MEASUREMENTS OF IRR1 FUEL DEPLETION VALIDATING WHOLE-CORE MONTE-CARLO SIMULATIONS OF MORE THAN THIRTY YEARS OF OPERATION

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ABSTRACT

We developed and implemented a novel experimental method of measuring ^{235}U depletion in irradiated fuel assemblies and compare the measurement results to burnup-coupled Monte-Carlo core simulations. The new experimental method relies on gamma spectroscopic measurements, which were performed on irradiated HEU MTR fuel used in IRR1 (Israel Research Reactor 1, Soreq NRC). Using the new method, ^{235}U depletion was determined from the transparency increase of the fuel assemblies with increasing depletion to 155keV gamma's emitted from a Rhenium source situated beneath the fuel assembly. In addition, ^{137}Cs activity was determined by analyzing the obtained spectroscopic measurements. Comparison between the experiment and core burnup simulations of IRR1 revealed a very good match in the axial, as well as the total fuel depletion and the ^{137}Cs activity.

1. Introduction

Acquiring accurate knowledge of the fuel depletion in a research reactor is a challenging task. In particular, in the Israeli Research Reactor 1 (IRR1) in Soreq (a 5MW HEU MTR-type fuel reactor) the difficulty arises, among other reasons, due to the nature of irregular operation with varying core layouts and strong absorbers (control rods) introduced at the upper half of the core at specific core positions that changed throughout its operational history. Thus, the flux distribution was non-homogenous and the burnup of each fuel assembly (FA) is significantly affected by the specific irradiation history during the last ~35 years.
Within the auspices of IAEA's CRP T12029 ("Benchmarks of Computational Tools against Experimental Data on Fuel Burnup and Material Activation for Utilization, Operation and Safety Analysis of Research Reactors") an experimental campaign was conducted, in parallel to an intensive calculation effort, to both measure and calculate the fuel depletion and current ^{137}Cs activity in the different FA's of the IRR1 reactor.

The most common method for the experimental determination of MTR Research Reactor FA burnup is based on measuring the rate of gamma-rays emissions from long-lived fission products from the FA (e.g., ^{137}Cs, having $T_{1/2}$ = 30.1 years, and emits gamma's at 661.7keV) as described in [1-3]. However, this method has two main shortcomings:
1. It requires an accurate way of determining the detection efficiency of the germanium detector (i.e., calibration), which is needed for converting the measured count rate (CPS) to surface activity in units of Ci/cm^2. Efficiency calibration depends on the geometrical settings, the intrinsic efficiency of the detector and the self-absorption of the FA's. With that regard, we can't use the standard solution, which is to measure the ratio of activities for ^{137}Cs and ^{134}Cs.

This is because the irradiation time of the FAs in IRR1 is much longer then the $T_{1/2}$ of ^{134}Cs (2.06 years), such that one cannot accurately deduce the burnup from its activity measurement [4].

2. For some FAs, the irradiation in IRR1 even exceeds the $T_{1/2}$ of ^{137}Cs. Actually, for research reactors it is not uncommon to have FAs that were irradiated during long periods, or that removed and later re-entered to the core during different periods, in the order of years, or even decades, thus the translation from ^{137}Cs activity to burnup requires the knowledge of each FA's history in order to determine the actual burnup.

In order to deal with these two shortcomings, we developed and implemented an innovative method, in which the ^{235}U content was determined using the attenuation of low-energy gamma ray from an external source, which passed through the FA. Since the attenuation of gamma's through Uranium is more significant than through its fission products (see Fig. 1), the higher the FA's depletion is, the more "transparent" it becomes.

The experimental activity involved two independent measurements, of ^{137}Cs activity and of ^{235}U depletion (by the new gamma transparency method) on five FA's that were previously irradiated in the reactor. The measurements were done along the central axis of the FA's with 20 measurement points per FA, thus obtaining the axial depletion profile, as well as an integral value for the depletion of the entire FA.

In parallel to the experimental campaign, a computational effort was made to calculate the burnup of the last 30 years history of IRR1, by using a coupled MCNP-DRAGON code (named "MUTZAV") with a detailed 3D model of the cores. For each of the 180 core configurations in IRR1's history, MCNP [5] was used to solve for the neutron flux in each burnup region, and consequently running DRAGON's [6] depletion module to calculate the burnup of the neutronically-important nuclides (actinides, fission products, poisons and structural components) and the ^{137}Cs content in the fuel meat slab. Following this step, the material composition for each meat burnup zone in the FAs was updated and used as a starting point for the next repeated calculation with the consequent core configuration [7].

