OAK RIDGE National Laboratory

Status of ORNL TSL evaluations

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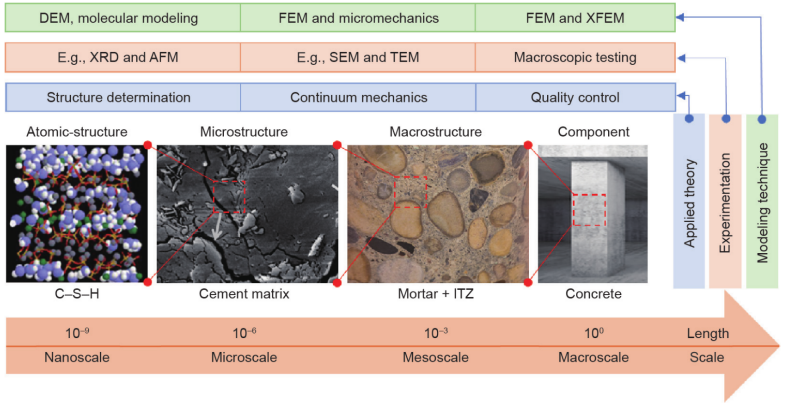
Overview

- Modeling of concrete
- Updates of polyethylene evaluation
- Initial transmission measurements at ORNL

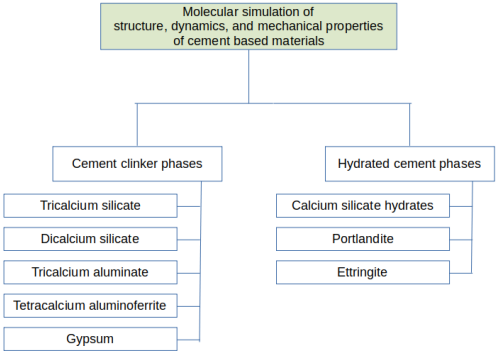
Modeling of concrete

- Work on ORNL ND11 task started towards the end of FY2022 Q3.
- Concrete is a extremely hard material to model on the atomistic level.



Reproduced from [1]. Schematic of multiscale description of a cementitious system. DEM: discrete element method; FEM: finite element method; XFEM: extended finite element method; XRD: X-ray diffraction; AFM: atomic force microscope; SEM: scanning electron microscopy; TEM: transmission electron microscopes; ITZ: interfacial transition zone.

Modeling of concrete

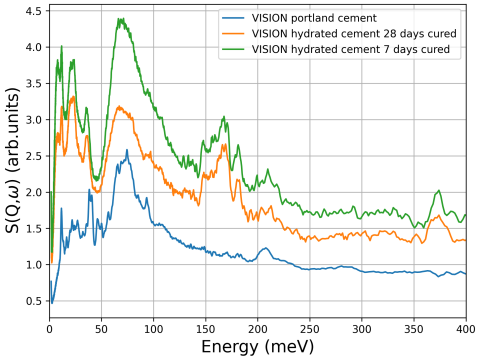


Reproduced from [1]. Molecular simulation of cement-based materials.

- Cement clinker phases have already been modeled, including gypsum.
- Unlike previous solid materials of interest to NCSP, there is no set structure for calcium-silicate-hydrate (CSH), which is 50%-65% by volume of hydrated cement paste, but we have been using different models available in the literature for modeling.
- Portlandite (CH) and Ettringite will be modeled as well.

