Characterization of neutron flux spectra in the irradiation sites of a 37 GBq $^{241}$Am-Be isotopic source

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Abstract

For the applicability of instrumental neutron activation analysis (NAA) technique, an irradiation unit with a 37 GBq $^{241}$Am-Be neutron source was installed at Institute of Nuclear Sciences of Ankara University. Design and configuration properties of the irradiation unit are described. It has two different sample irradiation positions, one is called site #1 having a pneumatic sample transfer system and the other is site #2 having a location for manual use. In order to characterize neutron flux spectra in the irradiation sites, the measurement results were obtained for thermal ($\Phi_{th}$) and epithermal neutron fluxes ($\Phi_{epi}$), thermal to epithermal flux ratio ($f$) and epithermal spectrum shaping factors ($z$) by employing cadmium ratios of gold (Au) and molybdenum (Mo) monitors. The activities produced in these foils were measured by using a p-type, 44.8% relative efficiency HPGe well detector. For the measured $\gamma$-rays, self-absorption and true coincidence summing effects were taken into account. Additionally, thermal neutron self-shielding and resonance neutron self-shielding effects were taken into account in the measured results. For characterization of site #1, the required parameters were found to be $\Phi_{th} = (2.11 \pm 0.05) \times 10^{9}$ n cm$^{-2}$ s$^{-1}$, $\Phi_{epi} = (3.32 \pm 0.17) \times 10^{10}$ n cm$^{-2}$ s$^{-1}$, $f = 63.6 \pm 1.5$, $z = 0.045 \pm 0.009$, respectively. Similarly, those parameters were measured in site #2 as $\Phi_{th} = (1.49 \pm 0.04) \times 10^{9}$ n cm$^{-2}$ s$^{-1}$, $\Phi_{epi} = (2.93 \pm 0.15) \times 10^{10}$ n cm$^{-2}$ s$^{-1}$, $f = 50.9 \pm 1.3$ and $z = 0.038 \pm 0.008$. The results for $f$-values indicate that good thermalization of fast neutrons on the order of 98% was achieved in both sample irradiation sites. This is because an optimum combination of water and paraffin moderator is used in the present configuration. In addition, the shielding requirements are met by using natural boron oxide powder (5.5 cm) and boron loaded paraffin layers against neutrons, and a 15 cm thick lead bricks against gamma-rays from source and its surrounding materials.

1. Introduction

Over the past sixty years, instrumental neutron activation analysis (NAA) has been used in nuclear analytical laboratories as a powerful technique. This is because NAA allows user to make non-destructively multi-element analysis in a variety of the samples [1]. Additionally, in recent years, automation of the counting systems and data evaluation process have greatly increased the sample throughput and the standardization of the used procedures gives a chance to many NAA laboratories to accomplish the results with a high degree of accuracy and reliability. To this end, especially reactors and neutron generators are most useful neutron production sources since they provide high neutron fluxes at irradiation sites. However, the irradiation units composed of isotopic neutron sources such as $^{241}$Am-Be, $^{238}$Pu-Be, $^{252}$Cf(sf), etc., are also commonly used in NAA applications and small scale research activities although they have low neutron fluxes compared to those of either reactors or generators. The main advantage of these sources is that they have long half-lives, thus giving stable fluxes in the irradiation sites.

