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Nondestructive burnup measurements by gamma-ray spectroscopy on spent fuel elements of the RP-10 research reactor

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A B S T R A C T

Gamma-ray spectroscopy is an important nondestructive method for the qualification of irradiated nuclear fuels. Regarding research reactors, the main parameter required in the scope of such qualification is the average burnup of spent fuel elements. This work describes the measurement, using nondestructive gamma-ray spectroscopy, of the average burnup attained by Material Testing Reactor (MTR) fuel elements irradiated in the RP-10 research reactor. Measurements were performed at the reactor storage pool area using ^{137}Cs as the only burnup monitor, even for spent fuel elements with cooling times much shorter than two years. The experimental apparatus was previously calibrated in efficiency to obtain absolute average burnup values, which were compared against corresponding ones furnished by reactor physics calculations. The mean deviation between both values amounts to 6%.

1. Introduction

The RP-10 is a 10 MW pool type nuclear research reactor, moderated and cooled by light water, located at the Instituto Peruano de Energía Nuclear (IPEN/Peru). After its first criticality in 25/11/1988, the RP-10 research reactor followed a weekly schedule of operation between Friday and Saturday during approximately 10 h at 7 MW, with the other days reserved to operation during 2–3 h at low power (320 kW) for activation analysis experiments. This routine changed in the beginning of the 2000s so that, until the end of 2007, operation took place every week on Sunday during 6 h at 10 MW and on the other days during 2–3 h at 320 kW. Currently, the reactor is operated on Thursday and on Friday during 5 h at 10 MW, whereas the other days of the week comprise low power operations on the same basis already mentioned.

The core of the RP-10 research reactor employs 29 plate-type usually designated as Material Testing Reactor (MTR) fuel elements, as well as 5 fork-type control rods of silver-indium-cadmium alloy (Ag–In–Cd alloy in proportion of 80%–15%–5% respectively) with stainless steel cladding. Fuel elements designed to permit insertion of a control rod are named control fuel elements, whereas all the others are named standard fuel elements. Aluminum-cladded graphite reflectors and beryllium reflectors are positioned around the reactor core.

Each MTR fuel element is constituted by plane parallel fuel plates, mounted mechanically between 2 lateral aluminum holders with grooves. A plane fuel plate contains a meat, where the nuclear fuel is located, surrounded by aluminum cladding. Standard fuel elements have 16 plane parallel fuel plates, whereas control fuel elements have 12 plane parallel fuel plates. The total thickness of a fuel plate is 0.176 cm and the meat thickness is equal to 0.100 cm. The external cladding thickness of the first and the sixteenth fuel plates of a standard fuel element is 0.045 cm. The gap between successive plates of a standard fuel element is 0.330 cm. Each fuel plate has an active length of 61.500 cm and an active width of 6.275 cm. Overall dimensions of a fuel element are (7.620 × 8.124) cm by 95.730 cm high. Fig. 1 presents the diagram of a standard MTR fuel element irradiated in the RP-10 research reactor.

Regarding active length and width, control fuel elements present the same design parameters of the standard ones, except for the fact that the first, second, fifteenth and sixteenth fuel plates are replaced by massive aluminum plates with thickness equal to 0.127 cm. Each pair of massive aluminum plates is separated by a gap of 0.610 cm, whereas the most internal massive aluminum plates and the most external fuel plates are 0.240 cm apart.

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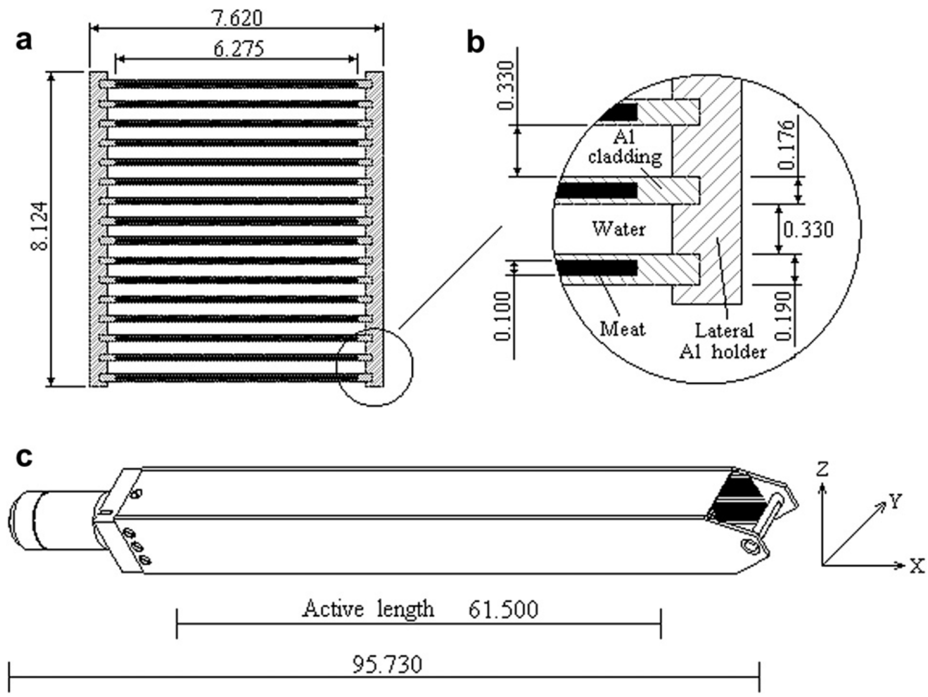

Fig. 1. Standard MTR fuel element irradiated in the RP-10 research reactor: (a) cross-sectional diagram; (b) detailed structure of successive fuel plates; (c) isometric view. The position of the reference frame used is indicated. All dimensions are given in cm.

Currently, the nuclear fuel employed in the RP-10 research reactor is U_3O_8 dispersed in an aluminum matrix, containing 2.30 gU/cm^3, whose uranium has 19.75% enrichment in ^{235}U. This Low Enriched Uranium (LEU) nuclear fuel was made in Germany (NUKEM).

There are 7 standard and 2 control fuel elements stored under water in the racks of the reactor storage pool, all of them containing the LEU nuclear fuel described above. First irradiation of all these fuel elements was carried out on 25/11/1988. The oldest fuel elements under wet storage were definitively withdrawn from the reactor core on 12/06/2003, whereas the newest ones are in the storage pool since 23/08/2007.

