Advancements in Validation of TSLs through Inelastic Neutron Scattering and Transmission Measurements

Kemal Ramić[1], Chris W. Chapman[1], Goran Arbanas[1], Jesse Brown[1], BJ Marshall[1], Luke Daemen[1], Doug Abernathy[1], Alexander Kolesnikov[1], Yongqiang Cheng[1], Anibal Ramirez Cuesta[1], Daniel Siefman[2], Yaron Danon[3], Dominik Fritz[3]

[1]Oak Ridge National Laboratory

[2]Lawrence Livermore National Laboratory

[3]Rensselaer Polytechnic Institute

TSL evaluation and validation lingering questions

- Are theoretical calculations accurate enough to be the sole basis of TSL evaluation process?
- How does evaluators usage of atomistic calculations and understanding of material influences the evaluation process?
- How much variability is there really in Inelastic Neutron Scattering (INS) spectra for different samples of the same materials?
  + Phonon spectrum is a property of the bulk material
  + Different elements have different scattering lengths, combined with their concentrations
- Should INS measurements (qualitative test of inelastic physics which is the basis of TSL evaluations) be dismissed as part of the TSL evaluation process?

Neutron scattering sampling in neutronics codes

- How do CE Monte Carlo neutronics codes use TSLs for scattering?
  - Sampling the distance to next collision (transmission!)
  - Sampling the reaction ratio probability
  - Sampling the exit energy and angle distributions (TSL!)
- We test these with:
  - Experimental transmission
  - Experimental inelastic scattering ($S(\alpha, \beta)$)

Scattering cross sections are based on phonon spectrum

- Inelastic scattering (coherent plus incoherent):

In the incoherent and Gaussian approximation, the $S(\alpha,\beta)$, as expressed in NJOY LEAPR module, in terms of phonon expansion can be written as:

$$S(\alpha,\beta) = e^{-\alpha\lambda}\sum_{n=0}^{\infty}\frac{1}{n!}\alpha^{n}\frac{1}{2\pi}\times\int_{-\infty}^{\infty}e^{i\beta\hat{t}}\left[\int_{-\infty}^{\infty}P(\beta')e^{i\beta'\hat{t}}e^{-\beta'/2}d\beta'\right] \tag{1}$$

where:

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)} \quad \text{and} \quad W = \frac{\int_{-\infty}^{\infty}P(\beta)e^{-\beta/2}d\beta}{AkT} \tag{2}$$

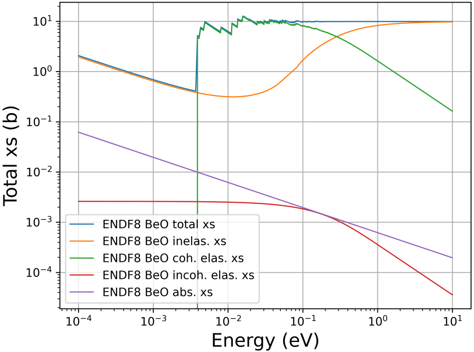
with $\rho(\beta)$ as the phonon spectrum.

- Coherent elastic scattering:

$$\sigma^{coh} = \frac{\sigma_c}{E}\sum_{E_i>E}f_i e^{-2WE_i} \tag{3}$$

- Incoherent elastic scattering:

$$\sigma^{incoh} = \frac{\sigma_b}{2}\left(\frac{1-e^{-4WE}}{2WE}\right) \tag{4}$$



- **As we can see all the scattering components are dependent on the inelastic physics through $\rho(\beta)$ and W (Debye-Waller factor)**

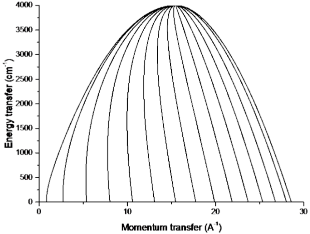
OAK RIDGE
National Laboratory

Atomistic calculations of phonon spectrum

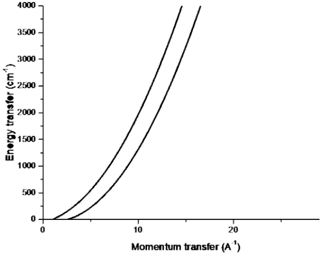
- Molecular dynamics (MD), ab-initio molecular dynamics (AIMD), and density functional theory (DFT) calculations are used typically for phonon spectrum calculations. DFT calculations are basis for coherent one-phonon inelastic scattering physics for strong coherent scatterers like carbon in graphite.
- The basis for these calculations are inter-atomic potentials.
- The potentials are only approximations to actual solution of Schrödingers equation, and are tailored to reproduce specific experimental quantities.
- Reproducing specific experimental quantities (i.e. lattice constants) does not mean other quantities will be reproduced well.
- The initial structure for non-crystaline materials is relatively unknown, and it is usually dependent on the evaluators choice of inter-atomic potential and evaluators understanding of the material.
- Surely with so many unknowns measurements need to be part of the evaluation and validation process?

INS measurements can validate $S(\alpha, \beta)$

- Direct geometry spectrometers (ARCS and SEQUOIA):



- Indirect geometry spectrometers (VISON):



+ Measured quantity $S(Q, \omega)$ is directly related to $S(\alpha, \beta)$:

$$S(\alpha, \beta) = k_B T \; exp\left(\frac{-\hbar\omega}{2k_B T}\right) S(Q, \omega) \tag{5}$$

where T is the temperature, and k_B is the Boltzmann constant.

