

Low energy neutrons from a $^{239}\text{PuBe}$ isotopic neutron source inserted in moderating media

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Several neutron applications share a common problem: the neutron source design. In this work MCNP computer code has been used to design a moderated $^{239}\text{PuBe}$ neutron source to produce low energy neutrons. The design involves the source located at the center of a spherical moderator. Moderator media studied were light water, heavy water and a heterogeneous combination of light water and heavy water. Similar moderating features were found between the 24.5 cm-radius container filled with heavy water (23.0-cm-thick) and that made with light water (3.5-cm-thick) plus heavy water (19.5-cm-thick). A $^{239}\text{PuBe}$ neutron source inserted in this moderator produces, at 27 cm, a neutron fluence of 1.8×10^{-4} n-cm⁻² per source neutron, with an average neutron energy of 0.34 MeV, where 47.8 % have an energy ≤ 0.4 eV. A further study of this moderator was carried out using a reflector medium made of graphite. Thus, 15-cm-thickness reflector improves the neutron field producing a neutron fluence of 1.1×10^{-3} n-cm⁻² per source neutron, whose average neutron energy is 0.098 MeV, where 75.8 % have an energy ≤ 0.4 eV.

Keywords: Isotopic neutron source; moderation; Monte Carlo method.

Las aplicaciones de los neutrones comparten un mismo problema: el diseño de la fuente. En este trabajo el código MCNP se utilizó para diseñar una fuente de neutrones moderada, a base de una fuente isotópica de $^{239}\text{PuBe}$, con el propósito de producir neutrones de baja energía. En el diseño, la fuente se colocó en el centro de un moderador esférico. Los medios moderadores estudiados fueron agua ligera, agua pesada y una combinación heterogénea de agua ligera con agua pesada. Características similares en la moderación se observaron para el contenedor de 24.5 cm de radio lleno con agua pesada (23.0 cm de espesor) y con contenedor lleno heterogeneamente con agua ligera (3.5 cm de espesor) y con agua pesada (19.5 cm de espesor). Una fuente de $^{239}\text{PuBe}$ en este último moderador produce a 27 cm una fluencia de 1.8×10^{-4} n-cm⁻² por cada neutrón emitido por la fuente, con una energía promedio de 0.34 MeV, donde el 47.8% tiene una energía ≤ 0.4 eV. Un estudio posterior sobre el moderador heterogéneo se realizó para determinar el efecto de utilizar un medio reflector de grafito. Así un reflector de 15 cm de espesor mejora el campo de neutrones de baja energía ya que se obtienen 1.1×10^{-3} n-cm⁻² por cada neutrón emitido por la fuente, la energía promedio es de 0.098 MeV, donde el 75.8 % tiene una energía ≤ 0.4 eV.

Descriptores: $^{239}\text{PuBe}$; moderación; Monte Carlo; neutrones.

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1. Introduction

Neutron applications are defined by the energy spectrum of the source: fast neutrons are used in radiobiological research, and radiotherapy [1,2], variable energy neutrons are applied to calibrate health physics instruments [3,4], to study the spectral shifting produced by filters [5] and to analyze the spatial and energy distribution [5,6]. On the other hand, epithermal neutrons are used in Boron Neutron Capture Therapy, BNCT, to treat brain tumor cancer [7-10]. Thermal neutrons are used in Neutron Capture Synovectomy [11], Neutron Activation Analysis [12-18], to produce standard neutron fields [19], and to obtain nuclear resonance fluorescence [20].

Neutrons are produced in nuclear reactors, particle accelerators and by isotopic neutron sources. Isotopic neutron sources, like ^{252}Cf , $^{241}\text{AmBe}$, $^{239}\text{PuBe}$, etc, yield neutrons with specific intensity and energy distribution. These are known for their simplicity of installation, operation and low price compared with nuclear reactors or neutron generators [21]. Isotopic neutron sources have complex spec-

tra [22], if the application requires thermal neutrons a moderator is used to produce as many thermal neutrons as possible. Moderators like paraffin [12-15, 20], polyethylene [16, 18, 23], light water [4, 24], heavy water [9, 10, 17, 19], and other rich hydrogen content materials [25] have been used.

