LLNL-JRNL-749383

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April 11, 2018

Journal of Radioanalytical and Nuclear Chemistry

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NOTICE THIS IS PART OF A SPECIAL ISSUE!

SI: Methods and Applications of Radioanalytical Chemistry (MARC XI)

MARC XI Log Number: 477

Retrospective Neutron Spectrum Determination of a (30 MeV D, Be) Source using the Multi-Foil Activation Technique and STAYSL-PNNL

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Abstract

Retrospective characterization of a (30 MeV D, Be) neutron source was performed employing multi-foil activation and STAYSL-PNNL. Experimental reaction rates were calculated from gamma spectroscopy measurements of irradiated foils and MCNP provided the guess spectrum. Adjusted spectra were evaluated through activation calculations for a stainless-steel target using FISPACT-II. Adjusted spectra showed limited dependence on the dosimetry reactions and provided minor improvements in activation calculations. Omitting reflected neutrons in the guess spectrum generated poor activation results and the limited number of dosimetry reactions introduced doubt in the adjusted spectra. A dedicated neutron spectrometry experiment and a more detailed simulation is required.

Keywords

STAYSL-PNNL; neutron spectral adjustment; foil activation; d+Be neutron source; MCNP; FISPACT-II

Introduction

The Crocker Nuclear Laboratory at the University of California-Davis campus (UC Davis) maintains a 76-inch cyclotron capable of producing neutrons through the d+Be family of neutron sputtering reactions. To date, little work has been done to characterize this neutron source, and current experiments are relying on legacy data [1]. Neutron production from a d+Be source at high deuteron beam energies produces fast neutrons through the (d, n), (d, 2n), (d, np), and (d, 2np) reactions on Be-9, with the average neutron energy being slightly less than 0.4*E_d, where E_d is the deuteron beam energy [2, 3].

A well characterized neutron spectrum is required for studying radiation effects and performing other nuclear physics experiments due to the energy dependence of neutron cross sections. The use of simulations with deterministic or stochastic computational codes also requires a validated neutron energy distribution as an input parameter to obtain meaningful results. The act of determining the neutron energy distribution, termed neutron spectrometry, may be performed through the analysis of recoil nuclei from neutron scattering, reaction-induced charged particle emission, or threshold material activation, among other methods [4, 5]. The multi-foil activation technique is often chosen for its convenience and applicability to all neutron fields. However, the multi-foil activation technique requires the proper selection of activation materials and computational methods to solve the inverse neutron spectrum unfolding problem [3, 5–8]. Iterative and least-squares methods of solving the unfolding problem also require an initial guess spectrum, usually provided by Monte Carlo codes.

This work used the STAYSL-PNNL suite of modules to perform retrospective least-squares spectral adjustment based on foil activation experiments performed using the UC Davis (30 MeV D, Be) neutron source [9]. The initial guess neutron spectrum was obtained from a simple model of the irradiation setup using the Monte Carlo N-Particle version 6.1 (MCNP) radiation transport code [10]. STAYSL-PNNL was selected because of its ability to handle neutron energies above 20 MeV, a situation that is encountered when using high deuteron beam energies. The objective of this work was to determine if the simplicity of the simulation and the limited number of activation foils was sufficient for retrospective determination of the neutron energy distribution, or if a dedicated neutron spectrometry experiment is required. The adjusted neutron spectra were then evaluated in activation calculations for a stainless-steel target using the FISPACT-II code [11].

Experimental

Foil Irradiations

A series of four foil activation experiments were conducted using the d+Be source at UC Davis with Al, Au, Co, Cu, Fe, Ni, and W activation foils obtained from Shieldwerx [12]. A stainless-steel type 304 (SS) foil from Goodfellow was included in one run as well [13]. Each experiment used a beam energy of 30 MeV and beam current of 10 µA. Irradiation times were either 4.0 h or 7.0 h in duration. A summary of the experimental parameters is given in Table 1. Gamma-ray spectroscopy was used to measure activities of selected reactions, listed in Table 2, found in the International Reactor Dosimetry and Fusion File (IRDFF) library version 1.05 [14]. Measurements were performed on a suite of HPGe detectors and analyzed using GAMANAL, with each sample having multiple independent counts and decay times up to 17 d [15]. The independent counts were used to calculate a weighted average of the activity at the end of irradiation for each reaction product listed in Table 2.

Table 1 Foils, irradiation time, and beam characteristics for each activation experiment.