We used this model to calculate the measured quantities for comparison. Overall, we found a very good agreement between the calculated quantities and the measured ones, both for the ^{235}U depletion and for the ^{137}Cs activity. Thus, both the new burnup measurement method and the MUTZAV calculation were validated.

This paper is organized as follows: Chapter 2 describes the experimental method used to determine the fuel depletion and ^{137}Cs activity in the FAs. In Chapter 3 describes the computational code "MUTZAV" and the 3D model which was used to deplete the neutronically-important nuclides of IRR1 core from 1980 until mid-2017. Chapter 4 presents the main results and comparison between simulations and experiment. Finally, Chapter 5 presents a brief summary and the main conclusions of this work.

2. Measurements of Depletion and ^{137}Cs Activity in IRR1 FA's

We required a gamma source that has high activity and a suitable gamma emission energy. The chosen external source was a natural Rhenium target irradiated in the IRR1, which generated ^{188}Re (from ^{187}Re (n,g) reaction) that emits 155 keV and 633 keV gamma rays (among others). To reduce systematic uncertainties, we measured the gamma transparency of two reference FAs with known Uranium content, namely, a Fresh FA (zero depletion); and a dummy FA (similar geometry, but without Uranium).

When an interrogated FA with unknown depletion is measured, the gamma intensity of the Rhenium source is attenuated to an intermediate degree between those in the measurements

of the fresh FA and a dummy FA. We have shown [6] that the intensity of measured 155keV gamma's through a depleted FA can be directly related to the actual depletion:

$$dep = c \cdot \frac{ln\left(\frac{I_{dep}}{I_{fre}}\right)}{ln\left(\frac{I_{nofuel}}{I_{fresh}}\right)} \quad , \tag{1}$$

where $I_{dep}, I_{fresh}, I_{nofuel}$ are the 155keV gamma intensities measured at the detector for the interrogated FA, a fresh FA and a dummy FA, respectively, and with $c \triangleq \left(\eta \frac{\sigma_f}{\sigma_a}\left(1 - \frac{\mu_{FP}}{\mu_U}\right)\right)^{-1}$ where $\eta, \sigma_f, \sigma_a, \mu_U$ and μ_{FP} are the fresh fuel enrichment, the microscopic fission and absorption cross sections and gamma attenuation coefficients of Uranium and an average fission product (determined by the weighted average of attenuation coefficients over the cumulative fission products distribution of thermal ^{235}U fission), respectively. For the specific case of IRR1 fuel we obtain $c = 1.70 \pm 0.01$. Finally, we note that the ^{235}U density is deduced from the measured depletion by $\rho_{U235} = \rho_{U23}^{Fresh}(1 - dep)$ where the mass density of the fresh FA is $\rho_{U235}^{Fresh} = 0.67 \pm 0.03\ \mathrm{g/cm^3}$.

The average depletion in the whole FA is determined by

$$\overline{dep} = \frac{\int_{z_i}^{z_f} dep(z)dz}{z_f - z_i} \cong \frac{\sum_{j=1}^{N-1} dep(z_j)(z_{j+1} - z_j)}{z_f - z_i} \quad , \tag{2}$$

Where z is the axial position along the fuel assembly, measured at *N* positions.

^{137}Cs activity was determined from the Net Count Rate of 662keV photons in the detector as follows:

$$A_{Cs137} = \frac{I_{662keV}}{\varepsilon Y} \quad , \tag{3}$$

Where $I_{662keV}, Y, \varepsilon$ are the Net Count Rate, the gamma emission probability and detection efficiency, respectively. The efficiency includes the geometrical efficiency and the detector inner efficiency. In this study, efficiency at 662keV (for ^{137}Cs gamma's) was determined experimentally using a 633keV gamma's emitted from a ^{188}Re source with a known activity (to within ±1%) and then corrected for the difference (correction of ~5% only) between the two gamma energies using a Monte-Carlo simulation of the experimental setup that included the detector (see [8] for detailed description).