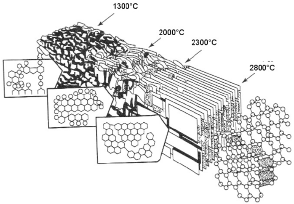
+ **This means that we can directly measure what we store in ENDF TSL files.**

Are only atomistic calculations enough?

- ENDF/B-VIII.1.b1 has 5 different graphite libraries: crystalline, Sd (crystalline), 10%, 20%, and 30% porosity reactor graphite

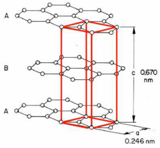
- What is graphitization process?
  + Graphitization is the process of heating amorphous carbon for a prolonged period of time, **rearranging the atomic structure to achieve an ordered crystalline structure** that is typical of solids.

Evolution of graphitization process, reproduced from [1]:

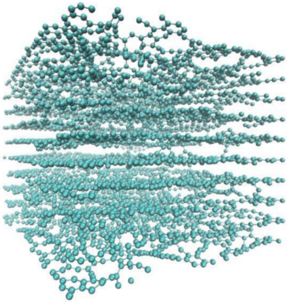


How does all this manifest itself in inelastic scattering measurements?

- Crystal structure for Crystalline and Sd graphite [2]:



- Porous structure for 30% porosity graphite [2]:



Measured graphite information

Graphite	Grain size [µm]	Density [g/cm^3]	Porosity [%]
PGA	800	1.70	25
G347a	50	1.85	17.8
IG-110	20	1.77	21.6
NBG-18	1600	1.85	17.8
PCEA	360	1.83	18
Mersen 2114	13	1.81	10
POCO-AXF-5Q	5	1.78	20
POCO-ZXF-5Q	1	1.78	20

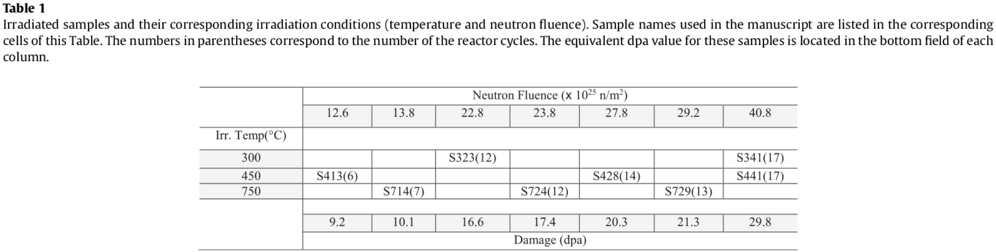
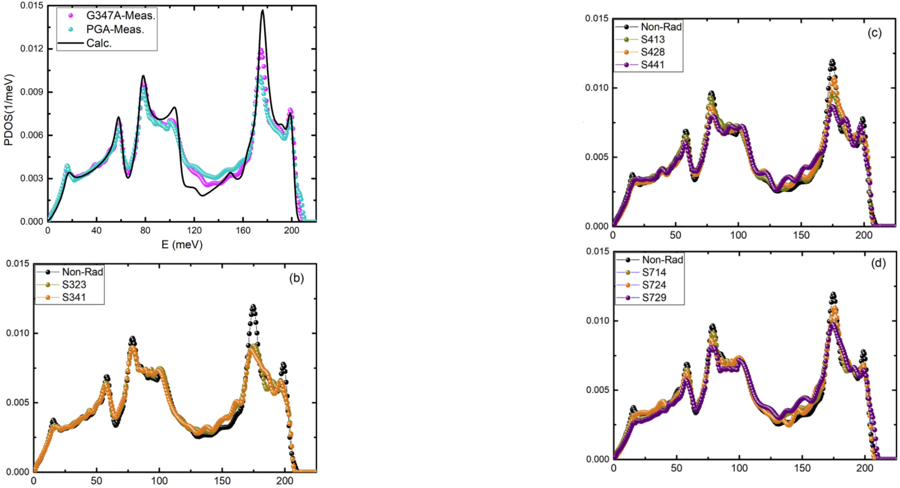


Table 1
Irradiated samples and their corresponding irradiation conditions (temperature and neutron fluence). Sample names used in the manuscript are listed in the corresponding cells of this Table. The numbers in parentheses correspond to the number of the reactor cycles. The equivalent dpa value for these samples is located in the bottom field of each column.

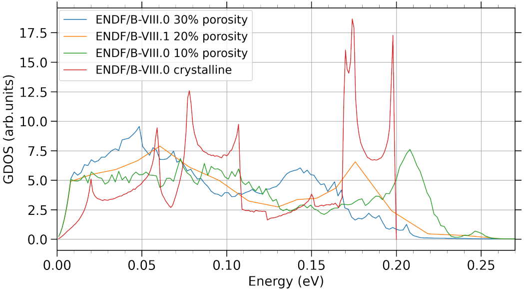
	Neutron Fluence (x 10^{25} n/m^2)						
	12.6	13.8	22.8	23.8	27.8	29.2	40.8
Irr. Temp(°C)							
300			S323(12)				S341(17)
450	S413(6)				S428(14)		S441(17)
750		S714(7)		S724(12)		S729(13)	
	9.2	10.1	16.6	17.4	20.3	21.3	29.8
	Damage (dpa)						

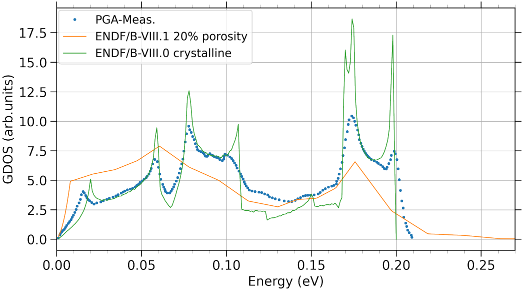
Atomistic calculations ≠ enough, INS measurements = essential

- In [3] and [4] phonon spectrum was measured at ARCS spectrometer for two non-irradiated samples of nuclear grade graphite (G347a and PGA) as well as eight samples of irradiated G347a, at different temperatures and neutron fluences.



