Fuel burnup calculation and measurement at IEA-R1 research reactor

L.A.A. Terremoto, C.A. Zeituni, J.A. Perrotta, A.T. e Silva, J.E.R. da Silva, M. Yamaguchi, J.B.M. Tondin, R. Frajndlich

Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN-SP)
Travessa R, 400 – Cidade Universitária; Caixa Postal 11049; CEP 05422-970
São Paulo – SP BRASIL

Abstract

The availability of burnup data is an important requirement in any systematic approach to the enhancement of safety, economics and performance of a nuclear research reactor. This work presents the main features of the neutronic calculations and gamma-ray spectroscopy measurements employed to determine the burnup of Material Testing Reactor (MTR) fuel elements irradiated in the IEA-R1 research reactor. Burnup values obtained by means of both methods show good agreement within the experimental error limits.

1. Neutronic Calculations

The IEA-R1 Research Reactor is a multipurpose reactor. It has been used for basic and applied research in nuclear área, training and also for radioisotopes production. Figure 1 shows the standard configuration of the IEA-R1 reactor core with 21 standard fuel elements and 4 control elements. The standard fuel elements numbered 149 to 152 contain 1.9 gU/cm^3; 153 to 164 and the control fuel elements 179 and 180 contain 2.3 gU/cm^3 while the remainders contain 3.0 gU/cm^3. All fuel elements are of LEU (low enriched uranium) type, with 19.75 % enrichment. Figure 2 shows the ^{235}U burnup distribution (October 2001).

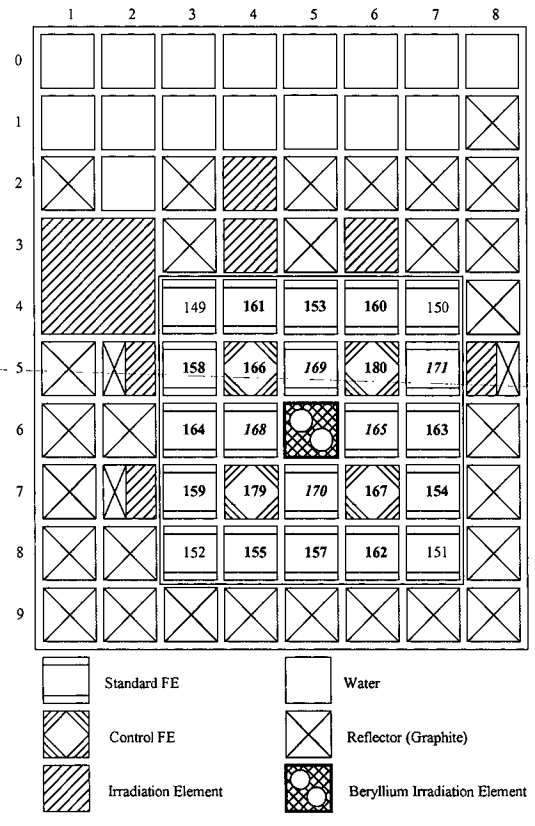

Figure 1. IEA-R1 reactor core configuration.

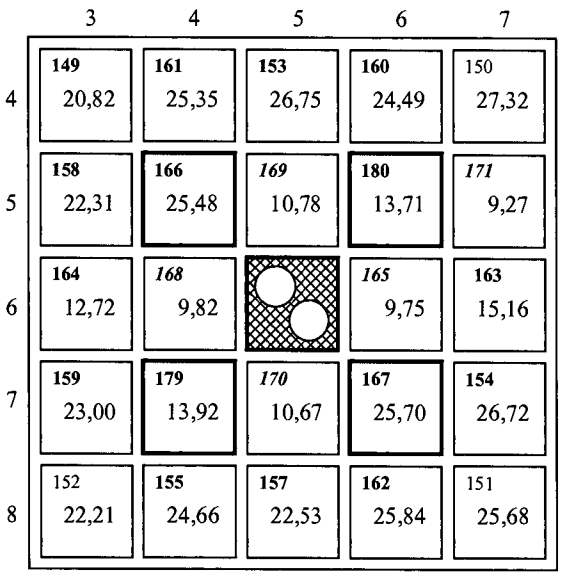

Figure 2. ^{235}U percentual burnup distribution.

The neutronic calculational methodology regarding the IEA-R1 reactor is based on LEOPARD [1, 2] and HAMMER-TECHNION [3] programs for cross section generation, 2DB [4] program for the core and burnup calculations in a two-dimensional geometry and CITATION [5] program for a three-dimensional analysis to obtain effective multiplication factor, neutron flux and power density distributions, integral and differential control rod worth, reactivity coefficients and kinetic parameters.

Design Criteria. The purpose of the design criteria for core assembling is to assure that the reactor operation is safe and controlled so that the reactor can be shut down and held subcritical for all operational states and that safety limits are not exceeded. Thus, the following criteria have been established based on the Safety Series-35 [6]:

- At least 200 % of the maximum reactivity excess shall be available in the reactivity control mechanisms. This criterion assures a shutdown margin of 100 % over the reactivity excess;

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- The reactor shall be maintained subcritical when the most reactive control rod is fully withdrawn and the others are fully inserted (stuck-rod criterion). In this condition the effective multiplication factor shall be less than 0.98;
- The maximum rate of addition of positive reactivity shall be less than 35 pcm/s; and
- The temperature and void reactivity coefficients shall be negative.

