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GAMMA-RAY SPECTROSCOPY ON IRRADIATED FUEL RODS

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ABSTRACT

The recording of gamma-ray spectra along an irradiated fuel rod allows the fission products to be qualitatively and quantitatively examined. Among all nondestructive examinations performed on irradiated fuel rods by gamma-ray spectroscopy, the most comprehensive one is the average burnup measurement, which is quantitative. Moreover, burnup measurements by means of gamma-ray spectroscopy are less time-consuming and waste-generating than burnup measurements by radiochemical, destructive methods. This work presents the theoretical foundations and experimental techniques necessary to measure, using nondestructive gamma-ray spectroscopy, the average burnup of irradiated fuel rods in a laboratory equipped with hot cells.

1. INTRODUCTION

Nondestructive gamma-ray spectroscopy on irradiated fuel rods, by means of qualitative and quantitative analysis of fission products, enables the burnup profile to be determined, the average burnup to be measured, the power distribution to be estimated and the fuel pellets stack within the rod to be accurately located. The basic experimental apparatus required to perform this nondestructive examination in a laboratory equipped with hot cells is shown schematically in Fig. 1.

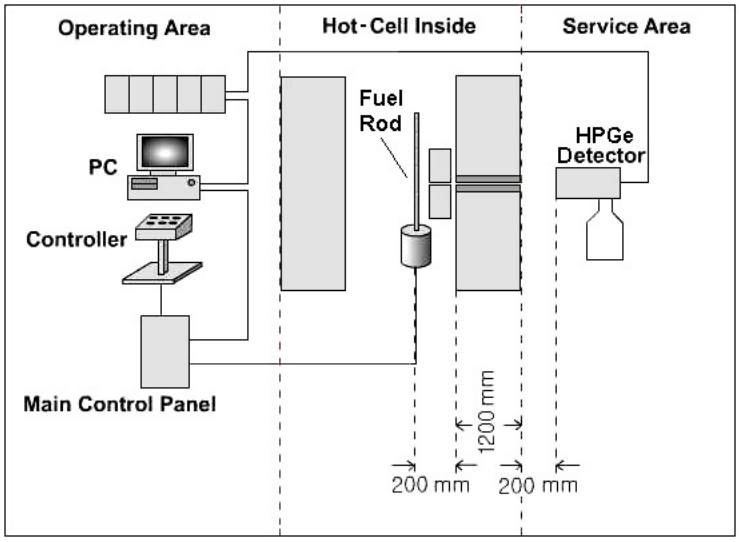

Figure 1. Basic experimental apparatus for gamma-ray spectroscopy on irradiated fuel rods.

The irradiated fuel rod to be studied moves in front of a HPGe detector. A collimation system positioned between the rod and the detector permits gamma-ray emissions from a restricted volume of the rod to be selected. Information coming from the detector is amplified, classed in terms of energy and analyzed.

Among all nondestructive examinations performed on irradiated fuel rods by gamma-ray spectroscopy, the most comprehensive one is the average burnup measurement, which is quantitative. In order to obtain the necessary information, complete gamma-ray spectra are accumulated as a function of the fuel rod active length. The net number of counts (net area) under the full-energy peaks is determined, giving a quantitative measurement of the amount of each radioactive fission product present at a specific location. These amounts can be related to the total activity of a given fission product, used as burnup monitor [1], which enables the determination of the total number of fission events and therefore the average burnup of the fuel rod.

However, such quantitative measurements require: *a*) previous energy and efficiency calibration of the experimental apparatus, using calibration sources or a standard fuel rod; *b*) a rigorous control of geometry and configuration of the system; *c*) accurate evaluation of all attenuation effects involved; *d*) detailed knowledge about the irradiation history of the fuel rod. Nevertheless, burnup measurements by means of nondestructive gamma-ray spectroscopy are less time-consuming and waste-generating than burnup measurements by radiochemical, destructive methods.

This work presents the theoretical foundations and experimental techniques necessary to measure, using nondestructive gamma-ray spectroscopy, the average burnup of irradiated fuel rods in a laboratory equipped with hot cells.

2. BURNUP MONITORS

The most suitable fission products for use as burnup monitor in gamma-ray spectroscopy measurements depend on the fuel rod history concerning irradiation and cooling periods [1], according to data presented in Table 1.

