

MEASUREMENT AND CALCULATION OF THE RATE OF NUCLEAR REACTION OF RADIOACTIVE CAPTURE ALONG OF THE RADIUS OF UO₂ FUEL PELLETT AT IPEN/MB-01 REACTOR

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ABSTRACTS

This work presents the measures of the nuclear reaction rates along of the radial direction of the fuel pellet by irradiation and posterior gamma spectrometry of a thin slice of fuel pellet disk of UO₂. From its irradiation the rate of radioactive capture in the ²³⁸U as a function of the radius of the pellet disk is showed, using diverse lead collimators of changeable diameters. Simulating the fuel pellet, a thin 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 hour at the power level of 100 watts. The nuclear reaction of radioactive capture nuclear rate occurs in the atoms of ²³⁸U that when absorbs a neutron transmutes into ²³⁹U of half-life of only 23 minutes. Thus it is opted for the detection of the ²³⁹Np, radionuclide derivate of the radioactive decay of the ²³⁹U and has a measurable half-life (2,335 days). The monitored gamma photo peak is centered in the energy of 277.6 keV being emitted with a gamma emission probability of 14,38%. In the gamma spectrometry 11 collimators have been used for different diameters. Then of the gamma spectra made in function of the diameter (radius) of the irradiated UO₂ fuel pellet disk is possible to get the average value of the counting for each different collimators, function of the specific pellet radius . These values are directly proportional to the radioactive capture nuclear reaction rates. This work presents some calculated values of rate of nuclear reaction of radioactive capture along of the radial direction of the fuel pellet obtained by Monte Carlo methodology using the MCNP-4C code.

1. INTRODUCTION

Experiments involving determination of the reaction rates in the fuel pellets are fundamental importance to correlate theory and experiment mainly concerning calculation methods and related nuclear data libraries. For a long time, experiments involving reaction rates measurements have been carried out worldwide. The most famous spectral indices measurements are the ones 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 indice ²⁸ρ. This spectral indice provide the ratio to epithermal to thermal neutron capture in ²³⁸U. The epithermal calculated value is obtained by calculations of self-shielding factors at resonances of the ²³⁸U using methods as NORDHEIM [2] and BONDARENKO [3] that overestimate the radioactive capture reaction rate in this neutron energy range. The estimation of the nuclear reaction rate along of the radius of fuel pellet of nuclear reactor is made by MCNP code and its libraries of nuclear associated data such as ENDF/B, JENDL, JEFF, etc.

Therefore the experimental measurements besides 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 of the radius fuel pellet and to compare with calculations by MCNP-4C Code [4].

2. EXPERIMENTAL DESCRIPTION

The IPEN/MB-01 reactor is a zero power reactor especially designed for measurement of a wide range of reactor physics parameters to be used a benchmark experimental data for checking the calculation methodologies and related nuclear data libraries commonly used in the field of reactor physics. This facility consist of a array of 28x26 UO₂ 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) was irradiated at the central position of the core. Exactly in the axial active fuel quote 94 mm was inserted a very thin UO₂ pellet (about 0,5 mm thickness) to measure a capture nuclear reaction rate The thin pellet (natural Uranium) was put between the ninth and tenth axial fuel pellet. This position was choice because it is enough far to fell the control rods disturbance.

The experimental fuel rod was irradiated for 1 hour in the maximum power level (100 watts). After 19 hours the experimental fuel rod was withdraw of the core and leaved to gloved box to its dismounting to get the thin experimental pellet. Immediately the dismounting start the gamma spectrometry of the thin pellet. For each counting is used a different collimator. The collimators diameters used have a diameter varying 0.5 mm since 4 mm until 8.5 mm . The diameter of the thin pellet is 8.49 mm exactly the same of the UO₂ fuel pellet of the reactor. Each gamma spectrometry data acquirement was made for 900 seconds and in the minimum were acquired 32 gamma spectra to each diameter collimator. The gamma spectrometry measure the counts rate centered at 277.6 keV with a gamma emission probability of the 14.38%. This gamma energy is emitted by ²³⁹Np that has a half-Life of 2,335 days. This radionuclide occurs by decay of the ²³⁹U atoms a direct product of the nuclear reactions between the neutrons and the target nucleus of the ²³⁸U. Using the expression (1) is possible to estimate [6] the absolute radioactive capture nuclear reaction rate (C8) at the diameter measured by different collimators (see Figure 1).