Neutronic Calculational Methodology. Figure 3 shows the schematic diagram of the neutronic calculational methodology for the IEA-R1 reactor core. The fuel cross section is obtained with LEOPARD program (version modified by Michigan University, which includes a plate geometry option) using a standard cell model (fuel, cladding and moderator) with an extra region to take into account other parts of the fuel element. The LINXS, CONVERB and CONVERD programs are to prepare cross sections to 2DB format.

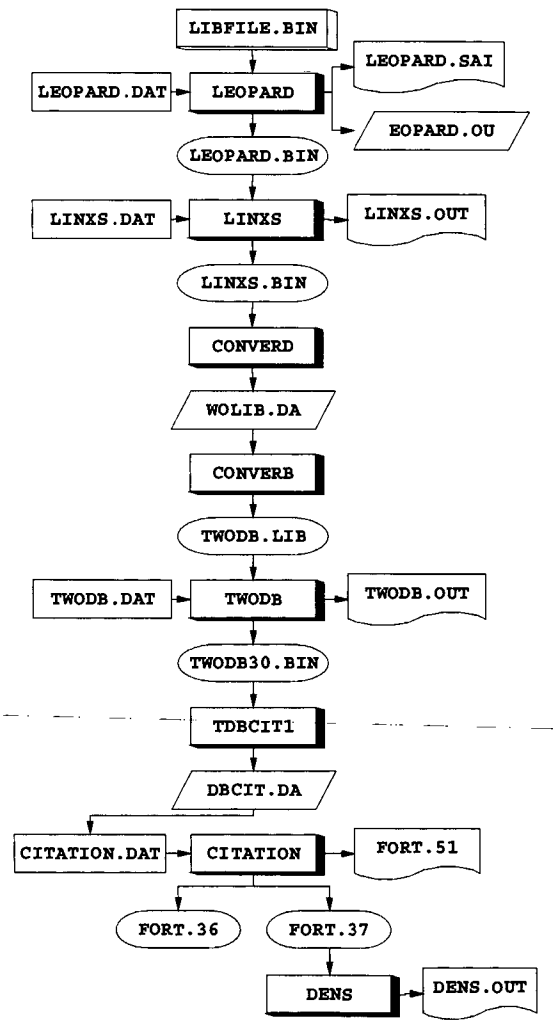

Figure 3. Neutronic calculational methodology.

The HAMMER-TECHNION program is employed to generate the cross sections for the non-fuel regions, such as control rods and reflectors.
The reactor power history is simulated with 2DB program in a two-dimensional model. This process generates the ^{235}U burnup distribution and a macroscopic cross sections file for CITATION program.
Finally, three-dimensional calculations are performed using CITATION program for effective multiplication factor, neutron flux and power density distributions, integral and differential control rod worth, reactivity coefficients and kinetic parameters. The DENS.OUT file contains the power density distribution for thermal hydraulic analysis.

2. Inventory of Nuclear Materials

The system of nuclear material safeguard has the main objective of permitting an inventory of each and every item covered under the definition of nuclear material, either stored or in use in the primacies of reactor. In the IEA-R1 reactor, complete records of the uranium burnup and plutonium production are maintained. Total mass of the element and its fissile isotope ^{235}U are registered in the case of uranium while for plutonium only the total mass of the element is registered. In the following we present some definitions to better understand the terms normally used:

Nuclear material: The elements uranium, thorium and plutonium as well as their chemical compounds are considered nuclear materials.
Installation: Any place where the nuclear material is used handled or stored.
Accountability area: Physical space where the quantity of nuclear material present can be determined with certain precision for the purpose of establishing material balance.
Safeguard agreement: Agreement between one or more countries, and the International Atomic Energy Agency (IAEA), requiring commitment of these countries not to use, for military purposes, material, equipment and technical information obtained within the agreement of cooperation.

The reactor manager is responsible for the safeguard and accountability of the nuclear material within the accountability area. The responsibility for the movement of nuclear material to areas external to the accountability area is of the safeguard service of the installation, duly authorized by the safeguard service of the National Nuclear Energy Commission (CNEN).

Since the beginning of its operation in 1957, the inventory of the nuclear material in the reactor IEA-R1 is made by calculating the uranium burnup and plutonium production in the fuel elements used in different configurations of the core. The calculations are carried out at the end of each month, after each change in the configuration of the reactor core, or during routine inspection visits from CNEN, AIEA and Brazil-Argentine Agency of Accounting and Control of Nuclear Material (ABACC).

