A simple gamma spectrometry method for evaluating the burnup of MTR-type HEU fuel elements

T. Makmal [a,b], O. Aviv [c], E. Gilad [a,*]

[a] The Unit of Nuclear Engineering, Ben-Gurion University of The Negev, Beer-Sheva 84105, Israel
[b] Nuclear Physics and Engineering Division, Soreq Nuclear Research Center, Yavne 81800, Israel
[c] Radiation Safety Division, Soreq Nuclear Research Center, Yavne 81800, Israel

ABSTRACT

A simple method for the evaluation of the burnup of a materials testing reactor (MTR) fuel element by gamma spectrometry is presented. The method was applied to a highly enriched uranium MTR nuclear fuel element that was irradiated in a 5 MW pool-type research reactor for a total period of 34 years. The experimental approach is based on in-situ measurements of the MTR fuel element in the reactor pool by a portable high-purity germanium detector located in a gamma cell. To corroborate the method, analytical calculations (based on the irradiation history of the fuel element) and computer simulations using a dedicated fuel cycle burnup code ORIGEN2 were performed. The burnup of the MTR fuel element was found to be 52.4 ± 8.8%, which is in good agreement with the analytical calculations and the computer simulations. The method presented here is suitable for research reactors with either a regular or an irregular irradiation regime and for reactors with limited infrastructure and/or resources. In addition, its simplicity and the enhanced safety it confers may render this method suitable for IAEA inspectors in fuel element burnup assessments during on-site inspections.

© 2016 Elsevier B.V. All rights reserved.

1. Introduction

Knowledge of nuclear fuel element (FE) burnup data is essential to ensure nuclear reactor safety and to enhance reactor operation and performance. In this paper, FE burnup is defined as the fraction of initial ^{235}U nuclei that underwent fission, and FE depletion is defined as the fraction of initial ^{235}U nuclei that underwent either fission or radiative capture. These two quantities, linked via the capture-to-fission ratio [1], are equivalent to each other via the energy-per-fission quantity. Burnup is usually measured, via destructive or non-destructive methods, in units of the total energy produced by the fuel normalized by the initial weight of heavy metals in the fuel elements, e.g., MWd/kgU.

Destructive methods for evaluating FE burnup are characterized by good accuracy without the need for detailed historical irradiation data [2–4]. However, these methods require complex and expensive infrastructure, entail substantial risk of exposure to high radiation levels, and involve chemical processes and expertize that rarely exist in research reactor facilities. Non-destructive assays (NDA), on the other hand, are safer and can be executed practically in any nuclear reactor, but they are usually less accurate [4–6]. One

of the most common NDA methods for evaluating fuel burnup is gamma spectrometry [3–5,7–9]. Here, the overall activity of a specific fission product, whose production rate is proportional to the fission rate (e.g., ^{137}Cs), is determined and later used to derive FE burnup using the irradiation history [5,8,10–12]. NDA methods have no significant radiation safety issues and usually require relatively short measurement durations (minutes).

The determination of burnup using NDA methods based on HPGe gamma-ray spectrometry, using fission product activities as monitors, has become very popular since the 1970's [8,13–15]. Most studies focused on spent fuel from NPPs, e.g., PWRs [16–23], BWRs [16,24–30], Pebble bed [31,32], and MOX fuel [18,26,33]. Fewer studies focused on RRs fuel, e.g., TRIGA [34–38], MTR [15,39], and other [38,40,41]. Moreover, some studies were devoted to high burnup spent fuel [16,19–22,30] and many studies involved validation by comparison to destructive chemical methods [20,21,25,27,30] and to numerical simulations [17–19,21–23,28,29,31,32,36–39,41]. Finally, most studies utilized special purpose gamma-ray spectrometry measurement systems [15–20,26,27,29,30,33–37,39,41,42]. For a comprehensive list of references see the review by Parker 2015 [43] and refs therein.

The vast majority of studies in this field address FEs from nuclear reactors with regular irradiation regimes, such as nuclear power plants, isotope production facilities and research reactors. A regular irradiation regime, in which reactor power, cycle lengths,

---

and cooling period durations do not change dramatically over time, enables the use of an alternative approach to evaluate fuel burnup, i.e., based on the measured activity ratios of fission and activation products, e.g., $^{134}Cs/^{137}Cs$ or $^{154}Eu/^{137}Cs$. However, the activation products ^{134}Cs and ^{154}Eu are relatively short lived, and as such, they are not applicable to determining the absolute burnup of FE that underwent irregular operation regimes and/or that were subjected to long irradiation periods (e.g., > 20 years). They are, however, applicable for determining the relative axial distribution of the burnup along the FE.

This study investigated the use of gamma spectrometry, a simple, NDA method, to evaluate the burnup of a MTR nuclear FE that was irradiated at Israeli Research Reactor #1 (IRR1), located at the Soreq Nuclear Research Center, for a period of 34 years. The experimental approach was based on remote measurements of the gamma spectrum of the FE using a high-purity germanium (HPGe) detector. Gamma spectra were obtained for the entire FE and for different axial segments. The experimental results were compared both to analytical calculations and to computer simulations with the well-known ORIGEN2 burnup code [44].

2. Israeli research reactor #1 (IRR1)

The measurements were carried out at IRR1, a 5 MW swimming pool type reactor operated by the Israel Atomic Energy Commission (IAEC) since June 1960 [45] (Fig. 1). Pool nominal dimensions are 11.0 m × 6.1 m × 9.7 m, and it contains 400 m^3 of deionised water ("light water"). The reactor core is fueled by 24–30 highly enriched (93%) uranium (HEU) plate-type MTR FEs. Each FE contains 23 parallel fuel plates mounted between two lateral aluminum holders. Overall FE dimensions are 7.6 cm × 8.0 cm × 87.3 cm, with an active length of 60 cm. Each fuel plate measures 0.051 cm thick, and it is enclosed in 0.038 cm thick aluminum cladding. According to the fuel manufacturer (CERCA, France), cladding integrity is guaranteed up to a burnup of 90%. However, due to various operational considerations, e.g., fuel depletion and flux distribution in the core, IRR1 safety regulations permit maximal FE burnup that is much lower than the manufacturer's limit.

