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ARTICLE

Fullerite Nano-materials as a Moderator in Neutron Irradiators with Radium Sources: Feasibility and Advantages

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Abstract: In this study, the feasibility of using fullerite nano-materials as a moderator in ^{226}Ra-Be neutron irradiators has been theoretically investigated, for the first time. Thermal, intermediate and rapid neutron flux in irradiation channels was calculated using the MCNP5 code when a fullerite nano-material was used as a moderator. The simulation results were then compared with other simulation results performed when paraffin was used as a moderator. The comparison showed that using fullerite instead of paraffin as a moderator results in an increase in the total number of irradiation neutrons by more than twice in average (240 %) for each direction inside the irradiator. This increase is distributed as follows: 27.84 %, 87.84 % and 124.32 % thermal, intermediate and rapid neutrons, respectively. The previous distribution indicates a significant increase in the intermediate and fast neutron flux. This is considered as an additional advantage of using the ^{226}Ra-Be neutron irradiator with a fullerite moderator. The irradiator can then be used not only to irradiate the materials whose irradiation requires thermal neutrons, but also those that require medium- to high-energy neutrons.

Keywords: Neutronic irradiator, Ra-Be radiation source, Cadmium, Neutron flux, MCNP5-beta code.

PACS: Neutrons diffusion and moderation, 28.20.Gd, Moderators (nuclear reactors), 28.41.Pa.

Introduction

Many countries began applying nanotechnologies in the nuclear industry since the 1970s [1, 2], which included: reactor moderators [3], high-density nuclear fuel with nanotechnology additives [4] and the activation of thermal hardening processes by nano-technology additions as one of the trends in technology innovation for new types of nuclear reactor fuel, such as plutonium oxides, uranium and nitrides for fast novel neutron reactors [5]. In recent years, the discovery of a new form of carbon nano-particle called fullerenes has attracted the interest of researchers to gain new basic knowledge of matter and the potential for its practical implementing applications [6]. Fullerenes are carbon molecules forming pentagonal and hexagonal rings and this term is currently applied to a broad class of atomic carbon molecules of general formula C_n (n even) that have a closed hollow polyhedron shape [7]. The most famous fullerenes are C_{60} and C_{70}. Other fullerene molecules that contain different numbers of carbon atoms from 36 to 540 and more have been manufactured and studied [7]. Among all known fullerene molecules, the C_{60} molecule is the most symmetrical molecule to date. It consists of sixty carbon atoms located on a spherical surface with a diameter of ~ 1 nm, as shown in Fig. 1.

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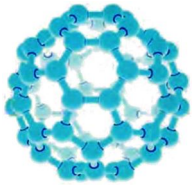

FIG. 1. Fullerene molecule for C_{60}.

C_{60} molecules are arranged spatially under certain conditions, where they are placed in crystal lattice nodes. Structures of a solid phase formed on the basis of fullerene molecules are called fullerites [8]. The C_{60} fullerite crystal has a FCC type cubic mesh structure with a constant value of 1.42 nm, while its density is 1.65 ± 0.03 g/cm^3 indicating a stable substance in the air. It does not dissolve and does not degrade even, after which it begins to sublime. Fig. 2 shows the lattice structure of a C_{60} crystal, where C_{60} molecules are mainly bound in the crystal by the van der Waals forces [8].

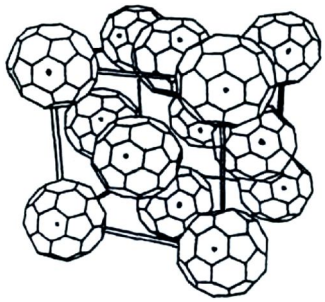

FIG. 2. C_{60} crystalline lattice structure.

Fullerite moderator was used with U^{235} to produce a homogeneous subcritical nuclear reactor [9]. According to the authors of [9], the reactor becomes subcritical when the fullerite moderator is transformed into a diamond-like form under pressure.