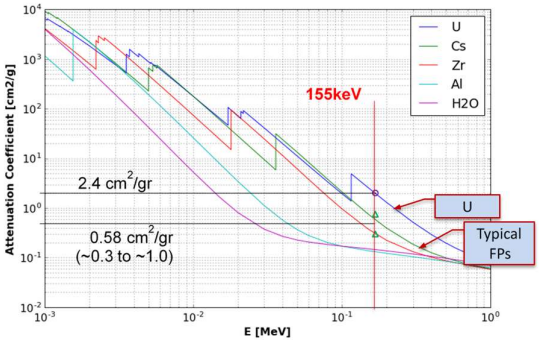

Figure 1. Attenuation coefficient as a function of gamma energy (values obtained from NIST Standard Reference Database: http://www.nist.gov/pml/xcom-photon-cross-sections-database) for the different materials existing in a Fuel Assembly (U, Al, H_2O, typical fission products). The relatively high-contrast of attenuation between U and typical Fissions Products, FPs (Cs, Zr) is shown for 155keV gamma ray.

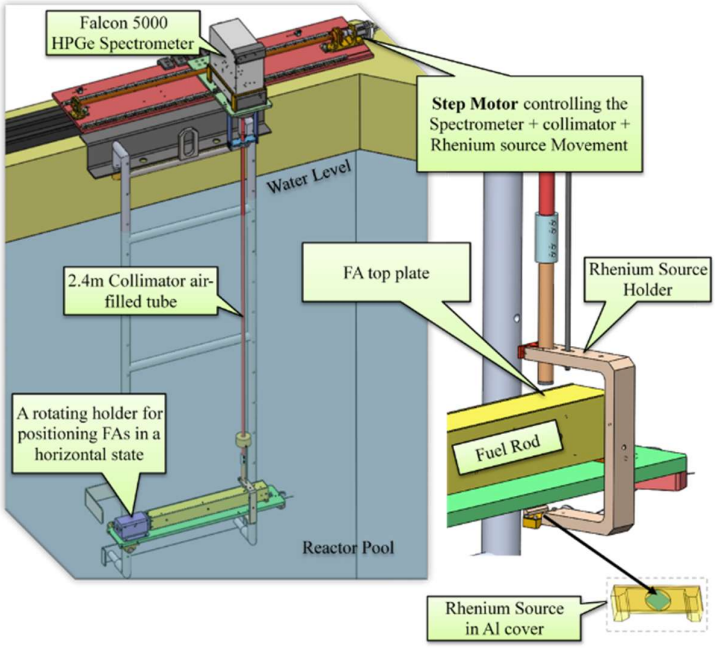

Figure 2. Experimental setup located in the reactor pool. The setup consists of a mechanism to position fuel rods horizontally and a sub-assembly with HPGe detector aligned with an air-filled collimator and an activated Rhenium source (^{188}Re) beneath the fuel rod. This entire sub-assembly is mounted on linear movers with a rotating shaft connected to a step motor controlling the position relative to the fuel rod. The relative positions of the collimator along the fuel rod were known within 0.1 cm

We performed simultaneous measurements of both ^{137}Cs activity as well as gamma-ray FA transparency as depicted in Figure 2, thus obtaining the two independent measurement at the same time. The experimental uncertainty was below 5% for ^{137}Cs activity measurements, and about 2% in the determination of the total depletion using the gamma transparency method. It is worth noting that the uncertainty in ^{137}Cs activity is mainly systematic (calibration, attenuation correction) while with the gamma transparency method it is mainly statistical (typically 5% statistical uncertainty of the net count rate at each data point). Thus, the accuracy of this new method can be further improved simply by obtaining longer measurements, without changing anything in the experiment itself.

3. Description of MUTZAV (Coupled MCNP-DRAGON code)

A coupling between MCNP-4B and DRAGON's depletion module was developed using PYTHON to improve the computational accuracy of the IRR1 operation properties that are related to the fuel burn-up. The Monte-Carlo simulation generates 69-group fluxes for each depletion zone (in our case, an IRR1 fuel rod is typically split into 15 depletion zones, where only the fuel meat is depleted in the simulations). Each zone is then depleted in DRAGON using WLUP (WIMS Library Update Project) 69-group cross-sections and depletion chains, for a total of 91 nuclides (23 actinides, 48 fission products and 20 burnable absorbers). The isotopic compositions of each zone in the MCNP core file are updated accordingly and the next Monte-Carlo simulation is performed. This code was first validated against similar research reactors (IAEA MTR, and Ohio-State University Research Reactor), using both code-to-code as well as code-to-experiment comparisons. [7].