Run #	Beam Energy (MeV)	Beam Current (uA)	Run Time (h)	Foils								
				Al	Au	Co	Cr	Cu	Fe	Ni	W	SS-304
Run 1	30	10	4.00	X	X			X	X	X		
Run 2			4.00	X	X			X	X	X		
Run 3			7.00		X	X	X		X	X		
Run 4			7.00		X	X	X				X	X

Table 2 Dosimetry reactions from IRDFF-1.05 for the foils used in this work.

Foil	Target	Ejectile	Residual	Foil	Target	Ejectile	Residual
Al	Al-27	a	Na-24	Co	Co-59	2n	Co-58
Fe	Fe-54	a	Cr-51		Co-59	3n	Co-57
	Fe-58	g	Fe-59		Co-59	g	Co-60
Ni	Ni-60	p	Co-60		Co-59	p	Fe-59
Cu	Cu-63	a	Co-60		Co-59	a	Mn-56
W	W-186	g	W-187	Au	Au-197	2n	Au-196
					Au-197	g	Au-198

MCNP Guess Spectrum

MCNP version 6.1 was used to model the d+Be neutron source at UC Davis. The model consisted of a Be-9 cylinder with a Cu jacket, the target frame, and the target foil. The Be-9 cylinder was 2.54 cm long with a 0.635 cm radius. The Cu jacket was 20 µm thick on all sides of the cylinder. The target frame was modeled as a rectangular parallelepiped 5.08 cm high, 3.71 cm wide, and 0.795 cm thick, with a 1.34 cm radius cutout through the center. The sample foil had a radius of 0.635 cm and was 0.0254 cm thick. The target frame composition was modeled as muscovite [16]. Natural element descriptions were used for the Be cylinder and Cu jacket, while the target foil was treated as a void. The geometry of the model is shown in Fig. 1, with all components aligned to share the central axis of the Be-9 cylinder. The physical characteristics of the room in which this neutron source is housed were not considered in this work. The consequence of this is that there is no estimation of the contribution of reflected neutrons in this simulation.

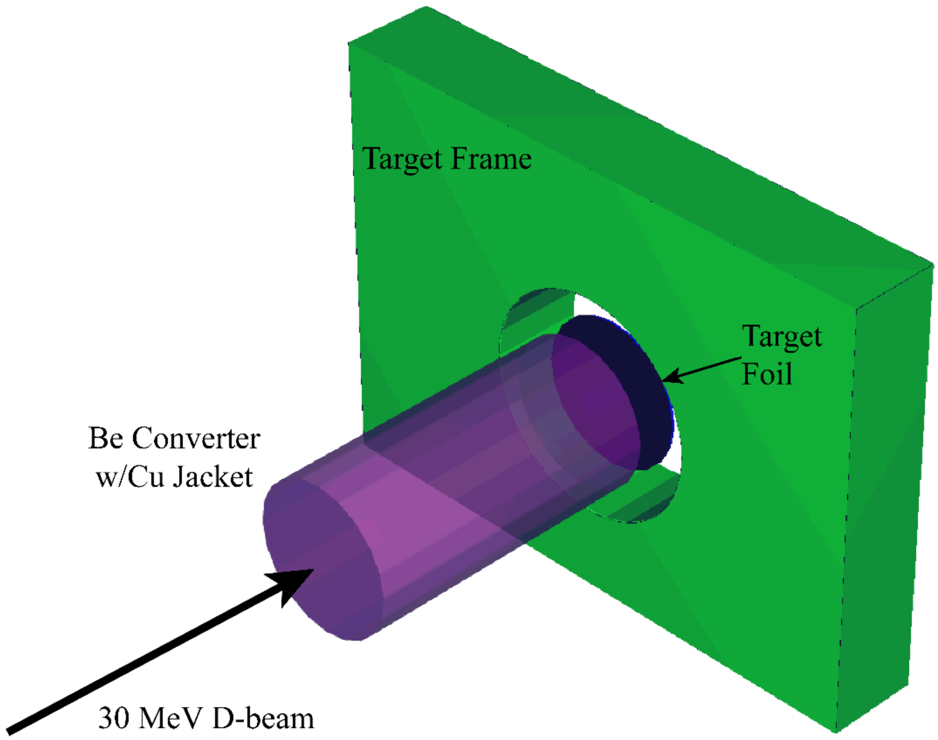

Fig. 1 A simple version of the activation geometry for the d+Be source at the University of California Davis Crocker Nuclear Laboratory showing the Be neutron converter with Cu jacket, muscovite target frame, and void target foil.

The MCNP simulation used a monodirectional and monoenergetic deuteron source at 30 MeV and ran 5×10^9 source particles to ensure good statistics. The CEM03.03 and LAQGSM03.03 physics models were used to handle deuteron interactions. The F4 tally was used to tabulate the neutron spectrum as seen by the target foil, using 1000 equal unit lethargy bins up to 60 MeV. The simulated neutron spectrum is shown in Fig. 2 as the neutron energy group probability distribution, along with the legacy data for comparison.

