Proposed Methodology for Evaluating and Validating TSLs

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CSEWG2022 – 1 November 2022

Overview

- Recent efforts in validating thermal neutron scattering cross sections:
  - Differential cross section measurements at ORNL for:
    - Evaluation [1]
    - Validation [2,3,4,5]
  - Total cross section measurements at RPI [6,7]
  - Pulsed-neutron die-away experiments at LLNL [8]
  - Integral criticality experiments by LLNL [9]
- Here, we propose a methodology for not only validating thermal scattering files that utilizes all available experimental data, but also for evaluating new libraries as demonstrated on polystyrene

Evaluation

- Initial configuration of polystyrene (C_8H_8) used to calculate phonon properties using VASP, Phonopy, and OClimax
- Phonon density of states (PDOS) was then optimized using NCrystal [11] and Dakota [12] using the following:
  - Process PDOS using NCrystal to calculate both the total cross section and VISION spectra
  - Calculate χ^2 of these datasets compared to experimentally measured total cross section and VISION spectra
  - Use Dakota to vary the PDOS based on the calculated χ^2 metric until convergence is achieved
- Samples in experiment were of varying chain lengths & tacticity, which previously showed no impact on cross section [13]

Evaluation

- PDOS chosen to vary because it affects all forms of thermal neutron scattering:

$$\frac{d^2\sigma}{dE_f d\Omega} = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}}\, e^{\frac{-\beta}{2}}\, S(\alpha,\beta)$$

  - Inelastic

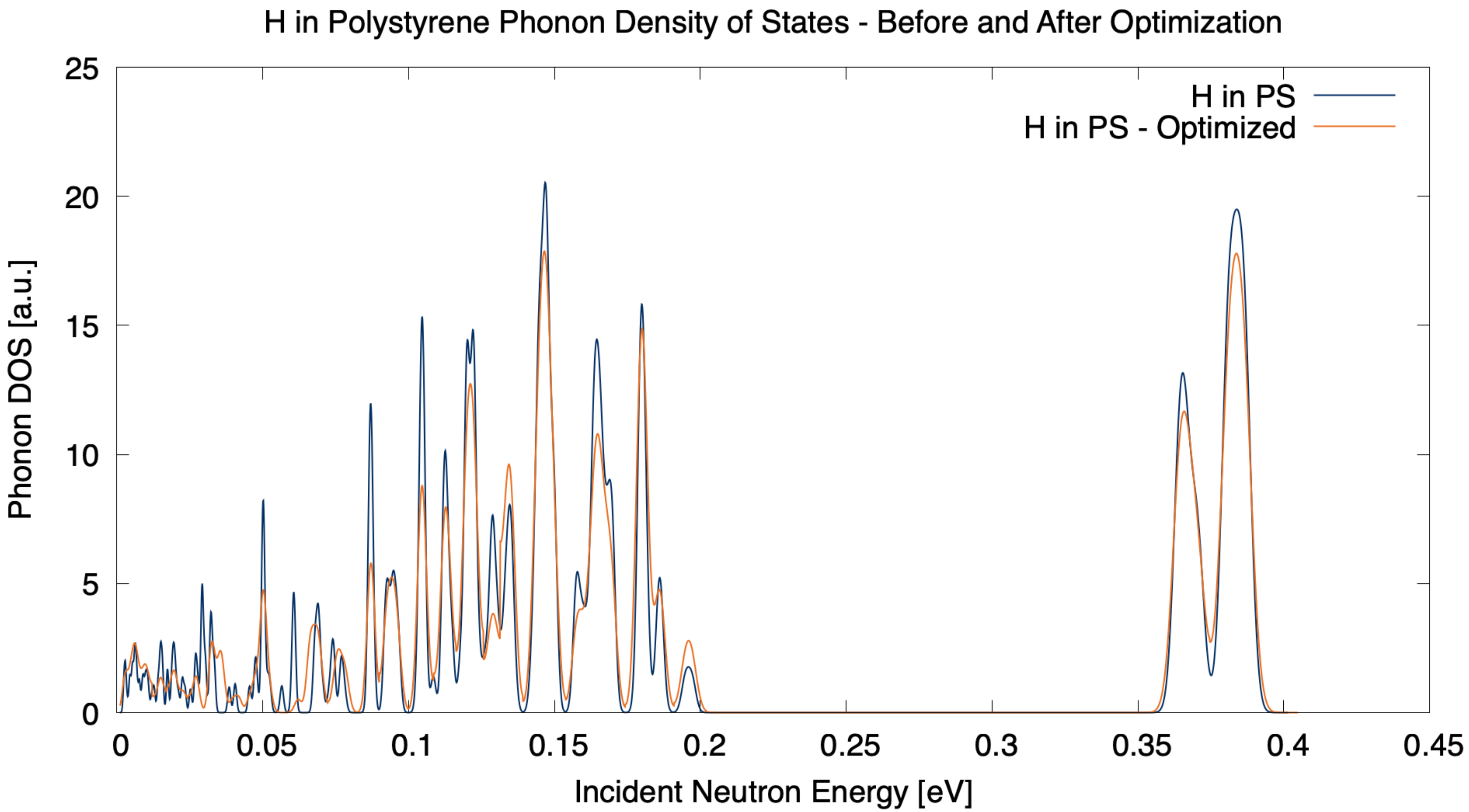
$$S(\alpha,\beta) = e^{-\alpha\lambda}\sum_{n=0}^{\infty}\frac{\alpha^n}{n!}\frac{1}{2\pi}\int_{-\infty}^{\infty} d\hat{t}\, e^{i\beta\hat{t}}\left[\int_{-\infty}^{\infty} d\beta'\, P(\beta')e^{i\beta'\hat{t}}e^{-\beta'/2}\right]$$