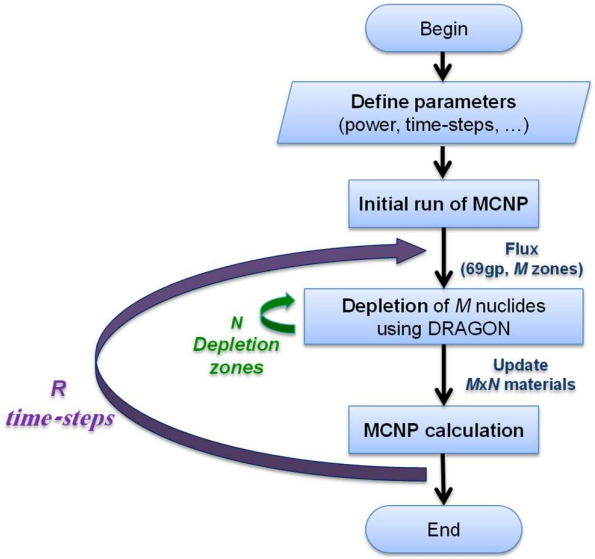

Figure 3. Computational scheme description of the coupled code MUTZAV

An IRR1 MCNP input file was created. Each FA consisted of 5 axial x 3 depletion zones (2 for side plates and one region for the central 21 plates) and described schematically in Figure 4. Using an automated Python script, 180 core layouts of IRR1 (described in detail in CRP T12029 [9], and illustrated schematically in Figure 5) were then burnt sequentially with 1 million neutron histories per burnup cycle, assuming constant 5MW power and absorber blades partially inserted at a fixed height of 75% (which was their typical position during the last 30 years, however with significant variations. Thus, this is an approximation). The total burnup during this period was 97.6 GWh.