The Monte Carlo method provides a computer-based technique to simulate the interactions of particles in a medium. The improving of computers speed and reduction of their prices makes Monte Carlo methods a cost effective procedure to design radiation application devices.

During simulation bodies are modeled, in shape, size and elemental composition, as close as possible to actual situations. The quality of Monte Carlo calculations depend from the number of histories utilized, the quality of involved cross sections and from the resemblance between model and actual situation.

In this work a Monte Carlo study, using the MCNP code, version 4B [26], was carried out to investigate the performance of light water, H_2O , heavy water, D_2O , and a heterogeneous combination of H_2O and D_2O moderators in the aim

to produce low energy neutrons, $E \leq 0.4$ eV, from a $^{239}\text{PuBe}$ neutron source, to be used for Boron Neutron Capture Synovectomy basic research.

2. Methods

Monte Carlo study, using the MCNP code version 4B [26], was carried out to investigate the performance of 3 moderator media, H_2O , D_2O , and a heterogeneous combination of H_2O and D_2O moderators to produce a low energy neutron field, from a $^{239}\text{PuBe}$ neutron source.

The moderator was modeled as three concentric spherical shells, in the center of which a 1.0 cm-radius sphere was used to represent the neutron source. The first spherical shell, with a 1.5 cm-radius (0.5-cm-thick) was filled with air, the second spherical shell is 5.0 cm-radius (3.5-cm-thick). The third spherical shell was modeled with 6 radii: 6.0, 7.0, 11.5, 16.5, 21.5 and 24.5-cm. The second and third spherical shells were filled with two types of moderators, H_2O and D_2O . In a different case the second shell was filled with H_2O and the third shell with D_2O . The moderator with best performance was studied adding a graphite neutron reflector.

The neutron spectrum was tallied at 27 cm from source center using a 0.5 cm radius detector. Neutron tally was carried out using 25 energy bins. Tallies in the first energy bin contains those neutrons whose energy is less or equal to 4.140×10^{-7} MeV. Nevertheless MCNP code can be utilized to obtain point-like neutron spectra in this investigation was decided to do few channels neutron spectra calculations in order to reduce the computation time and because the eventual experimental benchmark will be carried out with a multisphere neutron spectrometer. Multisphere neutron spectrometer, also known as Bonner spheres, allows to measure neutron spectra with the same energy bin array as used during calculations [27-29]. Figure 1 shows the moderator model used. In this study the number of histories were large enough to assure a statistical uncertainty, in each energy bin, less or equal to 5 %.

The quality of Monte Carlo simulation depend from the quality of materials' cross sections. Neutron and gamma cross sections are permanently evaluated and compiled in cross section libraries. In this study neutron interaction cross sections were taken from ENDF/B-VI library, that contains the most updated cross sections available for MCNP calculations [30-32]. Chemical binding and crystalline effects of H_2O , D_2O and graphite during thermal neutron scattering were taken into account using $S(\alpha, \beta)$ treatment [33].

Nevertheless $^{239}\text{PuBe}$ sources have been substituted by other neutron sources, such as $^{241}\text{AmBe}$ and ^{252}Cf , still there are several installations that use them. $^{239}\text{PuBe}$ neutron source features have been extensively studied [34-39]. Its neutron spectrum, shown in Fig. 2, was used as the source term during Monte Carlo calculations [38, 40, 41].

According to Allen and Beynon [9] a good moderating material should have the following properties:

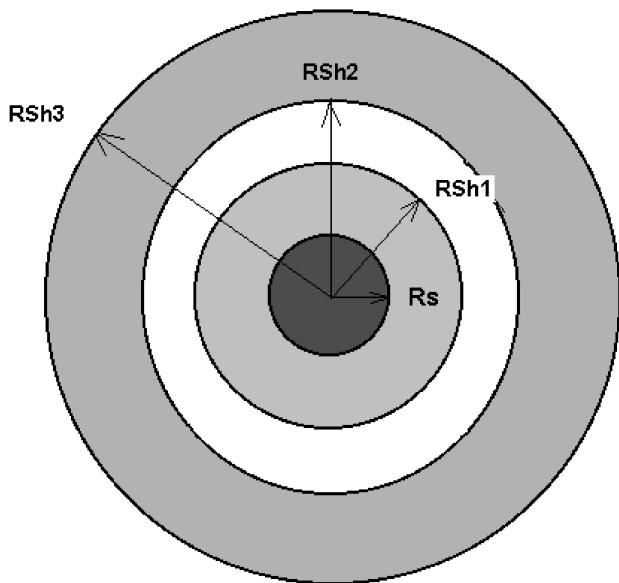


FIGURE 1. Moderator model. R_s is the source radius (1.0 cm), R_{Sh1} is the first shell radius (1.5 cm) filled with air, R_{Sh2} is the second shell radius (5.0 cm) filled with moderator and R_{Sh3} radius (6.0, 7.0, 11.5, 16.5, 21.5 and 24.5 cm) filled with moderator.

