± 2.5 %.

3.2. *Comparison of results from the various methods for fission power determination*

The peak burn-up values obtained for fuel rods irradiated in CALLISTO are compiled in table I, while the peak linear power values obtained for fuel rods being transient tested in the PWC-CCD device are compiled in table II.

Table I: Peak burn-up values obtained for fuel rods irradiated in the CALLISTO PWR simulator in BR2, according to the various methods.

Fuel Rod	Peak burn-up [GWd·t_M^{-1}]		
	Thermal Balance	γ-Spectrometry	Radiochemistry
UN1 (UO_2)	23.6 ⟨−8.4⟩[a]	25.8	25.9 ⟨+0.4⟩[a]
MN1 (MOX)	25.3 ⟨+5.3⟩	24.0	23.5 ⟨−2.1⟩
MN2 (MOX)	45.0 ⟨−1.8⟩	45.8	45.2 ⟨−1.3⟩
MN3 (MOX)	57.0 ⟨+4.4⟩	54.6	54.3 ⟨−0.5⟩

[a] The values indicated between ⟨⟩ are the percent differences with respect to the γ-spectrometry

Table II: Peak linear fission power values obtained for fuel rods transient tested in the PWC-CCD device in BR2, according to the various methods.

Fuel Rod	Peak linear fission power [W·cm^{-1}]		
	Thermal Balance	γ-Spectrometry	Fluence-dosimetry
UR1 (UO_2)	477.5 ⟨+1.1⟩[a]	472.4	479.4[b] ⟨+1.5⟩[a] 428.1[c] ⟨−9.4⟩
UR2 (UO_2)	476.1 ⟨+2.9⟩	462.9	386.7[b] ⟨−16.5⟩ 382.7[c] ⟨−17.3⟩
MR1 (MOX)	439.6 ⟨−4.2⟩	458.8	421.7[b] ⟨−8.1⟩ 385.7[c] ⟨−15.9⟩
MR2 (MOX)	468.3 ⟨+4.7⟩	447.2	- 398.5[c] ⟨−10.9⟩

[a] The values indicated between ⟨⟩ are the percent differences with respect to the γ-spectrometry
[b] These values are deduced from the Co dosimetry measurements
[c] These values are deduced from the Fe dosimetry measurements

One observes:

- for the peak burn-up values:
  - an excellent agreement between the γ-spectrometry and the radiochemistry results – this is also the case for other samples, not considered in the present paper;

  - a good agreement between the γ-spectrometry results and those obtained by the thermal balance method;
- for the peak linear fission power:
  - a good agreement between the γ-spectrometry results and those obtained by the thermal balance method;
  - the agreement between the γ-spectrometry (or thermal balance) and the fluence dosimetry results lies within 20 %, the agreement being better for the Co than for the Fe dosimeters – the fluence-dosimetry seems to systematically underestimate the fission power – this method still suffers from the cumulation of calculation, modelling and measurement errors and is still under refinement.[7]

Conclusions

Using appropriate fission indicators and applying appropriate calibration, γ-spectrometric post-irradiation measurements allow to determine the fission power, both in terms of burn-up and of linear fission power, of fuel rods within an estimated accuracy of ± 6 % as derived from the measurement/calculation formalism.
Application of the γ-spectrometry methodology to well characterized fuel irradiations within the BR2 materials testing reactor proves excellent agreement with the radiochemistry method (within ± 2 %), often considered as the reference determination, as well as a good agreement with the thermal balance method (within ± 8 %, which is reasonable with regard to the estimated precision of the individual methods).

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