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Characterization of a neutron irradiation facility using Monte Carlo simulations and activation dosimetry

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Abstract In the present work, the fluence rate at the sample irradiation positions for a neutron irradiation system based on two $^{239}\text{Pu}(\alpha,n)^9\text{Be}$ radionuclide neutron sources of nominal total activity of 444 GBq was derived. The measurements were performed using a set of activation dosimetry foils comprising gold, indium, iron, zinc, cobalt and aluminum and a high purity germanium based gamma spectrometry system. The measurements were supported by Monte Carlo simulations performed using the MCNP6 code. The results of the study showed that the neutron emission rate of the sources was $(3.41 \pm 0.53) \times 10^7$ n/s and the neutron fluence rate at the central sample position was $(5.70 \pm 0.80) \times 10^5$ cm²s⁻¹. Although radionuclide neutron sources cannot achieve the high fluence rates encountered in nuclear research reactor and accelerator facilities, they provide a simple and cost effective method for fast neutron irradiation of samples under constant neutron fluence rate conditions. The system will be used for neutron dosimetry and material irradiation studies.

Keywords neutron source, activation dosimetry, Monte Carlo simulations

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INTRODUCTION

Radionuclide neutron sources provide a simple and cost effective method for fast neutron irradiation under constant neutron fluence rate [1]. A neutron irradiation facility was developed by modifying an existing Prompt Gamma Neutron Activation Analysis (PGNAA) system for bulk samples analysis [2]. The modifications included utilization of two $^{239}\text{Pu}(\alpha,n)^9\text{Be}$ radionuclide sources of nominal total activity of 444 GBq and installation of a holder enabling bilateral irradiation of the samples (Fig. 1). Although the sources nominal activity was specified, the actual neutron emission rate was not known, since it depends on several factors, such as the source capsule size and the packing density of Pu and Be powders [3,4]

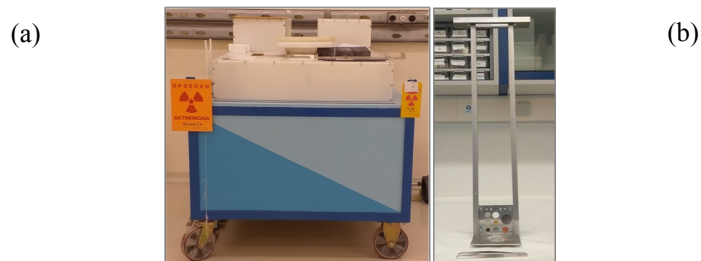


Figure 1. (a) Irradiation system, (b) Sample holder with activation foils.

Scope of the present work was the evaluation of the source emission rate and the neutron fluence rate at the sample irradiation positions, using activation dosimetry foils and Monte Carlo simulations. The applied neutron dosimetry methodology was taken from ref. [5].

EXPERIMENTAL

The set of activation foils used, the nuclear reactions employed and their threshold energies, the half-lives of the product isotopes, as well as the energy and intensity of the emitted gamma rays are shown in Table 1.

Table 1. Foils used and characteristics of the nuclear reactions

Foil	Reaction	Threshold (MeV)	T _{1/2}	Energy (keV)	Intensity (%)
Gold	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	-	2.695 d	411.8	95.62
Indium	$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	0.5	4.5 h	336.2	45.90
Iron	$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	4.9	2.5789 h	846.7	98.85
Zinc	$^{64}\text{Zn}(n,p)^{64}\text{Cu}$	2.0	12.701 h	1345.7	0.48
Cobalt	$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$	5.2	2.5789 h	846.7	98.85
Aluminum	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	4.9	14.997 h	1368.6	99.99
	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	3.8	9.458 min	843.7	71.80

Irradiations were performed in two experimental cycles of 7 d and 13 d, respectively. The gamma-ray measurements were performed using a HPGe detector of 85% relative efficiency. The Full Energy Peak Efficiency (ε) of the germanium detector used was experimentally determined and it is presented in Fig. 2. The experimental data were fitted with the empirical relationship of Eq. 1.

