Status of ORNL TSL evaluations

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Scattering cross section (xs) components in thermal range

- 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:

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

where:

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

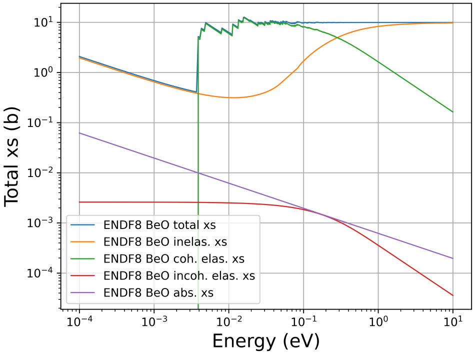
with $\rho(\beta)$ as the phonon spectrum.

- Coherent elastic scattering:

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

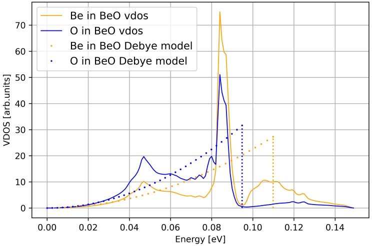
- Incoherent elastic scattering:

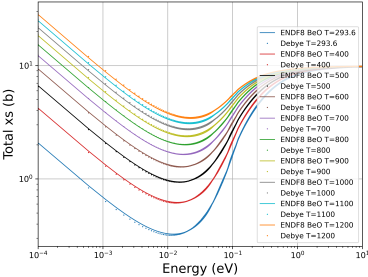
$$\sigma^{incoh} = \frac{\sigma_b}{2}\left(\frac{1-e^{-4WE}}{2WE}\right) \quad (4)$$

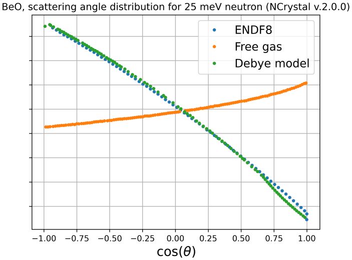


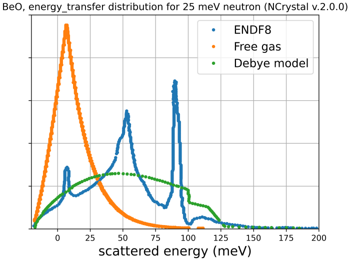
- **As we can see all the scattering components are dependent on the inelastic physics through $\rho(\beta)$ and W (Debye-Waller factor)**

The Physics of Thermal Neutron Inelastic Scattering





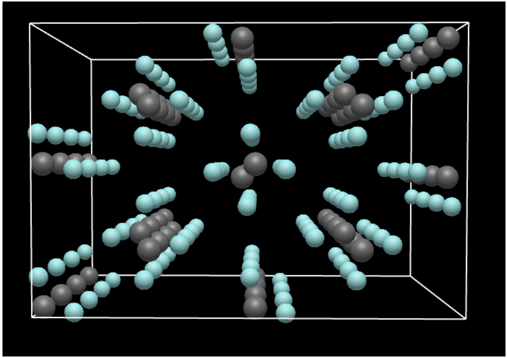




Preliminary work on Teflon

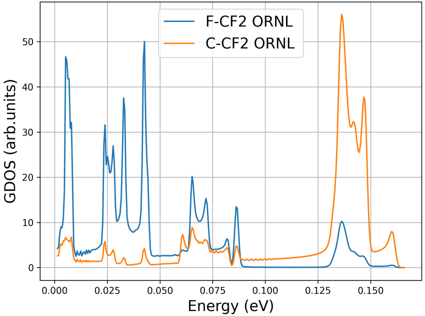
- Polytetrafluoroethylene (PTFE, Teflon) is a fluorocarbon solid, as it is a high-molecular-weight polymer consisting wholly of carbon and fluorine. PTFE crystallizes in orthorombic P_{nma} space group 63 (just like polyethylene).
- To verify both MD and AIMD calculations were performed. The chains of PTFE were heated above the melting point, equilibriated, and cooled down.

