FAST NEUTRON SCATTERING MEASUREMENTS WITH LEAD

A. Youmans, J. Brown, A. Daskalakis, N. Thompson, A. Weltz, and Y. Danon
Rennselaer Polytechnic Institute
Corresponding Address
youmaa@rpi.edu

D. Barry, B. Epping , G. Leinweber, M. Rapp
Bechtel Marine Propulsion Corp., Knolls Atomic Power Laboratory, P.O. Box 1072,
Schenectady, NY, 12301-1072 USA

ABSTRACT

Fast neutron scattering from natural lead was measured at the Rensselaer Polytechnic Institute's (RPI) Gaerttner Linear Accelerator Center (LINAC). This measurement was performed using eight detectors positioned at different solid angles, with neutron events recorded in time of flight. This experiment used liquid scintillator EJ301 detectors and a tantalum photo-neutron target, which were optimized for the energy range between 0.5 MeV and 20 MeV. A digital data acquisition system was used for pulse shape analysis to discriminate gamma rays from neutrons. These measurements are used to assess the accuracy of data libraries such as the Evaluated Nuclear Data File (ENDF), Japanese Evaluated Neutron Data Library (JENDL), and the Joint Evaluated Fission and Fusion File (JEFF). The resonance structure in natural lead was clearly observed in the scattering data. Back angle measurements showed a difference between measurements and MCNP calculations using the ENDF/B-VII.1 library. These neutron scattering data will improve the accuracy of cross section libraries used in simulations with lead, such as lead criticality benchmarks, reflector materials, and Gen IV reactors with lead coolant.

KEYWORDS
elemental Pb, benchmark data, angular distribution, neutron experiment

1. INTRODUCTION

Lead is an element used in current nuclear reactors as a shielding and reflector material and in proposed advanced reactors like SSTAR and other lead-cooled reactors [1]. Studies have indicated that possible inaccuracies in the lead scattering cross section cause significant variances in calculated reactor parameters [2]. Some inelastic scattering measurements in the MeV energy range have been done at Argonne National Lab with Pb-204 [3]. However, Pb-204 is the least abundant isotope in natural lead. The RPI LINAC has the capability to measure neutron scattering in lead at discrete angles over the high energy range (0.5 MeV-20 MeV) and compare these measurements to existing evaluations such as the Evaluated Neutron Data File (ENDF) [4], Joint Evaluated Fission and Fusion (JEFF) [5], and the Japanese Evaluated Nuclear Data Library (JENDL) [6]. There are slight differences between the cross section evaluations, especially in the angular scattering distributions. Monte Carlo N-Particle 6 (MCNP6) [7] is used to perform these comparisons by modeling the scattering experiment using different data libraries for the lead sample and comparing those results to the measured data. Using this measurement to improve the evaluated nuclear data libraries would impact benchmarking simulations and decrease the error resulting from uncertainties associated with nuclear data.

2. EXPERIMENTAL SETUP

A series of four different experimental setups were used to get a comprehensive analysis of the accuracy of lead scattering data evaluations in ENDF/B-VII.1, JEFF 3.1, and JENDL 4.0. Two sample thicknesses, 3 cm and 5 cm, were measured with two sets of angles, 45, 70, 100, and 150 degrees as well as 30, 60, 110, and 150 degrees. During each measurement of 50-60 hours, two samples were moved into and out of the neutron beam using an automated sample changer. An open beam and a 7 cm carbon sample were measured for 6 minutes each and a natural lead sample was measured for 19 minutes. This cycle was repeated so that slight changes in the LINAC conditions over time would not affect the measurement. The carbon sample was used to assess the quality of the measurement and data analysis and as an indication of the systematic experimental error of this method.

2.1. Detectors

Two liquid scintillator (EJ-301) detectors were placed at each solid angle, totaling eight detectors used for each measurement. The detectors placed at 150 degrees were kept in the same location for all the measurements to ensure consistency and because often the back angle measurements have the greatest discrepancy with the evaluations. An illustration of the experimental setup is shown in Figure 1.

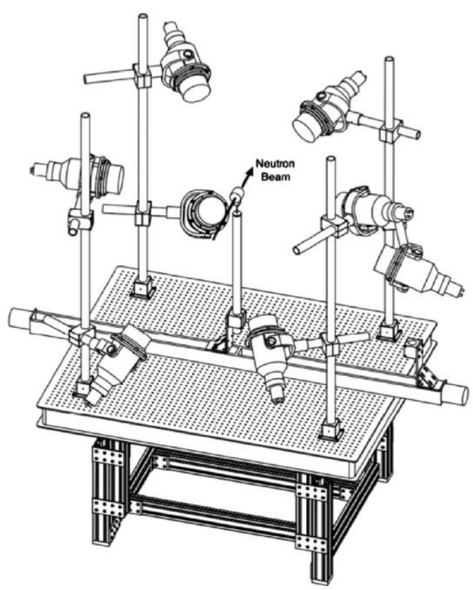

Figure 1. Scattering measurement experimental setup including eight EJ-301 detectors and a sample on a linear sample changer.