The necessity of measuring nondestructively the average burnup of these spent fuel elements was the main reason for IPEN/Peru to design, fabricate, install and operate a gamma scanning system, which is similar to those depicted in previous works (Rasmussen et al., 1966; Robinson et al., 1988; Wang et al., 1990; Kestelman and Guevara, 1988; Terremoto et al., 2000; Henríquez et al., 2001; Pereda et al., 2004) for use in gamma-ray spectroscopy on irradiated MTR fuel elements. Peruvian nuclear regulatory authorities require that experimental (gamma-ray spectroscopy) as well as theoretical (reactor physics calculations) results must be considered in order to check conformity of the discharge average burnup with the technical specifications of the nuclear fuel producer.

Installed at the reactor storage pool area, the gamma scanning system employs the gamma-ray spectroscopy method, which is based on analysis of spectra that result from collimation and detection of gamma-rays emitted during the decay of some radioactive fission products contained in the spent fuel elements. To obtain the necessary information, complete gamma-ray spectra are accumulated as a function of axial and transversal positions. The net number of counts (area) under the full-energy peak of 661.6 keV is determined, giving a quantitative measurement of the amount of ^{137}Cs present at a specific location. These amounts can be related to the total absolute activity of ^{137}Cs, used as burnup monitor, which enables the measurement of the total number of fission events and therefore the fuel element absolute average burnup.

However, the measurement of absolute average burnup of MTR fuel elements by gamma-ray spectroscopy 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 spent fuel element and accurate evaluation of all attenuation effects involved (Kestelman and Guevara, 1988; Terremoto et al., 2000; Henríquez et al., 2001; Pereda et al., 2004).

2. Theory

2.1. Attenuation effects

Gamma-rays emitted by radioactive fission products are attenuated as they emerge from the irradiated nuclear fuel and traverse successive layers of different materials. Consequently, all attenuation effects must be evaluated in order to measure the absolute gamma activity of spent fuel elements. This evaluation is carried out calculating the associated corrections regarding a spent MTR fuel element positioned horizontally during the measurements, with the surface of the fuel plates upwards.

The first correction to be calculated arises from the attenuation of gamma-rays traversing the plate meat where they are emitted, an effect called self-attenuation. During the measurements, the detector-to-plate distance remains large and unchanged. In such far-field configuration, the self-attenuation correction is given by (Debertin and Helmer, 1988)

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

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.

A second correction is due to attenuation of gamma-rays passing through the fuel plates and the water between them, because measurements have to be performed at the reactor storage pool. Taking into account the mentioned far-field configuration during the measurements and considering that all fuel plates of an element are identical and were irradiated under the same conditions, the correction 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 (16th) fuel plate, results

$$K^{16-j} = \left(e^{-2\mu_{\mathrm{Al}}a} \cdot e^{-\mu_{\mathrm{a}}b} \cdot e^{-\mu s}\right)^{16-j} \tag{2}$$

where μ_{Al} is the aluminum linear attenuation coefficient, μ_{a} is the water linear attenuation coefficient, a is the aluminum cladding thickness and b is the distance between two successive fuel plates of the element (this distance is filled with water).

Another correction to be evaluated originates from the attenuation of gamma-rays after traversing the upper aluminum cladding of the last fuel plate as well as the water layer between the last fuel plate and the bottom edge of the collimator tube. In this case, the correction due to attenuation is given by

$$k_2 = e^{-\mu_{\mathrm{Al}}c} \cdot e^{-\mu_{\mathrm{a}}C} \tag{3}$$

where c is the upper aluminum cladding thickness of the last fuel plate and C is the distance between the last fuel plate and the bottom edge of the collimator tube.

In this work, corrections due to attenuation of gamma-rays traversing the aluminum window that closes the collimator tube and the air contained inside the collimator tube, contrary to a previous burnup determination experiment (Terremoto et al., 2000), are not evaluated explicitly, because the efficiency calibration of the experimental apparatus already includes them.

The total correction due to attenuation effects can be obtained from the product of all corrections calculated by means of expressions (1)–(3).

2.2. Burnup monitor activity

Considering the total correction due to attenuation effects and using the reference frame shown in Fig. 1, if $\rho\,(x, y)$ is the specific activity of the irradiated fuel plates at the point (x, y), the number of counts per unit of time registered by the detector owed to the jth fuel plate, when the collimator tube is positioned over the point (x, y), is equal to

$$Q_j(x,y) = \rho(x,y) s I_\gamma A_j \varepsilon_j k_1 k_2 K^{16-j} \tag{4}$$

where I_γ is the absolute emission intensity of the gamma-ray, A_j is the area defined by the detection solid angle on the central plane of the jth plate meat and ε_j is the detector absolute efficiency for gamma-rays of a given energy and for the geometrical configuration embracing jth plate meat, collimator tube and detector during the measurements.

As a consequence of the activity of all the 16 irradiated fuel plates forming a spent fuel element, the total number of counts per unit of time, registered by the detector, results

$$Q(x,y) = \sum_{j=1}^{16} Q_j(x,y) = \rho(x,y) s\, I_\gamma\, k_1\, k_2 \sum_{j=1}^{16} A_j \varepsilon_j\, K^{16-j} \tag{5}$$

The total activity of the same spent fuel element due to a given burnup monitor is

$$D = 16\, l\, w\, s\, \overline{\rho} \tag{6}$$

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 and $\overline{\rho}$ is the average specific activity of each fuel plate.

Regarding the detector, it registers a total number of counts per unit of time whose average value $\overline{Q}$ is obtained by means of measurements performed along the active length and along the active width of the spent fuel element, been related to $\overline{\rho}$ by the expression

$$\overline{Q} = \overline{\rho}\, s\, I_\gamma\, k_1\, k_2 \sum_{j=1}^{16} A_j \varepsilon_j\, K^{16-j} \tag{7}$$

A combination of expressions (6) and (7) indicate that the total activity of a spent fuel element due to a given burnup monitor can be written as

$$D = \frac{16\, l\, w\, \overline{Q}}{I_\gamma\, k_1\, k_2 \sum_{j=1}^{16} A_j \varepsilon_j\, K^{16-j}} \tag{8}$$

Once the total correction due to attenuation effects has been considered, expression (8) shows that the product $A_j\varepsilon_j$ and the parameter $\overline{Q}$ must be measured in order to determine experimentally the total activity D of the spent fuel element concerning a given burnup monitor.

