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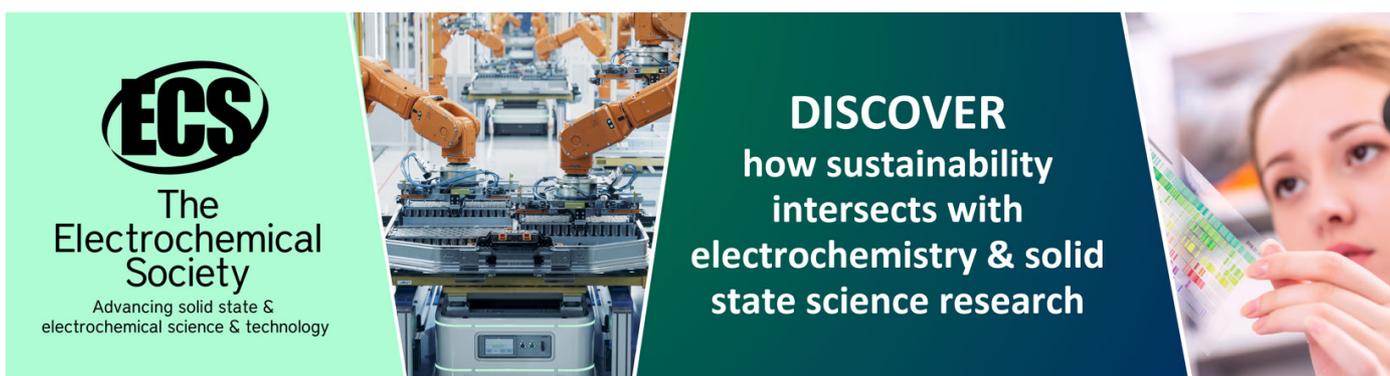
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Development of single sphere spectrometer with gold foil detector for neutron spectrometry

Rasito¹ and Bunawas²

Center for Applied Nuclear Science and Technology – BATAN, Jl. Tamansari 71
Bandung, Indonesia

¹Nuklindolab, Jl.Ir. Djuanda No. 5A Blok B Kav. P, Ciputat Timur, Tangerang
Selatan, Indonesia

E-mail: rasito@batan.go.id

Abstract. We developed a single sphere spectrometer (SSS) with a gold foil detector for neutron spectrometry. The detector of SSS is seven gold foil with 11 mm diameter and 0.18 mm thickness, which is placed in the polyethylene sphere of 30 cm diameter. The response of the detector was calculated for neutron in energy range 1×10^0 MeV up to 1×10^7 MeV using the Monte Carlo method by the MCNPX code system. The calculations were done based on the input of the SSS geometry model, source radiation, and fluence tally models. The validation of this SSS response was done with measure the neutron spectra from ^{252}Cf standard source at the neutron calibration laboratory of the Center for Technology of Radiation Safety and Metrology, National Nuclear Energy Agency, and compared with Bonner Sphere Spectrometer (BSS) LiI(Eu). Based on this comparison, it was known that the SSS Au foils measurement differs from the BSS LiI(Eu) by about 1%.

1. Introduction

Neutron fields ranging in energy from the thermal domain to tens or hundreds of MeV are a common characteristic of several facilities in the nuclear, medical, industrial, or research fields [1]. These neutron fields may be intentionally generated, as in nuclear plants or fast neutron irradiation facilities, or maybe a parasitic effect, as in accelerator-based cancer therapy. The neutron energy being distributed over ten or more orders of magnitude, an accurate measurement cannot prescind from the knowledge of the neutron spectrum, thus requiring the use of a spectrometer that responds over such a broad energy interval. Of the many neutron spectrometry techniques available to the scientific community, only the Bonner Sphere Spectrometer (BSS) fulfills this requirement. In neutron fields, BATAN has been applied to the neutron spectrometry method using Bonner Sphere Spectrometer (BSS) using LiI(Eu) detector [2].

The Bonner sphere spectrometer is still the only existing instrument that is able to respond from thermal energies up to hundreds of MeV neutrons, even if the energy resolution is known to be limited. In addition, it has a notable disadvantage that is the need to irradiate the spheres sequentially. This produces two main consequences: (1) the irradiation sessions are time-consuming because a BSS is typically composed of more than ten spheres, and (2) the BSS could never be adopted as a real-time spectrometer. The performance of the BSS in terms of energy interval would be highly desirable in most measurement applications of industry, research, and medical field. However, its use on a large scale is prevented by the inability to provide real-time monitoring [3].