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)}$$

  - Elastic

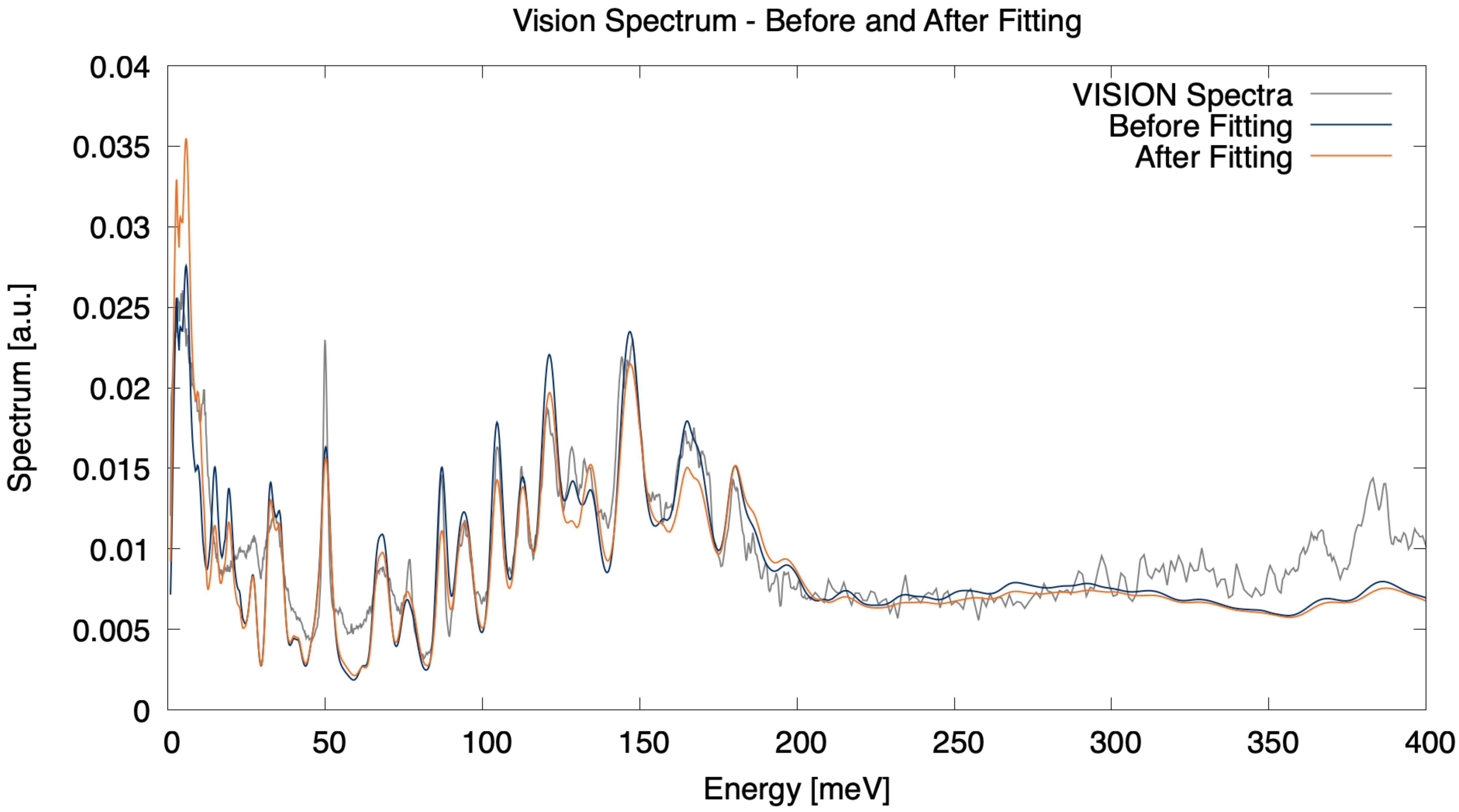
$$\sigma_{el}^{inc}(E) = \frac{\sigma_b}{2}\left(\frac{1-e^{4WE}}{2WE}\right) \qquad \sigma_{el}^{coh}(E) = \frac{\sigma_c}{E}\sum_{E_i<E} f_i e^{4WE_i} \qquad W = \frac{\int_{-\infty}^{\infty} d\beta\, P(\beta)e^{-\beta/2}}{AkT}$$

