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Matthew R. Halstead

Sangjin Lee

Indiana University - Bloomington

James C. Petrosky

Air Force Institute of Technology

Abigail A. Bickley

Indiana University - Bloomington

Paul Sokol

Indiana University - Bloomington

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Union of Compact Accelerator-Driven Neutron Sources I & II

Neutron energy spectrum characterization on TMR-1 at the Indiana University neutron source

Matthew R. Halstead^{a*}, Sangjin Lee^b, James Petrosky^a, Abigail Bickley^a, Paul Sokol^{b,c}

^a*Air Force Institute of Technology, 2950 Hobson Way, Wright-Patterson AFB, OH 45433, USA*

^b*Center for the Exploration of Energy and Matter, Indiana University, 2401 N Milo B. Sampson Lane, Bloomington, IN 47408, USA*

^c*Department of Physics, Indiana University, 2401 N Milo B. Sampson Lane, Bloomington, IN 47408, USA*

Abstract

The energy spectrum of the Neutron Radiation Effects Program (NREP) beam line, Target-Moderator-Reflector-1 (TMR-1), at Indiana University has not been previously characterized. The facility has a unique proton source with variable pulse length (15-600 μ s) and energy (13 MeV). Thus, it can produce a unique and tailored neutron beam when incident on a beryllium target. Through a combination of MCNP-X particle simulations, neutron activation experiments, and application of a spectrum unfolding code (SAND-II), the neutron source is characterized. Eight activation foils and wires were irradiated in the target area and the gamma activity measured. This information was used in an unfolding code, SAND-II, to deconvolve the spectrum, using the MCNP simulations as a basis for the spectral fitting.

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Keywords: NREP, MCNP-X, neutron activation analysis, SAND-II

1. Introduction

The high cost of maintenance and security of nuclear materials has led to an increasing desire to replace reactor-based neutron sources with non-uranium based alternatives. These sources often have

* Corresponding author. Tel.: 717-440-1287; fax: 937-255-2921.

E-mail address: Matthew.Halstead@afit.edu.

added benefits such as the ability to produce a hardened neutron spectrum and short pulse width, without the concerns of proliferation or maintenance of radioactive sources.

The Neutron Radiation Effects Program (NREP) beam line of the Low Energy Neutron Source (LENS) at the Indiana University Center for Exploration of Energy and Matter (CEEM) is one such alternative that is specifically designed for short pulse neutron radiation effects research. The beam line produces neutrons via (p,n) reactions in natural beryllium foil targets, with neutron energies up to 11 MeV. The facility is designed so that the proton beam can be incident on one of two Target-Moderator-Reflector (TMR) stacks. The neutron scattering target, TMR-2, uses a series of water and cryogenic methane moderators to produce a high flux of thermal and cold neutrons primarily used in condensed matter and materials science neutron research [1]. The radiation effects target, TMR-1, uses a water cooled beryllium target and a series of reflectors to produce a hard spectrum of neutrons for electronic radiation experiments. The TMR-1 cavity spectrum has not been characterized and is the subject of this paper.

1.1. Facility Description

The proton source at the IU CEEM produces protons by stripping electrons from hydrogen gas. The low-energy protons are accelerated through a 3 MeV radio-frequency quadrupole and two drift tube linear accelerators where they reach an energy of 13 MeV. Typically, the beam is operated at 20 mA, with a repetition rate of 20 Hz and an approximately square proton pulse with a width from 15 to 600 μ s.

Neutrons are produced via a (p,n) reaction in a natural beryllium target. The target foil is 1.2 mm thick and was chosen to maximize neutron production in the beryllium while minimizing the possibility of proton capture and subsequent hydrogen gas creation. The range of 13 MeV protons in beryllium is calculated to be 1.28 mm. The target is cooled with a continuous flow of water to facilitate maximum heat transfer during full-power beam operation, which would otherwise fracture the target due to thermal expansion.