- PTFE structure:

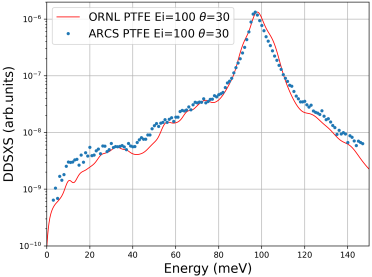


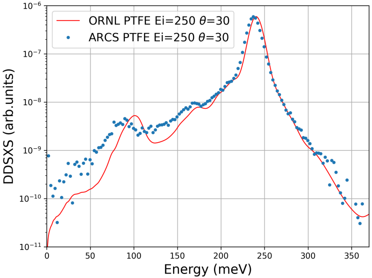
Preliminary work on Teflon

- Frozen phonon calculations for C and F in PTFE were performed using VASP, Phonopy and oClimax. The resulting phonon spectra:



- Inelastic neutron scattering (INS) measurements at ORNL SNS:





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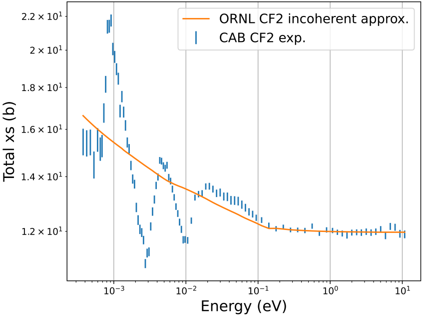
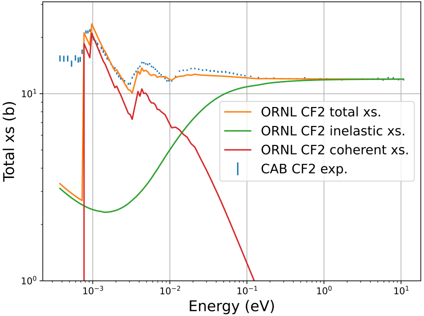
Preliminary work on Teflon

- PTFE in incoherent approximation and with coherent elastic:





- We ran some fast spectra benchmarks (as expected there is no impact):

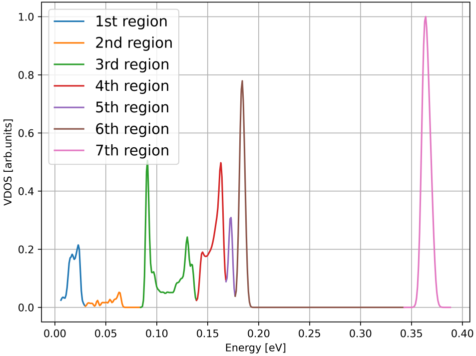
	benchmark		ENDF8 free gas				ORNL			
name	keff	unc	keff	unc	st.dev away	C/E	keff	unc	st.dev away	C/E
HMF-007-032	0.9941	0.0012	1.00445	0.00013	8.625	1.01041	1.00445	0.00013	8.625	1.01041
HMF-007-033	0.9977	0.0019	1.01353	0.00013	8.33157	1.01586	1.01353	0.00013	8.33157	1.01586
HMF-007-034	0.9959	0.0017	1.01669	0.00013	12.22941	1.02087	1.01669	0.00013	12.22941	12.22941

- The library was submitted to NNDC for inclusion in ENDF8.1
- Planned new INS and transmission measurements at VISION and RPI.

Polyethylene (PE) evaluation

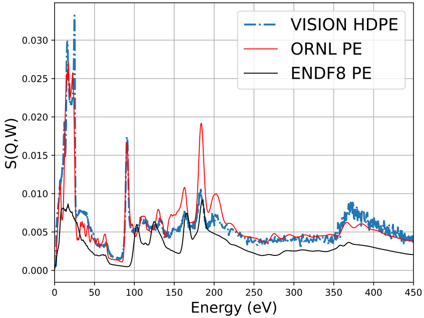
- Motivation:
  - New transmission measurements at RPI, as well as criticality benchmarks from LLNL.
  - High Density Polyethylene (HDPE) is a up to 80% crystalline material, and it crystalizes in the same structure as Teflon.
  - ORNL PE evaluation was optimized with respect to differential measurement at VISION spectrometer at SNS, as well as transmission measurement from RPI. See Chris talk, as well as [1,2].

- Optimization summary:
  - We used NCrystal [3] and Dakota [4] to minimize χ^2 between the RPI transmission data and the evaluation. The weights for each distinct region of GDOS were varied by Dakota, where total cross section is calculated by NCrystal at exactly same energy grid as the experimental cross section data, and χ^2 was calculated. The process was repeated until optimal minimal difference in χ^2 was achieved. We have further expanded the method by inclusion of VISION neutron spectrometer experimental data in the process. 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 procedure. The method described in [2] has been adapted to work with NCrystal.