Additionally, in many instrumental NAA applications to determine elemental concentrations in samples, the well-established $k_0$ standardization method is often used. In the $k_0$-method, only a single isotope is used as a comparator standard to determine the element concentrations in the sample. Today, the $k_0$-method, are commonly employed at various NAA laboratories using different approaches and it has many advantages such as an economy of irradiation and counting costs, the achievement of high throughput and high accuracy and traceability [2]. However, the application of the $k_0$-method necessitates knowledge of the neutron spectrum shape characteristics [3]. It was also shown that the spectrum...
parameters vary considerably over the volume of the irradiation site [4]. This is true that if the used cavity is very large, thus leading to flux depression. Thus, a complete characterization of the neutron fields is a must for the accurate determination of the nuclear parameters such as effective resonance energies, thermal neutron and epithermal neutron cross sections for the isotopes via the neutron capture \((n,\gamma)\) reactions. For instance, lack of adequate characterization of the neutron spectra are the reasons for such discrepant values on the neutron induced reactions [5,6]. Hence, in this work, neutron flux spectra and flux parameters will be characterized in two different irradiation sites of an \(^{241}\text{Am-Be}\) neutron source by taking into account all possible precautions together with use of the newest measurement protocols. For the characterization of an irradiation facility, it is necessary to determine thermal neutron flux (\(\Phi_{\text{th}}\)) and epithermal neutron flux (\(\Phi_{\text{epn}}\)), thermal-to-epithermal flux ratio (\(f\)) and epithermal neutron spectrum shape factor (\(x\)). These are the essential flux parameters for the characterization of a neutron field. Where the shaping factor, \(x\) is a degree of, non-ideality of the epithermal neutron flux distribution in real neutron spectra, which can often be represented by \(1/E^{1+x}\) in an epithermal neutron energy range [7]. Of these parameters, the values of \(x\) and \(f\) are dependent on the irradiation site of a neutron source and they can be determined unless there is a change in the geometry of the neutron source [2]. \(x\) is independent of energy, can be positive or negative, depending on the neutron source configuration, moderator material, geometry of irradiation site [8]. In case of poor thermalization, neutron spectrum is shifted to higher energies, then \(x\) is negative and inversely if the neutrons tend to be more thermalized then \(x\) becomes positive [9].

In determining the reactor neutron spectrum parameters, the \(^{95,97}\text{Zr}^{198}\text{Au}\) method has been used to characterize the neutron spectrum. However, the \(\text{Zr}^{198}\text{Au}\)-monitor approach is not deemed as a very suitable monitor pair for daily application in between regular samples under routine conditions. This monitor pair does not give the correct results for monitoring \(f\) and \(x\) value at the irradiation positions when low fluxes are available from isotopic neutron sources [10]. This is mainly due to the measurement of the very low induced activities of \(^{95,97}\text{Zr}\) and \(^{198}\text{Au}\). Especially in low flux neutron fields, it is difficult to measure the induced activity of \(^{97}\text{Zr}\) since the net area of the 743 keV peak of \(^{97}\text{Zr}\) will be too poor [11].

Alternatively, \(^{197}\text{Au}\) and \(^{98}\text{Mo}\) monitor pair is usable for the epithermal flux measurement using the \(^{197}\text{Au}(n,\gamma)^{198}\text{Au}\) and \(^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}\) reactions [10]. \(^{197}\text{Au}\) is an ultimate monitor isotope because of good nuclear and decay properties. However, the second monitor \(^{98}\text{Mo}\) has also a relatively high resonance integral cross section of \(I_0 = 6.96\) b when compared to \(^{94}\text{Zr}\) monitor with \(I_0 = 0.271\) b thus it would give sufficient peak area results for 140.5 keV (4.52%) analytical peak with a good statistical quality when the Cd ratio method is carried out in low intense neutron fields. This is because \(^{98}\text{Mo}\) has a relatively large \(Q_0\)-value (i.e., \(I_0/q_0 = 53.1\)) for the monitoring of epithermal activation [12] and it has a much longer half-life of 65.976 h [13] for the measurement of the radioactivity produced in Mo-foil.

The first aim of this paper is to describe the physical aspects of an irradiation unit with a 37 GBq \(^{241}\text{Am-Be}\) neutron source, installed at Institute of Nuclear Sciences of Ankara University for the applicability of instrumental NAA technique. The second aim is to make the characterization of neutron flux spectra and flux parameters in two different irradiation sites of a \(^{241}\text{Am-Be}\) neutron source by taking into account all possible precautions together with an application of the newest measurement protocols. For this, the design and configuration properties of the whole irradiation unit are described in the following section. For the characterization of the neutron spectra in site \#1 and site \#2, thermal neutron flux (\(\Phi_{\text{th}}\)) and epithermal neutron flux (\(\Phi_{\text{epn}}\)), thermal to epithermal flux ratio (\(f\)) and epithermal spectrum shaping factors (\(x\)) are determined as the essential parameters. Measurements were performed by using cadmium ratios of Au and Mo foils as monitors.