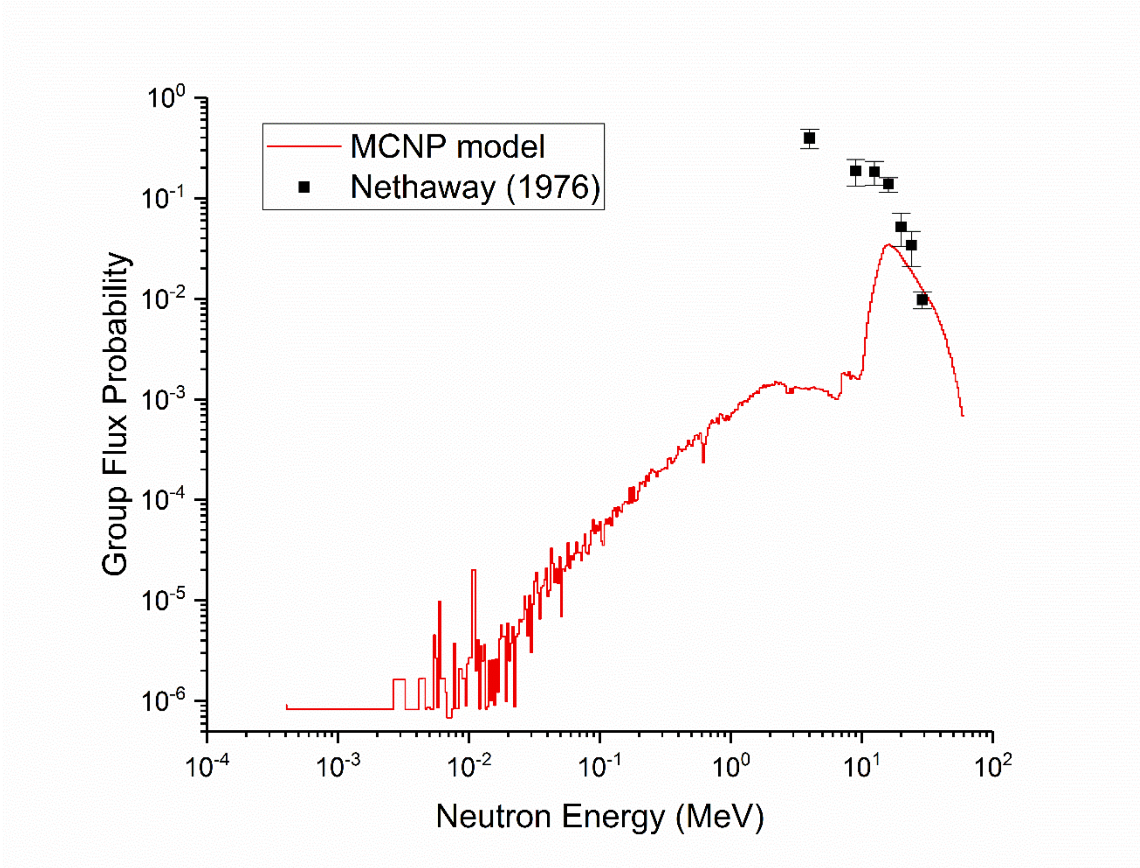

Fig. 2 The simulated neutron spectrum (red) from an MCNP simulation using an F4 tally with 1000 equal unit lethargy bins up to 60 MeV and the legacy data for the d+Be source [1]; MCNP error bars have been omitted for clarity.

Spectral Adjustment and Activation Calculations

The STAYSL-PNNL software suite is a collection of modules used to determine self-shielding and irradiation history correction factors, calculate reaction rates based on experimentally measured activities, and then perform spectral adjustment on a user supplied guess spectrum using a least-squares approach [9]. The suite of modules utilizes the cross-section and covariance data available in the IRDFF-1.05 library. The irradiation characteristics detailed in Table 1 were used as input to the Beam Correction Factors (BCF) module to correct for irradiation history. The SigPhi Calculator used the BCF output, experimental specific activities, gamma self-shielding factors, and foil composition data to calculate reaction rates relative to the number of target atoms for each reaction. The

SHIELD module used foil and irradiation characteristics to calculate neutron self-shielding factors. This work used the pre-compiled 725 energy group structure cross-section and covariance data files, based on IRDFF-1.05, generated from the NJOY99/NJpp modules [17]. Output from the SigPhi Calculator and the SHIELD module were used with the 725 group data files and the MCNP guess spectrum in Fig. 2 as input for the STAYSL-PNNL least-squares adjustment module.