Table 1. Main properties of the fission products used as burnup monitors in gamma-ray spectroscopy measurements on irradiated fuel rods: fuel rod history, fission product used as standard burnup monitor, half-life $T_{1/2}$, energy E_γ and absolute emission intensity I_γ of the main emitted gamma-rays, fission yield for ^{235}U, ^{238}U and ^{239}Pu by thermal neutrons y_t and by fast neutrons y_r

Fuel rod history		Burnup monitor	$T_{1/2}$	E_γ (keV)	I_γ	^{235}U		^{238}U	^{239}Pu	
Irradiation	Cooling					y_t (%)	y_r (%)	y_r (%)	y_t (%)	y_r (%)
< 40 d	> 9 d	$^{140}Ba/^{140}La$	12.75 d	1596.5	1.099	6.30	6.13	5.98	5.56	5.29
< 200 d	> 40 d	^{95}Zr	64.03 d	724.2	0.437	6.49	6.37	5.13	4.89	4.66
				756.7	0.554					
≥ 1000 d	< 2 yr	$^{144}Ce/^{144}Pr$	284.9 d	2185.7	0.007	5.48	5.28	4.50	3.74	3.74
> 1800 d	≥ 2 yr	^{137}Cs	30.14 yr	661.6	0.851	6.21	6.16	5.97	6.62	6.49

Modern equipment, employing HPGe detectors coupled to Compton suppression systems for nuclear spectroscopy applications involving ultra-high input counting rates, enables the use of ^{137}Cs as burnup monitor regardless the fuel rod history.

3. FUEL ROD ACTIVITY

The total activity of a fuel rod due to a given burnup monitor is:

$$D = \pi \cdot r_0^2 \cdot L \cdot \overline{\rho} \tag{1}$$

where r_0 is the fuel pellets radius, L is the fuel rod active length and $\overline{\rho}$ is the average value of the specific activity in the fuel pellets.

The average value of the total number of counts registered by the HPGe detector (net area) per unit of time is obtained as a result of measurements performed at various locations along the fuel rod active length and is related to the average value of the specific activity in the fuel pellets by the expression:

$$\overline{Q} = \overline{\rho} \cdot v \cdot I_\gamma \cdot \varepsilon_\gamma \cdot k_1 \cdot k_2 \cdot k_3 \tag{2}$$

where v is the fraction of the fuel rod volume defined by the detection solid angle (function of geometry and configuration of the experimental apparatus), I_γ is the gamma-ray absolute emission intensity (number of gamma-rays with a given energy emitted per disintegration), ε_γ is the intrinsic efficiency of the HPGe detector for gamma-rays of a given energy, k_1 is the self-attenuation correction, k_2 is the correction due to gamma-rays attenuation after traversing the cladding and k_3 is the correction due to gamma-rays attenuation after traversing the air.

In order to take into account heterogeneities in the distribution of burnup monitor atoms in the fuel matrix, measurements along the fuel rod active length must be performed for angles of 0°, 45°, 90°, 135°, 180°, 225°, 270° and 315°. For each one of these angles, a measurements-set will be obtained, whose arithmetical mean constitutes a partial result. Finally, the uncertainty-weighted mean [2] of the eight partial results will be the average value $\overline{Q}$.

Combining both equations shown before, the total activity of a fuel rod due to a given burnup monitor can be rewritten as follows:

$$D = \frac{\pi \cdot r_0^2 \cdot L \cdot \overline{Q}}{v \cdot I_\gamma \cdot \varepsilon_\gamma \cdot k_1 \cdot k_2 \cdot k_3} \tag{3}$$

4. CORRECTIONS DUE TO ATTENUATION EFFECTS

Evaluation of effects caused by attenuation of gamma-rays is necessary to determinate the absolute gamma activity of irradiated fuel rods. Corrections due to attenuation effects need

two hypotheses to be calculated, both based on overall geometry and configuration of the experimental apparatus [1]:

· Once the detector is positioned far away from the point where the gamma-ray was emitted, the radiation is strongly directed perpendicularly to the fuel rod axis.

· Once the geometry and the configuration of the system remain unchanged, the detection solid angle is the same for every point of the fuel rod.