Modeling of concrete

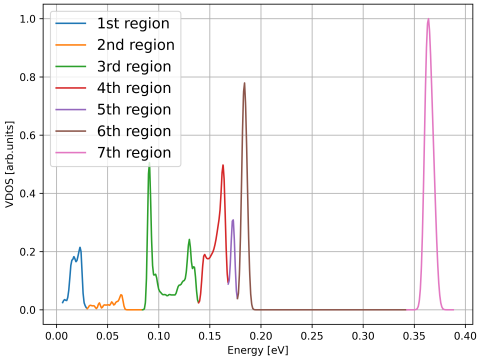
- So what is our plan?:
  1. Assume (and test) that thermal neutron scattering law of concrete is linear combination of percentages of the phonon spectra of the hydrated cement and aggregates (i.e. quartz, calcite, dolomite)
  2. Mix a specific composition of concrete, i.e. 70% quartz and 30% hydrated cement (with 50%-50% mixture of water and portland cement)
  3. Measure inelastic spectra at VISION of aggregates, portland cement, hydrated cement, and concrete (after a month of curing). We have some preliminary data already, and we will create own samples of hydrated cement and concrete using different portland cement, and possibly different compositions.
  4. Measure transmission of hydrated cement and concrete at RPI.
  5. Possibly measure transmission of different compositions of hydrated cement and concrete at ORNL.



Polyethylene (PE) evaluation

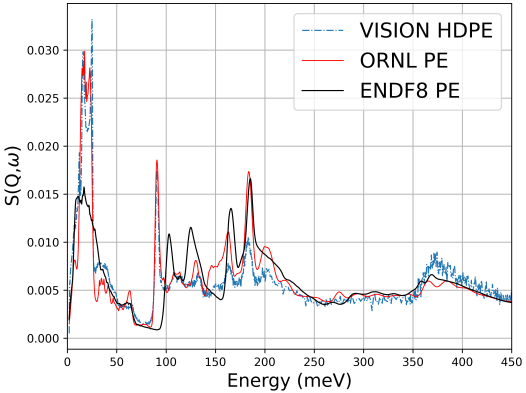
- Motivation:
  - New transmission measurements at RPI, as well as criticality benchmarks from LLNL.
  - ORNL PE evaluation was optimized with respect to differential measurement at VISION spectrometer at SNS, as well as transmission measurement from RPI. See Chris talk, as well as [2,3].

- 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

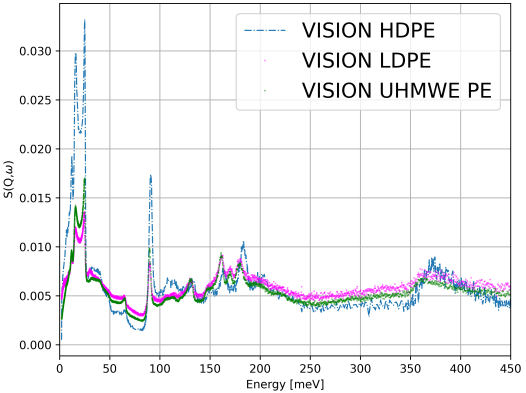


Polyethylene evaluation validation- INS measurements at ORNL

- VISION HDPE comparison:



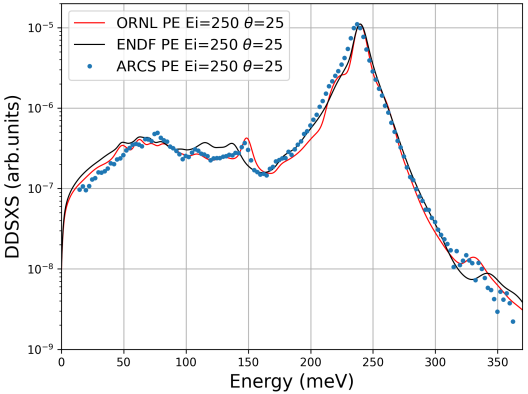
- VISION HDPE, LDPE, and UHMW comparison:



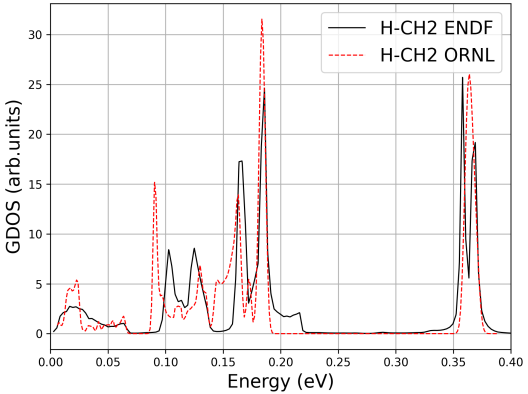
* **ORNL PE has better agreement with the shape of all different PEs!**

Polyethylene evaluation validation- INS measurements at ORNL

- ARCS HDPE comparison:

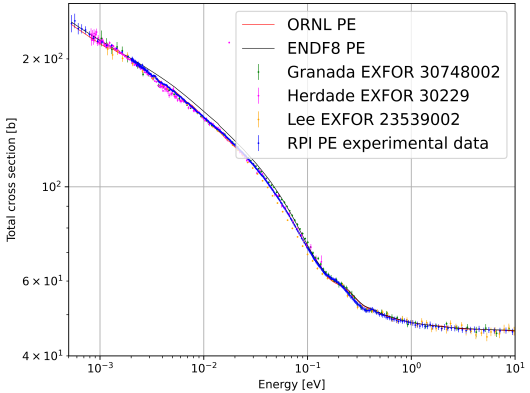


- Phonon spectra comparison:

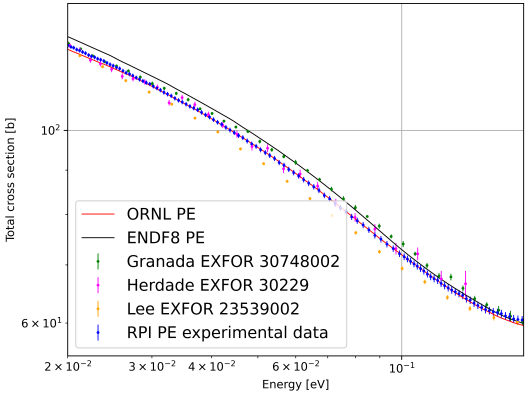


Polyethylene evaluation validation- Transmission measurements at RPI

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

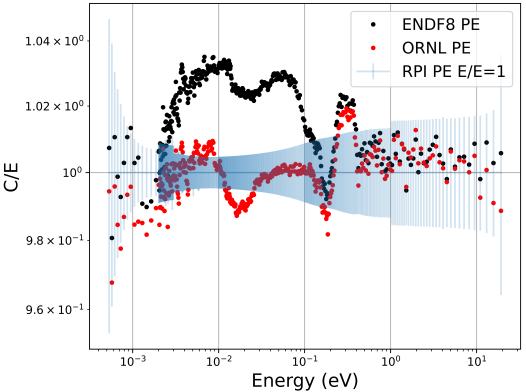


- Total xs comparison (2e-2 to 2e-1 eV):

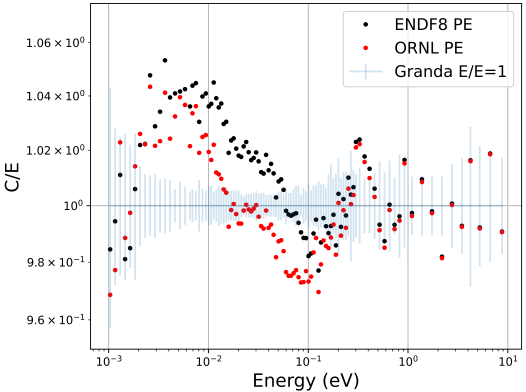


Polyethylene evaluation validation- Transmission measurements at RPI

- C/E comparison to new RPI data:



- C/E comparison to Granda data:



Polyethylene evaluation validation- Pulsed neutron die-away measurements

- New pulsed neutron die-away measurements at LLNL [4]