Atomistic calculations ≠ enough, INS measurements = essential

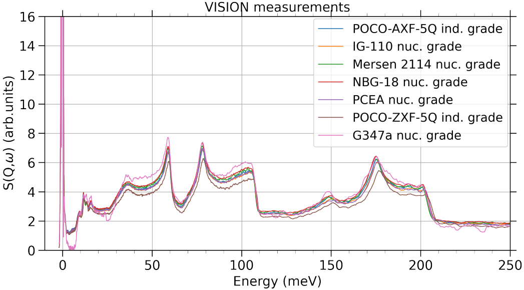


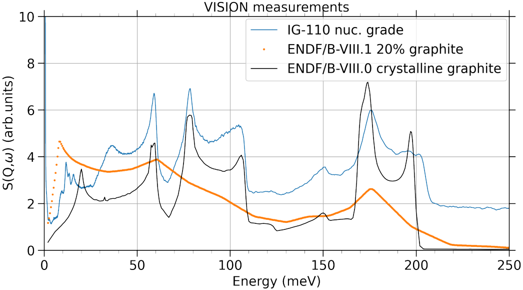


- **INS measurements of different grades of nuclear graphite are consistent**, and in line with expectation that INS spectra due to graphitization process should be similar to INS of perfect graphite.
- **Porous graphite libraries are not consistent with INS measurements**, and show impact of relying on just theoretical calculations and modeling inconsistent with the material characterization and understanding!

Atomistic calculations $\neq$ enough, INS measurements $=$ essential

- Our collegues at ORNL have measured multiple different grades of graphite at VISION spectrometer as well:

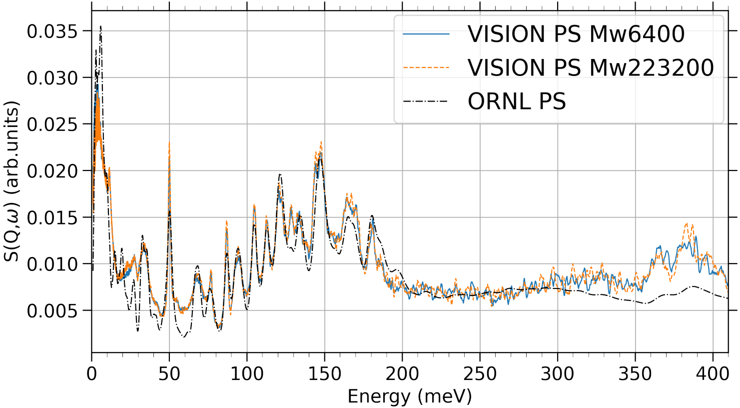




- **INS measurements of different grades of nuclear graphite are consistent**
- **Porous graphite libraries are not consistent with INS measurements**
- INS measurements show that phonon spectrum is not so variable with changing composition of the samples, and **INS measurements are integral to TSL evaluation and validation.**

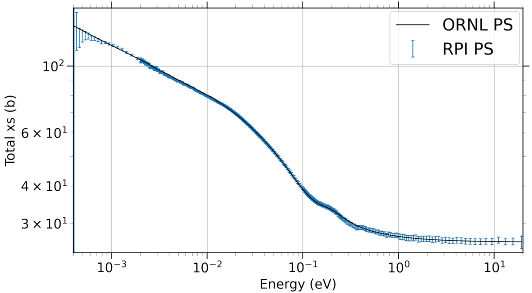
Oak Ridge National Laboratory

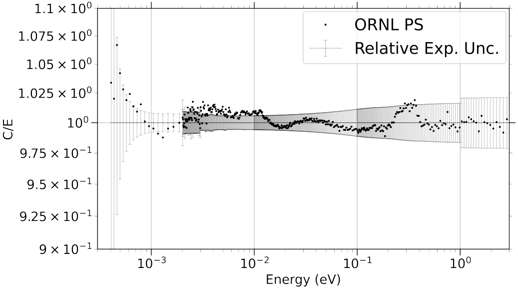
Integral Performance ≠ Differential Performance
Example: Polystyrene & Polyethylene



- Structurally different polystyrene (different molecular weights, Mw) have almost identical inelastic spectra.

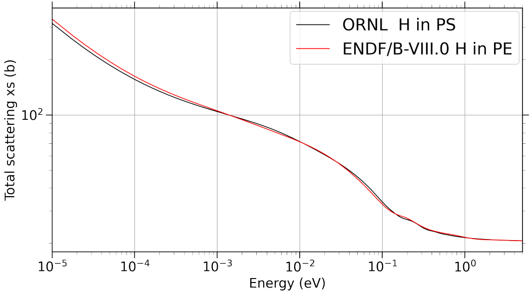
Integral Performance $\neq$ Differential Performance
Example: Polystyrene & Polyethylene

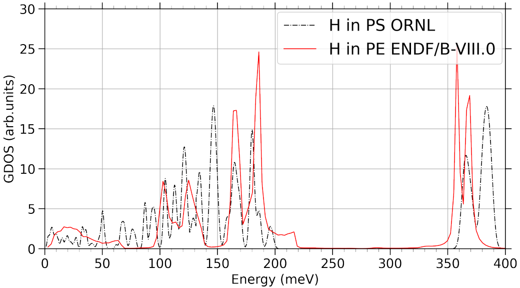




- Excellent agreement with RPI transmission measurement. Ideally for validation purposes multiple transmission measurements should be used.