The first correction to be calculated arises from the attenuation of gamma-rays traversing the UO_2 fuel pellet where they are emitted. This effect is called self-attenuation and, considering a large cylindrical source, the self-attenuation correction is given by [3,4]:

$$k_1 = \frac{I_1(2\mu r_0) - L_1(2\mu r_0)}{\mu r_0} \tag{4}$$

where μ is the linear attenuation coefficient of UO_2 for a given gamma-ray energy, I_1 is the modified Bessel function of order 1 and L_1 is the modified Struve function of order 1. These functions are defined by means of the following convergent infinite series [5]:

$$I_1(x) = \left(\frac{x}{2}\right) \cdot \sum_{n=0}^{\infty} \frac{(x^2/4)^n}{n! \cdot \Gamma(n+2)}$$

$$L_1(x) = \left(\frac{x}{2}\right)^2 \cdot \sum_{n=0}^{\infty} \frac{(x/2)^{2n}}{\Gamma(n+3/2) \cdot \Gamma(n+5/2)} \tag{5}$$

where the gamma functions, being n an integer number, are given by the expressions [5]:

$$\Gamma(n+1) = n!$$

$$\Gamma(n+1/2) = \frac{(2n)!}{2^{2n} \cdot n!} \cdot \sqrt{\pi} \tag{6}$$

The second correction to be calculated arises from the attenuation of gamma-rays traversing the cladding of the fuel rod. In this case, the correction due to attenuation is given by:

$$k_2 = e^{-\mu_c \cdot s} \tag{7}$$

where μ_c is the linear attenuation coefficient of the cladding material for a given gamma-ray energy and s is the cladding thickness.

The third correction to be calculated arises from the attenuation of gamma-rays traversing the air. In this case, the correction due to attenuation is given by:

$$k_3 = e^{-\mu_{ar} \cdot \ell} \tag{8}$$

where μ_{air} is the linear attenuation coefficient of the air for a given gamma-ray energy and ℓ is the distance between the fuel rod cladding and the HPGe detector window.

5. CALIBRATION PROCEDURE

Regarding burnup measurements on fuel rods in hot cells, an alternative calibration standard can be made manufacturing a hollow tube with exactly the same dimensions and material of a fuel rod active length, fulfilling it completely with an aqueous solution of a burnup monitor (for example, ^{137}Cs) whose specific activity is well known and sealing it immediately afterwards.

The total activity of such calibration standard due to the burnup monitor will be:

$$d = \pi \cdot r_0^2 \cdot L \cdot \bar{a} \tag{9}$$

where $\bar{a}$ is the specific activity of the aqueous solution contained in the calibration standard.

Once positioned exactly at the same geometry and configuration that were employed during the measurements on the fuel rod, the average value of the total number of counts registered by the HPGe detector (net area) per unit of time is obtained as a result of measurements performed at various locations along the calibration standard and is related to the specific activity of its aqueous solution by the expression:

$$\bar{q} = \bar{a} \cdot v \cdot I_\gamma \cdot \varepsilon_\gamma \cdot k_1^{'} \cdot k_2 \cdot k_3 \tag{10}$$

where $k_1^{'}$ is the self-attenuation correction of gamma-rays traversing the water among which they are emitted. The value of $k_1^{'}$ is calculated by means of the same expression for k_1, but using the linear attenuation coefficient of water μ_w for a given gamma-ray energy.

Aiming the reduction of the experimental uncertainties, measurements along the calibration standard length must be performed for angles of 0°, 45°, 90°, 135°, 180°, 225°, 270° and 315°. For each one of these angles, a measurements-set will be obtained, whose arithmetical mean constitutes a partial result. Finally, the uncertainty-weighted mean [2] of the eight partial results will be the average value $\bar{q}$.

Combining both equations shown before regarding the calibration standard, its total activity due to a given burnup monitor can be rewritten as follows:

$$d = \frac{\pi \cdot r_0^2 \cdot L \cdot \bar{q}}{v \cdot I_\gamma \cdot \varepsilon_\gamma \cdot k_1^{'} \cdot k_2 \cdot k_3} \tag{11}$$

The ratio of a fuel rod activity to the calibration standard activity gives the result:

$$D = \frac{\overline{Q} \cdot k_1^{'} \cdot d}{\overline{q} \cdot k_1} \tag{12}$$

which enables the direct determination of the total activity of a fuel rod due to a given burnup monitor, based only on parameters that can be either measured or calculated.

