CALIBRATION OF INDIUM RESPONSE FUNCTIONS IN AN Au–In-BSE SYSTEM UP TO 800 MeV

Zhonglu Wang^{1,3,*}, Rebecca M. Howell¹, Eric A. Burgett², Stephen F. Kry¹, Nolan E. Hertel²
and Mohammad Salehpour¹

¹Department of Radiation Physics, The University of Texas M. D. Anderson Cancer Center, 1515 Holcombe Blvd., Houston, TX 77030, USA

²Neely Nuclear Research Center, G. W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA

*Corresponding author: zhonglu.wang@caritaschristi.org

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Calibration of the response functions of a gold (Au)–indium (In) dual foil Bonner sphere extended (BSE) system was described. The response of the In and Au foil of the system was calculated using MCNPX code with different activation cross-sectional libraries: (ACTL and ENDF VI for gold and ACTL and 532DOS2 for In). To verify and correct the calculated response functions the Bonner sphere set (BSS) was irradiated using ²⁵²Cf and ²⁴¹AmBe sources of known neutron strengths for neutrons ranging from thermal to 20 MeV, and was irradiated at the 800-MeV neutron beam of the Los Alamos Neutron Science Center. The neutron spectrum of the 800 MeV beam was determined using time-of-flight (TOF) technique. We observed that the uncertainty of activation cross section in the resonance region can result in great uncertainty in the MCNPX-calculated response functions of activation foil-based BSS. The MCNPX-calculated response functions must be corrected using neutron sources of known spectrum and strength.

INTRODUCTION

Proton irradiation for cancer therapy is an increasingly popular treatment modality due to its highly conformal nature. However, neutrons are generated during patient treatment through the interaction of protons with the accelerator structure, treatment room and patient. These neutrons have no therapeutic value, but rather present a health risk to the patient as they irradiate non-target tissues and can thereby induce secondary malignancies. It is essential to study these neutrons to understand and mitigate when possible the risk they pose to the patient. While several investigators have used Monte Carlo calculations to investigate the secondary neutron spectra from proton radiotherapy, a limited number of measurements are reported in the literature. This is primarily due to the difficulties in measuring these high-energy neutrons as their energy will be as high as that of the protons used for radiotherapy, up to 250 MeV. The challenge arises in that most neutron detectors have limited response >20 MeV.

Bonner spheres have been used more than any other neutron spectrometer because the system responds over a very large energy range (thermal energies to 20 MeV) and has a nearly isotropic response.⁽¹⁾ The measurement system consists of thermal neutron detectors placed in the centres of

polyethylene (PE) moderating spheres with diameters ranging from 2" to 12".⁽²⁾ Higher energy neutrons are detected with the larger spheres; however, the larger spheres in the standard Bonner sphere set respond with similar sensitivities to neutrons >10 MeV and have strongly decreasing sensitivities >20 MeV.^(1,2)

Birattari *et al.*^(3–5) developed an extended range neutron rem counter LINUS by adding a shell of high-atomic-number (high-Z) material in the moderator. The high-Z materials have large (n,xn) cross sections and increase the number of neutrons in the detection system while reducing their energy, thereby increasing their chance of being moderated in the PE and being detected. Based on this concept, studies have investigated recently the use of high-Z material shells in conjunction with standard PE spheres to differentiate and increase the high-energy response of neutron spectrometers^(6–10). Wiegel and Alevra⁽⁶⁾ developed such a system, known as *NEMUS*. The system is commercially available and uses a ³He-filled proportional counter within 12 PE spherical shells and four PE spheres that contain copper or lead inlets⁽⁶⁾. This system has been successfully used to measure high-energy neutron spectra in low-intense neutron field⁽¹¹⁾; however, it is of limited utility for measurements in intense or pulsed fields as dead-time losses arise from pulse pile-up.

