CANDU AND TRIGA FUEL BURN-UP DETERMINATION USING AXIAL AND TOMOGRAPHIC GAMMA-SCANNING[*]

R. I. DOBRIN[1], T. CRACIUNESCU[2], M. PAVELESCU[3]

[1]Institute for Nuclear Research Pitesti, P.O. Box 78, Campului Str., 1, Mioveni, Romania,
E-mail: relu.dobrin@nuclear.ro
[2]National Institute for Lasers, Plasma and Radiation Physics, Bucharest, Romania,
E-mail: teddy@infim.ro
[3]Academy of Romanian Scientists, Bucharest, Romania,
E-mail: mpavelescu2002@yahoo.com

Abstract. Sufficiently accurate determination of fuel burn-up in power reactors is indispensable in optimizing fuel management for reasons related to both safety and economy. The Post-Irradiation Examination Laboratory (PIEL) from INR Pitesti is the only Romanian laboratory with hot cells and the capability to perform direct burn-up measurements of spent fuel rods. The most accurately approaches for burn-up determination consist in destructive radiochemical methods, but this means a rather costly and time consuming chemistry process. More attractive are the non-destructive gamma spectrometric methods, which are fast, cheap, and preserve the fuel integrity. The paper shows the methodologies used at PIEL for burn-up determination of more then one hundred of TRIGA HEU, TRIGA LEU and CANDU type fuel rods, using axial and radial gamma-scanning methods. Also, a computer program developed at PIEL for fast burn-up evaluation and an experimental result are presented in some details.

Key words: fuel rod, burn-up evaluation, gamma scanning, tomographic reconstruction.

1. INTRODUCTION

During irradiation in a nuclear power plant, the composition and physical characteristics of the fuel change continuously from its initial physical properties, and this has important consequences on the fuel cycle. At the end of its life in the reactor, the fuel needs to be characterized as accurately as possible for reasons related to both safety and economy. The main investigations carried out are oriented towards verifying the fuel cladding integrity and determining the fissile content and the fuel burn-up (BU), the latter being an indicator of the fuel cycle efficiency. The burn-up of nuclear fuel is related to the lifetime of fuel in the core

and it is given by the total amount of thermal energy that it has produced per unit of fissile material in the reactor. The common unit used to measure the burn-up is MWh/kg or GWd/t. Another definition of burn-up is the number of fissions per 100 heavy nuclide atoms initially present in the fuel, expressed in percentage. Being related to the inventory of fission products (FPs) formed in the matrix of the fuel, the burn-up is usually determined by measuring the content of an element that results from the fission process. The most accurately approaches for burn-up determination consist in destructive radiochemical methods, but this means a rather costly and time consuming chemistry process. For this reason, the non-destructive gamma spectrometric methods are more attractive, because they are fast, cheap, and preserve the fuel integrity. Moreover, additional information about behavior of the irradiated fuel, related to axial and radial distribution of the FPs activity, the migration of volatile FPs inside the fuel rod, fuel cladding integrity etc. are obtained using axial and tomographic gamma-scanning methods.

2. METHODS

As defined above, the burn-up of an irradiated fuel rod can be expressed as:

$$\mathrm{BU} = N_f \cdot \frac{E_f}{m_U}, \tag{1}$$

where: N_f is total number of fissions, E_f is energy release per fission and m_U is metallic uranium mass in stack fuel rod.

Ideally the result can be obtained by measuring the amount of fissile material left in the fuel when the initial enrichment of the fuel is known. Practically the procedure is not so easy.

When a fuel is irradiated in a reactor, the abundances of heavy element isotopes change as a result of neutron capture and fission. From these changes, using the equations of evolution of heavy metals implied in the irradiation process, one can deduce the number of fissions that have taken place (for TRIGA and CANDU-type fuel rods the main heavy metals are: ^{235}U, ^{238}U and ^{239}Pu).

