Braz J Phys (2011) 41:38–43
DOI 10.1007/s13538-011-0002-9

NUCLEAR PHYSICS

Measurements of Fission and Radioactive Capture Reaction Rates Inside the Fuel of the Ipen/MB-01

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Received: 18 August 2010 / Published online: 14 April 2011

Abstract This work presents the measures of the nuclear reaction rates along the radial direction of the fuel pellet by irradiation and posterior gamma spectrometry of a thin slice of fuel pellet of UO_2 at 4.3% enrichment. From its irradiation, the rate of radioactive capture and fission had been measured as a function of the radius of the pellet disk using a Ortec GMX HPGe detector. Lead collimators had been used for this purpose. Simulating the fuel pellet in the pin fuel of the IPEN/MB-01 reactor, a thin UO_2 disk is used, being inserted in the interior of a dismountable fuel rod. This fuel rod is then placed in the central position of the IPEN/MB-01 reactor core and irradiated during 1 h under a neutron flux of 5×10^8 n/cm^2 s. In gamma spectrometry, 10 collimators with different diameters have been used; consequently, the nuclear reactions of radioactive capture that occurs in atoms of ^{238}U and the fission that occurs on both ^{235}U and ^{238}U are measured in function of 10 different regions (diameter of collimator) of the UO_2 fuel pellet disk. Nuclear fission produces different fission products such as ^{143}Ce with a yield fission of 5.9% which decay is monitored in this work. Corrections in geometric efficiency due to introduction of collimators on HPGe detection system were estimated using photon transport of MCNP-4C code. Some calculated values of nuclear reaction rate of radioactive capture and fission along the radial direction of the fuel pellet obtained by Monte Carlo methodology, using the MCNP-4C code, are presented and compared to the experimental data showing very good agreement.

Keywords Fission rate · Radioactive capture rate · Nuclear fuel

1 Introduction

Experiments involving the determination of the reaction rates in the fuel pellets are of fundamental importance to correlate theory and experiment, mainly concerning calculation methods and related to nuclear data libraries. For a long time, experiments involving reaction rate measurements have been carried out worldwide. The most famous spectral index measurements are the performed in the TRX and BAPL critical facilities, selected by the CSEWG [1], as benchmarks. Historically, there has been a long-standing problem related to the over prediction of the spectral indices $^{28}\rho$. These spectral indices provide the ratio of epithermal to thermal neutron capture in ^{238}U. The epithermal calculated value is obtained by calculations of self-shielding factors, at resonances of the ^{238}U, using methods such as NORDHEIM [2] and BONDARENKO [3], which overestimate the radioactive capture reaction rate in this neutron energy range. Another spectral index of great importance is $^{25}\delta$. It provides the ratio of epithermal to thermal neutron fission in ^{235}U, and it is commonly used as a base to test benchmarks. Generally,

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estimations of the nuclear reaction rate along the radius of the nuclear reactor fuel pellet are made by MCNP code and its libraries of nuclear associated data such as ENDF/B, JENDL, JEFF, and others.

Therefore, the experimental measurements, although very rare and difficult, are very important to estimate the level of accuracy and precision of the calculations methodology and its nuclear data libraries. This work aims to measure the relative nuclear reaction rate along the radius fuel pellet and to compare with calculations performed by MCNP-4C Code [4] and give some preliminary values of $^{28}\rho$ and $^{25}\delta$.

2 Experimental Description

The IPEN/MB-01 reactor is a zero power reactor, especially designed for measurements of a wide range of reactor physics parameters, to be used as benchmark experimental data for verifying the calculation methodologies and related nuclear data libraries, commonly used in the field of reactor physics. This facility consists of an array of 28 × 26 UO_2 fuel rods, 4.3% enriched and clad by stainless steel (type 304), inside a light water tank. A complete description of the IPEN/MB-01 reactor may be found elsewhere [5].

The experiments were carried out at the asymptotic region of the reactor core. An experimental fuel rod (similar to the one used in the reactor) is irradiated at the central position of the core. Exactly in the axial active fuel quote, 94 mm of a very thin UO_2 pellet (about 0.5 mm thickness) is inserted to measure nuclear capture and fission reaction rate. This thin pellet (4.3% enriched uranium) is inserted between the ninth and tenth axial fuel pellet. This position was chosen because it is enough far to feel the control rods disturbance. Two irradiations are necessary to determinate reaction rates induced by thermal neutrons. In the first irradiation, a cadmium glove is used in the experimental fuel rod to discriminate thermal and epithermal neutrons, and in the second irradiation, nothing is used to block the thermal neutrons.