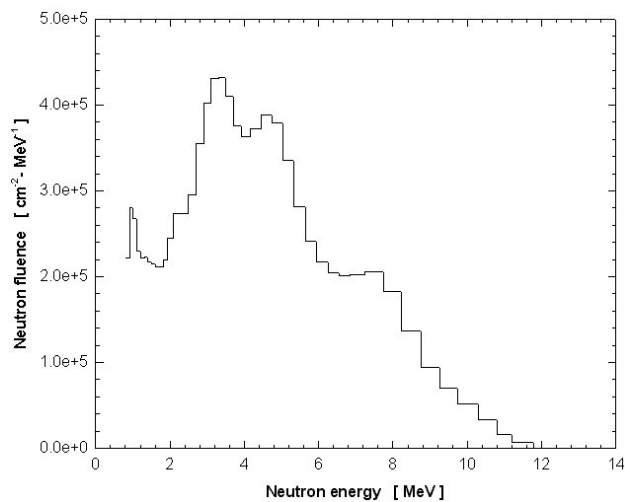


FIGURE 2. $^{239}\text{PuBe}$ neutron source spectrum.

- large scattering cross section,
- large average increase in lethargy per collision,
- small neutron capture cross section.

Even when H_2O , in comparison with D_2O , is ruled out by the third criterion, H_2O was included in this study, because its cost is low and it is easy to obtain.

The MCNP output includes per energy bin, the neutron fluence per source neutron; with this, the average neutron energy, the percentage of low energy neutrons, the dose,

the dose equivalent and the quality factor were calculated [36, 39]. The average neutron energy was calculated using,

$$\bar{E} = \frac{\sum_i \phi_i E_i}{\sum_i \phi_i}, \tag{1}$$

here ϕ_i is the neutron fluence in the i^{th} energy bin E_i , the percentage of low energy neutrons was calculated using Eq. (2), where ϕ_1 is the neutron fluence in the first energy bin

$$\% n_{TH} = \frac{\phi_1}{\sum_i \phi_i} 100. \tag{2}$$

The dose was calculated using the fluence-to-dose conversion factors h_D , from NCRP 38 [37], using

$$D = \sum_i \phi_i h_{D_i}. \tag{3}$$

The dose equivalent was determined using the fluence-to-dose equivalent conversion factors h_H , from NCRP 38, using,

$$H = \sum_i \phi_i h_{H_i}. \tag{4}$$

The quality factor was obtained using the dose equivalent to absorbed dose ratio, $Q = H/D$.

From all cases studied, the moderator that produced the larger percentage of low energy neutrons and the lower contribution of epithermal and fast neutrons, was selected to analyze the effect of adding a graphite reflector. Three different reflector thickness were analyzed, 5, 10 and 15 cm. The reflector was modeled as a spherical shell with 38.5 cm inner radius, leaving a 14 cm-thickness spherical shell as irradiation zone. The neutron fluence was tallied at 27 and at 100 cm from the center of the spherical shells.

MCNP calculations were also carried out for ²³⁹PuBe bare in air. Neutron fluences were calculated at 27.0 and 100.0 cm. This last result was used to compare with experimental and calculated data published in literature.