Evaluation



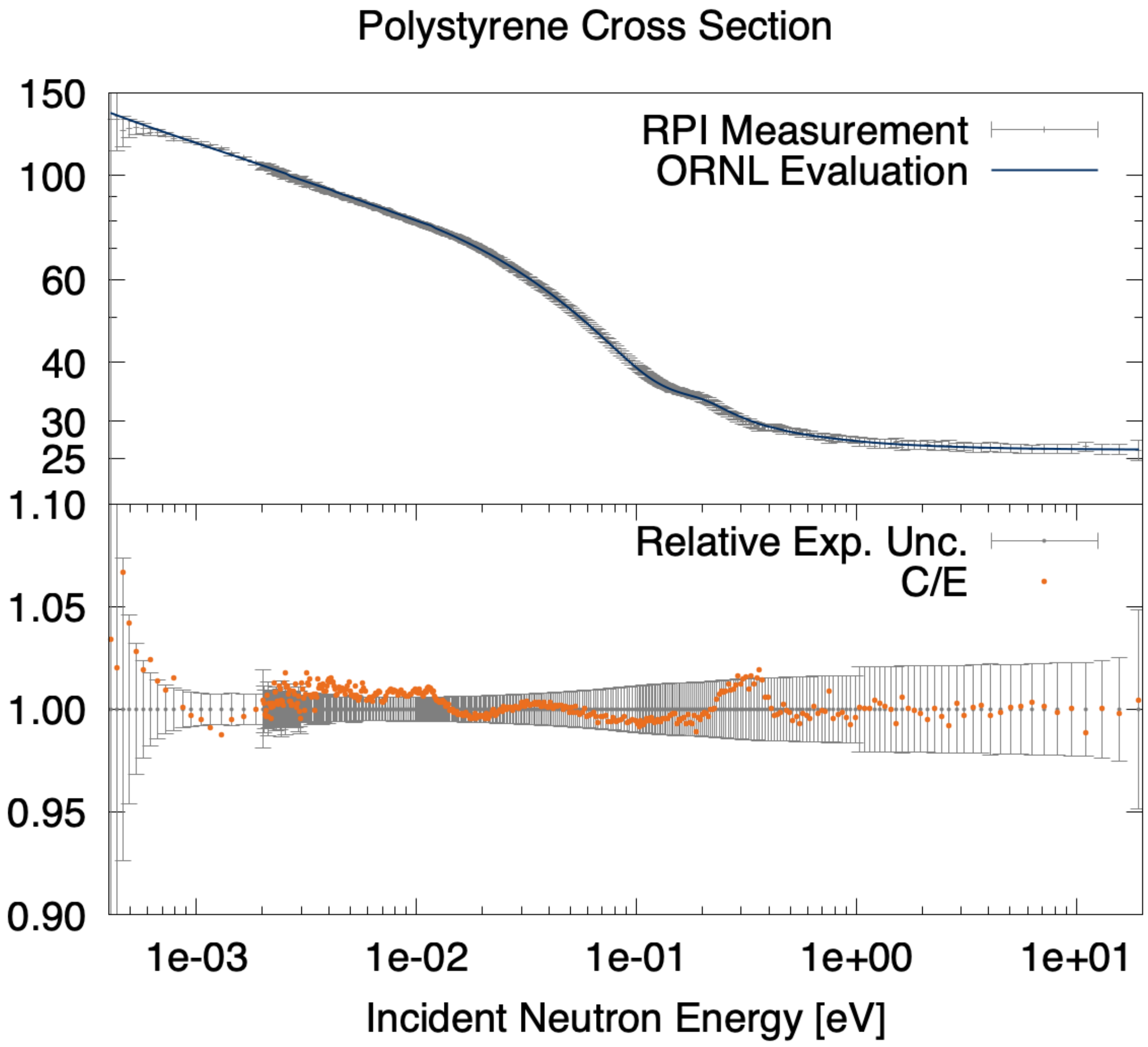
H in Polystyrene Phonon Density of States - Before and After Optimization
H in PS
H in PS - Optimized
Phonon DOS [a.u.]
Incident Neutron Energy [eV]
0
5
10
15
20
25
0
0.05
0.1
0.15
0.2
0.25
0.3
0.35
0.4
0.45

Evaluation – Differential Scattering



Evaluation – Total Cross Section

- Comparison of evaluation to experiment straightforward
- Transmission measurements carried out at RPI
  - Used for evaluation; cannot be used for validation
- Ideally, multiple samples/experiments would be used
  - Some for validation, some for evaluation



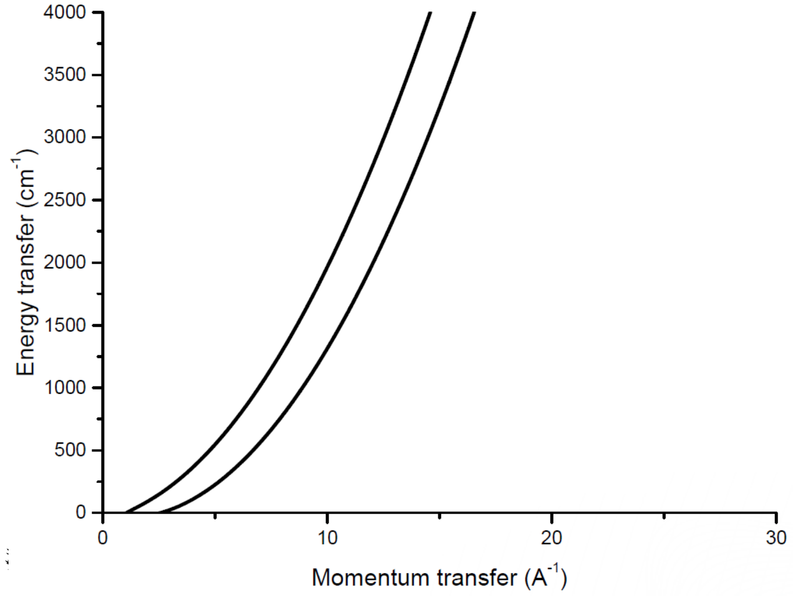
Validation – Double Differential Scattering Cross Sections