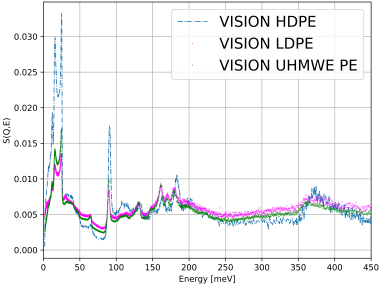
Polyethylene evaluation validation- INS measurements at ORNL

- VISION HDPE comparison:

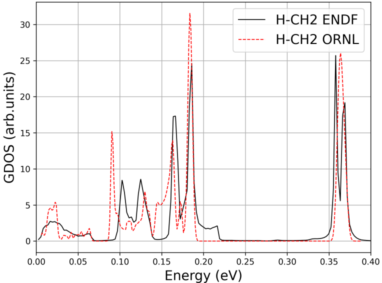


- ORNL PE has better agreement with the shape of all different PEs!

- VISION HDPE, LDPE, and UHMW comparison:



- Phonon spectra comparison:



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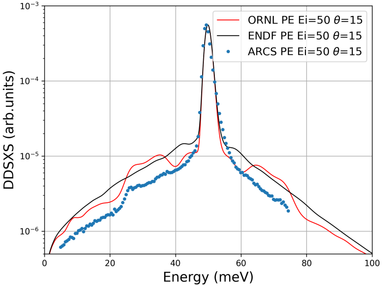
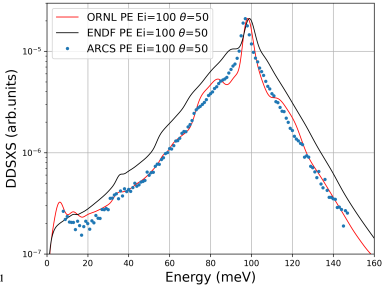
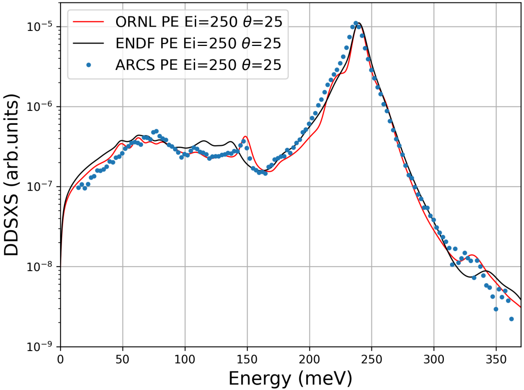
Polyethylene evaluation validation- INS measurements at ORNL

- ARCS HDPE comparison:



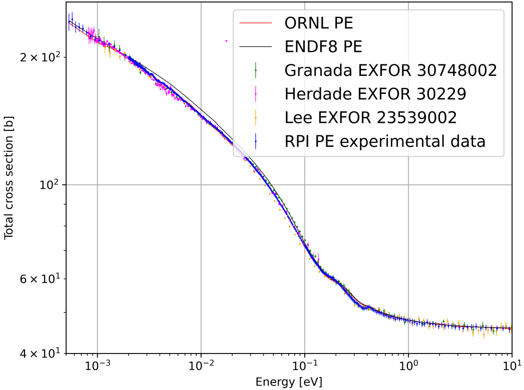




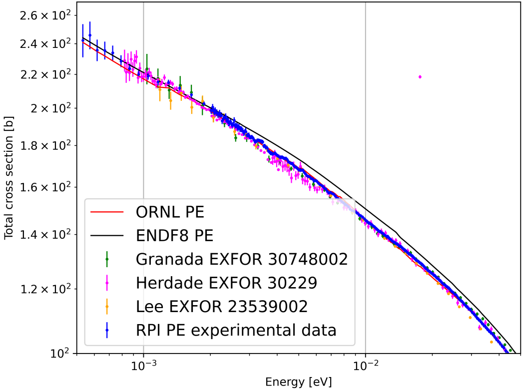
- ORNL PE has better agreement with DDSXS data from ARCS.

