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# RESOLVED RESONANCE REGION ANALYSIS OF $^{206}$Pb, $^{207}$Pb, and $^{208}$Pb FOR NEXT GENERATION LEAD-COOLED FAST SYSTEMS

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**Abstract.** Recently, great interest has been generated in using lead as a coolant for fast neutron systems and as a result it is important to investigate the ENDF/B-VIII.0 [6] isotopic evaluations that comprise stable lead. To this end, resonance parameters for $^{206}$Pb, $^{207}$Pb, and $^{208}$Pb were re-evaluated because their resolved resonance regions extend beyond 0.5 MeV meaning resonance parameters used in reconstructing cross sections and elastic scattering angular distributions impact fast systems. The impact of resonance parameters is demonstrated by the differences between the evaluations in predicting experimental results from the fully modeled RPI Quasi-Differential Scattering Experiment[10] via MCNPv6.2 [12]. In addition, MCNP KCODE calculations of lead-sensitive fast spectra critical benchmarks showed variations of keff on average of 400 pcm, caused solely from differences in elastic scattering angular distributions in $^{208}$Pb. Re-evaluation entailed fitting data with the program SAMMY[18].

## 1 INTRODUCTION

Lead fast reactors (LFRs) are one of many GEN IV design concepts that may improve upon conventional light water reactors (LWRs). Examples of such improvements are larger amounts of passive safety and higher thermal efficiencies. Unfortunately, deficiencies in lead nuclear data are leading to poor calculation accuracy for systems analogous to LFRs. Not only does this impede development of LFRs like those concepts by Westinghouse [2] and Hydromine [3], but also the development of accelerator driven systems like MYRRHA [4]. To remedy this, the Department of Energy, Nuclear Energy University Program sponsored the evaluation of the natural isotopes of lead in order to improve predictive performance of lead cooled fast systems [5].

### 1.1 Neutron Modelling of LFR-Similar Systems

Before any commercial reactor can reach full NRC licensing, there needs to be extensive testing and validation of neutron and thermal-hydraulic models of similar-spectra test systems. Neutron modeling can be done using Monte Carlo, deterministic transport, and/or diffusion theory codes, all of which use cross section data from ENDF libraries such as ENDF/B-VIII.0 [6]. A straight forward sanity check for reactor engineers is to analyze the available critical experiments with similar spectra and composition to their proposed system. In the case of LFRs, engineers may look for fast spectrum systems that use lead as reflectors or interstitial material. Figure 1 shows calculated $k_{eff}$ over experimental measurements for a variety of benchmarks from the International Criticality Safety Experiment Benchmark Project (ICSBEP) Handbook [7] with ENDF/B-VIII.0 [6], JENDL-5.0[8], and JEFF-3.3 [9] cross sections. Figure 1 shows that ENDF/B-VIII.0 libraries display a trend of low C/E values for fast spectrum benchmarks that contain high amounts of lead. Contrasting, JENDL-5.0 and JEFF-3.3 calculate on average 500 pcm higher than ENDF/B-VIII.0 which results in either good agreement or slight over prediction. What's causing this disagreement and which library is a better representation of lead? These were the questions needing to be answered for this evaluation work.

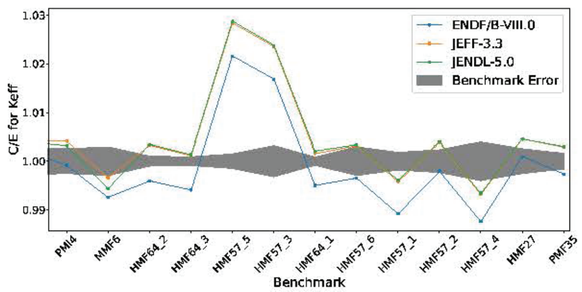


**Figure 1.** Comparison C/E values for ENDF/B-VIII.0, JENDL-5.0, and JEFF-3.3. ENDF/B-VIII.0 under predicts while JENDL-5.0 and JEFF-3.3 perform better but over-predict a few benchmarks (HMF64, PMF35).