In this research, the feasibility of a use of fullerite nano-material as a moderator in ^{226}Ra-Be paraffin neutron irradiators has been investigated, for the first time. The viability of using fullerite nano-material moderator was demonstrated theoretically by using the MCNP5 code [10]. It can be used to calculate neutron flux values in each irradiation channel of ^{226}Ra-Be irradiator available at the Physics Department of the Faculty of Science, Damascus University. The neutron flux was calculated in two conditions. These are when paraffin and then fullerite nano-material was used as a neutron moderator. Neutron flux in each irradiated channel of the two studied states was then compared.

Modeling of ^{226}Ra-Be Irradiator

The ^{226}Ra-Be irradiator is one of the isotopic neutron irradiators that are used to obtain fast, intermediate and thermal neutron flux. The irradiator was made by the PHYWE German company. It consists of an isotope source ^{226}Ra-Be and a paraffinic moderator surrounded by a 4 mm thick parallelepiped steel container of dimensions 50 x 50 x 60 cm^3. The container is covered with a rectangular steel lid. The moderator in its turn contains ten cylindrical irradiation channels and a rectangular gap that is dedicated to insert a cadmium plate. Each channel is surrounded by a 1 mm thick and 2.2 cm diameter steel liner. The rectangular gap is also surrounded by a 1 mm thick steel liner. Five of the ten irradiation channels are distributed around the ^{226}Ra-Be neutron source on the circumference of a 10 cm radius. Three of the remaining five channels are situated on the ox axis at 15, 20 and 25 cm away from the source. The last two channels are located at diagonal distances of 20 and 25 cm from the radium-beryllium source. Positions of the irradiation channels, the cadmium plate and the ^{226}Ra-Be neutron source are shown in Fig. 3 and described in Table (1).

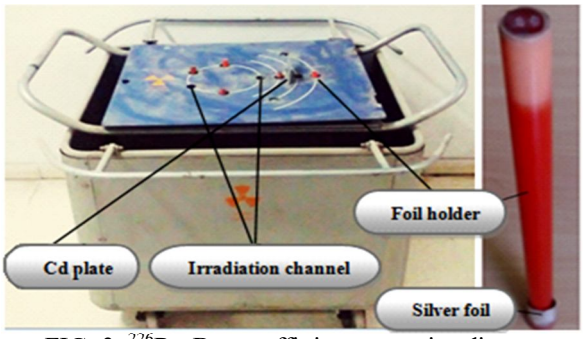

FIG. 3. ^{226}Ra-Be paraffinic neutron irradiator.

The ^{226}Ra-Be neutron source consists of a homogeneous mixture of the ^{9}Be and ^{226}Ra isotopes, where beryllium forms most of its material (Ra to Be ratio is 1/5). The mixture weight and the source's activity are 3.5 mg and 9.09×10^4 n/s, respectively. The ^{226}Ra-Be neutron source is surrounded by a 7 cm length and 2 cm diameter double-walled nickel-steel cylindrical tube. The tube was itself surrounded by a 7 cm length and 4 cm diameter cylindrical lead. Fig. 4 illustrates (a) the spectrum of the neutron source ^{226}Ra-Be according to reference [11] and (b) the spectrum from which values used in the simulation of this research were obtained.

TABLE 1. ^{226}Ra-Be radioactive physical properties that have been applied in the simulation with MCNP5 code.

Name	Shape	Name	Distance from ^{226}Ra-Be source (cm)	Notice
Steel container	Parallel rectangles	Steel container		Contains paraffin moderator
Cadmium	Plate	Cadmium	17.5	On ox axis
Channel number	Distance of 2 cm diameter cylindrical irradiation channels from ^{226}Ra-Be source (cm)			
1		10		On oy axis
2		10		On ox axis
3		-10		On oy axis
4		-10		In the xoy plane
5		-10		In the xoy plane
6		15		On ox axis
7		18.023		In the xoy plane
8		20		On ox axis
9		25		In the xoy plane
10		25		On ox axis
Sample holder	Plastic cylinder with 30 cm length and 0.5 cm radius			Length inside the paraffin is 20 cm.
^{226}Ra-Be Source				
^{226}Ra-Be mixture weight	3.5 mg	The mixture is housed within a double-walled nickel-steel cylindrical tube with 7 cm length and 2 cm diameter.		
^{226}Ra to ^{9}Be ratio	1/5			
Lead barrier	Cylinder with 7 cm length and 2 cm diameter surrounds the tube.			
Lead holder	Cylinder with 18 cm length and 4 cm diameter.			

