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N. Kotb
Radiation Protection Department, Hot Laboratories Centre, Atomic Energy Authority, Egypt, n.kotb87@yahoo.com

M. Tohamy
Experimental Nuclear Physics Department, Nuclear Research Centre, Atomic Energy Authority, Egypt

A. Solieman
Experimental Nuclear Physics Department, Nuclear Research Centre, Atomic Energy Authority, Egypt

T. El - Zakla
Radiation Protection Department, Hot Laboratories Centre, Atomic Energy Authority, Egypt

T. Amer
Physics Department, Faculty of Science, Al - Azhar University, Girls Branch, Cairo, Egypt

See next page for additional authors

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CHARACTERIZATION OF ^{241}AM-BE NEUTRON SOURCE USING THRESHOLD FOIL ACTIVATION TECHNIQUE

N. A. Kotb[a*], M.M. Tohamy[b], A. H. M. Solieman[b], T. El-Zakla[a], T. Z. Amer[c], S. Elmeniawi[c] and M. N. H. Comsan[b]

[a] *Radiation Protection Department, Hot Laboratories Centre, Atomic Energy Authority, Egypt*
[b] *Experimental Nuclear Physics Department, Nuclear Research Centre, Atomic Energy Authority, Egypt*
[c] *Physics Department, Faculty of Science, Al-Azhar University, Girls Branch, Cairo, Egypt*
[*]Corresponding author: n.kotb87@yahoo.com

ABSTRACT

The aim of the study is to characterize ^{241}Am-Be isotopic neutron source with 5 Ci activity and nominal neutron yield 1.1×10^7 n/s. The emitted neutrons were measured employing threshold multi-foil activation technique using the following reactions: ^{115}In(n,n'), ^{58}Ni(n,p), ^{64}Zn(n,p), ^{27}Al(n,p), ^{56}Fe(n,p), ^{59}Co(n,α), ^{24}Mg(n,p), and^{27}Al(n,α). The threshold reactions were selected based on their half-lives, adequate threshold energy and cross-section. The reaction rate of each sample foil is measured by detecting the gamma ray spectra produced as a result of reaction products decay. A hyper pure germanium detector (HPGe) is used for gamma-ray spectrum measurements. The obtained spectra were analyzed using Genie2000 software and the associated system absolute efficiency was calculated using Efftran software. Obtained reaction rates were compared with available published data.

Keywords: ^{241}Am-Be neutrons, HPGe, threshold reactions, activation technique, reaction rate.

1. INTRODUCTION

Isotopic Neutron Sources (INSs) such as ^{241}Am-Be, ^{226}Ra-Be, and ^{239}Pu-Be are widely used in a variety of applications such as neutron activation analysis [1-4]. Each INS has a specific strength and spectrum. The need for the characterization of INSs is essential and plays an important role in the shielding design and determination of their potential uses. Different techniques are used for neutron spectrum characterization such as Bonner Sphere Spectrometer [5-8], and foil activation technique [9-16]. There are several advantages of using foil activation technique in neutron field characterization, measurements and mapping due to their small size, insensitivity to gamma radiation, low cost and their ability to tolerate different conditions of measurements [17].

The threshold reactions such as (n, p), (n, α), (n, 2n) and (n, n′) can be used to determine the neutron flux density and spectrum in the fast energy region using different unfolding methods [18,19]. The present work aims to study the use of threshold activation foils to characterize ^{241}Am-Be isotopic neutron source. Different materials are used in our experiment as threshold detectors. The criteria for the selection of materials used as detectors depend on the following: (1) suitable half-life of the reaction product, (2) type of reaction product and possibility of detecting the emitted radiation, (3) high reaction cross-section and appropriate effective threshold energy aiming to cover the entire range of source neutron energies, and (4) availability of the materials with low cost and high degree of purity.

2. MATERIALS AND METHODS:

2.1. Description of ^{241}Am-Be irradiating system:

The irradiating system consists of three components; ^{241}Am-Be source, shielding house and source handling system. The ^{241}Am-Be neutron source has activity 5 Ci (185 GBq) and nominal neutron emission rate $2.2\text{x}10^6$ n/Ci. The source has a cylindrical shape with external dimensions of 22 cm long and 4.4cm diameter. The active volume at the center of cylindrical source contains AmO_2-Be powder that is doubly encapsulated inside welded stainless-steel layers.

The neutron house is a wooden cubic box of dimensions $60\times60\times60$ cm^3. A layer of 6 cm paraffin wax mixed with 20% boric acid (H_3BO_3) was placed in the outer sides (faces) of

the cube, while the core is filled with paraffin wax mixed with 5% boric acid. At the center of the cube, a horizontal cylindrical tube with inner diameter 4.8 cm was built for neutron source to easily move inside it. The box outer layer composed of 2 cm lead surrounds the cubic structure to attenuate the gamma radiation.