2. Experimental

2.1. Design and configuration of the neutron irradiation unit

As seen in Fig. 1, the present irradiation unit contains a 37 GBq \(^{241}\text{Am-Be}\) neutron source placed in water tank surrounding with paraffin. Additionally it was shielded by the paraffin mixed with boron oxide and only boron oxide powder against neutrons. Lead bricks are used for gamma radiation shielding. In the present configuration, the source is originally encapsulated in a cylindrical aluminum sheath with 4.2 cm diameter and 13 cm height. The source sheath is then encased in the 0.4 cm thick-polyamide cylinder box with an internal diameter of 4.5 cm and an internal height of 15 cm to fix in the middle of the water tank by means of a grab handle. The de-ionized water was filled in a cylindrical plastic tank having a height of 100 cm and 95 cm diameter, which was mainly usable for thermalization of the fast neutrons. To support and configure of the whole irradiation geometry, the plastic water tank containing the source was placed in a wooden made enclosure box. It was assumed that wood is a practical choice since it contains H and C atoms, also for additional neutron moderation. The inside dimensions of the wooden box is 125 cm × 125 cm × 100 cm box. Top cover of the box is also filled by boron oxide powder for neutron shielding. In final installation, laying lead bricks is also performed on the flat top cover against gamma radiations. Additionally, the
moderation of fast neutrons (up to 12.1 MeV for $^{241}$Am-Be neutron source) was also enhanced by adding the melted paraffin surrounding the plastic tank. The second layer was made by using boron oxide loaded paraffin to absorb the thermalized neutrons. The opening between the lateral sides of the wooden box is filled with natural boron oxide to absorb completely the remaining thermal neutrons, where the boron oxide used is 5.5 cm in thickness. The best shielding for the neutrons is achieved by this design consideration. In order to shield the gamma rays from the source and the surrounding materials, Chevron type (zig-zag) lead bricks are put up a wall with thickness of 15 cm thickness around the wooden box. From point of the radiation protection, the whole neutron irradiation unit was installed in a room behind the laboratory, and licensed by Turkish Atomic Energy Authority according to the permissible dose limits.

The source is fixed between two irradiation channels. There are two sample irradiation positions in these channels, one of them is here called site #1 having a pneumatic sample transfer system or named “rabbit” sample transfer system. The other irradiation position is called site #1 having a location for manual use. In site #1 line, the pneumatic sample transfer system is provided with an air-compressor by use of rubber hoses. A metal screen is mounted to the lower part of the pipe to ensure that the rabbit is always placed in the middle height of the source. However, the compressed air flows through the metal screen to relieve it in each run of the pneumatic system as shown in Fig. 2.

The rabbit transfer capsules for the pneumatic tube irradiations are made of polyethylene and the shape of the rabbit is also shown in Fig. 2 in which the physical dimensions are given in millimeters. The lids of the capsules are rounded adequately to facilitate its movement in the plastic hose via the compressed air. The rabbit sample transfer system is controlled manually by means of a control switch in the laboratory. Additionally, for manual use a strength plastic pipe with a 117.5 cm height and 5.2 cm diameter is placed and fixed near by the $^{241}$Am-Be neutron source in water tank. The bottom end of the pipe is sealed for water-proof. Depending on the intended purpose, any sample is irradiated at a desired height when it is sent from top to bottom end of the plastic pipe by means of a guided locator.