3. Results

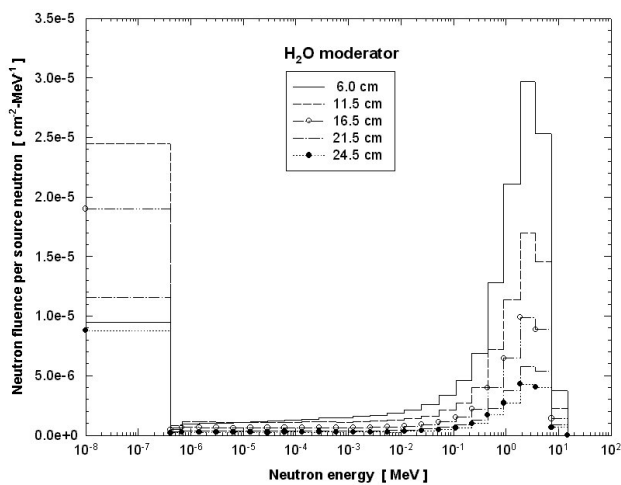
The neutron spectra at 27 cm originating from a ²³⁹PuBe source located at the center of a H₂O moderating medium with different radii (6.0, 11.5, 16.5, 21.5 and 24.5 cm) are shown in Fig. 3. As the H₂O thickness is increased fast neutrons are moderated shifting the spectra into lower energies. In Table I the total neutron fluence per source neutron, the percentage of low energy neutrons, average energy, absorbed dose, equivalent dose and quality factors in function of moderator radius are shown for six moderator radii. From these results we can notice that with the 6.0 cm-radius moderator, the total neutron fluence at 27 cm is quite similar to the total neutron fluence in air(see Table IV) (1.4×10^{-4} neutrons-cm⁻²/source neutron), this means that neutron loss is negligible. Figure 4 shows the neutron spectra produced by ²³⁹PuBe neutron source inside D₂O moderator media, its dosimetric features are shown in Table II. Comparing the total neutron fluences, produced by the 6.0 cm-radius H₂O and D₂O moderators, we can notice that the values are practically the same, nevertheless the percentage of low energy neutrons is larger for the H₂O moderator. Also, the fraction of low energy neutrons is larger for 6.0, 7.0, 11.5, and 16.5 cm H₂O moderator radii. The total neutron fluence is less than the total fluence using the D₂O moderator. This is because absorption cross section for H₂O is larger than for D₂O. Probable explanation of this behavior is as follows: For small scattering angles between neutrons and low atomic nuclei, as hydrogen and deuterium, the neutrons lose approximately the same amount of energy. For larger scattering angles, neutrons lose a larger amount of energy when they collide with hydrogen in comparison with collisions with deuterium. Thus, the H₂O moderator inner shell provides the presence of thermal neutrons before neutron absorption becomes relevant.

TABLE I. Neutron spectra features, at 27 cm, produced by a ²³⁹PuBe neutron source inserted at the center of a H₂O-based-moderator media.

Radius [cm]	Neutron Fluence [n-cm ⁻² /sn]	Low energy neutrons [%]	Average Energy [MeV]	Absorbed Dose [Gy]	Equivalent Dose [Sv]	Quality Factor [Sv/Gy]
6.0	1.39E(-4)	6.81	2.20	4.32E(-15)	3.61E(-14)	8.35
7.0	1.36E(-4)	10.35	2.04	3.99E(-15)	3.30E(-14)	8.27
11.5	1.04E(-4)	23.51	1.69	2.61E(-15)	2.09E(-14)	8.00
16.5	6.48E(-5)	29.34	1.62	1.55E(-15)	1.22E(-14)	7.88
21.5	3.83E(-5)	30.26	1.65	9.19E(-16)	7.20E(-15)	7.83
24.5	2.85E(-5)	31.15	1.63	6.78E(-16)	5.31E(-15)	7.83

TABLE II. Neutron spectra features, at 27 cm, produced by a $^{239}\text{PuBe}$ neutron source inserted at the center of a D_2O -based-moderator media.

Radius [cm]	Neutron Fluence [n-cm ⁻² /sn]	Low energy neutrons [%]	Average Energy [MeV]	Absorbed Dose [Gy]	Equivalent Dose [Sv]	Quality Factor [Sv/Gy]
6.0	1.42E(-4)	0.02	2.28	4.56E(-15)	3.86E(-14)	8.45
7.0	1.42E(-4)	0.07	2.09	4.27E(-15)	3.57E(-14)	8.37
11.5	1.46E(-4)	2.21	1.38	3.15E(-15)	2.47E(-14)	7.84
16.5	1.54E(-4)	12.68	0.83	2.33E(-15)	1.62E(-14)	6.95
21.5	1.68E(-4)	30.81	0.48	1.86E(-15)	1.10E(-14)	5.92
24.5	1.86E(-4)	43.19	0.34	1.74E(-15)	9.15E(-15)	5.25

FIGURE 3. Neutron spectra, at 27 cm, produced by a $^{239}\text{PuBe}$ neutron source inserted at the center of a set of 6.0, 11.5, 16.5, 21.5 and 24.5-cm-radii H_2O moderator media.

