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ON MODELING A PLUTONIUM-BERYLLIUM NEUTRON SOURCE

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Abstract. In the present work, the neutron yield of a Pu-Be neutron source has been calculated by using two methods, calculation with the SOURCES4C code developed at LANL and via an equation, respectively. The present work aims to determine and compare the two methods in obtaining the yields and spectra of an (α, n) type neutron source. The source used for this work is being held at IFIN-HH, and was well experimentally characterized in previous work. The results of the present work compare well with the experimental data.

Key words: Sources4C, Pu-Be neutron source, neutron spectra.

1. INTRODUCTION

Neutron sources have been in use for quite some time, and the need of accurate accounts for nuclear material, especially plutonium and uranium, is necessary for IAEA safety and safeguards. Following recent events, there have been modifications to safeguarding laws, and precise measurements of Americium and Plutonium present in old neutron sources scattered all over the world are required. The isotopic content is an important information for further certification and accountancy of nuclear materials. One such source, a Pu-Be source, is present at National Institute for R&D in Physics and Nuclear Engineering (IFIN-HH) and has been characterized experimentally by P.-A. Söderström et.al. [1], both by γ-tagged and in full energy spectra. The reaction principle on which this type of source is based is:

$$^{239}_{94}\mathrm{Pu} + ^{9}_{4}\mathrm{Be} \rightarrow ^{235}_{92}\mathrm{U} + ^{4}_{2}\alpha + ^{9}_{4}\mathrm{Be} \rightarrow ^{235}_{92}\mathrm{U} + ^{12}_{6}\mathrm{C} + ^{1}_{0}n$$

The reaction will produce a clean neutron spectrum of multiplicity one, partially in coincidence with 4.43 MeV γ-ray from excited $^{12}_{6}\mathrm{C}$. In the article by P.-A. Söderström et.al. they obtained gamma-ray spectra by using high-purity germanium (HPGe) detector with 60% relative efficiency, placed at 30 cm from the detector. For the emitted neutrons from the source, they [1] used ELIGANT-TN setup, which consists of 28 ^{3}He detectors counters with a 12-bar pressure arranged onto three rings, and all detectors are placed in a polyethylene matrix. They obtained a mean energy of 3.25(17) MeV and a source activity of $2.220(5)\cdot10^{5}$ neutrons per second. The work of Bagi et. al. [3] is a good starting point for investigating such sources as the authors provide much information with regard to this type of sources; they also used other sources of similar contents [4].

2. THEORY

The Sources4C code was developed at Los Alamos National Laboratory by W.B. Wilson et.al. [2]. It is capable of calculating neutron sources homogenous, interface, monoenergetic α-beam, and three-region interface (TRI) type problems. Homogeneous sources consist of α-emitting and low-z material mixtures, and typical examples include "PuBe" neutron sources and actinide solutions. interface problems imply a composite material consisting of two separate slab regions (source and target), such as a plated actinide or storage device. Similarly, the three-region interface problems assume a thin slab of low-z target material sandwiched between

α-emitting source material and low-z target material. A beam problem calculates the neutron source from a monoenergetic α-beam incident on a low-Z slab. Systems that include combinations of these problems, however, must be run separately and subsequently compiled by the user. The slowing and stopping of α-particles in a material are described by the material's stopping power [2],

$$SP(E) = \frac{-\mathrm{d}E}{\mathrm{d}x} \tag{1}$$

which yields an α particle's energy loss per unit path length x. The energy loss of an α particle of initial energy E_α in travelling a distance L can be determined from the stopping power [2]

$$\Delta E = E_a - E'_a = \int_0^L \left(\frac{-\mathrm{d}E}{\mathrm{d}x} \right) \mathrm{d}x \tag{2}$$