The evolution of a gamma-radioactive fission product X at any time during the irradiation can be expressed approximately by the following equation:

$$\frac{\mathrm{d}\,_Z^A X}{\mathrm{d}t} = y_{_Z^A X} \cdot \sigma_{f(HM)} \cdot \Phi \cdot N_{HM} + \sigma_{c(_Z^{A-1}X)} \cdot \Phi \cdot {}_Z^{A-1}X - (\lambda_{_Z^A X} + \sigma_{c(_Z^A X)} \cdot \Phi) \cdot {}_Z^A X, \tag{2}$$

where: N_{HM} is the number of heavy metals nuclides in the volume of the irradiated fuel rod, Φ is the neutron flux, y_X is the fission yield of the fission product considering that it is approximately the same from each type of fissile nuclides, $\sigma_{f(HM)}$ is the fission cross section of the fissile nuclides, λ is the decay constant of

the considered fission product, $\sigma_{c\left(_{Z}^{A}X\right)}$ and $\sigma_{c\left(_{Z}^{A-1}X\right)}$ are the neutron capture cross sections of the fission product and respectively its capture precursor.

Usually a fuel rod is irradiated in irradiation steps. Each step is characterized by specific values of the neutron flux and time of irradiation. The total sequence of active and passive irradiation steps forms the reactor's operating history. In principle, using Eq. 2 for each irradiation step, the number of fissions and consequently fuel burn-up can be calculated. Unfortunately the actual values of the neutron flux generally are not available. Usually only values proportional to the real ones can be obtained from the reactor records:

$$F = a \cdot \Phi .   \tag{3}$$

In order to compensate this lack of information gamma spectrometric analysis of some specific FPs (so-called burn-up monitors) can be used and burn-up determination can be performed indirectly using their spontaneous gamma-rays emission.

Generally, gamma spectrometry is a non-destructive, rapid, and low cost method that serves to determine the activity of all emitting fission products still present in a fuel rod at the time of measurement. It requires the calibration of the gamma spectrometric installation in absolute values, which means the determination of the efficiency as a function of energy for given experimental conditions, using adequate gamma sources. Also, it is necessary to determine the transmission phenomena of the gamma radiation emitted, that means that the attenuation and self-attenuation properties of the fuel and the structures surrounding the fuel have to be determined.

A fission product can be used as a quantitative measure of burn-up, if the following conditions are met [1]:

– The fission product should be characterized by near-to equal fission yields for the major uranium and plutonium fissile nuclides. If the yields are substantially different, the effective fission yield will depend on the reactor's operating history.

– The neutron capture cross section of the fission product must be low enough so that the observed fission product concentration is due only to heavy element fission and not to secondary neutron capture reactions.

– The fission product half-life should be long compared to the fuel irradiation time, so that the quantity of fission products present is approximately proportional to the number of fissions.

– The fission product gamma rays must be of relatively high energy (proper above 500 keV) so as to be able to escape to a useful degree from the fuel rod.

^{137}Cs is the most widely accepted as BU monitor since it adequately satisfies all of above conditions [2, 3]. ^{137}Cs has a long half life time (30.09 years) as compared to the irradiation times (usually max. few years). Therefore, its decay plays only a minor role for long irradiation times and can be easily corrected using

simple mathematics [3]. In addition, the cross section of ^{137}Cs for neutron capture (n,γ) is very small with a value of about 0.25 barns and, thus, this neutron reaction is negligible. The generation of ^{137}Cs from ^{136}Xe through neutron capture and subsequently γ-decay can be neglected likewise since the cross section for this neutron reaction of about 0.23 barns is also very small. Therefore, generation and decay of ^{137}Cs in the reactor is almost only determined by the fission of ^{235}U and ^{239}Pu and behaves linearly as a function of the burn-up. The fission yield of ^{137}Cs is relative high (6.3·10^{-2}) and approximately the same for each fissile heavy metal isotope and is hence representative of the total number of fissions. Finally, another rationale to use ^{137}Cs as a burn-up monitor is that its short living daughter nuclide ^{137m}Ba has a very clear gamma peak at 661.66 keV, which can easily be identified by gamma spectrometry; due to the short half life time of ^{137m}Ba, the activity of ^{137m}Ba and ^{137}Cs is proportional. In this case, the follow simple relationship for burn-up calculation can be used:

$$\mathrm{BU} = \frac{N_{Cs137}}{\gamma_{Cs137}} \cdot \frac{E_f}{m_U}, \tag{4}$$

where: N_{Cs137} is the total number of ^{137}Cs nuclides in the fuel (experimental measured) and γ_{Cs137} is the fission yield of the ^{137}Cs from the fissile heavy metals in the fuel.