2.2. Foil irradiations and measurements of the produced activities

Before establishing the fixed irradiation positions, a series of foil activation experiments were carried out for the determination the maximum neutron flux distribution around the $^{241}$Am-Be neutron source by using standard Au foils. Since the ultimate goal of the present irradiation unit is to utilize in some INAA irradiations for the samples, some irradiations and measurements were performed repeatedly to determine accurately the maximum available thermal neutron flux position in two different irradiation sites.

The $\alpha$-shape parameters of the epithermal neutron spectrum distribution at irradiation site #1 and site #2 were determined by using cadmium ratios of the foils, Au and Mo. These foils have high purity of greater than 99.95%, 0.0508 mm (0.002 inch) thickness and 12.7 mm (0.5 inch) diameter, purchased from Shieldwerx, A Division of Bladewerx LLC, Rio Rancho, NM, USA. The $^{197}$Au and $^{98}$Mo monitors were activated easily by $(n,\gamma)$ reaction. The irradiations were, in turn, performed using two set of samples: one with and the other without 1 mm thick Cd cylinder box whose diameter is 13 mm and height is 20 mm. The masses of Au and Mo foils are in order of 0.09–0.13 g. They were exposed to the neutrons in the irradiation sites in which the sample positions are always fixed. The foils were always irradiated within and without 1 mm thick cylinder Cd box to obtain Cd-ratio values.

The gamma-ray spectrometer used in the measurements consists of a 44.8% relative efficient, p-type HPGe well detector (Canberra, GCW4023), a spectroscopy amplifier (Canberra 2025) and a 14 bit ADC/MCA analyzer (Canberra Multiport II). The detector has a resolution of $\sim 2$ keV at 1332.5 keV ($^{60}$Co). The well of the present detector has a 23 mm diameter and a 35 mm depth

Fig. 2. The arrangement of a $^{241}$Am-Be neutron source and a pneumatic irradiation site.
to accommodate the sample tubes. It has a standard lead shield made of 10 cm thick, jacketed by a 9.5 mm carbon steel outer housing. The inside of lead is graded by a 1 mm thick Sn and a 1.6 mm thick Cu liners to attenuate low energy X- and gamma rays.

The gamma-ray spectra were acquired in a 4096 MCA channels. The amplifier gain was set to 0.75 keV/channel to cover about 3000 keV energy range. The full-energy peak detection efficiency, $r_p$, was calculated by using GESPECOR® (Ver 4.2) software that uses a Monte Carlo simulation [14].

The induced activities of Au and Mo foils were counted in the well of the detector by means of 1 mm thick polystyrene tube to obtain more counting efficiency within about 4m geometry. Thus good statistical quality of data were achieved from the acquired spectra because of both well counting geometry and the chosen longer acquisition periods. Background measurements were always subtracted from the sample spectra. The net counts for the peaks of interest were evaluated by means of Genie-2000 software and manually calculation from the sample spectra.

The thermal neutron self-shielding ($G_n$) and resonance neutron self-shielding ($G_{epi}$) for the irradiated foils were calculated by using Nisle’s approximation [15]. To do this, the required nuclear data, e.g., resonance parameters, absorption, scattering, total microscopic cross-sections, etc. were obtained from JENDL-4.0 [16], ENDF/B VII [17–19] and NuDat [13] online libraries. For the present counting geometry, gamma-ray self-absorption ($F_s$) and true coincidence summing ($F_c$) effects were calculated by a GESPECOR® (Ver 4.2) software that uses MCNP code simulation. Self-absorption correction factors $F_s$ were calculated using mass attenuation coefficients that were taken from NIST [20]. The efficiency data for the well counting geometry were also calculated from GESPECOR® (Ver 4.2) software built-in KORDATEN database.