Moderator studies are mostly carried out using a single moderator [4-6, 9-21, 23]. A comparison of the obtained results with H_2O and the D_2O suggests us to study the moderating features of the heterogeneous combination of both moderators ($\text{H}_2\text{O}/\text{D}_2\text{O}$). In this case the second spherical shell (5.0 cm-radius) was filled with H_2O and the third spherical shell, with variable radii, was filled with D_2O . Calculated neutron spectra are shown in Fig. 5, while the total number of neutrons per source neutron at 27 cm from the $^{239}\text{PuBe}$ center, the percentage of low energy neutrons and the spectrum average energy are shown in Table III. Neutron fluence and dosimetric features are approximately alike to the D_2O moderator, except the percentage of low energy neutron, however the $\text{H}_2\text{O}/\text{D}_2\text{O}$ moderator has 495.3 cm³ less heavy water.

Plots of the calculated spectra for the same size, but different type moderators are shown in Figs. 6, 7 and 8 where moderating effect can be observed. In Fig. 6, the 6.0 cm-radius container filled with of H_2O (4.5 cm H_2O thickness)

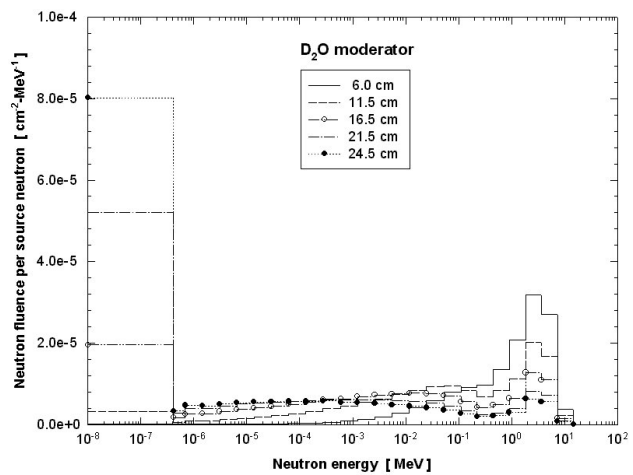
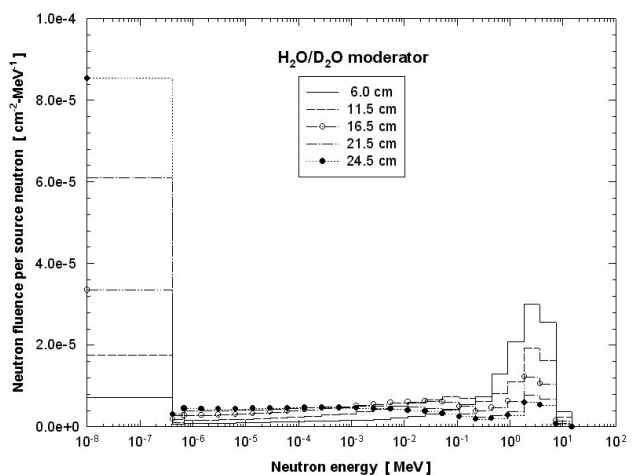
FIGURE 4. Neutron spectra, at 27 cm, produced by a $^{239}\text{PuBe}$ neutron source inserted at the center of a set of 6.0, 11.5, 16.5, 21.5 and 24.5-cm-radii D_2O moderator media.FIGURE 5. Neutron spectra, at 27 cm, produced by a $^{239}\text{PuBe}$ neutron source inserted at the center of a set of 6.0, 11.5, 16.5, 21.5 and 24.5-cm-radii $\text{H}_2\text{O}/\text{D}_2\text{O}$ moderator media.

TABLE III. Neutron spectra features, at 27 cm, produced by a ²³⁹PuBe neutron source inserted at the center of a H₂O/D₂O-based-moderator media.

