Analysis by Monte Carlo of thermal neutron flux from a $^{241}\text{Am}/^9\text{Be}$ source for a system of trace analysis in materials

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HIGHLIGHTS

- Design of an enhanced thermal neutron source for use in trace detection in substances or materials, through NAA method.
- The thermal neutron irradiation system is using a $^{241}\text{Am}/^9\text{Be}$ source.
- Different configurations of polyethylene cylinders were simulated to choose the one giving the highest thermal neutron flux.
- The performance was evaluated with the MCNP6 code and validated with laboratory measurements with a miniature BF_3 detector.

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ABSTRACT

Neutron techniques to characterize materials have a wide range of applications, one of the major developments being the identification of terrorist threats with chemical, biological, radiological, nuclear and explosives (CBRNE) materials. In this work, a thermal neutron irradiation system, using a $^{241}\text{Am}/^9\text{Be}$ source of 111 GBq inside polyethylene cylindrical moderators, has been designed, built and tested. The geometry of moderator and the neutron source position were fixed trying to maximize the thermal neutrons flux emitted from the system. Therefore, the system is in fact a thermalized neutron source taking advantage of the backscattered neutrons, achieving thermal fluence rates of up to $5.3 \times 10^2 \text{ cm}^{-2} \text{ s}^{-1}$, with dominantly thermal spectra. Samples can be placed there for several hours and thereafter be measured to identify their component elements by NAA (Neutron Activation Analysis).

Through Monte Carlo techniques employing the MCNP6 code (Pelowitz et al., 2014), four different configurations with polyethylene cylinders were simulated to choose the most adequate geometry. The theoretical model was then replicated in the neutronics hall of the Neutron Measurements Laboratory of the Energy Engineering Department of Universidad Politécnica de Madrid (LMN-UPM), carrying out experimental measurements using a BF_3 neutron detector. A high agreement between MCNP6 results and the experimental values measured was observed.

Consequently, the system developed could be employed in future laboratory experiments, both for the identification of trace substances by NAA and for the calibration of neutron detection equipment.

1. - introduction

The detection of substances using neutrons is a non intrusive method with multiple applications in fields like agriculture, industry, mining and, of course, in security applications (Llovera et al., 2000; Datema et al., 2002). Neutron activation analysis (NAA) techniques can

provide both qualitative and quantitative information about material composition, but it is necessary to have a significant flux of particles, frequently obtained from reactors or accelerators (Travesí, 1975). When the pristine source intensity is low, the neutron flux can be increased by using backscattering of neutrons through collisions (Datema et al., 2002; Bedogni et al, 2016, 2017; Didi et al., 2017).

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NAA methods to identify substances are based on the irradiation of bulk materials with neutrons. When the target material captures neutrons, gamma rays are emitted promptly (10^{-14} to 10^{-12} s), with energies in the range from about 100 keV to 10 MeV, corresponding with the excitation energies of the nuclei formed after neutron capture reactions, which in turn are equal to the binding energy of the added neutron. The materials composition usually include hydrogen (H), carbon (C), nitrogen (N) and oxygen (O), elements whose emission spectrum is fully identified (Grodzins, 1991; Buffler, 2004). While the analysis of the gamma radiation spectrum allows to identify the component elements (Reilly et al., 1991), the intensity of the emitted gamma radiation depends on the amount of each isotope. The detection of these gamma rays is the principle of prompt gamma NAA (PGNAA) and it requires adequate gamma spectrometers (Travesí, 1975; Turhan, 2004; Uchai et al., 2008).

Alternatively, activated samples can be measured in the laboratory after neutron irradiation to identify the component elements by gamma spectrometry of their decay (Travesí, 1975). This is sometimes called delayed gamma neutron activation analysis (DNAA).