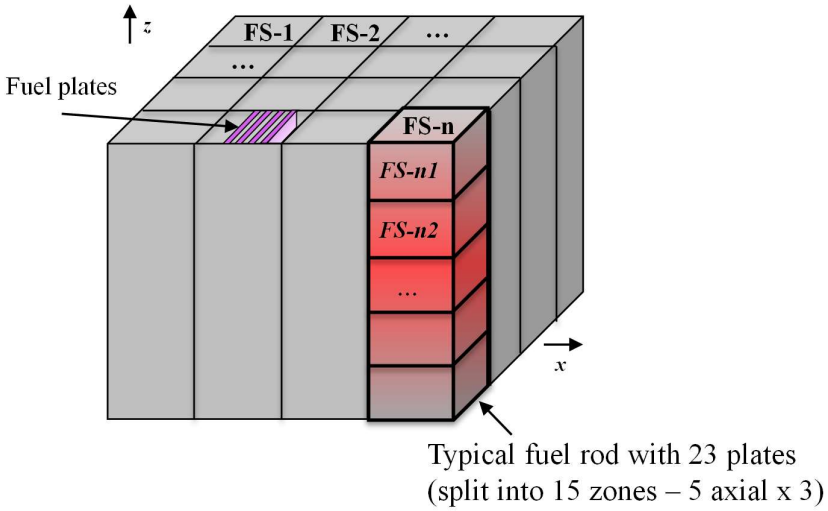

Figure 4. Schematic whole-core three-dimensional discretization of IRR1 FAs

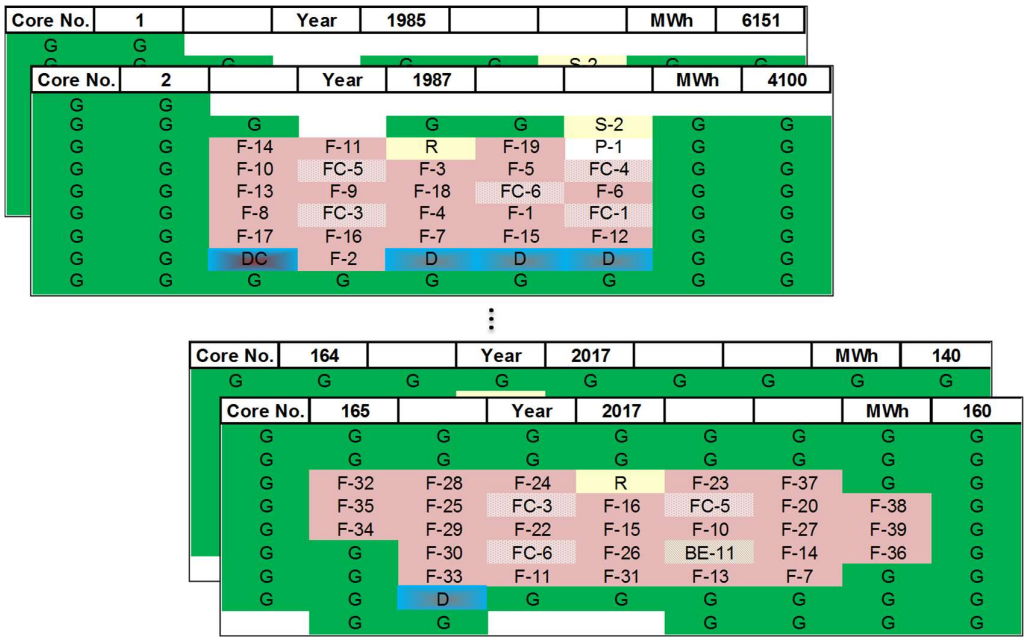

Figure 5. Examples of IRR1 computed cores. F and FC denote a standard FA and special FA's (i.e., partial fuel assembly containing control rods), BE-11 is a Beryllium rod with control blades, G is graphite and R is the regulating rod. D, DC are irradiation sites

4. Results

In this chapter we put together both the results obtained using the experimental methods as well as the MUTZAV simulations, in order to benchmark the accuracy of new core calculation and also to evaluate the accuracy of the commonly used approximation in estimating the ^{235}U depletion from ^{137}Cs activity measurement.

4.1. Experimental Data Analysis

For each of the five FAs measured (see Table I), 20 spectra measurements were obtained at discrete positions along the axial center of the FA. A typical gamma spectrum is presented in Figure 6. It can be seen that the typical long-lived FPs (Fission Products) are observed, as well as the gammas emitted from the Rhenium source positioned below the FA (137keV of ^{186}Re and 155keV of ^{188}Re) and the long-lived activation products (604keV from ^{134}Cs and 1274keV from ^{154}Eu). We focus our attenuation only on 662keV and 155keV gammas, allowing us to compute the ^{137}Cs activity and the depletion with the gamma transparency method, respectively.

Table I. List of Measured Fuel Assemblies with initial fissile mass and first and last irradiation dates

FA	Initial U235 mass [g]	First Irradiation	Last Irradiation
FS-2	280.5	1975	5/1990
FS-5	281.4	1976	2/2011
FS-17	282.6	1984	5/3/2017
FS-18	282.2	1984	10/2013
FS-19	282.1	1987	20/10/2016

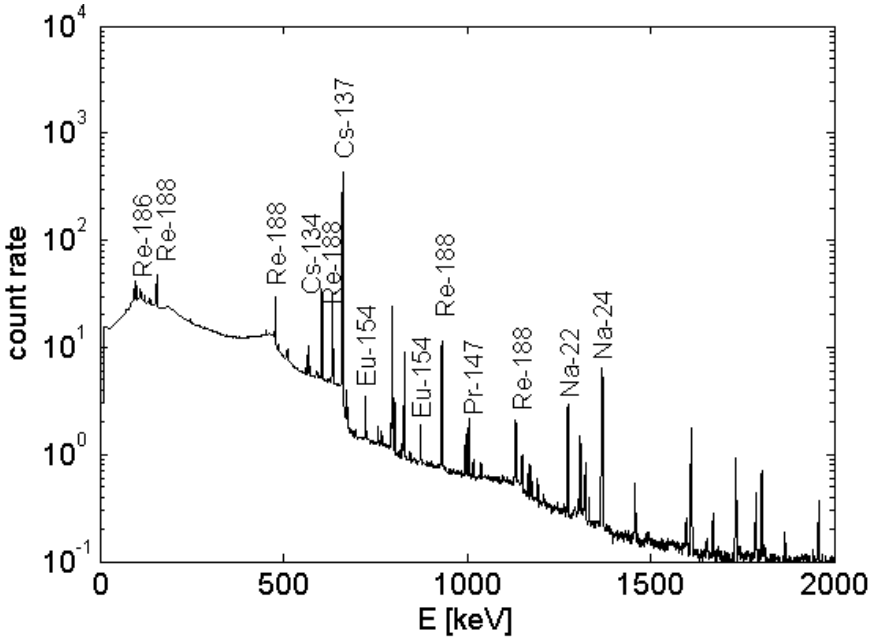

Figure 6. Example of a spectroscopic measurement in the middle of FS-18.