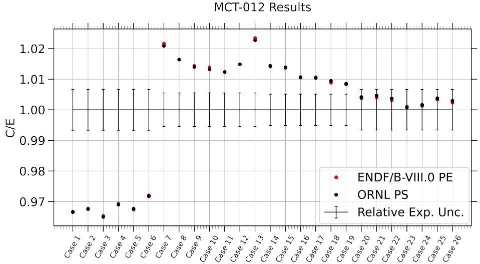
Integral Performance ≠ Differential Performance
Example: Polystyrene & Polyethylene

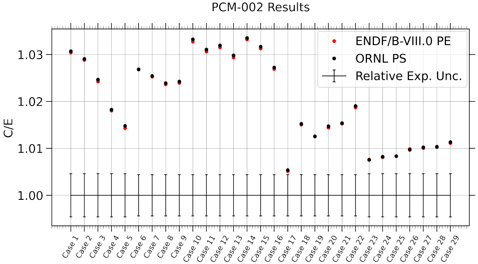




- PS and PE have almost identical total cross section for hydrogen.
- PS and PE have significantly different phonon spectrum.

Integral Performance $\neq$ Differential Performance
Example: Polystyrene & Polyethylene



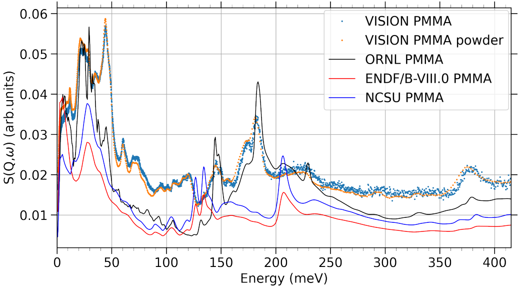


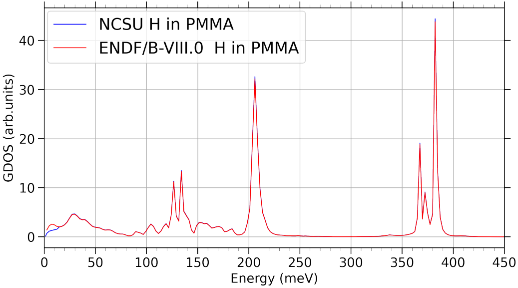
	MCT-012 χ^2	PCM-002 χ^2
Polyethylene	9.12	24.12
Polystyrene	9.13	24.7

Table 1: Calculated χ^2 value using PS and PE TSLs for MCT-012 and PCM-002 benchmarks.

- **This demonstrates importance of INS and transmission measurements, because with different phonon spectra we can calculate total cross section nearly identically, as well as critical benchmarks.**

Lucite evaluation - INS validation

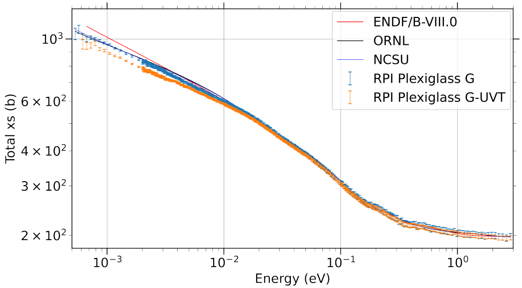


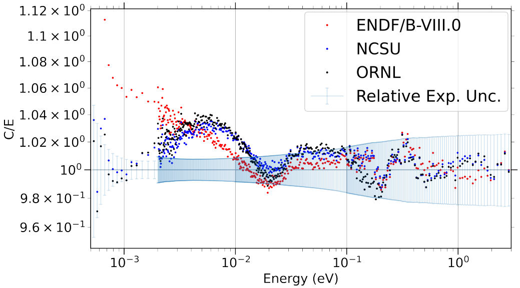


TSL	χ^2
ENDF/B-VIII.0	1.24E+6
NCSU	8.32E+5
ORNL	4.94E+5

- Different PMMA samples, powder and sheet (with additives) have almost identical INS spectra.
- ORNL has a better agreement with INS measurement.
- NCSU updated the phonon spectrum yet agreement with INS measurements was not significantly improved.

Lucite evaluation - Transmission validation

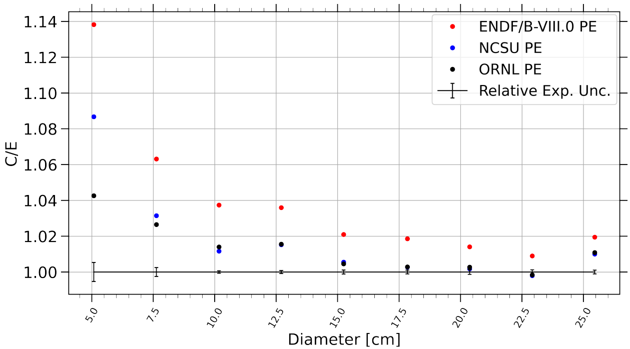




TSL	Plexiglass G χ^2
ENDF/B-VIII.0	2.939E+7
NCSU	2.9322E+7
ORNL	2.9315E+7

- Plexiglass G and G-UVT samples have different total cross section due to additives.
- ORNL TSL has an overall better agreement with Plexiglass G (common form) than both updated NCSU and ENDF/B-VIII.0 evaluations.