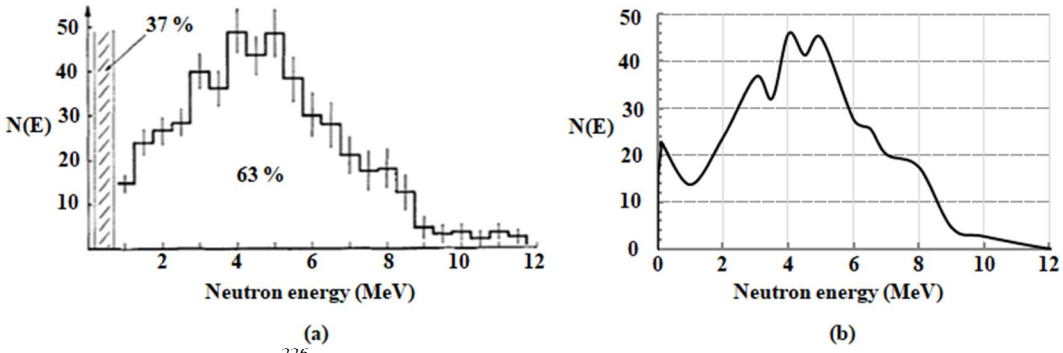

FIG. 4. Neutrons spectrum of ^{226}Ra-Be source (a) according to reference [11] and (b) the values used in the simulation.

MCNP5 code was used to model ^{226}Ra-Be paraffinic irradiator. This code is applied widely around the world to model neutral particle radiography, photon, electron or coupled neutron, photon and electron transport, for three-dimensional or time-dependent general geometry [10]. The ^{226}Ra-Be neutron source within the SDEF source ID card provided in the MCNP5 code has been described as a point source located at (0,0,0); the centre of the Cartesian coordinate system is supported in the modeling. The considered point source is at a 25 cm depth from the top surface of the irradiator. It is also 35 cm and 25 cm away from the right and the left irradiator surface, respectively (Fig. 5).

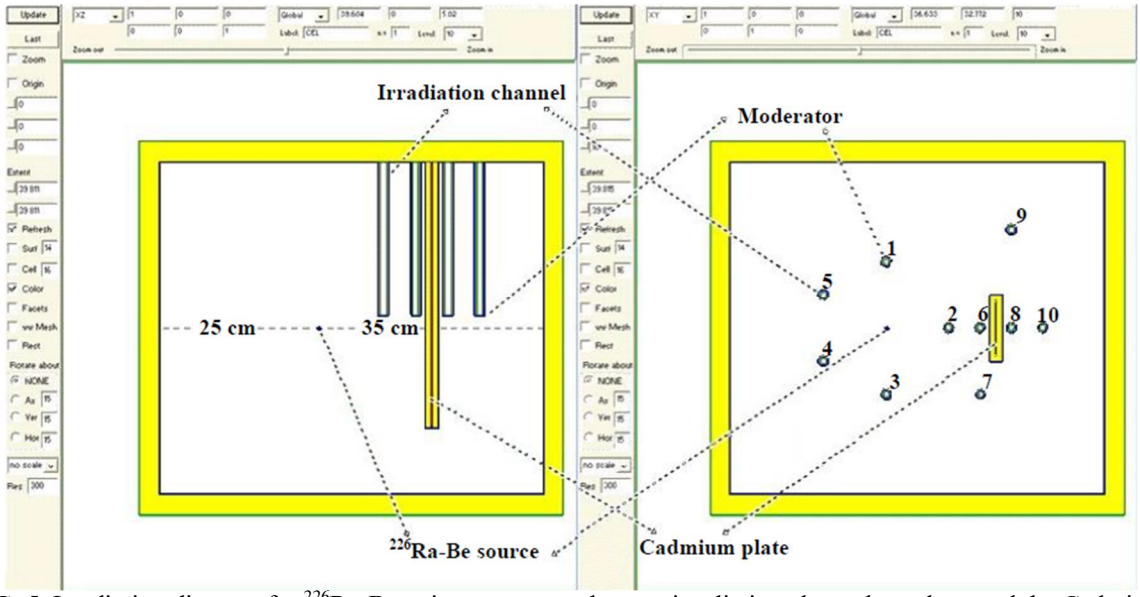

FIG. 5. Irradiation diagram for ^{226}Ra-Be point source, moderator, irradiation channel numbers and the Cadmium plate (Cd) in the order that was used in the input entry file of the MCNP5 code.