The specific activity of $^{99}$Mo was determined by 140.511 keV gamma ray emission from the $^{99}$Mo($\beta^-$,$^{99m}$Tc[$10])$ reaction. The specific activities of $^{99}$Mo were measured, in turn after a bare Mo foil and a Mo foil within Cd-box irradiations. The reason for the use of 140.511 keV gamma ray emissions in the activity determinations, is the most intense gamma-ray of its decay product, $^{99m}$Tc. However, it is known that the parent isotope $^{99}$Mo also contributes to the 140.511 keV peak of $^{99m}$Tc [10]. Hence, For deducing only, after the total specific activity of $^{99}$Mo and $^{99m}$Tc was measured from the mixed 140.511 keV peak, only $^{99}$Mo specific activity can be deduced according to the calculation method given in literature [21,22], because the measured activity is a sum of parent–daughter decay case.

On the other hand, the $\alpha$-shape factor of the neutron spectrum can be determined with a sufficient accuracy by using cadmium ratio measurements of $^{197}$Au and $^{99}$Mo monitors, as described in the previous work [10]. It is an important point that the newest nuclear data involved in dual monitor method should be used. The $^{99}$Mo($n$,$\gamma$) $^{99m}$Tc monitor reaction gives relatively low induced activity due to low neutron fluxes from isotopic neutron sources, however, it is still a usable monitor because it has a reasonably good decay properties and nuclear data mentioned above. The used well-type Ge detector was also provided the good counting statistics for the measurement of the 140.511 keV peak from the $^{99}$Mo activity produced in the foil, especially for the case of cadmium covered foil irradiations.

### 2.3. Measurements of cadmium ratio of Au and Mo foils, $R$

For an isotope irradiated by neutrons, the cadmium ratio, $R$, can be easily calculated using induced activities of the isotope by the following equations:

$$ R = \frac{A_{sp}}{A_{sp}/F_{Cd}} $$

with $A_{sp} = \left\{N_p/(wSDCT_{m})\right\}_{bare}$ and $A_{sp}^+ = \left\{N_p/(wSDCT_{m})\right\}_{Cd}$

$A_{sp}$ denotes specific activities after irradiations. The superscripts "+" and "−" indicate the irradiation cases; Cd-covered and bare foil irradiation, respectively. $w$ is the element mass in the foil. After the end of the coating procedure: $N_p$ denote net number of counts under the full-energy peak collected during live time $t_{irr}$; $S = 1 - e^{-\gamma t_{irr}}$ is saturation factor with $\gamma = \text{decay constant}$, $t_{irr} = \text{irradiation time}$; $D = e^{-\gamma t_0}$ is decay factor with $t_0 = \text{decay time}$; $C = [1 - \exp(-\lambda t)]/\lambda t_0$ is measurement factor correcting for decay during the real time, $t_0$ for a measurement. $F_{Cd}$ is epithermal neutron transmission factor which is required to correct for absorbed epithermal neutrons by the cadmium shield. For the measured gamma-rays, the calculated self-absorption factors, TCS correction factors and other nuclear decay data used in the analyses are given in Table 1.

### 2.4. The ratio of thermal flux to epithermal flux, $f$

In neutron field there are always a thermal and epithermal components. This quantity gives the information about the available thermal flux and epithermal neutron flux in the irradiation site. Then the ratio of the thermal flux to epithermal flux, $f$, can be calculated by using cadmium ratio of the monitor isotope as follows:

$$ f = \frac{\Phi_{0\alpha}}{\Phi_{epi}} = \frac{\Gamma_{0\alpha}}{\Gamma_{epi}} \frac{G_{epi}}{G_{th}} (R - 1) $$

where $\Phi_{0\alpha}$ and $\Phi_{epi}$ are, respectively, thermal and epithermal neutron fluxes, $\sigma_0$ is thermal neutron cross section, $g$ is Westcott correction factor which is unity for $^{197}$Au and $^{99}$Mo. The $G_{epi}$ and $G_{th}$ are thermal and epithermal neutron self-shielding correction factors, respectively.