In 2015, the first time BATAN developing a new neutron spectrometer i.e., Single Sphere Spectrometer (SSS) using gold (Au) foil detector. This device consists of a spherical moderator embedding seven thermal neutron detectors arranged in positions along the X-axis. The first idea of a multi-detector single sphere neutron spectrometer was developed by Bedogni et al. in the INFN-CIEMAT collaboration [4]. This included a moderating polyethylene sphere with thermal neutron detectors arranged along three perpendicular axes. The sphere diameter is 30cm, and the detector positions are placed at fixed radial distances (0, 3, 6, 9, 10.5, 12, 14 cm) along the three axes X, Y, and Z, the total number of measurement positions is 37 [1]. Measurements with this spectrometer are quite accurate but require a very long time. Besides requiring 37 detector response curves, it also requires 37 times for gamma counting. To overcome time efficiency, in the design of BATAN, try to reduce the number of detectors to 7 pieces. In BATAN design, the sphere diameter is 30cm, and the detector positions are placed at fixed radial distances (0, 3, 6, 9, 10.5, 12, 14 cm) but along the X-axes only.

The SSS has been designed using the Monte Carlo transport code MCNPX with seven positions. The MCNPX has been used to calculate the response matrix of the final configuration. As described below, the response of Au foils has been modeled as thermal neutron detectors with the idea of building a passive prototype to check the spectrometer response matrix experimentally. The simulated SSS readings have been unfolded with the UMG unfolding code in the absence of detailed pre-information and the unfolded spectra compared with the reference spectra i.e., using calibrated BSS LiI(Eu). The results are satisfactory and allowed approving the production of a prototypal spectrometer based on Au activation foils.

2. Method

The SSS was developed through five steps i.e., designing detector and moderator, modeling of a neutron source, calculation of neutron response function, measurement of a neutron source, and spectra unfolding. This spectrum will be compared with neutron measurement with calibrated BSS. Neutron response of SSS Au foils detector calculated using MCNPX through three steps i.e., the preparation of input files, the execution of the code, and the interpretation of the *output*.

2.1. Detector and moderator

The moderator of a single sphere spectrometer that developed in the BATAN Neutron Laboratory made from polyethylene with 30 cm diameters. The seven detectors made from Au foil with 11 mm × 0.18 mm used as thermal neutron detectors located at long the X-axis of SSS. Design of a single sphere spectrometer of BATAN, as shown in figure 1.

Gold is among the most commonly used materials for neutron flux monitors. It offers several advantages, e.g., it can be produced in great purity; it yields high activities due to its large thermal cross-section for neutron capture in the nuclear reaction $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$; ^{198}Au has a very convenient half-life of 2.7 days [5].



Figure 1. A prototype of SSS with 7 Au foils

The response of the Au activation foils has been calculated as the number of $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reactions within the foil volume normalized per unit incident fluence, using the track-length scoring option for the fluence (F4 tally), i.e.:

$$N_{(n,\gamma)} = \int dE \Phi_E V \rho_{at} \sigma_{(n,\gamma)} \quad (1)$$

where $\sigma_{(n,\gamma)}$ is the microscopic cross-section for (n,γ) reactions in Au, V is the volume of the foil, ρ is the atomic density (atoms per unit volume), Φ_E is the energy distribution of neutron fluence, and $N_{(n,\gamma)}$ is the number of (n,γ) reactions in the considered volume.

The Au activation foils have a diameter of 1.1 cm, thickness 0.018 cm, and purity higher than 99.9%. The density of the foils is 19.276 g cm⁻³, thus giving $V_{\text{rat}}=5.888 \times 10^{-4}$ b⁻¹ cm⁻¹. The moderator materials are polyethylene, (CH₂)_n 0.955 g cm⁻³ in density. Atomic composition and physical data of different elements utilized to build the model were obtained from the Compendium of Material Composition Data for Radiation Transport Modeling.

2.2. Model of the neutron source

In these calculations, the Au foil body was oriented parallel to the source-sphere axis. The sphere-detector combination was irradiated with a neutron beam produced by a disk-shaped neutron source. Then, a monoenergetic neutron source whose neutrons were directed towards the polyethylene sphere is shown in figure 2. Irradiations were carried out using 28 monoenergetic neutron sources with energy range from 1×10^6 MeV up to 1×10^8 MeV. These calculations were performed with the Monte Carlo code MCNPX version 2.6.0 using the LA150 cross-section library. The response was defined as the number of $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reactions per incident neutron fluence based on the track length estimate of detector flux normalized to one starting particle.