Calculations are performed using the computer program MBRPLU.BAS, developed by the team of reactor operators. The method used in the calculation requires information on the reactor operation time for each day, the dissipated energy (E_d) in MWh for this period and the accumulated energy dissipation (E_{da}) during each month or whatever period considered. This information is routinely registered in the reactor operation logbook. The value of E_{da} is used for the calculation of the ^{235}U burnup. Computer programs HAMMER-TECHNION [3] and CITATION [5] are used to obtain the cellular calculation and the thermal neutron flux respectively at each fuel element position in the grid plate of the reactor core. These values are employed to calculate the ratio $\phi_{th}^{i}/\bar{\phi}$ between the thermal neutron flux at any given fuel element position i and the average neutron flux. For each position, one also determines the ratio between the mass (M_i) of ^{235}U at position i and the total mass (M_t) of ^{235}U in the core. The ^{235}U burnup factor is determined from the following expression:

$$B_i = \frac{M_i}{M_t} \cdot \frac{\phi_{th}^{i}}{\bar{\phi}} \qquad (1)$$

The burnup factor B_i is normalized to give:

$$B_{in} = \frac{B_i}{\sum_{i=1}^{n} B_i} \qquad (2)$$

where n is the number of fuel elements in the core and B_{in} is the normalized burnup factor.

The amount, in grams, of the ^{235}U burnup in the fuel element at any given position i is therefore obtained by means of the expression:

$$B_{235}\,(g) = B_{in} \cdot E_{da} \cdot 0.051667 \qquad (3)$$

where the factor 0.051667 is the ^{235}U burnup mass for each MWh of dissipated energy of the reactor.

To calculate the ^{238}U burnup mass, the following equation is used:

$$B_{238}\,(g) = B_{235} \cdot f \qquad (4)$$

where f is a factor that depends on the uranium enrichment of the fuel element, being 0.848 for 93% enrichment and 0.924 for 20% enrichment.

The ^{239}Pu produced mass is calculated by means of the following expression:

$$P_{239}\,(g) = B_{235} \cdot g \qquad (5)$$

where the factor g takes values of 0.001613 and 0.077420 respectively for 93% and 20% uranium enrichment of the fuel elements.

The MBRPLU.BAS program finally generates a data file containing details of uranium burnup and plutonium production in each fuel element, which can be printed out in the form of a report "Fuel Element Situation as of __/__/__".

As part of the necessary documentation for the safeguard of nuclear material, maps identifying all the locations within the reactor building, where fuel elements or fission chambers are stored or used, including the reactor pool, safe box and dry storage silos located at the first floor of the reactor building, are prepared and printed out. A general logbook is maintained, where the inventory data is registered every month, after every change of the core configuration or whenever there is an inspection visit. After every change of reactor core configuration or change in the position of any fuel element, a report on *Individual Data of Fuel Elements* must be filed.

The transfer of nuclear material between accountability areas or installations requires a prior completion of the "*Authorization of Transference of Nuclear Material*" form accompanied by a "*Notification of Transference of Nuclear Material*" form [7].

A total of 56 fuel elements and 10 fission chambers are either in use or stored, inside the reactor building. All of them are covered by the safeguard agreement. During the period 1999-2000 six reactor configuration changes were made. The uranium burnup and plutonium production [8] data are shown in Table 1.

Table 1. Uranium burnup and plutonium production in the IEA-R1 reactor during the years 1999-2000.

Configuration	^{238}U (g)	^{235}U (g)	^{239}Pu (g)
206	107.06	115.82	8.97
207	79.72	86.32	6.68
208	0	0	0
209	0	0	0
210	241.80	261.64	20.26
210A	68.13	73.74	5.71
Total	496.71	537.52	41.62

The core configurations 208 and 209 had no uranium burnup as the reactor was operated at low power (20 W) for specific experiments. During the year 1999-2000 the reactor received 14 new fuel elements, two among them were control elements. These fuel elements were fabricated in the fuel fabrication center of IPEN-CNEN/SP. Twelve normal fuel elements were of U_3Si_2-Al dispersion with a density of 3.0 gU/cm^3 while the control elements were of U_3O_8-Al dispersion with a density of 2.3 gU/cm^3. Only six of these fuel elements were used in the reactor during this period. The uranium enrichment in all these elements is 20%.

On November 3, 1999, under an agreement between IPEN-CNEN/SP and the United States Department of Energy (US-DOE), 127 spent fuel elements, containing 67154.54 g of ^{238}U and 15101.67 g of ^{235}U were transported to U.S.A. These spent fuel elements, accumulated since the

beginning of the reactor operation in 1957, were stored in racks inside the reactor pool as well as in the dry storage silos located at the first floor of the reactor building.

During 1999-2000 two inspections were carried out by AIEA and ABACC.

3. Gamma-ray Spectroscopy Measurements

Introduction. The irradiation of fuel elements gives rise to radioactive fission products that, in the absence of cladding failures, remain inside the meat of each fuel plate. Gamma-ray spectroscopy is a nondestructive method based on analysis of spectra that result from collimation and detection of gamma-rays emitted in the decay of most radioactive fission products. In the IEA-R1 reactor, gamma-ray spectroscopy measurements were performed at the reactor pool area.

To obtain the necessary information, complete gamma-ray spectra are accumulated as a function of axial and transversal positions of the irradiated fuel element. The net areas under the full-energy peaks are determined, giving a quantitative measurement of the amount of each radioactive fission product present at a specific location. These amounts can be related to the total absolute activity of a given fission product, used as burnup monitor, which enables the determination of the total number of fission events and therefore the fuel element absolute burnup.

Such determination, however, requires the previous energy and efficiency calibration of the experimental apparatus, a rigorous control of its geometry, detailed knowledge about the irradiation history of the fuel element, precise record of its cooling time, accurate evaluation of all attenuation effects involved and low pool background.

