Artificial Neural Networks applied in the Spectrometry of a $^{239}\text{Pu}$-Be source


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Abstract. To explore the potential use of a neutron source and to define the procedure to handle it under safety conditions, features like neutron spectrum and the ambient dose equivalent of the source must be known. The aim of this work was to determine the spectrum, the total fluence rate and the ambient dose equivalent of a 185 GBq $^{239}\text{Pu}$-Be neutron source. Using Monte Carlo methods the spectrum, the total fluence rate, and the ambient dose equivalent of a $^{239}\text{Pu}$-Be were calculated. The spectrum was calculated at 50, 100, 200 and 300 cm from the source in air using MCNP X and MCNP 4C codes. The neutron spectrum was also obtained, at 100 cm, using a Bonner sphere spectrometer whose count rates were used to unfold the neutron spectrum, the unfolding was carried out using an Artificial Neural Network for neutron spectrometry. With the spectrum, the total neutron fluence and the ambient dose equivalent were determined. Calculated results were compared with measured values where Monte Carlo results were smaller than those measured. These differences were attributed to the presence of $^{241}\text{Pu}$ during the source manufacturing. In order to match calculated and measured quantities a 0.102 w/o of $^{241}\text{Pu}$ was estimated. After corrections the differences between calculated and experimental results were 1%. This result shows the advantages of using Artificial Neural Networks technology in the unfolding of neutron spectrum using as a single piece of information the count rates of a Bonner sphere spectrometer.

KEYWORDS: Artificial Neural Networks, Unfolding, Bonner sphere spectrometer, Neutrons.

1. Introduction

Since its discovery [1, 2] to nowadays, neutrons have been widely utilized in several areas of science and technology. Neutrons are naturally and artificially generated; environmental neutrons are mainly produced as a product of nuclear reactions occurring between cosmic rays and atmospheric nuclei, and in $(\alpha, n)$ reactions with environmental nuclei, where $\alpha$-particles come from the decay of heavy nuclei. The artificial production of neutrons includes nuclear fission in sub and critical reactions, nuclear fusion, nuclear reactions produced in accelerators and in isotopic neutron sources. [3, 4]. Regardless the way of generation, neutrons have an energy distribution, also known as neutron spectrum ($\Phi(E)$), that is an important feature for neutron dosimetry.

Small size, compactness, portability, easy to handle and the no need of high voltage are the main advantages of isotopic neutron sources. On the other hand the small neutron strength, the emission of photons, and the lost of neutron strength due to decay, are few of its drawbacks. In these type of sources neutrons are produced by $(\gamma, n)$ or $(\alpha, n)$ exoenergetic nuclear reactions. In this group is also included those sources that produce neutrons through spontaneous fission of some heavy nuclei, like $^{252}\text{Cf}$. [4]

Few of the reported uses of these sources include neutron activation analysis, teaching, and calibration. [5-7] For calibration the International Organization for Standardization (ISO), recommends $^{241}\text{AmBe}$, $^{241}\text{AmB}$, unmoderated $^{252}\text{Cf}$, and $\text{D}_2\text{O}$-moderated $^{252}\text{Cf}$ sources. [8]

In order to use a neutron source for calibration, design safety protocols, and to explore potential applications require knowing several features such as the elemental concentration, strength of neutron

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yield, the spectrum, and the effect of laboratory conditions upon neutron spectrum and dosimetric quantities. This information is also required to establish the safety and radiological protocols.

In the $^{239}$PuBe source a mixture of the $\alpha$-emitter $^{239}$Pu with $^9$Be produce neutrons through the nuclear reaction $^9$Be($\alpha$, n)$^{12}$C*, where $^{12}$C* is left in excited state emitting 4.4 MeV photons whose dose contribution must be also known. In this type of sources impurities of $^{241}$Pu tend to increase the neutron yield because $^{241}$Pu decays to $^{241}$Am, that emits alpha particles that contributes to ($\alpha$, n) reactions in the mixture. Thus, the neutron source will tend to increase its neutron yield with time. The neutron yield of a $^{239}$PuBe source, with initial source strength of $7.94 \times 10^6$ s$^{-1}$, with different concentrations of $^{241}$Pu impurities is shown in Fig. 1. [9].

**Figure 1:** $^{239}$PuBe neutron yield in time due to $^{241}$Pu impurities.

The aim of this work is to determine the yield, fluence rate, neutron spectrum, and the ambient dose equivalent due to neutrons of a $^{239}$PuBe neutron source. Here, the count rates of a BSS were used to unfold the neutron spectrum using an Artificial Neural Network for spectrometry. This study also includes calculations with Monte Carlo methods and to compare them with the measured quantities.

