Nuclear Inst. and Methods in Physics Research, A 903 (2018) 224–233

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Depletion measurements using a new gamma transparency method of irradiated MTR-type fuel assemblies

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ARTICLE INFO

Keywords:
MTR fuel
Spectroscopic measurements
Rhenium
Gamma transparency
Burnup
Depletion

ABSTRACT

An innovative method for the determination of the depletion in research reactor fuel assemblies (FAs) based on gamma transparency was developed, tested, and presented. The basic concept is the assessment of the uranium content from the attenuation of gamma rays passing through the FA, which is placed between a gamma-emitting source and a spectroscopic detector. With increasing depletion, more uranium transforms into fission products, which causes a significant decrease in the gamma-rays attenuation at energies higher than ~116 keV, thus increasing the count rate at the detector. The attenuation from the fuel assembly was compared to that of a fresh (highest known uranium content, most opaque to gamma-rays) and dummy (Aluminum-only; most transparent) fuel assemblies, and subsequently, a logarithmic interpolation to determine the current uranium content was performed. In this study, a low-energy gamma ray of 155 keV (emitted from ^{188}Re) was used, which exhibited significant contrast between the attenuation through the uranium versus its fission products, while obtaining sufficiently high count-rate at the detector at relatively short count durations. The ^{188}Re source was generated prior to the experimental campaign through the ^{187}Re(n,γ) reaction by irradiation of natural rhenium in IRR1 (Israel Research Reactor 1, 5MW open-pool HEU MTR-type fuel, Soreq Nuclear Research Center, Israel). Five FAs of different irradiation histories previously used in the IRR1 reactor were studied. The determined depletions were found to be in the range of 47%–59%, with absolute uncertainties of 2%–3%. The axial distribution of the depletion along the FAs was determined with high resolution (20 axial points per FA) using the low-energy photons and found to be in good agreement compared to the axial distributions obtained from fission products such as ^{137}Cs. Experimental results of the present study were also compared to results of a different experiment that used the traditional gamma ray spectroscopy technique (i.e., using ^{137}Cs activity) for one of the studied fuel assemblies (FS-5), giving good agreement in both the axial depletion distribution, as well as the whole assembly depletion. Thus, the method described in this paper demonstrates improved accuracy compared to other non-destructive spectroscopy-based methods. Furthermore, we managed to obtain spectroscopic signatures of relatively short-lived fission products such as ^{140}Ba ($T_{1/2}$ = 12.7 days) from an FA that was irradiated 65 days before the measurement.

1. Introduction

Knowledge of nuclear fuel depletion is essential to ensure nuclear reactor safety and to enhance reactor operation and performance. In this paper, depletion is defined as the fraction of initial ^{235}U nuclei that underwent either fission or radiative capture and burnup is defined as the fraction of initial ^{235}U nuclei that underwent fission only. Another motivation to accurately determine the fuel depletion is related to decreasing uncertainties in criticality safety calculations (e.g., burnup credit), thus optimizing spent fuel storage facilities and transport casks. Finally, verification of operator-declared data on the current fissile content in the nuclear fuel is required for safeguards purposes. Both destructive and non-destructive methods were widely used to determine depletion in nuclear fuel elements [1,2]. The destructive methods, which are based on chemical methods involving the fuel dissolution, usually exhibit good accuracy in the depletion [3–5]. However, these methods are not suitable in most cases since the fuel is destroyed and further utilization in the reactor is not possible. Therefore, determination of depletion (or burnup) by non-destructive methods is often sought. These methods are predominantly based on gamma-ray spectrometry,

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https://doi.org/10.1016/j.nima.2018.06.083
Received 9 April 2018; Received in revised form 27 June 2018; Accepted 29 June 2018
Available online 4 July 2018

i.e., using fission product activities as burnup indicators requires the knowledge of the fuel irradiation history [6–12]. The vast majority of studies focused on nuclear power plants [8,11–15] and fewer works dealt with research reactors [16–19]. Many studies were validated against the traditional, destructive chemical methods [9,15,20] and to numerical simulations [9–13,19].

For research reactors using MTR-type fuel, the most common experimental method to measure Fuel Assembly (FA) depletion is based on measuring the rate of gamma rays emitted from long-lived fission products (FPs) inside the FA [6,7,10,13,15]. Either the measurement of the activity of a radionuclide (e.g., ^{137}Cs, which has a half-life of 30.1 years and emits gamma rays at 662 keV) is used or the activity ratio between an FP and an activation product (e.g., ^{154}Eu/^{137}Cs), where one was created in proportion to the fuel depletion and the other — to its square.

However, accurate determination of the depletion from these measurements is challenging because:

1. It requires accurate calibration of the detection efficiency of the ^{137}Cs in the spectroscopic detector, which is needed for transforming the measured count rate to its activity (e.g., in units of Ci). This calibration depends on the geometrical settings, the intrinsic efficiency of the detector, and the self-absorption of the gamma rays inside the FA itself.
2. The knowledge of the irradiation history of each FA is required, including the power distributions inside each core layout where the FA was used. In research reactors, where the irradiation regime can be irregular (i.e., non-continuous irradiation periods separated by varying cooling periods of months or even years), with strongly absorbing control rods that distort the shape of the flux in the reactor, this information is far from trivial.