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## 2 LEAD ISOTOPE EVALUATIONS

Beginning the evaluation work involved determining the contributing isotopes and reactions to a variety of critical, shielding, and benchmark systems [5]. Thirty-six critical and shielding benchmarks were evaluated from ICSBEP, with the shielding benchmarks proving to be largely insensitive to lead cross sections. Rensselaer Polytechnic Institute (RPI) Quasi-Differential Scattering Experiments [10] involving lead and sensitivities from DICE [11], further aided investigation into which isotopes were contributing the most to system uncertainties. In the end, models of these systems with MCNP[12] KSEN, and MCNP tallies determined that $^{208}$Pb elastic scattering is the largest contributor followed by $^{207}$Pb and $^{206}$Pb elastic scattering. This is no surprise since lead is known to have a low capture cross section and its near double magic nucleus makes the threshold energy for inelastic scattering quite high, leaving elastic scattering the main reaction channel. Table 1 contains the abundance, upper limit of the resolved resonance region and s-wave level spacing for ENDF/B-VIII.0. Note that $^{208}$Pb is over 50% of natural lead and because of its double magic nucleus has a wide level spacing and easily resolved resonances. Below are summaries of the current adjustments made to the isotopic evaluations, $^{204}$Pb was left out of the work since it accounts for only 1.4% of natural lead and most systems are insensitive to it. Studying the $^{208}$Pb cross sections for elastic scattering,

**Table 1.** Natural abundances, upper limit of the resolved resonance region (RRR), and s-wave level spacing ($D_o$) calculated from resonance parameters from ENDF/B-VIII.0 library of isotopes that comprise lead (natural).

| Isotope | Abund. [%] | RRR[keV] | $D_o$ [keV] |
|---|---|---|---|
| Pb-204 | 1.4 | 50 | 2.172 |
| Pb-206 | 24.1 | 900 | 37.1 |
| Pb-207 | 22.1 | 470 | 30 |
| Pb-208 | 52.5 | 1000 | 400 |

it appeared all evaluations were fit to the same ORELA transmission measurement by Carlton[13] meaning that, besides minor differences in capture reactions, the elastic scattering should be the same. However, looking at RPI quasi-differential data it was noted that the ratio of forward-to-back scattering would change with neutron library, pointing to the issue of elastic scattering angular distributions (ESAD). This was confirmed when ENDF/B-VIII.0 resonance parameters were used in NJOY21[14] to calculate new ESADs that greatly improved predictions of the RPI scattering experiment. Figure 2 shows a back-scatter detector of current libraries and Figure 3 displays new ESADs per isotope. Now that it was known that the ESAD was causing a large difference in the scattering experiment and that NJOY21 was capable of generating new ESADs via the Blatt-Biedenharn (BB) formalism [15], two things must be addressed. First, the BB formalism uses resonance parameters to directly calculate the angular distributions and therefore one must trust the quantum number assignments of the resonance parameters to a high degree. This means re-evaluating parameters to differential data on Experimental Nuclear Reaction Data (EXFOR) [16]. Secondly, the BB treatment requires the use of resonance parameters. Therefore, to improve the elastic scattering cross sections, the RRR was extended to 1.5 MeV, limited by the resolution of the data.

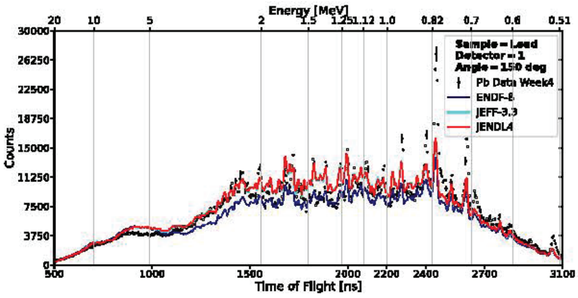


**Figure 2.** Simulated count rate and scattering data of back-scatter detector of RPI Quasi-Differential Scattering experiment with lead sample. MCNP simulation is normalized via carbon sample.