The voltage of each detector was set using a Na-22 calibration source to align all detectors to the same channel observed for the 511 keV peak. The detector gains were recalibrated every 24 hours. EJ-301 detectors are able to discriminate between gamma rays and neutrons because the pulse of a gamma ray has a faster fall time than that of a neutron [8]. This is due to the different interactions that these particles undergo in the $CH_{1.213}$ material of the detector. A gamma ray undergoes Compton scattering with electrons in the detector, while a neutron undergoes proton scattering. The light given off by the gamma interaction dies down faster than the light given off by a neutron interaction. Lower energy pulses with a lower pulse integral have a higher probability of being misclassified than higher energy pulses, so a misclassification factor was previously developed as a function of pulse integral [8].

2.2. Linear Accelerator Conditions

The LINAC electron beam conditions were: 11 microamps current, 55 MeV energy, and 7 ns pulse width. The electron beam was directed to a water cooled tantalum target. In the target, electrons undergo bremsstrahlung (electron slowing-down) interactions, which produce x-rays. These photons interact with the tantalum in the target in a (γ, n) reaction, which is an isotropic source of neutrons. These neutrons stream through a collimated flight path to the scattering station 30 meters away from the target. The photoneutron source has an evaporation spectrum in the range from 0.5 to 20 MeV in this experiment. Three neutron fission detectors were used to monitor the neutron beam conditions and normalize the data in post-processing.

3. DATA COLLECTION AND ANALYSIS

Scattering data were collected over a total of four weeks. One computer was dedicated to collecting and saving data, while another computer was used to analyze data. The raw data was analyzed using programs developed at RPI to classify pulses as gammas or neutrons using pulse shape analysis (PSA).

3.1. Acqiris Data Acquisition System

Data were recorded using an 8-bit, 1 GHz digital Agilent-Acqiris AP240 digitizer system, which has been used previously at RPI [9]. The digital data allowed pulse shape analysis to be used to discriminate gammas from neutrons in post processing [8]. Each event was saved with the time of the event, the detector number, and the pulse waveform sampled at 1 ns resolution for 120 ns.

3.2. Post Processing

Raw data was post-processed using C++ and Python codes written at RPI [8]. This process outputs grouped data in time of flight (TOF) after classifying pulses using PSA. The background is subtracted and runs are monitor normalized. A gamma misclassification factor is also applied to the data [8].

3.3. Comparing to MCNP Simulations

MCNP6 simulations of the experimental setup were performed using lead scattering cross section data from ENDF/B-VII.1, JEFF 3.1, and JENDL 4.0 for the lead sample. All other materials in the simulation of the experiment used ENDF/B-VII.1 cross section data. The neutron flux and detector efficiency for each of the eight detectors used have been measured using prior in-beam experiments [8]. These simulations were normalized and compared to measured lead scattering data. A comparison between simulations using each of the three data libraries and the experimental data was made to determine which of the evaluated data libraries is most accurate.

The normalization factors were calculated for each detector using the measured and simulated carbon data. Each detector had its own normalization factor, which was then used in the analysis of the lead data. Slight detector efficiency shifts were corrected by fitting a simulation of the carbon measurement to the carbon scattering data. This efficiency shift calculated using the carbon data was then applied to the analysis of the lead scattering data. The systematic uncertainty of this post processing technique thus depends on the ability to accurately measure the carbon scattering. This is shown in Figure 2 for scattering at 30 degrees.

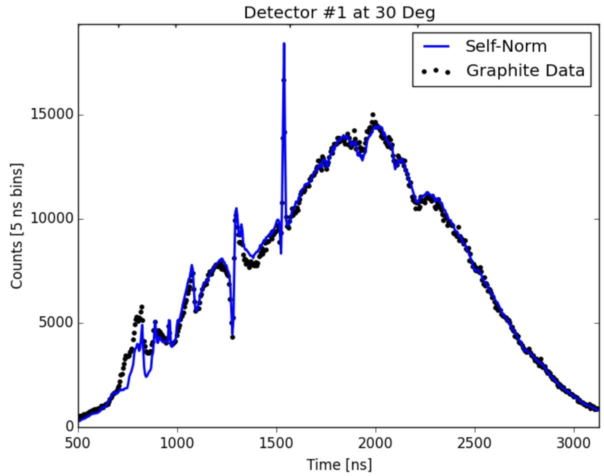

Figure 2. Measured carbon scattering data at 30 degrees compared to MCNP6 simulation using ENDF/B-VII.1 cross section data.