The FISPACT-II code was used to evaluate the MCNP guess spectrum and the STAYSL-PNNL adjusted spectra through activation calculations on the SS foil. The guess and adjusted spectra were converted to the 709 energy group structure in FISPACT-II and used the JEFF-3.2 library for activation calculations [18]. The irradiation in FISPACT-II was defined to match Run 4 which included the SS foil. The irradiation time was 7.0 h and the flux magnitude was calculated to be $3.43(17)\times10^{10}$ n cm^{-2} s^{-1}, based on the n/d production ratio determined by MCNP and the deuteron beam current. Calculated/Experiment ratios of activation rates were used to compare the different spectra.

Results and Discussion

Adjusted Neutron Spectra

All STAYSL-PNNL results had large chi-squared values, signaling low confidence in the results. This was likely caused by the limited scope of the reactions used as constraints for the least-squares adjustments. Additional reactions were available from IRDFF-1.05, based on the foils used, but are subject to interference from multiple reaction pathways to common activation products in natural abundance element foils. This issue is more pronounced for higher energy neutron sources, as discussed by Greenwood [19]. The retrospective nature of this work did not allow for the pre-selection of a sufficient number of dosimetry reactions and the work was further limited by what was actually detected. To appropriately solve the neutron unfolding problem, a proper number of dosimetry reactions with reaction thresholds covering the anticipated energy range is required. Based on the current version of the IRDFF library, isotopically pure target foils are also required to increase the number of reactions available for selection.

The initial MCNP guess spectrum and adjusted neutron spectra from each run are plotted in Fig. 3 as group flux probabilities using the 725 energy group structure. All spectra show the same general shape, with the adjusted spectra being lower in magnitude than the initial MCNP guess. At lower neutron energies, 10^{-4}-10^{-2} MeV, the adjusted spectra probabilities are comparable to the initial MCNP spectrum. Also, the adjusted spectra are nearly identical and show two strong peaks at neutron energies of approximately 6 keV and 11 keV. These peaks are strongest for Run 3 (green), followed by Run 2 (yellow), then by Run 1 (blue), and finally Run 4 (gray). Further comparison between spectra showed that the average neutron energy shifted slightly downward from 18.948(16) MeV in the MCNP spectrum to an average of 17.35(9) MeV in the adjusted spectra. The average neutron energy for all adjusted spectra agreed within 1-sigma uncertainty.