Radius [cm]	Neutron Fluence [n·cm ⁻² /sn]	Low energy neutrons [%]	Average Energy [MeV]	Absorbed Dose [Gy]	Equivalent Dose [Sv]	Quality Factor [Sv/Gy]
6.0	1.40E(-4)	5.11	2.20	4.37E(-15)	3.66E(-14)	8.37
7.0	1.41E(-4)	6.37	2.03	4.10E(-15)	3.40E(-14)	8.28
11.5	1.43E(-4)	12.16	1.35	3.04E(-15)	2.36E(-14)	7.77
16.5	1.49E(-4)	22.31	0.82	2.24E(-15)	1.55E(-14)	6.93
21.5	1.63E(-4)	37.45	0.48	1.79E(-15)	1.06E(-14)	5.90
24.5	1.79E(-4)	47.77	0.34	1.67E(-15)	8.78E(-15)	5.25

and the heterogeneous, made with 3.5 cm thickness of H₂O and 1 cm thickness of D₂O, produce low energy neutrons, while the same container filled with D₂O does not. In Fig. 7 the 16.5 cm-radius container filled with H₂O or D₂O produces approximately the same amount of low energy neutrons, while H₂O moderator produces less total neutrons because neutron absorption is occurring. In Figure 8 the 24.5 cm radius container with D₂O or H₂O/D₂O produce a larger amount of low energy neutrons in comparison with H₂O, where neutron absorption is larger. From these results we found that H₂O moderator has poorer features compared with the D₂O and H₂O/D₂O moderators. These last two moderators have approximately similar moderating properties, but H₂O/D₂O moderator utilizes less heavy water than the D₂O moderator. This leads us to select the 24.5 cm-radius H₂O/D₂O moderator to study the effect of adding a reflector. The reflector's effect was calculated using three different graphite's thickness, 5, 10 and 15 cm. Graphite reflector reduces the energy of the neutrons and returns back some of those leaking out.

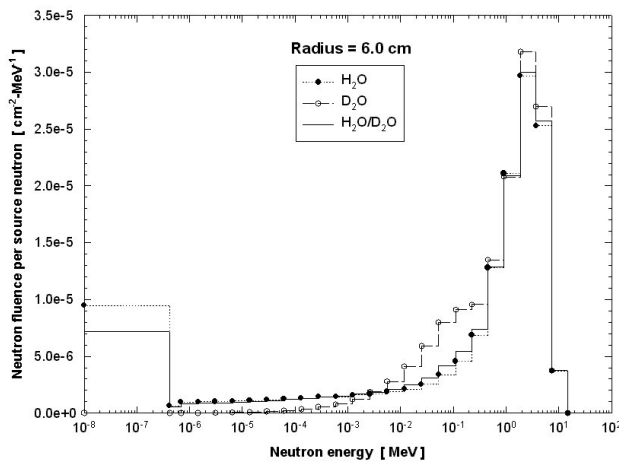


FIGURE 6. Neutron spectra, at 27 cm, produced by a ²³⁹PuBe neutron source inserted at the center of 6.0-cm-radius moderator media (H₂O, D₂O and H₂O/D₂O).

Neutron energy spectra were calculated at 27 cm from the center of the neutron source, and at 57 cm outside the moderator with reflector. ²³⁹PuBe neutron spectra, at 27 cm, inserted in H₂O/D₂O 24.5-cm-radius moderator, with and

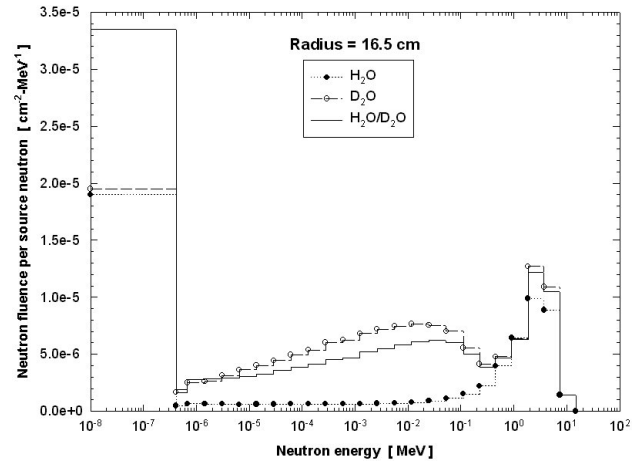


FIGURE 7. Neutron spectra, at 27 cm, produced by a ²³⁹PuBe neutron source inserted at the center of 16.5-cm-radius moderator media (H₂O, D₂O and H₂O/D₂O).