2.3. Burnup determination

Whereas the values of the product $A_j\varepsilon_j$ are measured during the efficiency calibration of the experimental apparatus, the parameter $\overline{Q}$ is obtained by means of gamma-ray spectroscopy measurements on each one of the spent fuel elements.

Gamma-ray spectroscopy measurements on each one of the spent fuel elements are performed with the same duration and following parallel rows along the active length. Every row embraces many gamma-ray spectra measured on points located at approximately regular intervals. For all spectra, the net number of counts (area), under the full-energy peak that corresponds to a given burnup monitor, is determined directly afterward.

The average value of the number of counts for each row is determined integrating the net number of counts along the active length and, subsequently, dividing the result by the total active length l of the spent fuel element. In order to obtain the average number of counts under the full-energy peak of interest for the whole measurement, an arithmetical mean of the average values for all rows is calculated. Thereafter, the spent fuel element is turned 180° around its axis and the entire measurement is repeated. As a result, two values of $\overline{Q}$ are obtained, one for each side of a spent fuel element.

Another method employed to obtain $\overline{Q}$ consists of measuring only the central axis row along the active length and along the active width of a spent fuel element, calculating the average number of counts per unit of time for both rows, adding them together and subtracting the result from the number of counts per unit of time measured at the central point of the spent fuel element (Kestelman and Guevara, 1988). Although this method has been tested quite successfully (Kestelman and Guevara, 1988; Terremoto et al., 2000; Henríquez et al., 2001), the different procedure adopted in the present work to determine the value of $\overline{Q}$ intends to get a more detailed picture of the burnup distribution across the spent fuel elements and to reduce the total relative error for average burnup measurement by means of gamma-ray spectroscopy.

Replacing the obtained value of $\overline{Q}$ in expression (8) and using the radioactive decay law, the total number of burnup monitor nuclei present in the spent fuel element, immediately after the end of the last irradiation period, becomes

$$N_0 = \frac{16\, l\, w\, \overline{Q}}{\lambda\, I_\gamma\, k_1\, k_2 \sum_{j=1}^{16} A_j \varepsilon_j\, K^{16-j}}\, e^{\lambda t_c} \tag{9}$$

where λ is the decay constant of the burnup monitor and t_c is the time interval between the end of the last irradiation period of the fuel element and the start of the gamma-ray spectroscopy measurements on it.

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

$$\Delta U = \frac{N_0\, m_0}{Y\, N_U^0} f \tag{10}$$

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 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 (Bibichev et al., 1979):

$$f = \frac{\lambda \sum_{i=1}^{n} P_i t_i}{\sum_{i=1}^{n} P_i e^{-\lambda \tau_i}\left(1 - e^{-\lambda t_i}\right)} \tag{11}$$

where P_i is the average relative power corresponding to the ith irradiation period (been $\sum_{i=1}^{n} P_i = 1$), 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.

Finally, the combined use of equations (9)–(11) determines the fissioned mass of ^{235}U in the spent fuel element, whereas the ratio $\Delta U/m_0$ furnishes its average burnup.

This procedure is carried out separately for both sides of a spent fuel element and the overall value of its average burnup results from an uncertainty-weighted mean (Helene and Vanin, 1981) of the two values obtained.

The overall value of the average burnup was measured in 3–5 different dates for each spent fuel element, in order to test the reproducibility of the measurements. At the end, for a given spent fuel element the final value of the average burnup results from an uncertainty-weighted mean (Helene and Vanin, 1981) of the 3–5 overall values measured.

3. Experiment

3.1. Experimental apparatus

The gamma scanning system of IPEN/Peru consists of collimator tube, adjustable stainless steel holders, x–y motion device made up of stainless steel base equipped with two perpendicular crank driven mechanisms, high-purity germanium (HPGe) detector together with fast suitable electronics and an online microcomputer data acquisition module. The holders are employed in the positioning of the collimator tube between the spent fuel element and the HPGe detector. This configuration enables the determination of gamma emission rate of a specific fuel volume and avoids the system overflow concerning data acquisition.

During the measurements, the spent fuel element remains over a stainless steel platform located under the water of the reactor storage pool, while the detector and other electronic components are installed permanently outside the water at the reactor pool hall. The collimator tube extends from 0.35 ± 0.05 cm above the surface of the last plate of the spent fuel element up to 3.0 cm sideways below the HPGe detector, perpendicularly to the detector axis and 1.8 cm away from the detector aluminum window (position that corresponds to the center of the germanium crystal side surface).

The collimator tube consists of central part, upper collimator and bottom collimator. The central part is an aluminum cylindrical pipe with a length of 310.1 cm, an external diameter of 3.2 cm and a wall thickness of 0.5 cm. Lead collimators cladded with aluminum are attached to both ends of the central part by means of mortises. The upper collimator is removable and has a length of 5.9 cm, an external diameter of 3.2 cm and a central collimating aperture with diameter of 0.6 cm. The bottom collimator has a length of 18.5 cm, an external diameter of 5.0 cm and a central collimating aperture with diameter