The measurements and calculations were done on fuel element 5 (FE5), which was selected due to its long and highly irregular irradiation history and its (expected) high burnup level. FE5 was irradiated intermittently between the years 1977 and 2011 under a wide variety of core configurations and in both the center and along the periphery of the core. Therefore, FE5 can be considered an excellent representative of an "average FE", and as such, the exact core configuration during each irradiation cycle can be
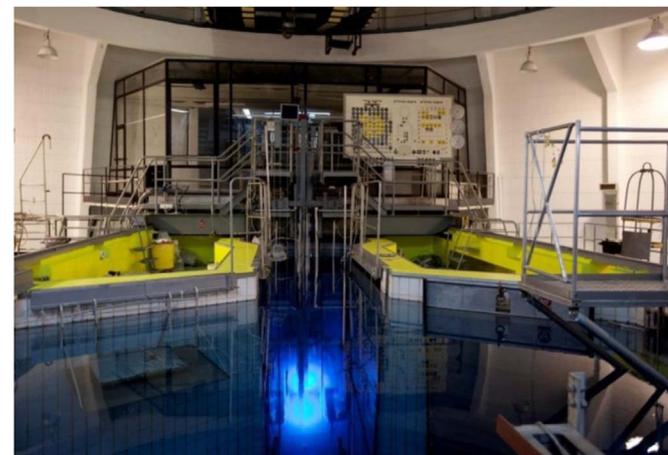
neglected. Furthermore, since the FEs consist of 93% HEU, the power was dominated by the thermal fission of ^{235}U, and rendering the contributions from other fissile materials negligible [45].

3. Burnup evaluation by analytic calculation

The burnup of a FE is the ratio between the amount of fissile material consumed by fission and the initial amount of fissile material. The initial amount of ^{235}U in FE5 was approximately 12.23 g per plate and 281.40 g for the entire FE, with associated uncertainty of less than 0.05 g [46]. The amount of ^{235}U consumed by fission was derived principally from the irradiation periods and the corresponding power levels. The estimation of the yearly power relied on two documentation sources: the first comprised monthly reports that specified the amounts of energy (in MWH) produced by IRR1 during the different irradiation cycles, and the second comprised yearly and semi-yearly operational reports on the status of the reactor's nuclear fuel inventory and the amount of energy produced (in MWD).

Two assumptions were necessary. First, FE5 was considered to be representative of an average FE because over time it had been shifted between most of the positions in the reactor's core (i.e., 13 out of 20 possible conventional sites during 34 years). The second assumption was that 1 MWD is equivalent to the consumption of a total of 1.22 g^{235}U, which is the ^{235}U consumption value calculated for a typical FE in IRR1 [47]. During the first 15 years of FE5 irradiation period, the average power experienced yearly by it, shown schematically in Fig. 2, exhibited only minor fluctuations. Beginning in the early 1990s, however, FE5 irradiation was characterized by irregularities, including significant changes in the power level and in the lengths of the irradiation periods, including periods where FE5 was not irradiated (i.e., was outside the core, e.g., 2005–2007). These changes resulted from the shuffling of FE5 location in the core. Based on the irradiation history and the assumptions above, the total amount of ^{235}U consumed in FE5 between 1977 and 2011 was 159.9 g. The uncertainty in the amount of final fissile material was estimated using the relative uncertainty in the information about the reactor's irradiation history, which was 5%. Therefore, the overall burnup for FE5 is $48.9 \pm 2.4\%$.
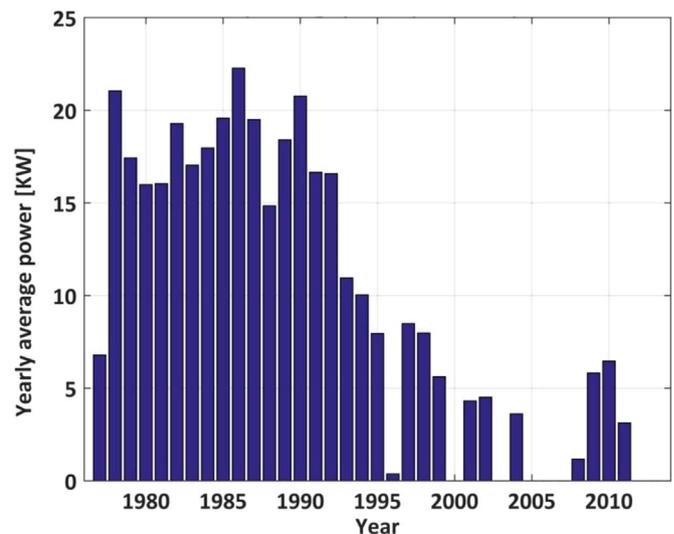

Fig. 1. Front view of IRR1.

Fig. 2. Average power produced yearly by FE5 in IRR1 between 1977 and 2011. During 2000, 2003, 2005–2007 and since 2012 FE5 was outside the core.

4. Burnup evaluation by gamma spectrometry

The burnup of FE5 and its axial profile were evaluated based on the radioactive fission and on the activation products ^{137}Cs, [134]Cs, and [154]Eu ("burnup indicators"). Key properties of these radionuclides within the FEs – e.g., low neutron-capture cross sections (including precursors) and low migration (including precursors) – render them suitable for burnup evaluation [8,48]. Moreover, relying on these properties ensures that only a weak dependence exists between fission yields and reactor operating history and between radionuclide activity ratios and burnup. Likewise, this approach also ensures that the fission product density is proportional to the number of fissions, which minimizes the decay corrections. In addition, ^{137}Cs, [134]Cs, and [154]Eu have high energy and resolvable characteristic gamma-rays, properties that make them good candidates for burnup evaluation by gamma spectrometry methods.

The fission product ^{137}Cs is linearly dependent on the integral neutron flux, and as such, it is less dependent on the irradiation history. The comparable durations of the irradiation period and of the half-life of [137]Cs (30.1 y) dictated the inclusion of a significant decay correction. Moreover, the accumulation of ^{137}Cs in the fuel shows that it has weak sub-linear dependence on FE burnup. In contrast, the half-lives of [134]Cs (2.06 y) and [154]Eu (8.60 y), significantly shorter than the 34-year irradiation period of FE5, make them unsuitable to be used for evaluations of the absolute burnup of FE5, which was instead performed using [137]Cs. However, [134]Cs and [154]Eu were used to evaluate the relative axial burnup and to study its dependence on these shorter lived activation products. The axial distributions of [154]Eu and ^{137}Cs contain revealing information about the entire irradiation history, including the axial distribution of the power density in the FE during irradiation. Likewise, the axial distribution of [134]Cs is similarly informative, but for a much shorter period of approximately 10 years. Finally, the axial distributions of both [154]Eu and [134]Cs are proportional to that of the burnup in FE5 under two (reasonable) assumptions. First, the axial power distribution that FE5 experienced during its long irradiation history is similar to an average axial power distribution, and second, this average axial power distribution did not change dramatically during the irradiation period.