To accommodate the measurement of high-energy neutrons in intense or pulse beams, Burgett⁽¹⁰⁾ described a Bonner sphere extension (BSE) that used

³Present address: Hudner Oncology Center, Saint Anne's Hospital, 795 Middle Street, Fall River, MA 02721, USA.

copper, lead and tungsten shells in conjunction with the standard regular Bonner sphere (rBS) set (six spheres: 2", 3", 5", 8", 10" and 12"). While the detector system can accommodate an active detector ($^6\text{Li}(\text{Eu})$), it can also be used with a well-established passive detection system in pulsed-beam conditions: gold (^{197}Au) activation foils⁽¹⁰⁾. Although this detector system functions well⁽¹²⁾, the Au foils have a relatively low activation cross section and a long half-life, reducing the sensitivity of the system. While this is not usually a problem, a system with increased sensitivity is advantageous when measurement time is limited. Such limited time for measurement of a pulsed-beam is common for medical proton accelerator, where the vast majority of the beam time is dedicated to patient treatment and quality assurance. To accommodate the beam time limitations, it was desired to use indium (In) activation foils. The In foil increases the sensitivity of the system because the reaction $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ has a higher activation cross section and the $^{116\text{m}}\text{In}$ has a shorter half-life (54 min). The In foils could be reasonably incorporated into the detection system through the use of a dual-foil system that simultaneously uses both Au and In activation foils. A dual-foil detector system used with the standard Bonner sphere set (BSS) has been previously calibrated at low energy using ^{252}Cf and americium-beryllium ($^{241}\text{AmBe}$) neutron sources⁽¹³⁾.

In the current study we used the dual-foil detector system in conjunction with the BSE, hereafter referred to as the Au–In BSE system. We calculated response functions specifically for the extended spheres in the Au–In BSE. The most accurate working response functions are determined through a combination of calculations and measurements in calibrated monoenergetic neutron beams⁽¹⁾. However, there are few facilities with such beams and, therefore, neutron sources with wider energy distributions are often used. In the current work measurements were performed in a high-energy neutron beam-line at the Los Alamos Neutron Science Center (LANSCE) to correct the magnitude of the calculated response functions for the Au and In foils in each sphere of the Au–In BSE.

METHODOLOGY

Description of Au–In BSE

The Au–In BSE design is a combination of the extended spheres described by Burgett⁽¹⁰⁾ and a dual-foil holder described by Wang *et al.*⁽¹³⁾. The design of the BSE multisphere is briefly described in the following section. Further details can be found in a thesis by Burgett⁽¹⁰⁾. The existing commercially available Bonner spheres served as the basis for the design, then concentric shells of copper (Cu), tungsten (W), and lead (Pb) were added in various combinations with the existing 3" and 5" Bonner spheres

(designated small and large assemblies based on the inner Bonner sphere size). The small assembly consists of a standard 3" Bonner sphere, surrounded by a 1.5-mm thick Al shell (76.2 mm (3") inner diameter (ID) and 127 mm (5") outer diameter (OD)) filled with Cu powder, W granulates and Pb solid or and further encased in a PE sphere with an 203.2 mm (8") OD. The large assembly has a similar design but used a 5" Bonner sphere surrounded by a 1.5-mm thick Al shell (127 mm (5") ID and 177.8 mm (7") OD) filled with Cu, Pb or W and encased in a PE sphere with a 304.8 mm (12") OD. Both assemblies can be used with or without the outer PE sphere. A dual-foil holder and assembly of a small BSE are shown in Figure 1. Twelve spheres with different response functions can be obtained by combinations of the metal and polyethylene shells and the 3" and 5" regular PE spheres. For convenience, each combination of the BSE was named with three components in the following manner: the first component is a number, 3 or 5, indicating the size of the regular BS in the centre, the second component is the symbol of the shell metal, (Cu, Pb or W), the third component is a letter, B (stands for bare, no polyethylene covering the metal shell) or P (stands for polyethylene shell covering the metal shell). For example, 3PbB means a 3" BS covered by lead shell without polyethylene shell covering outside the lead shell.

A customised PE holder was designed to simultaneously accommodate one In foil and one Au foil placed orthogonally to each other. The holder that fits into the centre of a Bonner Sphere, resembles a standard $^6\text{Li}(\text{Eu})$ detector is shown in Figure 1. An Au foil is placed in the slit and an In foil is placed on top of the holder.