- Direct comparison to double differential measurements is difficult for several reasons:
  - Large number of datapoints (~1e5-1e6 points per incident energy)
  - Experimental uncertainties are normally very small, leading to unrealistic chi-squared values
- Several ways to combat these issues
  - Slice data into several 2-D sets (e.g., DDXS at specific scattering angles)
  - Integrate data over angle/energy
- Additionally, visual representation of comparisons of 3-D data not aesthetically appealing
  - Currently, we integrate over Q to generate a 2-D set

Validation – Double Differential Scattering Cross Sections

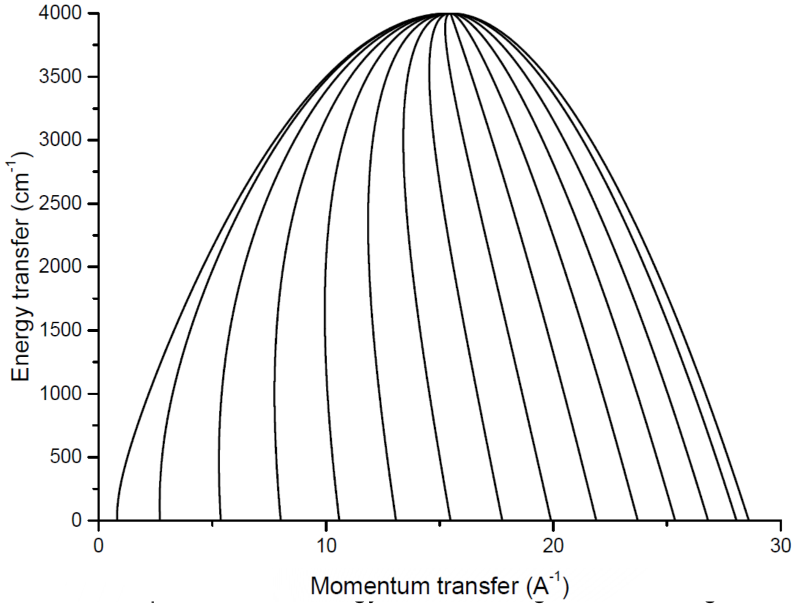
- Indirect Geometry
  - Currently used for evaluation
- White beam of neutrons hits sample
- Scattered neutrons reflected off graphite blocks to two detectors for forward- and backward-scattering angles
- Graphite blocks configured to scatter neutrons at 4 meV

- Direct Geometry
  - Currently used for validation
- User chooses incident energy; Fermi choppers rotate to select energy from white beam
- Detector & data acquisition system (DAS) setup measures final energy and scattering angle