$$C8 = \frac{{}^{U9}\lambda - {}^{Np}\lambda}{{}^{U9}\lambda} \frac{{}^{Np}\lambda \cdot {}^{Np}C \cdot \exp({}^{Np}\lambda \cdot te)}{{}^{Np}f\gamma \cdot {}^{Np}I \cdot {}^{Np}\eta \cdot [1 - \exp(-{}^{Np}\lambda \cdot ti)] [1 - \exp(-{}^{Np}\lambda \cdot tc)]} \quad (1)$$

Where, ^{Np}λ Constant decay of the ²³⁹Np; ^{U9}λ the constant decay to ²³⁹U, ^{Np}C integral counting of the gamma energy of 277.6 keV, ^{Np}f_γ the self-shielding factor at 276,6 keV from ²³⁹Np, ^{Np}I the gamma emission probability to gamma energy of the 277.6 keV from ²³⁹Np; ^{Np}η global efficiency to gamma photopeak of 276.6 keV from ²³⁹Np, te the wait time to counting, ti the time irradiation and tc the time of counting.

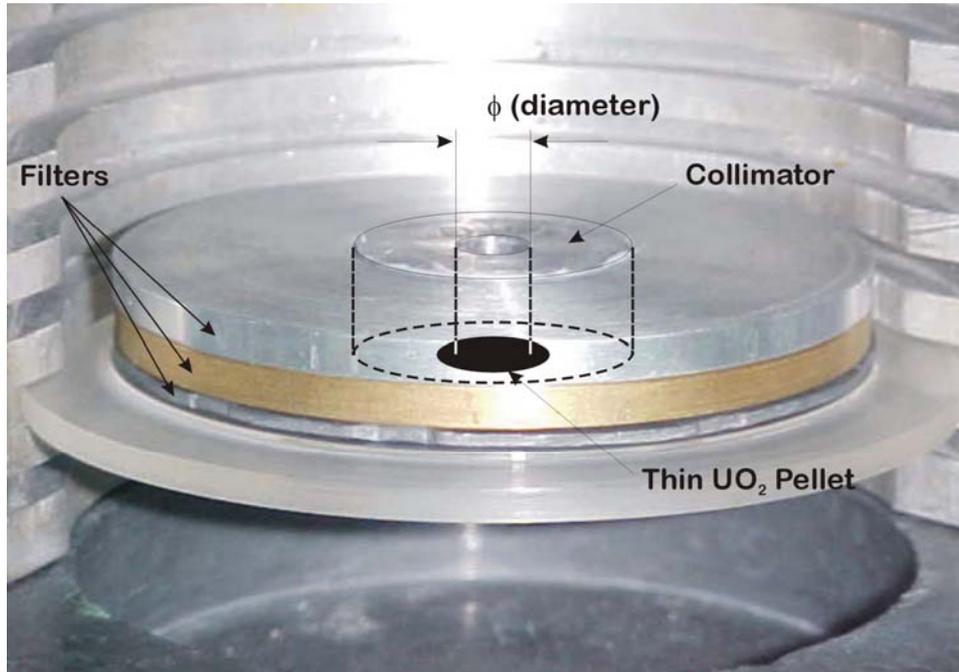


Figure 1 – Collimator used to measure gamma fotopeak of the ^{239}Np (277.6 keV) formed by radioactive capture nuclear reaction in the thin pellet irradiated in the fuel rod.

3. EXPERIMENTAL RESULTS

The Relative Radioactive Capture reaction along of the radius of the Thin UO_2 pellet irradiated has been measured by its gamma spectrometry using several collimators with different diameters (see Figure 1 and Table 1). Then the problem was to estimate de counting rate in the finished of irradiation (C_0) discounting the dead time. This parameter was estimate using the expression (2), where t_c is the Life time counting (without dead time) in the gamma spectrometry.

$$C_0 = \frac{{}^{Np} \lambda \cdot C \cdot \exp({}^{Np} \lambda \cdot t_e)}{[1 - \exp(-{}^{Np} \lambda \cdot t_c)]} \quad (2)$$

The Figure 2 show an example to bare thin UO_2 pellet irradiated and the gamma spectrometry made by 65 acquisition data using a 6 mm diameter collimator.