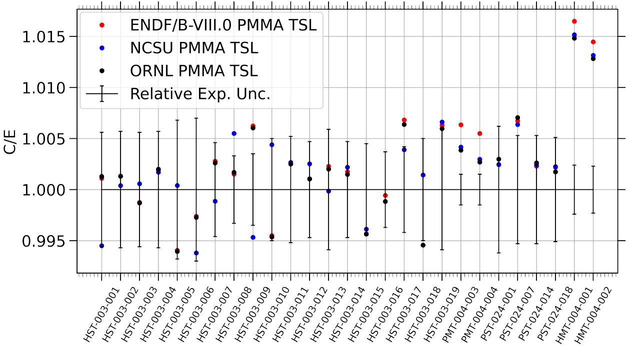
Lucite - Pulsed neutron die-away validation



TSL	χ^2
ENDF/B-VIII.0	6975.1
NCSU	1141.4
ORNL	1029.7

- ORNL TSL performs significantly better than ENDF/B-VIII.0 evaluation.
- ORNL and NCSU perform similarly due to their similar total cross sections.
- Similar to critical benchmarks there is a high sensitivity in changes to the total cross section, which may not be justifiable in comparison with inelastic measurements.

Lucite - Critical benchmarks validation



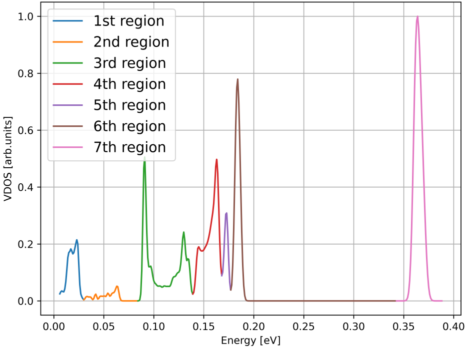
TSL	χ^2
ENDF/B-VIII.0	132.73
NCSU	97.51
ORNL	93.41

- ORNL TSL performs better including new HMT-004 benchmarks from LLNL
- **This demonstrates importance of INS and transmission measurements, because with different phonon spectra we can calculate total cross section similarly, as well as critical benchmarks.**

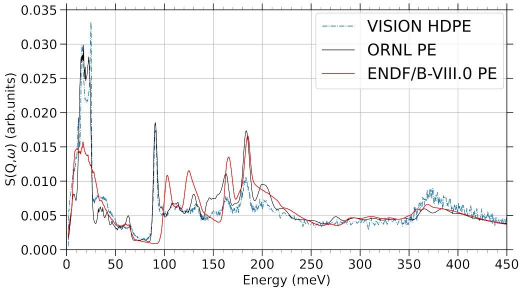
Polyethylene (PE) evaluation

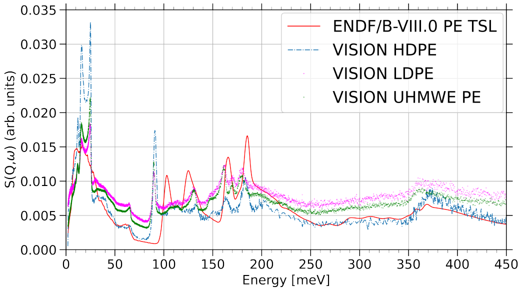
- Motivation:
  - New transmission measurements at RPI, as well as criticality benchmarks from LLNL and LANL.
  - ORNL PE evaluation was optimized with respect to differential measurement at VISION spectrometer at SNS, as well as transmission measurement from RPI.

- Optimization summary:
  - assign weights for each distinct region of GDOS and vary them by Dakota
  - calculate χ^2 with respect to VISION INS measurement. ENDF files can be directly compared to the VISION data by extracting $S(\alpha, \beta)$ at specific (α, β) values measured in VISION experiment and applying well-know VISION experimental resolution.
  - calculate χ^2 with respect to RPI transmission measurement
  - repeat the process until combined χ^2 is minimized



Polyethylene validation- INS measurements at ORNL



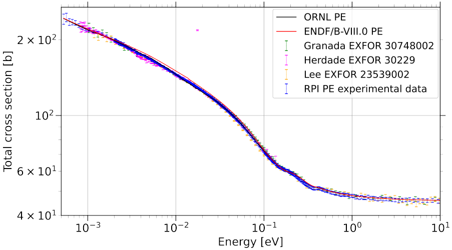


TSL	VISION HDPE χ^2
ENDF/B-VIII.0	8.80E+4
ORNL	4.36E+4

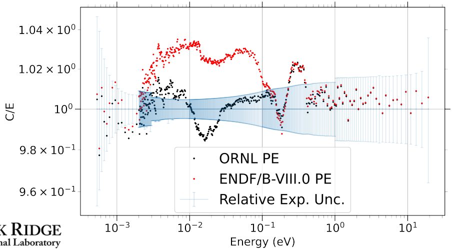
* **ORNL PE has better agreement with the shape of all different PEs!**

Polyethylene validation- Transmission measurements at RPI

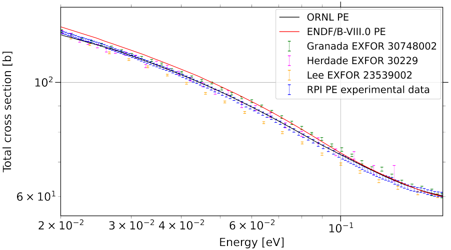
- Total xs comparison (5e-4 to 10 eV):