Response function calculations

The response functions of the Au–In BSE were computed up to 1 GeV using the MCNPX code⁽¹⁴⁾. The geometry of the sphere set, including Al shell and air gap between holder and sphere, were modelled in detail. From 5.0×10^{-10} to 1.0×10^{-8} MeV, response functions were calculated in 10 equal lethargy bins per decade; from 1.0×10^{-8} to 1.0×10^3 MeV, response functions were calculated in 20 equal lethargy bins per decade. The extended neutron cross-sectional library, LA150⁽¹⁵⁾, was used in the response function calculations.

Two (n, γ) cross-sectional libraries⁽¹⁶⁾ were used to tally the neutron capture reaction rates of ^{198}Au and $^{116\text{m}}\text{In}$. For the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction tally, cross sections from the Evaluated Nuclear Data File (ENDF/V) and the ACTL library were used. For the $^{115}\text{In}(n,\gamma)^{116\text{m}}\text{In}$ reaction tally, cross sections from the ACTL library and the 532DOS2 library were used. For the $^{116\text{m}}\text{In}$ calculation using the ACTL library, two MCNP MT numbers, 102 and

Figure 1. A dual-foil holder and the assembling process of a small BSE without and with an outer PE shell (an Au foil is being inserted in the middle slit).

1102, were used to calculate the two states of $^{116\text{m}}\text{In}$ with half-lives of 2.18 s and 54.41 min, respectively. MCNP MT number describes the interaction type in the Monte Carlo simulation, e.g. 102 describes (n, γ) interaction⁽¹⁴⁾. Because $^{116\text{m}}\text{In}$ decays completely from its 2.18-s half-life state to its 54.41-min half-life state within 1 min, the results of the two tallies were summed together to represent the quantity of $^{116\text{m}}\text{In}$ with a half-life of 54.41 min.

Measurements

Correction of the response functions <20 MeV

The response function correction experiments using ^{252}Cf and $^{241}\text{AmBe}$ neutron sources were conducted at the high bay area of Neely Nuclear Research Centre, Georgia Institute of Technology. The neutron spectral fluence rates at the location of the measurement consisted of two components: the neutrons directly coming from the source and the

neutrons from room return and air scattering (RR&AS). The spectral fluence of neutrons directly from the source was determined from the neutron source strength and the neutron spectrum from International Standard ISO-8529⁽¹⁷⁾. The RR&AS neutron spectral fluence was determined by the well-established $^6\text{LiI(Eu)}$ detector-based Bonner sphere system. Please refer to ref. (13) for the correction procedure. The corrected response functions here after will be referred to as the ^{252}Cf and $^{241}\text{AmBe}$ -corrected response functions, R_{ij}^{CA} , where i stands for the identification number of the BS, and j for the energy group, as described in ref. (13).

Correction of the response functions >20 MeV

For neutrons >20–800 MeV, the response functions of the system was corrected using measurement of the neutron beam of WNR 15 flight path at 90 m station at the LANSCE. The neutron spectrum was

determined using time-of-flight (TOF) method. Neutrons <20 MeV could not be determined using TOF method due to the long flight path and consequent overlap of the low-energy neutrons from the previous pulse with the current pulse.

The TOF neutron spectrum normalised to cm^{-2} per 1000 fission chamber counts is shown in Figure 2 (see ref. 10 for detailed normalisation method). The neutron spectra shown in Figure 2 were regrouped according to energy structure of the response function.

The activation rate of Au foil was very low due to the low intensity of neutron fluence and the limited beam time, so only the In foils were counted, though the Au and In foils were irradiated at the same time.