Basic Experimental Technique. The experimental apparatus for gamma-ray spectroscopy [9] consists of collimator tube, lead shielding, high-purity germanium (HPGe) detector together with suitable fast electronics and an on-line microcomputer data acquisition module. Attainment of absolute burnup values requires previous energy and efficiency calibration of the detection set (collimator tube + HPGe detector).

The detection set was positioned over an aluminum base fixed at the pool border of the IEA-R1 research reactor, and equipped with two perpendicular crank driven mechanisms forming a x-y frame that enables the movement of the detection set either parallel or normal to the axial direction of an irradiated fuel element horizontally positioned.

In order to perform the gamma-ray spectroscopy measurements, an irradiated fuel element previously selected is hoisted up from the spent fuel storage rack and brought to an immerse stainless steel platform, where it was positioned horizontally with the fuel plates surface-upwards. An angle steel welded at the platform ground assures reproducibility in the positioning of the irradiated fuel element. Gamma-ray spectroscopy measurements are performed in the configuration shown schematically in Figure 4.

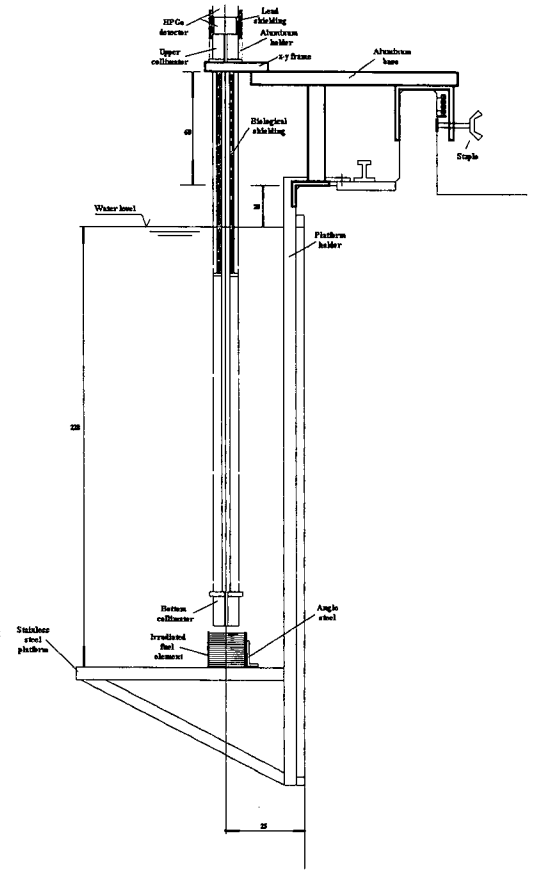

Figure 4. Cross-sectional diagram of the experimental apparatus for gamma-ray spectroscopy installed at the pool area of the IEA-R1 research reactor. Dimensions are in cm.

The collimator tube is positioned between the irradiated fuel element and the detector in order to enable the determination of the gamma emission rate of a specific volume, as well as to avoid the system overflow concerning data acquisition. In the configuration shown in Figure 4, the distance between the bottom collimator window and the last plate of the fuel element was determined using a scale and a radiation-resistant underwater camera.

The HPGe detector employed has a volume of 130 cm^3, with 1.71 keV resolution and 26.1 % relative efficiency for the gamma-ray of ^{60}Co [10]. The gamma-ray energy range taken for the analysis was from 50 to 2800 keV.

Calibrations. In order to identify the full-energy peaks present in every spectrum obtained from measurements on irradiated fuel elements, energy calibrations of the HPGe detector were carried out immediately before and also after the experiments, using punctiform calibration sources.

The efficiency calibration of the detection set (collimator tube + HPGe detector) was measured for the fixed detection geometry and the gamma-ray energies of interest (emitted in the decay of burnup monitors). The absolute efficiency

of the detection set was determined measuring separately the intrinsic efficiency ε_γ of the HPGe detector as a function of the gamma-ray energies and the area a_j of the circle defined by the solid angle on the central plane of the jth plate meat of a given irradiated fuel element.

In order to measure the parameter ε_γ, the HPGe detector was fixed at the upper part of a cylindrical lead shielding with a side opening that enables the one-by-one positioning of punctiform calibration sources inside. During the measurements, the distance between the center of the HPGe detector window and the center of each calibration source was constant. The obtained values of the intrinsic efficiencies at the gamma-ray energies emitted by the calibration sources were fitted by a function to give the value of ε_γ at the energies of interest [9].

The areas a_j were measured using an experimental arrangement in which the detection set was positioned horizontally in front of a strong ^{137}Cs source of known activity. The distances between the aluminum window, located at the bottom edge of the collimator tube, and the front part of the ^{137}Cs source were adjusted to reproduce the relative positioning between the same aluminum window and the center of each plate of a fuel element during the measurements performed at the reactor pool [9].

Burnup Determination. Once the values of ε_γ and a_j are known, it is necessary to measure the parameter $\overline{Q}$, that gives the average value for the total number of counts per unit of time registered in the detector. It must be measured to determinate experimentally the total activity of the irradiated fuel element due to a given burnup monitor and, thereafter, the fissioned mass of ^{235}U in this fuel element.

The ratio between fissioned and initial masses of ^{235}U in a fuel element furnishes its burnup.