In order to deal with these two shortcomings, a new experimental method was developed and implemented in the IRR1 in Soreq Nuclear Research Center. In this new method, ^{235}U content was determined from direct measurement of the attenuation of low-energy gamma rays that passed through the FA. For gamma rays in the low energy range (~116–200 keV), the attenuation coefficient is significantly higher for uranium compared to its fission products, as can be seen in Fig. 1. As an example, the difference in the attenuation coefficient reaches ~400% for the photon energy of 155 keV. Thus, the more an FA was depleted, the more "transparent" it became.

This paper is organized as follows: in Section 2 we derive the relation between gamma ray attenuation through an FA and the depletion, followed by description of the experimental setup and present the results in Sections 3 and 4, respectively. We summarize and conclude key findings in Section 5.

2. Materials and methods

2.1. Gamma transparency method

The attenuation of photons passing through a material having a thickness w is described by:

$$I = I_0 e^{-\mu\rho w} \tag{1}$$

where I_0 and I are the intensities before and after passing through a material, respectively, μ is the macroscopic attenuation coefficient, and ρ is the mass density (the product $\mu\rho$ is known as the linear attenuation coefficient). In the low-energy range, the interaction between photons and matter is dominated by the photoelectric effect, which strongly depends on the atomic number (Z) and energy (E), according to the relation $\mu_{Pe} \propto Z^4/E^3$ [22]. Thus, a strong absorption in heavy elements (e.g., uranium) is achieved, compared to the much lighter fission products. In order to determine the uranium content of an FA, this large contrast can be exploited by comparing the transparency to gamma

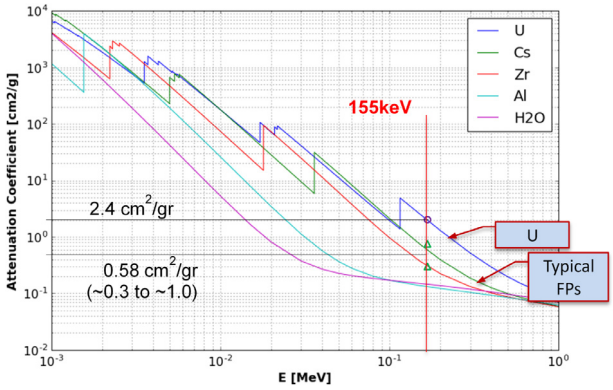

Fig. 1. Attenuation coefficient as a function of photon energy [21] for the most abundant materials content in an irradiated fuel assembly (i.e., U, Al, H_2O and typical fission products, such as cesium and zirconium). The relatively high-contrast of attenuation between U and typical fissions products (Cs, Zr) is shown for 155 keV gamma ray.

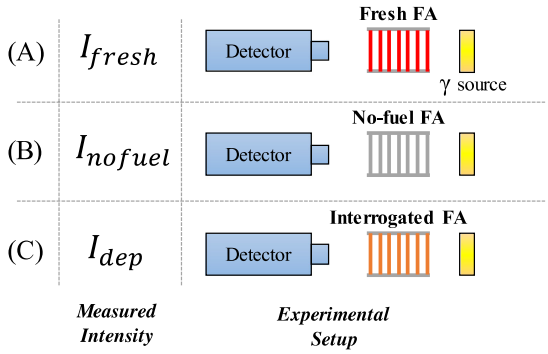

Fig. 2. Conceptual experimental setup and required measurements in three configurations: (A) Fresh FA (zero depletion); (B) Dummy FA (same geometry, but without Uranium); (C) Interrogated FA (unknown depletion).

rays in the energy range described above, through the interrogated FA and through calibration FAs with known materials composition. The measurements were done in the following manner: 1. A high intensity radioactive source, which emits gamma rays in the relevant energy range, was produced, 2. A spectroscopic detector was used to measure the count rate of the gamma rays emitted from the radioactive source and passing through (A) a fresh FA (i.e., zero depletion), (B) A dummy element ("no-fuel") having equal geometry and composition that does not contain uranium, and, (C) the interrogated FA, for which we want to determine the depletion, as illustrated in Fig. 2.

The chosen radioactive source was a natural rhenium foil irradiated in the IRR1 reactor (described in the next section and refs. [10,23,24]), which generated ^{188}Re (via ^{187}Re(n,γ) reaction) that emitted 155 keV gamma rays. This source was chosen for two primary reasons: (1) Energy of the gamma rays is within the U/FP high contrast region (Fig. 1), and (2) Relatively high neutron capture cross section and short product half-life, resulting in an activation rate that yielded sufficient gamma count at the detector.

The count rates observed by the detector for the three FAs (Fig. 2) are defined as I_{fresh}, I_{nofuel} and I_{dep}, respectively. The count rate through the interrogated FA with depletion dep is given by

$$I_{dep} = I_{fresh} \cdot \exp\left[\left(\rho^0_{U5} - \rho_{U5}(dep) - \rho_{U6}(dep)\right) \cdot \mu_U \cdot w_{meat} - \rho_{FP}(dep) \cdot \mu_{FP} \cdot w_{meat}\right] \tag{2}$$

where $\rho^0_{U5}, \rho_{U5}, \rho_{U6}$, are the densities of the fresh ^{235}U, and the depleted ^{235}U, ^{236}U, and Fission Products, ρ_{FP}. w_{meat} is the fuel meat thickness,