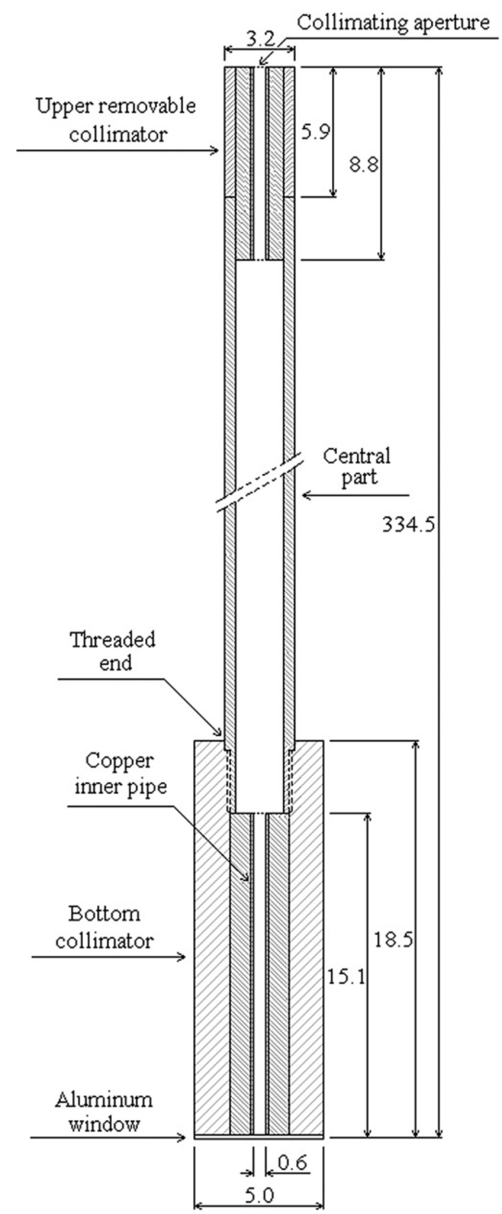

Fig. 2. Cross-sectional diagram of the collimator tube used in the gamma-ray spectroscopy measurements. All dimensions are given in cm.

of 0.6 cm closed by an aluminum window with thickness of 0.1 cm, which can be opened when the collimator tube is outside the water in order to check the alignment of the gamma scanning system. Therefore, the total length of the collimator tube is 334.5 cm. Fig. 2 shows schematically the cross-sectional diagram of the collimator tube used in the gamma-ray spectroscopy measurements.

After been collimated, gamma-rays are detected by the HPGe detector together with fast suitable electronics composed of amplifier, high voltage supply, multichannel analyzer and BIN. All these electronic components of the gamma scanning system are manufactured by CANBERRA. The HPGe detector has a volume of 51.39 cm^3, with 1.68 keV resolution and 10.1% relative efficiency for the 1332.5 keV gamma-ray of ^{60}Co. The gamma-ray energy range taken for the analysis is from 50 keV to 2800 keV. Acquisition of gamma-ray spectra is performed with the multichannel analyzer coupled to a microcomputer through an S100 control interface.

3.2. Efficiency calibration

In order to perform the efficiency calibration, the collimator tube and the electronic components of the gamma scanning system were removed from the reactor storage pool and transported to a laboratory, where they were assembled in a configuration that reproduces exactly their relative positioning during the measurements. Shortly thereafter, the aluminum window of the bottom collimator was opened and the alignment of the system was checked using a low intensity laser beam. Careful adjustments were made manually until the correct alignment has been attained.

Following the alignment, the aluminum window of the bottom collimator was closed and a 5.21×10^8 Bq calibration source of ^{137}Cs, sealed in a small stainless steel cylindrical case with active diameter of 0.6 cm, was placed on a short ruled rail along the collimator tube central axis, in front of the aluminum window of the bottom collimator. The absolute efficiency of the system was measured as a function of the distance between the bottom collimator window and the upper circular surface of the calibration source, embracing the range from 0 to 9 cm. This efficiency calibration function was employed to determine the absolute values of the product $A_j\varepsilon_j$ at the center of each plate meat of a spent fuel element during the measurements performed at the reactor storage pool.

The exclusive utilization of ^{137}Cs in the efficiency calibration procedure was consequence of a decision to use this radionuclide as the only burnup monitor. Two circumstances led to such decision: the lack of calibration sources of other burnup monitors (Terremoto et al., 2000) with shape suitable for the efficiency calibration procedure and the intent to test the adequacy of using ^{137}Cs as burnup monitor even for spent fuel elements with cooling times much shorter than two years.

After the efficiency calibration has been concluded, the collimator tube and the electronic components of the gamma scanning system were transported back to the reactor storage pool and assembled there once again. The energy calibration of the system was then carried out according to the standard procedure (Knoll, 1989), using punctiform calibration sources of ^{137}Cs, ^{60}Co and ^{152}Eu, as well as the high energy gamma-rays of ^{24}Na, an activation product (Perrotta et al., 1998; Terremoto et al., 2000; Zeituni et al., 2004) present with very low activity in the reactor storage pool.

3.3. Test of the efficiency calibration reproducibility

A procedure created in IPEN/Peru was employed to test the efficiency calibration reproducibility after the complete assembling of the gamma scanning system at the reactor storage pool.

According to this procedure, the sealed calibration source of ^{137}Cs previously used in the laboratory was encased tightly in a notch machined at the centre of a squared Plexiglas holder containing a circular mortise with the same external diameter as the bottom collimator and a long nylon string attached to each one of its four corners.

Promptly thereafter, the holder was immersed in the reactor storage pool and coupled to the bottom collimator using the strings, which were then symmetrically tied to the upper structure of the stainless steel holders of the gamma scanning system.

This arrangement, whose main components are shown schematically in Fig. 3, assures the precise positioning of the calibration source at the center of the aluminum window of the bottom collimator. Under such configuration, the efficiency of the gamma scanning system was determined by 3 complete measurements with duration of 30 min live time each, and the average result was compared with the one given by the efficiency calibration function for the distance equal to 0 cm.

Such procedure was repeated before every set of gamma-ray spectroscopy measurements on each spent fuel element. All results obtained from efficiency measurements performed at the reactor storage pool showed good agreement with the corresponding one obtained from the efficiency calibration function within an experimental uncertainty of approximately 2%.