Using equations (1) and (3), we obtained the local values of the depletion (using gamma transparency method) and the ^{137}Cs activity per cm^2, respectively, for each of the 5 FA's. Typical axial profiles are depicted in Figure 7. It is evident that the axial distribution of ^{137}Cs represents quite accurately the true fuel depletion. This fact is far from being trivial (as it is a comparison between two completely independent measurements). Furthermore, as the control blades are inserted from the upper part of the reactor, the flux is pushed downward and we indeed see higher depletion (and Cesium activity) at the lower half of the FAs. It is also interesting to observe the increased burnup effect (higher ^{137}Cs activity and increased gamma transparency) near the fuel edges (z < 4 and 54 < z) caused by the higher moderator to fuel ratio. Further outside of the active region of the plate there are neither ^{137}Cs signal, while the Rhenium signal increases due to lack of ^{235}U attenuation, Hence the ^{137}Cs activity as resulted from equation (3) goes to zero there, while the depletion resulted from equations (1) goes dramatically up.

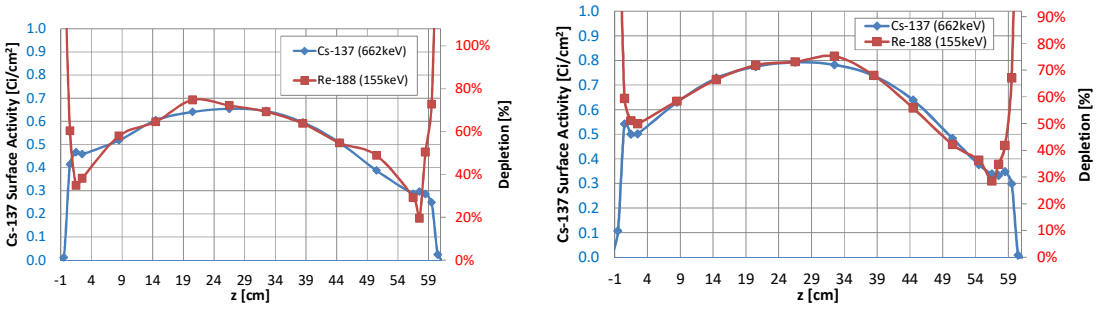

Figure 7. Axial distribution of ^{137}Cs Activity (per surface area) and Depletion, using the gamma transparency method for FS-5 (Left) and FS-18 (Right). The secondary y-axes ranges were adjusted so that the mean depletion and mean ^{137}Cs activity yield the same height ($y_{2max} = \overline{A_{Cs137}}/\overline{Dep}$). Note that beyond z < 0 cm and z > 59 cm there was no Uranium in the FA, thus ^{137}Cs Activity goes to zero and depletion "explodes" (since gamma transparency through the Aluminium is much higher).

4.2. Comparison between Experiment and Simulations

We now focus our attention on comparison between the experimental data and the MUTZAV simulations: both for the FAs ^{235}U mass content and for the ^{137}Cs activity.
In Figure 8 we present the current ^{235}U mass in each of the measured FA's. The experimental results and the results for the simulation match well for all FA's, within their uncertainties. As we obtained local depletion measurements and computed depletion along 5 axial regions for each FA, a detailed comparison is presented in Figure 9 for two of the measured FA's (the other measured FA's gave similar results). Indeed, there is a good agreement in the axial depletion profiles, suggesting that the approximation of calculating the burn up with constant height of the control rods in the MUTZAV simulations (75% height) is reasonable.