Similarly, the distance traveled in slowing from E_α to E'_α is

$$L = \int_{E_\alpha}^{E'_\alpha} \frac{1}{\left(\frac{\mathrm{d}E}{\mathrm{d}x} \right)} \mathrm{d}E = \int_{E'_\alpha}^{E_\alpha} \frac{1}{\left(\frac{-\mathrm{d}E}{\mathrm{d}x} \right)} \mathrm{d}E \tag{3}$$

During the slowing down of the α particles within the material, neutrons may be produced by (α, n) reactions with the nuclides in the material. The probability of an (α, n) interaction with nuclide i by an α particle of energy E traveling from x to $x+\mathrm{d}x$ is [2]:

$$N_i \sigma_i(E) \mathrm{d}x = \frac{N_i \sigma_i(E) \mathrm{d}E}{\left(\frac{\mathrm{d}E}{\mathrm{d}x} \right)} \tag{4}$$

where N_i is the atom density of nuclide I and σ_i is the microscopic (α, n) cross section for nuclide i. The probability of (α, n) interaction with nuclide i by an α-particle that slowed from E_α to E'_a is then[2]:

$$P_i\left(E_\alpha \to E'_\alpha\right) = \int_{E_\alpha}^{E'_\alpha} \frac{N_i \sigma_i(E)}{\left(\frac{\mathrm{d}E}{\mathrm{d}x} \right)} \mathrm{d}E = \int_{E'_\alpha}^{E_\alpha} \frac{N_i \sigma_i(E)}{\left(\frac{-\mathrm{d}E}{\mathrm{d}x} \right)} \mathrm{d}E \tag{5}$$

Thus, the probability of an α-particle undergoing an (α, n) reaction with nuclide I before stopping in the material is given by the thick-target neutron production function [2]

$$P_i\left(E_\alpha\right) = \int_0^{E_\alpha} \frac{N_i \sigma_i(E)}{\left(\frac{-\mathrm{d}E}{\mathrm{d}x} \right)} \mathrm{d}E \tag{6}$$

The stopping cross section (ε) is defined as,

$$\varepsilon(E) = \frac{-1}{N} \frac{\mathrm{d}E}{\mathrm{d}x} \tag{7}$$

where N is the total atom density of the material. The quantities p_i and P_i can now be expressed in terms of the stopping cross section [2]:

$$p_i\left(E_\alpha \to E'_\alpha\right) = \frac{N_i}{N} \int_{E'_\alpha}^{E_\alpha} \frac{\sigma_i(E)}{\varepsilon(E)} \mathrm{d}E \tag{8}$$

and

$$P_i\left(E_\alpha\right) = \frac{N_i}{N} \int_0^{E_\alpha} \frac{\sigma_i(E)}{\varepsilon(E)} \mathrm{d}E \tag{9}$$

Generally, any material involved in a homogeneous problem will be composed of any number of different elements (e.g., Li, Be, or Pu). The stopping cross section $\varepsilon(E)$ of a material composed of J elemental constituents may be calculated using the Bragg-Kleeman relationship [2, 6]:

$$\varepsilon(E) \cong \frac{1}{N}\sum_{j=1}^{J} N_j \varepsilon_j(E) \tag{10}$$

where

$$N = \sum_{j=1}^{J} N_j \tag{11}$$

A fraction of the decay of nuclide k within a material may be via α particle emission. This fraction (F_k^{α}) of alpha decay may occur with the emission of one of L possible α particle energies. The intensity (F_{kl}^{α}) is the fraction of all decays of nuclide k resulting in an α particle of energy E_l; and thus [2]

$$F_k^{\alpha} = \sum_{l=1}^{L} f_{kl}^{\alpha} \tag{12}$$

Therefore, the fraction of nuclide k decays resulting in an (α, n) reaction in a thick-target material containing I nuclides with non-negligible (α, n) cross sections is [2]:

$$R_k(\alpha, n) = \sum_{l=1}^{L} f_{kl}^{\alpha} \sum_{i=1}^{I} P_i(E_l) \tag{13}$$