The experimental fuel rod is irradiated for 1 h, in the maximum power level (100 W). After 19 h, the experimental fuel rod is withdrawn from the core and taken to the gloved box, for its dismounting so that a thin experimental pellet is obtained. Immediately after the dismounting, the gamma spectrometry of the thin pellet is started in an Ortec GMX-40210 HPGe detector. For each counting, a different collimator is used. The collimators used have diameters of 2.692, 3.823, 4.68, 5.373, 6.002, 6.577, 7.126, 7.654, 8.125, and 8.492 mm. Each collimator increases the exposed area in approximately 5.7 mm^2. The diameter of the thin pellet is 8.49 mm, exactly the same of the reactor UO_2 fuel pellet. Each gamma spectrometry data acquirement is made for 900 s, and in the minimum achievement, 40 gamma spectra were acquired to each collimator diameter. The gamma spectrometry measures the counts rate centered at 277.6 keV, with a gamma emission probability of 14.38% for neptunium 239 (half-life of 2.335), which is the product of neutron capture of ^{238}U, and it measures the counts rate centered at 293.3 keV, with a gamma emission probability of 42.8% for cerium 143 (half-life of 33.7 h), formed by fission of ^{235}U with yield of 5.9373%.

The radionuclide ^{239}Np occurs by decay of the ^{239}U atoms, a direct product from the nuclear reactions between the neutrons and the target nucleus of the ^{238}U. Using the expression (1), it is possible to estimate [6, 7] the absolute radioactive capture nuclear reaction rate (C_8), at the diameter measured by different collimators (see Fig. 1).

$$C_8 = \frac{{}^{U9}\lambda - {}^{Np}\lambda}{{}^{U9}\lambda} \times \frac{{}^{Np}\lambda \cdot {}^{Np}C \cdot \exp\left({}^{Np}\lambda \cdot t_e\right)}{{}^{Np}f\gamma \cdot {}^{Np}I \cdot {}^{Np}\eta \cdot \left[1 - \exp\left(-{}^{Np}\lambda \cdot t_i\right)\right]\left[1 - \exp\left(-{}^{Np}\lambda \cdot t_c\right)\right]} \tag{1}$$

where ${}^{Np}\lambda$ is the constant decay of the ^{239}Np, ${}^{U9}\lambda$ is the constant decay to ^{239}U, ${}^{Np}C$ is the integral counting of the gamma energy of 277.6 keV, ${}^{Np}f\gamma$ is the self-shielding factor at 277.6 keV from ^{239}Np, ${}^{Np}I$ is the gamma emission probability to gamma energy of the

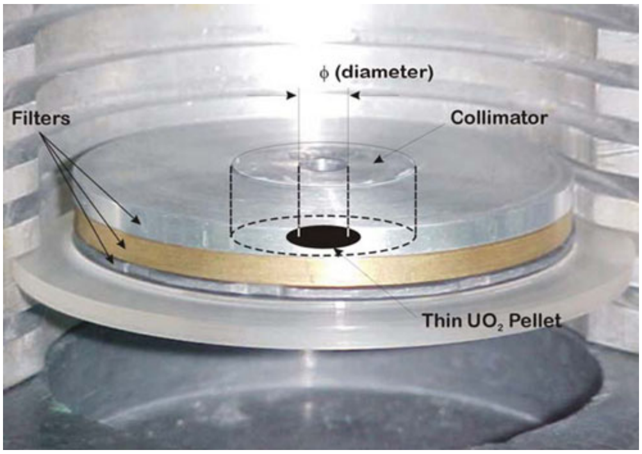

Fig. 1 Collimator used to measure gamma photopeak of the ^{239}Np (277.6 keV) formed by radioactive capture nuclear reaction in the thin pellet irradiated in the fuel rod

