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Thermal Scattering Law Evaluations in Progress

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North Carolina State University & Naval Nuclear Laboratory

Introduction

- Evaluations of new TSLs are underway
  - Naval Nuclear Laboratory
    - $H(UH_3)$
    - $H(PuH_2)$
  - North Carolina State University
    - H (in heavy paraffinic oil)
    - F Li Be (in FLiBe)
    - Al O (in Al_2O_3)

H-UH_3

- Uranium Hydride (UH_3) has been used in several historical critical experiments
  - G. A. Linenberger, et al., *Nucl. Sci. Eng.*, **7**, 44-57 (1960).
  - HEU-COMP-INTER-003, “Reflected Uranium-Hydride Critical Assemblies”
- β-UH_3 stable at room temperature and above
  - 8 molecules (32 atoms) per unit cell
  - 6.643 Å lattice constant
- H-UH_3 TSL developed using first-principles or *ab initio* lattice dynamics (AILD) approach
  - VASP to calculate interatomic Hellman-Feynman forces for crystal structure using GGA+U
  - PHONON to determine dispersion relations and phonon density of states (PDOS)
  - H-UH_3 TSL evaluated in incoherent approximation using NJOY/LEAPR

HEU-COMP-INTER-003

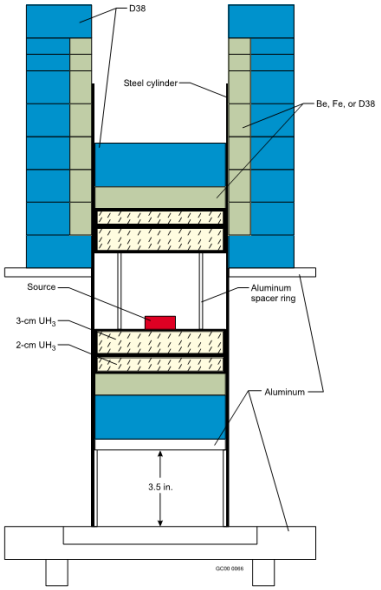


Figure 2. Schematic of the Doubly Reflected Assembly.

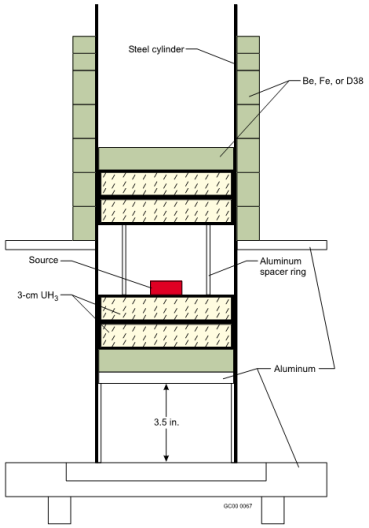
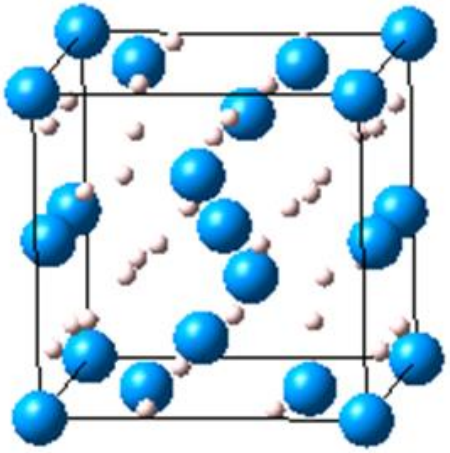
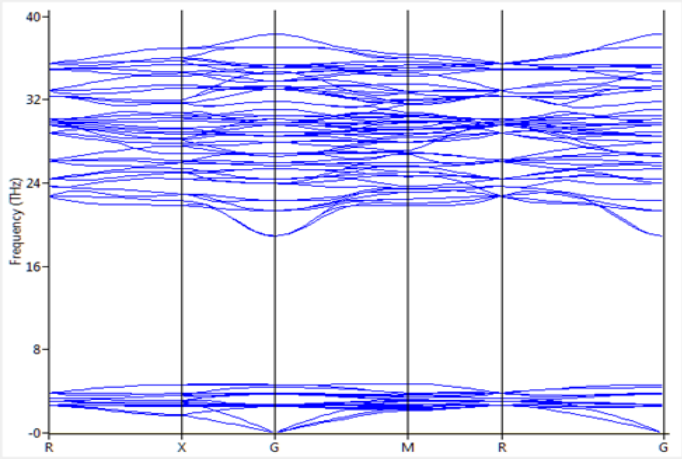


Figure 3. Schematic of a Singly Reflected Assembly.