In order to determine the correction factors for the response functions >20 MeV, the neutron spectrum <20 MeV should be determined first. The regular BSS (rBSS) was used whose response functions were already corrected using ^{252}Cf and $^{241}\text{AmBe}$ sources of known strength, to determine the neutron spectrum <20 MeV. Since the response functions of the rBS to high-energy neutrons (>20 MeV) were small, the contributions to the reaction rates from these high-energy neutrons should also be small. Nevertheless, it must be accounted for to quantify the response. The TOF-measured neutron spectrum (>20 MeV) was convoluted, $\Phi_{>20}$, with the ^{252}Cf and $^{241}\text{AmBe}$ -

corrected response functions, R_{ij}^{CA} , of the six rBSS to obtain the high-energy neutron contributions to the reaction rates, $A_{i(>20)}^{\text{C}}$, and subtracted these contributions from the measured values, $A_{i(\text{total})}^{\text{M}}$, to obtain the reaction rates from neutrons <20 MeV, $A_{i(<20)}^{\text{C}}$. The neutron spectral fluence <20 MeV, $\Phi_{<20}$, was determined by unfolding these $A_{i(>20)}^{\text{C}}$ data using MAXED code⁽¹⁸⁾ with the MCNPX simulated neutron spectrum as the default spectrum.

Once $\Phi_{<20}$ was determined, the spectrum was convoluted with the rBS response function to determine the reaction rates produced by neutrons <20 MeV for each of the six rBSs. Next, the rBS reaction rates from neutrons >20 MeV was obtained by subtracting $A_{i(>20)}^{\text{C}}$ from $A_{i(\text{total})}^{\text{M}}$. The correction factors for response functions >20 MeV were determined by equation (1):

$$\begin{aligned} \text{CF}(>20)_i &= \frac{A_{i(\text{total})}^{\text{M}} - A_{i(<20)}^{\text{C}}}{A_{i(>20)}^{\text{C}}} \\ &= \frac{A_{i(\text{total})}^{\text{M}} - \sum_{j(<20 \text{ MeV})} R_{ij}^{\text{CA}} \cdot \Phi_j}{\sum_{j(>20 \text{ MeV})} R_{ij}^{\text{CA}} \cdot \Phi_j} \quad (1) \end{aligned}$$

The response functions of the six rBSs were corrected according to their correction factors. The above procedure was performed iteratively until $\Phi_{<20}$ converged.

Figure 2. TOF-measured neutron spectrum of the LANSCE 800-MeV neutron beam (black thick line and labelled as 'TOF (regrouped)', neutron spectrum <20 MeV is determined using regular BS (labelled as 'Unfold (regular BSS)') and the unfolded neutron spectrum with corrected response of the Au-In BSE (labelled as 'Unfold (RF-final)').

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The determined $\Phi_{<20}$ was then combined with the TOF-measured neutron fluence rate to form the complete neutron spectral fluence rate of the LANSCE 800-MeV neutron beam at the

measurement location. Because the cross sections <150 MeV were relatively accurate due to the availability of the evaluated neutron cross-sectional library, LA150, in the MCNPX code, the correction

Figure 3. MCNPX-calculated response functions of (a) ^{198}Au and (b) $^{116\text{m}}\text{In}$ for the BSEs of 3PbB and 3PbP to demonstrate the effects of different activation cross-sectional libraries on the response calculations.

of response functions >20 MeV was separated into two parts: from 20 to 150 MeV and >150 MeV. The uncertainties of the MCNPX-calculated response functions >150 MeV are much larger due to the lack of cross-sectional library, so the response functions >150 MeV were subjected to more corrections.

The response functions in the range of 20–150 MeV were smoothed to avoid abrupt changes in adjacent groups. The neutron spectral fluence rates <150 MeV, $\Phi_{<150}$, were convoluted with the ^{252}Cf and $^{241}\text{AmBe}$ -corrected response functions, R_{ij}^{CA} , to obtain the reaction rate, $A_{i(>150)}^{\text{C}}$. The correction

Figure 4. ^{252}Cf and $^{241}\text{AmBe}$ source-corrected response functions of $^{116\text{m}}\text{In}$ in the Au–In BSE system.

factors for response functions >150 MeV were determined by equation (2).

$$\begin{aligned} CF(>150)_i &= \frac{A_{i(\text{total})}^M - A_{i(<150)}^C}{A_{i(>150)}^C} \\ &= \frac{A_{i(\text{total})}^M - \sum_{j(<150 \text{ MeV})} R_{ij}^{CA} \cdot \Phi}{\sum_{j(>150 \text{ MeV})} R_{ij}^{CA} \cdot \Phi} \end{aligned} \quad (2)$$