4.1. Experimental setup

The experimental setup used in this study is presented in Fig. 3. FE5 was remotely moved by a motorized lever into a fixed position inside the reactor pool and facing the detector. Its position was monitored via an underwater camera. The detector used in this study was a portable HPGe detector (model Falcon 5000, Canberra) with a relative detection efficiency of 22% and a resolution of 1.9 keV at gamma energy of 1332.5 keV (^{60}Co). The HPGe detector was situated in a "gamma cell" (commonly found in research reactors) separated from the pool by a thin aluminum window (at a depth of 8.46 m). The Falcon 5000 is equipped with built-in signal processing units (amplifier and multi-channel analyzer) and a high voltage power supply. The detector was remotely controlled by a dedicated tablet (connected by a LAN cable). The Genie 2000 software [49] was used for data acquisition and analysis. The detection efficiency for the gamma-ray energy of the burnup indicator, [137]Cs (661.7 keV), was calculated using the in situ object counting system (ISOCS), which is Monte-Carlo software based on comprehensively validated MCNP code [50,51]. The geometry and the material composition of the experimental setup, including the detector's inner structure, the radiation source, and their relative positions, were used as input for the ISOCS software. In the framework of this study, efficiency calculations were validated from measurements of a cylindrically shaped ^{60}Co calibration source with a total activity of 108 ± 5 Ci and whose dimensions were 40.6 cm × Ø0.6 cm [52]. The source was inserted into the reactor pool and measured under conditions similar to those used for FE5 [47].

Two types of measurements were carried out. The first was used to determine the total burnup. Here, the distance between the FE and the gamma cell's aluminum window was 191.5 cm and the distance between the aluminum window and the detector was 17 cm (see Fig. 3). A rectangular lead wall measuring 61 cm × 61 cm × 15 cm with a small aperture (6 cm × 15 cm × 20 cm) was positioned between the detector and the aluminum window. The aperture dimensions and the detector-source distance provided a solid angle that was sufficient to cover the entire FE. The second measurement was used to determine the burnup distribution along the main axis of FE5. Here, a lead wall (61 cm × 61 cm × 15 cm) with a narrow aperture (4 cm × 20 cm × 15 cm), positioned in the gamma cell
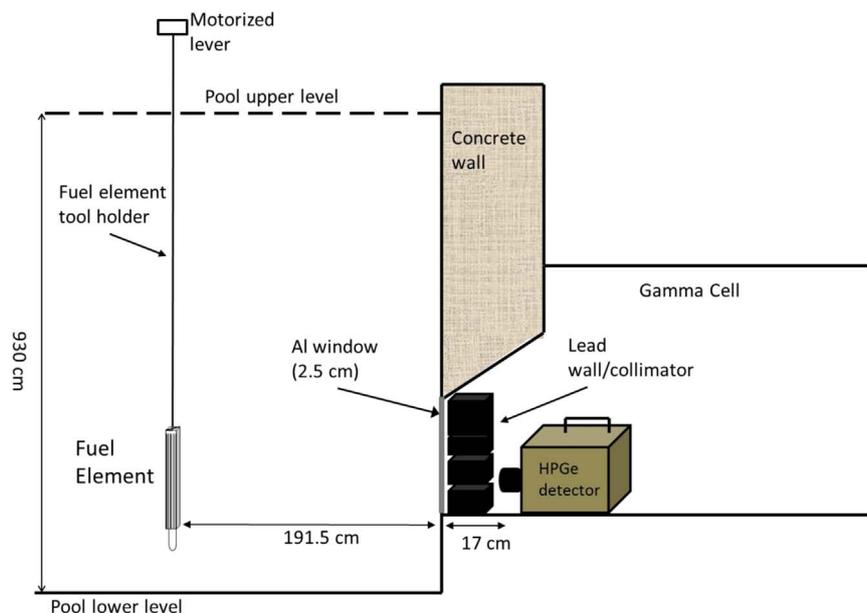

Fig. 3. Scheme of the experimental setup (see text for details).

between the detector and the aluminum wall, was used to focus on discrete sections of FE5. The aperture dimensions and the detector-source distance allowed measurements to be made of six non-overlapping axial segments (10 cm each). The vertical position of FE5 was varied by a dedicated tool holder. The distance between FE5 and the aluminum window was 158 cm and the distance between the aluminum window and the detector was 108 cm.

The simple shielding arrangements described above guaranteed a sufficient signal-to-noise ratio, i.e., via the attenuation of high energy gamma-rays and secondary events (caused by the interactions of particles emitted from FE5 with the surrounding media).

4.2. Determination of the burnup

The burnup (BU) of a FE is defined as the ratio between the number of ^{235}U nuclei that underwent fission in the FE, N_{fiss}, and the initial number of ^{235}U nuclei in the FE, N_0,

$$BU = \frac{N_{fiss}}{N_0}. \tag{1}$$

The initial number of ^{235}U nuclei per FE is specified by the manufacturer, $N_0 = (7.210 \pm 0.002) \times 10^{23}$. The number of ^{235}U nuclei that underwent fission, N_{fiss}, is related to the number of fission product indicators (N_{ind}) in the FE normalized by the fission yield (Y) and by the decay-buildup correction factor (f) based on the irradiation history, according to

$$N_{fiss} = \frac{N_{ind} \cdot f}{Y}. \tag{2}$$

For ^{137}Cs, $Y = 6.26\%$ [12]. The decay correction factor, f, can be expressed by the Bibichev correlation function [11,53] given by

$$f = \frac{\lambda \cdot \sum_{i=1}^{n} P_i \cdot t_i}{\sum_{i=1}^{n} P_i \cdot e^{-\lambda \cdot \tau_i} \cdot (1 - e^{-\lambda \cdot t_i})}, \tag{3}$$

where P_i is the average relative power fraction corresponding to the nth irradiation period ($\sum_{i=1}^{n} P_i = 1$), n is the total number of irradiation periods, t_i is the duration of the ith irradiation period, τ_i is the time interval between the end of the ith irradiation period and the end of the last irradiation period, and λ is the decay constant of the fission product. The parameters P_i, t_i, and τ_i were calculated for each irradiation period i from IRR1 documentation history. Accounting for the measurement period and the associated uncertainty evaluated from the irradiation history data, the decay correlation factor for ^{137}Cs was found to be 1.8 ± 0.1.