The last equation indicates a linear relationship between the burn-up and the concentration of the isotope ^{137}Cs. More precisely, including and the case of long irradiation times, the relationship between BU and N_{Cs137} is [3]:

$$\mathrm{BU} = \frac{N_{Cs137}(t_{irr})}{\gamma_{Cs137} \cdot N_{HM}} \cdot C_{corr}(t_{irr}), \tag{5}$$

where: N_{HM} is the number of heavy metals atoms, t_{irr} is the irradiation time and C_{corr} is the correction term:

$$C_{corr} = t_{irr} \cdot \lambda_{Cs137} \cdot \left(1 - \mathrm{e}^{(-\lambda_{Cs137} \cdot t_{irr})}\right)^{-1}. \tag{6}$$

Due to the relatively long half-life of ^{137}Cs, this correction term is small for short irradiation times (*i.e.* for an irradiation time of 3 years, the correction amounts to about 3%), but its contribution is more significant for relatively long irradiation times. In this case, using ^{137}Cs as burn-up monitor, the general equation of FPs evolution (Eq. 2) becomes more adequate for burn-up calculation. Also, in case of shorter cooling times, the total gamma-ray activity is dominated by the short-lived radionuclides (^{103}Ru ($T_{1/2}$=39.35d), ^{95}Zr (64.02d) – ^{95}Nb (35.06d), ^{140}Ba (12.8d) – ^{140}La (40.22h), ^{144}Ce (284.3d) – ^{144}Pr (17.28min) etc.) activities of which depend mostly on the reactor operating history and hence the Eq. 2 must be used for a correct burn-up determination.

For a fast burn-up evaluation in any of the above described situations a computer program (called EVOLUT) has been developed at PIEL. The input data

consists on the one hand of the experimental evaluation of the number of nucleus of a fission product (burn-up monitor) X_{exp} at the end of the irradiation and on the other hand of the history of irradiation (the time length and relative value of the neutron flux for each step of irradiation). Using the irradiation history (the relative values F are used instead of the real flux values Φ) Eq. 2 allow the calculation of the nucleus number X_{calc}. The constant a (see Eq. 3) is iteratively adjusted until $X_{calc} = X_{exp}$ and in this way, using Eq. 3, the real flux values Φ can be calculated for the whole irradiation history. Then they can be used to determine the total number of fissions and consequently the burn-up. The values Φ are used also in Eq. 2 to calculate the evolution of each fission product during the irradiation.

EVOLUT is able to deal with several burn-up monitors: ^{137}Cs, $^{134}Cs/^{137}Cs$, ^{103}Ru, $^{95}Zr-^{95}Nb$, $^{140}Ba-^{140}La$ and $^{144}Ce-^{144}Pr$.

3. EXPERIMENTAL

For experimental determination of the number of nucleus of fission products selected as burn-up monitors the well known gamma-scanning technique [4] is used at PIEL (INR – Pitesti). This is a non-destructive, rapid and low cost method to determine the axial profile of the FPs activity along the fuel rod and was previously described [5]. The gamma scanning system consists of a vertical fuel rod-positioning machine equipped with step-by-step motors, a collimator set in the hot cell shielding wall, a HPGe detector, a multichannel analyzer with a 200 MHz ADC and 4096 channel memory and a control desk equipped with digitally pulse counters to display the fuel rod position. The collimator includes three rectangular slits having the width of 50 mm and the aperture size of 0.1 mm, 0.25 mm and 0.5 mm. The slits are made of tungsten alloy and their position can be horizontal or vertical.

The fuel rod is axially scanned using a horizontally slit having a certain aperture (usually 0.5 mm) and a step of fuel rod movement of the same size and the isotopic gamma activity profiles are registered along the fuel rod.

A ^{124}Sb source prepared in the laboratory, whose gamma spectrum is composed of many lines, serves to give the relative detection efficiency and a certified ^{137}Cs source (273 mCi ±4.5% to 18.08.1980) is used for cross-checking of the absolute value.