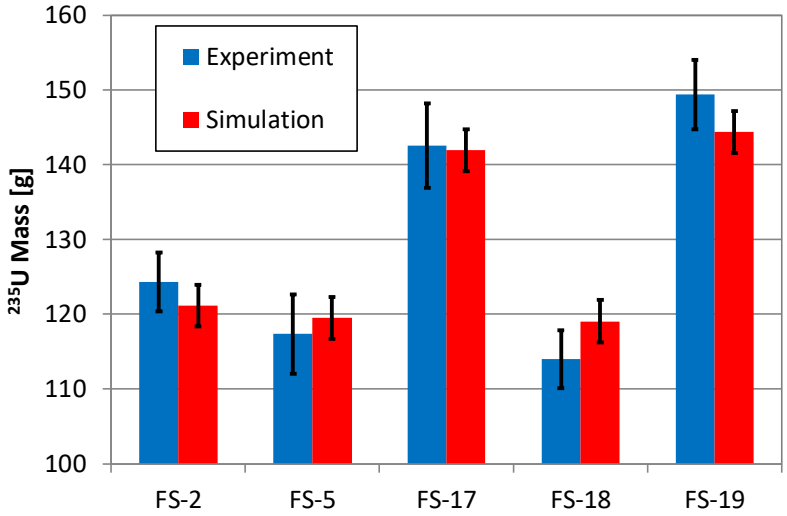

Figure 8. ^{235}U mass in each FA. Comparison between experiment (gamma transparency method) and MUTZAV simulation of IRR1 history until mid-2017.

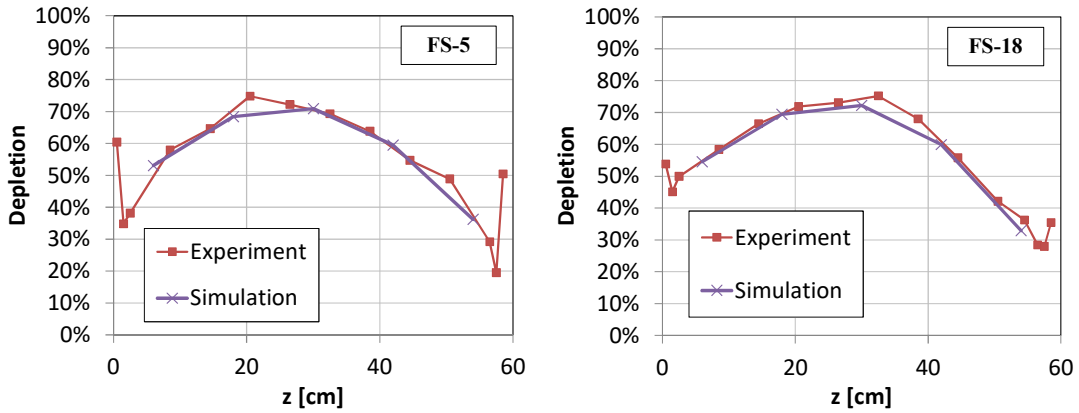

Figure 9. Axial ^{235}U depletion distribution within two FA's. Comparison between experiment (gamma transparency method) and MUTZAV simulation of IRR1 history until mid-2017. Note that near z ~ 0 and 60 cm, the experimental measurement of the depletion appears to increase due to two effects: higher moderator/fuel ratio causes increased local flux, and initial Uranium density that is not homogeneous near the meat edges.

The total ^{137}Cs activity that was measured in the FA's was compared with the simulations as can be seen in Figure 10. Again, the results match well, with a slightly lower measured activity compared to the simulations (by about 3%, on average) suggesting perhaps a systematic bias caused by the 662keV detection efficiency calibration process.

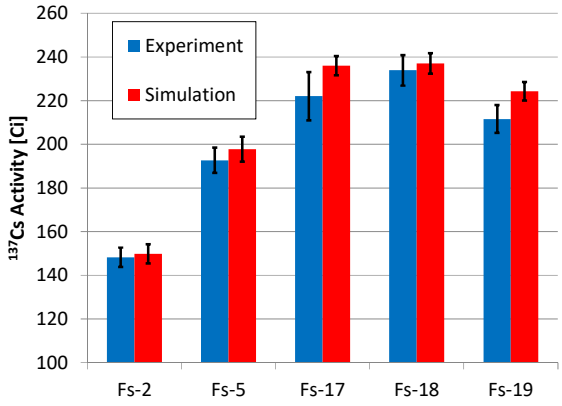

Figure 10. Total ^{137}Cs activity in each FA. Comparison between experiment (662keV gamma's) and MUTZAV simulation of IRR1 history until mid-2017.