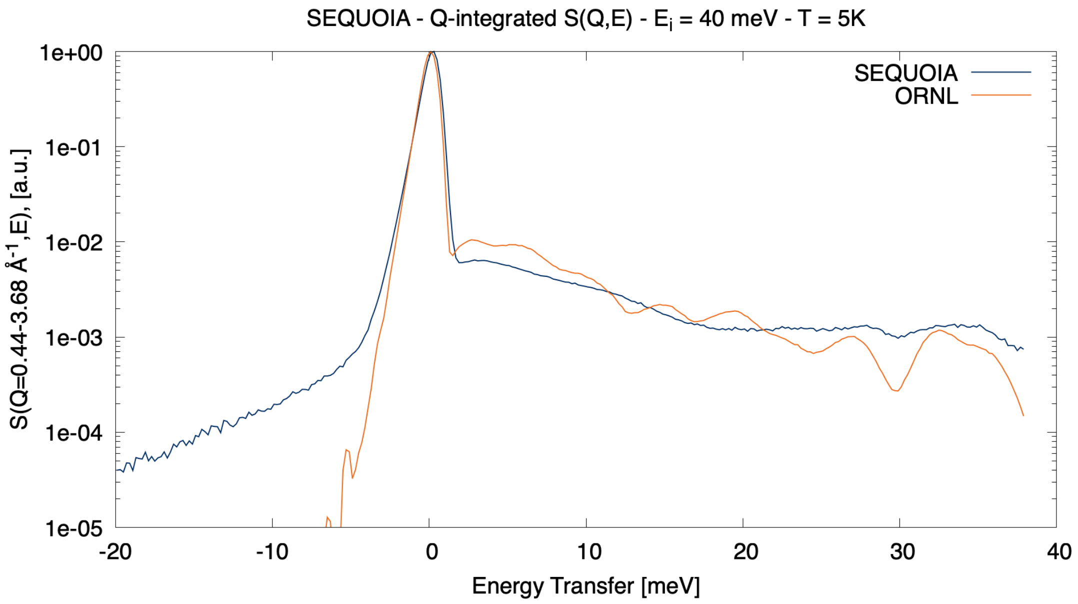


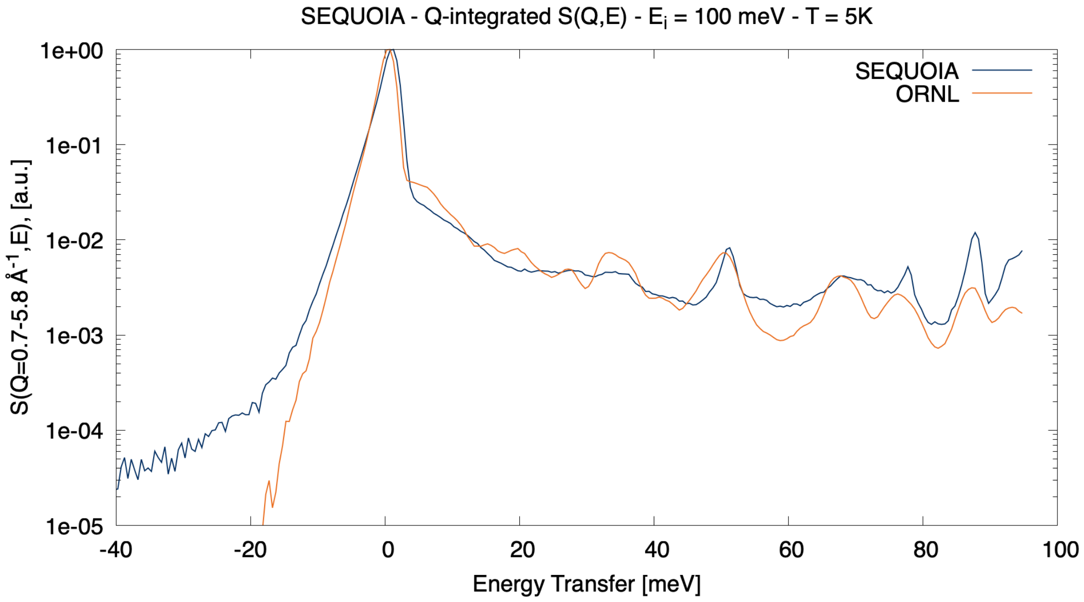
Complimentary measurement techniques

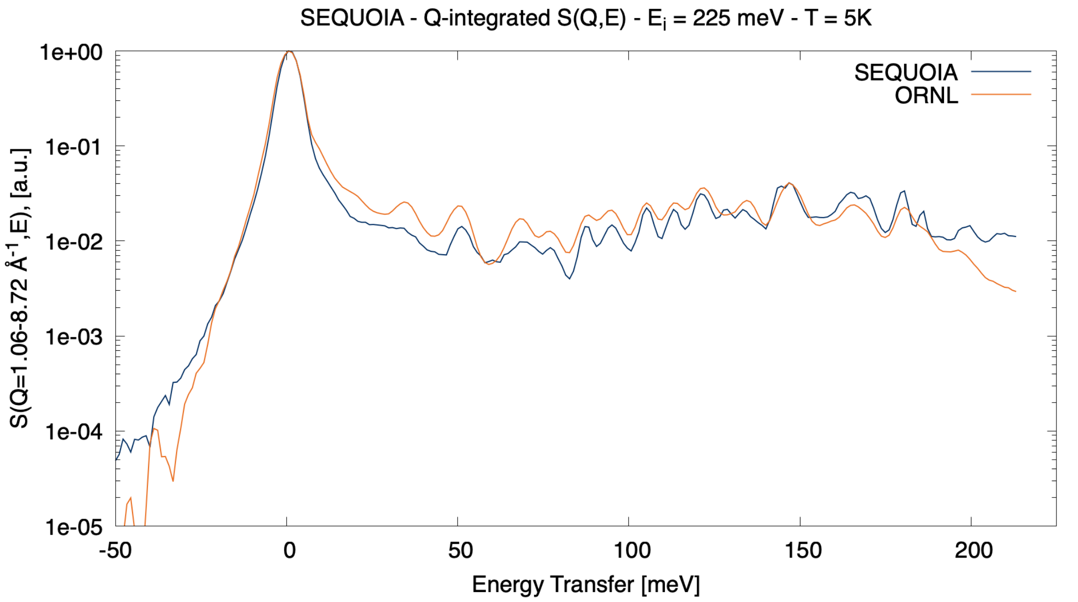
$$S(Q,E) = \frac{1}{k_B T} e^{\frac{-\beta}{2}} S(\alpha,\beta)$$