Neutron Flux Calculation within the Irradiation Channels ^{226}Ra-Be

The F4 (Tally) card provided in the MCNP5 code was used to compute the entire neutron flux in each channel using: 1. paraffin and 2. fullerite with densities of 0.904 g / cm^3 and 1.65 g / cm^3, respectively as a moderator. Fig. 6 shows the variation of the neutron flux in the irradiator channels for the neutron energy (calculated by MCNP5 code for each lethargy unit) when the paraffinic moderator was used.

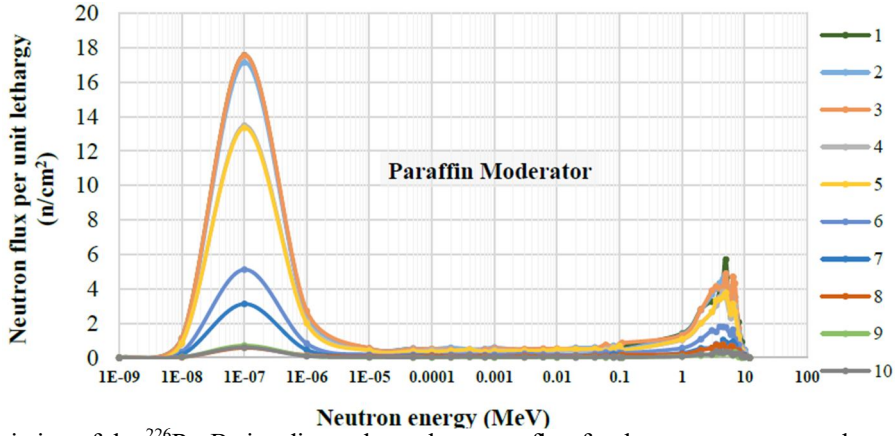

FIG. 6. Variation of the ^{226}Ra-Be irradiator channel neutron flux for the neutron energy when the paraffin moderator was used.

Fig. 7 shows the variation of the neutron flux in the irradiator channels for the neutron energy (calculated by MCNP5 code for each lethargy unit) using the fullerite moderator.

Fig. 8 shows a comparison of average neutron flux rate in all ten irradiation channels shown in Figs. 6 and 7 with paraffin and fullerite as moderator, respectively.

Fig. 9 shows a comparison of average neutron flux rate in the cadmium plate where both paraffin and fullerite were used as a moderator for neutrons. The cadmium plate absorbs the thermal neutrons to produce fast neutrons. This reduces the thermal neutron flux in the irradiation channels behind it.

Fig. 10 shows a comparison of the total neutron flux in each of the ten irradiation channels separately for the use of paraffin and fullerite as a neutron moderator.

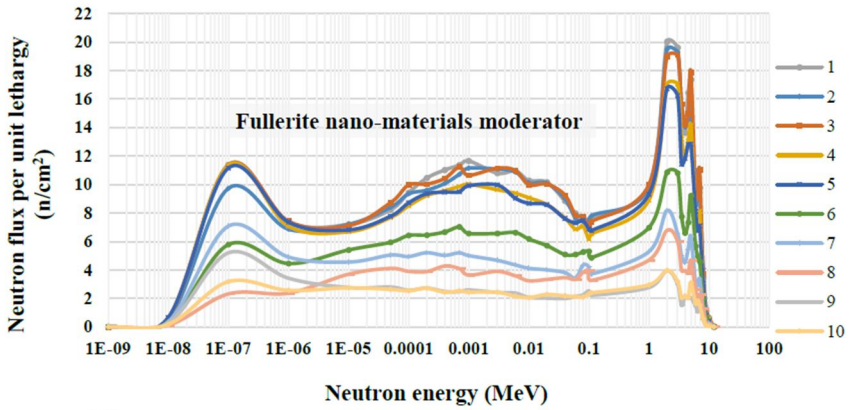

FIG. 7. Variation of the ^{226}Ra-Be irradiator channel neutron flux for the neutron energy when the fullerite (C_{60}) moderator was used.