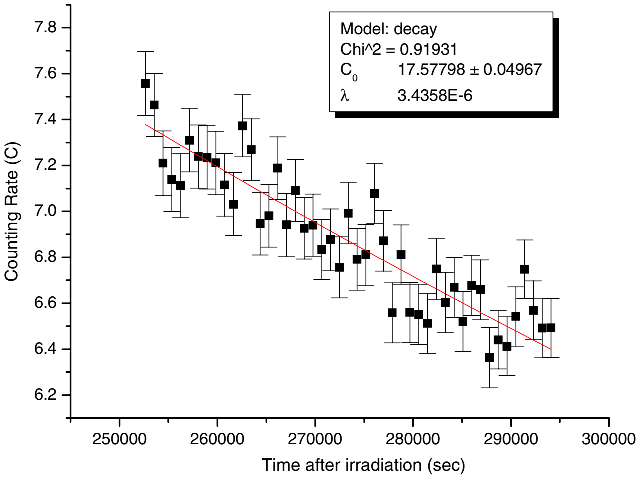

Fig. 2 Determination of C_0 to bare thin UO_2 pellet using a 6-mm-diameter collimator

277.6 keV from ^{239}Np, $^{\mathrm{Np}}\eta$ global efficiency to gamma photopeak of 277.6 keV from ^{239}Np, t_e is the wait time to counting, t_i is the time irradiation, and t_c is the time of counting.

Using the expression (2) is possible to estimate [6, 8, 9] the absolute fission nuclear reaction rate (F) at the diameter measured.

$$F = \frac{C_0}{{}^{\mathrm{Ce}}Y \cdot {}^{\mathrm{Ce}}f\gamma \cdot {}^{\mathrm{Ce}}I \cdot {}^{\mathrm{Ce}}\eta \cdot \left[1 - \exp(-{}^{\mathrm{Ce}}\lambda . \cdot t_i)\right]} \qquad (2)$$

where $^{\mathrm{Ce}}f\gamma$ is the self-shielding factor for the rod in the photopeak of 293.3 keV for ^{143}Ce, $^{\mathrm{Ce}}I$ is the gamma emission probability for this energy , $^{\mathrm{Ce}}\eta$ is the global efficiency for 293.3 keV, $^{\mathrm{Ce}}\lambda$ is the decay constant for ^{143}Ce, $^{\mathrm{Ce}}Y$ is the fission yield, and t_i is the irradiation time.

The introduction of collimators in the Ortec GMX HPGe detector system causes a reduction in its geometric efficiency [10]. So the count values obtained during the counting process should be corrected. For that, the system was modeled using the code MCNP-4C. A source of photons was included in this modeling to compare the efficiencies between measurements with and without the collimators. The ratio between these calculated efficiency make it possible to obtain correction factors for each of collimators. The efficiencies were determined using the f8 tally (pulse height tally) which counts the number of particles that have deposited a certain bin of energy in the germanium crystal. Statistical uncertain derived from this correction factors used a correlation factor of 1.

3 Experimental Results

The relative reaction rate, along the radius of the thin UO_2 pellet irradiated, was measured by its gamma spectrometry, using several collimators with different diameters (see Fig. 1). Then, the problem is to estimate the counting rate in the end of irradiation (C_0), discounting the dead time. This parameter was estimated using the expression (3), where t_c is the life time counting (without dead time) in the gamma spectrometry.

$$C_0 = C \cdot \exp({}^{\mathrm{Np}}\lambda \cdot t_e) \qquad (3)$$

Figure 2 shows an example of the gamma spectrometry made by acquisition 48 spectrum data using the 6.577-mm-diameter collimator building the ^{239}Np decay curve.

Table 1 Relative nuclear reaction rate of radioactive capture in UO_2 disk irradiated without cadmium covered (bare) at central position of the IPEN/MB-01 reactor core—axial quote 94 mm