In this work, a thermal neutron irradiation system, using a $^{241}\text{Am}/^{90}\text{Be}$ source of 111 GBq inside polyethylene cylindrical moderators, has been designed, built and tested. These kind of neutron sources are widely used in many applications, including substances identification both in university laboratories and research centers (Uchai et al., 2008). $^{241}\text{Am}/^{90}\text{Be}$ sources are made of a compact mixture of finely powdered AmO_2 with Be (Lessing and Glagolenko, 2011; Didi et al., 2017). ^{241}Am has a half life of 432.17 ± 0.66 years; its radioactive decay produces two main radiations: an alpha particle of 5.486 MeV with 85.2% yield, and a gamma radiation with an energy of 59.54 keV with a yield of 35.9% (Eckert and Ziegler, 2010). $^{241}\text{Am}/^{90}\text{Be}$ produces neutrons with energies between 0.8 to 10 MeV with a mean energy of 3.2 MeV. The neutron production yield is estimated to be $7.0 \cdot 10^{-5}$ neutrons per alpha. (Vega Carrillo and Martinez Ovalle, 2016).

To increase the activation of nuclei it is necessary to moderate the fast neutrons released by the source to energies below 0.4 eV, since the capture cross section of isotopes of interest is usually much larger for thermal than for fast neutrons. For example, the thermal neutron ($E = 0.025$ eV) capture cross section of Nitrogen is 0.075 b while for 1 keV neutrons it is only $3.77 \cdot 10^{-4}$ b (NEA, 2019). Therefore, neutron moderators are required in activation techniques. Graphite and polyethylene show an excellent attenuation behavior moderating neutrons. Graphite produces less gamma rays compared with polyethylene, but it requires greater thickness (Bedogni et al., 2016) since the average energy lost by neutrons in each collision in graphite is lower than in polyethylene. The values of thermal neutron ($E = 0.025$ eV) cross sections for these two materials are shown in Table 1. The conclusion is that hydrogen rich materials usually offer the best performance in thermalizing neutrons (Vega Carrillo et al., 2007; Bedogni et al., 2016).

In summary, the main goal of this work has been to design a system able to reach the necessary flux of thermal neutrons for activation techniques or for the calibration of neutron detection equipment, by thermalizing fast neutrons released from the $^{241}\text{Am}/^{90}\text{Be}$ source in high density polyethylene (HDPE). The concept takes advantage of back scattering of neutrons in a HDPE cavity surrounding the neutron source, and it has some similarity to that described in (Bedogni et al., 2016) but, in essence, it is smaller and simpler, offering the advantage of its

easier construction and lower cost when small samples need to be analyzed. The system was simulated by Monte Carlo methods using MCNP6 code (Pelowitz et al., 2014), and the results obtained were compared with experimental measurements carried forward in the neutronics hall of the Neutron Measurements Laboratory of the Energy Engineering Department of Universidad Politécnica de Madrid (LMN UPM).

2. - Materials and methods

The main components and materials used in this work have been a $^{241}\text{Am}/^{90}\text{Be}$ neutron source of 111 GBq nominal activity, with a neutron emission intensity of $5.83 \pm 0.14 \cdot 10^6$ neutrons $^{-1}$ (Vega Carrillo et al., 2009), four cylinders of HDPE with different sizes used as moderator, and a miniature BF_3 neutron detector.

The first part of the work was focused in performing Monte Carlo simulations, in order to optimize the system geometry and configuration. The source from the LMN UPM was modeled with its real dimensions: outside diameter 1.48 cm and length 4.64 cm, three layers capsule made of 0.21 cm thickness stainless steel, 0.1 cm thickness lead and 0.2 cm thickness aluminum (Gallego et al., 2004).

The simulations were carried out with the MCNP6 code (Pelowitz et al., 2014), using F5 tallies, 222 groups neutron spectra, and ENDF/B VII cross section library for neutrons of $E < 20$ MeV. In order to reduce uncertainties to less than 5%, 10^8 histories were processed. The transport of thermal neutrons in polyethylene was included through the scattering matrix treatment, S (α, β) (Pelowitz et al., 2014).