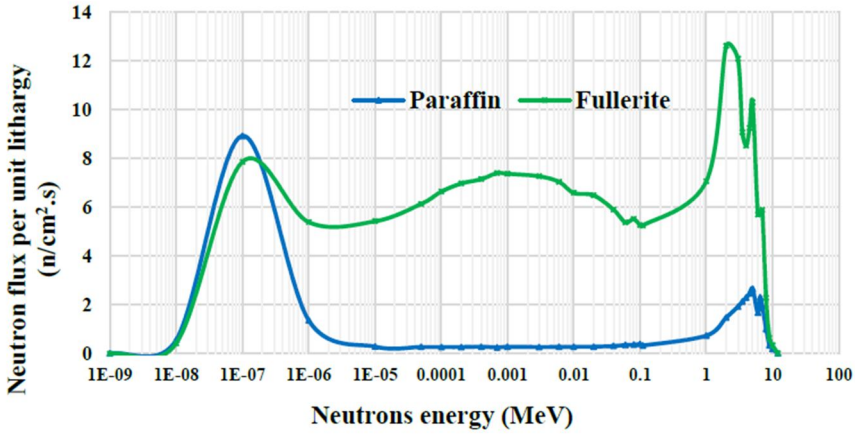

FIG. 8. A comparison of average neutron flux rate in all ten irradiation channels with paraffin and fullerite as moderator, respectively.

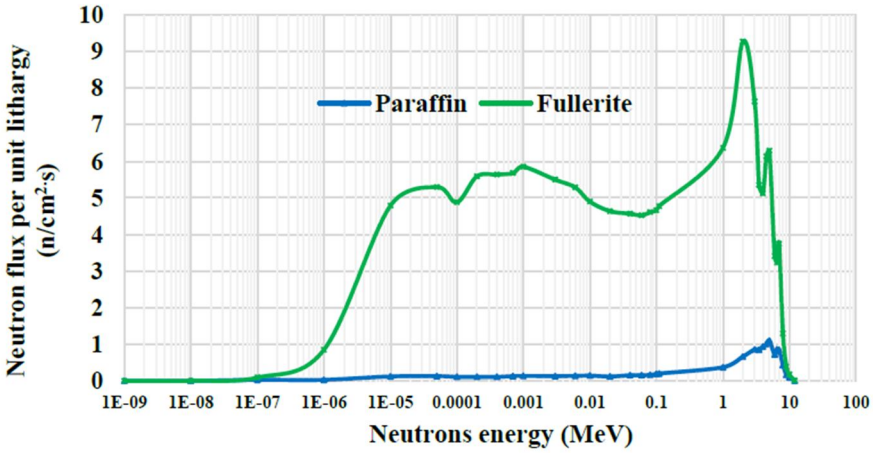

FIG. 9. A comparison of average neutron flux rate in the cadmium plate with paraffin and fullerite as moderator, respectively.