- C/E comparison to new RPI data:



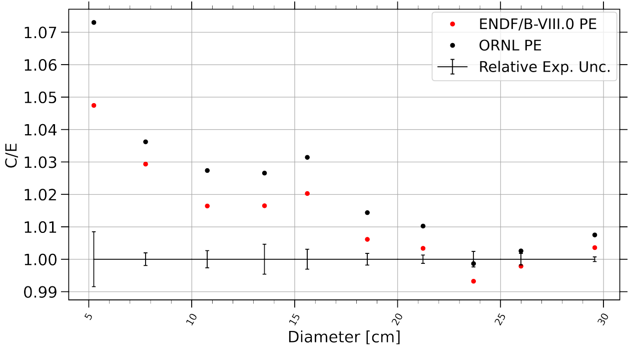
- Total xs comparison (2e-2 to 2e-1 eV):



Transmission exp.	ENDF/B-VIII.0 χ^2	ORNL χ^2
RPI	1.31E+8	4.28E+7
Herdade	4.73E+8	4.59E+8
Lee	1.9443	1.9421
Granda	2.33E+7	2.57E+7

Polyethylene validation- Pulsed neutron die-away measurements

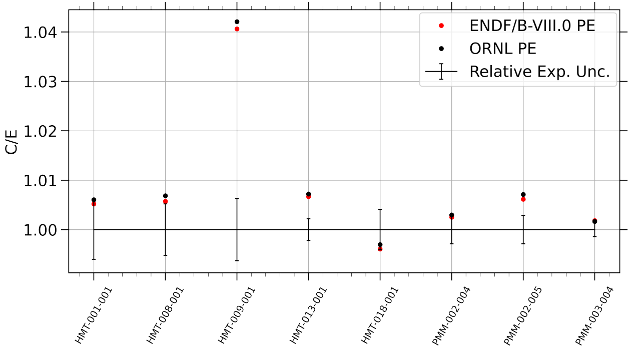
- New pulsed neutron die-away measurements at LLNL.



TSL	χ^2
ENDF/B-VIII.0	404.09
ORNL	901.29

- New ORNL evaluation reduces, compared to ENDF/B-VIII.0, total neutron scattering cross section resulting in an increase in PNDA α compared to ENDF/B-VIII.0. ENDF/B-VIII.0 and ORNL calculated α values are within 1-2% of each other.

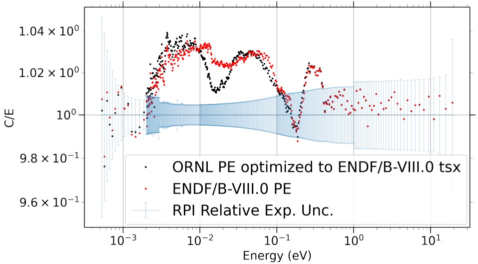
Polyethylene validation- Integral criticality benchmarks

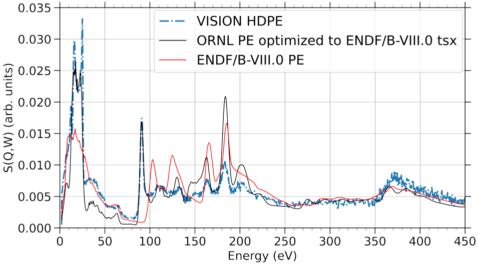


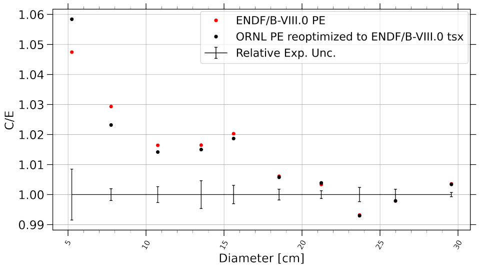
TSL	χ^2
ENDF/B-VIII.0	60.83
ORNL	67.38

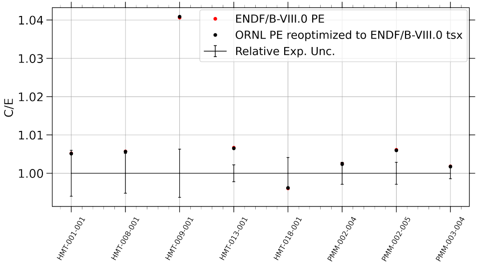
- ORNL evaluation reduces, compared to ENDF/B-VIII.0, total neutron scattering cross section resulting in an increase in neutron multiplication factor k_{eff}.
- For most benchmarks k_{eff} for calculated values is anywhere from 200- 4000 pcm away from experimental values, which is indicative of possible issues with nuclear data besides TSL.
- As presented by Catherine Percher during mini-CSWEG2023 these benchmarks are really not that thermal and not a good test for TSLs.