The Neutronics Hall facility of LMN UPM was characterized using Monte Carlo calculations and verified through experimental procedures. Monte Carlo calculations included detailed models of the irradiation bench, the hall and the neutron source [Vega Carrillo et al., 2012], as it is shown in Fig. 1.

In each configuration, a new cylinder was added to the cylinder of the previous one. For example, in configuration 3, cylinder 3 was added outside cylinders 1 and 2. However, in configuration 4, the last cylinder (4) was added inside cylinder 1. In this way, each added cylinder increased the thickness of moderating material.

Using MCNP6 F5 tally, thermal neutron fluence rates ($E < 0.4$ eV) were estimated at different points out of the device along the main axis of symmetry. The calculated total fluence rate spectra were multiplied by the source strength. The center of the source was taken as the reference point and calculations and measurements were made at distances $Z = 29, 30, 31, 32, 33, 34, 35, 36$, and 37 cm, respectively (36 simulations), as indicated in Fig. 2.

Once the system was studied with MCNP6 code, the assembly was

Fig. 1. Scheme of the MCNP6 model of the irradiation bench at LMN-UPM with the Am/Be source and HDPE moderator cylinders.

Table 1
Thermal neutron ($E = 0.0253$ eV) cross section of hydrogen and graphite [from NEA, 2019].

Element	σ_s (barn)	σ_a (barn)
Hydrogen	30.08	0.3327
Graphite	4.95	0.00386

Fig. 2. Scheme of the geometry, materials and detector distances considered in the MCNP6 simulations. Up left: configuration 1, 1 HDPE cylinder 1.2 cm thickness surrounding the source. Up right: configuration 2, 2 HDPE cylinders with 2.45 cm total thickness. Low left: configuration 3, 3 HDPE cylinders with 4.95 cm total thickness. Low right: configuration 4, with an added cylinder of 1.3 cm thickness inside.

built in the in the irradiation bench of the neutronics hall of LMN UPM (Fig. 3), performing the appropriate measurements to obtain experimental data.

A miniature BF_3 thermal neutron detector (Nuclear Chicago model NC 201 inside the probe NC 202, with 0.48 cm $\varnothing$ x 1.27 cm length; 96% enriched in ^{10}B at 700 mm internal pressure) was selected due to its very high thermal neutron absorption cross section for $^{10}\text{B}_5$ (3843.5 b) (NEA, 2019), and also because the kinetic energy of the charged particles (2 MeV) released, is fairly easy to detect (Tsoulfanidis and Landsberger, 2015).

Concerning radiation protection, the laboratory staff uses individual

dosimeters and area monitors for neutron and gamma radiation are available to control dose. However, laboratory staff should not stay around the irradiation bench during irradiations, because the transfer of the source to the irradiation bench and the irradiation time is automated with a pneumatic system controlled by a PLC (Programmable Logic Controller). Therefore, the dose records during the experiments were negligible.

3. results

Results of the MCNP6 calculations for configurations 1, 2 and 3 are

Fig. 3. Irradiation bench at the LMN-UPM neutronics hall with the HDPE cylinders and BF_3 neutron detector.

Fig. 4. Calculated thermal neutron ($E < 0.4 \text{ eV}$) fluence rate vs. source-to-detector distance for configurations 1, 2 and 3.

shown in Fig. 4, comparing the values of neutron flux with thermal energy ($E < 0.4 \text{ eV}$) at different irradiation points from the center of the source. It can be seen how increasing the HDPE thickness also increases the backscattered thermalized neutron flux at all the reference points considered.

Additionally, Fig. 5 displays the results obtained for configurations 3 and 4. It is observed that the additional internal HDPE cylinder added for configuration 4 is not optimal, because neutron captures in this added cylinder reduce the thermal flux. Consequently, configuration 3 is the one preferred.