Once the test of the efficiency calibration reproducibility has been successfully completed, the calibration source of ^{137}Cs was removed from the reactor storage pool and hoarded in a lead shielding located faraway outside the water, while a spent fuel

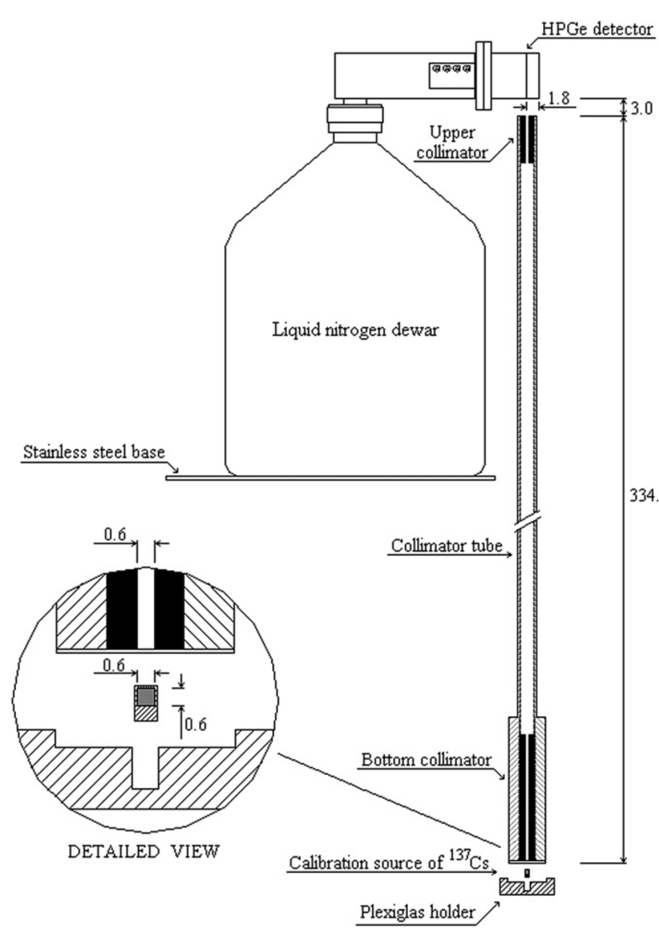

Fig. 3. Schematic diagram of the main components of the arrangement assembled in order to test the efficiency calibration reproducibility. All dimensions are given in cm.

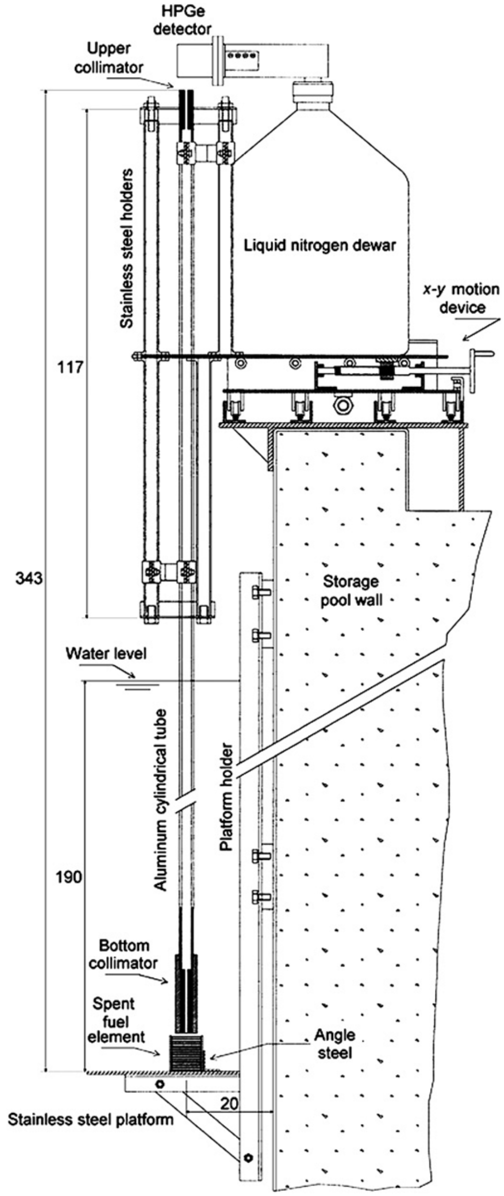

Fig. 4. Cross-sectional diagram of the gamma scanning system installed at the storage pool area of the RP-10 research reactor. All dimensions are given in cm.

element was positioned over an immerse stainless steel platform for measuring.

3.4. Gamma-ray spectroscopy measurements on spent fuel elements

The gamma scanning system has a stainless steel base fixed at the border of the reactor storage pool and equipped with two perpendicular crank driven mechanisms forming a x–y frame that enables the movement of the detection set (collimator tube + HPGe detector) either parallel or normal to the axial direction of a spent fuel element horizontally positioned.

In order to perform the gamma-ray spectroscopy measurements, a spent fuel element previously selected was hoisted up from the rack located at the bottom of the reactor storage pool and brought to an immerse stainless steel platform, where it was positioned horizontally at a depth of 1.9 m with the fuel plates

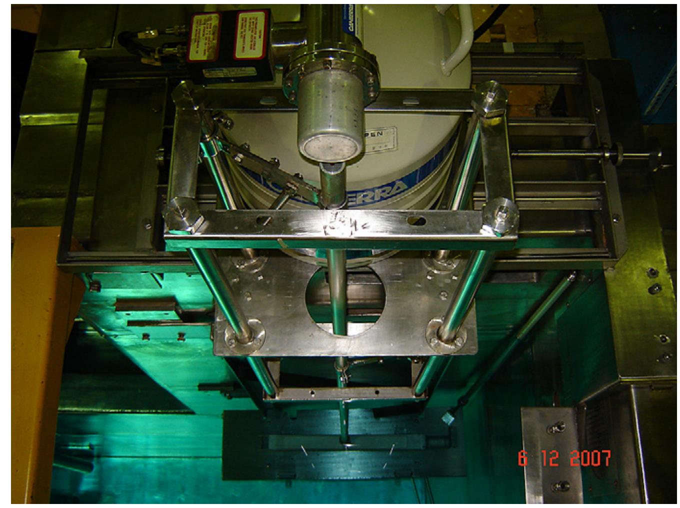

Fig. 5. Photograph of the gamma scanning system, showing the detection set (collimator tube + HPGe detector) positioned for measurement at the central point of a spent fuel element.

surface-upwards and perpendicular to the collimator tube axis. Reproducibility in the positioning of the spent fuel element was assured by angle steels welded at the platform ground. Gamma-ray spectroscopy measurements were performed in the configuration shown schematically in Fig. 4.

In this configuration, the distance between the bottom collimator window and the last plate of the fuel element, as already mentioned, was $C = 0.35 \pm 0.05$ cm. The uncertainty is compatible with recommendations (Rasmussen et al., 1966) that the reproducibility of the relative positioning between the bottom collimator window and the last plate of the fuel element must be assured within an error lower than $\pm$ 0.16 cm.