- Optimizing the cross section with respect to ENDF:



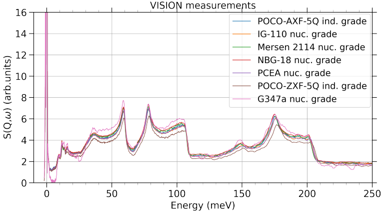






Summary & Conclusions

- INS measurements are not as variable based on sample composition, because phonon spectra is a properly of the bulk material.
- **INS and transmission measurements are and should be the most significant part of TSL validation**, as they are the fundamental quantities used by Monte Carlo neutronics codes.
- **Relaying solely on theoretical atomistic calculations for TSL evaluation is not enough**, they are highly dependent on inter-atomic potentials (which are not the most accurate depending on the quantity trying to be reproduced) and evaluators knowledge and understanding of the material being studied.
- Crit. benchmarks - While extremely useful for validation of nuclear data at all energies, critical benchmarks are not the best tool to provide a definitive answer on conflicting TSLs
- - **INS and transmission measurements need to be the basis of validation of TSLs**



Acknowledgements

- This work was supported by the Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy.
- This research used resources at the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory.
- This research used resources of the Compute and Data Environment for Science at ORNL, which is supported by DOE SC under Contract No. DE-AC05-00OR22725.
- Computational resources were also provided by the Rensselaer Polytechnic Institute Center for Computational Innovations, more specifically the Artificial Intelligence Multiprocessing Optimized System supercomputer.
- This research used resources of the National Energy Research Scientific Computing Center (NERSC), a U.S. Department of Energy Office of Science User Facility operated under Contract No. DE-AC02-05CH11231.

References

[1] Delhaes, P., “Carbon Science and Technology - From Energy To Materials”, Hoboken, NJ: John Wiley Sons, Inc., pp.35-38., 2012

[2] A. Hawari and V. Gillete. “Inelastic Thermal Neutron Scattering Cross Sections for Reactor-grade Graphite”, .Nuc. Data Sheets, 118:176–178, 2014

[3] I. I. Al-Qasir et al., “Vacancy-driven variations in the phonon density of states of fast neutron irradiated nuclear graphite”, Carbon, 168:42–54, 2020.ISSN: 0008-6223

[4] I. I. Al-Qasir et al., “Neutron thermalization in nuclear graphite: A modern story of a classic moderator”, Ann. Nucl. Energy, 161:108437, 2021.ISSN: 0306-4549

Additional slides: SANS xs

- **Small angle neutron scattering (SANS)** - relevant for materials like nanodiamonds and porous graphite, which arises due to differing scattering length densities across the material (i.e. pores vs carbon).

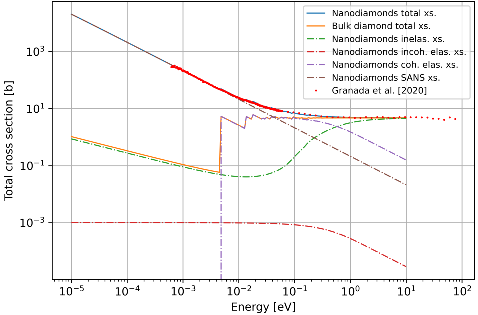
$$\sigma(k_0) = \frac{\sigma_0}{2k_0^2} \int_0^{2k_0} qI(q)\mathrm{d}q. \qquad (6)$$

This cross section as a function of the wavevector k_0 and I(q), the SANS structure factor, is used by Monte-Carlo codes during run-time to compute the macroscopic total cross section and sample the distance to next collision. If there is a collision, the code randomly picks the reaction based on the ratio of its cross section to the total one. For different types of scattering, the outgoing energy and direction are sampled according to the underlying physics. In the case of SANS, there is no energy exchange, but the direction still need to be determined.

- There is a need for SANS format in ENDF or GNDS
- Already implemented in OpenMC and MCNP, MCNPX, and in new NCrystal v3.0.0 release

]

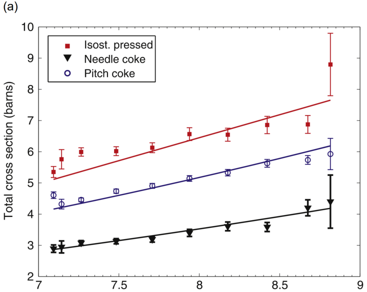
- Inelastic scattering (coherent plus incoherent)
- Coherent elastic scattering
- Incoherent elastic scattering



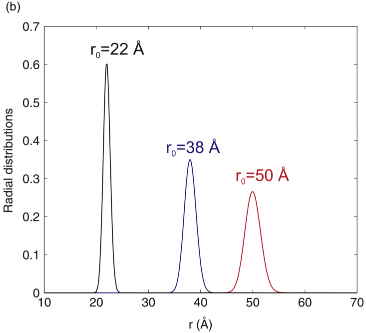
* K.Ramić, J. I. Marquez Damian, et al.”Advances in Nuclear Data and Software Development for the HighNESS Project”, 14th International Topical Meeting on Nuclear Applications of Accelerators

Additional slides: Porous graphite

- In “*Model for neutron total cross-section at low energies for nuclear grade graphite”, Nuclear Instruments and Methods in Physics Research Section A, Volume 971*, V. M. Galvàn Josa et al., have shown that the **magnitude of SANS xs component is directly related to the pore sizes** for nuclear graphite.

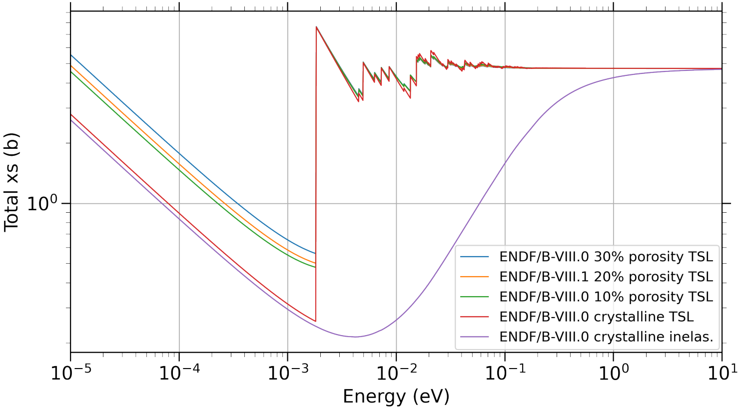


- Fig. a) Experimental neutron total cross-section data (dots) as a function of neutron wavelength, and the corresponding fitted curves.