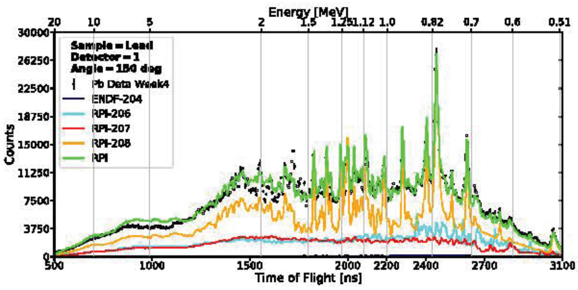


**Figure 3.** Simulated and scattering data of back-scatter detector of RPI Quasi-Differential Scattering experiment with lead sample. Improvement in scattering prediction below 1 MeV points. RPI evaluations are simply ENDF/B-VIII.0 evaluations with ESAD calculated from resonance parameters using Blatt-Biedenharn.

### 2.1 $^{208}$Pb

Since $^{208}$Pb is the main contributor to both critical and scattering systems, a significant portion of evaluation time was spent on this particular isotope. Changes to $^{208}$Pb include adding a direct capture component to the capture cross section, extending the resolved resonance region from 1 to 1.5 MeV, and recalculating the ESADs. A direct/semi-direct component was used to adjust the capture cross section to known Maxwellian Averaged Cross Sections (MACS(30 keV)) [17]. The average radiation width of resonances were reduced compared to ENDF/B-VIII.0 while an MF-3 background was added to the ENDF, together this reduces the capture worth of $^{208}$Pb for the RPI evaluation by a factor of two compared to ENDF/B-VIII.0. Though half of the previous capture, this has little to no effect on integral systems because the capture reaction is already 1,000 times smaller than elastic scattering. Extending the resolved resonance region was a significant undertaking and utilized the fitting code SAMMY [18] with MCNP simulations of the RPI scattering experiment to assign reasonable spin assignments. Figure 4 shows the current evaluations without resonances past 1 MeV and the RPI evaluation which has resonances out to 1.5 MeV.

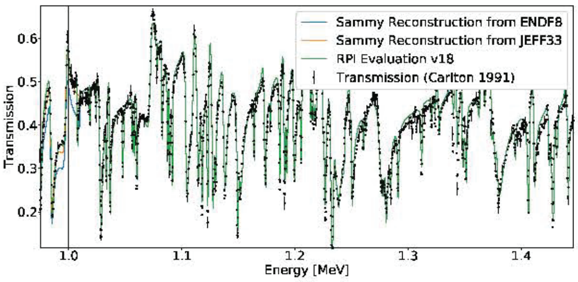


**Figure 4.** Calculated transmission from resolved resonance parameters of $^{208}$Pb in SAMMY against Carlton[13] data, JEFF-3.3 and ENDF/B-VIII.0 do not have resonance parameters past the black line at 1 MeV, while RPI goes out to 1.5 MeV.

## 2.2 $^{206}$Pb

Updates to the $^{206}$Pb isotope focused on adjusting the radiation widths of resonances, which constrain the neutron capture cross section. Over 50 minor p-wave and d-wave capture resonances were added to the evaluation that were observed in capture yield data from Borella[19] and Domingo-Pardo[20]. Additional studies into resonance statistics plots such as the cumulative level plot and neutron strength functions determined that these minor resonance have little to no impact on system performance but do present a more realistic model.

## 2.3 $^{207}$Pb

Evaluating $^{207}$Pb looked to verifying the capture cross section to new measurements by Domingo-Pardo [21] made after the release of ENDF/B-VIII.0. Additional work was done to adjust theoretical resonances pertaining to the bound levels of the nucleus as determined by thermal neutron scattering lengths. In actuality, this involved switching the neutron widths of the j$\pm\frac{1}{2}$ for the s-wave resonances.

# 3 IMPACT OF EVALUATION

With the resolved resonance region evaluations performed, the resonance parameters for all three isotopes were fed into NJOY21, with the RECONR module enabled to use the Blatt-Biedenharn formalism, to get new ESAD. The elastic angular distributions calculated from this step change the average scaResttering angle, mu-bar, at a much higher energy frequency than current evaluations. High energy quasi-differential scattering data largely validates the improvement from the new ESAD, $^{208}$Pb is the largest contributor to the large scattering fluctuations with only minor contributions visible from new $^{206,207}$Pb ESAD. The impact of the minor Pb isotopes cannot be understood without additional validation systems. To alleviate this, shielding and critical benchmarks, RPI Quasi-Differential Scattering Experiment [10], and RPI Lead Slowing Down Spectrometer measurements [22] were selected as validation systems for the evaluation. For brevity's sake, critical experiments were chosen to provide the most obvious observable impact from the evaluation.