Fig. 5 presents a photograph of the gamma scanning system, where a spent fuel element positioned for measurement is shown.

Gamma-ray spectroscopy measurements on each one of the spent fuel elements were performed following 3 equidistant parallel rows along the active length: a row at the central axis and 2 side rows. The distance between successive rows was 2.5 cm. Every row embraced 24 gamma-ray spectra, each one obtained from a measurement with duration of 150 s of live time performed on a measuring point. The dead time of the HPGe detector remained always below 4% in all measurements.

For each gamma-ray spectrum obtained from measurements performed across the 3 rows along the active length of a spent fuel element, the net number of counts (area) under the full-energy peak of 661.6 keV was determined online by means of the commercial software Genie™ 2000, developed by CANBERRA and commonly used in the analysis of gamma-ray spectra.

The average value of the number of counts for each row was determined integrating the net number of counts along the active length and, subsequently, dividing the result by the total active length $l = 61.5$ cm of the spent fuel element. In order to obtain the

Table 1
Main properties of ^{137}Cs: half-life $T_{1/2}$, average yield Y for fission of ^{235}U by thermal neutrons, energy E_γ and absolute emission intensity I_γ of the gamma-ray emitted (Reus and Westmeier, 1983; Tasaka et al., 1983; Compilation and Evaluation of Fission Yield Nuclear Data, 2000).

Burnup monitor	$T_{1/2}$	Y (%)	E_γ (keV)	I_γ
^{137}Cs	30.14 years	6.21 ± 0.03	661.6	0.851

Table 2
Mass attenuation coefficient (μ/d), density (d) and linear attenuation coefficient (μ) for aluminum, water and meat material, considering gamma-rays of 661.6 keV (Knoll, 1989; Hubbell and Berger, 1968).

Material	μ/d (cm^2/g)	d (g/cm^3)	μ (cm^{-1})
Al	0.0749	2.690	0.2015
H_2O	0.0894	1.000	0.0894
Dispersion U_3O_8 in Al	0.1035	4.358	0.4510

average number of counts under the full-energy peak of 661.6 keV for the whole measurement, an arithmetical mean of the average values for the 3 rows was calculated. Thereafter, the spent fuel element was turned 180° around its axis and the entire measurement was repeated.

All spent fuel elements currently at the storage pool of the RP-10 research reactor have been submitted to gamma-ray spectroscopy measurements. The corresponding data were registered for later reduction in order to obtain the average value of the total number of counts per unit of time $\overline{Q}$ and, thereafter, to determine the average burnup of the spent fuel element (see part 2).

3.5. Algorithm utilized for data reduction

The algorithm utilized at IPEN/Peru for data reduction employs the equations already presented in detail (see part 2) adapted to the software MATHCAD. Besides the values of $A_j\varepsilon_j$ obtained from the efficiency calibration function (see part 3, Section 3.2), these equations include parameters related to the main design characteristics of the spent fuel elements (see part 1).

Operation records of the RP-10 research reactor were also examined in order to reconstruct the detailed irradiation history of each spent fuel element. Calculations performed with the software EXCEL enabled the use of this information to obtain the correction factor f (see part 2, Section 2.3), whose value was employed as input of the algorithm.

Following the decision to use ^{137}Cs as the only burnup monitor (see part 3, Section 3.2), its relevant properties regarding gamma-ray spectroscopy were reviewed (Reus and Westmeier, 1983; Tasaka et al., 1983; Compilation and Evaluation of Fission Yield Nuclear Data, 2000), listed in Table 1 and included in the algorithm.

Moreover, special attention was dedicated to the properties of structural materials of each spent fuel element and water concerning the attenuation of gamma-rays with energy of 661.6 keV. These properties (Knoll, 1989; Hubbell and Berger, 1968), also included in the algorithm, are joined in Table 2.

4. Results and discussion

Gamma-ray spectroscopy measurements on spent fuel elements were performed only during long maintenance periods in which the RP-10 research reactor did not operate, in order to reduce the pool background caused mainly by the activation product ^{24}Na ($T_{1/2} = 15.02$ h) generated in the operating reactor core (Perrotta et al., 1998; Terremoto et al., 2000; Zeituni et al., 2004).

A typical gamma-ray spectrum obtained from measurements on spent fuel elements is presented in Fig. 6, resulting from a run of 3600 s of live time carried out at the central point of the fuel element NN 008 only 106 days after the end of the last irradiation period, with its peaks identified in Table 3. Counting time was longer than employed for ordinary measurements in order to provide an undoubted identification of the photons detected. The full-energy peak of 661.6 keV appears clearly and undistorted in the spectrum, with an uncertainty of 0.2% in the net number of counts and a full width at half maximum of 1.5 keV. This evidence corroborates the reliable use of ^{137}Cs as burnup monitor even for spent fuel elements with cooling times much shorter than two years, as reported more recently (Henríquez et al., 2001; Pereda et al., 2004, 2008).

The areas under the full-energy peaks of 661.6 keV, presented for each measurement row as a function of the active length of a spent fuel element, constitute its burnup profiles, because ^{137}Cs activity is directly proportional to the integrated neutron flux (Phillips, 1991) and therefore to burnup (Matsson and Grapengiesser, 1997). As an example, Fig. 7 shows the burnup profiles for the fuel element NN 008, obtained from measurements of 150 s of live time performed 175 days after the end of the last irradiation period.

Once finished all measurements on every spent fuel element currently stored in the racks of the reactor storage pool, the final

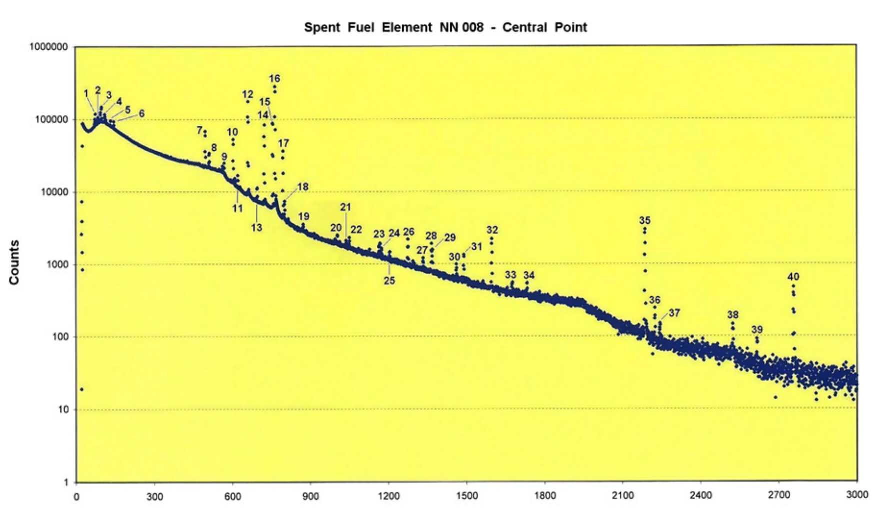

Fig. 6. Gamma-ray spectrum obtained from measurement of 3600 s of live time performed at the central point of the spent fuel element NN 008 with 106 days of cooling time. The origin of each peak is identified in Table 3.