The large value of the density of the nuclear fuel involves the absorption of a significant number of emitted photons. So a correct estimation of the self-absorption coefficient (K_a), defined as the ratio between the number of photons which are not absorbed in the fuel and the total number of emitted photons is essential for an accurate gamma activity calculation. The self-absorption coefficient can be calculated if the shape of the distribution of the fission product in the cross-section of the fuel rod is known: beginning from the source, which is

described by the distribution of the fission product, a large number of photons are followed up, individually, simulating their interaction with the material. The shape of the distribution is achieved, from radial gamma-scanning measurements, using a computed tomography approach based on a maximum entropy algorithm [6]. Otherwise, the tomography proved to be one of the most reliable and efficient tool for the determination of isotope distribution in irradiated fuel rods [7, 8]. Practically, a radial profile is obtained moving the fuel rod transversally (step-by-step at a regular interval of the same size as the aperture of the slit) in front of a vertical slit (usually 0.25 mm) of the collimator (the entire diameter of the rod is covered). The measurement is repeated for five equidistant projection angles (72^0) and a reconstruction grid of about 50×50 pixels is obtained.

Thus, using axial and radial gamma-scanning profiles the activity and consequently the nucleus number of a fission product X_{exp} at the end of the irradiation can be experimental determined. Considering an adequate gamma-line at energy E, X_{exp} is calculated as:

$$X_{\exp}{}^{E} = \frac{\sum A_i^{E}}{\lambda^{E} \cdot s^{E} \cdot t_m \cdot \varepsilon^{E} \cdot K_a{}^{E} \cdot e^{-\lambda \cdot t_d}}, \qquad (7)$$

where: $\sum A_i$ is the total net counts in the fuel rod (determined from the axial gamma-scanning profile), λ is the decay constant, s is the gamma emission probability, t_m is the measurement live time, ε is the detection efficiency, K_a is the self-absorption coefficient and t_c is the cooling time (elapsed time between end of irradiation and the moment when the gamma spectrometric analysis was performed).

4. RESULTS AND DISCUSSIONS

EVOLUT was successfully used in the analysis of more then one hundred of CANDU-type, TRIGA-HEU and TRIGA-LEU fuel rods at PIEL. The uncertainty of the results is under 10% (it must be mentioned that only the uncertainty of the certified ^{137}Cs gamma standard source used for the efficiency calibration is 4.5%). We report here the results obtained for a CANDU-type fuel rod (5% ^{235}U enrichment) irradiated in the 14 MW(th) TRIGA materials testing reactor from INR Pitesti during 26 months. The irradiation history is composed by 146 active irradiations steps and 146 passive ones (conventional, the last one is the cooling time). The time length of each step is represented in Fig. 1 and the correspondingly relative flux values F are represented in Fig. 2.

As the gamma spectrometric analysis was performed after a cooling time of 45 days several burn-up monitors, with long, medium and short half-times were suitable to be measured and used for burn-up evolution: ^{137}Cs, ^{103}Ru, ^{95}Zr, ^{95}Nb, ^{140}La. The burn-up values using EVOLUT are listed in Table 1.
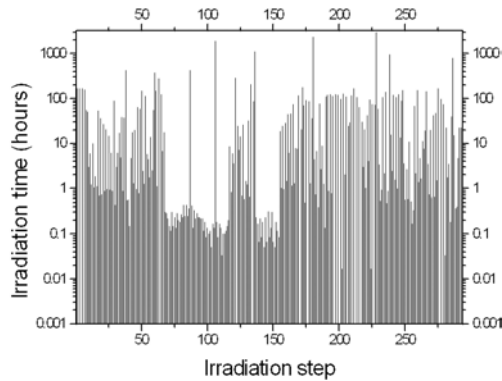
Fig. 1 – Irradiation history: the time length for each step of irradiation.
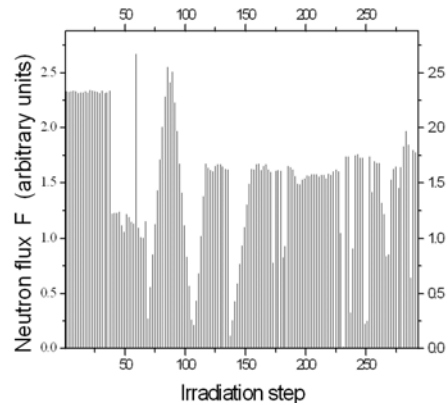

Fig. 2 – Irradiation history: the relative value of the neutron flux for each step of irradiation.