The number of fission product indicators can be obtained directly from a measurement of the fission product activity in the FE, given by

$$N_{ind} = \frac{A \cdot e^{\lambda \cdot t_c}}{\lambda}, \tag{4}$$

where t_c is the time that elapsed between the end of the last irradiation and when the measurement was made (being 2.62 years for FE5). The specific activity A of a burnup indicator is given by Eq. (5) (which neglects decay corrections during the measurement),

$$A = \frac{R}{Q \cdot \varepsilon}, \tag{5}$$

where R is the ratio between the net photo-peak area of the fission product's characteristic gamma-ray and the measurement live-time (i.e., count rate), and Q is the gamma-ray emission probability, being $84.99 \pm 0.20\%$ for ^{137}Cs for the gamma-ray energy of 661.7 keV [54]. Finally, ε is the detection efficiency, which includes attenuation factors accounting for the various media the gamma-

ray passes through between the source and the detector. For the first measurement set, the detection efficiency for the gamma-ray energy of 661.7 keV, derived from Monte-Carlo simulation, was found to be $(1.4 \pm 0.1) \times 10^{-13}$ counts/γ. The uncertainty of the detection efficiency was estimated by propagating the uncertainty along the distance between the FE and the aluminum window (± 1 cm). Finally, the explicit relation between the burnup and the measured photo-peak count rate of ^{137}Cs for FE5 is

$$BU = \frac{R \cdot e^{\lambda \cdot t_c} \cdot f}{Y \cdot \varepsilon \cdot P \cdot \lambda \cdot N_0} = 0.4757 \cdot R \tag{6}$$

4.3. Experimental results

Two sets of gamma-ray measurements were performed. The first set was used to determine the absolute burnup of FE5 via the activity of the fission product ^{137}Cs (Fig. 4, top). The spectrum was acquired for 5 min with a relatively low dead time of less than 2%. The peaks associated with ^{137}Cs (661.7 keV), ^{134}Cs (795.9 keV), and ^{154}Eu (1274.4 keV) are clearly distinguishable from the Compton continuum and from those of other radionuclides typically found in a reactor pool environment after operation (e.g., ^{24}Na). All of the fuel elements (including FE5) were located more than 6 m from the gamma cell, and a background measurement revealed no ambient contributions within the region-of-interest around the burnup indicator's photo-peaks (dashed blue lines along the bottom images in Fig. 4).

The count rate for ^{137}Cs was found to be 1.09 ± 0.10 s^{-1}, which was translated to an activity of 243 ± 22 Ci using Eq. (5). Thus, the absolute burnup, determined based on the activity measurement and the irradiation history according to Eq. (6), was found to be $52.4 \pm 8.8\%$. The dominant contributions to the uncertainty in the burnup measurement originated mainly from the statistical uncertainty in the measured count rate (9%), the uncertainty in the calculated detection efficiency (7%) and the uncertainty in the irradiation history (5%).

The second set of measurements was used to determine the relative axial burnup distributions of FE5. To minimize any systematic uncertainties, all measurements were performed in sequence and during identical live times (4 min). Fig. 5 shows the normalized photo-peak count rates (661.7 keV, 795.9 keV and 1274.4 keV) of the corresponding burnup indicators (^{137}Cs, ^{134}Cs, and ^{154}Eu) that appear in the measured gamma-ray spectrum (Fig. 4). The horizontal error bars represent uncertainties in position and the vertical error bars represent statistical uncertainties in the photo-peak count rates. The relative axial distributions exhibit similar non-symmetric cosine-like axial profiles around the center of FE5. This result is consistent with the IRR1 safety analysis report and the position of the control blades in the upper part of the core [45]. The burnup in the FE is about 40% higher at its center than along its edges, an effect that has been observed in other studies [11,53]. Nevertheless, the resulting relative axial profiles of the three burnup indicators are significantly different, especially along the edges of the FE. Due to its relatively long half-life decay constant, only minor losses have occurred in the (integral) irradiation information stored in the ^{137}Cs signal since 1977. In contrast, the signals of ^{154}Eu and ^{134}Cs, which have shorter half-lives, store (integral) irradiation information of later, much shorter time periods. The profile of ^{137}Cs, therefore, is flatter than those of ^{154}Eu and ^{134}Cs, and the profile of ^{154}Eu is flatter than that of ^{134}Cs.