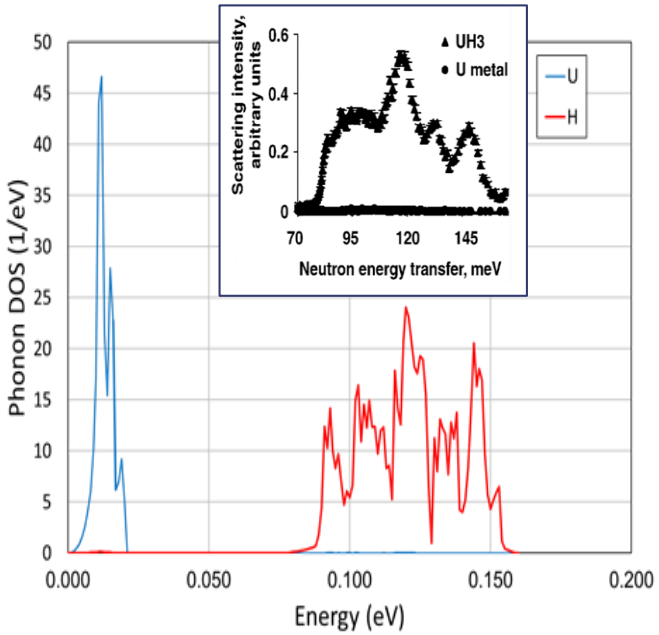


β-UH_3 Unit Cell

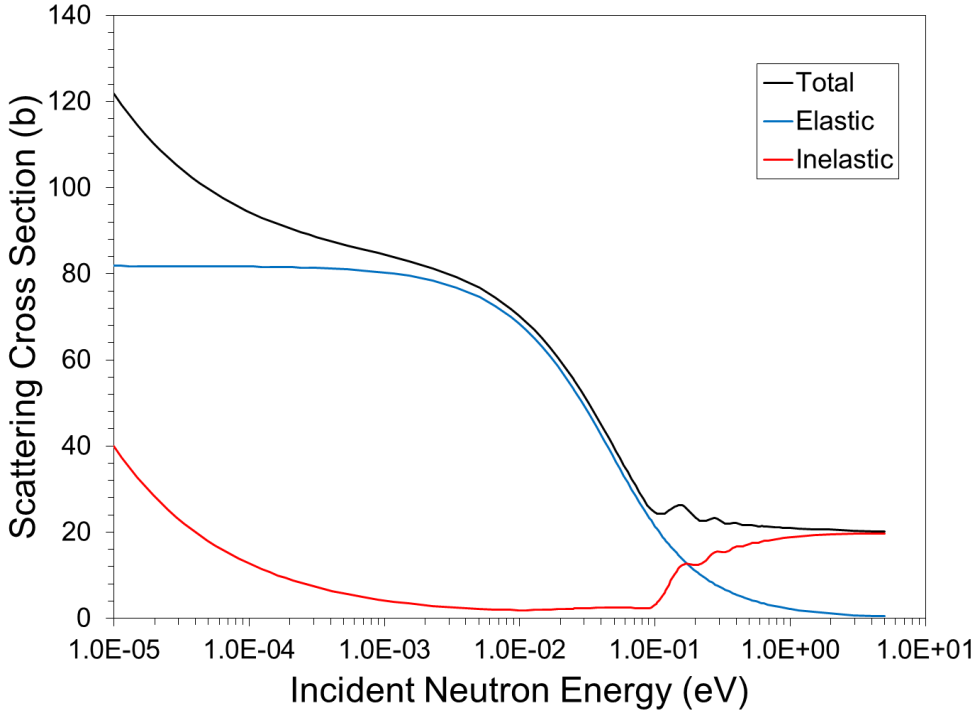
H-UH_3



Calculated dispersion relation for UH_3



Agreement between calculated and measured Phonon DOS



Total, elastic, and inelastic scattering cross section for H-UH_3 at 293.6 K generated by NDEX