Safe handling of the source was achieved using special apparatus insuring taking it out easily of its housing system for irradiation and inserting it back to the center of the housing for storage. During irradiation, the source was taken out of the housing to a distance of 50cm and fixed on a wooden table with dimensions of 50cm wide and 120cm length. The experimental arrangement of the irradiating system including the neutron irradiator, neutron house as well as the neutron source handling system is presented in figure 1 [20].

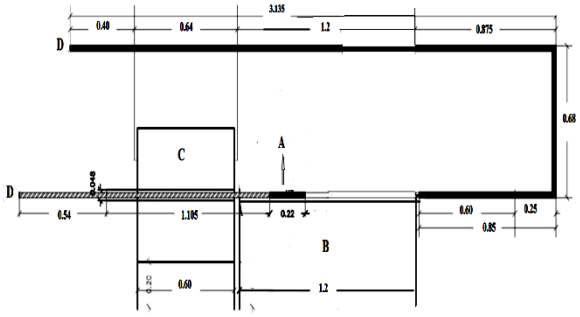

Fig. 1: The components of the irradiating system. A is the ^{241}Am-Be neutron irradiator, B is the wooden table, C is the neutron house, and D is the source handling system. All dimensions are in (m).

2.2. Threshold detectors:

Seven different foils were used in current study with different cross-sections and gamma-ray energy emission. They were exposed to a flux of neutrons for a period of time depending on the half life of the product nucleus. The irradiation times of In(n,n') reaction was 22.5h, Ni(n,p) reaction was 164h, Zn(n,p) reaction was 69h, Al(n,p) reaction was 1h, Fe(n,p) reaction was 5h, Co(n,α) reaction was 23h, Mg(n,p) reaction was 43h and Al(n,α) reaction was 45h. The foils were placed during irradiation in contact with the source at its center. The nuclear data of the used threshold activation reactions are summarized in Table 1. Figure 2 presents the cross-sections as a function of neutron energy of the used threshold reactions [21].

Table 1. Nuclear data of the used threshold reactions.

Activation reaction	Radioactive product	Half-life	Gamma energy (keV)	Gamma probability (%)
^{115}In(n, n')	^{115m}In	4.49 h	336.24	45.80
^{58}Ni(n, p)	^{58}Co	70.86 d	511.00 810.78 863.95 1674.73	29.50 99.00 7.00 5.00
^{64}Zn(n, p)	^{64}Cu	12.70 h	511.01 1345.84	34.50 0.50
^{27}Al(n, p)	^{27}Mg	9.46 m	843.74 1014.42	71.80 28.00
^{56}Fe(n, p)	^{56}Mn	2.58 h	846.77 1810.77 2113.12	98.90 27.20 14.30
^{59}Co(n, α)	^{56}Mn	2.58 h	846.77 1810.77 2113.12	98.90 27.20 14.30
^{24}Mg(n, p)	^{24}Na	14.96 h	1368.63 2754.03	100.00 99.90
^{27}Al(n, α)	^{24}Na	14.96 h	1368.63 2754.03	100.00 99.90

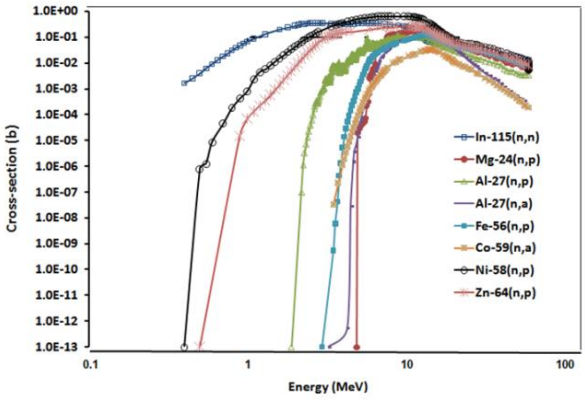

Fig.2: Cross sections as function of neutron energy for the threshold detectors [21].

2.3. High Resolution Gamma Spectroscopy System:

The measurements of foils activity are obtained from the gamma-ray emission that results from the decay of the target foil. The emitted gamma-rays were measured using high resolution gamma-ray spectrometry system equipped with germanium (HPGe of 70% relative efficiency) detector manufactured by ORTEC, adjustable High-Voltage Power Supply (ORTEC; model 459, 0-5kV) and amplifier (TENNELEC; model TC 240). The measuring

system was connected to a personal computer-based data acquisition system, which has a digital-Multi-Channel-Analyzer (CANBERRA; model Multi Port II, 8096 channel).