$$\varepsilon = \frac{1}{E} (a + b \cdot \ln(E)^{-1} + c \cdot \ln(E)^{-2} + d \cdot \ln(E)^{-3} + e \cdot \ln(E)^{-4} + g \cdot \ln(E)^{-5} + h \cdot \ln(E)^{-6}) \quad (1)$$

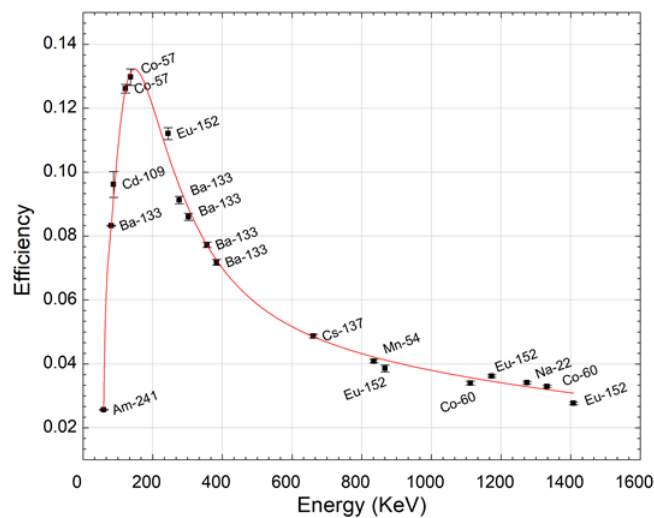


Figure 2. HPGe detector Full Energy Peak Efficiency as a function of photon energy. The line represents empirical fit to the experimental data using eq. 1 ($r^2=0.998$).

The saturation activity in each foil was determined using Eq. 2, based on the net counts (C) at the photopeak, decay constant (λ), detector Full Energy Peak Efficiency (ε_γ), gamma intensity (f_γ), irradiation (t_{irr}), decay (t_d) and counting (t_c) time. Corrections for self-absorption of gamma-rays (G_γ) and true coincidence summing (f_{tcc}) were taken into account.

$$A_{\text{sat,exp}} = \frac{C}{\varepsilon_\gamma f_\gamma G_\gamma f_{\text{tcc}}} \frac{\lambda}{(1 - e^{-\lambda t_{\text{irr}}}) e^{-\lambda t_d} (1 - e^{-\lambda t_c})} \quad (2)$$

SIMULATIONS

A detailed model of the irradiation system (Fig. 3) was produced using the Monte Carlo code MCNP6.1 [6]. ENDF71x and IRDFF v.1.05 cross section data libraries were used for neutron transport and nuclide activation calculations, respectively. The neutron energy spectrum of the source was taken from ref. [2]. Calculations were performed using the MCNP F4 tally providing the average neutron fluence ($\text{cm}^{-2} \cdot \text{n}^{-1}$) and the FM multiplier tally to get the number of produced nuclei per unit volume from corresponding reactions in each foil ($\text{cm}^{-3} \cdot \text{n}^{-1}$). The results of the calculations provided the foil saturation activity per source neutron, as shown in Eq. 3 where N_t is the number of target nuclei, $\sigma(E)$ the cross section, λ the decay constant and $\phi(E)$ the neutron fluence.