2. Materials and Methods

The $^{239}$PuBe source was manufactured using 79.979 grams of $^{239}$Pu with a nominal activity of 185 GBq. The source consists in three 2.59 cm-diameter cylinders, two cylinders are 5.56 cm-length and one is 3.68 cm-length. Cylinders are kept together inside of an aluminium container. In the original data sheet of the source no information was given for impurities and source strength. This source is part of the subcritical reactor Nuclear Chicago model 9000.

2.1. Calculations

A detailed model of the neutron source was built, neutron transport calculations were carried out with the MCNP 4C and MCNP X 2.4.0 codes [10, 11] and the neutron spectra at 50, 100, 200, and 300 cm in air were calculated. In these calculations the source term was taken from the International Atomic
Using the spectra, $\Phi_E(E)$, obtained with MCNP 4C, the total fluence, $\phi$, was calculated using equation 1, and the ambient dose equivalent, $H^{*}(10)$, was determined using equation 2.

\[
\phi = \int_{E_{min}}^{E_{max}} \Phi_E(E) \, dE
\]

\[
H^{*}(10) = \int_{E_{min}}^{E_{max}} \Phi_E(E) \, h^{*}(10)(E) \, dE
\]

Here, $h^{*}(10)(E)$ are the fluence-to-dose conversion coefficients for $H^{*}(10)$, that were taken from NCRP 74. [13]

2.2. Measurements

Using a Bonner sphere spectrometer (BSS) with a 0.4 Ø × 0.4 cm $^6$LiI(Eu) scintillator, the neutron spectrum, at 100 cm from the source, was obtained. Polyethylene spheres utilized during measurements were 0, 5.08, 7.62, 12.7, 20.32, 25.40, and, 30.48 cm-diameter. From the BSS count rates the total fluence rate, $H$, and $H^{*}(10)$ were estimated using Artificial Neural Networks technology [14, 15].

The artificial neural network for spectrometry (ANNS) was trained using 181 known neutron spectra and their respective BSS count rates. This ANNS is able to output the neutron spectrum using as input the experimental count rates of the BSS without the need of having any clue about the features of the neutron field that is measured, therefore, no initial guess spectrum is required to do the unfolding, as mostly all available unfolding codes require. The features of this artificial neural network have been published elsewhere. [14, 15]

BSS measurements were carried out in an open space, with no walls, and at a height above ground of 200 cm in order to avoid the neutron room return [16, 17]. Spectrum, total fluence rate, and the dose equivalent obtained experimentally were compared with the Monte Carlo results obtained with MCNP 4C code.

In the aim to match those quantities a correction factor due to the presence of $^{241}$Pu was estimated.

3. Results and Discussion

The neutron lethargy spectra of $^{239}$PuBe, calculated at different distances in air with both Monte Carlo codes, are shown in Figs. 2, 3, 4, and 5. The Monte Carlo results are given for a source with strength of 1 s$^{-1}$. In these set of figures can be noticed that there are slightly differences between the spectrum obtained with MCNP 4C and MCNP X, those differences are attributed to cross sections libraries utilized by both codes. However, the differences are not relevant. For all distances total fluence behaves as $1/r^2$ rule meaning that the inclusion of air in the Monte Carlo calculations did no modifies the spectrum.

In Fig. 6 are shown, the MCNP 4C calculated and the BSS with ANNS measured, $^{239}$PuBe neutron spectrum at 100 cm, it can be noticed that both spectra looks alike. Neutron spectra used during the ANNS training have neutrons in the epithermal region, this could be the reason why measured spectra show the presence of few neutrons in the energy region.
Figure 2: $^{239}$PuBe neutron spectrum at 50 cm in air

Figure 3: $^{239}$PuBe neutron spectrum at 100 cm in air
Figure 4: $^{239}$PuBe neutron spectrum at 200 cm in air

Figure 5: $^{239}$PuBe neutron spectrum at 300 cm in air
Figure 6: Measured and MCNP 4C-calculated neutron spectrum of $^{239}$PuBe at 100 cm

From Monte Carlo calculations to 100 cm of distance the neutron source produces $\phi = 9.54 \times 10^{-6}$ cm$^{-2}$-s$^{-1}$ and $H^*(10) = 1.37 \times 10^{-5}$ µSv/h, these values correspond to a neutron source strength of 1 s$^{-1}$. The corresponding ambient dose equivalent-to-fluence conversion factor is 398.4 pSv-cm$^2$ for $H^*(10)$ that agrees in 5% to experimental conversion factor reported in the literature. [18]

Using the mass of $^{239}$Pu and its half life, 24110 ± 30 years [19], the original activity of $^{239}$Pu was 185 GBq, thirty nine years latter this activity has been reduced in 0.1% due decay. Using the specific yield factors for $^{239}$PuBe sources given by Hoste [20] and the NCRP [9], the mean neutron source strength is $7.94 \times 10^6 \pm 0.09\%$ s$^{-1}$. This neutron yield was utilized with the Monte Carlo results to obtain $\phi$ and $H^*(10)$ resulting in 76 ± 1 cm$^{-2}$-s$^{-1}$, and 108.9 ± 1.4% µSv/h respectively.