Table 3
Number of each peak in the spectrum of Fig. 6, together with the corresponding photon energy, radionuclide identification and origin (Reus and Westmeier, 1983).

Number	Energy (keV)	Radionuclide	Origin
1	74.969	–	Lead Kα
2	84.8	–	Lead Kβ
3	98.439	–	Uranium Kα
4	111.0	–	Uranium Kβ
5	133.5	^{144}Ce	Fission product
6	145.4	^{141}Ce	Fission product
7	497.1	^{103}Ru	Fission product
8	511.0	–	Annihilation e^-/e^+
9	569.3	^{134}Cs	Fission product
10	604.7	^{134}Cs	Fission product
11	621.9	^{106}Ru	Fission product
12	661.6	^{137}Cs	Fission product
13	696.5	^{144}Ce	Fission product
14	724.2	^{95}Zr	Fission product
15	756.7	^{95}Zr	Fission product
16	765.8	^{95}Nb	Fission product
17	795.8	^{134}Cs	Fission product
18	801.9	^{134}Cs	Fission product
19	873.2	^{154}Eu	Fission product
20	1004.8	^{154}Eu	Fission product
21	1038.6	^{134}Cs	Fission product
22	1050.3	^{106}Ru	Fission product
23	1167.9	^{134}Cs	Fission product
24	1173.2	^{60}Co	Activation product
25	1204.8	^{91}Y	Fission product
26	1274.5	^{154}Eu	Fission product
27	1332.5	^{60}Co	Activation product
28	1365.1	^{134}Cs	Fission product
29	1368.5	^{24}Na	Activation product
30	1460.8	^{40}K	Natural background
31	1489.2	^{144}Ce	Fission product
32	1596.5	^{140}Ba	Fission product
33	1674.7	–	SE ^{144}Ce 2185.7 keV
34	1731.9	–	DE ^{24}Na 2753.9 keV
35	2185.7	^{144}Ce	Fission product
36	2223.25	–	H (n, γ) D
37	2242.9	–	SE ^{24}Na 2753.9 keV
38	2521.7	^{140}Ba	Fission product
39	2614.6	^{208}Tl	Natural background
40	2753.9	^{24}Na	Activation product

values obtained for the average burnup were directly compared against corresponding ones furnished by reactor physics calculations (Determination of Research Reactor Fuel Burnup, 1992; Padilla, 2008a,b), with the results shown in Table 4.

The reactor physics calculations necessary to obtain the average burnup for each spent fuel element were performed by means of the computer codes CITATION, WIMSD4 and a utilitarian named WIMCIT. Cross sections are generated using WIMSD4 and provided as input data for CITATION, which is employed to calculate the average burnup in MWD/MTU achieved by every fuel element while inside the reactor core. Regarding each spent fuel element, this value of the average burnup is employed as input for WIMSD4 in order to calculate the corresponding isotopic composition and, therefore, to obtain the percentage of the initial number of ^{235}U nuclei that undergone fission. Calculations with WIMSD4 are performed in one-dimensional slab geometry considering reflective boundary conditions (null neutronic current) and critical effective multiplication factor, whereas calculations with CITATION are performed in two-dimensional Cartesian geometry surrounded by 20 cm of water and also considering reflective boundary conditions (Determination of Research Reactor Fuel Burnup, 1992; Padilla, 2008a,b).

Results shown in Table 4 demonstrate, for gamma-ray spectroscopy measurements on each spent fuel element, a good reproducibility within the experimental error limits. Moreover, the average relative experimental uncertainty for the final values amounts to merely 3.1%. The main contributions to this overall uncertainty arise from the efficiency calibration and from the evaluation of attenuation effects, as usual in nondestructive absolute average burnup measurements by gamma-ray spectroscopy. In the case of measurements performed on spent fuel elements of the RP-10 research reactor, the average relative experimental uncertainties in the efficiency calibration and in the evaluation of attenuation effects are both approximately equal to 2%.

Data presented in Table 4 also include the ratio gamma-ray spectroscopy final value/reactor physics calculations of the average burnup for each spent fuel element. The mean deviation between measured and calculated values of the average burnup for the spent fuel elements of the RP-10 research reactor is 6.0%.

Analogous results have been obtained from other recent burnup measurements using nondestructive gamma-ray spectroscopy on irradiated plate-type MTR fuel elements, although employing the activity ratio technique, which enables calculation of the average burnup using measurements of quotients between activities of $^{95}Zr/^{137}Cs$ and $^{134}Cs/^{137}Cs$ (Iqbal et al., 2001) or $^{134}Cs/^{137}Cs$ alone (Ansari et al., 2007), based on the fact that buildups of ^{95}Zr, ^{137}Cs and ^{134}Cs during irradiation are, respectively, directly proportional