Polyethylene evaluation validation- Transmission measurements at RPI

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



- Total xs comparison (5e-4 to 5e-2 eV):



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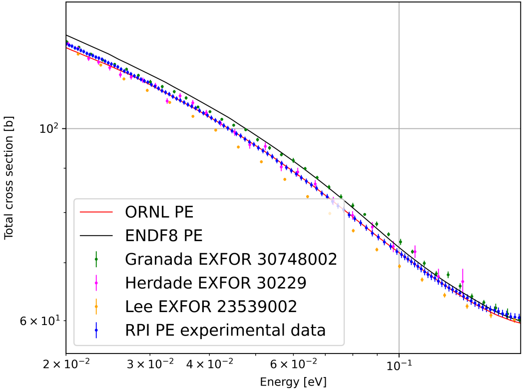
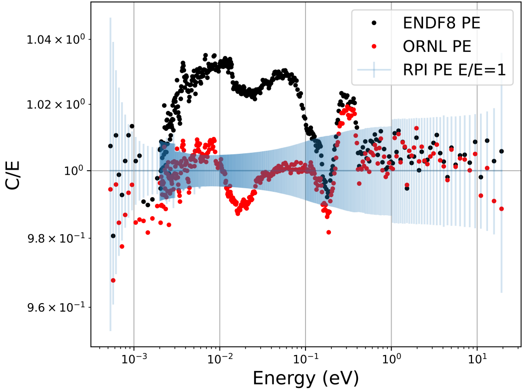
Polyethylene evaluation validation- Transmission measurements at RPI

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



- C/E comparison to new RPI data:



Polyethylene evaluation validation- Pulsed neutron die-away measurements

- New pulsed neutron die-away measurements at LLNL [5]

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

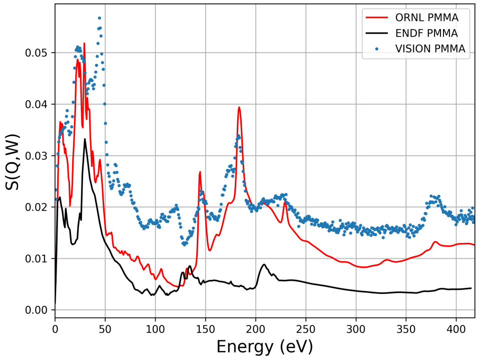
- We have tried to identify all relevant thermal benchmarks that contain Polyethylene in International Handbook of Evaluated Criticality Safety Benchmark Experiments 2020, as well as new preliminary LLNL benchmarks [6] (thanks to Catherine Percher).

	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
PMT-004-001	1.00000	0.00150	0.99799	0.00008	1.34	0.99799	0.99974	0.00008	0.17333	0.99974
PMT-004-002	1.00000	0.00150	0.99959	0.00008	0.27333	0.99959	1.00276	0.00008	1.84	1.00276

- New ORNL evaluation reduces, compared to ENDF8, total neutron scattering cross section resulting in an increase in neutron multiplication factor k_{eff}.
- Note: PMT-004-001 has thinner PE (better agreement with ORNL) and PMT-004-002 has thicker sheets of PE (better agreement with ENDF-8. Thinner sheets are expected to be more sensitive to variation in scattering energy and angle distributions.
- Since we saw the same increase in PNDA calculated values, where only material cross sections were used, we suspect this increase possibly comes from erroneous cross sections at energies above thermal region.

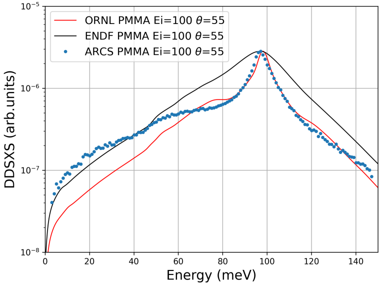
Lucite (PMMA) evaluation validation- INS measurements at ORNL

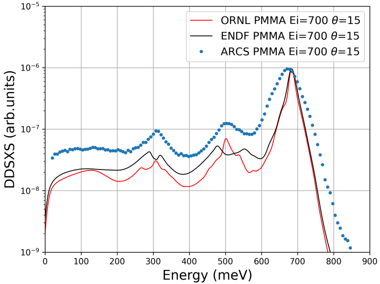
- VISION PMMA comparison:



- ORNL PMMA has better agreement with differential INS data!