The $\overline{Q}$ value is obtained by means of measurements performed along the active length (X-axis) and along the active width (Y-axis) of the fuel element. These measurements define a function of two variables, with typical profiles like the ones shown in Figure 5.

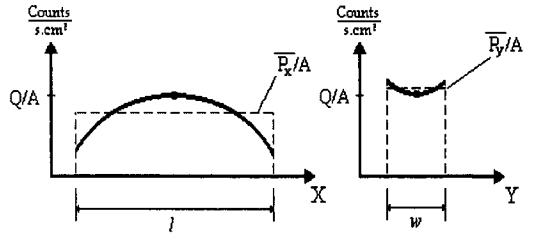

Figure 5. Representation of typical burnup profiles obtained from gamma-ray spectroscopy measurements on irradiated MTR fuel elements. The parameters indicated are employed to calculate the average value $\overline{Q}$.

Applying a property of two-variable functions [11] to the experimentally obtained profiles, one finds

$$\overline{Q} = \overline{P}_X + \overline{P}_Y - Q \tag{6}$$

where $\overline{P}_X$ is the average number of counts per unit of time obtained in measurements along the fuel element active length, $\overline{P}_Y$ is the average number of counts per unit of time obtained in measurements along the fuel element active width and Q is the number of counts per unit of time measured at the central point of the fuel element.

Using this formulation for $\overline{Q}$ together with the radioactive decay law, the total number of burnup monitor nuclei present in a standard IEA-R1 fuel element (which has 18 plane parallel fuel plates), immediately after the end of the last irradiation period, becomes [9]

$$N_0 = \frac{18 \cdot l \cdot w \cdot \left(\overline{P}_X + \overline{P}_Y - Q\right)}{\lambda \cdot I_\gamma \cdot \varepsilon_\gamma \cdot k_1 \cdot k_2 \cdot k_3 \cdot \sum_{j=1}^{18} a_j K^{18-j}} e^{\lambda \cdot t_c} \tag{7}$$

where l is the active length of each fuel plate of the element, w is the active width of each fuel plate of the element, λ is the decay constant of the burnup monitor, t_c is the interval between the end of the last irradiation period and the start of the measurements (usually known as cooling time), I_γ is the absolute emission intensity of the gamma-ray emitted in the decay of the burnup monitor, ε_γ is the intrinsic efficiency of the HPGe detector for the gamma-ray emitted in the decay of the burnup monitor and a_j is the area defined by the detection solid angle on the central plane of the jth plate meat.

The parameters designated by k_1, k_2, k_3 and K are correction factors due to attenuation effects [9]:

$$k_1 = \frac{1 - e^{-\mu \cdot s}}{\mu \cdot s} \tag{8}$$

is the self-attenuation correction factor, that results from the attenuation of gamma-rays traversing the plate meat where they are emitted, where s is the plate meat thickness and μ is the linear attenuation coefficient, for a given gamma-ray energy, of the fuel material contained in the plate meat;

$$k_2 = e^{-\mu_{Al} \cdot a} e^{-\mu_a \cdot C_1} e^{-\mu_{Al} \cdot C_2} \tag{9}$$

is the correction factor that arises from the attenuation of a gamma-ray after traversing the upper aluminum cladding of the last fuel plate (whose thickness is a), the water layer between the last fuel plate and the bottom edge of the collimator tube (whose thickness is C_1) and afterwards the aluminum window that closes the collimator tube (whose thickness is C_2), where μ_{Al} is the aluminum

linear attenuation coefficient and μ_a is the water linear attenuation coefficient;

$$k_3 = e^{-\mu_{air} \cdot L} \quad (10)$$

is the correction factor that takes into account the attenuation of the gamma-ray when it passes through the air contained inside the collimator tube before it strikes the detector, where μ_{air} is the linear attenuation coefficient of air and L is the total length of the collimator tube, and

$$K = e^{-2 \cdot \mu_{Al} \cdot a} e^{-\mu_a \cdot b} e^{-\mu \cdot s} \quad (11)$$

is the correction factor that corresponds to the attenuation of a gamma-ray, emitted in the meat of the jth fuel plate and passing through successive layers until reaching the upper surface of the meat contained inside the last (18th) fuel plate, where b is the distance between two successive plates of a fuel element.

The fissioned mass of ^{235}U in the irradiated fuel element is given by

$$\Delta U = \frac{N_0 m_0}{y N_U^0} f \quad (12)$$

where N_U^0 is the initial number of ^{235}U atoms in the fuel element, m_0 is the initial mass of ^{235}U in the fuel element, y is the average yield of the burnup monitor in the fission of ^{235}U [9] and f is a correction factor that takes into account the decay of burnup monitor nuclei occurred during different irradiation periods and powers, which is given by the following expression [12]:

$$f = \frac{\lambda \sum_{k=1}^{n} P_k t_k}{\sum_{k=1}^{n} P_k e^{-\lambda \cdot \tau_k} \left(1 - e^{-\lambda \cdot t_k}\right)} \quad (13)$$

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

Finally, the combined use of equations (7), (12) and (13) determines the fissioned mass of ^{235}U in the irradiated fuel element, while the ratio $\Delta U/m_0$ furnishes its burnup.