The value for $P_i(E_l)$ will be determined using the discrete form of Eq. (9),

$$P_i(E_l) = \frac{N_i}{N}\sum_{g=1}^{G-1}\frac{1}{2}\left[\frac{\sigma_i^{g+1}}{\varepsilon^{g+1}} + \frac{\sigma_i^{g}}{\varepsilon^{g}}\right]\left(E^{g+1} - E^{g}\right) \tag{14}$$

where g, $g+1$, $g-1$, are the discrete energy groups per alpha decays $\sigma_i^1 = \sigma_i(0)$, $\sigma_i^G = \sigma_i(E_l)$, $\varepsilon^1 = \varepsilon(0)$, and $\varepsilon^G = \varepsilon(E_l)$ (i.e., the energy range has been discretized into $G-1$ energy groups). It is important to note that calculation of the (α, n) neutron source per decay of nuclide k requires accurate knowledge of the discrete energy (α, n) cross section for each target nuclide (σ_i^g), discrete energy stopping cross section (ε^g) for all elemental constituents, atom fraction (N_i/N) for each target nuclide, the intensity for emission of each of L α-particles (f_{kl}^{α}), and the energy of each of the L α-particles (E_l) [2]. All of these equations are taken into account when the calculations are performed by the code SOURCES4C.

The user is advised to investigate the source(s) prior to simulations, and an appropriate isotopic content would make the results closer to actual experimental results. In the case of Plutonium source, the short half-life ^{238}Pu isotope imposes decay corrections, and the activity affects the source yield.

The theoretical yield of 3.8×10^5 n/cm^2/s has been obtained with equation (15), from the article of Runnalls et al. This equation is based on the Baerg suggestions, which were not published at the time of Runnalls et al. article:

$$\frac{n}{n_{max}} = N \cdot S_{\mathrm{Be}} \Big/ \left(N \cdot S_{\mathrm{Be}} + S_{\mathrm{Pu}}\right) \tag{15}$$

where $n/n_{\max}$ = fraction of maximum theoretical neutron yield, N = Be:Pu atom ratio, S_{Be} = Be linear atomic stopping power = 0.63, S_{Pu} = Pu linear atomic stopping power = 4.60, the values for linear stopping power were given in [5].

3. RESULTS AND DISCUSSIONS

In the present work, we simulated with the aid of SOURCES4C code both neutron yields and spectra and, theoretically, with the help of a equation, the neutron yield of the Pu-Be neutron source. The neutron source used for this work is the same with that from the work of P.-A. Söderström et al. [1]. This work presents a comparison with their experimental neutron yield and spectra.

In the present work, the computer code SOURCES-4C was used to obtain (α, n) neutron spectra of the specified source. Figure 1 presents the obtained spectra coming from (α, n) reactions, Fig. 2 presents the spectra coming from Spontaneous Fission of the Plutonium isotopes, while Fig. 3 presents the total neutron spectrum of the source. SOURCES-4C code can calculate separately the yields and spectra from (α, n) reaction and spontaneous fission fractions.

Table 1 presents the important data about the isotopes present in the source being modelled, according to data from literature and Ref. [1]. There are three isotopes of Plutonium present in the source and which have been taken into account.

Table 1

Isotopic content of the source at the specified date

% Composition (20/11/2019) 12:00:00 AM					
Nr.Crt.	**Isotope Content**	**Source %**	**Nr.Crt.**	**Isotope Content**	**Source %**
1	**Pu-237**	**0.00**	**5**	**Pu-241**	**0.00**
2	**Pu-238**	**1.00**	**6**	**Pu-242**	**0.00**
3	**Pu-239**	**75.00**	**7**	**Am-241**	**0.00**
4	**Pu-240**	**24.00**			
	Total	**100.00**			