### 2.5. The epithermal spectrum shape factor, $\alpha$

Epithermal spectrum shape factor, $\alpha$ is an essential parameter in a non-ideal, real epithermal neutron spectrum [7]. It is related to order of non-ideality of the epithermal neutron flux distribution which is approximated by $\Phi(E) = 1/E^{1+\alpha}$. Accordingly, a resonance integral $I_q(\alpha)$ for a real epithermal neutron spectrum also characterized by $\alpha$-shape factor [23,24]. The information in detail about $\alpha$-shape factor were given in our previous works [25–28]. According to the previous works [29–33], $\alpha$-shape factor is related to other parameters such as the effective resonance energy ($E_r$), resonance integral to thermal neutron cross section ratio, $Q_d(\alpha) = [\pi I_q(\alpha)/\sigma_0]$ in a real $1/E^{1+\alpha}$ epithermal flux distribution:

$$ \ln \left( \frac{Q_d}{Q_0} \right) = \frac{0.429}{(E_r/cd) 0.429} - 0.629 = \alpha \cdot \ln E_r $$

It seems that $\alpha$-shape factor exists in both sides of the above Eq. (4), which can be solved by using an iteration procedure. To do this, both sides of Eq. (4) is set to an initial $\alpha$-value and then the procedure is repeated by changing the new $\alpha$-values until it converges to a constant value, as previously shown [34–36].

### 3. Results and discussion

For the characterization of an $^{241}$Am-Be source neutron irradiation unit, we have determined four essential parameters, namely: thermal to epithermal flux ratio, $f$ and epithermal neutron spectrum shape factor, $\alpha$ thermal neutron flux, $\Phi_{0\alpha}$ and epithermal neutron flux, $\Phi_{epi}$. Measurements were carried out in two different
To this end, the induced activities in these Au and Mo foils were measured by using a well-type HPGe detector. For the measured γ-rays, self-attenuation and true coincidence summing effects in the used foils were taken into account. Additionally, thermal neutron self-shielding and resonance neutron self-shielding effects were taken into account in the present determinations. In case of $^{98}$Mo($n$,γ)$^{99m}$Tc monitor reaction, the specific activities were measured from the peak area of the most suitable gamma ray of 140.511 keV (89.1%) from mother-$^{99m}$Tc. This is a suitable analytical peak among the others emitted from $^{99m}$Tc for the measurement. Although it is an intense (89.1%) gamma ray emission, it also mixes from the emission $^{99}$Mo($\gamma$)$^{99m}$Tc mother–daughter decay, thus it yielded to obtain the more accurate activity determination of $^{99m}$Tc after made the necessary branching ratio correction. For site #1 (pneumatic line) and site #2 (manually use position), the measured thermal neutron flux were found to be $\Phi_{th} = (2.11 \pm 0.05) \times 10^7$ n cm$^{-2}$ s$^{-1}$ and $\Phi_{th} = (1.49 \pm 0.04) \times 10^7$ n cm$^{-2}$ s$^{-1}$, epithermal neutron flux were $\Phi_{epi} = (3.32 \pm 0.17) \times 10^7$ n cm$^{-2}$ s$^{-1}$ and $\Phi_{epi} = (2.93 \pm 0.15) \times 10^7$ n cm$^{-2}$ s$^{-1}$, epithermal neutron spectrum shape factors were found to be $\alpha = 0.045 \pm 0.009$ and $\alpha = 0.038 \pm 0.008$ and thermal-to-epithermal flux ratios were found to be $f = 63.6 \pm 1.5$ and $f = 50.9 \pm 1.3$, respectively. The total uncertainties were estimated by using normal error propagation law by taking into uncertainty sources. In flux measurements, total uncertainties are order of 2.4–5.5, however the uncertainties in $\alpha$ are generally higher, i.e., 20–21% as expected.

In conclusion, a new irradiation unit was characterized in terms of basic flux parameters using the well-established protocols. The present irradiation sites will be used in instrumental NAA of some elements in the samples in future. In future NAA applications, it is planned to use $k_0$-NAA standardization method because the $k_0$-NAA software (Kayzero for Windows Ver.2, 2005) has already commercially purchased for our laboratory and also $k_0$-IAEA free software can be used for the comparison.

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**References**