5. Burnup evaluation by simulations

To further verify the experimental results, the dedicated burnup code ORIGEN2 was used to assess FE5 burnup. This code
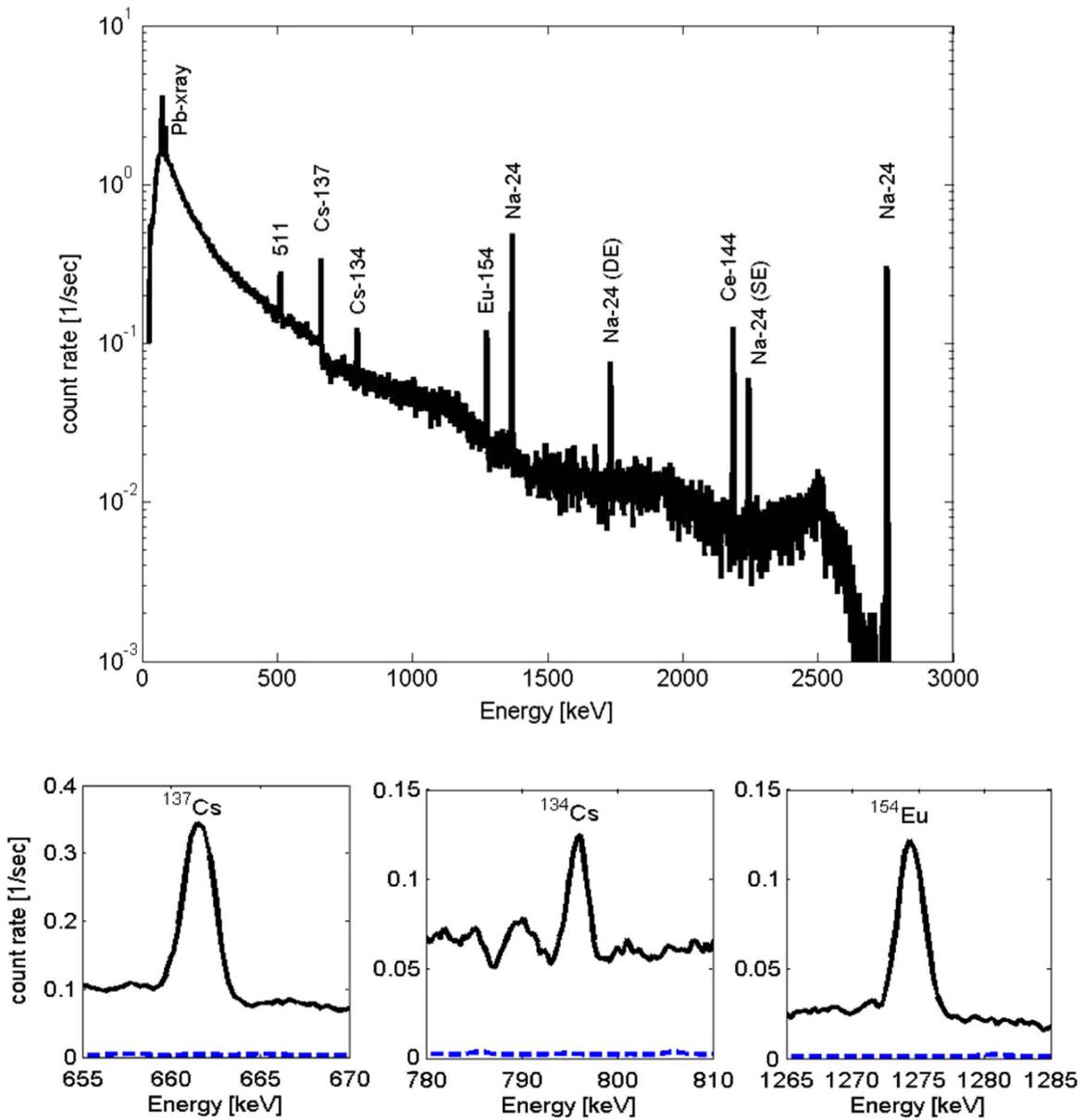
Fig. 4. A typical gamma-ray spectrum for FE5 positioned in IRR1 (top) and a zoom-in on the main photo-peaks of ^{137}Cs, ^{134}Cs and ^{154}Eu (bottom). The bottom figures also show the background signal (dashed blue line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
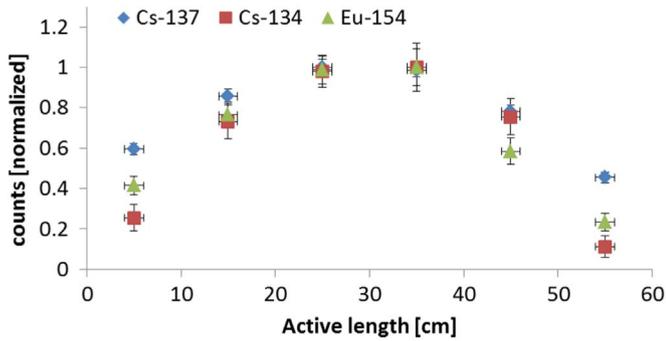

Fig. 5. Relative burnup for ^{137}Cs, ^{134}Cs and ^{154}Eu along the active length of FE5.

Table 1
Geometry and composition of a fuel element.

Component	Dimensions	Isotope	Density [g/cc]
Fuel plate (meat)	$0.051 \times 6.23 \times 60$ cm^3	^{235}U	6.41781E-1
		^{238}U	4.83046E-2
		Al	2.46444E+0
Cladding	0.038 cm	Al	2.70000E+0
Water	0.105 cm	H$_2$O	9.95670E-1
Support plate	0.475 cm	Al	2.70000E+0

includes various reactor models and cross section libraries for actinides, fission products and activation products (together with their half-lives), and fission yields for both neutrons and photons [44]. The dimensions and isotopic composition of each component in the FE are presented in Table 1. The code, which solves the burnup equations using the matrix exponential method, calculates both mass and activity for a very wide range of materials and isotopes, including fission products, actinides and activation products, and as such, it accounts for most of the known types of nuclear reactions. By default, the code implements the point reactor model for burnup calculation. The input file thus describes 23 fuel plates that constitute a total initial amount of 281.4 g of ^{235}U (93% enrichment), two additional support plates, and water
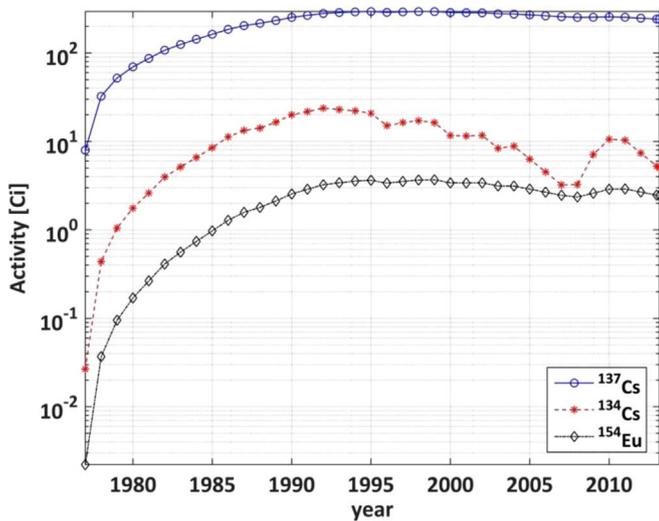
Fig. 6. Calculated activities of ^{137}Cs, ^{154}Eu, and ^{134}Cs in FE5 between 1977 and 2011.

flowing between the plates. The irradiation history, including the yearly average power level per FE (shown in Fig. 2) was used in the calculations.

