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Neutrons doses determination inside the thermal column of the TRIGA Mark III Nuclear Reactor

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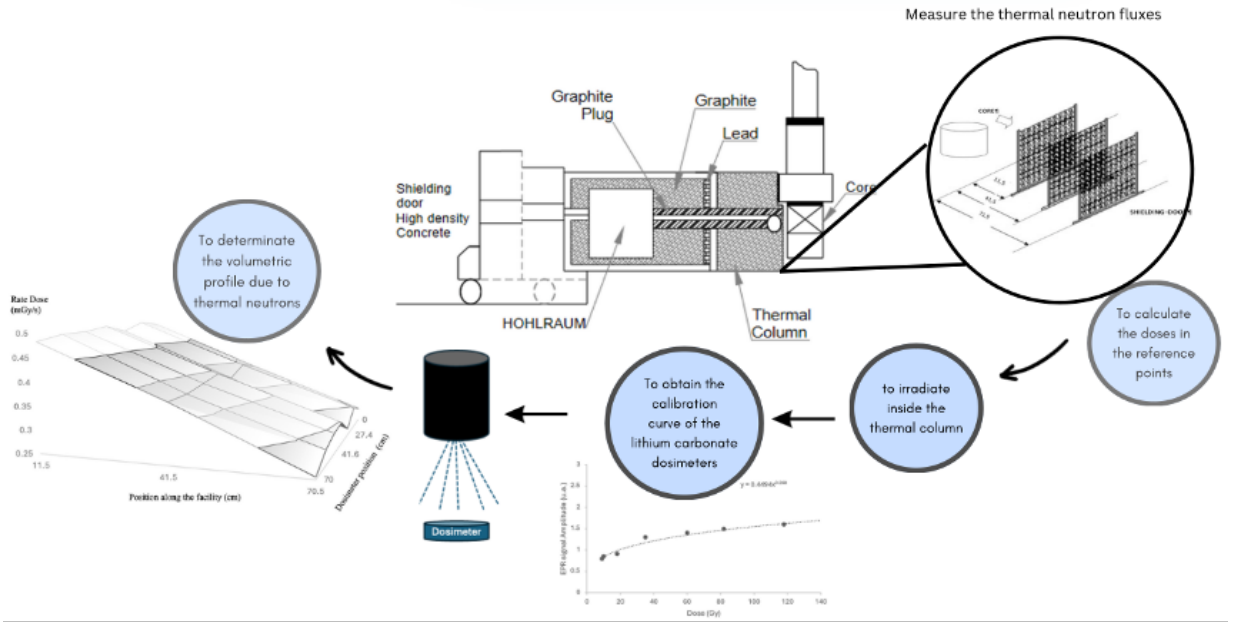
Graphical abstract



Abstract

Determination of the volumetric dose profile for the irradiation installation in the thermal column of the nuclear reactor is a current problem that requires attention in nuclear engineering. This paper presents a proposal that considers the use of Lithium Carbonate pellets as dosimeters and using the Electronic Paramagnetic Resonance spectroscopy technique to measure the absorbed dose within the thermal column. With the above, neutron dosimetry was carried out in the thermal column of the TRIGA Mark III Reactor, for which it was necessary to establish the appropriate calibration curve. The results obtained show the relevance of the strategy in determining the volumetric dose profile proposed in this paper.

Keywords: *Lithium Carbonate Dosimeter; TRIGA Mark III; Thermal Column; Paramagnetic Resonance.*

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Determinación de dosis de neutrones dentro de la columna térmica del Reactor Nuclear TRIGA Mark III

Resumen
La determinación del perfil de dosis volumétrica para la instalación de irradiación en la columna térmica del reactor nuclear es un problema al que hoy en día se presta atención en la ingeniería nuclear. Este artículo presenta una propuesta que considera el uso de pellets de Carbonato de Litio como dosímetros y el uso de la técnica de espectroscopía de Resonancia Paramagnética Electrónica para medir la dosis absorbida dentro de la columna térmica. Con lo anterior se realizó dosimetría de neutrones en la columna térmica del Reactor TRIGA Mark III, para lo cual fue necesario establecer la curva de calibración adecuada. Los resultados obtenidos muestran la relevancia de la estrategia en la determinación del perfil de dosis volumétrica propuesta en este trabajo.