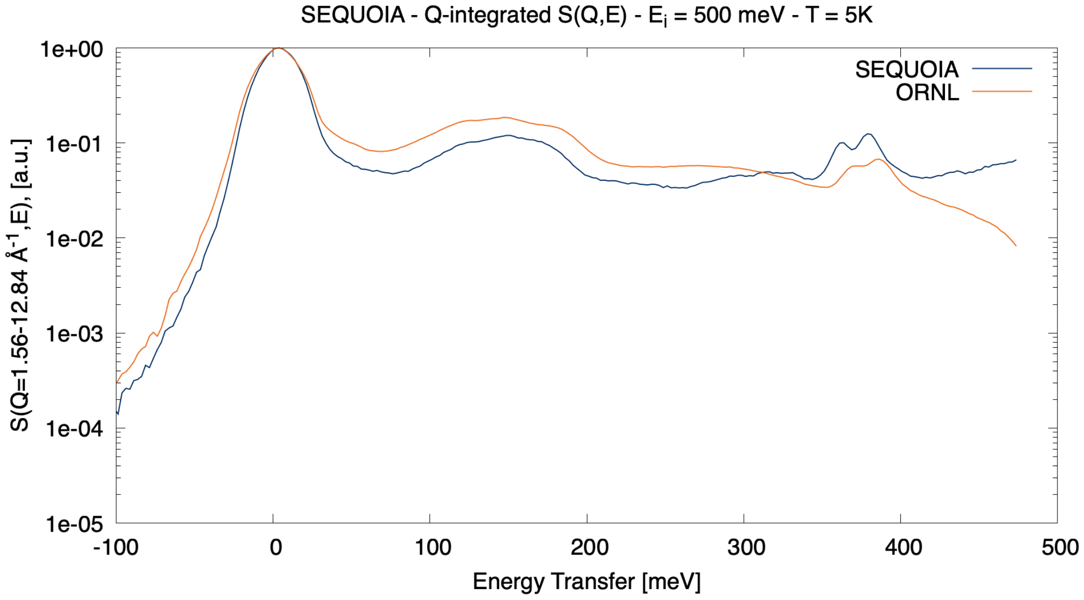


Validation – Double Differential Scattering Cross Sections



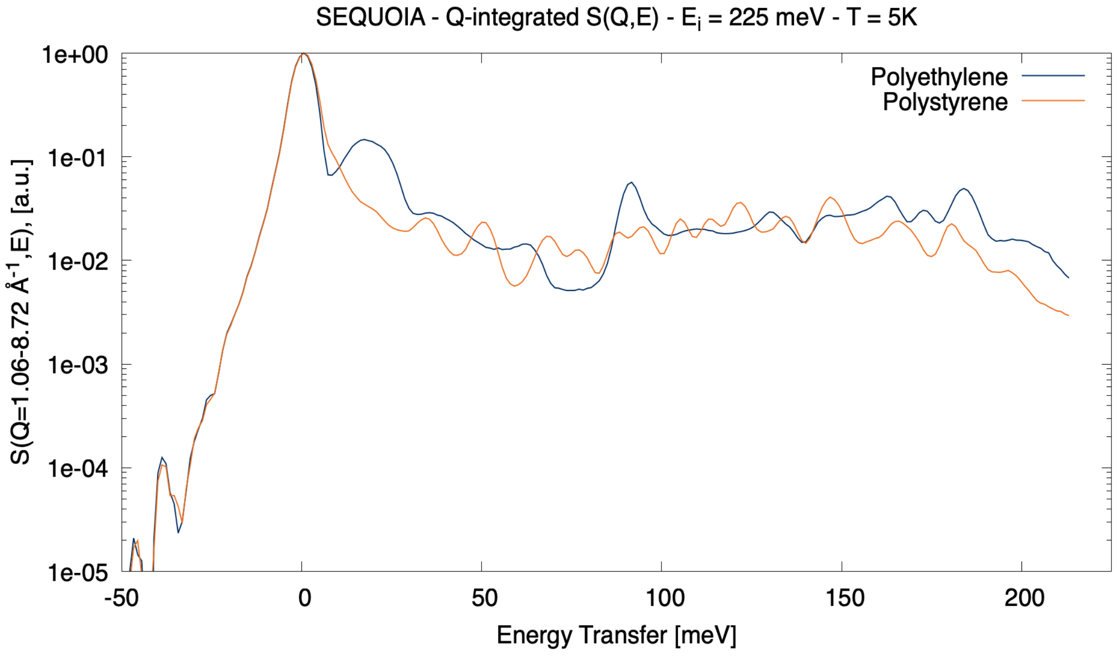


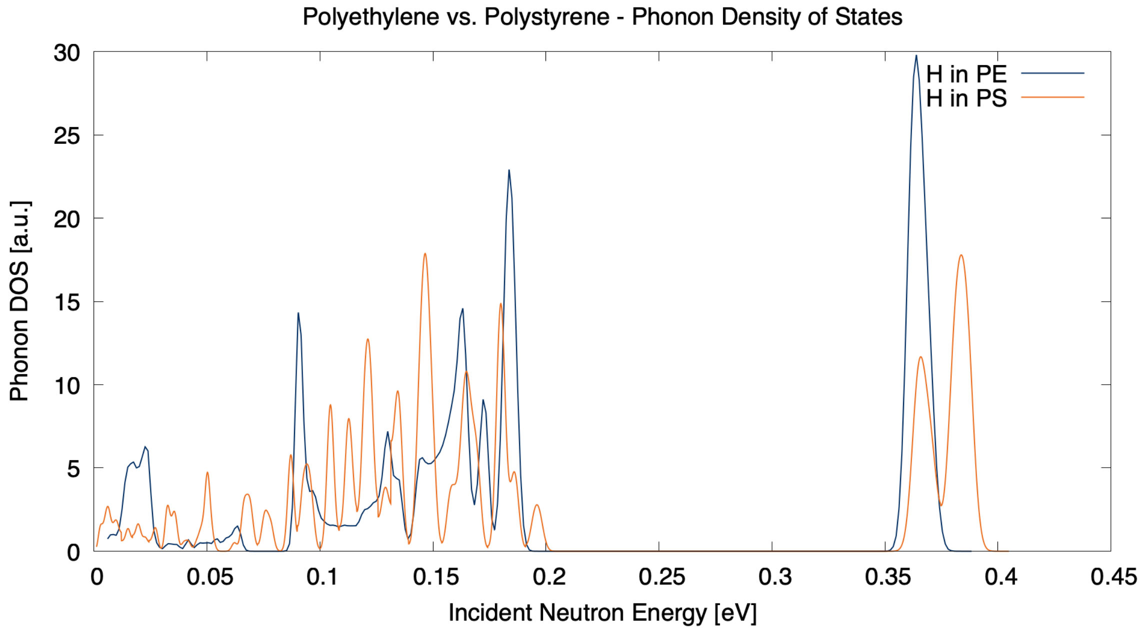


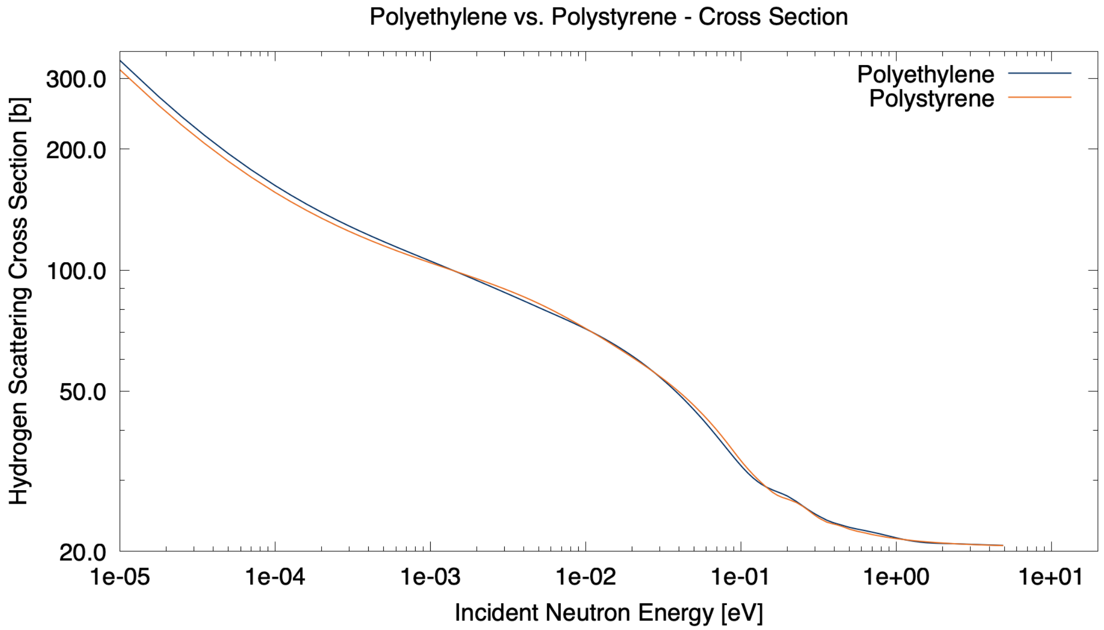