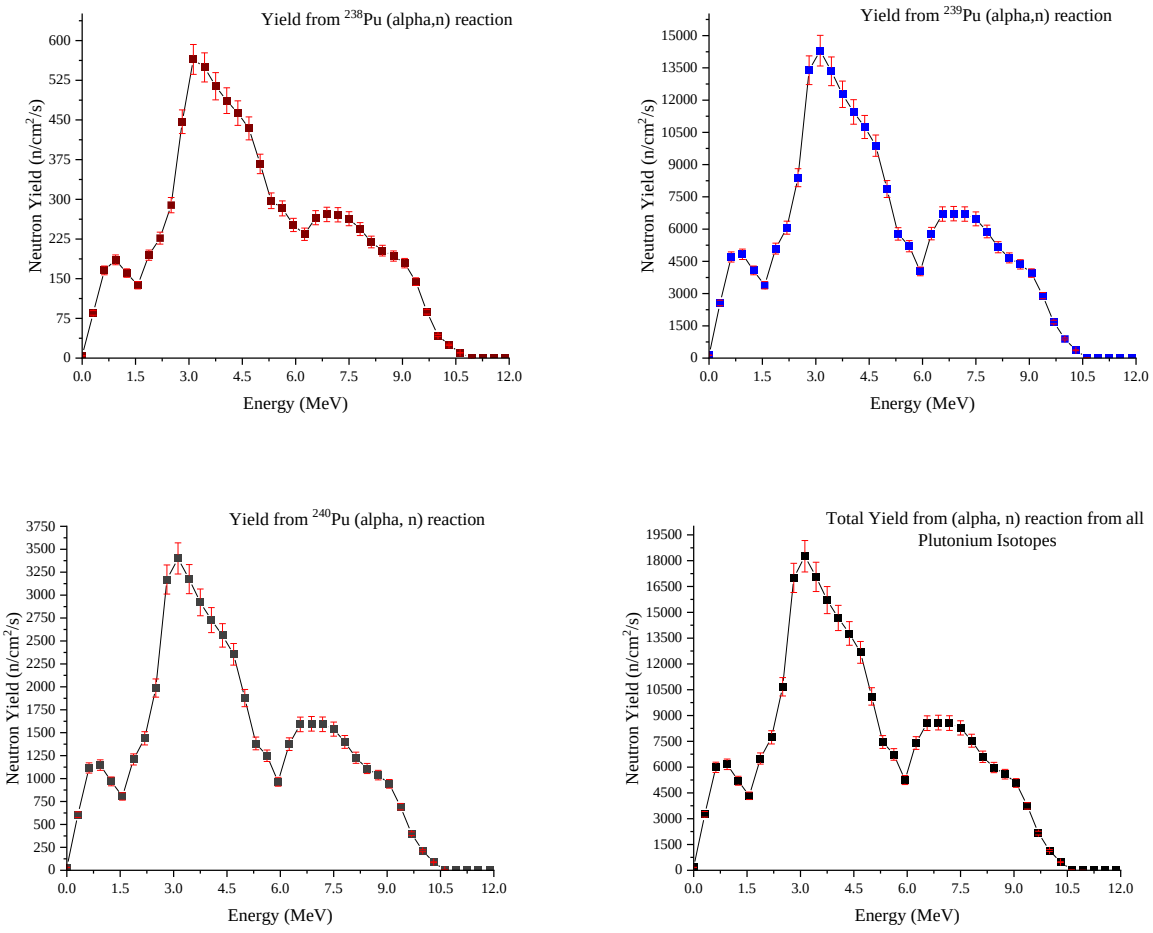

Fig. 1 – Neutron yields originating from (α,n) reaction, calculated using SOURCES4C code for each isotope separately.

For the comparison with experimental data, error bars of ± 5% are thought to be a realistic basis. Figure 3 presents a comparison between the experimental data of [1] and the results obtained from SOURCES4C code. Errors for the experimental points of ±5% are also represented. In the case of equation used in determining the theoretical neutron yield of the source we only made a single calculation.