- ARCS PMMA comparison:





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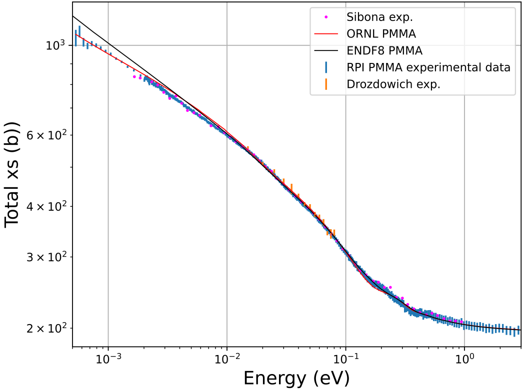
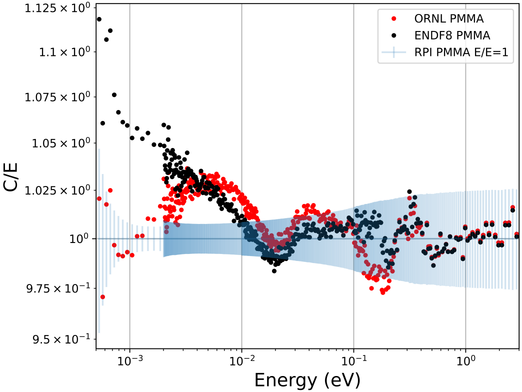
Lucite (PMMA) evaluation validation- Transmission measurements at RPI

- Total xs comparison :



- C/E comparison to new RPI data:



Lucite (PMMA) evaluation validation- Pulsed neutron die-away measurements

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
25.4632	5.896	0.006	6.01071	0.00163	0.01946	5.95977	0.00310	0.01082
22.9171	6.365	0.008	6.42224	0.00077	0.00899	6.35454	0.00188	-0.00164
20.3707	6.884	0.009	6.98083	0.00167	0.01407	6.90339	0.00221	0.00282
17.8373	7.653	0.008	7.79460	0.00692	0.01850	7.67492	0.00122	0.00286
15.2441	8.821	0.01	9.00550	0.00372	0.02092	8.86147	0.00278	0.00459
12.7057	10.611	0.009	10.99313	0.00367	0.03601	10.77586	0.00659	0.01554
10.1765	13.804	0.01	14.31974	0.02078	0.03736	13.99727	0.00868	0.01400
7.6267	20.051	0.05	21.31716	0.01373	0.06315	20.58287	0.03546	0.02653
5.08	33.911	0.18	38.59880	0.08583	0.13824	35.35671	0.08024	0.04263

- New ORNL evaluation calculates PNDA α significantly better compared to ENDF8, especially at smaller diameters, where sensitivity to TSL is increased.