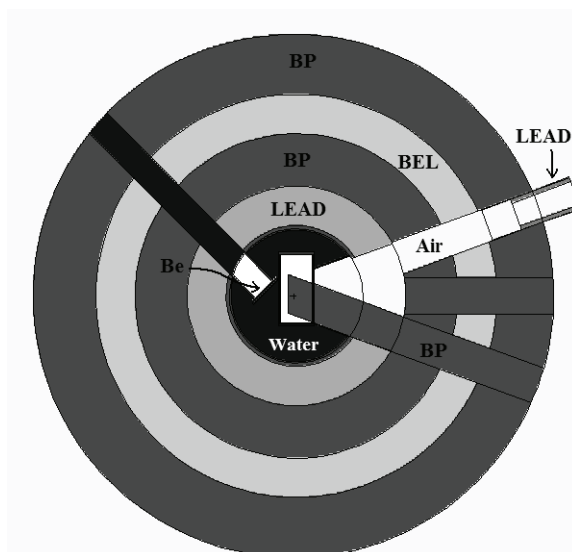


Fig. 1. TMR-1 simulation schematic in MCNP-X radiation transport code. “BP” signifies 2.142% borated polyethylene, “BEL” is short for borated epoxy lead bricks, and “Be” shows where the beryllium target is positioned.

The neutrons born in the beryllium target must travel through several materials layers before entering the experimental sample irradiation cavity. The first material traversed is a ~6 mm water layer, which is the primary heat removal mechanism for the target. Next is a thin aluminum layer, which is the water containment. This is followed by a more substantial water layer (~10 cm) which acts as a reflector, contained in a thin aluminum containment vessel. These layers of water instigate considerable neutron energy down-scattering, thus softening the neutron spectrum. The water reflector can be drained if necessary, but it was not during the present measurements.

The TMR stack is made from concentric cylinders of lead and borated plastics. It is designed to minimize the number of neutrons and gammas escaping the sample cavity. It has the capability of fitting three beam ports; however, one is currently filled with a plastic plug. Of the two that remain, one has a lead collimator and is used for neutron radiography experiments. The second has a plastic stringer system that facilitates the placement of items to be irradiated, such as neutron activation foils or electronic components. These are located approximately 6 cm from the target.

1.2. Characterization Approach

The present research follows two paths. First, a model of the neutron beam facility was developed using the particle transport code, MCNP-X (ver. 2.6.0) [2], which provided an estimate of the expected spectrum. The spectrum was then measured by multiple-element neutron activation analysis. A high-purity germanium detector with associated gamma measurement software was used to measure the induced gamma activity spectrum in each sample material. The resulting activity data provided input into a spectrum unfolding code, Spectrum Analysis by Neutron Detectors, Version 2 (SAND-II) [3].

2. Simulation

The TMR-1 beam line was modeled in the Monte Carlo N-Particle Extended (MCNP-X) radiation transport code environment. In the simulation, 13 MeV protons were normally incident on a 1.2 mm thick foil of natural beryllium. All relevant aspects of the environment were considered, to include all TMR layers and the water reflector. Interaction cross sections for all materials were based upon Evaluated Nuclear Data File (ENDF) inputs [4].

For the simulation, three point-detector flux tallies were used. One was at the center of the sample cavity, a second was along the plastic stringer beam port at 50 cm from the center, and a third was placed at 110 cm along the same line out of the beam port. Figure 1 represents a schematic of the TMR in simulation space. The beryllium target is indicated by a black arrow in the figure.

Figure 2 presents the differential neutron energy flux with an inset figure of the fractional standard deviation (FSD). Three point-detector tallies were recorded: one at the face of the stringer (labeled "0cm"), one 50 cm, and one 110 cm along the stringer axis. The spectrum recorded by the 0 cm flux tally was used as the initial guess for the SAND-II unfolding routine. The FSD for neutron energies less than 3.4×10^{-9} MeV is greater than the recommended value of 0.05. Therefore, the perturbation of the spectrum in this range is more significant than other energy regions.

3. Experiment

Neutron activation experiments were carried out on-site using high purity wires and foils with threshold reactions that covered a majority of the expected spectrum. Five wires/foils were positioned on the plastic stringer face (~9 cm from the Be target) at the locations presented in Figure 3. These locations were selected in order that the spatial variation of the neutron energy spectrum could be determined.