$$A_{\text{sat,calc}} = \lambda N_t \int_E \sigma(E) \phi(E) dE \quad (3)$$

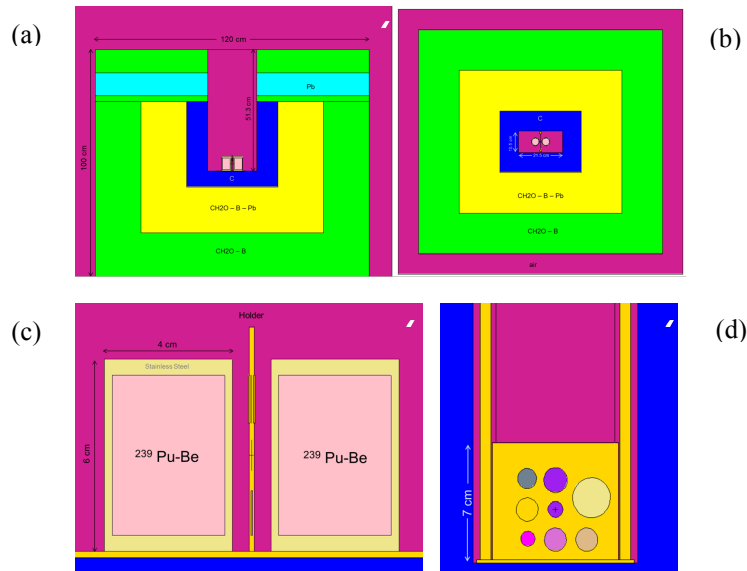


Figure 3. Model of the irradiation facility: (a) vertical view, (b) horizontal view, (c) holder positioned between the sources, and (d) holder and samples.

RESULTS AND DISCUSSION

The source neutron emission rate was evaluated as the ratio of the experimentally over the calculated activity on each foil using Eq. 4.

$$\text{Source emission rate} = \frac{A_{\text{sat,exp}}}{A_{\text{sat,calc}}} (\text{s}^{-1}) \quad (4)$$

The derived source emission rate was $(3.41 \pm 0.53) \times 10^7 \text{ n} \cdot \text{s}^{-1}$. It is noted that the evaluated source emission rate is $\sim 17\%$ higher than the value of $(2.9 \pm 0.3) \times 10^7 \text{ n} \cdot \text{s}^{-1}$ predicted on the basis of neutron yield of 65 ± 6 per 10^6 α -particles emitted from ^{239}Pu [3]. The total and above 0.5 MeV neutron fluence rate at the sample positions are shown in Fig. 4.

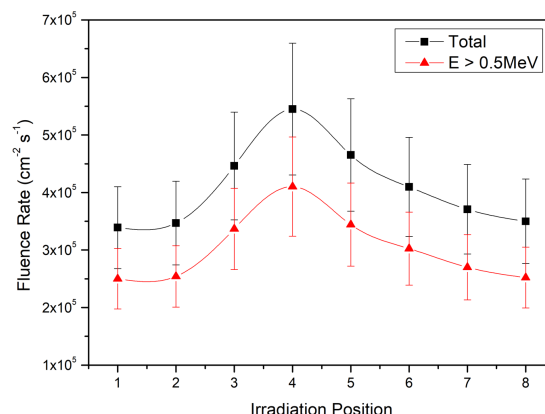


Figure 4. Neutron fluence rate at the irradiation positions (Note: eye catching lines)

The neutron spectra and yields of (α , n) type radionuclide neutron sources are known to be altered by secondary neutron interactions in the source and capsule materials as well as by the source preparation method. Therefore, insufficient knowledge of the ^{239}Pu –Be sources characteristics (powder mixture composition, packing density and exact capsule dimensions) affects the neutron spectrum of the emitted neutrons [3]. Such inaccuracies are of particular importance for neutron energies below 1 MeV, where no accurate neutron spectroscopy experimental data are usually available. Moreover, experimental methods, such as the Manganese bath technique, which may provide an accurate determination of the source neutron yield, require specialized equipment and are generally difficult to implement.

In this work, the activation foil technique was applied to determine the neutron yield of the source and the fluence rate at the sample irradiation positions of a neutron irradiation system based on Pu–Be sources. Future work will be directed toward reconstruction of the spectrum on the basis of the different energy responses of the activation foils. The irradiation system will be used for neutron dosimetry and material irradiation studies.

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