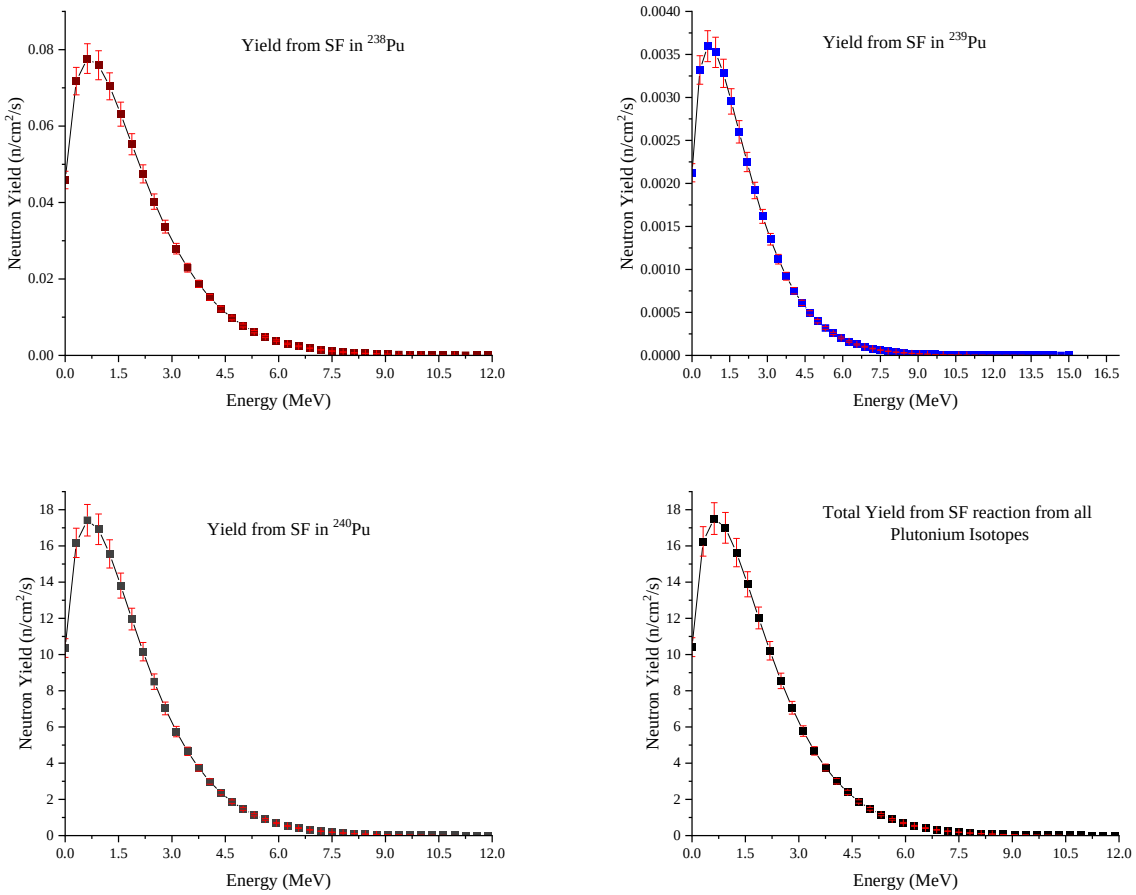

Fig. 2 – Neutron yields originating from Spontaneous Fission calculated using SOURCES4C code for each isotope separately.

The results from Fig. 3 are plotted from data obtained with the SOURCES4C code and experimentally in the work of P.-A. Söderström et.al. [1], the code has been implemented with the capability to breakdown the yields and neutron spectra, in present case three Pu isotopes present in the sample.

For our source, we have obtained the following:

- from (α, n) reaction fraction the yield of 2.684×10^5 n/cm^2/s,
- from spontaneous fission fraction yield 1.56×10^2 n/cm^2/s.