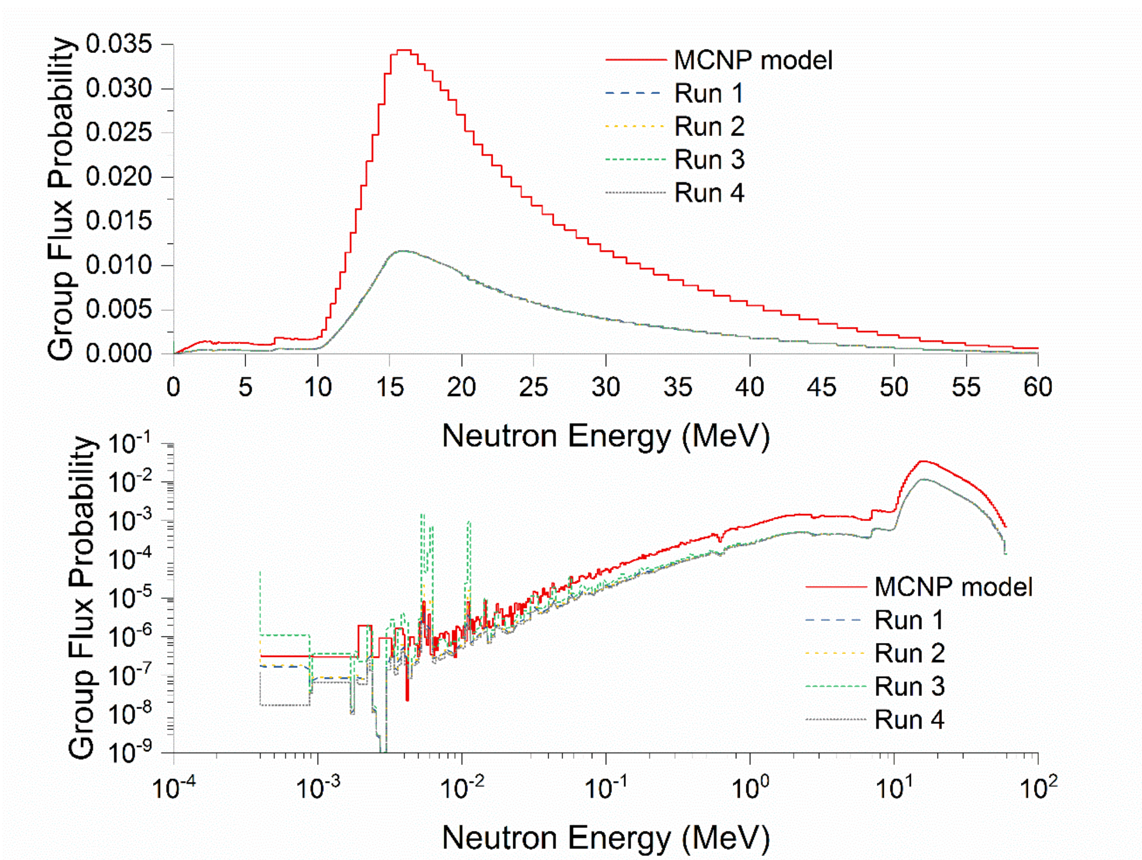

Fig. 3 Comparison of neutron energy group flux probability distributions on linear (top) and log (bottom) scales for the MCNP guess spectrum (red, line), and the STAYSL-PNNL adjusted spectra for Run 1 (blue, dash), Run 2 (yellow, dot), Run 3 (green, short dash), and Run 4 (gray, short dot).

The cumulative probabilities of each run, along with the MCNP guess spectrum, are plotted in Fig. 4. The MCNP spectra shows a slightly broader neutron energy distribution than the adjusted spectra, with a greater percentage of neutron energies falling outside the 10-20 MeV range. The cumulative distributions are additional evidence of just how similar each of the adjusted spectra are. The similarities between runs show that differences in the limited number of dosimetry reactions used as constraints each in run do not significantly influence the adjusted spectra, placing more importance on the quality of the initial guess spectrum. However, more work is needed to confirm this point since the chi-squared values were unacceptable.