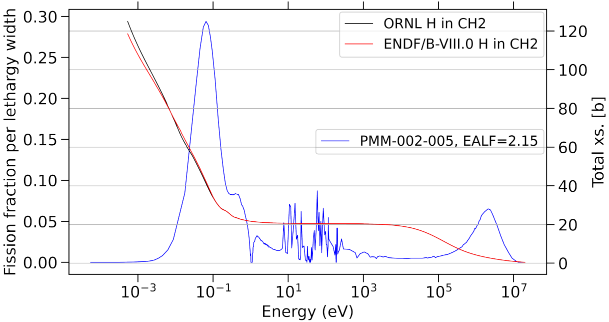
- Fig. b) Pore sizes.

Additional slides: Porous graphite



Additional slides: Fission density spectra in benchmarks

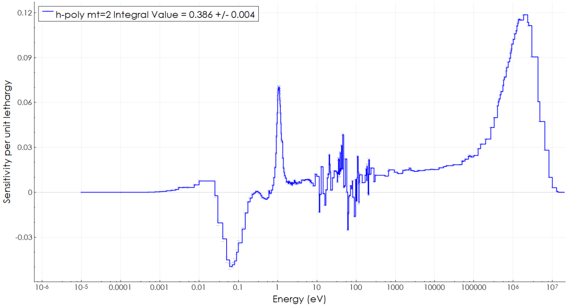
- Three bin spectral definition from ICSBEP Handbook provides a rough quantification of the neutron energy spectrum
- TEX thermal cases are not very thermal
- PU-MET-THERM-002-005 has about 66% fissions induced by thermal neutrons, corresponding to an EALF of 2.15 eV



- **How effective are these benchmarks at differentiating among different 1H-poly TSL evaluations?**

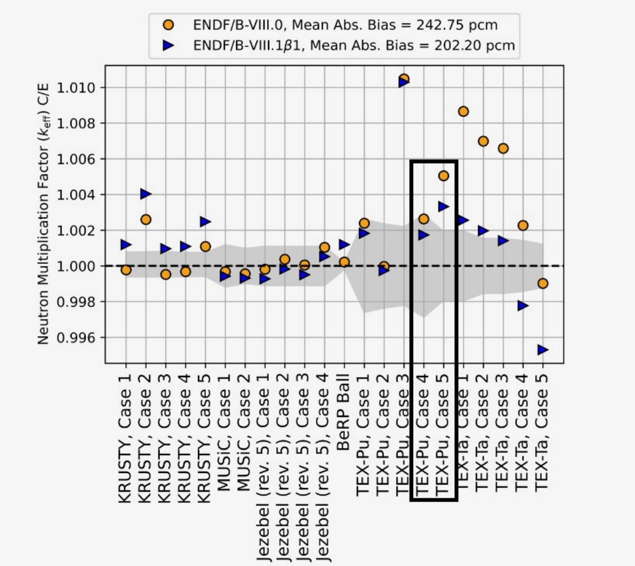
Additional slides: Sensitivity data confirm lack of sensitivity

- Majority of sensitivity is above 10 eV
  - TSL not even being used
- Misprediction of k_{eff} for this benchmark is unlikely to be driven by TSL
  - CAUTION: This is based on the integrated 1D scattering sensitivity

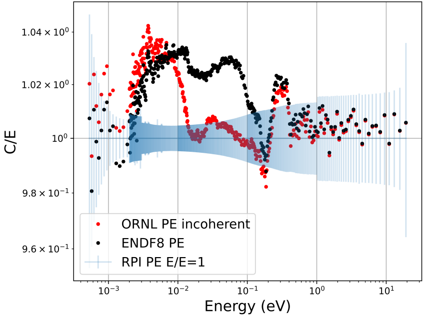


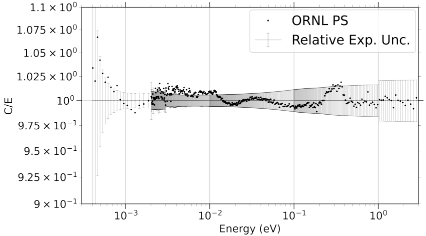
+ **Good news:** new experiments with thicker moderator plates have been performed and evaluated
  - PMT-004 evaluations approved at April TRG meeting for inclusion in 2023 ICSBEP Handbook
  - Two cases include polyethylene
  - Thermal fission fraction of these two cases is 72.9% and 74.6%
  - Patience: these two cases will likely be useful and available within the next year
+ **Less good news:** Only two points, both with Pu, and likely highly correlated

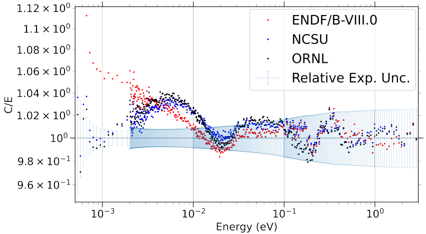
Additional slides: LANL ENDF/B-VIII.1.b1 testing



Additional slides







OAK RIDGE National Laboratory