The measured lead data compared to simulations using different cross section libraries is shown in Figure 3 at 30 degrees and 150 degrees. There are some discrepancies between the measured data and the simulations. At 30 degrees, ENDF/B-VII.1 and ENDF/B-VI.8 overpredict scattering between 1250 ns and 1500 ns (3.1 MeV to 2.2 MeV) and all the libraries underestimate resonances in the lower energy region (<1 MeV). At 150 degrees, ENDF/B-VII.1 and ENDF/B-VI.8 underpredict scattering and all the libraries underestimate some of the resonances, especially a large resonance at 2460 ns (817 keV).

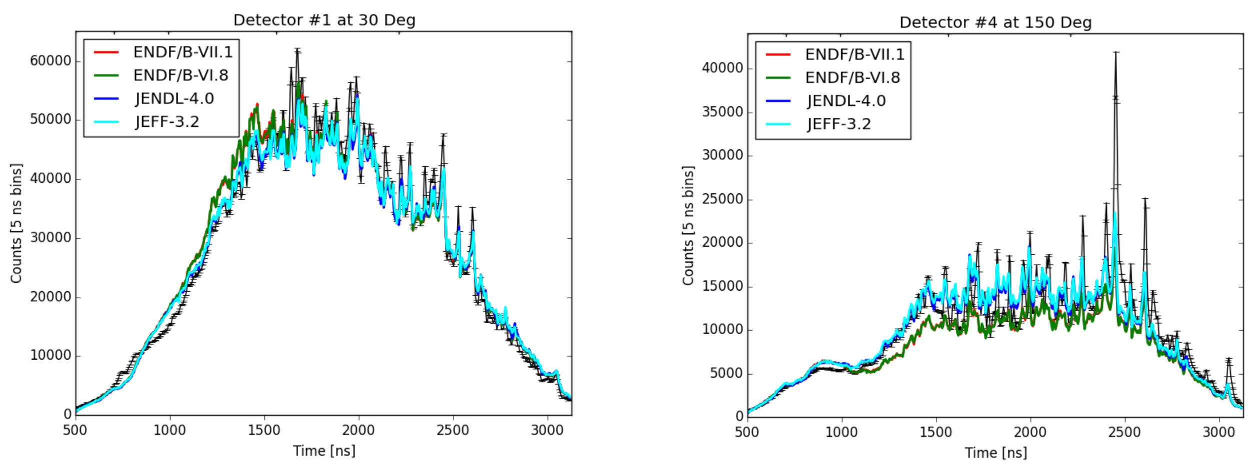

Figure 3. Measured lead data at a solid angle of 30 degrees (left) and 150 degrees (right) compared to normalized simulated data.

4. RESULTS

A computation divided by experimental value (C/E) was used to quantify agreement between the evaluated data libraries and the experimental data. The total average C/E values for each data library are shown in Table 1. The values closest to 1.000 for each angle are bolded.

Table I. Total C/E values averaged from 0.5 MeV to 20 MeV

Angle	C(ENDF/B-VII.1)/E	C(JENDL 4.0)/E	C(JEFF 3.2)/E
30	**1.011**	0.983	**0.989**
45	0.883	0.911	**0.916**
60	0.962	**0.993**	**1.006**
70	**1.084**	1.071	**1.086**
100	1.093	**1.015**	1.020
110	**0.999**	0.941	0.947
150	0.841	**1.010**	1.029