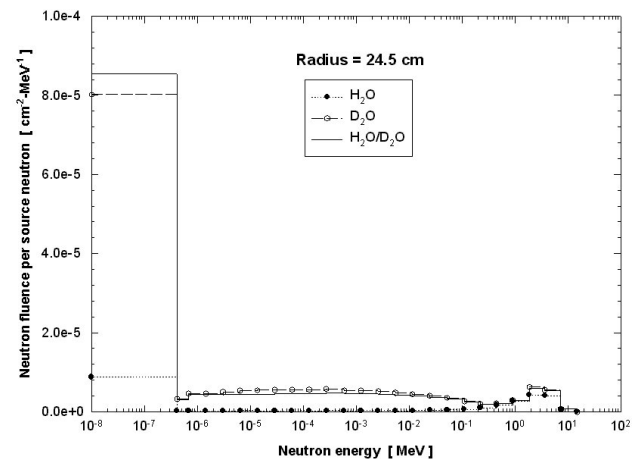


FIGURE 8. Neutron spectra, at 27 cm, produced by a ²³⁹PuBe neutron source inserted at the center of 24.5-cm-radius moderator media (H₂O, D₂O and H₂O/D₂O).

without graphite reflector are shown in Fig. 9. In Table IV the neutron spectra features are shown. Using 15 cm-thickness graphite reflector drastically reduces fast and epithermal neutrons in comparison with the moderator media without reflector, while the percentage of low energy neutrons increase from 47.8 % to 75.8 %. Total fluence per source neutron is 1.1×10^{-3} for moderator with reflector, meanwhile is 1.8×10^{-4} for the moderator without it. At 100 cm from the center of this moderator with reflector there is a neutron fluence of $1.04 \times 10^{-5} \text{ cm}^{-2}/\text{source neutron}$ and a dose equivalent of $2.75 \times 10^{-16} \text{ Sv}$. This dose is 14 times less than the dose produced by the bare source.

Bare $^{239}\text{PuBe}$ spectrum in air was also calculated, it was compared with experimental published results [41, 43, 44]. All spectra have the same features: peak is between 10^{-2} to 15 MeV and peak maximum is located between 1.9 to 3.7 MeV.

The dose equivalent per unit fluence and the average neutron energy were used to compare with results published in literature [45, 46], the comparison is shown in Table V, where the agreement is good.

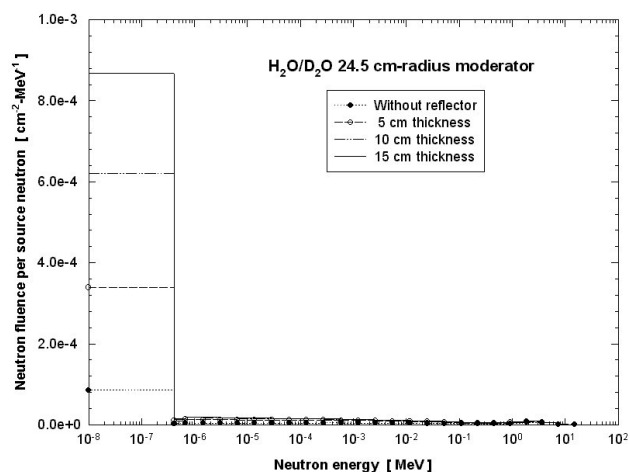


FIGURE 9. Neutron spectra, at 27 cm, produced by a $^{239}\text{PuBe}$ neutron source inserted at the center of $\text{H}_2\text{O}/\text{D}_2\text{O}$ 24.5-cm-radius moderator, without and with graphite reflector (5, 10, and 15-cm-thickness).

TABLE IV. Neutron spectra features, at 27 cm, produced by a $^{239}\text{PuBe}$ neutron source bare in air and inserted at the center of the $\text{H}_2\text{O}/\text{D}_2\text{O}$ -based-moderator (24.5-cm-radius), without and with Graphite reflector (5, 10, and 15-cm-thick).