Palabras clave: *Dosímetro de Carbonato de Litio; TRIGA Mark III; Columna Térmica; Resonancia Paramagnética.*

Determinação de doses de nêutrons na coluna térmica do Reator Nuclear TRIGA Mark III

Resumo
A determinação do perfil de dose volumétrica para instalação de irradiação na coluna térmica do reator nuclear é um problema que atualmente vem recebendo atenção na engenharia nuclear. Este artigo apresenta uma proposta que considera o uso de pastilhas de Carbonato de Lítio como dosímetros e o uso da técnica de espectroscopia de Ressonância Paramagnética Eletrônica para medir a dose absorvida dentro da coluna térmica. Com o exposto, foi realizada a dosimetria de nêutrons na coluna térmica do Reator TRIGA Mark III, para a qual foi necessário estabelecer a curva de calibração adequada. Os resultados obtidos mostram a relevância da estratégia na determinação do perfil de dose volumétrica proposto neste trabalho.

Palavras-chave: *Dosímetro de Carbonato de Lítio; TRIGA Mark III; Coluna Térmica; Ressonância Paramagnética.*

Introduction

The use of lithium carbonate (Li_2CO_3) as a material for thermal neutron dosimetry has been already established [1]. The performance of the Li_2CO_3 dosimeters is based on quantitative yield evaluation of free radicals produced by the irradiation of this material with thermal neutrons. In addition to the alpha-particles and tritium recoil ions, the irradiation with neutrons produces carboxy radical ions CO_3^- and CO_2^- , and the proposed method in this paper is based on quantitation of the stable radical ions CO_2^- by the spectrometric technique Electronic Paramagnetic Resonance (EPR), which provides a signa that is proportional to the received dose. It is important to point out some of the useful properties of the Li_2CO_3 by means of the EPR spectrometry [2]:

1. A broad dose range from 0 to 225 Gy that does not generate undesirable signals (background).
2. The manufacture of Li_2CO_3 dosimeters is easier than other types of dosimeters [3].

In this study commercial Li_2CO_3 for manufacturing the pellets was used. The method is based on the nuclear reaction that occurs when the ^6Li absorbs thermal neutrons to produce α particles and tritium nuclei, as show in the reaction of Equation 1 [4]. The cross section for this reaction is 940 b, and the energies of the reaction products have been estimated in $2.73\ MeV$ (tritium) and $2.05 MeV$ (α).

$$^6Li + n_{th} \rightarrow ^3H + ^4He + 4.78\ MeV \qquad (1)$$

On the other hand, the EPR or electronic spin resonance (ESR) is a sensitive spectroscopic technique to quantify unpaired ions. Generally, it involves free radicals of organic molecules or ions of transition metals for inorganic compounds [1]. Since the most stable molecules have a closed-shell configuration, with all spins paired, it allows knowing the energy levels of each of the molecular orbitals under the influence of the ionic interactions in the compound.
The energy of interaction with the magnetic field of the electrons is greater that of the nuclei, so that, the weaker external magnetic fields the higher electromagnetic frequencies are required. In a magnetic field of $0.3\ T$, the EPR occurs around 10 GHz.

It is advisable to give some characteristics about the irradiation facility used to perform the dosimeter irradiation. The Thermal Column (CT) has an empty cavity called *hohlraum*, where it is possible irradiate massive samples. This facility has the peculiarity of allowing irradiation with almost only thermal neutrons. Due to the thermal column is an aluminum cube built in the concrete shielding structure, lined inside with graphite blocks, as it is shown in Figure 1. The purpose of this graphite covering is to moderate the fast neutrons tol thermal energies, also it has a lead wall to minimize the gamma radiation field inside the CT. This allos to neglect the gamma and fast neutron components.