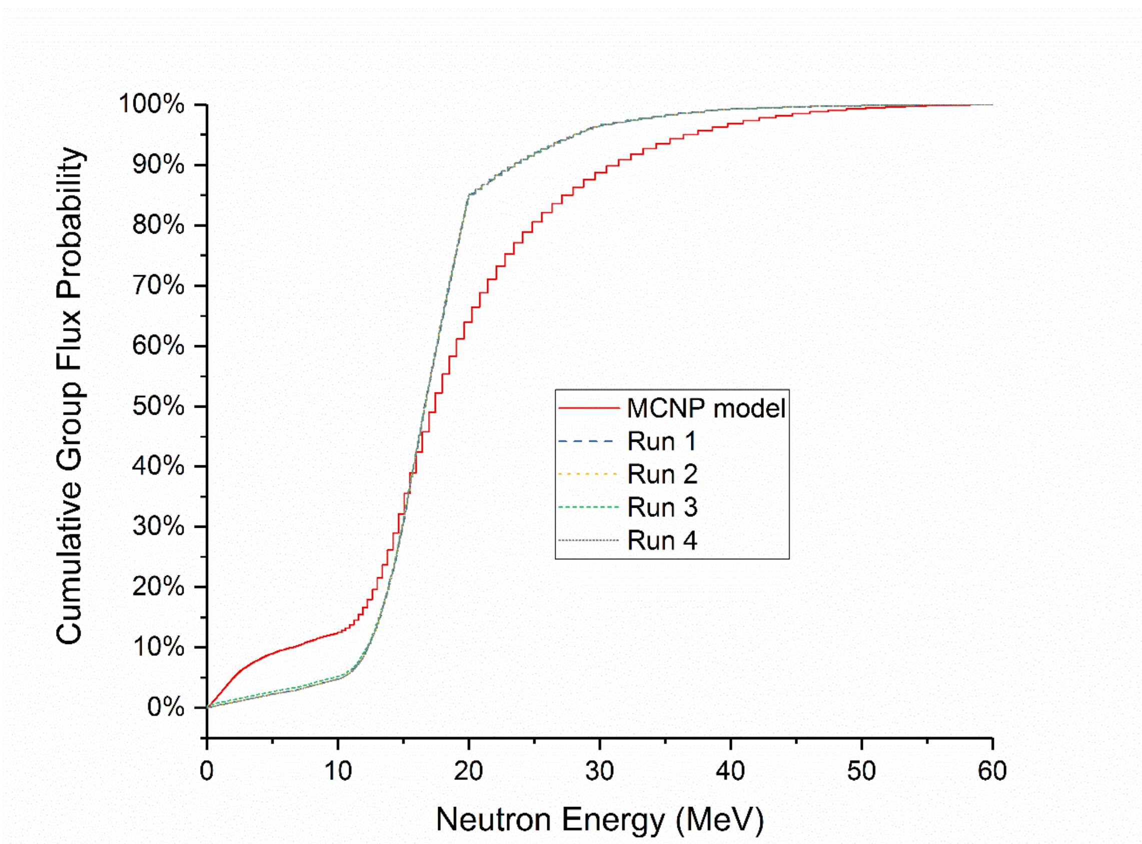

Fig. 4 Cumulative group flux probability plots for the MCNP guess spectrum (red, line) and the STAYSL-PNNL adjusted spectra for Run 1 (blue, dash), Run 2 (yellow, dot), Run 3 (green, short dash), and Run 4 (gray, short dot).

FISPACT-II Calculations

The use of FISPACT-II for activation calculations required re-binning of the MCNP guess spectrum and STAYSL-PNNL spectra to the 709 energy group structure. Changing the energy group structure caused a minor distortion of the adjusted spectra in the 1-10 MeV range for the adjusted spectra. An example of the degree of distortion is shown in Fig. 5 for Run 3. Additionally, the probabilities of the lowest energy neutrons were elevated when re-binning from the STAYSL-PNNL 725 group structure to the FISPACT-II 709 group structure. The cross sections of individual reactions should be examined to determine any effects of the re-binning process. The MCNP guess spectrum also showed some distortion due to re-binning, this time in the 10-30 MeV range. The result was that the most probable neutron energy was shifted a few MeV higher.

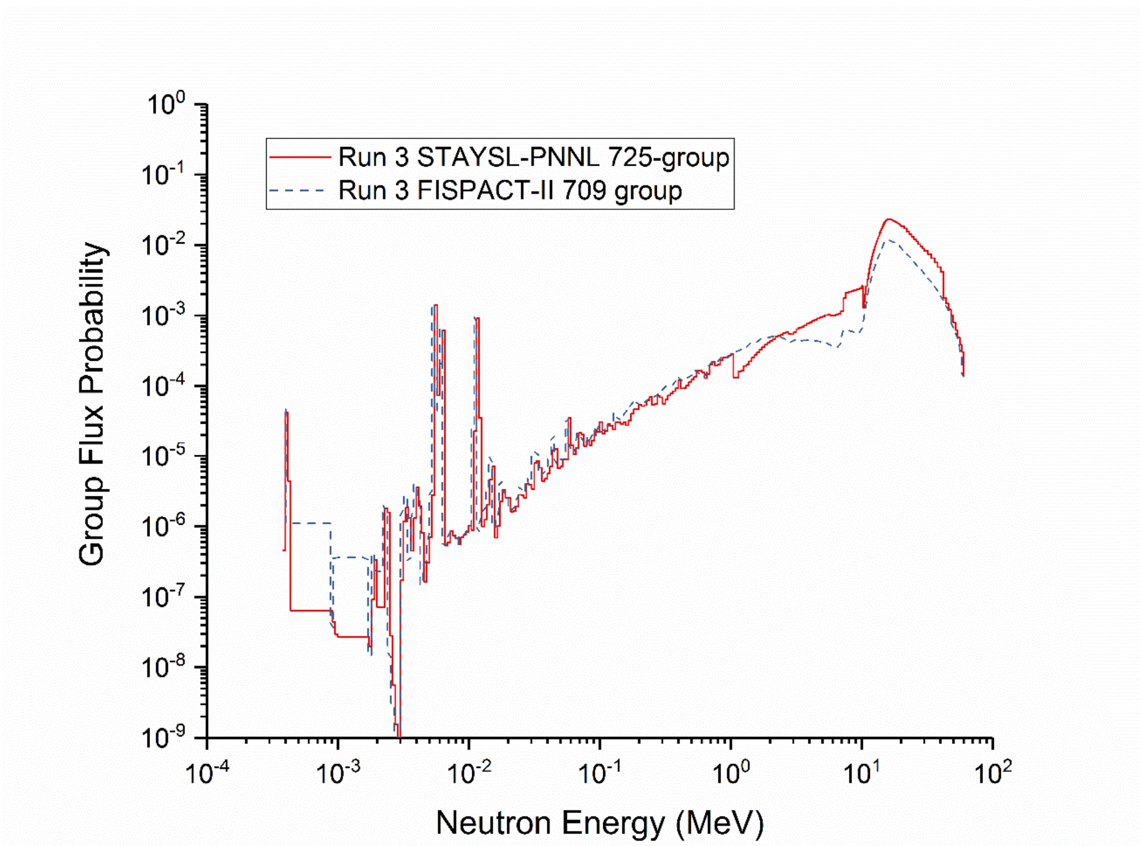

Fig 5 Comparison of the Run 3 STAYSL-PNNL adjusted spectra in 725 energy groups (red, line) and the spectrum re-binned to the FISPACT-II 709 energy group structure (blue, dash).