Due to its long half-life ^{137}Cs ($T_{1/2}$ = 30.09 y) is practically not influenced by the passive steps of the irradiation. So the evolution of the total burn-up (see Fig. 3) and the evolution of the number of ^{137}Cs nuclides (see Fig. 4) are similar and the best burn-up value is obtained in this case. A more complicated evolution can be observed in Fig. 5 for ^{95}Zr ($T_{1/2}$ = 64.0 d) and a dramatic one for ^{140}La ($T_{1/2}$ = 40.22 h for ^{140}La and $T_{1/2}$ = 12.8 d for ^{140}Ba, its precursor) in Fig. 6. Even in these cases good values of burn-up are obtained. The relative discrepancy of the results, in respect with the burn-up value evaluated by more accurately mass spectrometry method [9], that is 217.2 MWh/kgU, is under 6% (see the last row in Table 1).

Table 1

Burn-up values using EVOLUT

FP	^{103}Ru	^{137}Cs	^{95}Zr	^{95}Nb	^{140}La
BU (MWh/kgU)	207.9	216.1	212.7	209.3	228.8
Relative Discrepancy (%) BU_{EVOLUT} vs. BU_{MS}	4.3	0.5	2.1	3.7	5.4

The simplest formula for burn-up calculation (Eq. 4) has been applied at PIEL whenever was possible. In Fig. 7 are presented some burn-up values (calculated using Eq. 5) as a function of the correspondingly experimental determined ^{137}Cs activity for a series of CANDU-type and TRIGA LEU fuel rods, irradiated in the TRIGA reactor from INR Pitesti and measured along few years. As can be seen in Fig. 7, the theoretical linear relationship between the burn-up and the inventory of the ^{137}Cs isotope was very good met experimentally for different types of irradiated fuel rods and for different power histories. The linear relationship holds for a wide range of burn-ups, from 120 to about 2 000

MWh/kgU and the result can be interpreted as an additional argument to consider that burn-up measurement was done correctly.
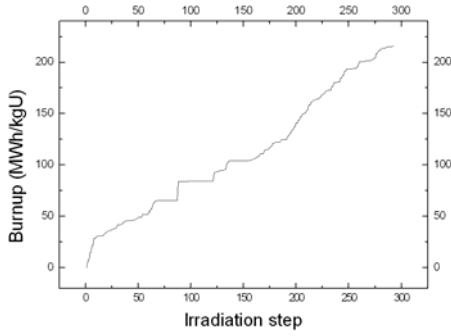

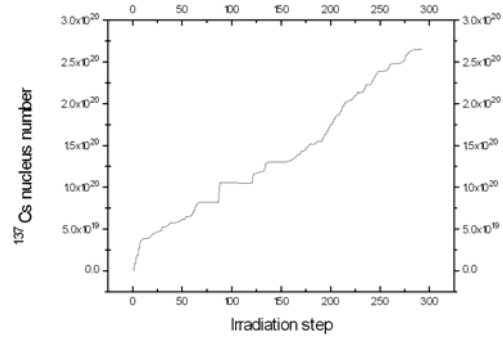Fig. 3 – Burn-up evolution along irradiation history.

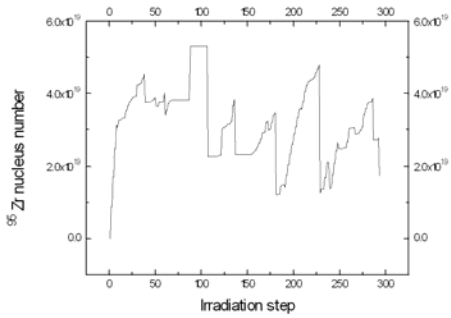Fig. 4 – Evolution of number of ^{137}Cs nuclides along irradiation history.

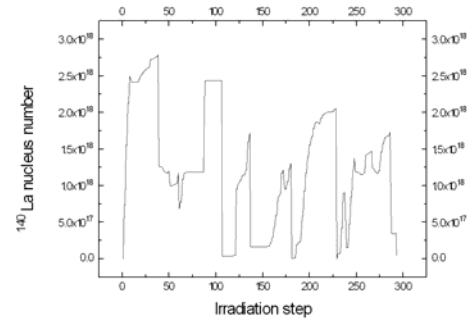Fig. 5 – Evolution of number of ^{95}Zr nuclides along irradiation history.