Fig. 6 shows the calculated concentrations and activities of ^{137}Cs, ^{134}Cs, and ^{154}Eu, and Fig. 7 shows the mass of ^{235}U and the burnup of FE5. Thus, the calculated burnup of FE5 at the time of the experimental measurements was found to be, according to ORIGEN2, 49.1 ± 2.5%.

The calculated activity and activity ratio of the radionuclides ^{134}Cs and ^{154}Eu in FE5 between 1977 and 2011 are shown in Fig. 8. Due to the irregular irradiation regime and the short decay time scales compared to the length of the total irradiation period, the activity ratio of these radionuclides is not injective, which further supports the statement that this quantity cannot be used to determine fuel burnup.

6. Summary and discussion

This study presents the burnup evaluation of an MTR type fuel element irradiated for a period of 34 years in an irregular irradiation regimes using gamma spectrometry. The burnup was assessed via two independent calculation-based approaches. The
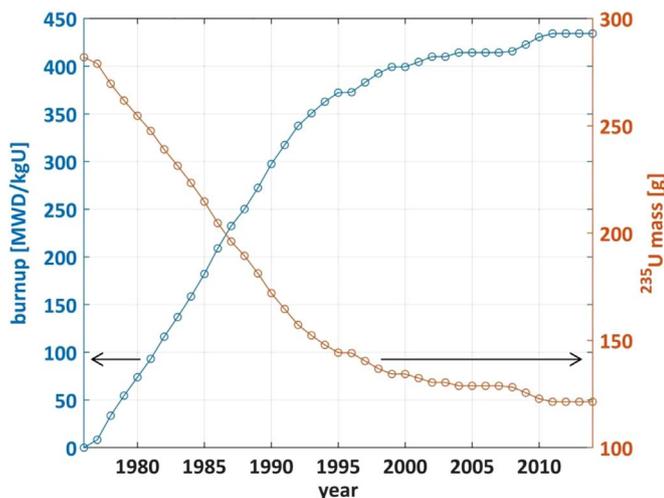

Fig. 7. Calculated burnup (blue) and ^{235}U mass (red) of FE5 between 1977 and 2011. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
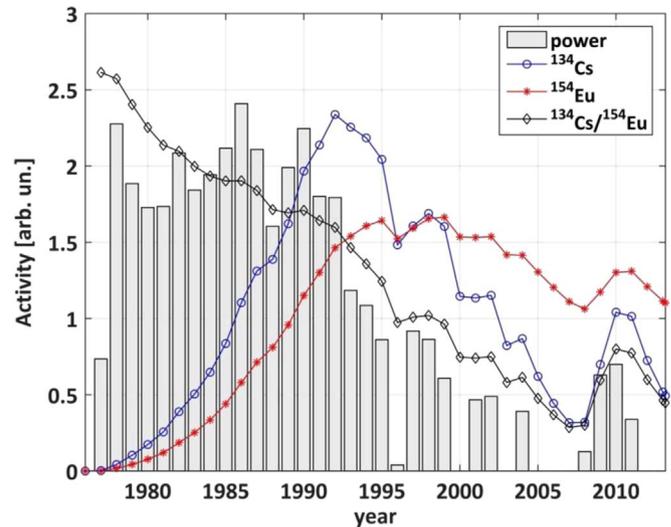

Fig. 8. Calculated activities of ^{154}Eu (red) and ^{134}Cs (blue) and their normalized ratio (black) in FE5 between 1977 and 2011. The average power per element per year is schematically depicted with bars. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2
Summary of burnup results for FE5 using three independent methods.

Method	Burnup [%]
Analytical	48.9 ± 2.4
Experimental	52.4 ± 8.8
Simulations (ORIGEN2)	49.1 ± 2.5

measured and calculated burnup results for FE5 are summarized in Table 2. The assumptions used in this paper (i.e., average yearly power and the use of FE5 as an average element) seem to be sufficient for the evaluation of fuel element burnup using a simple and quick method with good accuracy. According to Table 2, the deviation between the calculated value (by simulations) and the measured value is 5.2% for the burnup of FE5 and that the measured value is higher than the calculated values. This result is consistent with previous studies showing, under more complex experimental settings, deviations of up to 8% between calculated and experimental values [12].

The burnup evaluation method presented in this paper is unique for two reasons. First, the experimental setup and the measurement procedures are simple, inexpensive, safe and highly flexible, and can be deployed and implemented relatively quickly. Moreover, no special purpose systems were constructed for this experiment, and the infrastructures used are common in most existing pool type research reactors around the world. Second, the measurements can be carried out by the reactor's operating team. Likewise, International Atomic Energy Agency inspectors can also exploit this method as a simple and immediate tool for evaluating the burnup levels of fuel elements during safety inspections.

Another important conclusion arising from this study refers to the adequacy of the calculation scheme used to verify the experimental results. The calculation scheme used in this study is based on several important assumptions. The first one is that FE5 is representative of an average FE because over time it had been shifted between most of the positions in the reactor's core. This assumption, although seems bold, is proved to be sufficiently accurate for FE5. However, it can be generalized to any FE in any research reactor that was irradiated for a long period in many different locations in the core. The second assumption is that although the reactor was operated for 2–3 days a week for 6–7 h

each time, the burnup calculation can rely on a much coarser temporal resolution, e.g., using yearly power averages per FE. This assumption was also verified and the use of coarse temporal resolution, e.g., a year, substantially simplifies the calculations without a significant toll on accuracy. The third assumption is that 1 MWD is equivalent to the consumption of a total of 1.22 g ^{235}U, and not the thermal reactors' approximate value of 1.25 g ^{235}U/MWD. This value was calculated for a typical FE in IRR1 and has significant effect on the interpretation of the experimental results. Hence, it is highly recommended to calculate this consumption rate and not use approximate values.

Finally, it was demonstrated that the use of activity ratio of different isotopes, either measured or calculated, cannot be used to estimate the absolute burnup of FEs in research reactors characterized by highly irregular irradiation regime. A detailed description of the calculation schemes, including a comparison between different Monte Carlo codes, different burnup modules and different geometrical models used in this study will be published in a complementary paper.

Acknowledgments

References

[1] J.R. Lamarsh, Introduction to Nuclear Reactor Theory, Addison-Wesley, 1966.