Radius of the collimator used to sample the UO_2 disk (mm)	Relative radioactive capture nuclear reaction rate (C_0)[a]	Normalized values of C_0 (%)	Absolute radioactive capture nuclear reaction rate (C_8)[b]	Normalized calculated relative radioactive capture nuclear reaction rate (MCNP-4C)
2.692 ± 0.005	6.37 ± 0.83	0.062 ± 0.008	$(2.21 \pm 0.288) \times 10^5$	0.081 ± 0.009
3.823 ± 0.005	14.24 ± 0.64	0.139 ± 0.006	$(4.92 \pm 0.224) \times 10^5$	0.163 ± 0.013
4.68 ± 0.005	24.58 ± 0.63	0.239 ± 0.006	$(8.48 \pm 0.227) \times 10^5$	0.247 ± 0.016
5.373 ± 0.005	35.98 ± 0.74	0.351 ± 0.008	$(1.24 \pm 0.027) \times 10^6$	0.333 ± 0.018
6.002 ± 0.005	44.34 ± 0.80	0.433 ± 0.008	$(1.53 \pm 0.030) \times 10^6$	0.421 ± 0.020
6.577 ± 0.005	54.31 ± 0.88	0.53 ± 0.009	$(1.87 \pm 0.034) \times 10^6$	0.512 ± 0.023
7.126 ± 0.005	64.98 ± 0.95	0.634 ± 0.010	$(2.24 \pm 0.037) \times 10^6$	0.607 ± 0.025
7.654 ± 0.005	75.10 ± 1.05	0.733 ± 0.012	$(2.59 \pm 0.041) \times 10^6$	0.708 ± 0.027
8.125 ± 0.005	84.22 ± 1.08	0.822 ± 0.012	$(2.90 \pm 0.043) \times 10^6$	0.820 ± 0.029
8.492 ± 0.005	102.43 ± 1.07	1 ± 0.013	$(3.53 \pm 0.046) \times 10^6$	1 ± 0.032

[a] ^{239}Np (277.6 keV)
[b] 100 W power level—active fuel quote of 94 mm at central fuel rod

Table 2 Relative nuclear reaction rate of radioactive capture in UO_2 disk irradiated with cadmium covered at central position of the IPEN/MB-01 reactor core—axial quote 94 mm

Radius of the collimator used to sample the UO_2 disk (mm)	Relative radioactive capture nuclear reaction rate (C_0)[a]	Normalized values of C_0 (%)	Absolute radioactive capture nuclear reaction rate (C_8)[b]	Normalized calculated relative radioactive capture nuclear reaction rate (MCNP-4C)
2.692 ± 0.005	4.74 ± 0.40	0.070 ± 0.006	$(1.645 \pm 0.141) \times 10^5$	0.076 ± 0.010
3.823 ± 0.005	9.53 ± 0.42	0.141 ± 0.006	$(3.293 \pm 0.148) \times 10^5$	0.153 ± 0.015
4.68 ± 0.005	15.86 ± 0.40	0.235 ± 0.006	$(5.473 \pm 0.145) \times 10^5$	0.231 ± 0.018
5.373 ± 0.005	22.90 ± 0.47	0.339 ± 0.007	$(7.904 \pm 0.175) \times 10^5$	0.312 ± 0.021
6.002 ± 0.005	28.32 ± 0.52	0.419 ± 0.008	$(9.776 \pm 0.196) \times 10^5$	0.3955 ± 0.024
6.577 ± 0.005	34.46 ± 0.57	0.510 ± 0.009	$(1.189 \pm 0.022) \times 10^6$	0.482 ± 0.026
7.126 ± 0.005	41.52 ± 0.64	0.614 ± 0.011	$(1.433 \pm 0.025) \times 10^6$	0.574 ± 0.029
7.654 ± 0.005	47.96 ± 0.71	0.710 ± 0.012	$(1.655 \pm 0.028) \times 10^6$	0.673 ± 0.031
8.125 ± 0.005	52.35 ± 0.71	0.775 ± 0.012	$(1.807 \pm 0.029) \times 10^6$	0.788 ± 0.034
8.492 ± 0.005	67.58 ± 0.72	1 ± 0.0133	$(2.332 \pm 0.031) \times 10^6$	1 ± 0.038

[a] ^{239}Np (277.6 keV)

[b] 100 W power level—active fuel quote of 94 mm at central fuel rod

The final results of relative and absolute (C_8) radioactive capture nuclear reaction rate, with MCNP-4C correction and normalized by total counts to maximum diameter collimator (8.49 mm), may be seen in Table 1. Relative calculated values of radioactive capture along the radial direction of the fuel pellet was obtained using the MCNP-4C code [4] (Monte Carlo method), with the ENDF-BVI.8 nuclear data library; these data are also presented in Table 1. The same procedures were used to irradiate a thin UO_2 pellet, in the same experimental conditions and axial quote (94 mm), but covered with cadmium. The cadmium was used around the dismountable fuel rod and centered at quote 94 mm. The cadmium glove used was 7 cm long and 0.5 mm thick. The results obtained, after gamma spectrometry with MCNP-4C correction [4], may be seen in Table 2. Relative calculated values of radioactive capture along the radial direction of the fuel pellet with cadmium cover are also presented.