Figs. 6–10 show the neutron spectra of the bare $^{241}\text{Am}/^9\text{Be}$ neutron source and with the four configurations of HDPE cylinders considered, obtained with MCNP6. The figures are plotting the energy distribution

Fig. 5. Calculated thermal neutron ($E < 0.4 \text{ eV}$) fluence rate vs. source-to-detector distance for configurations 3 and 4.

Fig. 6. MCNP6 total calculated neutron spectra at different source-to-detector distances (bare Am/Be source).

Fig. 7. MCNP6 total calculated neutron spectra at different source-to-detector distances (Configuration 1 – HDPE thickness 1.2 cm).

Fig. 8. MCNP6 total calculated neutron spectra at different source-to-detector distances (Configuration 2 – HDPE thickness 2.45 cm).

of neutron fluence at nine irradiation points.

From MCNP6 calculations, it can be seen that the best source moderator configuration between those studied is the number 3 (HDPE

Fig. 9. MCNP6 total calculated neutron spectra at different source-to-detector distances (Configuration 3 – HDPE thickness 4.95 cm).

Fig. 10. MCNP6 total calculated neutron spectra at different source-to-detector distances (Configuration 4 – inside additional cylinder).

Fig. 11. Comparison between MCNP6 calculation results and experimental measurements with the BF_3 detector for configuration 3 (HDPE thickness 4.95 cm).

thickness 4.95 cm). Therefore, it was chosen to be reproduced experimentally at LMN UPM. Thermal neutron flux was measured with the miniature BF_3 detector. The comparison between the experimental

measurements and the results obtained in the MCNP6 calculation for configuration 3 at the different points considered is shown in Fig. 11. It can be seen a good agreement for all the points except for the closest two were simulations indicate a higher flux, so it can be said that experimental measurements validated the Monte Carlo calculations.

4. - Conclusions

Several simulations were carried out with MCNP6 code to choose the best HDPE moderator configuration design and geometry and the neutron source position, looking to maximize the thermal neutron flux in the irradiation area where the sample to analyze must be placed. In each configuration, a new HDPE cylinder was added to the cylinders of the previous one. Taking the source center as reference point, the neutron fluxes and energy spectra were estimated at several points.

Between the five configurations analyzed – bare $^{241}\text{Am}/^{9}\text{Be}$ source plus four additions of HDPE cylinders– the one giving maximum thermal fluence rates was configuration number 3 (Fig. 2). Results show that low energy neutrons increase when more HDPE cylinders are added, through scattering on the moderator allowing to achieve the highest thermal fluence rates (Fig. 4).

The spectra plot in Figs. 6 10 show that configuration number 3, with cylinder 16.5 cm of diameter and 4.95 cm of thickness achieves the best thermal neutron component, with a flux at the closest point of $5.35 \times 10^2 \text{ cm}^{-2} \text{ s}^{-1}$ (Fig. 4). If compared with configuration 4 (Fig. 5), with an added cylinder inside cylinder 1, the thermal neutron flux is smaller ($1.32 \times 10^2 \text{ cm}^{-2} \text{ s}^{-1}$), due to capture of thermal neutrons in the internal cylinder (Hsu and Kearfott, 1999), comparable to previous work developed by Bedogni (2016) or Zhang (2013). Monte Carlo simulations are very useful to design a source moderator setup that increases the thermal neutron output efficiency.

The experimental irradiation on the LMN UPM bench (Fig. 3) give measured thermal neutron flux comparable with the results obtained through MCNP6 code, thus validating the Monte Carlo calculations. However, for the first two points the measured values are slightly lower than the calculated ones, probably due the wall effect of the BF_3 detector, because of its small size (Knoll, 2000; Nasir et al., 2018).

To conclude, the device optimized by Monte Carlo method using MCNP6 code, experimentally tested and validated at the neutronics hall of LMN UPM, could be used hereafter in detection of trace of substances or materials, by NAA (Neutron Activation Analysis) method. The simple design makes this device a potentially interesting tool to characterize elements or to test and calibrate thermal neutron detectors.

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