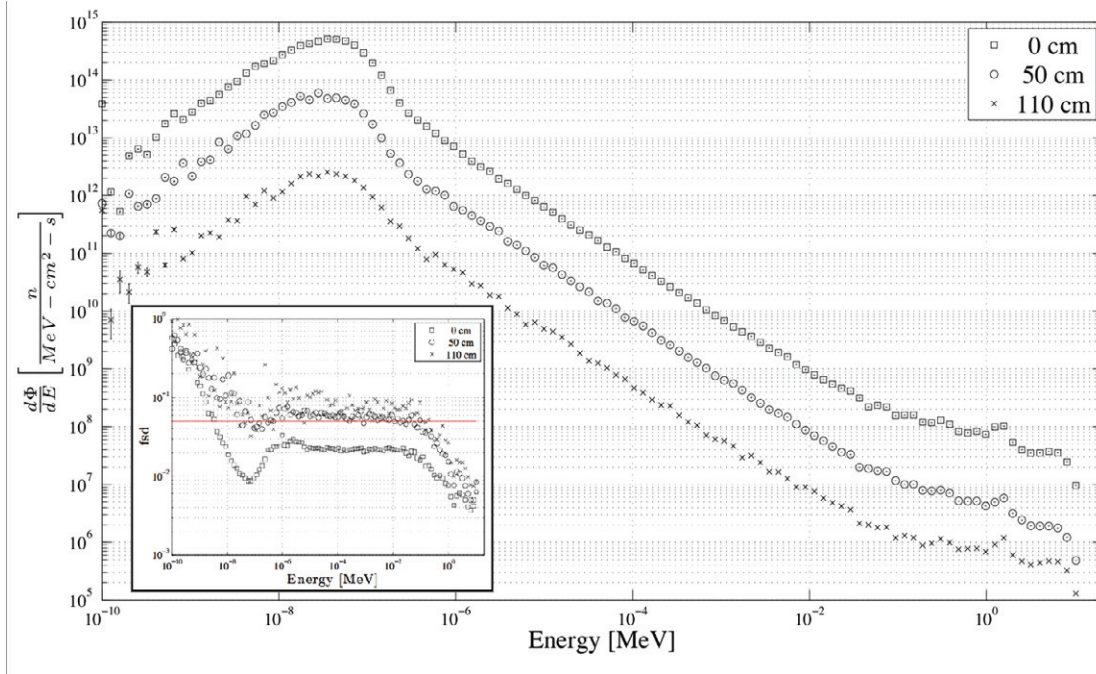


Fig. 2. MCNP-X results after simulating 3.7×10^8 protons incident on the beryllium target and measured at 0, 50, and 110 cm from the center of the cavity. INSET: Plot of fractional standard deviation for the simulation.

3.1. Gamma Detector Calibration

All the materials irradiated had at least one daughter isotope that decayed by gamma emission. To measure the daughter products' radioactivity, a Canberra model GC7020 high-purity germanium (HPGe) detector was used. Before beginning the neutron activation experiments, the gamma detection system was calibrated. First, a 1-hour background count measurement was taken and the background count subtracted from each subsequent measurement. Then, a 1-hour energy calibration was done using a NIST-traceable tri-nuclide source made of two isotopes of europium and one of cesium. Finally, a detector efficiency calibration was accomplished. The results of these three calibrations were then used when measuring the daughter products' gamma emissions.

3.2. Neutron Activation Analysis

Each sample was irradiated for 30 to 120 minutes. The various reactions considered in this research are presented in Table 1. The materials labeled with *N/A* as the threshold energy are considered full-spectrum rather than threshold detectors. The Qtool calculator [5] was used to compute all threshold energies.

After irradiation, each material was removed from the sample staging and was suspended either 8 or 23 cm from the HPGe detector face. This was required in order to minimize detector dead-time, while maximizing the total counts and obtaining the best possible statistics. For measurements, a 30 to 120 minute counting interval was used.