Figure 3 shows the comparison between calculated and experimental percentage of nuclear reaction rate of radioactive capture, along the diameter of the UO_2 disk, irradiated at central position of the core, exactly in the axial active fuel (quote 94 mm). As the same,

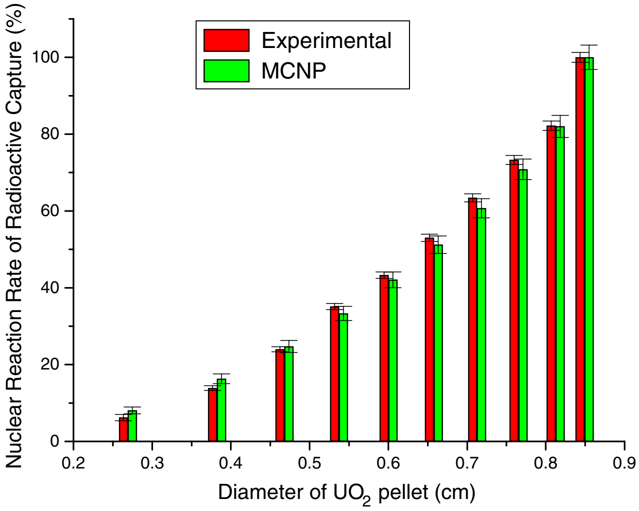

Fig. 3 Percentage of nuclear reaction rate of radioactive capture, along the diameter of the UO_2 pellet, irradiated at central position of the core

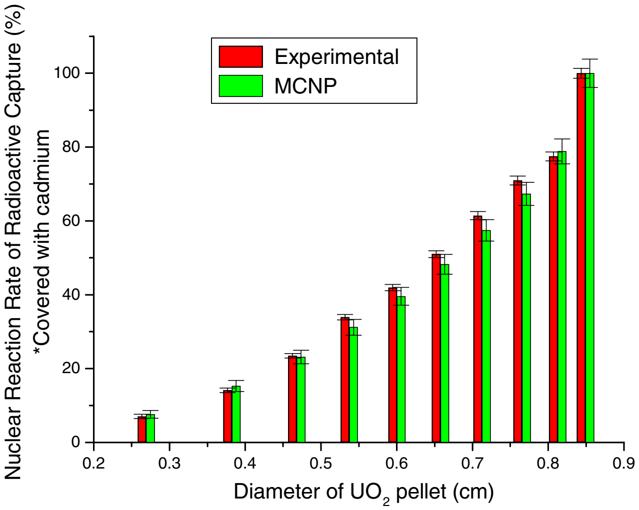

Fig. 4 Percentage of nuclear reaction rate of radioactive capture, along the diameter of the UO_2 pellet irradiated, covered with cadmium glove, at central position of the core

Table 3 Relative nuclear reaction rate of fission in UO_2 disk, irradiated without covered cadmium (bare), at central position of the IPEN/MB-01 reactor core—axial quote 94 mm