[2] J.S. Kim, Y.S. Jeon, S.D. Park, B.C. Song, S.H. Han, J.G. KIM, Dissolution and burnup determination of irradiated U–Zr alloy nuclear fuel by chemical methods, Nucl. Eng. Technol. 38 (3) (2006) 301.

[3] ASTM C1769-15, Standard Practice for Analysis of Spent Nuclear Fuel to Determine Selected Isotopes and Estimate Fuel Burnup, ASTM International, West Conshohocken, PA, 2015.

[4] EGADSNF, Spent nuclear fuel assay data for isotopic validation - State-of-the-art Report, Paris, France: OECD, NEA/NSC/WPNCS/DOC(2011)5, 2011.

[5] J.R. Phillips, Irradiated fuel measurements, in: D. Riely, N. Ensslin, H. Smith Jr., S. Kreiner (Eds.), Passive Nondestructive Assay of Nuclear Materials, Nuclear Regulatory Commission, 1991, pp. 529–562, NUREG/CR-5550, LA-UR-90-72.

[6] A. Lebrun, G. Bignan, Nondestructive assay of nuclear low-enriched uranium spent fuels for burnup credit application, Nucl. Technol. 135 (3) (2001) 216.

[7] B.B. Bevard, J.C. Wagner, C.V. Parks, M. Aissa, Review of Information for Spent Nuclear Fuel Burnup Confirmation, NUREG/CR-6998, ORNL/TM-2007/229, Nuclear Regulatory Commission, 2009.

[8] S.T. Hsue, T.W. Crane, W.L. Talbert, Jr. J.C. Lee, Nondestructive Assay Methods for Irradiated Nuclear Fuels LA-6923 (ISPO-9) 1978.

[9] IAEA, Practices and developments in spent fuel burnup credit applications, IAEA-TECDOC-1378, 2003.

[10] S.A. Ansari, M. Asif, T. Rashid, K.G. Qasim, Burnup studies of spent fuels of varying types and enrichment, Ann. Nucl. Energy 34 (2006) 641.

[11] M.V. Mora, A.G. Padilla, J.L. Palomino, L.A. Terremoto, Nondestructive burnup measurements by gamma-ray spectroscopy on spent fuel elements of the RP10 research reactor, Prog. Nucl. Energy 53 (2011) 344.

[12] IAEA, Determination of research reactor fuel burnup, IAEA-TECDOC-633, 1992.

[13] F.L. Lisman, W.J. Maeck, J.E. Rein, Determination of the nuclear fuel burn-up from fission product analysis, Nucl. Sci. Eng. 42 (2) (1970) 215.

[14] P. Popa, M. De Coster, D. Langela, Burn-up determination of nuclear fuels by high resolution gamma spectrometry, track formation in solid-state detectors, and neutron dose measurements, Nucl. Technol. 9 (5) (1970) 755.

[15] J.R. Phillips, T.R. Bement, K. Kaieda, E.G. Medina, Nondestructive Verification of Relative Burn-up Values and Cooling Times of MTR Irradiated Fuel Elements, Los Alamos National Laboratory, 1979.

[16] D.D. Cobb, J.R. Phillips, G.E. Bosler, G.W. Eccleston, J.K. Halbig, C.R. Hatcher, S.T. Hsue, Nondestructive Verification and Assay Systems for Spent Fuels, Los Alamos National Laboratory, 1982.

[17] C. Willman, A. Hakansson, O. Osifo, A. Backlin, S.J. Svard, Nondestructive assay of spent nuclear fuel with gamma-ray spectroscopy, Ann. Nucl. Energy 33 (2006) 427.

[18] C. Willman, A. Hakansson, O. Osifo, A. Backlin, S.J. Svard, A nondestructive method for discriminating MOX fuel from LEU fuel for safeguards purposes, Ann. Nucl. Energy 33 (2006) 766.

[19] S. Caruso, M. Murphy, F. Jatuff, R. Chawla, Validation of ^{134}Cs, ^{137}Cs and ^{154}Eu single ratios as burnup monitors for ultra-high burnup UO$_2$ fuel, Ann. Nucl. Energy 34 (2007) 28.

[20] S. Caruso, I. Günther-Leopold, M.F. Murphy, F. Jatuff, R. Chawla, Comparison of optimised germanium gamma spectrometry and multicollector inductively coupled plasma mass spectrometry for the determination of ^{134}Cs, ^{137}Cs and ^{154}Eu single ratios in highly burnt UO$_2$, Nucl. Instrum. Methods Phys. Res. A 589 (3) (2008) 425.

[21] K.J. Park, J.S. Ju, J.S. Kim, H.S. Shin, Y.B. Chun, H.D. Kim, Determination of burnup and Pu/U ratio of PWR spent fuels by gamma-ray spectrometry, Nucl. Eng. Technol. 41 (10) (2009) 1307.

[22] I.C. Gauld, M.W. Francis, Investigation of Passive Gamma Spectroscopy to Verify Spent Nuclear Fuel Content, in: Proceedings of the 51st Annual Meeting of the Institute of Nuclear Materials Management (INMM), Baltimore, MD, 2010.

[23] C.T. Nguyen, I. Almási, Z. Hlavathy, J. Zsigrai, L. Lakosi, P. Nagy, T. Parkó, I. Pós, Monitoring burn-up of spent fuel assemblies by gamma spectrometry, IEEE Trans. Nucl. Sci. 60 (2) (2013) 1107.

[24] S. Matsuura, H. Tsuruta, T. Suzaki, H. Okashita, H. Umezawa, H. Natsume, Non-destructive gamma-ray spectrometry on spent fuels of a boiling water reactor, J. Nucl. Sci. Technol. 12 (1) (1975) 24.

[25] H. Natsume, H. Okashita, H. Umezawa, S. Okazaki, T. Suzuki, M. Ohnuki, T. Sonobe, Y. Nakahara, S. Ichikawa, S. Usuda, S. Matsuura, H. Tsuruta, T. Suzaki, T. Komori, S. Tamura, K. Gunji, K. Tamura, Gamma-ray spectrometry and chemical analysis data of JPDR-I spent fuel, J. Nucl. Sci. Technol. 14 (10) (1977) 745.