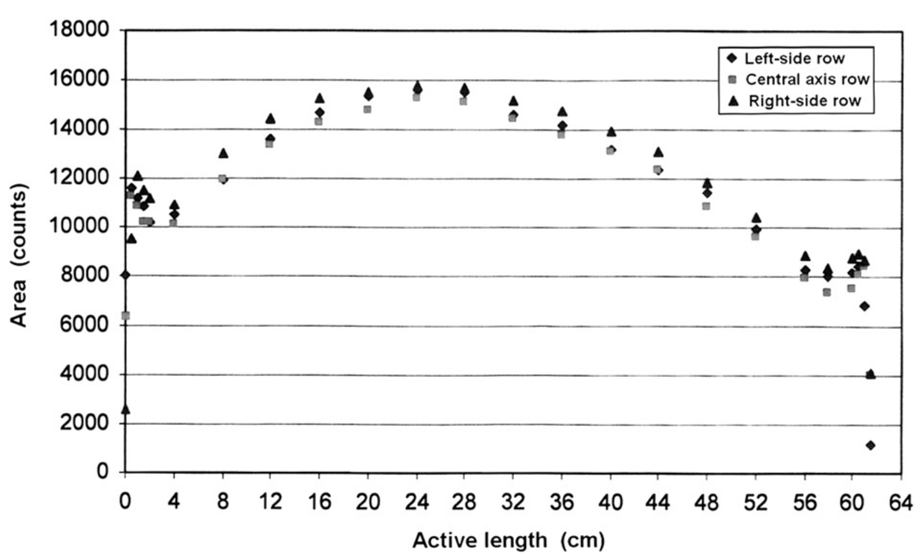

Fig. 7. Experimental burnup profiles for the spent fuel element NN 008, obtained from measurements of 150 s of live time, performed with 175 days of cooling time and following three equidistant parallel rows along the active length. The distance between successive rows is 2.5 cm.

Table 4
Average burnup values obtained by means of gamma-ray spectroscopy measurements compared against corresponding ones furnished by reactor physics calculations.

Spent fuel element	Irradiation history (days)	Storage date (d/month/yr)	Measuring date (d/month/yr)	Average burnup		Ratio of burnup values (measured/calculated)
				Gamma-ray spectroscopy (%)	Reactor physics calculations (%)	
NN 001	316	15/03/2006	16/11/2006	46.83 ± 3.69	49.52	0.929 ± 0.024
			09/03/2007 14/03/2007	45.95 ± 2.56		
			21/06/2007	46.85 ± 2.59		
			09/08/2007	46.19 ± 2.56		
			22/11/2007	44.70 ± 2.48		
			Mean value	46.00 ± 1.20		
NN 002	275	12/06/2003	15/11/2006	42.84 ± 3.38	47.32	0.907 ± 0.025
			16/03/2007	42.14 ± 3.32		
			21/06/2007	44.01 ± 2.44		
			10/08/2007	43.84 ± 2.43		
			28/11/2007	41.60 ± 2.31		
			Mean value	42.94 ± 1.19		
NN 003	275	12/06/2003	09/11/2006	44.29 ± 3.49	47.63	0.912 ± 0.032
			17/03/2007	42.89 ± 3.38		
			05/07/2007	43.71 ± 2.42		
			17/08/2007	42.60 ± 3.34		
			Mean value	43.42 ± 1.52		
NN 004	275	12/06/2003	28/10/2006	48.20 ± 3.80	49.70	0.959 ± 0.031
			09/03/2007	47.15 ± 3.72		
			05/07/2007	48.31 ± 2.68		
			23/08/2007	47.05 ± 2.61		
			Mean value	47.66 ± 1.53		
NN 008	317[a]	23/08/2007	07/12/2007	44.61 ± 3.50	45.34	0.993 ± 0.032
			14/02/2008	45.30 ± 2.52		
			24/04/2008	44.57 ± 2.48		
			16/07/2008	45.69 ± 3.58		
			Mean value	45.00 ± 1.45		
NN 009	320	13/07/2006	13/03/2008	46.35 ± 2.57	47.04	0.998 ± 0.032
			30/05/2008	48.42 ± 3.80		
			05/02/2009	47.28 ± 2.64		
			16/04/2009	46.28 ± 3.65		
			Mean value	46.97 ± 1.51		
NN 015	333	23/08/2007	16/12/2009	40.91 ± 2.27	45.95	0.920 ± 0.030
			08/02/2010	43.20 ± 2.40		
			16/03/2010	42.80 ± 2.38		
			Mean value	42.26 ± 1.36		
NC 003[b]	327	16/03/2007	11/04/2008	42.67 ± 2.37	45.34	0.926 ± 0.033
			17/07/2008	42.20 ± 2.35		
			03/12/2008	40.27 ± 3.18		
			Mean value	41.97 ± 1.48		
NC 005[b]	316	15/03/2006	05/12/2007	46.36 ± 2.58	50.93	0.918 ± 0.026
			06/03/2008	47.21 ± 2.62		
			29/05/2008	46.00 ± 3.62		
			04/02/2009	46.15 ± 3.67		
			17/04/2009	47.50 ± 2.64		
			Mean value	46.77 ± 1.30		

[a] Remained outside the reactor core between 16/11/2001 and 04/09/2002.
[b] Control fuel elements.

to the neutron flux, to the neutron fluence and to the square of the neutron fluence (Iqbal et al., 2001). According to these experiments, the mean deviation between measured and calculated values of the average burnup for 5 spent fuel elements is 5.9% (Iqbal et al., 2001; Ansari et al., 2007).

5. Conclusion

Nondestructive gamma-ray spectroscopy was employed in order to measure the absolute average burnup of all spent MTR fuel elements irradiated in the RP-10 research reactor and currently stored under water in the racks of the reactor storage pool. The performed measurements embrace 7 standard and 2 control MTR fuel elements.

Measurements were carried out at the reactor storage pool area using ^{137}Cs as the only burnup monitor, even for spent fuel elements with cooling times as short as 106 days. Extensive tests concerning the reproducibility of measurements exhibited good results.

As a result of the measurements, values of the average burnup were obtained with a mean relative experimental uncertainty of 3.1%. Compared against corresponding ones furnished by reactor physics calculations, a mean deviation of 6.0% arose, which is compatible with results that have been obtained from other recent burnup measurements using nondestructive gamma-ray spectroscopy on irradiated plate-type MTR fuel elements. This evidence indicates that it is not necessary to wait approximately 2 years after the definitive withdrawal of a fuel element from the reactor core in order to perform average burnup measurements based on ^{137}Cs.

Successful accomplishment of all requisites necessary for absolute gamma-ray spectroscopy measurements, as described along the present work, demonstrates that the gamma scanning system of IPEN/Peru is fully adequate to measure the average burnup of spent MTR fuel elements and, consequently, can be employed for this purpose in the RP-10 research reactor.

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