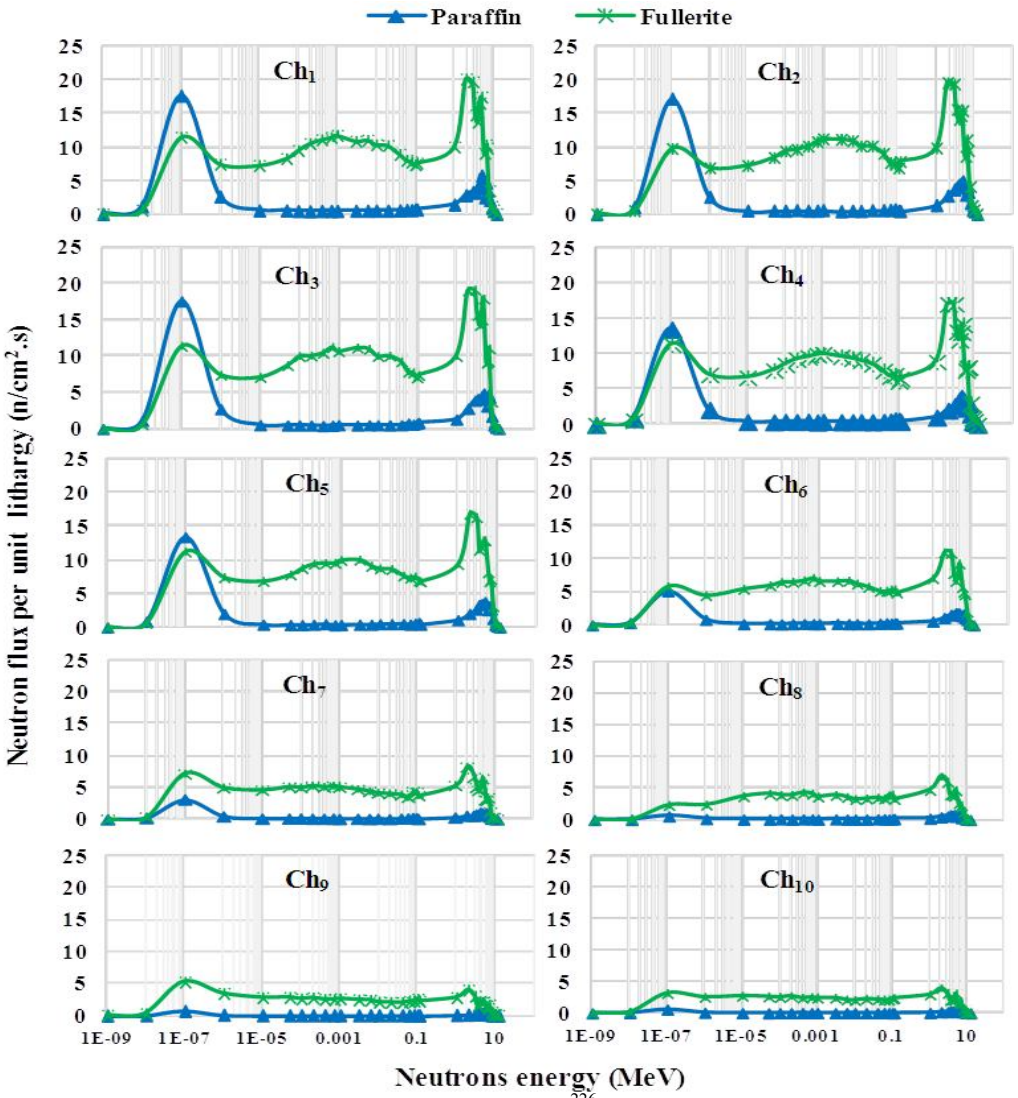

FIG. 10. Variation of the neutron flux in each channel of the ^{226}Ra-Be irradiator for the neutron energy where both paraffin and fullerite were used as neutron moderator, respectively.

Results and Discussion

Figs. 8 and 9 show that there is a significant change in the distribution of the intermediate and fast neutron fluxes in the irradiation channels and the cadmium plate for using fullerite instead of paraffin as a neutron moderator. In the intermediate energy field, it is observed that the neutron flux is virtually non-existent in all channels when the paraffin moderator is used. This is because most of the fast neutrons slow down to thermal energy within the paraffin moderator. Paraffin moderator also absorbs a large number of neutrons that pass through it. Paraffin has a high capability (cross-section) of moderating neutrons from a fast to a thermal range. It also has a large absorption cross-section for thermal neutrons, which leads to loss of a significant number of these neutrons as a result of being absorbed during the moderation process. The intermediate neutron flux increases clearly in the case of using fullerite as a moderator and its maximum value ranges between 2 - 11 n/cm^{2}s. This indicates that a ^{226}Ra-Be irradiator with fullerite moderator can irradiate substances requiring only intermediate to fast energy neutrons, such as iron, nickel, aluminum, thorium and cobalt [12].

On the other hand, there are no significant differences in all ten irradiation channels within the thermal range between the two neutron flux curves of the fullerite and paraffin moderators. However, it can be concluded from Fig. 10 that:

1. It is preferred to use paraffin as a moderator for the channels close to the source (less than 10 cm). The peak of the thermal neutron flux for the paraffin moderator is higher in this case.