Results and Discussion. Initially, pool background measurements were performed and the full-energy peaks in the corresponding gamma-ray spectrum were identified using the energy calibration. No fission products at all were found. Only activation products and some isotopes of the natural background [13] were detected [14].

Among the activation products, ^{58}Co ($T_{1/2}$ = 70.92 d), ^{60}Co ($T_{1/2}$ = 5.27 yr) and ^{65}Zn ($T_{1/2}$ = 244.1 d) are always present in water samples collected from the reactor pool [15]. They are generated by oxidation (chemical reaction) and neutron absorption (nuclear reaction) occurred at the cladding and holder of the reactor control rods, both made of metallic nickel (cases of ^{58}Co and ^{60}Co), or at structures of the heat exchangers (case of ^{65}Zn).

However, if the pool water is collected with the reactor under operation or thereupon the reactor shutdown, the activation product ^{24}Na ($T_{1/2}$ = 15.02 h) is largely predominant. It is generated by the nuclear reaction $^{27}Al(n,\alpha)^{24}Na$, occurred in the aluminum cladding of fuel and reflector elements, as well as in aluminum structures of the reactor core, during the irradiation [15].

Finally, the activation product ^{110m}Ag ($T_{1/2}$ = 249.9 d) is generated by means of radiative capture of one neutron by the isotope ^{109}Ag, which forms 48.17 % of the natural silver [16], metal that constitutes 80 % of the alloy used in the reactor control rods. This silver, activated in the reactor core, dilutes in the pool water as monovalent cations, once the silver oxide is slightly soluble [17]. However, on the contrary of the other activation products, ^{110m}Ag is not detected in pool water samples, but in metallic surfaces over which this isotope is deposited probably by means of displacement chemical reactions. Hence, ^{110m}Ag was detected over the aluminum-cladding surface of the fuel plates of almost all stored fuel elements [15].

The pool background measurements also demonstrated that prominent Compton continua from high-energy gamma-rays, mainly the ones emitted in the decay of activation products, can obscure full-energy peaks from gamma-rays emitted in the decay of burnup monitors and, as a consequence, would distort the result of the gamma-ray spectroscopy measurements on irradiated fuel elements. Once the most important activation product concerning this issue is ^{24}Na, it was decided that gamma-ray spectroscopy measurements on irradiated fuel elements would be performed only during long maintenance periods in which the reactor did not operate.

In the beginning, the irradiated fuel elements, selected for gamma-ray spectroscopy measurements, were assorted in two groups, according to the cooling time and fission product used as burnup monitor. Fuel elements with cooling times equal or longer than two years were monitored using ^{137}Cs, whereas for shorter cooling times the utilized burnup monitor was $^{144}Ce/^{144}Pr$.

The duration of a gamma-ray spectroscopy measurement at each selected point of the fuel element varied according to the burnup monitor employed. If the chosen burnup monitor was ^{137}Cs,

each measurement had the duration of 600 s of live time. However, if the suitable burnup monitor was $^{144}Ce/^{144}Pr$, each measurement had the duration of 3600 s of live time. A gamma-ray spectrum was obtained from each measurement. As a consequence, complete gamma-ray spectra were accumulated as a function of axial and transversal positions for a given irradiated fuel element.

An additional test of the experimental method was performed later, measuring the burnup of two partial fuel elements manufactured by IPEN and irradiated in the IEA-R1 reactor. These partial fuel elements present the same project parameters of the standard ones also fabricated by IPEN, except for the total number of fuel plates: the fuel element IEA-128 has two fuel plates (the first and the 18th ones), whereas the fuel element IEA-129 has ten fuel plates (the first, 3rd, 5th, 7th, 9th, 10th, 12th, 14th, 16th and 18th ones). Massive aluminum plates of the same thickness replaced the other fuel plates of these elements. Each measurement on these partial fuel elements was longer than corresponding ones performed on standard fuel elements also monitored using ^{137}Cs, requiring 900 s of live time (IEA-129) and 1200 s of live time (IEA-128).

As an example of gamma spectra obtained from measurements on irradiated fuel elements monitored with ^{137}Cs, Figure 6 shows the gamma spectrum from a run of 600 s of live time carried out at the central point of the fuel element 111.

It is important to observe that the full-energy peak from 661.6 keV, that corresponds to the gamma-ray emitted in the decay of ^{137}Cs, is the most prominent in the gamma spectrum.

For each gamma spectrum obtained from measurements along the active width and along the active length of an irradiated fuel element, the net number of counts (area) under the full-energy peak from 661.6 keV was determined using the computer program IDEFIX [18].

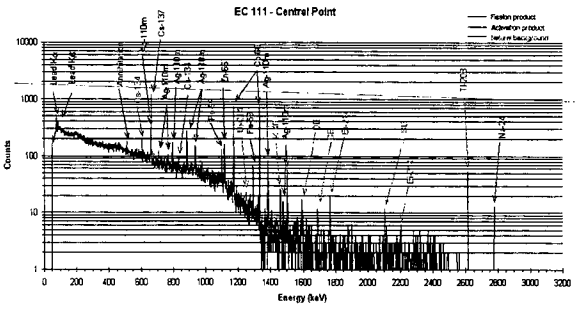

Figure 6. Gamma-ray spectrum obtained from measurements of 600 s of live time performed at the central point of the irradiated fuel element 111. The origin of each peak is indicated.