Radius of the collimator used to sample the UO_2 disk (mm)	Relative fission nuclear reaction rate (C_0)[a]	Normalized values of C_0 (%)	Absolute fission nuclear reaction rate (F)[b]	Normalized calculated relative fission nuclear reaction rate (MCNP-4C)
2.692 ± 0.005	9.63 ± 0.87	0.103 ± 0.009	$(1.109 \pm 0.102) \times 10^5$	0.093 ± 0.005
3.823 ± 0.005	16.56 ± 0.61	0.179 ± 0.007	$(1.902 \pm 0.074) \times 10^5$	0.188 ± 0.005
4.68 ± 0.005	27.27 ± 0.68	0.293 ± 0.008	$(3.128 \pm 0.086) \times 10^5$	0.284 ± 0.008
5.373 ± 0.005	38.75 ± 0.78	0.416 ± 0.010	$(4.444 \pm 0.104) \times 10^5$	0.381 ± 0.010
6.002 ± 0.005	47.86 ± 0.84	0.514 ± 0.019	$(5.489 \pm 0.117) \times 10^5$	0.480 ± 0.011
6.577 ± 0.005	57.46 ± 0.91	0.618 ± 0.012	$(6.59 \pm 0.132) \times 10^5$	0.580 ± 0.012
7.126 ± 0.005	66.93 ± 0.98	0.720 ± 0.013	$(7.677 \pm 0.146) \times 10^5$	0.682 ± 0.013
7.654 ± 0.005	76.74 ± 1.05	0.825 ± 0.015	$(8.802 \pm 0.160) \times 10^5$	0.786 ± 0.014
8.125 ± 0.005	83.53 ± 1.03	0.898 ± 0.015	$(9.581 \pm 0.166) \times 10^5$	0.892 ± 0.015
8.492 ± 0.005	93.01 ± 1.07	1 ± 0.016	$(10.668 \pm 0.178) \times 10^5$	1 ± 0.016

[a] ^{143}Ce (293.3 keV)

[b] 100 W power level—active fuel quote of 94 mm at central fuel rod

Table 4 Relative nuclear reaction rate of fission in UO_2 disk, irradiated with covered cadmium (bare), at central position of the IPEN/MB-01 reactor core—axial quote 94 mm

Radius of the collimator used to sample the UO_2 disk (mm)	Relative fission nuclear reaction rate (C_0)[a]	Normalized values of C_0 (%)	Absolute fission nuclear reaction rate (F)[c]	Normalized calculated relative fission nuclear reaction rate (MCNP-4C)
2.692 ± 0.005	–[b]	–[b]	–[b]	0.099 ± 0.006
3.823 ± 0.005	2.39 ± 0.15	0.189 ± 0.013	$(0.273 \pm 0.019) \times 10^5$	0.199 ± 0.008
4.68 ± 0.005	3.92 ± 0.13	0.311 ± 0.012	$(0.448 \pm 0.018) \times 10^5$	0.298 ± 0.010
5.373 ± 0.005	5.56 ± 0.16	0.440 ± 0.015	$(0.636 \pm 0.023) \times 10^5$	0.398 ± 0.012
6.002 ± 0.005	6.84 ± 0.16	0.542 ± 0.017	$(0.782 \pm 0.025) \times 10^5$	0.497 ± 0.013
6.577 ± 0.005	8.08 ± 0.17	0.640 ± 0.0190	$(0.924 \pm 0.029) \times 10^5$	0.598 ± 0.015
7.126 ± 0.005	9.02 ± 0.18	0.715 ± 0.020	$(1.032 \pm 0.032) \times 10^5$	0.698 ± 0.016
7.654 ± 0.005	10.03 ± 0.25	0.795 ± 0.025	$(1.148 \pm 0.039) \times 10^5$	0.798 ± 0.017
8.125 ± 0.005	11.08 ± 0.22	0.878 ± 0.025	$(1.267 \pm 0.038) \times 10^5$	0.899 ± 0.018
8.492 ± 0.005	12.62 ± 0.25	1 ± 0.028	$(1.443 \pm 0.044) \times 10^5$	1 ± 0.019

[a] 143Ce (293.3 keV)

[b] Low counts during gama spectrometry

[c] 100 W power level—active fuel quote of 94 mm at central fuel rod

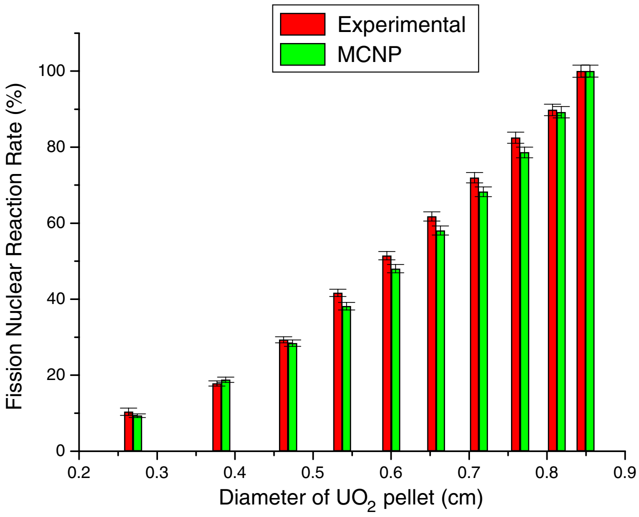

Fig. 5 Percentage of nuclear fission reaction rate along the diameter of the UO_2 pellet, irradiated at central position of the core