The final results can be sawn in the Table 1 where is showed too the relative radioactive capture nuclear reaction rate normalized by total counts to maximum diameter collimator (8,5 mm). Thus in this situation all diameter foil is sampled and the counts (277.6 Kev to ^{239}Np) are proportional to nuclear reaction rate.

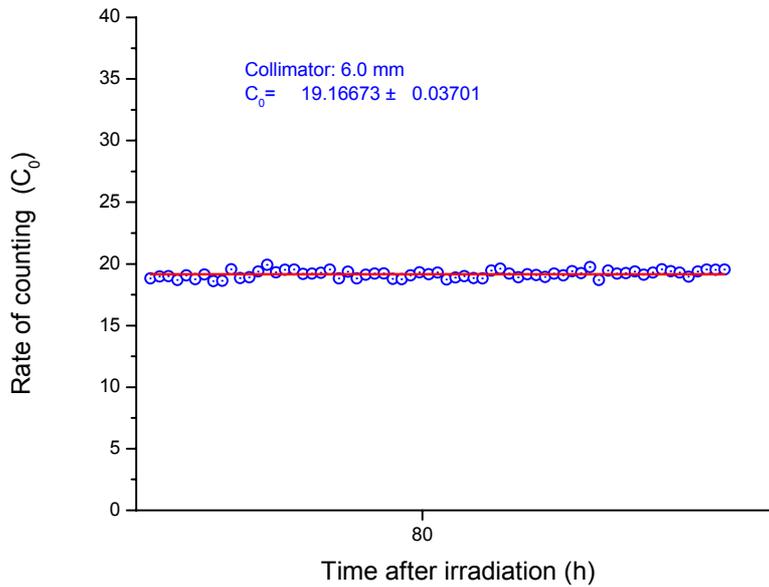


Figure 2. Determination of C_0 to bare Thin UO₂ pellet using a 6 mm diameter collimator.

Table 1. Relative Nuclear Reaction Rate of radioactive Capture to Thin UO₂ Pellet 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 Thin Pellet Disk (mm)	Nominal Collimator Diameter (mm)	Relative Radioactive Capture Nuclear Reaction Rate – Equation (2) (C_0) *	Normalized Values of C_0	Absolute Radioactive Capture Nuclear Reaction Rate – Equation (1) (C8) **
0.1498 ± 0.0019	3.0	1.5025 ± 0.0836	$0.0354 \pm 0,0019$	$(5.27 \pm 0.29). 10^4$
0.1959 ± 0.0015	4.0	4.3874 ± 0.1033	$0.1034 \pm 0,0024$	$(1.54 \pm 0.36) 10^5$
0.2235 ± 0.0017	4.5	5.7679 ± 0.1122	$0.1359 \pm 0,0026$	$(2.02 \pm 0.40).10^5$
0.2610 ± 0.0018	5.0	12.4472 ± 0.1778	$0.2932 \pm 0,0042$	$(4.36 \pm 0.63).10^5$
0.2735 ± 0.0018	5.5	13.9493 ± 0.2284	$0.3286 \pm 0,0054$	$(4.92 \pm 0.81).10^5$
0.2980 ± 0.0023	6.0	19.1553 ± 0.2830	$0.4513 \pm 0,0067$	$(6.72 \pm 1.00).10^5$
0.3233 ± 0.0017	6.5	23.4970 ± 0.3103	$0.5536 \pm 0,0073$	$(8.24 \pm 1.09).10^5$
0.3468 ± 0.0034	7.0	27.2740 ± 0.3579	$0.6425 \pm 0,0084$	$(9.61 \pm 1.26) .10^5$
0.3780 ± 0.0011	7.5	32.4800 ± 0.9110	$0.7652 \pm 0,0092$	$(1.15 \pm 0.14).10^6$
0.4038 ± 0.0021	8.0	38.4771 ± 0.4994	$0.9065 \pm 0,0117$	$(1.36 \pm 0.18).10^6$
0.4275 ± 0.0019	8.5	42.4474 ± 0.8332	$1.0000 \pm 0,0196$	$(1.50 \pm 0.29).10^6$

* ²³⁹Np (277,6 KeV); ** 100 watts power level – active fuel quote of 94 mm at Central fuel rod.