Lucite (PMMA) evaluation validation- Integral criticality benchmarks

	benchmark		ENDF8				ORNL			
name	keff	unc	keff	unc	st.dev away	C/E	keff	unc	st.dev away	C/E
HST-003-001	1.0016	0.0056	1.00271	0.00023	0.19821	1.00111	1.00283	0.00022	0.21964	1.00123
HST-003-002	1.0016	0.0057	1.00293	0.00023	0.23333	1.00133	1.00322	0.00022	0.28421	1.00162
HST-003-003	1.0014	0.0056	1.00015	0.00027	0.22321	0.99875	1.00014	0.00027	0.22500	0.99874
HST-003-004	1.0009	0.0057	1.00283	0.00026	0.33860	1.00193	1.00248	0.00025	0.27719	1.00158
HST-003-005	1.0021	0.0068	0.99615	0.00027	0.87500	0.99406	0.99598	0.00028	0.90000	0.99389
HST-003-006	1.0013	0.007	0.99869	0.00027	0.37286	0.99739	0.99865	0.00027	0.37857	0.99735
HST-003-007	1.0006	0.0046	1.00339	0.00022	0.60652	1.00279	1.00354	0.00023	0.63913	1.00294
HST-003-008	1.0003	0.0033	1.00181	0.00026	0.45758	1.00151	1.00154	0.00026	0.37576	1.00124
HST-003-009	0.9996	0.0035	1.00584	0.00026	1.78286	1.00624	1.00579	0.00027	1.76857	1.00619
HST-003-010	1.0011	0.005	0.99659	0.00028	0.90200	0.99549	0.99648	0.00028	0.92400	0.99539
HST-003-011	0.9997	0.0052	1.00222	0.00027	0.48462	1.00252	1.0027	0.00027	0.57692	1.00300
HST-003-012	1.0006	0.0047	1.00166	0.00023	0.22553	1.00106	1.00169	0.00023	0.23191	1.00109
HST-003-013	1.0004	0.0059	1.00268	0.00023	0.38644	1.00228	1.00237	0.00023	0.33390	1.00197
HST-003-014	1.0005	0.0047	1.00225	0.00023	0.37234	1.00175	1.00233	0.00023	0.38936	1.00183
HST-003-015	1	0.0045	0.99567	0.00023	0.96222	0.99567	0.99535	0.00023	1.03333	0.99535
HST-003-016	1.0002	0.0037	0.99963	0.00026	0.15405	0.99943	0.99998	0.00026	0.05946	0.99978
HST-003-017	0.9994	0.0042	1.00622	0.00026	1.62381	1.00682	1.00593	0.00026	1.55476	1.00653
HST-003-018	1.0009	0.005	0.99546	0.00027	1.08800	0.99456	0.99553	0.00028	1.07400	0.99463
HST-003-019	0.9991	0.0059	1.00539	0.00027	1.06610	1.00630	1.00521	0.00026	1.03559	1.00612
PCM-002-010	1	0.0044	1.03134	0.00021	7.12273	1.03134	1.02976	0.00022	6.76364	1.02976
PCM-002-023	1	0.0046	1.00871	0.00023	1.89348	1.00871	1.00766	0.00023	1.66522	1.00766
PMT-004-003	1	0.0015	1.00634	0.00009	4.22667	1.00634	1.00402	0.00009	2.68000	1.00402
PMT-004-004	1	0.0015	1.0055	0.00008	3.66667	1.00550	1.00282	0.00008	1.88000	1.00282

- ORNL has better agreement with differential experimental data, as well as with new LLNL (by $\approx$ 200-300 pcm) and most old benchmarks.

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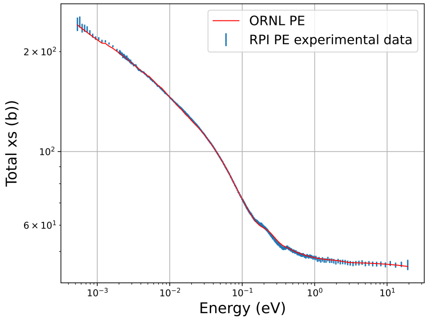
Conclusions

- A new TSL library for Teflon was created and submitted to NNDC for inclusion in ENDF8.1. More experimental characterization is 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 experimental data, and possibly points to issues with other cross section data for integral benchmarks.
- A new TSL library for Lucite (including C and O in PMMA files) was created and submitted to NNDC for inclusion in ENDF8.1. The library has been thoroughly validated with differential experimental data, as well with integral benchmarks.
- Proposed methodology for evaluation and validation has been successfully applied and demonstrated on Polyethylene and Lucite TSL libraries.



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.
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- 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] 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/

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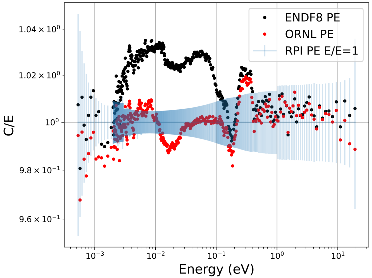
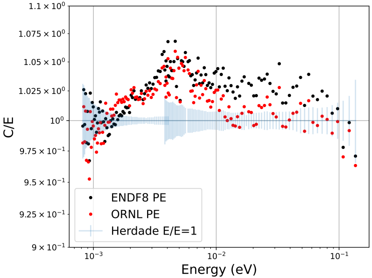
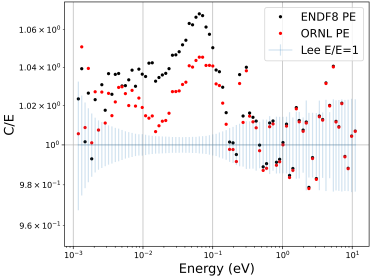
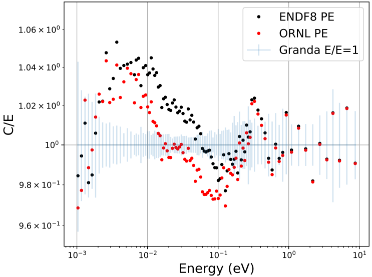
Backup slides









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