In every gamma spectra, the full-energy peak from 661.6 keV is always preceded by another one, much smaller and very close, identified as correspondent to the most intense gamma-ray emitted in the decay of the activation product ^{110m}Ag (E_γ = 657.7 keV, I_γ = 0.947). Hence, it was necessary to fit a Gaussian function to each one of the full-energy peaks of a doublet using the computer program IDEFIX [18], in order to separate correctly the areas, as shown in Figure 7.

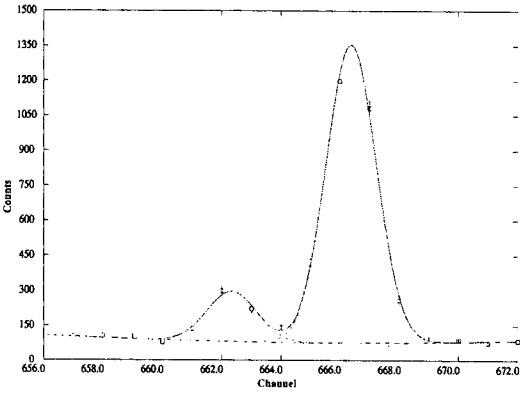

Figure 7. Gaussian functions fitted to each full-energy peak of the doublet with energies 657.7 keV (^{110m}Ag) and 661.6 keV (^{137}Cs) using the computer program IDEFIX [18], in order to separate the areas of these peaks in the gamma-ray spectrum obtained from measurement at the central point of the fuel element 111.

The areas under the full-energy peaks from 661.6 keV, presented as a function of the active length and active width of an irradiated fuel element, constitute its burnup profiles. As an example, Figure 8 shows the burnup profiles for the fuel element 111.

The values of the areas were used to calculate the parameter $\overline{Q}$ and, thereafter, the total activity of an irradiated fuel element due to the burnup monitor ^{137}Cs, as well as the fissioned mass of ^{235}U in this fuel element. Finally, the ratio between fissioned and initial masses of ^{235}U in this fuel element furnishes its burnup. All these calculations employed to determine burnup from experimental data were made using the software EXCEL [19].

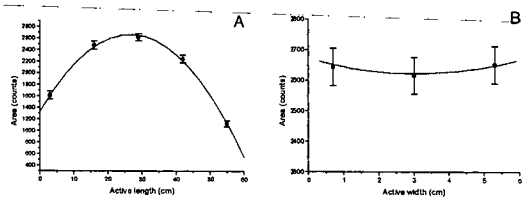

Figure 8. Experimental burnup profiles for the fuel element 111: (A) burnup profile along the active length; (B) burnup profile along the active width. The curves joining the points are just to guide the eye.

A resemble procedure was adopted for measurements on irradiated fuel elements monitored with $^{144}Ce/^{144}Pr$, but in this case each selected point was measured during 3600 s of live

time. Figure 9 shows the gamma spectrum from a measurement carried out at the central point of the fuel element IEA-131.

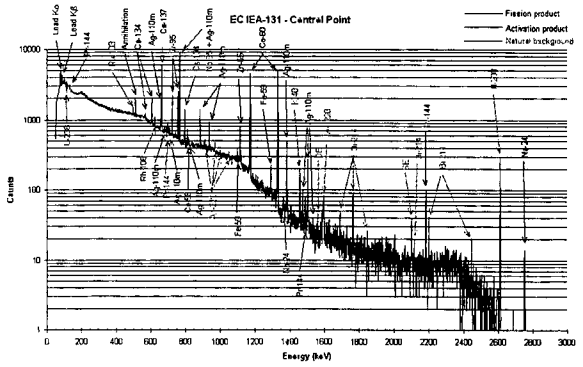

Figure 9. Gamma-ray spectrum obtained from measurements of 3600 s of live time performed at the central point of the irradiated fuel element IEA-131. The origin of each peak is indicated.

In this spectrum, one observes many full-energy peaks from gamma-rays emitted in the decay of short-lived fission products, like ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Rh and also the adopted burnup monitor ^{144}Pr, whose full-energy peak of 2185.7 keV arises isolated and very clearly. The burnup profiles for the fuel element IEA-131 are shown in Figure 10.

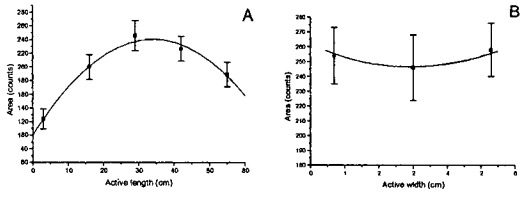

Figure 10. Experimental burnup profiles for the fuel element IEA-131: (A) burnup profile along the active length; (B) burnup profile along the active width. The curves joining the points are just to guide the eye.

Once finished the measurements on every irradiated fuel elements previously selected, the obtained burnup values were directly compared with corresponding ones provided by neutronic calculations [20, 21], with the results shown in Table 2.

Concerning standard fuel elements, the average relative experimental uncertainty for measurements monitored with ^{137}Cs is 7.5 %, whereas for measurements monitored with ^{144}Ce/^{144}Pr this uncertainty reaches 16.3 %.