The same way has been irradiated a thin UO₂ Pellet in the same experimental conditions and axial quote (94 mm), but with Cadmium Covered. The Cadmium Covered has been used around the dismantable fuel rod and centered at quote 94 mm. The Cadmium Glove used had 20 cm of the length and 0,5 mm of thickness. The results obtained after gamma spectrometry can be seen in the Table 2.

Table 2 – Relative Nuclear Reaction Rate of radioactive Capture to Thin UO₂ Pellet 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 Thin Pellet Disk (mm)	Nominal Collimator Diameter (mm)	Relative Radioactive Capture Nuclear Reaction Rate – Equation (2) (C ₀) *	Normalized Values of C ₀	Absolute Radioactive Capture Nuclear Reaction Rate – Equation (1) (C8) **
0.1498 ± 0.0019	3.0	1.0727 ± 0.0797	0.0354 ± 0,0019	(3.72 ± 0.44) x 10 ⁴
0.1959 ± 0.0015	4.0	3.0987 ± 0.01669	0.1034 ± 0,0024	(1.08 ± 0,09) x 10 ⁵
0.2235 ± 0.0017	4.5	4.6137 ± 0.1113	0.1359 ± 0,0026	(1.60 ± 0,15) x 10 ⁵
0.2610 ± 0.0018	5.0	8.6685 ± 0.1871	0.2932 ± 0,0042	(3.01 ± 0,28) x 10 ⁵
0.2735 ± 0.0018	5.5	9.7806 ± 0.1735	0.3286 ± 0,0054	(3.39 ± 0,32) x 10 ⁵
0.2980 ± 0.0023	6.0	13.5529 ± 0.2654	0.4513 ± 0,0067	(4.70 ± 0,44) x 10 ⁵
0.3233 ± 0.0017	6.5	15.4352 ± 0.2505	0.5536 ± 0,0073	(5.35 ± 0,50) x 10 ⁵
0.3468 ± 0.0034	7.0	19.5322 ± 0.3704	0.6425 ± 0,0084	(6.78 ± 0,64) x 10 ⁵
0.3780 ± 0.0011	7.5	23.8980 ± 0.3632	0.7652 ± 0,0092	(8.29 ± 0,78) x 10 ⁵
0.4038 ± 0.0021	8.0	28.6718 ± 0.5216	0.9065 ± 0,0117	(9.95 ± 0,94) x 10 ⁵
0.4275 ± 0.0025	8.5	30.0378 ± 0.6913	1.0000 ± 0.0325	(1,04 ± 0,10) x 10 ⁶

* ²³⁹Np (277,6 KeV); ** 100 watts power level – active fuel quote of 94 mm at Central fuel rod.

The Figure 3 shows the percentage of Nuclear Reaction Rate of Radioactive Capture along of the Diameter of the UO₂ thin pellet irradiated at Central Position of the Core exactly in the axial active fuel quote 94 mm). The same way the Figure 4 show the experimental results obtained to UO₂ Thin Pellet irradiated with the Lead Glove of 20 cm length and 0,5 thickness. To both cases it was used to fit the experimental data the Boltzmann function with good results.

This function is the same used to estimate the calibration curve of the control rods of the IPEN/MB-01 reactor and has itself showed good results when the Physical phenomenology involve neutronic absorption.