Experiment			ENDF8			ORNL		
Diameter (cm)	alpha (1/ms)	std (1/ms)	alpha (1/ms)	std (1/ms)	Bias (C-E)/E	alpha (1/ms)	std (1/ms)	Bias (C-E)/E
29.57784	6.946	0.005	6.97088	0.00210	0.00358	6.99815	0.00055	0.00751
25.99247	7.252	0.013	7.23654	0.00086	-0.00213	7.27069	0.00157	0.00258
23.68864	7.532	0.018	7.48085	0.00215	-0.00679	7.52198	0.00193	-0.00133
21.23407	7.791	0.01	7.81726	0.00337	0.00337	7.87079	0.00265	0.01024
18.52972	8.333	0.015	8.38402	0.00282	0.00612	8.45288	0.00606	0.01439
15.61121	9.133	0.028	9.31830	0.00351	0.02029	9.42010	0.01530	0.03144
13.53452	10.187	0.047	10.35499	0.00298	0.01649	10.45767	0.00469	0.02657
10.7503	12.484	0.033	12.68892	0.01861	0.01641	12.82581	0.01330	0.02738
7.75713	17.887	0.035	18.41186	0.00930	0.02934	18.53495	0.01412	0.03622
5.24282	29.273	0.248	30.66189	0.10354	0.04745	31.41043	0.13845	0.07302

- New ORNL evaluation reduces, compared to ENDF8, total neutron scattering cross section resulting in an increase in PNDA α compared to ENDF8. ENDF8 and ORNL calculated α values are within 1-2% of each other.
- 14 MeV neutron source is used, so no other cross section data used besides material in the problem.

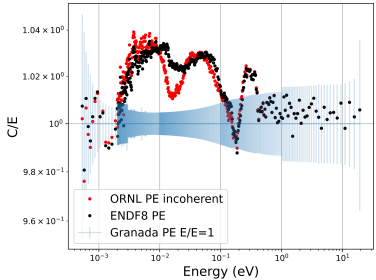
Polyethylene evaluation validation- Integral criticality benchmarks

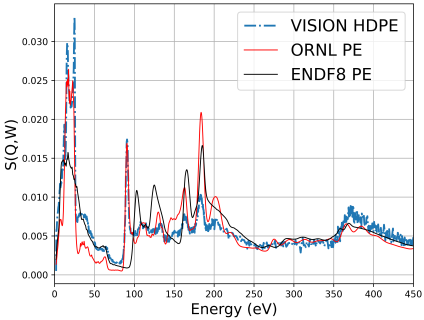
- We have tried to identify all relevant thermal benchmarks that contain Polyethylene in International Handbook of Evaluated Criticality Safety Benchmark Experiments 2021.

	benchmark		ENDF8				ORNL			
name	keff	unc	keff	unc	st.dev away	C/E	keff	unc	st.dev away	C/E
HMT-001-001	1.00100	0.00600	1.00621	0.00022	0.86833	1.0052	1.00705	0.00022	1.00833	1.00604
HMT-008-001	1.00090	0.00520	1.00662	0.00017	1.1	1.00571	1.00776	0.00017	1.31923	1.00685
HMT-009-001	1.00090	0.00630	1.04156	0.00017	6.45396	1.04062	1.04303	0.00017	6.6873	1.04209
HMT-013-001	1.00060	0.00220	1.0073	0.00016	3.04545	1.00669	1.00782	0.00016	3.28181	1.00721
HMT-018-001	1.00380	0.00410	0.99985	0.00017	0.96341	0.99606	1.00076	0.00017	0.74146	0.99697
PMM-002-004	1.00083	0.00287	1.00335	0.00009	0.878	1.00252	1.00381	0.00009	1.03833	1.00298
PMM-002-005	0.9994	0.00287	1.00554	0.00009	2.13937	1.00614	1.00651	0.00009	2.47735	1.00711
PMM-003-004	1.00116	0.00142	1.00299	0.00008	1.28873	1.00183	1.00282	0.00008	1.16901	1.00166

- New ORNL evaluation reduces, compared to ENDF8, total neutron scattering cross section resulting in an increase in neutron multiplication factor k_{eff}. ORNL library is demonstrably more accurate on differential and transmission level. For most benchmarks k_{eff} for calculated values is anywhere from 300- 4000 pcm away from experimental values, and this could be related specific HDPE measured, and points to a question how sensitive these measurements are to TSL.