The lower precision obtained in burnup measurements monitored with ^{144}Ce/^{144}Pr is a consequence of the low absolute emission intensity for the high-energy gamma-ray analyzed (E_γ = 2185.7 keV, I_γ = 0.007), which enhances the uncertainty of the area under the corresponding full-energy peak, because the total number of detected gamma-rays and therefore of recorded counts tend to be low at this energy. Relative uncertainties up to 30 % were obtained measuring with ^{144}Ce/^{144}Pr the burnup of nuclear fuels irradiated at power reactors [22, 23].

Table 2. Burnup values obtained by means of gamma-ray spectroscopy measurements (GRS) compared with corresponding ones provided by neutronic calculations (NC). IEA-128 and IEA-129 are partial fuel elements.

Fuel Element	Storage Date	Burnup Monitor	GRS (%)	NC (%)
84	10/06/1996	^{137}Cs	45.24 ± 3.81	46.92
86	05/05/1997	^{144}Ce/^{144}Pr	50.17 ± 9.93	48.40
88	31/12/1986	^{137}Cs	38.25 ± 2.88	40.50
91	12/12/1994	^{137}Cs	46.75 ± 3.26	47.63
92	09/11/1981	^{137}Cs	37.21 ± 2.92	37.56
93	09/07/1992	^{137}Cs	35.31 ± 3.13	39.93
96	09/07/1992	^{137}Cs	40.25 ± 3.38	40.51
98	19/12/1994	^{137}Cs	45.98 ± 3.13	48.28
101	30/06/1989	^{137}Cs	35.91 ± 3.18	39.08
105	16/10/1995	^{137}Cs	46.73 ± 3.19	47.44
107	17/09/1994	^{137}Cs	44.77 ± 3.15	48.30
108	27/09/1993	^{137}Cs	40.58 ± 2.89	46.15
111	04/09/1995	^{137}Cs	46.43 ± 3.03	47.37
112	04/09/1995	^{137}Cs	44.47 ± 3.04	46.31
IEA-123	16/10/1995	^{137}Cs	46.56 ± 3.36	45.93
IEA-124	16/10/1995	^{137}Cs	47.95 ± 3.51	45.61
IEA-126	10/06/1996	^{137}Cs	47.66 ± 3.34	46.17
IEA-128	07/01/1991	^{137}Cs	12.02 ± 4.25	12.58
IEA-129	30/09/1993	^{137}Cs	17.77 ± 1.78	18.44
IEA-130	09/09/1997	^{144}Ce/^{144}Pr	36.77 ± 5.13	36.10
IEA-131	09/09/1997	^{144}Ce/^{144}Pr	28.92 ± 4.67	31.46
IEA-132	09/07/1997	^{144}Ce/^{144}Pr	30.88 ± 4.77	27.34

Using the data exposed in Table 2, the ratio between burnup values obtained by gamma-ray spectroscopy and neutronic calculations was determined for each fuel element, with the results show in Figure 11. For most fuel elements, a good agreement within the experimental error limits is observed.

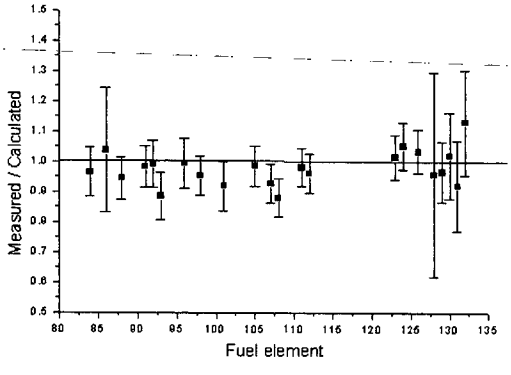

Figure 11. Ratio of measured/calculated burnup values for each fuel element.

Other burnup measurements performed recently exhibit analogous results.

Measurements carried out using a planar minidetector of cadmium telluride (CdTe) and

employing the ratio of ^{134}Cs and ^{137}Cs activities together with isotopic correlations, furnished burnup results that, compared with the ones declared by the reactor operators, agree within an uncertainty of 10 % [24].

On the other hand, a compilation embracing a very large number of burnup measurements monitored with ^{137}Cs, and performed on nuclear fuels irradiated at power reactors, show results that, compared with corresponding ones provided by neutronic calculations, differ at the utmost by 8 % [25].

5. Conclusions

Fuel burnup at the IEA-R1 research reactor is determined by means of neutronic calculations, aiming mainly core assembling and refueling as well as accountability and safeguard purposes.

Nondestructive gamma-ray spectroscopy measurements were carried out independently on 22 MTR spent fuel elements stored inside the reactor pool. The measurements were performed at the reactor pool area. They embrace fuel elements with different fuel materials, cooling times, initial enrichment grades and total number of fuel plates.

Fuel elements with cooling times equal or longer than two years were monitored using ^{137}Cs, whereas for shorter cooling times the utilized burnup monitor was $^{144}Ce/^{144}Pr$.

The obtained burnup values were compared with corresponding ones provided by neutronic calculations and good agreement within the experimental error was observed between them.

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Luis Antonio Albiac Terremoto

De: "Claudinei Pracidelli" <cpracide@ipen.br>
Para: <laaterre@net.ipen.br>
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