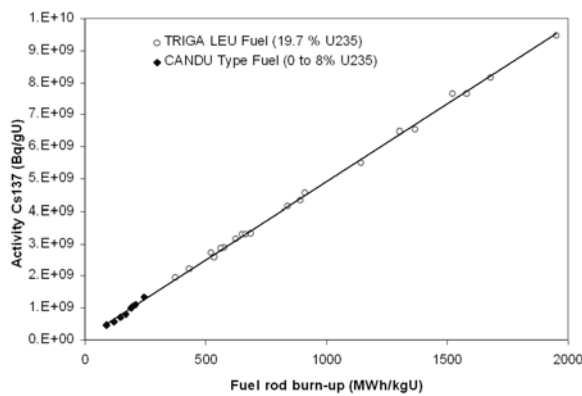Fig. 6 – Evolution of number of ^{140}La nuclides along irradiation history.

Fig. 7 – ^{137}Cs inventory as a function of burn-up.

5. CONCLUSIONS

We appreciate that the computer code developed at PIEL is a useful tool for burn-up evaluation based on non-destructive gamma spectrometry methods, with a good accuracy (< 10%). Extensive experiments were performed in order to validate the method described in this paper and the implementation. EVOLUT can be used on an usual PC and the results are obtained in a few minutes. It has an original and user-friendly graphical interface and it provides also output in script MATLAB files for graphical representation and further numerical analysis. The computer program needs simple data and it is valuable especially when a large number of burn-up analyses are required quickly.

The theoretical linear relationship between the burn-up and the inventory of ^{137}Cs isotope was verified for a wide range of burn-ups and for different types of fuel rods irradiated in the same reactor, which confirms the correctness of the burn-up measurements made over time at PIEL.

REFERENCES

1. J. R. Phillips *et al.*, *Quantitative Determination of Fission Products in Irradiated Fuel Pins Using Non-Destructive Gamma Scanning*, Anal. Chem., **47**, 71 (1975).
2. C. Devida, M. Betti, P. Peerani, E. H. Toscano and W. Goll, *A Quantitative Burn-up Determination: A Comparision of Different Experimental Methods*, Proceedings "HOTLAB" Plenary Meeting 2004, Norway, 106-113.
3. D. Freis, R. Nasyrow and E. H. Toscano, *Burn-Up Determination of High Temperature Reactor Spherical Fuel Elements by Gamma Spectrometry*, Proceedings HTR2006: 3rd International Topical Meeting on High Temperature Reactor Technology 2006, South Africa .
4. T. Pineira, Rep. CEA R 4944, CEA C'entre d'etude de Cadarache, 1978.
5. M. Parvan, I.L. Tuturici, R. Dobrin, M. Popov, R. Radulescu, V. Toma, *The Post-Irradiation Examination of CANDU Type Fuel Irradiated in the Institute for Nuclear Research TRIGA Reactor*, 3rd Research Coordination Meeting on Examination of Water Reactor Fuel (ED-WARF- II) 1995, Russia.
6. Al. Alexa, T. Craciunescu, G. Mateescu and R. Dobrin, *The Tomographic Maximum Entropy Method in the 3-D Analysis of Nuclear Fuel Pins*, Journal of Nuclear Materials, **218**, 139–142 (1995).
7. S. Caruso, M.F. Murphy, F. Jatuff, R. Chawla, *Determination of within-rod caesium and europium isotopic distributions in high burn-up fuel rods through computerised gamma-ray emission tomography*, Nuclear Engineering and Design, **239-7**, 1220–1228 (2009).
8. S. Jacobsson, C. Andersson, A. Hakansson, A. Backlin, *A tomographic method for verification of the integrity of spent nuclear fuel assemblies*. I: *Simulation studies*, Nuclear Technology, **135-2**, 131–145 (2001).
9. *** Standard Method of Test for Atom Percent Fission in Uranium and Plutonium Fuel (Mass Spectrometry Method), American National Standard Institute, ASTM E 244.