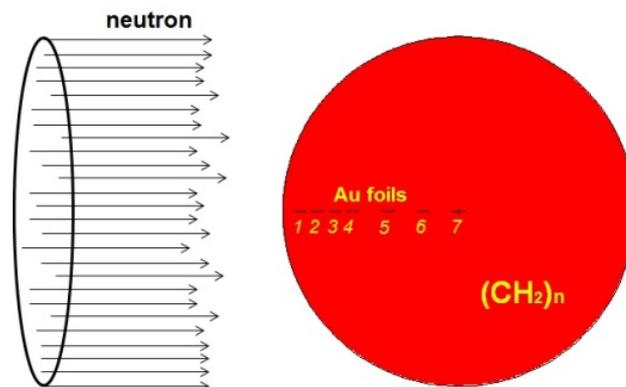


Figure 2. Disk-shape neutron source with parallel direction

2.3. Response calculations

The response of i detector as energy function is obtained by using this equation [6]:

$$\begin{aligned} R_i(E) &= \frac{S_i N}{\rho} \int_0^E \sigma_{(n,\gamma)} \Phi_E dE \\ &= C_i \int_0^E \sigma_{(n,\gamma)} \Phi_E dE \end{aligned} \quad (2)$$

where $S_i = \frac{\pi}{4} d_i^2$ (cm²) is the source surface area considered sphere diameter d , N (5.88×10^{23} atoms-cm⁻³) is the Au foil volumetric atom density, ρ (19,2 g cm⁻³) is its mass density, and C_i is the multiplier constant.

In MCNPX simulation, the detector response is calculated using track-length tally (F4) for neutrons (cm^{-2}). Tally of *F4* can score cell fluence ($\text{particles}/\text{cm}^2$) and *statistical errors*. The response function can be calculated by multiply the fluency with the multiplication factor. The *multiplication factor* using a tally multiplier (FM) is one of the most important properties to convert fluence to response function. The FM value is neutron field area (cm^2) \times atomic density of detector (10^{24} atoms cm^{-3}) divided by detector density (g cm^{-3}). The value of response function (cm^2/g) will be resulted from fluence (cm^{-2}) \times FM (cm^2/g). The correct form to write the FM card in MCNPX input is FM4 (Ci 2 102). The multiplier constant Ci of 2.159 and 102 is the identification number generally used by the ENDF/B-VI library to classify the radiative capture reaction (n,γ). According to the MCNPX requirement, N should be used in the unit of 10^{24} cm^{-3} since the cross-section tabulated values are directly taken by the code in barn units ($1 \text{ barn} = 10^{-24} \text{ cm}^2$).

3. Results and discussion

3.1. The response of SSS Au foils

MCNPX with SSS geometry, radiation source, and fluence input models was run using a computer with processor intel core i5 2.3 GHz and RAM 4 GB. In this work, we use a number of particle histories 1×10^6 . We assumed that this number of particle histories was enough because of the result of the statistical error in MCNPX output less than 3% for each response value. The MCNPX output is neutron fluence on the Au foil detectors as a function of initial neutron energy. The complete response function values of the whole SSS Au foils, as obtained with the biased MCNPX calculations, are given in figure 3. It can be clearly seen that the single-sphere can be chosen for this system may cover with a sufficient guarantee a very large neutron energy range (i.e., from thermal neutrons normally described by a Maxwellian energy distribution around 25 meV up to tens of MeV).

The MCNPX code is simulating the neutron transport from source to the materials based on cross-section data of elements in material for each incoming neutron energy. For neutron energies below 20 MeV, evaluated cross-sections are available that are validated against experimental data. But for neutron energies above 20 MeV, experimental cross-section data are scarce. Therefore intra-nuclear cascade (INC) and evaporation models are applied in these codes [7]. For this reason, every neutron transport code is based on theoretical nuclear models to describe interactions of neutrons with nuclei in the matter. Calculation of response functions using the Monte Carlo code and nuclear models used, which in turn may add to the uncertainty involved in SSS, in particular for neutron energies above 20 MeV [7].