To 100 cm the $\phi$ and $H^*(10)$, obtained from the BSS and the ANNS, are $81 \pm 4$ cm$^{-2}$-s$^{-1}$ and $114 \pm 5\%$ µSv/h respectively. These values compared with those obtained with the Monte Carlo calculations are larger in 7% for $\phi$ and 5% for $H^*(10)$.

Measured values are larger than those obtained with MCNP 4C code, probable explanation of the differences is attributed to the increase of neutron yield due to the presence of $^{241}$Pu as impurities during source fabrication. The amount of this isotope was estimated in 0.102% mass fraction of $^{241}$Pu. This value increases the Monte Carlo quantities in 6.16%. Doing the corrections, for the presence of $^{241}$Pu in the original source mixture, in the Monte Carlo
calculated values results in $8.43 \times 10^6 \pm 0.09\% \ s^{-1}$ for the source strength, at 100 cm the total neutron fluence rate is $81 \pm 1 \ cm^{-2} . s^{-1}$, and $H*(10)$ is $115.6 \pm 1.4\% \ \mu Sv/h$.

The correction applied to Monte Carlo results compares better to $81 \pm 4 \ cm^{-2} . s^{-1}$ for total neutron fluence rate, and to $114 \pm 5\% \ \mu Sv/h$ for $H*(10)$ obtained with the BSS with ANNS. By comparing these values the differences between the quantities corrected by $^{241}Pu$ concentration and those measured with the BSS with ANNS are 0 for $\phi$ and 1\% for $H*(10)$.

4. Conclusions

To handle a neutron source in terms of radiological protection, and to explore its possible applications quantities like the source strength, spectrum, fluence rate, and ambient dose equivalent are required. Therefore, these quantities must be known to characterize a neutron source. Those quantities have been determined for a $^{239}PuBe$ neutron source by means of Monte Carlo calculations and measurements performed using a BSS with ANNS.

The presence of impurities of $\alpha$-emitter isotopes in the mixture of ($\alpha, n$) sources produces, in the first 70 years after source-fabrication, an increase in the neutron yield, with the consequent increase in the $H*(10)$ and the need of a better shielding. In the case of a $^{239}PuBe$ source the presence of small amounts of $^{241}Pu$ isotope decaying, with a half-life of 13 years, by $\beta$-emission ends in $^{241}Am$ that, with a half-life of 432 years, decays emitting $\alpha$ particles, producing a build up of $^{241}Am$ ending in the increase of neutron output of $^{239}PuBe$ source.

By Monte Carlo calculations, where a detail model of the neutron source was utilized, the neutron spectrum, fluence rate, and $H*(10)$ were determined. From these calculations the resulting ambient dose equivalent-to-fluence conversion factors is $398 \ pSv-cm^2$ for $H*(10)$.

The $^{239}PuBe$ neutron spectra at 50, 100, 200, and 300 cm from the source in air were calculated using MCNP X and MCNP 4C. Small differences were found between the spectra that are attributed to the cross section libraries utilized for both codes. However, these differences are not significant.

At 100 cm from the source a BSS was located and the count rates measured for each sphere were collected and input in an Artificial Neural Network designed to unfold the neutron spectrum. By comparing this with the MCNP 4C calculated spectrum both look alike, however the presence of neutrons in the epithermal region show us the need to design a different ANNS where training data include spectra similar to isotopic neutron sources.

Resulting spectrum was utilized to estimate the total neutron fluence rate and the $H*(10)$. At first these quantities were different for those obtained through Monte Carlo calculations. The differences were attributed to the presence of $^{241}Pu$ during source manufacturing.

Corrections due to the $^{241}Pu$ presence were carried out and corrected values for source strength, fluence rate, and $H*(10)$ were estimated. These quantities compares well with those obtained with the BSS. Thus the neutron yield is $8.43 \times 10^6 \ s^{-1}$. To 100 cm the total neutron fluence rate is $81 \ cm^{-2}.s^{-1}$, and $H*(10)$ is $115.6 \ \mu Sv/h$ that compare well with $81 \ cm^{-2}.s^{-1}$, and $114 \ \mu Sv/h$ obtained using BSS with ANNS.
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