The calibration of the detector was performed before the analysis using a set of standard sources (^{60}Co, ^{137}Cs, ^{22}Na, ^{133}Ba, and ^{152,154,155}Eu). The analysis was made using the software Genie-2000.

Efftran software [22] was used to determine the efficiencies of the system considering dimensions of target foils, detector and measurement geometries. It implements the efficiency transfer method using Monte Carlo technique to transfer point-source efficiency into volumetric sample efficiency. The specification of the detector was defined as specified by the manufacturer.

3. RESULTS AND DISCUSSION:

The characterization of the neutron field was carried out using seven different activation foils as threshold detectors. The reaction rates were obtained from the gamma-ray emission from the decay of the reaction products.

The reaction rate (RR) induced in a threshold detector irradiated by the neutron source during time t_i and measured during time t_m after a cooling time t_c was derived from the following expression [1]:

$$RR\ (s^{-1}) = \frac{M}{m\,N_A}\frac{\lambda C}{(\varepsilon P_\gamma \theta(1-e^{-\lambda t_i})e^{-\lambda t_c}(1-e^{-\lambda t_m}))} \quad [3]$$

Where M is the atomic weight of sample (amu), λ is the radioactive decay constant (s^{-1}), C is the gamma-ray photo-peak counts, m is the mass of the sample (g), N_A is the Avogadro number, ε is the absolute efficiency, P_γ is the gamma-ray decay probability, θ is the isotopic abundance of the irradiated material.

Flux corresponding to different threshold reactions was estimated by dividing the measured reaction rate by the energy-averaged cross-sections. The energy-averaged cross-section ⟨σ⟩ was derived using the ^{241}Am-Be ISO 8529-1 spectrum and tabulated IRDF cross-section data for the isotopes of interest [21,23] according to the following equation [24].

$$\phi\ (cm^{-2}.s^{-1}) = RR / \langle\sigma\rangle \quad \textbf{[4]}$$

$$\langle\sigma\rangle = \frac{\int_{E_{thr}}^{E_{max}} \sigma(E)n(E)dE}{\int_{E_{thr}}^{E_{max}} n(E)dE} \quad \textbf{[5]}$$

Where σ(E) is the activation cross-section at energy E, n(E) is the neutron intensity at energy E, E_{max} is the maximum energy of the neutron in the spectrum, E_{thr} is the effective threshold energy calculated according to the follwing condition [25]:

$$\int_{E_{thr}}^{\infty} \sigma(E)\eta(E)dE = 0.95\int_{0}^{\infty} \sigma(E)\eta(E)dE \quad \textbf{[6]}$$

in which η(E) is the neutron spectral shape function

$$\eta(E) = \frac{n(E)}{\int_0^\infty n(E)d(E)} \quad \textbf{[7]}$$

Table 2 presents the results of measured reaction rates for the used foils, as well as various published data. Current data were obtained by neutron irradiation of monitor reactions utilizing ^{241}Am-Be source of $1.1x10^7$n/s nominal total emission rate, whereas Uddin data made use of ^{241}Am-Be neutron source with neutron yield of 7.5×10^6n/s [24]. The reaction rate data of Son and Tan were obtained employing Dalat nuclear research reactor (radial channel No. 4) with nominal power of 0.5 kW and $5.06x10^8$n/cm^2.s fast neutron flux as estimated by ^{58}Ni monitor reaction [16]. Azad data were obtained using a Spherical Uranium Converter (NISUS) with flux around $5x10^7$n/cm^2.s [26].

The values of measured reaction rates are affected not only by source flux, but also by the spectral shape function. ^{241}Am-Be sources' neutron spectra have multiple peaks at high energies with very little thermal neutron component (less than 1% of the total yield) **[23]**. On the other hand, Dalat reactor have Maxwelian shape around thermal neutrons follwed by 1/v fast neutron dependance. As outlined by Azad [26], NISUS uranium converter spectrum has a fast neutron maximum

value around 1 MeV and a long tail up till 11 MeV.

As is seen from table 2, **Uddin** isotopic neutron source has reaction rates of same order of magnitude compared with the current measurements. Likewise, fission neutrons of

Table 2: Reaction rate (s^{-1}) measurements for different threshold reactions.

Activation reaction	Current work	Uncertainty %	241Am-Be Neutron source [24]	Dalat research reactor [16]	NISUS facility [26]
$^{115}In(n, n')^{115m}In$	3.50E-20	8.66	3.76E-20	3.29E-16	0.12E-16
$^{58}Ni(n, p)^{58}Co$	2.89E-20	8.16	2.45E-20	3.04E-16	4.31E-16
$^{64}Zn(n, p)^{64}Cu$	8.96E-21	7.02	-	-	1.15E-16
$^{27}Al(n, p)^{27}Mg$	2.13E-21	7.03	-	-	1.40E-17
$^{56}Fe(n, p)^{56}Mn$	6.47E-22	7.27	-	8.95E-18	3.28E-18
$^{59}Co(n, \alpha)^{56}Mn$	5.02E-23	9.75	-	-	-
$^{24}Mg(n, p)^{24}Na$	7.36E-22	11.61	-	-	5.70E-18
$^{27}Al(n, \alpha)^{24}Na$	5.75E-22	7.17	-	1.72E-18	2.32E-18

Table 3: Flux values for different threshold reactions.