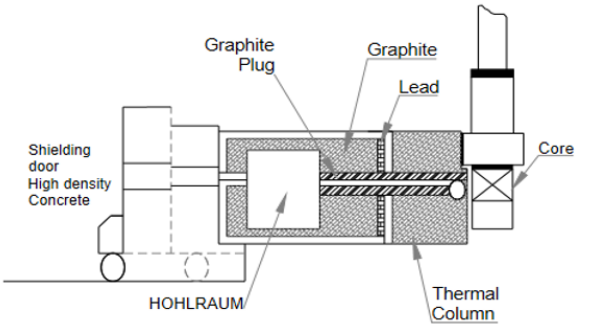

Figure 1. Scheme of the reactor irradiation facility, Thermal Column (CT).

The technique used to determine the thermal flux was the foil activation method, using pairs of foils made with high purity elements such as Au, In, Cu, etc. It is known that Cadmium is an effective neutron absorber with energies less than 0.4 eV.
In order to measure the thermal flux in a specific point it is necessary to irradiate one bared foil and other covered with cadmium, because the bared foil will be irradiated with neutrons of all energies of the neutron spectrum and the covered foil only with neutrons above the cadmium cutoff energy (> 0.4 eV).
In this way, the activity due to the thermal neutrons will be equal to the activity induced in the bare foil (neutrons of all energies) minus the activity of the covered foil.
The activation equation, that relates the subcadmium flux with the measured activation in both foils [5], is given by:

$$\phi_{cd} = \frac{Pa}{kN_A\sigma_c(1-e^{-\lambda t_i})}\left[\frac{A_d(t_d)e^{\lambda t_d}}{m_d} - \frac{A_{cd}(t_d)e^{\lambda t_d}}{m_{cd}}\right] \qquad (2)$$

The fluence Φ, is the number of particles dN incident on a surface with an area *da*, and its units are $cm^{-2}s^{-1}$, thus:

$$\phi = \frac{dN}{da} \tag{3}$$

The term flux has been employed for the quantity termed fluence rate. The fluence rate, Φ, is the increment of the fluence $d\Phi$ in the time interval dt, and its units are cm^{-2}-s^{-1} thus;

$$\phi = \frac{d\phi}{dt} \tag{4}$$

According that, fluence is determined multiplying the neutron flux by the irradiation time (in seconds) of each irradiated dosimeter and it is given in cm^{-2}. In order to calculate the dose H, the fluence is multiplied by the effective dose conversion coefficient for whole body exposure, $hE = 4E\text{-}12Sv\,\mathrm{cm}^2$ [6]. But the absorbed doses D in *Gy*, is required, then the quality factor *Q* must be considered to convert dose in Sv to *Gy*. Factor Q for thermal neutrons with average energy of 0.025 eV, [7] is given by Equation 5 for $E_n < 1\ MeV$:

$$Q = 2.5 + 18.2\ exp - \left[\frac{(InE)^2}{6}\right] \tag{5}$$

Then, for $E = 0.025\ eV \Rightarrow Q = 4.384$, as $H = DQ$, then $D = H/4.384$ in *Gy*. (6)

Experimental procedure

The procedure starts measuring the thermal neutron fluxes in some reference points of the thermal column. With these measurements it is possible to calculate the doses in those reference points.

After the flux measurements and their corresponding doses, it will be proceeded to irradiate inside the thermal column at the same distances, where the neutron fluxes and the lithium carbonate dosimeters were previously measured for different irradiation times, with the purpose to obtain the calibration curve of the lithium carbonate dosimeters.