- Optimizing the cross section with respect to ENDF:

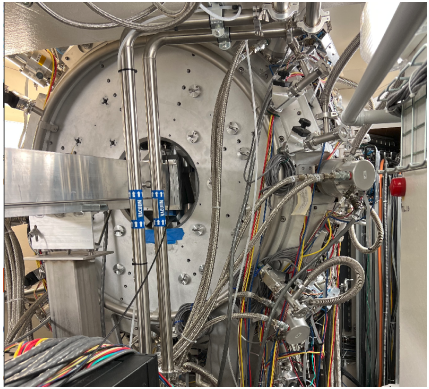




	benchmark		ENDF8				ORNL			
name	keff	unc	keff	unc	st.dev away	C/E	keff	unc	st.dev away	C/E
HMT-001-001	1.00100	0.00600	1.00621	0.00022	0.86833	1.0052	1.00613	0.00022	0.855	1.00512
HMT-008-001	1.00090	0.00520	1.00662	0.00017	1.1	1.00571	1.00656	0.00017	1.08846	1.00565
HMT-009-001	1.00090	0.00630	1.04156	0.00017	6.45396	1.04062	1.04181	0.00017	6.49365	1.04087
HMT-013-001	1.00060	0.00220	1.0073	0.00016	3.04545	1.00669	1.00710	0.00016	2.95455	1.00649
HMT-018-001	1.00380	0.00410	0.99985	0.00017	0.96341	0.99606	0.99999	0.00017	0.74146	0.9962
PMM-002-004	1.00083	0.00287	1.00335	0.00009	0.878	1.00252	1.00322	0.00009	0.83275	1.0023
PMM-002-005	0.9994	0.00287	1.00554	0.00009	2.13937	1.00614	1.00537	0.00009	2.08014	1.00597
PMM-003-004	1.00116	0.00142	1.00299	0.00008	1.28873	1.00183	1.00291	0.00008	1.23239	1.00175

Transmission measurements at ORNL

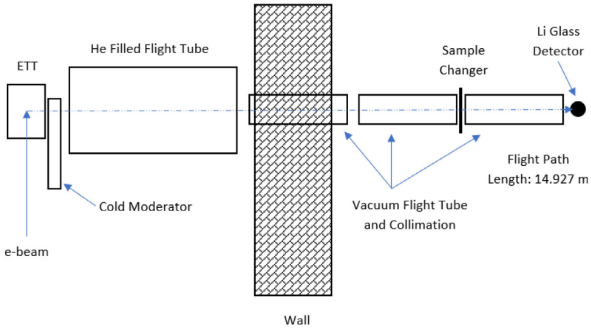
- As seen in Chris' talk and here, having differential inelastic neutron scattering (phonon) measurements as well as transmission measurements is important for validation, but also evaluation of TSL libraries.
- VISION instrument at Spallation Neutron Source at ORNL has been traditionally only used for INS measurements, but due to it's unique (indirect neutron spectrometer with incident white beam) capabilities it could be possibly used for transmission measurement in thermal region.
- It utilizes general-user proposal system for access to beam-time, and if the transmission capability could be developed, it would be of great benefit of NCSP and other programs to whom TSL libraries are of interest, with almost no cost.
- We have started work with the VISION instrument scientist to develop transmission capability as well.



Using VISION for transmission comes with challenges due to it unorthodox transmission setup.