[26] H. Tsuruta, T. Suzaki, S. Matsuura, Profiles of activity ratios of fission products, Cesium-134, Cesium-137 and Ruthenium-106 in low-enriched PuO$_2$-UO$_2$ fuel assemblies, J. Nucl. Sci. Technol. 14 (1) (1977) 43.

[27] T. Suzaki, S. Okazaki, H. Okashita, T. Suzuki, N. Kohno, M. Ohnuki, N. Shinohara, T. Sonobe, A. Ohno, K. Murakami, H. Umezawa, H. Tsuruta, S. Matsuura, Y. Asakura, S. Araya, T. Yamanouchi, Non-destructive and destructive measurements on burn-up characteristics of Japan Power Demonstration Reactor-I full-core fuel assemblies, J. Nucl. Sci. Technol. 23 (1) (1986) 53.

[28] H. Ezure, Calculation of atom ratios of ^{134}Cs/^{137}Cs, ^{154}Eu/^{137}Cs and Pu/U, burn-up and most probable production amount of plutonium in fuel assemblies of JPDR-1, J. Nucl. Sci. Technol. 27 (6) (1990) 563.

[29] S. Svard, A. Hakansson, A. Backlin, O. Osifo, C. Willman, P. Jansson, Non-destructive experimental determination of the pin-power distribution in nuclear fuel assemblies, Nucl. Technol. 151 (1) (2005) 70.

[30] J.E. Fast, J.W. Chenault, B.D. Glasgow, D.C. Rodriguez, B.A. VanDevender, L.S. Wood, Spent Nuclear Fuel Measurements, PNNL- 23561, 2014.

[31] W.H. Yan, L.G. Zhang, Z. Zhang, Z.G. Xiao, Feasibility studies on the burnup measurement of fuel pebbles with HPGe gamma spectrometer, Nucl. Instrum. Methds Phys. Res. A 712 (2013) 130.

[32] W.H. Yan, L.G. Zhang, Z. Zhang, Y. Zhang, Z.G. Xiao, Prototype studies on the nondestructive online burnup determination for the modular pebble bed reactors, Nucl. Eng. Des. 267 (2014) 172.

[33] K.V. Vrinda Devi, T. Soreng, J.P. Panakkal, H.S. Kamath, A non destructive technique for determination of U^{233} and Pu content in MOX fuel pins for fast reactors, Nucl. Eng. Des. 241 (2011) 2783.

[34] H. Böck, J. Hammer, Gamma spectrometry of TRIGA fuel elements, Atomkernenerg. Kerntech. 39 (3) (1981) 195.

[35] T.K. Wang, J.J. Peir, An iterative approach for TRIGA fuel burn-up determination using nondestructive gamma-ray spectrometry, Appl. Rad. Isot. 52 (1) (2000) 105.

[36] R. Khan, S. Karimzadeh, H. Böck, TRIGA fuel burn-up calculations and its confirmation, Nucl. Eng. Des. 240 (2010) 1043.

[37] S. Karimzadeh, R. Khan, H. Böck, Gamma spectrometry inspection of TRIGA MARK II fuel using cesium isotopes, Nucl. Eng. Des. 241 (2011) 118.

[38] R.I. Dobrin, T. Craciunescu, M. Pavelescu, CANDU and TRIGA fuel burn-up determination using axial and tomographic gamma-scanning, Romaniam Rep. Phys. 63 (4) (2011) 1009.

[39] P.H. Liem, S. Amini, A.G. Hutagaol, T.M. Sembiring, Nondestructive burnup verification by gamma-ray spectroscopy of LEU silicide fuel plates irradiated in the RSG GAS multipurpose reactor, Ann. Nucl. Energy 56 (2013) 57.

[40] L.K. Pan, C.S. Tsao, Estimation of burn-up in Taiwan research reactor fuel pins by using nondestructive techniques, Nucl. Technol. 102 (3) (1993) 313.

[41] J. Navarro, R. Aryaeinejad, D.W. Nigg, A feasibility study to determine cooling time and burnup of ATR fuel using a nondestructive technique and three types of gamma-ray detectors, in: Proceedings of the 14th International Symposium on Reactor Dosimetry, INL/CON-11-22168, Bretton Woods, NH, 2011.

[42] B. Barnes, J. Phillips, G. Waterbury, J. Quintana, J. Netuschil, A. Murray, Characterization of irradiated nuclear fuels by precision gamma scanning, J. Nucl. Mater. 81 (1–2) (1979) 177.

[43] H.M.O.'D. Parker, M.J. Joyce, The use of ionising radiation to image nuclear fuel: a review, Prog. Nucl. Energy 85 (2015) 297.

[44] S.B. Ludwig, J.P. Renier, Standard- and extended-burnup PWR and BWR reactor models for the ORIGEN2 computer code, ORNL/TM-11018, 1989.

[45] IRR1, Safety analysis report IRR1, 2013.

[46] C.E.R.C.A., Fiche Suiveuse Element - Element Combustible, Ref EFN/173 AD-PH/MV, 1967.

[47] T. Makmal, Burnup evaluation in a MTR fuel element by gamma-ray spectrometry (M.Sc. thesis), Ben-Gurion University of the Negev, Israel, 2015.

[48] J.R. Phillips, G.R. Waterbury, N.E. Vanderborgh, T.K. Marshall, Quantitative determination of fission products in irradiated fuel pins using non-destructive gamma scanning, Anal. Chem. 47 (1) (1975) 71.

[49] Canberra Industries, Genie2000 V3.1 User's Manual, 2006.
[50] Canberra industries, In Situ Object Counting System (ISOCS) Geometry Composer Version 4.2, 2009.
[51] P. Kalb, Comparability of ISOCS Instrument in Characterization at Brookhaven National Laboratory, New York, 2000.
[52] Certificate of Measurement, Cobalt 60 Irradiation source, Atomic Energy of Canada Limited, Commercial Products, Ottawa, Canada, 1981.
[53] L.A. Terremoto, C.A. Zeituni, J.A. Perrotta, J.E.R. da Silva, Gamma-ray spectroscopy on irradiated MTR fuel elements, Nucl. Instrum. Methods Phys. Res. A 450 (1999) 495.
[54] JNDC, Nuclear Data Library of Fission Products, (Online). Available: ⟨http://wwwndc.jaea.go.jp/cgi-bin/nuclinfo2010?55,137⟩.