Finally, the lithium carbonate dosimeter was irradiated in different points of the thermal column, in order to determinate the volumetric profile due to thermal neutrons.

Equipment used

For the experimentation carried out, the following equipment was used:

1. Reactor TRIGA Mark III with a maximum power of 1 MWth.
2. Spectrometer, Varian Brand, Model *E-15*.
3. Scales, OHAUS Brand, Model Mark Explorer.
4. Gamma spectrometry system.

Determination of the neutron flux in the Hohlraum

In general terms, the irradiation of the activation foils was made in the following way:

1. **Gold foils preparation.** First, the gold foils were cleaned with alcohol, and then rinsed with demineralized water. Then, the foils were weighe with the digital analytical scale.
2. **Neutron flux measurement.** Figure 2 shows how the neutron flux is measured at points chosen at different distances from the bottom of the thermal column.

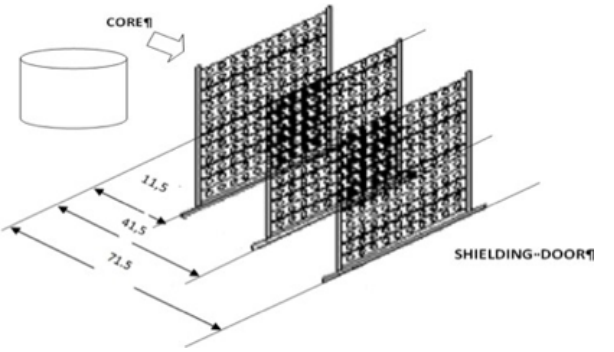

Figure 2. The three different positions, where the support of the dosimeters were located.

One pair of gold foils wasfixe on the surface of the support one of them covered with two cadmium plates and the other without the cadmium plates.

Later the support was placed inside the thermal column at one of the distances previously elected to irradiated the pair of foils at 1 MWh during 20 minutes. This was repeated for each of the three different positions as indicated in Figure 2, in order to measure the neutron fluxes at each position.

After the irradiation, the foils were removed to measure their activities, using the Gamma Spectrometry System. With the measured activities for each foil and the parameters included in the Equation 2, Using the computational program CAFLU [8] the fluxes (subcadmiun and epicadmiun) can be calculated (specially developed to solve Equation 2, in several points).

Measurement of doses

Dosimeters of Li_2CO_3 were chosen to measure the absorbed dose inside the irradiation facility *hohlraum* as used previously in reference [1].

3. **Sample preparation.** Dosimeters were prepared by blending the Li_2CO_3 salt with silicone (RTV gel, Mar. Dow Corning), to have samples of Li_2CO_3 with better consistency. Then the homogeneous mixture was introduced into open-ended cylindrical molds with a plastic syringe. The batch was ready for the extraction from the molds after 24 h. The batch consisted of 105 samples. The samples were cylindrical pellets (10-mm long, 3.9 mm in diameter), each one contained 80 mg of Li_2CO_3.
4. **Sample irradiation.** The main stage of this work was to irradiate several dosimeters of Li_2CO_3 in different points inside the thermal column, previously determined, all of this was carried out to obtain the dose profile in the thermal column [9]. In this way the Li_2CO_3 pellets were irradiated in a similar way as the irradiation of the gold foils. The irradiation was carried out as follows:

- A set of 105 samples was divided into 21 groups, each containing five pellets.
- The radiation times were set and are shown in Table1.

Table 1. Irradiation times for the Li_2CO_3 dosimeters.

Group	Irradiation time (h)
A	0.0833, 0.166, 0.25, 0.33, and 0.5
B	10, 20.0, 40.0, 60.0, 80.0, and 100.0
C	1.0, 2.0, 3.0, 4.0, and 5.0
D	120.0, 140.0, 160.0, 180.0, and 200.0

- All groups of samples were irradiated, to determinate the time dependent response for this kind of dosimeters [9].
- To measure the dose space distribution, the support was moved at three different positions (Figure 2) along the *hohlraum* axis.
- The irradiations were of 20 minutes at 1 MWh power for each position.
- The EPR measurements for each set of five dosimeters were averaged.