4.3. Investigation of ^{137}Cs-based depletion estimation

We now turn our attention to the often-used method of depletion estimation from ^{137}Cs activity. This method is promptly employed for research reactors [3, 4], and it assumes uniform power distribution in each core, thus eliminating the need for neutronic simulations. Under this assumption, the depletion is given by:

$$\widetilde{dep} = \frac{\sigma_a}{\sigma_f}\frac{A_{\mathrm{Cs137}}\, e^{\lambda t_c}}{Y\, N_0} f \ , \qquad (4)$$

Where A_{Cs137} is the activity of Cs-137 in the fuel, λ is the decay constant of Cs-137 ($T_{1/2} = 30.1\ years$), t_c is the time interval between the end of the last irradiation period of the fuel element and the gamma-ray spectroscopy measurements, Y is the fission yield of ^{137}Cs (6.26%), N_0 is the atomic density of ^{235}U for fresh fuel. $f = \frac{\sum_{i=1}^{n} P_i t_i}{\sum_{i=1}^{n} P_i e^{-\lambda\tau_i}\left(1-e^{-\lambda t_i}\right)}$ is known as the Bibichev correction factor, which takes into account the decay of Cs-137 since it was created from fissions until measured (P_i is the average relative power corresponding to the *i*th irradiation period, *n* is the total number of irradiation periods during the whole irradiation history of the fuel element, t_i is the duration of the ith irradiation period and τ_i is the time interval between the end of the ith irradiation period and the end of the last irradiation period).

It can be seen below in Table II that for our particular case, employing this method yields reasonable results in the depletion calculation, with discrepancies between 3 and 12% in the absolute depletion (while the measurement uncertainty is less than 5%). The reason for the discrepancies lies in the calculation of the Bibichev correction factor, which assumes uniform depletion rate for each FA in all cores. For IRR1, since most of the measured FA's were originally introduced near the core's center and moved toward the periphery near their end-of-life, this created lower depletion rates and thus lower ^{137}Cs production rate for these FA's in later cores (lower flux rates). Overall, this created a trend of depletion under-estimation as is seen in the last column of Table II.

Table II. Comparison between exact depletion measurement and the commonly used Cs-137–based depletion approximation

FA	^{137}Cs Activity	Bibichev factor	Cs-based estimated Depletion[1]	Measured Depletion	Depletion Difference (Cs_based - Measured)
FS-2	148	2.20	44%	56%	-12%
FS-5	193	1.92	50%	58%	-8%
FS-17	222	1.53	46%	49%	-3%
FS-18	234	1.60	51%	60%	-9%
FS-19	212	1.49	43%	47%	-4%

5. CONCLUSIONS

This paper presented spectroscopic measurements of IRR1's MTR fuel, with analysis of the ^{137}Cs activity and of the fuel depletion. The latter was determined by a newly developed technique - based on measurement of gamma transparency. We measured five FAs that were previously irradiated in the IRR1 reactor. The gamma transparency method (described in Chapter 2) required also the measurement of the attenuation through a fresh FA and a dummy FA, to accurately determine the current Uranium content in the fuel and thus, the depletion. This method has the advantage of using the available neutron flux in the reactor itself to irradiate the source (Rh), making the method feasible for usage in many other reactors.
Each FA was measured at 20 axial points, thus obtaining the depletion profile as well as an integral value of the entire FA. The overall uncertainty for the determined depletion for all FAs was below 5%.
In parallel, a computational effort was undertaken to simulate the depletion of 180 core layouts of IRR1 using MCNP coupled with DRAGON's depletion module. The cores were modelled to contain 5 axial zones for each FA and burnt according to the reactor's history description in the past 37 years at a constant 5MW power rate (without taking into account in details the irregular operational history within of each cycle, and the varying of control rods heights) with a total burnup of about 100 GWh.
Comparison between the simulations and experiment revealed a very good match in the total fuel depletion as well as in the ^{137}Cs activity in all 5 FA's. Furthermore, comparison between the axial distributions of the depletion showed that the constant height assumption was enough to give satisfactory results.

Finally, we checked the often-employed ^{137}Cs-based depletion approximation for the five measured FA's and found its accuracy to be within 12% (absolute depletion), which is primarily due to the inaccuracy of the assumption of uniform power per FA that is used in this approximation.
The results pave the way for future work that will make our developments more ubiquitous as tools for fuel control, management and inventory verification.

ACKNOWLEDGMENTS

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[1] Using eq. (4)

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