Reaction	Effective threshold energy (MeV)	Flux ($cm^{-2}s^{-1}$)	Uncertainty (%)
$^{115}In(n, n')^{115m}In$	1.8	1.04E+05	10.53
$^{58}Ni(n, p)^{58}Co$	2.8	5.97E+04	10.13
$^{64}Zn(n, p)^{64}Cu$	3.1	4.90E+04	9.23
$^{27}Al(n, p)^{27}Mg$	4.3	4.19E+04	9.48
$^{56}Fe(n, p)^{56}Mn$	5.9	2.49E+04	9.43
$^{59}Co(n, \alpha)^{56}Mn$	6.3	7.38E+03	11.45
$^{24}Mg(n, p)^{24}Na$	6.7	9.16E+03	9.25
$^{27}Al(n, \alpha)^{24}Na$	6.9	8.56E+03	9.29

Dalat reactor and NISUS converter have same data orders, reflecting the similarities of fluxes and spectral shapes.

The estimated integral-fluxes are presented in table 3. It is clear that reactions of lower threshold energy can respond to a wider range of energies yielding higher flux values (see figure 3). Therefore $^{115}In(n,n')$ flux can properly represent the fast neutron part of ^{241}Am-Be emitted neutrons covering the whole energy range from 1.8 MeV to 6.9 MeV.

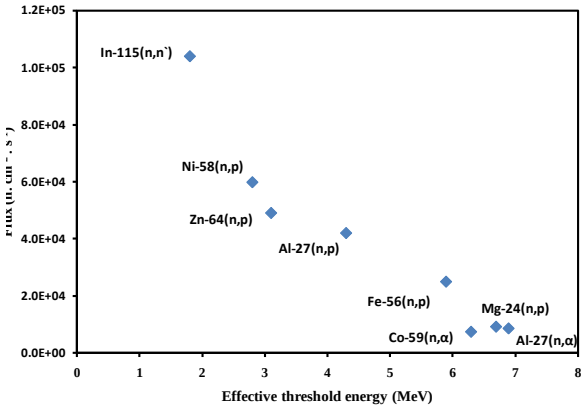

Fig. 3: Estimated integral-fluxes for different threshold detectors.

4. CONCLUSION:

In the present work, ^{241}Am-Be neutron source with activity 5 Ci and nominal neutron yield 1.1×10^{7} n/s has been characterized using threshold activation detectors and HPGe based gamma spectrometer. The used reactions cover effective-threshold-energy range from 1.8 to 6.9 MeV detecting the neutron energy up to the ISO-8529 limit of 11 MeV. The threshold detectors have been successfully applied to map the neutron flux over the entire energy range of the source-neutrons. The obtained results with data [24,26] comparisons were reveals that the differences of reaction rates could be explained in light of sources' flux and spectral shape. Thus, sources of different neutron origins (fission or reaction) show similarities in their induced reaction rates.

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الملخص العربى

الهدف من هذه الدراسة هو توصيف مصدر النيوترون النظائري من نوع أميرسيوم-بريليوم ذو النشاط الاشعاعى5كورى وفيض نيوترونى 1.1×10^{7} نيوترون فى الثانية. تم قياس النيوترونات المنبعثة باستخدام تقنية تنشيط الرقائق ذات طاقة العتبة باستخدام التفاعلات التالية:

^{115}In(n,n'), ^{58}Ni(n,p), ^{64}Zn(n,p), ^{27}Al(n,p), ^{56}Fe(n,p), ^{59}Co(n,α), ^{24}Mg(n,p), and^{27}Al(n,α)

تم اختيار تفاعلات العتبة بناء على نصف العمر،طاقة العتبة المناسبة والمقطع المستعرض. وقد تم قياس معدل تفاعل كل عينة من الرقائق عن طريق الكشف عن أطياف جاما الناتجة عن تحلل نواتج التفاعل.تم استخدام كاشف الجرمانيوم عالى النقاوة لقياسات طيف أشعة جاما. وقد تم حساب الكفاءة المطلقة المرتبطة بكل عينة باستخدام برنامج (Efftran). تمت مقارنة معدلات التفاعل المكتسبة مع البيانات المنشورة المتاحة.