The total yield calculated via SOURCES4C is 2.686×10^5 n/cm^2/s from both spontaneous reaction and (α, n) reaction.

The result from the eq. (15), which was presented earlier, gives only the total yield of a source based on specific information we have about the materials of the source; in our case, we need the stopping power in Plutonium and Beryllium, which were taken from [5] and used on our source and the total yield is 3.812×10^5 n/cm^2/s.

This yield is higher than both the experimental and the SOURCES4C predicted yields.

The reason may be that shielding factors were not considered in this approach, since we do not know what is inside the active material and how homogenous it is.

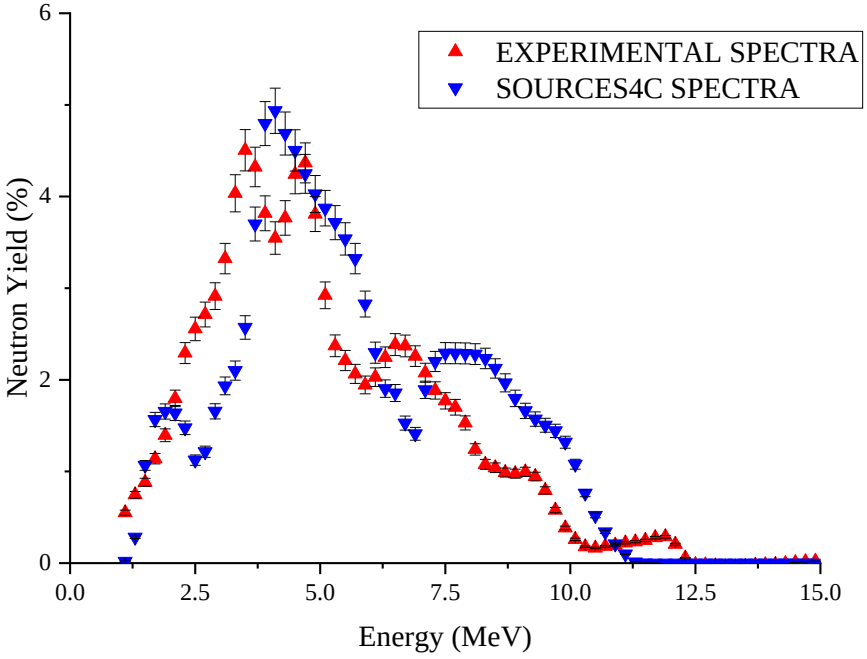

Fig. 3 – Comparison between the total neutron spectrum from experiment [1] (up triangles) and that of the present calculations with the SOURCES4C code (down triangles).

4. CONCLUSION

The software SOURCES4C and the equation used to determine the neutron yield of the source are in reasonably good agreement with the available data from the manufacturer of the source of 2.27×10^5 n/cm^2/s, and the result of 2.22×10^5 n/cm^2/s from Reference [1].

Our result calculated via the equation [15] is 3.812×10^5 n/cm^2/s, one reason for this discrepancy could be the fact that the equation used determines the ideal yield of the source without taking into account self-shielding factors, which may differ from the measurements conditions.

The SOURCES4C result approximates better the total yield of the source of 2.68×10^5 n/cm^2/s, the final result is a sum of all reactions (α, *n*; spontaneous fission process) and isotopes present in the source (Pu-238, Pu-239, Pu-240), the actual result could be improved by obtaining a source with a more refined isotopic concentration. However, in the present case, this is impossible because the source certification from the manufacturer does not contain very precise information about the isotopic content, in the work of P.-A. Söderström et al. they determine the concentration based on Makarova et al. tables. The neutron spectra from the code and obtained experimentally look rather similar. In conclusion, the calculation via SOURCES4C gives comparable results with the ones obtained in a real measurement of the source itself.

As final conclusion both equation [15] and SOURCES4C can be used to determine the age of a source/sample sources which are not certified or whose certification has been lost.

This approach may be recommended to characterize this type of neutron sources.

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