Transmission measurements at ORNL

- RPI transmission setup:



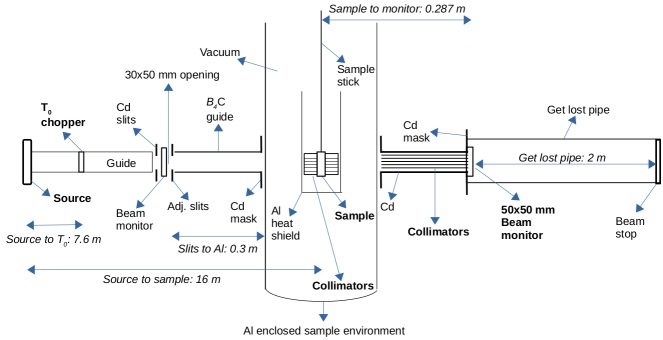
- Long distance ($\approx$ 15m from sample to detector) and well collimated beam
- Transmission calculated as:

$$T = \frac{C_s - B_s}{C_o - B_o} \quad (1)$$

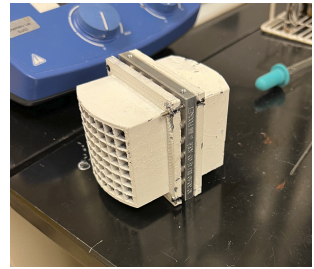
- For background characterization “double notch” technique is used (Cadmium and Indium)

Transmission measurements at ORNL

- ORNL VISION transmission setup:



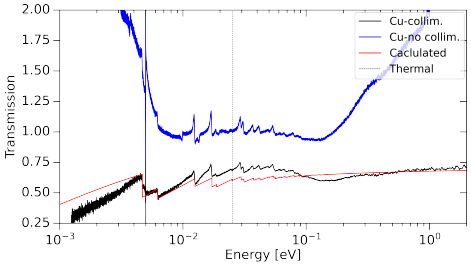
- Short distance (≈ 0.3m from sample to detector). Needs collimation (nylon 3D printed collimator painted by boron nitride paint):



- For background characterization “double notch” technique is used (Cadmium, Indium, and Erbium)
- Temperatures from ≈ 5 - 1300 K.

Transmission measurements at ORNL

- Measured transmission for Copper at VISION:

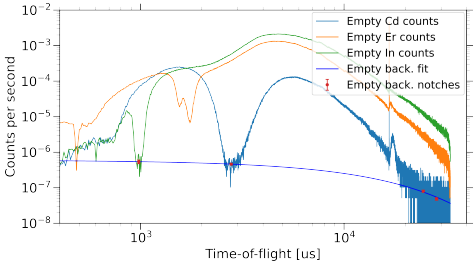


- No background was subtracted here.
- Collimator was not “black” enough.

Future work:

+ 3D print Boron Carbide collimators.
+ Obtain thicker Erbium and Cadmium notches.
+ Automate orientation of sample stick.

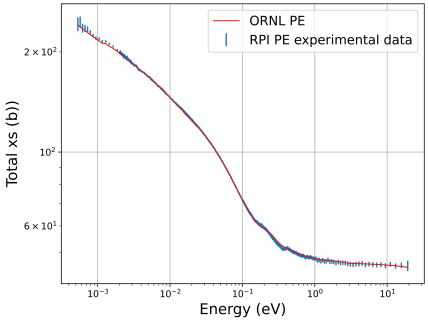
- Characterization of background:



- Erbium notch was not “black” enough.

Conclusions

- Literature review and initial modeling of concrete components have been performed. INS and transmission measurements of concrete and hydrated cement have been also planned.
- A new TSL library for Polyethylene (including C in PE files) was created and submitted to NNDC for inclusion in ENDF8.1. The library has been thoroughly validated with differential and transmission experimental data, and possibly points to the fact that integral benchmarks are specific to the material measured as well being not sensitive enough for validation of different TSLs.
- We have started developing a new transmission capability at ORNL that would greatly benefit NCSP program, and come at almost no cost.