Calculated/Experiment (C/E) results for the activation of the SS foil in Run 4 are shown in Table 3, with 1-sigma uncertainty, for each of the spectra discussed in this work. C/E values were calculated by taking the ratio of saturation activities (reaction rates) from FISPACT-II calculations and experimental gamma spectroscopy measurements. The C/E results based on the MCNP spectrum were consistently further away from the ideal value of 1 than those for the adjusted spectra. This indicated that spectral adjustment with STAYSL-PNNL did improve upon the initial neutron energy spectrum. Since the neutron distributions for all adjusted spectra were very similar, it is no surprise that the C/E values for Runs 1-4 were nearly identical.

Table 3 Calculated/Experiment ratio values with 1-sigma uncertainty from FISPACT-II calculations for activation of a stainless-steel foil.

Nuclide	MCNP	Run 1	Run 2	Run 3	Run 4
Co-56	0.19(8)	0.35(15)	0.35(15)	0.35(15)	0.35(15)
Co-57	0.276(8)	0.453(13)	0.453(13)	0.451(13)	0.453(13)
Co-58	0.116(4)	0.153(6)	0.153(6)	0.153(6)	0.153(6)
Co-60	0.228(10)	0.323(16)	0.323(16)	0.322(16)	0.323(16)
Cr-51	0.513(9)	0.580(15)	0.580(15)	0.578(15)	0.580(15)
Fe-59	0.093(8)	0.110(13)	0.110(13)	0.111(13)	0.110(13)
Mn-52	4.91(3)	1.805(10)	1.085(10)	1.806(10)	1.803(10)
Mn-54	0.361(6)	0.322(8)	0.322(8)	0.321(8)	0.322(8)
Mn-56	0.16(3)	0.21(4)	0.21(4)	0.20(4)	0.21(4)
Ni-57	0.300(9)	0.512(15)	0.512(15)	0.510(15)	0.513(15)
V-48	1.623(11)	0.608(4)	0.608(4)	0.608(4)	0.607(4)

Even though there was improvement in the C/E values from the MCNP spectrum to the adjusted spectra, they are still significantly different from the ideal value of 1. The poor C/E values of the FISPACT-II calculations highlight the need for a dedicated neutron spectrometry experiment. Additionally, the simulation needs to be expanded to include a definition of the room in which the neutron source is housed which will provide an estimate of the contribution of thermal/epithermal neutrons reflected by the room environment. A better estimation of the thermal/epithermal neutron contribution would lower the average neutron energy, providing a greater probability of low energy neutrons. The resulting effect would be a greater portion of neutrons available at energies that have larger cross-section values, thereby increasing the reaction rate of a given activation product.

Another concern is that the limited number of reactions used in this work did not provide sufficient constraints for the neutron spectrum unfolding problem, as evidenced by large chi-squared values. Additionally, limitations of the nuclear data available for the FISPACT-II may also contribute. Cross-section data may have contributed to errors since the data for many of the reactions producing the activation products in Table 3 do not exceed 20 MeV. For the adjusted spectra, the portion of the neutrons above 20 MeV is approximately 15% and may affect the results.

Conclusions

This work used multiple computation codes in an effort to retrospectively characterize the neutron spectrum of the (30 MeV D, Be) neutron source at the University of California-Davis Crocker Nuclear Laboratory through the multi-foil activation technique. An MCNP simulation provided an initial guess spectrum, the STAYSL-PNNL suite of modules generated adjusted spectra using a least-squares approach to fit experimentally measured activities, and FISPACT-II was used to evaluate the adjusted spectra through activation calculations, which were compared against experimental results.

Comparison of the STAYSL-PNNL adjusted spectra showed little dependence on the selected dosimetry reactions, with all four runs being nearly identical. The major difference between the adjusted spectra and the MCNP guess spectrum was the emergence of strong peaks in the adjusted spectra at neutron energies of approximately 6 keV and 11 keV. The spectral adjustment process also shifted a greater number of neutrons into the 10-20 MeV range relative to the MCNP spectrum, slightly lowering the average neutron energy from 18.948(16) MeV to 17.35(9) MeV.

The C/E results for the FISPACT-II activation calculations with the adjusted spectra showed minor improvements over those for the MCNP spectrum but were still significantly different that the ideal value of 1. The simplified definition of the simulation environment was the major hindrance, which omitted any contribution of low energy reflected neutrons from the MCNP guess spectrum. This resulted in an over estimation of the high neutron energy region of the spectrum and limited nuclide production rates due to lower corresponding cross-section values. Additionally, the adjustment process only had a small number of constraints, which led to large chi-squared values and placed doubt in the adjusted spectra.