## 3.1 Fast Critical Benchmarks

Suites of critical benchmarks are normally selected as the validation for nuclear data because these systems have gone through extensive characterization and provide a single low uncertainty value to compare against. The eigenvalue $k_{eff}$ provides an integral check on the whole cross section library in a single value. Figure 5 shows the performance of the international libraries and the RPI evaluation.

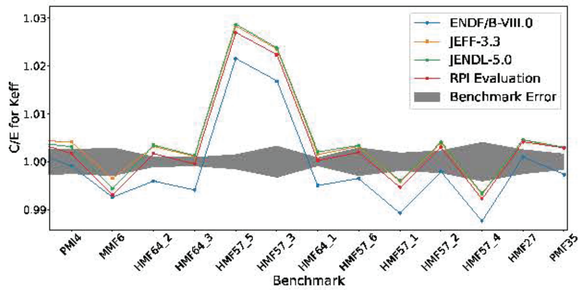


**Figure 5.** Comparison C/E values for international and RPI evaluations. RPI improves the ability to calculate the experimental $k_{eff}$ over all libraries for fast systems.

As stated previously these integral benchmarks are an important factor but, not the sole criteria in a list of checks for validating the cross section data. The models used could have any number of errors from geometries or deficiencies in the cross sections of other isotopes, resulting in scenarios where "fixing" the cross sections will not make C/E within uncertainty. Hence, the evaluator must look at the aggregate trends and note benchmarks such HMF57-3 and HMF57-5 [7] as potential outliers. Observing the trends from Figure 5 it can be seen that the RPI evaluation on average performs better than current libraries for fast spectra benchmarks. Table 2 shows a chi-square like statistic that compares the C/E value from the benchmarks and the benchmark uncertainty. Each benchmark's chi-square value is then weighted by the benchmarks total sensitivity to $^{208}$Pb and summed together to get the value shown. The outliers HMF57-3 and HMF57-5 are ignored in this calculation.

# 4 FUTURE WORK

Much of the work to date has been resolved resonance region evaluations with some refitting of the fast region.

**Table 2.** Performance of RPI evaluation and international libraries against fast benchmarks, chi-square statistic used to compute evaluation performance, and average departure from $K_{eff} \pm 1\sigma$.

| **Library** | **Fast Benchmarks** | $\bar{\Delta}K_{eff}[pcm]$ |
|---|---|---|
| ENDF-8.0 | 3.91 | 322 |
| JENDL-5.0 | 2.08 | 162 |
| JEFF-3.3 | 1.82 | 142 |
| RPI Evaluation | 1.75 | 142 |

A URR has been deemed not necessary in the evaluation scheme because of the limitations the evaluation technique has compared to RRR and fast region to accommodate updates to ESAD, elastic, and inelastic scattering reactions. A $^{208}$Pb fast region evaluation has been preformed by T. Kawano[23] of Los Alamos National Laboratory and the authors are collaborating to mesh the two evaluations. Covariance is the final component of the evaluation process and will happen once fast region fitting of $^{206,207}$Pb are finished.

## 5 CONCLUSIONS

Evaluated nuclear data is the foundation on which most neutron transport, and in extension the nuclear industry, is built on. Deficiencies in cross section libraries can have direct impacts on reactor design and operation. It was shown that cross section improvements of lead isotopes resulted directly to observable improvements in predicting lead reflected integral data, analogues to the much larger LFRs. Evaluation improvements include recalculating elastic scattering angular distributions for all isotopes, refitting capture cross sections for the major isotopes, extending the RRR for $^{208}$Pb, and adjusting bound resonance for $^{207}$Pb. Future work will be to re-evaluate fast regions for $^{206,207}$Pb and update covariance for all major isotopes.

## References

[1] T. K. Kim, C. Grandy, K. Natesan, J. Sienicki, R. Hill, "Research and Development Roadmaps for Liquid Metal Cooled Fast Reactors", ANL/ART-88, 2017.

[2] Status Report for LFR: https://aris.iaea.org/PDF/W-LFR-2020.pdf

[3] Status Report for LFR-AS-200: https://aris.iaea.org/PDF/Hydromine20LFR-AS-200-2020.pdf

[4] H. Abderrahim. "MYRRHA: A Multipurpose Experimental ADS for R&D objectives" SCK CEN. Presentation at EPAC 2004.