5. **Determination of free radical concentrations through EPR technics.** The dose measurements were performed by assessing the change in the free radical concentration in the Li_2CO_3 dosimeters [10]. For this purpose, the EPR spectrometer was used, operating in the *X* ray band. The spectrometer was equipped with a dual- resonance cavity operating in the TE104 mode, which enabled us to compare the amplitude of the signal of the sample (*S*), of an irradiated pellet positioned in the one half of the cavity, with the amplitude of the signal of the standard (*P*), composed of a synthetic ruby. In this way, the relative signal *P*/*S* of a pellet can be measured with a high accuracy.

   The relative concentration of free radicals was evaluated by comparing the peak-to-peak height ratio *D*/*S* [6]. The EPR operational settings are given in Table 2.

Table 2. The EPR operational settings.

Operating conditions for the EPR equipment.	
Central magnetic field	330 mT
Scan width	40 mT
Scan time	8 min
Magnetic field amplitude modulation	0.1 mT
Modulation frequency	100 kHz
Microwave power	2.0 mW (nominally 1.0 mW at half of the cavity)
Time constant	3.0 s

Results

Neutron fluxes

Table 3 shows the masses of the gold foils, irradiation and decay times, and measured counts, in order to calculate with the program CAFLU, the subcadmium and epicadmium neutron fluxes.

The nuclear parameters for each kind of material used as detection foils are given as data in the program CAFLU, then with the values contained in the first five columns of Table 3, and the total efficiency of the counting system ε = 0.014, the subcadmium and epicadmium fluxes were calculated.

Table 3. Subcadmium and epicadmium fluxes at three different positions in the central axis of the thermal column.

Position (cm)	Weight (g)	Time		Counts	S Subcadmium F flux (n-cm^{-2}s^{-1})	Epicadmium flux (n-cm^{-2}s^{-1})	Cadmium rate
		T_c (s)	T_d (h)				
11.5	0.0037 0.004	200 600	0.85 0.6	4572±68 162±44	5.51 E 10^{8*}	5. 63 E 10^{6*}	95*
41.5	0.0038 0.0073	200 600	0.68 0.46	3509±59 221±53	4.36 E 10^{8*}	4.58 E 10^{6*}	90*
71.5	0.0074 0.0044	180 600	2.11 2.2	9727±100 199±17	3.3 E 10^{8*}	3.44 E10^{6*}	91*

*There is no data for fluxes in 600 seconds, due to inaccurate statistics.

Outcomes for the dosimeter irradiations

To see the behavior of dosimeters with longer irradiation times, the dosimeters were irradiated for longer times. The readings obtained for the groups of dosimeters B, C and D are shown in Table 4.

Table 4. Subcadmium and epicadmium fluxes at three different positions in the central axis of the thermal column.

Signal EPR (u.a.)	Time (*h*)	Signal EPR (u.a.)	Time (*h*)
1.4908	1	2.5622	60
1.5891	2	2.7786	80
1.7051	3	2.7987	100
1.7874	4	2.8046	120
1.8512	5	2.8134	140
2.0456	10	2.8277	160
2.2478	20	2.8302	180
2.4461	40	2.8235	200

The data given in the previous table were plotted for the irradiation times from 1 to 200 hours vs the EPR signal values. Figure 3 shows that the dosimeters tend to saturation after 80 hours of irradiation.