A dedicated and carefully planned experiment is required to achieve satisfactory results for the adjusted neutron spectrum. Such an experiment would involve the careful selection of single reaction pathways for activation products in isotopically pure foils, due to current limitations in the IRDFF-1.05 library. This future experiment would be complemented by a fully defined simulation environment which includes the room geometry for the neutron source, allowing for an estimation of the reflected neutron contribution.

Acknowledgements

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. Nethaway DR, Van Konynenburg RA, Adams TM (1976) Measurement of the neutron spectrum on a thick beryllium target from the reaction of 30-MeV deuterons. Natl Tech Inf Serv UCID52024.
2. Wei Z, Yan Y, Yao ZE, et al (2013) Evaluation of the neutron energy spectrum, angular distribution, and yield of the 9Be(d,n) reaction with a thick beryllium target. DOI: 10.1103/PhysRevC.87.054605.
3. Stefanik M, Bem P, Majerle M, et al (2017) Neutron spectrum determination of d(20)+Be source reaction by the dosimetry foils method. DOI: 10.1016/j.radphyschem.2017.03.029.
4. Brooks FD, Klein H (2002) Neutron spectrometry-historical review and present status. Nucl Instruments Methods Phys Res A 476:1–11.
5. Vagena E, Theodorou K, Stoulos S (2018) Thick-foils activation technique for

neutron spectrum unfolding with the MINUIT routine—Comparison with GEANT4 simulations. DOI: 10.1016/j.nima.2018.01.025.

6. Maeda S, Tomita H, Kawarabayashi J, Iguchi T (2011) Fundamental study on neutron spectrum unfolding using maximum entropy and maximum likelihood method. Prog Nucl Sci Technol 1:233–236.

7. Hayes JW, Finn E, Greenwood L, Wittman R (2014) Characterization of a Thermo Scientific D711 D-T neutron generator located in a low-scatter facility. DOI: 10.1016/j.nima.2013.11.023.

8. Shahabinejad H, Hosseini SA, Sohrabpour M (2016) A new neutron energy spectrum unfolding code using a two steps genetic algorithm. DOI: 10.1016/j.nima.2015.12.028.

9. Greenwood LR, Johnson CD (2013) User Guide for the STAYSL PNNL Suite of Software Tools. PNNL-22253, Pacific Northwest National Laboratory, Richland, Washington.

10. Goorley T, James M, Booth T, et al (2012) Initial MCNP6 Release Overview. DOI: 10.13182/NT11-135.

11. Sublet J-CC, Eastwood JW, Morgan JG, et al (2015) FISPACT-II User Manual. In: Tech. Rep. UKAEA-R(11)11 Issue 7. http://fispact.ukaea.uk/. Accessed 23 Jan 2018.

12. Shieldwerx SWX-500 Series Activation Foils Specification Sheet. http://www.shieldwerx.com/assets/swx-5xx.pdf. Accessed 27 Mar 2017.

13. Goodfellow (2014) Stainless Steel - AISI 304 - Foil. goodfellowusa.com. Accessed 7 Mar 2017.

14. Zsolnay EM, Noy RC, Nolthenius HJ, Trkov A (2012) Summary description of the new International Reactor Dosimetry and Fusion File (IRDFF release 1.0). INDC(NDC)-0616. https://www-nds.iaea.org/publications/indc/indc-nds-0616-1.pdf. Accessed 24 Jan 2018.

15. Gunnink R, Niday JB (1972) Computerized quantitative analysis by gamma-ray spectroscopy (Gamanal), Vol. 1-4, UCRL-51061.

16. Muscovite Mineral Data. http://webmineral.com/data/Muscovite.shtml#.Wq4YKJdlAuU. Accessed 15 Jan 2018.

17. MacFarlane RE, Muir DW (1994) The NJOY Nuclear Data Processing System: Version 91. LA-12740-M, DOI: 10.2172/10115999.

18. Nuclear Energy Agency (2014) JEFF-3.2 evaluated data library. http://www.oecd-nea.org/dbforms/data/eva/evatapes/jeff_32/. Accessed 10 Jan 2018

19. Greenwood LR, Simakov SP, Trkov A (2017) INDC International Nuclear Data Committee testing and improving the International Reactor Dosimetry and Fusion File (IRDFF). INDC(NDS)-0731. https://www-nds.iaea.org/publications/indc/indc-nds-0731.pdf. Accessed 5 Feb 2018.