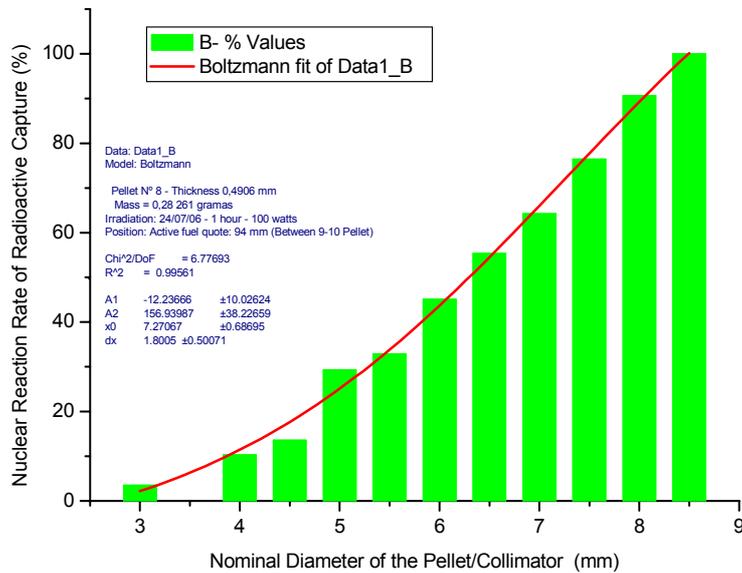


Figure 3. Percentage of Nuclear Reaction Rate of Radioactive Capture along of the Diameter of the UO₂ Pellet Irradiated at Central Position of the Core (quote 94 mm).

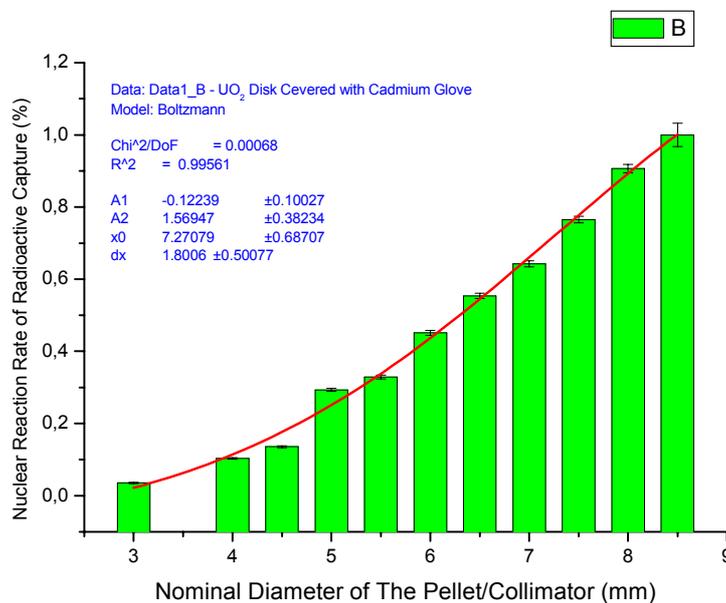


Figure 4 - Percentage of Nuclear Reaction Rate of Radioactive Capture along of the Diameter of the UO₂ Pellet Irradiated Covered with Cadmium Glove at Central Position of the Core (quote 94 mm).

4. CALCULATION BY MCNP-4 C CODE

The calculation methodology aimed to reproduce the experiment. Thus it was used for this purpose the MCNP-4C code [4] (Monte Carlo method) using the ENDF-BVI.8 nuclear data library and the results can be seen at Table 3 given below.

Table 3. Calculated values [7] of Relative Nuclear Reaction Rate of Radioactive Capture to bare Thin UO₂ Pellet irradiated in IPEN/MB-01 Reactor Core obtained by MCNP-4C Code.

Radius of the Fuel Pellet (cm)	Relative Nuclear Reaction Rate of Radioactive Capture	Normalize Values *
0.134239	4.69622×10^{-3}	0.0808
0.189842	4.78294×10^{-3}	0.1631
0.232508	4.90005×10^{-3}	0.2474
0.268477	4.99034×10^{-3}	0.3332
0.300167	5.10993×10^{-3}	0.4211
0.328816	5.31006×10^{-3}	0.5124
0.355162	5.52099×10^{-3}	0.6074
0.379684	5.85956×10^{-3}	0.7082
0.402716	6.53097×10^{-3}	0.8205
0.424500	1.04300×10^{-2} *	1.0000

* Normalize to Total Nuclear Reaction rate of the Fuel Pellet of larger radius;

** Experimental Values obtained from Boltzmann fit (Fig.1) to the same radius Pellet showed at the Table 3.

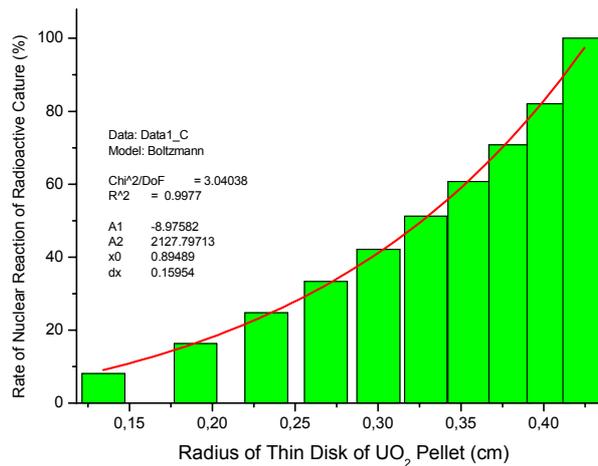


Figure 5. Percentage of Nuclear Reaction Rate of Radioactive Capture along of the Diameter of the UO₂ Pellet Irradiated at Central Position of the Core (quote 94 mm