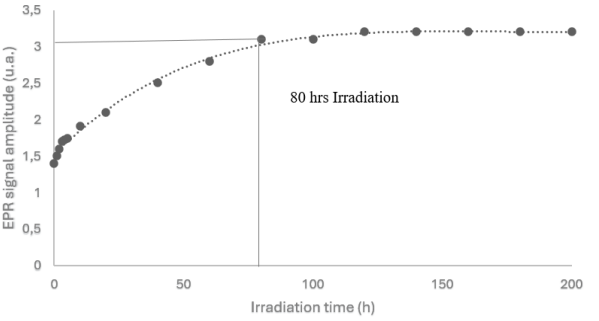

Figure 3. Graphic of the EPR signal amplitude as function of the irradiation time.

Dose determination

Table 5 shows the fluxes for the different irradiation positions in the *holhraum*. With these fluxes and the irradiation times for the irradiated dosimeters at different times, the total fluence for each group of dosimeters were calculate. Also, in the table their corresponding EPR signals are included and doses were calculated according Equation 5 and Equation 6.

By plotting the data given in Table 5, it is possible to get the calibration curve for this kind of dosimeters, which is shown in Figure 4.

Table 5. Data of fluence and their corresponding EPR signals.

	Fluence (cm^{-2})	EPR	Dose	Time	Fluence (cm^{-2})	EPR	Dose
(s)		(u.a.)	(Gy)	(s)		(u.a.)	(Gy)
300	1.31E+11	0.8422	0.1182	14400	6.28E+12	1.7874	5.6757
600	2.62E+11	0.8742	0.2365	18000	7.85E+12	1.8512	7.0946
900	3.92E+11	0.9062	0.3547	36000	1.57E+13	2.0456	14.1892
1200	5.23E+11	0.9382	0.3941	72000	3.14E+13	2.2478	28.3784
1800	7.85E+11	1.017	0.7095	144000	6.28E+13	2.4461	56.7567
3600	1.57E+12	1.4908	1.4189	216000	9.42E+13	2.5622	85.1351
7200	3.14E+12	1.5891	2.8378	288000	1.26E+14	2.7122	113.5135
10800	4.71E+12	1.7051	4.2568	360000	1.57E+14	2.7987	141.8918

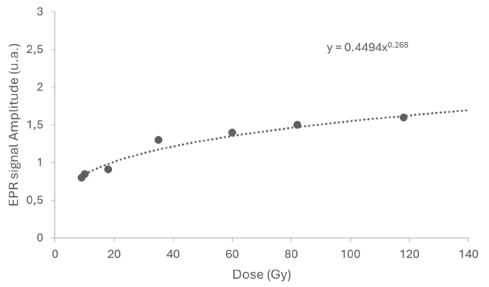

Figure 4. Calibration curve for the neutron thermal dosimeters of Li_2CO_3.

The graph above reports the amplitude of the EPR signal with dose. By adjusting with the potential H =4.49*10^{-1} EPR 2.68*10^{-1}, the corresponding dose value can be interpolated for any measured EPR value.

Dose profile measurements

Analyzing the calibration curve shown in the Figure 4, it was decided to irradiate the samples during 20 minutes to 1 MW, since it is considered that the response of the dosimeters is almost lineal in the range from 10 to 60 min, besides it is more convenient and faster to get the readings for all irradiated dosimeters. Doses measured with the dosimeters are included in Table 6.

With the data listed in Table 6, the graph shown in Figure 5 was obtained, where the three graphs correspond to the three different positions where the support with the dosimeters was located inside the thermal column.

As expected, Figure 5 shows that the highest rate dose is obtained near the back of the hohlraum and decreases as the distance to the back increases. With the purpose of getting a better idea of how the doses vary with the position in the thermal, a three-dimensional plane was plotted, showing a clearer image of the dose variation inside the irradiation facility, the thermal column (see Figure 6).

Table 6. Lateral dose distribution inside the thermal column to different distances 11.5, 41.5 and 71.5 cm.