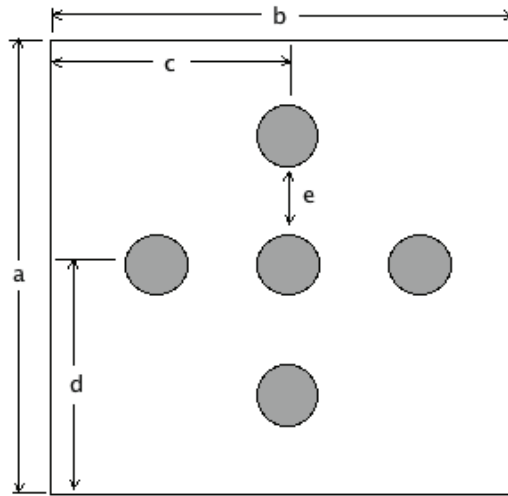


Fig. 3. The schematic for an irradiation sample of foil/wire measurements; dimensions: $a = 12.7\text{cm}$, $b = 14.6\text{ cm}$, $c = 7.3\text{ cm}$, $d = 6.8\text{ cm}$, $e = 3.8\text{ cm}$.

The isotope's total activity was calculated as the area under the associated gamma peak and calculated using a non-linear least-squares algorithm provided by the detector software to remove background effects [6]. The measured activity was then extrapolated to saturation [7] by

$$A_{\infty} = \frac{\lambda C}{(1 - e^{-\lambda t_0}) e^{-\lambda t_1} (e^{-\lambda t_1} - e^{-\lambda t_2})} \quad (1)$$

where λ is the isotopic decay constant, C is the measured number of decays during the counting time, t_0 is the irradiation time, $t_1 - t_0$ is the wait time between the end of irradiation and the beginning of counting, and $t_2 - t_1$ is the count time. The saturated activity resulting from the calculations above was used as input for the SAND-II unfolding routine.

4. SAND-II Spectrum Unfolding

The spectrum unfolding routine SAND-II utilizes an initial guess spectrum and compares the expected activity in materials, calculated using neutron cross section tables, with the measured saturated activity from neutron activation analysis. Comparing these two pieces of information, the program calculates correction factors, perturbs the guess spectrum, and then repeats the process until a solution criterion is met. For this research, two stopping criteria were used. First, the program would stop iteration if the average deviation of the difference between calculated and measured activities was less than 5%.

Failing this, the program stopped iteration if the average deviation shifted less than 1% over two consecutive iterations. Figure 4 presents the SAND-II results for the middle sample location, only. The dotted line shows the MCNP-X results used as input into SAND-II, while the circular dots show how the spectrum was perturbed based on the measured data.

Table 1. Materials selected for use in neutron activation analysis experiments. The threshold energy was calculated using Qtool.

#	Reaction	Half-life [s]	$E_{\text{threshold}}$ [MeV]	Geometry
1	$^{27}\text{Al}(n,p)^{27}\text{Mg}$	567.6	1.896	Wire
2	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	5.40×10^4	3.249	Wire
3	$^{59}\text{Co}(n,\alpha)^{56}\text{Mn}$	9.28×10^3	N/A	Wire
4	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$	1.66×10^8	N/A	Wire
5	$^{63}\text{Cu}(n,\gamma)^{64}\text{Cu}$	4.57×10^4	N/A	Wire
6	$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	1.61×10^4	N/A	Foil
7	$^{115}\text{In}(n,\gamma)^{116}\text{In}$	3.26×10^3	N/A	Foil
8	$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	9.28×10^3	2.966	Foil
9	$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	2.70×10^7	N/A	Foil
10	$^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$	3.85×10^6	N/A	Foil
11	$^{58}\text{Ni}(n,p)^{58}\text{Co}$	6.12×10^6	1.000	Wire
12	$^{109}\text{Ag}(n,\gamma)^{110\text{m}}\text{Ag}$	2.16×10^7	N/A	Foil
13	$^{197}\text{Au}(n,\gamma)^{198}\text{Au}$	2.33×10^5	N/A	Foil

The resultant spectrum displays a few key features to note. First, the entire spectrum is shifted 1 to 2 orders of magnitude higher than the MCNP-X model. Also, there is a “singularity” in the flux at the transition from thermal to epithermal, around 0.5 to 2 eV. This indicates that the thermal flux contribution is overestimated in the correction factor calculation, implying measurement error for one or more full-spectrum foils/wires. Additionally, in the epithermal region, resonances are evident that may be an artifact of the low-resolution cross section tables built into the SAND-II code structure. Finally, in the fast neutron region, another drop and rise in the flux is distinguished at 10 MeV. Since the maximum neutron energy is 11.15 MeV, the flux should go to zero at this point. However, a user cannot modify SAND-II’s preset energy range, so the program attempts to extrapolate values, which leads to the non-physical results past 11.15 MeV.