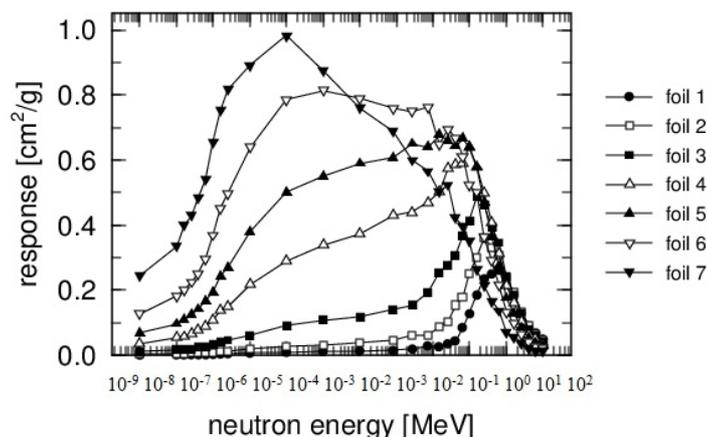


Figure 3. The neutron response function of SSS with 7 Au foil detectors.

However, the response functions at high neutron incidence energies are rather low due to the elastic scattering cross-sections with hydrogen nuclei that drop sharply above 10 MeV [6]. In this figure, the

calculated response functions are shown the seven response functions of Au foil detectors. The broad shape curves for the small foil number become narrower with a shift of the maximum response to higher energies when increasing the polyethylene sphere diameters. This fact reflects the physical effect of the neutron thermalization mechanism inside these narrow polyethylene. Each one of these curves should be understood as equivalent to a continuous function of the neutron incidence energy in the unfolding procedure.

The calculations were considered with the parallel neutron incidence direction (see figure 2) only, so this method not necessary for measurement with incident beam perpendicular (normal) to the gold foil plane or with a truly isotropic neutron emission. According to Amgarou et al. calculation's, percent deviations of inner gold foil responses given with parallel and normal neutron incidence directions from those obtained with isotropic emission are below 3.5% [6].

3.2. Measurement of ^{252}Cf neutron spectra

Measurement of ^{252}Cf neutron spectra using SSS Au foil is shown in Fig. 4. This measurement is compared with active BSS employing $^6\text{Li}(\text{Eu})$ detector. The BSS $^6\text{Li}(\text{Eu})$ counters have, in general, high neutron sensitivity. As an example, the response of the 20.31 cm diameter BSS sphere at 1 MeV neutron energy ranges is 0.1 - 3 cm^2 [8]. The active BSS, previously validated in several well-known neutron fields, is based on a cylindrical 4 mm \times 4 mm $^6\text{Li}(\text{Eu})$ scintillator. The experiment took place in the neutron laboratory (about 12 m \times 6 m \times 5 m) at 100 cm from the ^{252}Cf neutron source. The neutron fluence rate ranged in the interval 90 - 95 $\text{cm}^2 \text{ s}^{-1}$ [2].



Figure 4. Measurement of ^{252}Cf neutron spectra using SSS (a) and BSS (b).

The SSS Au foils, shown in figure 4b, operate with the same set of positions as the BSS. In every exposure, seven gold foil with diameter 11 mm, thickness 0.18 mm, and purity > 99.9% are positioned in the sphere with a specially designed polyethylene. Thermal and epithermal neutrons activate the ^{197}Au in natural Gold producing ^{198}Au , half-life 2.697 d with 0.411 MeV gamma emission. Due to its high energy and high yield, the gamma emission is certainly the most suitable for in situ measurements. The measurements of ^{198}Au activity using CAPRAC, a good type gamma counter with a NaI detector. Inside a neutron field, the foil detector produces activity per mass, C , that is related to the response matrix, $R_i(E)$, and the neutron spectrum, $\Phi(E)$, through the Fredholm integral equation of the first kind, shown in equation (3)

$$C = \int_0^E R_i(E) \Phi(E) dE \quad (3)$$

The SSS Au foil activity is expressed in terms of saturation specific activity. The procedure needed to obtain this quantity from activity the result of the gamma counting implies corrections for (1) the detector efficiency, (2) the decay between the end of the irradiation and the beginning of the measurement, (3) the decay during the measurement and (4) the fraction of the saturation activity

reached during the irradiation [8]. The last factor (saturation factor, F_{sat}) is given by equation (2) for constant rate sources:

$$F_{sat} = 1 - 2^{-\frac{t_{irr}}{T}} \quad (4)$$

where t_{irr} is the irradiation time and T is the half-life of the isotope.