Position (cm)	EPR (u.a.)	Dose rate (mGy/s) at 11.5 cm	EPR (u.a.)	Dose rate (mGy/s) at 41.5 cm	EPR (u.a.)	Dose rate (mGy/s) at 71.5 cm
0	0.9208	0.4891	0.8279	0.3812	0.4894	0.2798
7	0.9375	0.4980	0.8549	0.3937	0.5204	0.2975
21	0.9066	0.4815	0.8076	0.3719	0.4631	0.2648
35	0.9382*	0.4983	0.856	0.3942	0.5218	0.2983
49	0.9251	0.4914	0.8355	0.3847	0.5153	0.2946
63	0.9251	0.4914	0.8166	0.3760	0.4934	0.2821
70	0.909	0.4828	0.8152	0.3754	0.4457	0.2548

*The EPR signal is the same, the changes are the dose and the dose ratio.

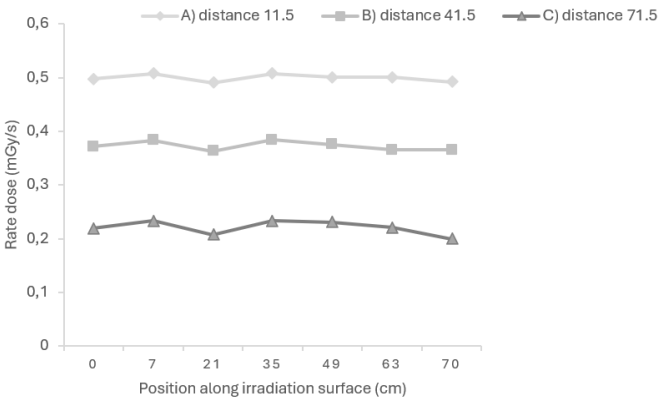

Figure 5. Profile of the rate doses inside the thermal column in three different distances.

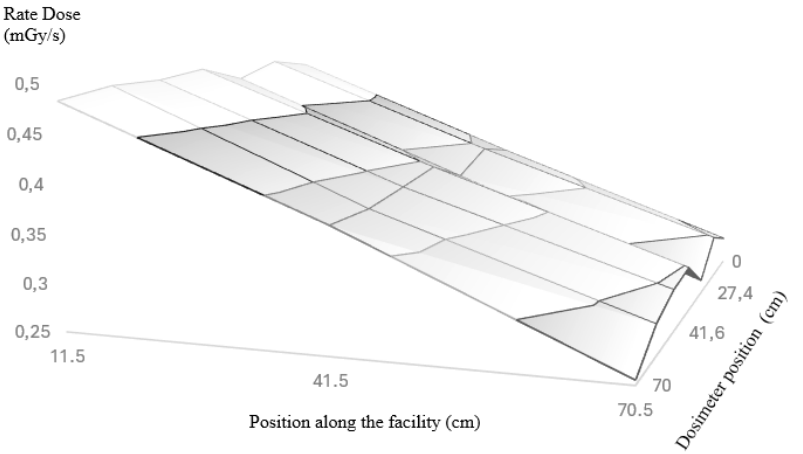

Figure 6. Dose variation along of the irradiation facility. Coordinates (x, y, z)

Summary and conclusions

First, the distribution of doses inside the thermal column was as expected higher in the back of the thermal column and decreases as the distance increases, something important is to note that the transversal doses for a given distance, along the facility, remain almost constant.

From the values of cadmium rate, given in Table 3, it can be seen that the cadmium rate in each point was around 90, this means there are almost one hundred subcadmium (thermal) neutrons for each epicadmium neutron, and these kinds of neutrons have resonance energies, only above the cadmium cutoff energy, in the range of *eV*, whereby the contribution of fast neutrons was negligible.

Finally, the dosimeters used resulted adequate to measure the doses induced by the neutron fluxes existing in the facility used. This kind of dosimeters would perform better with a little bit higher fluxes, because the irradiation times would be shorter.

For future measurements it would be recommendable to use an irradiation facility with a higher neutron flux, at least an order of magnitude more, in order to check up on if the mentioned dosimeters work adequately at higher rate of doses.

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