6. FISSIONED MASS

The total activity of a fuel rod is the one presented at the instant the irradiation is finished and is related to the measured activity by means of the law of radioactive decay:

$$D = D_0 \cdot e^{-\lambda \cdot t_c} \tag{13}$$

where λ is the radioactive decay constant of the burnup monitor and t_c is the time interval between the end of the fuel rod irradiation and the beginning of the measurements on it.

Another manner to write the equation above is:

$$D = \lambda \cdot N_0 \cdot e^{-\lambda \cdot t_c} \tag{14}$$

which, combined with the calibration results, furnishes the total number of burnup monitor atoms in the fuel rod immediately after the end of the irradiation:

$$N_0 = \frac{\overline{Q} \cdot k_1^{'} \cdot d}{\lambda \cdot \overline{q} \cdot k_1} \cdot e^{\lambda \cdot t_c} \tag{15}$$

Once the burnup monitor is produced in fission at a constant average yield, the mass of uranium that undergone fission is calculated by means of the following expression:

$$\Delta U = \frac{N_0 \cdot M_0}{y \cdot N_U^0} \cdot f \tag{16}$$

where y is the average fission yield of the burnup monitor (see Table 1), M_0 is the original total mass of uranium inside the fuel rod (including all isotopes), N_U^0 is the total number of uranium atoms inside the fuel rod before irradiation (including all isotopes) and f is a correction factor that takes into account the decay of burnup monitor nuclei occurred during different irradiation periods and powers, which is given by the following expression [6]:

$$f = \frac{\lambda \cdot \sum_{k=1}^{n} P_k \cdot t_k}{\sum_{k=1}^{n} P_k \cdot e^{-\lambda . \tau_k} \cdot (1 - e^{-\lambda . t_k})} \tag{17}$$

where λ is the radioactive decay constant of the burnup monitor, P_k is the average relative power corresponding to the *k*th irradiation period (been $\sum_{k=1}^{n} P_k = 1$), n is the total number of irradiation periods during the whole irradiation history of the fuel rod, t_k is the duration of the *k*th irradiation period and τ_k is the time interval between the end of the *k*th irradiation period and the end of the last irradiation period.

From the description of the parameters above, it is easy to realize that a detailed knowledge about the irradiation history of the fuel rod is required to calculate the value of the correction factor f.

The combined use of the equations obtained for N_0, ΔU and f enables the determination of the uranium mass of the fuel rod that undergone fission.

7. AVERAGE BURNUP

The average burnup of the fuel rod, expressed as the fraction of the total number of uranium atoms inside the fuel rod that undergone fission, is given by:

$$B = \frac{\Delta U}{M_0} \tag{18}$$

which is related to the total energy generated during the fuel rod irradiation, per unit of mass of the nuclear fuel, by [7,8]:

$$[\text{GWD / MTU}] = (9.6 \pm 0.3)\ x\ [B\ x\ 100] \tag{19}$$

where GWD / MTU means Giga Watts-Day per Metric Ton of Uranium (most common form employed to express the average burnup of nuclear fuels irradiated in power reactors).

8. CONCLUSIONS

The present work describes the theoretical foundations and experimental techniques necessary to measure, using nondestructive gamma-ray spectroscopy, the average burnup of irradiated fuel rods in a laboratory equipped with hot cells. An alternative calibration procedure is proposed in order to perform the measurements directly.

However, reliable results can be attained by this procedure only if the following requirements are fulfilled: *a*) the geometry and the configuration of the system remain unchanged, which demands a rigorous control of the positioning reproducibility for all components of the experimental apparatus; *b*) the HPGe detector and the associated electronic components are designed for nuclear spectroscopy applications involving ultra-high input counting rates, because measurements will be performed on irradiated nuclear fuels; *c*) the HPGe detector and the associated electronic components work under stable conditions; *d*) all pertinent attenuation corrections are precisely calculated; *e*) the irradiation history of the fuel rod is very well known.

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