Validation - Benchmarks

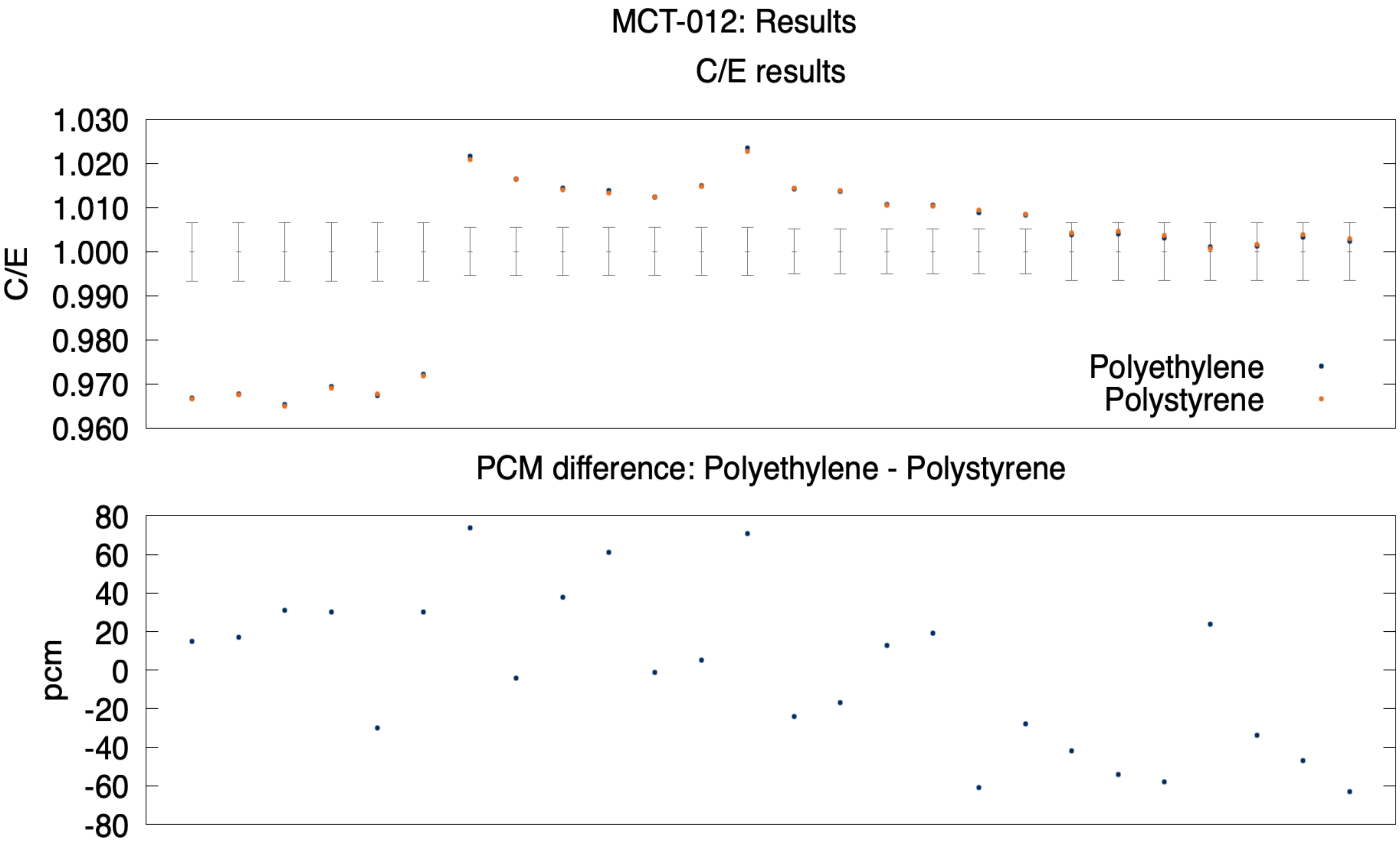
- Several ICSBEP benchmarks contain polystyrene
- Previously used polyethylene TSL library as a surrogate for polystyrene
  - Is this a good approximation?