5. Conclusions

The MCNP-X simulations have provided an initial guess spectrum that facilitates an adequate first-pass understanding of the spectrum that should be seen by irradiation foils. However, it is the goal of the author to build a correlating simulation environment in GEANT4 [8], a vector Monte Carlo transport code. The results of this comparison will be disclosed in a future forum. The simulations will serve as a baseline for future modification to the TMR. In this way, the results of any changes to the geometry, materials, or configuration can be analyzed in simulation space before making any changes to the TMR in physical space.

A significant amount of delay-gamma neutron activation analysis data has been recorded, processed, and stored. Currently, since the middle location was the only data analyzed, only a small portion of this data has been used. However, once analysis is complete on this data set, all five locations will be examined, which will lead to a better understanding of the spatial variation of the neutron flux. Further, a simple method of characterization has been developed that can be used in the future to recharacterize the neutron source after any changes are made to the geometry or configuration.

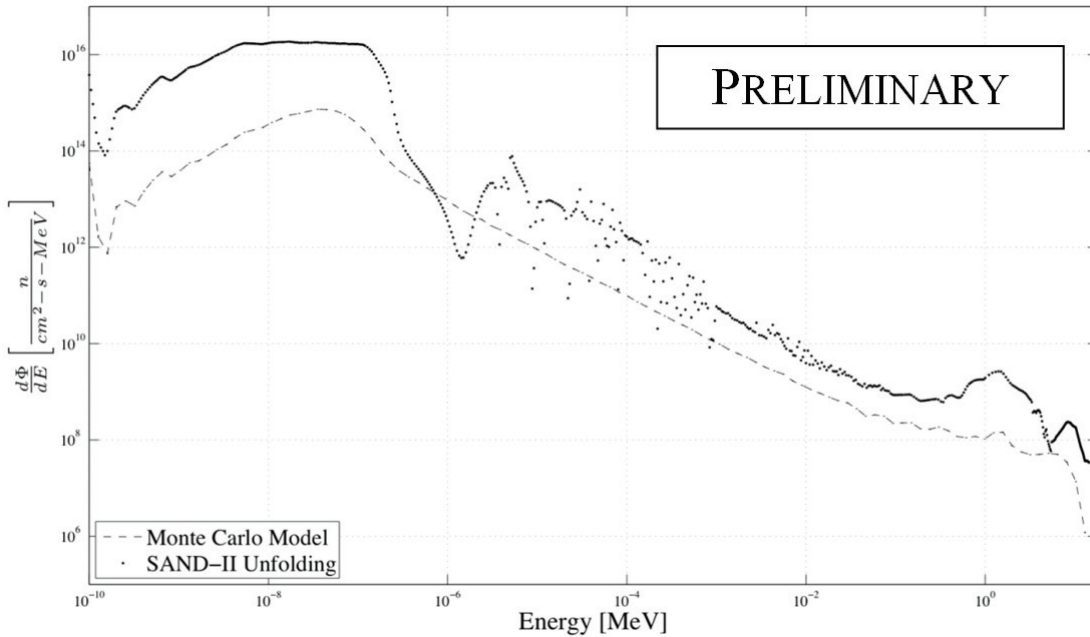


Fig. 4. Differential neutron flux spectrum for the center irradiation position.

The first step is to simulate the scenario in a particle transport code, either MCNP-X or GEANT4, and obtain an estimate of the neutron flux for the location of interest. Then, neutron activation analysis is done for a number of materials. Finally, SAND-II computes the perturbed spectrum based on the neutron activations.

It is important to note that the spectrum has not been verified, and the characterization of the spectrum presented is preliminary. Most importantly, the spectrum analysis procedure has been validated and can be used for future studies.

Acknowledgements

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