Case [cm]	Neutron Fluence [n-cm ⁻² /sn]	Low energy neutrons [%]	Average Energy [MeV]	Absorbed Dose [Gy]	Equivalent Dose [Sv]	Quality Factor [Sv/Gy]
Bare source	1.40E(-4)	0	4.32	5.94E(-15)	5.08E(-14)	8.54
Moderator without reflector	1.79E(-4)	47.8	0.34	1.67E(-15)	8.78E(-15)	5.25
Moderator with 5-cm-thick reflector	5.53E(-4)	61.3	0.14	3.94E(-15)	1.59E(-14)	4.05
Moderator with 10-cm-thick reflector	8.84E(-4)	70.3	0.09	5.80E(-15)	2.09E(-14)	3.60
Moderator with 15-cm-thick reflector	1.14E(-3)	75.8	0.07	7.21E(-15)	2.42E(-14)	3.36

TABLE V. Average neutron energy and dose equivalent produced by a bare $^{239}\text{PuBe}$.

Reference	Average neutron energy [MeV]	Dose equivalent [10 ⁻¹⁰ Sv-cm ⁻²]
Nachtigall, 1967	4.3 ± 0.2	3.52 ± 0.05
Anderson and Neff, 1972	4	3.55
Thorngate and Griffith, 1985	4.6	3.41
Griffith et al, 1990	not reported	3.95
NCRP, 1991	4.5 to 5	4.6 to 5.0
Buckner and Sims, 1992	4.57 ± 0.09	3.96 ± 0.04
Vega-Carrillo and Becerra, 2000	3.2	3.59
This work	4.32	3.61

4. Conclusions

The use of Boron Neutron Capture Therapy to treat brain cancer tumors requires a large number of epithermal neutrons that become thermal during its transport in the brain. Other

applications, like in vivo-hand neutron activation analysis of aluminum and neutron capture Synovectomy, require a large number of lower energy neutrons, because the body part under neutron bombardment is small and no substantial neutron moderation is achieved during neutron transport inside the joint. The use of Monte Carlo methods allows to design neutron irradiators with different purposes, during calculations neutron source and moderating materials can be tested, making this method an inexpensive way to optimize the final design. Some drawbacks of Monte Carlo methods that should be taken into consideration are, long computation times in order to achieve a valid statistical uncertainty, to know as exact as possible the elemental composition of materials to achieve realistic results, and to use updated and validated cross section data. In the aim to obtain low energy neutrons from a $^{239}\text{PuBe}$ neutron source, to carry out basic research for Neutron Capture Synovectomy a Monte Carlo study of three moderator media were performed. In all cases neutrons with larger energy are shifted to lower energies. With the H_2O moderator some neutrons are absorbed by hydrogen reducing the total fluence and the percentage of low energy neu-

trons in comparison with the D_2O moderator. However using a heterogeneous combination of $\text{H}_2\text{O}/\text{D}_2\text{O}$ moderators produces similar moderating effect than D_2O moderator, using less amount of heavy water. This is because to small scattering angles, neutron losses approximately the same amount of energy in collisions with hydrogen or deuterium. To larger scattering angles between neutrons and hydrogen, the neutron losses more energy in comparison with collisions with deuterium.

A spherical 24.5 cm-radius moderator media made of a heterogeneous combination of $\text{H}_2\text{O}/\text{D}_2\text{O}$ shows approximately the same moderation quality as the same size D_2O moderator. Per each neutron emitted by de $^{239}\text{PuBe}$, the 24.5 cm-radius $\text{H}_2\text{O}/\text{D}_2\text{O}$ moderator produce at 27 cm a neu-

tron fluence of $1.8 \times 10^{-4} \text{ cm}^{-2}$, with an average energy of 0.34 MeV, where 47.8 % have an energy $\leq 0.4 \text{ eV}$. To increase the total fluence and the low energy neutron contribution a graphite reflector was added. Thus, the heterogeneous moderator with 15-cm-thick graphite reflector produces, at 27 cm, a neutron fluence of $1.1 \times 10^{-3} \text{ cm}^{-2}$ per source neutron, with an average energy of 0.098 MeV, where 75.8 % have an energy $\leq 0.4 \text{ eV}$. This percentage is 1.6 times larger than the moderator without reflector.

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