2. There is no advantage in using either fullerite or paraffin moderator for the channels within a distance of 10 - 15 cm from the irradiation source. The thermal neutron flux is approximately the same for both moderators.
3. It is preferred to use fullerite as a moderator for channels located at distances greater than 15 cm from the source. The thermal neutron flux in paraffin medium begins to decrease at a distance greater than 15 cm until it practically approaches zero at a distance of 25 cm from the source.

Fig. 11 shows the variation of the thermal neutron flux as a function of the channel's distance from the irradiation source when using paraffin and fullerite, respectively, as a neutron moderator.

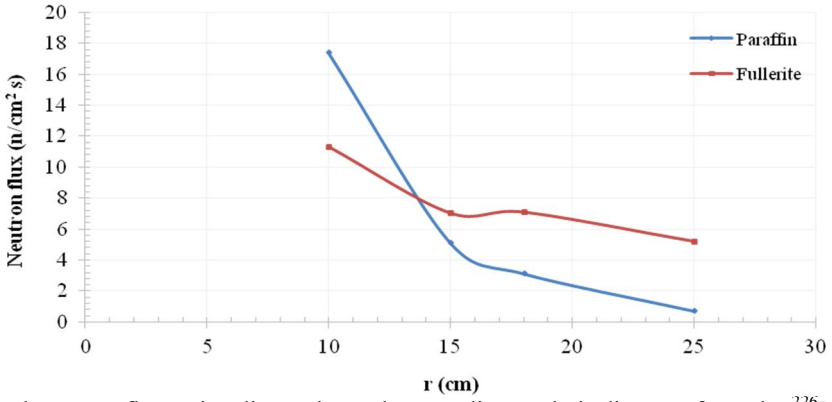

FIG. 11. Thermal neutron flux at irradiator channels according to their distance from the ^{226}Ra-Be irradiator when using paraffin and fullerite, respectively, as a neutron moderator.

Finally, It was also possible from Fig. 10 to calculate the average increase in neutron flux across the entire neutron energy range when fullerite is used instead of paraffin as a moderator. The average increase of neutron flux distributed in one direction was as follows: 27.84 %, 87.84 % and 124.32 % for the thermal, intermediate and rapid neutrons, respectively. Thus, the average total increase of the neutron flux was about 240% when replacing the paraffin as a moderator with fullerite moderator. This means that the number of irradiation neutrons rises by more than double when fullerite was used as a moderator. The simulation results, in addition to the weather and high temperature resistance of fullerite nanostructure, encourage the use of this type of moderator in radium source irradiators instead of paraffin.

Conclusion

The neutron flux in irradiation channels with fullerite as a moderator was calculated using the MCNP5 code for modeling the ^{226}Ra-Be neutron irradiator at Physics Department in the Faculty of Science at Damascus University. The neutron flux was also calculated when paraffin was used as a neutron moderator. The comparison between the calculated flux values showed some variations depending on the energy of the neutrons. The difference in the values of the thermal neutron flux also depends on the distance of the irradiation channel from the irradiator source. The value of flux in channels close to the irradiation source (at a distance less than 10 cm), where paraffin was used as a moderator, is slightly greater than that when using fullerite as a moderator. By contrast, the thermal neutron flux value in far channels (at a distance greater than 15 cm) increases if fullerite is used as a moderator. The upshots also confirm the presence of a strong moderation of fast and intermediate neutrons with large absorption of thermal neutrons in both the paraffin and the cadmium plate when using paraffin as a moderator in contrast to the use of fullerite for moderation. Thus, using fullerite as a moderator has many advantages. First, it provides the ability to irradiate materials of various types across the entire neutron spectrum. The magnitude of the irradiation neutron flux depends on the distance of the channel from the neutron source. Second, it absorbs fewer neutrons than paraffin. Finally, it has a solid structure and withstands extreme environmental conditions of high temperature and humidity. These advantages make fullerite more suitable than paraffin to be used as a moderator. This is still factual, although the thermal neutron flux in the channels close to the source is slightly better in the presence of paraffin than the same flux in the presence of fullerite.

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