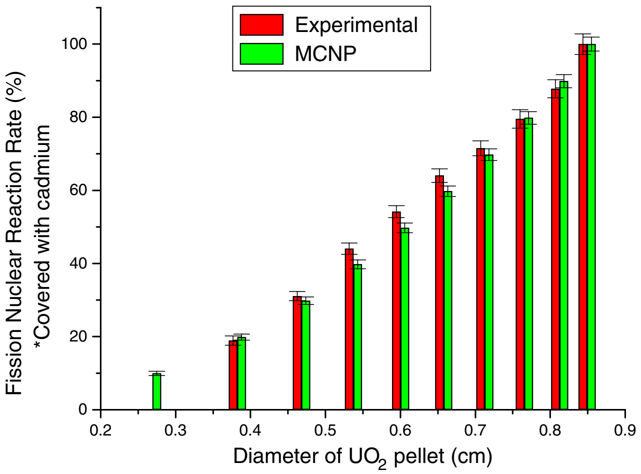

Fig. 6 Percentage of nuclear fission reaction rate along the diameter of the UO_2 pellet irradiated, covered with cadmium glove, at central position of the core

Fig. 4 shows comparison between calculated and experimental results obtained to UO_2 disk, irradiated with the cadmium glove.

Both total and epithermal rate of radioactive capture showed a high agreement between calculated and experimental data, differing mostly in the region between 5.373 and 7.126 mm.

The final results of the relative nuclear fission rate, with MCNP-4C correction and normalized by total counts to maximum diameter collimator (8.49 mm), may be seen in Table 3, without cadmium cover, and in Table 4, with cadmium cover. Thus, in this situation, the whole foil diameter is sampled and the counts (293.3 keV to ^{143}Ce) are proportional to the nuclear fission reaction rate. The cadmium glove was the same used to determine the radioactive capture in ^{238}U. Relative calculated values of fission along the radial direction of the fuel pellet was obtained using the MCNP-4C code [4] (Monte Carlo method), with the ENDF-BVI.8 nuclear data library; these data are also presented in Tables 3 and 4.

Figure 5 shows the comparison between calculated and experimental percentage of fission nuclear reaction rate, along the diameter of the UO_2 disk, irradiated at central position of the core, exactly in the axial active fuel (quote 94 mm). As the same, Fig. 6 shows comparison between calculated and experimental results obtained to UO_2 disk, irradiated with the cadmium glove.

The results obtained for nuclear fission rate have good agreement between experimental and calculated data in border region. Experimental data from collimators with 2.692 and 3.823 mm have high uncertain and should be examined carefully although that its agreement. Intermediate diameter collimators show the highest discrepancies where calculated values show systematical sub-estimation.

4 Conclusion

This work aims to show the present stage of nuclear reaction radioactive capture and fission rates measurements, along the nuclear fuel UO_2 pellet radius, of the IPEN/MB-01 reactor. This nuclear reaction rate measurement is very difficult to obtain experimentally because it requires the introduction of collimators in the HPGe gamma detection system causing a reduction in its geometric efficiency that should be corrected. The corrections obtained by MCNP-4C showed very good results when applied in experimental data.

The comparisons between experimental and calculated data showed very good agreement in the border regions of the UO_2 pellet and good results in the intermediate region. Collimators with small diameters showed high uncertain and should be examined carefully.

Actual results are important to be correlated with the calculation methodology. Then, the calculation results will be compared and its precision level will be estimated, using different nuclear data libraries.

Some improvements in the experimental measurements will be implemented. Titanium collimators with higher precision and more effective attenuating factors are beginning to be studied as possible substitutes for the actual lead ones. Irradiation with a higher power level will increase counts in collimators with smaller diameters reducing uncertain. Implementing these improvements in next experiments and make it reproductive would probably make these measurements a benchmark.

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