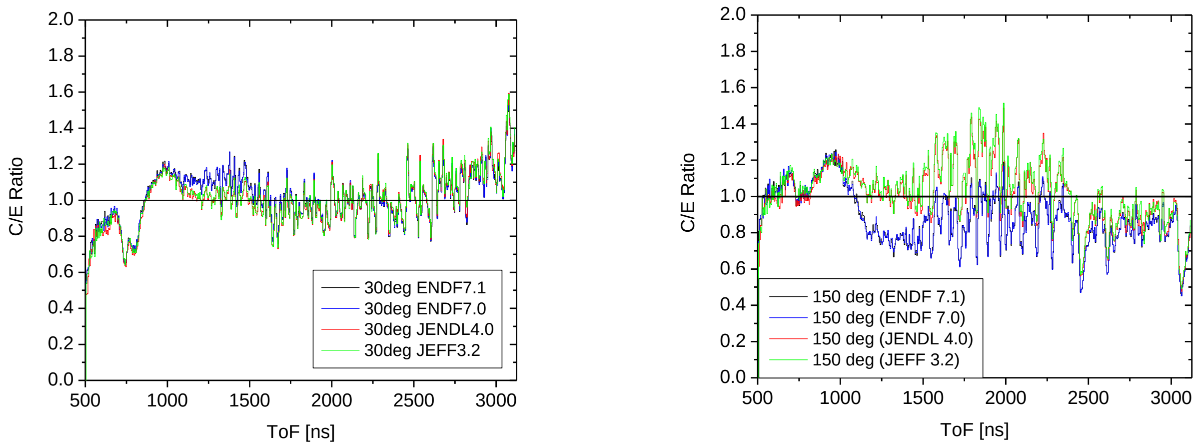

Figure 4. C/E values as a function of TOF for a detector at a solid angle of 30 degrees (left) and 150 degrees (right).

Figure 4 shows the C/E values for a detector at 30 degrees and at 150 degrees. At 30 degrees, there are discrepancies at low TOF (high energy) and some resonances are underpredicted at longer TOF (low energy). In most of the region of interest for this angle, the data agree reasonably well with the computed values. There is more discrepancy between measured data and computational results at the back angle. ENDF/B-VII.1 under estimates resonances. JEFF 3.2 and JENDL 4.0 both generally overestimate in the middle region, while underestimating certain resonances in the longer TOF (low energy) region.

Of particular interest is the resonance at 817 keV, which was observed to be overestimated at some angles (60, 70, 100 degrees) and underestimated at other angles (30, 150 degrees). This indicates that the smooth optical model used to calculate the angular distributions for MCNP6 does not correctly model the behavior of strong resonances. Figure 5 shows the data compared to the MCNP6 simulation of the experiment and the Pb-208 elastic scattering cross section, which is the same in this region in ENDF/B-VII.1, JEFF 3.2, and JENDL 4.0. The resonances in the cross section are clearly seen in the data. However, the measured data are much higher than the calculated data in the resonance.

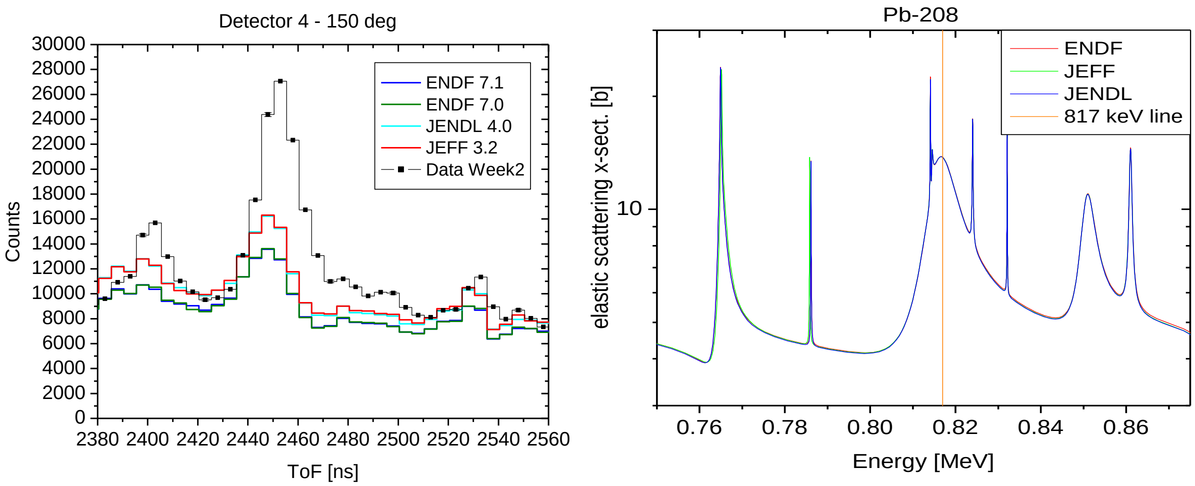

Figure 5. Measured and calculated natural lead scattering data zoomed around 817 keV (left) and elastic scattering lead cross section zoomed around 817 keV (right).

5. CONCLUSIONS

Lead scattering data were successfully measured at RPI in the 0.5 to 20 MeV energy region. The resonance structure of lead was observed. The measured data were compared to simulations of the experiment using different existing data libraries in order to determine which library best matched the data. JEFF 3.2 agrees best in the forward angles that were measured (30, 45, 60, 70 degrees); JENDL 4.0 agrees best at 100 and 150 degrees; and ENDF/B-VII.1 agrees best at 110 degrees. All libraries had discrepancies at certain resonances. One way to more accurately represent the angular distributions of lead in MCNP simulations may be to generate the angular distribution from the resonance parameters instead of the smooth optical model.

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