The neutron fluence rate in the measurement point was proportional to the Au foils activity. Using the step by step correction algorithm, the activity of each foil was corrected to obtain the saturation activity in the condition of reference intensity. The corrected specific activities are used as input data for the unfolding code, and the unfolding process provides (among other data) the fluence rate corresponding to the reference intensity. This value divided by neutron source activity provides the fluence per activity, which is directly comparable with the data measured with the BSS LiI(Eu) detector. From this measurement and using UMG 3.3 unfolding code obtained the ^{252}Cf neutron spectra, as shown in figure 5.

A little bit different in the results of neutron measurements using Au foil with $^6\text{LiI}(\text{Eu})$ can be influenced by several things, including the value of a cross-section of neutron capture reaction. In general, the neutron capture cross-section for the ^6Li much greater than 940 barn [9] [10], while ^{197}Au is 98.8 barn [11]. The difference value large enough cross-section of this result is a very different sense of both detectors, which in $^6\text{LiI}(\text{Eu})$ is almost 10 times more sensitive than Au. This difference will affect quite significantly, especially when measuring the low flux of neutrons. Some references say that the sensitivity Au would be good for neutron flux measurement, or their application is operatively limited to fluence rate higher than in the order of $10^3 \text{ cm}^2 \text{ s}^{-1}$ [8].

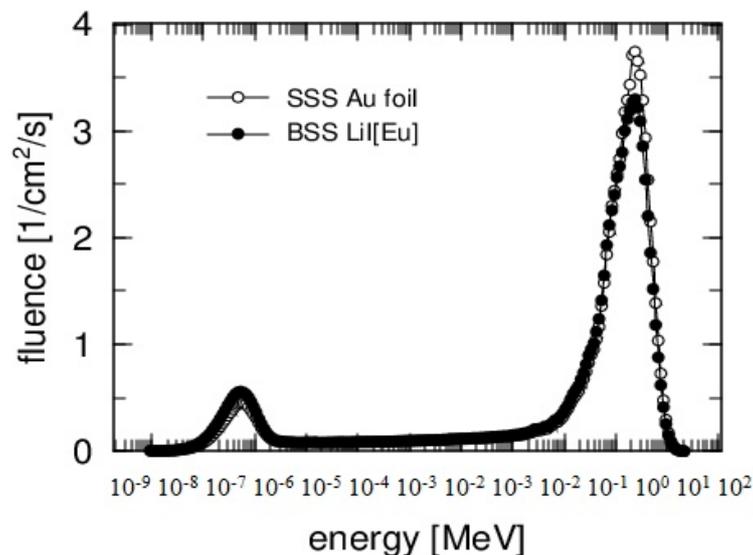


Figure 5. Neutron spectra of ^{252}Cf from SSS Au foils and BSS LiI(Eu).

The $^{197}\text{Au}(n,\gamma)$ cross-section is an established cross-section standard for thermal neutron energies (at 0.0253 eV) and for the energy range between 0.2 and 2.8 MeV. The cross-section at the epithermal neutron energy of $10^6 - 10^2$ MeV is the resonant area with 1550 barn [11]. The $^6\text{Li}(n,t)$ cross-section has a peak region at the fast neutron energy from 0.1 to 1 MeV. This difference will affect the measurement results, especially in the area of an epithermal and fast neutron, especially if accuracy in the calculation of the response matrix is less good.

The uncertainty in the fluence is the quadratic combination of the unfolding uncertainty and SSS calibration uncertainty. The unfolding uncertainty is the result of the propagation of the Au foils

activity uncertainties and detector response (MCNPX) uncertainty. The neutron fluence obtained with the SSS Au foils $92 \pm 11 \text{ cm}^2 \text{ s}^{-1}$. This value is coherent with the BSS LiI(Eu) is $91 \pm 9 \text{ cm}^2 \text{ s}^{-1}$. The neutron fluence value of SSS Au foils differs from the BSS LiI(Eu) by about 1%, which confirms the expected level of accuracy of the SSS Au foils.

4. Conclusion

The SSS neutron spectrometer has been a success to be developed in BATAN using seven of Au foil detectors. The SSS performance has been tested in the measurement of ^{252}Cf neutron spectra and compared with BSS LiI(Eu) calibrated. Based on this comparison, it was known that the perform of the SSS Au foil detector was in a good agreement.

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