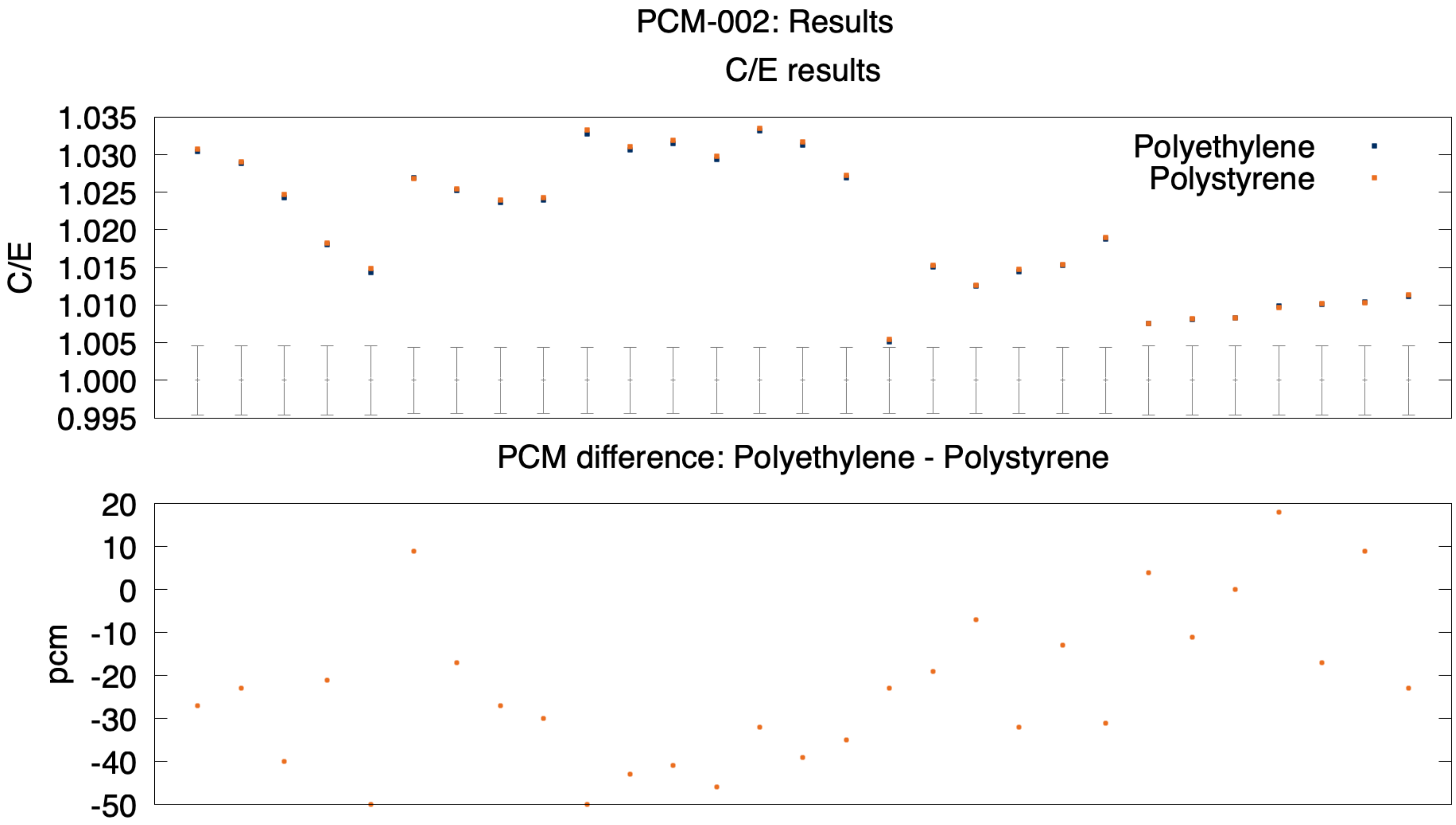


Validation - Benchmarks



Material	χ^2
Polyethylene	237.2
Polystyrene	237.5

Validation - Benchmarks



Material	χ^2
Polyethylene	700.6
Polystyrene	716.9

Conclusions

- Polystyrene evaluation conducted using multiple experiments (differential and integral)
- Proposed methodology shown to improve neutron transport
- Files submitted to NNDC for inclusion in ENDF/B-VIII.1 release
- MCT-012 & PCM-002 relatively insensitive to changes in $S(\alpha, \beta)$
- Differences in differential results don't always propagate to differences in integral results
  - Hence why evaluation of differential data important

Future Work

- Feedback on evaluation methodology welcome

- More varied experiments (different samples, different experiment types) -> more consistent results

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.

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