[5] Y. Danon, D Brown et al. "IMPROVEMENTS OF NUCLEAR DATA EVALUATIONS FOR LEAD ISOTOPES IN SUPPORT OF NEXT GENERATION LEAD-COOLED FAST SYSTEMS" MS-NE-2. DOE Technical Narrative 19-16739.

[6] D. A. Brown et al., "ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with Cielo-Project Cross Sections, New Standards and Thermal Scattering Data," Nucl. Data Sheets, 148, 1 (2018).

[7] J. Bess et al. "International Handbook of Evaluated Criticality Safety Benchmark Experiments" OECD-NEA, Print. July 2019.

[8] O. Iwamoto, N. Iwamoto, K. Shibata, A. Ichihara, S. Kunieda, F. Minato, and S. Nakayama, "Status of JENDL", EPJ Web of Conferences, 239, 09002-1-6(2020).

[9] A.J.M. Plompen, O. Cabellos, C. De Saint Jean, et al. The joint evaluated fission and fusion nuclear data library, JEFF-3.3", Eur. Phys. J. A56(2020)181.

[10] Amanda E. Youmans, J. Brown, A. Daskalakis, N. Thompson, A. Welz, Y. Danon, B. McDermott, G. Leinweber and M. Rapp, "Fast Neutron Scattering Measurements with Lead", AccApp 15, Washington, DC, pp. 355-360, November 10-13 2015.

[11] The DICE tool is available at: https://www.oecd-nea.org/science/wpncs/icsbep/dice.html

[12] C.J. Werner, J.S. Bull, C.J. Solomon, et al.,"MCNP6.2 Release Notes", LA-UR-18-20808 (2018).

[13] R.F. Carlton et al. "R-Matrix analysis of an ORELA measurement of n+208Pb total cross section from 78 to 1700 keV".Bulletin of the American Physical Society Ser.II, volume 36, page 1349, paper J10-10, 1991/04. 1991

[14] Jeremy Lloyd Conlin, A.C. Kahler, Austin P. McCartney and Daniel A. Rehn, "NJOY21: Next generation nuclear data processing capabilities", EPJ Web of Conferences 146, 09040 (2017).

[15] J.M. Blatt and L.C. Biedenharn, Rev. Mod. Phys. 24 (1952) 258

[16] N. Otuka, et al., "Towards a More Complete and Accurate Experimental Nuclear Reaction Data Library (EXFOR): International Collaboration Between Nuclear Reaction Data Centres (NRDC)", Nucl. Data Sheets 120, pp. 272-276 (2014); V.V. Zerkin, B. Pritychenko, "The experimental nuclear reaction data (EXFOR): Extended computer database and Web retrieval system", Nucl. Inst. and Meth. in Phys. Res. Sec. A: Accelerators, Spectrometers, Detectors and Associated Equipment 888, pp. 31-43 (2018).

[17] https://exp-astro.de/kadonis1.0/selementquery.php?isotope=208Pb

[18] N. M. LARSON, "Updated User's Guide for SAMMY: Multilevel R-Matrix Fits to Neutron Data Using Bayes Equations," ORNL/TM-9179/R8 ENDF-364/R2, Oak Ridge National Laboratory (Oct. 2008).

[19] Borella, A., Gunsing, F., Moxon, M., Schillebeeckx, P., and Siegler, P. (2007). "High-resolution neutron transmission and capture measurements of the nucleus $^{206}$Pb". Phys. Rev. C, 76, 014605.

[20] Domingo-Pardo, C., et al (2007). "Measurement of the radiative neutron capture cross section of $^{206}$Pb and its astrophysical implications". Phys. Rev. C, 76, 045805

[21] Domingo-Pardo, C., et al (2006)." Resonance capture cross section of $^{207}$Pb". Phys. Rev. C, 74, 055802.

[22] N. W. Thompson. "Measuring and Validating Neutron Capture Cross Sections Using a Lead Slowing-Down Spectrometer". Thesis. RPI 2017.

[23] T. Kawano. LANL. Private Communication, 2021-2022.