The same way the Table 4 show the calculated values of the nuclear reaction rate of radioactive capture to thin disk of UO₂ pellet irradiated with cadmium covered (Cadmium glove around the fuel rod centered at quote 94 mm). These lead glove used in this calculation has 5.0 cm of length and 0.5 mm of thickness.

Table 4. Calculated values of Relative Nuclear Reaction Rate of Radioactive Capture to bare Thin UO₂ Pellet irradiated in IPEN/MB-01 Reactor Core obtained by MCNP-4C Code [7].

Radius of the Fuel Pellet (cm)	Nuclear Reaction Rate of Radioactive Capture	Normalize Values *
0.134239	2.92723E-3	0.0760
0.189842	2.96066E-3	0.0769
0.232508	3.02122E-3	0.0784
0.268477	3.10577E-3	0,0806
0.300167	3.22422E-3	0,0837
0.328816	3.34879E-3	0.0869
0.355162	3.54742E-3	0.0921
0.379684	3.80316E-3	0.9872
0.402716	4.43155E-3	0.1150
0.424500	8.15385E-3	0.2117

* Normalize to Total Nuclear Reaction rate of the Fuel Pellet of larger radius;

** Experimental Values obtained from Boltzmann fit (Fig.1) to the same radius Pellet showed at the Table 4.

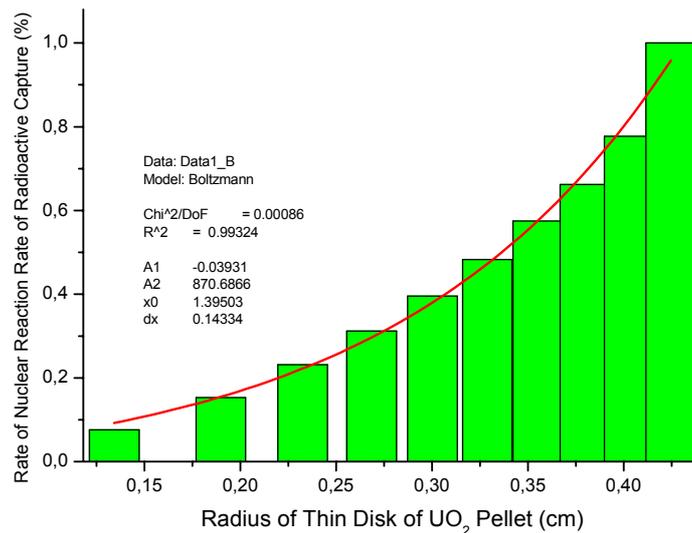


Figure 6 - Percentage of Nuclear Reaction Rate of Radioactive Capture along of the Radius of the UO₂ Pellet Irradiated with Cadmium Covered Glove at Central Position of the Core (axial quote 94 mm).

5. CONCLUSIONS

This work aims to show the actual stage of the measurements of nuclear reaction rates of radioactive capture along of the radius of fuel UO_2 pellet of nuclear fuel of the IPEN/MB-01 reactor. This nuclear reaction rate is very difficult to obtain by experimental way and the final results will be very important to correlate with calculation methodology. The future step of this work will be to make more acquisition of the experimental data. The goals is to make 3 irradiations in the minimum for each kind of thin disk of UO_2 pellet and verify the average behavior and the reproducibility of the measurements to compare with calculation results and estimate its precision level using different nuclear data libraries.

The preliminary results show a similar spatial distribution of the nuclear reaction rate along the radius of the thin UO_2 fuel pellet disk between calculated and measured values. When is fitted a Boltzmann function to both cases the results are good and show the same spatial tendency of spatial distribution of nuclear reaction rate along of the radius of the fuel pellet.

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