And the corrected response functions are as follows:

$$\begin{aligned} R_{ij} &= R_{ij}^{CA} \quad \text{for } j \leq 150 \text{ MeV} \\ R_{ij}^{CA} \cdot CF(>150)_i &\quad \text{for } j > 150 \text{ MeV} \end{aligned} \quad (3)$$

RESULTS

Response functions were calculated for all spheres and for both In and Au foils placed at the dual-foil holder using MCNPX code. The MCNPX-calculated response functions of 3PbB and 3PbP are shown in Figure 3. For Au foils, the response functions calculated using ENDF and ACTL activation cross-sectional libraries have significant difference, especially for small spheres (Figure 3(a)). For In foil, the response functions calculated using ACTL and 532DOS2 activation libraries also have big difference (Figure 3(b)), though not as big a difference as that of the Au foil response functions. These

discrepancies are mainly caused by the uncertainties of cross section in the resonance region and the uncertainty of the determined RR&AS neutron spectral fluence rate. More detailed explanation can be found in ref. (13).

The ^{252}Cf and $^{241}\text{AmBe}$ sources corrected In foil response function of each BSE is shown in Figure 4. Some of the BSE's response functions corrected using LANSCE 800-MeV neutron beam are shown in Figure 5. Comparing the response functions of the same BSE in Figures 4 and 5, large corrections were made using the LANSCE 800-MeV neutron beam.

The $^{116\text{m}}\text{In}$ activation rates of the 18 spheres (6 rBSs and 12 BSEs) were unfolded using the ^{252}Cf and $^{241}\text{AmBe}$ sources corrected response functions and the LANSCE 800-MeV neutron beam-corrected response functions, respectively, and the unfolded spectra are shown in Figure 2, which showed good agreement with the TOF-measured spectrum and the rBS unfolded spectrum (<20 MeV).

DISCUSSION

The sources of uncertainty in the MCNPX-calculated response function are statistical, neutron cross section and geometry modelling uncertainties. The statistical uncertainty was less than 1% for most energy groups. The geometry of the rBSs is simple and the modelling error is minor. So the response function's uncertainty is mainly from the neutron

Figure 5. Some of the work response functions of $^{116\text{m}}\text{In}$ in the Au-In BSE system (after ^{252}Cf and $^{241}\text{AmBe}$ source and LANSCE 800-MeV beam corrections).

cross-sectional uncertainty. In this study, the cross-sectional library, LA150, for neutron transport and two different neutron activation cross-sectional libraries for activation rate calculation were used (ENDF and ACTL for Au and ACTL and 532DOS2 for In foil). The big difference in the response function is caused by the different activation cross-sectional libraries, which was demonstrated in Figure 4.

After correction using neutron sources of ^{252}Cf and $^{241}\text{AmBe}$ of known strengths, the difference of the response functions calculated using different cross-sectional library, which is caused by the resonance cross-sectional uncertainty, is eliminated. However, the uncertainty of the neutron source strength, the uncertainty in the determination of activation rate and the uncertainty in the determined RR&AS neutron spectrum will be introduced in the response functions. The geometry of BSEs is more complex than the regular spheres and may have more geometry uncertainty due to incomplete modelling of air gap and homogeneity assumption of the metal shell (copper powder, tungsten granulates and melted lead were filled in the respective aluminum shell containers, so some degree of inhomogeneity may exist). This geometry modelling uncertainty is also included in the correction factors.

The correction of the response functions >20 MeV using the LANCE 800-MeV neutron beam is mainly an adjustment of neutron multiplication of the metal shells. The increase of the response function with neutron energy is less sharp than the prediction of MCNPX calculation. It is unclear how much of this is caused by errors in the modelling and how much is caused by errors in the interaction cross sections. But the corrected response functions for the Au-In foil-based BSE have provided a set of working response functions for measuring high-energy neutron spectrum for intense and pulsed beams.

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