Relevant experimental characterization needs to be part of the evaluation of new ENDF libraries as well as validation.

Acknowledgements

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- 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

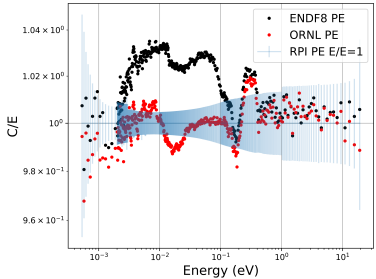
[1] A. A. Bahraq et al., “Molecular Simulation of Cement-Based Materials and Their Properties”, Engineering, Volume 15, 2022, Pages 165-178, https://doi.org/10.1016/j.eng.2021.06.023.

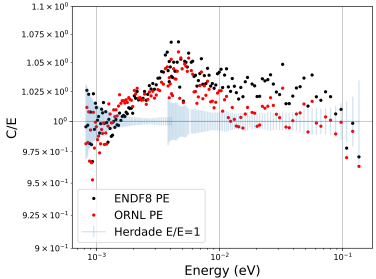
[2] K. Ramić et al., “Thermal Neutron Scattering Evaluation for Polyethylene from RPI/ORNL/ESS ”, CSEWG2021, Available at https://indico.bnl.gov/event/13121/contributions/57171/

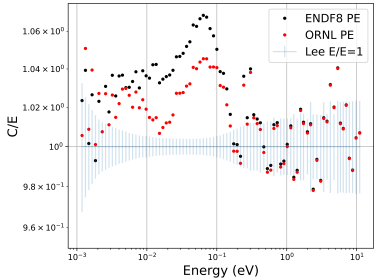
[3] C. Chapman et al., “Thermal Neutron Scattering Research at ORNL – ENDF File Validation”, NCSP TPR2022, Available at https://ncsp.llnl.gov/sites/ncsp/files/2022-02/59_tns_research_at_ornl_2022_tpr_final.pdf

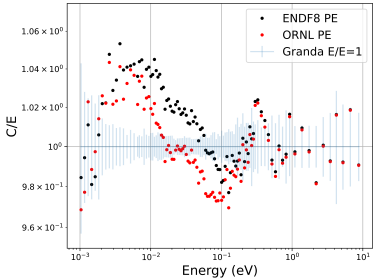
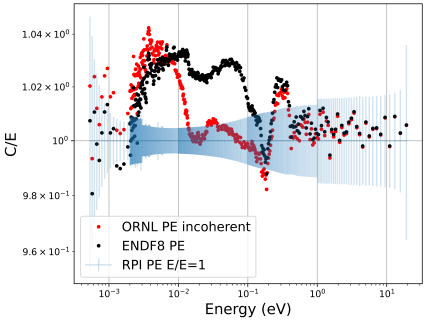
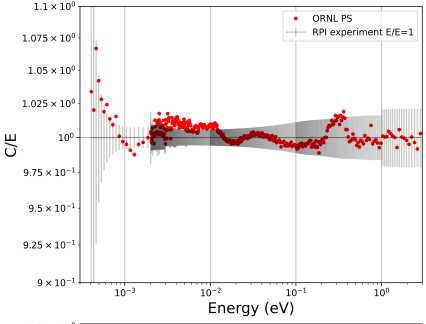
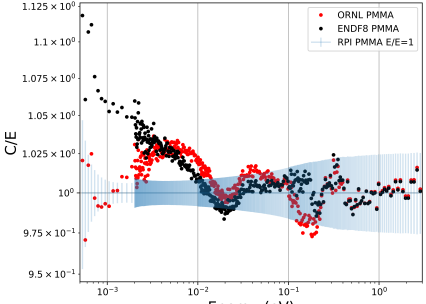
[4] D. Siefman, “Pulsed-Neutron Die-Away Experiments for Plastics”, ND2022, Available at https://indico.frib.msu.edu/event/52/contributions/943/

Backup slides









Backup slides