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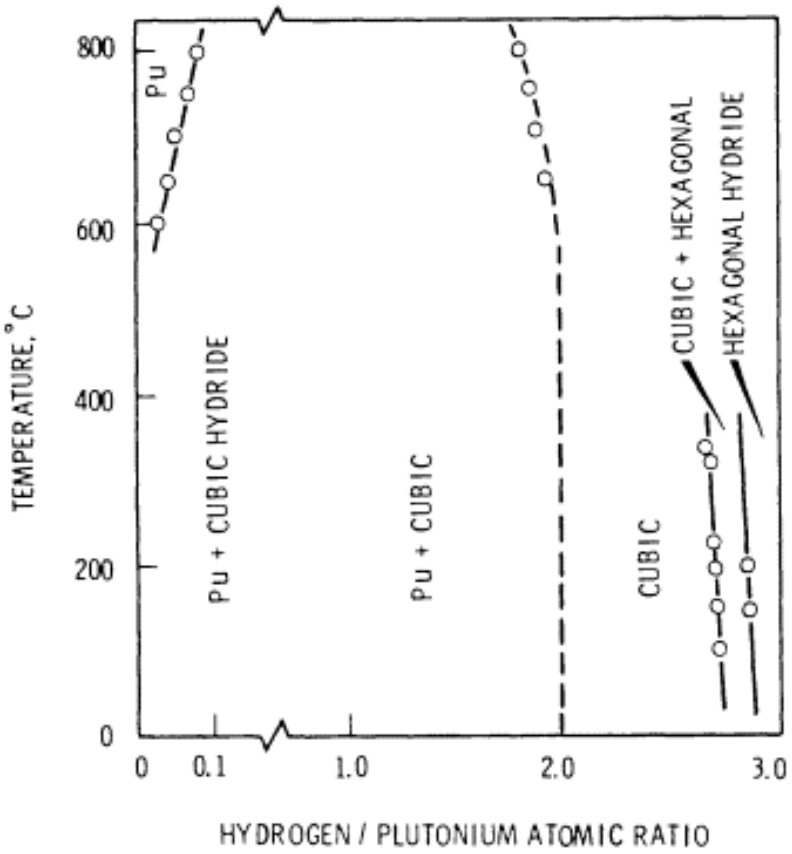
Plutonium-Hydrogen Phase Diagram

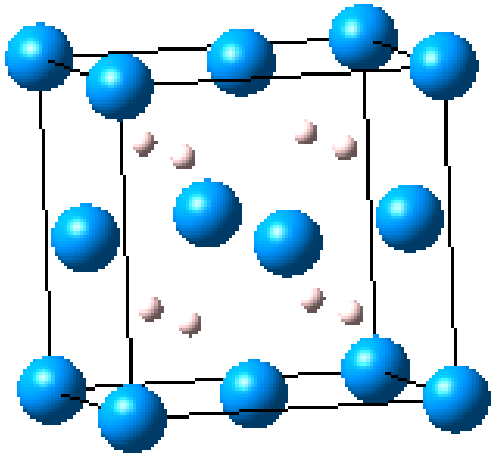
- Hydride/de-hydride processes used in some NNSA fuel cycle facilities
- Four PuH_x phases present
  - PuH_2 (FCC) for H/Pu ≤ 2.0
    - Two phase solid solution of Pu (metal) + PuH_2 (FCC)
  - PuH_{2+x} (FCC) for 2.0 < H/Pu < 2.75
    - Single phase solid solution
  - PuH_{2+x} (FCC) for 2.75 < H/Pu < 3-ε
    - Two phase solid solution, PuH_{2+x} (FCC) + PuH_3 (Hex)
  - PuH_3 (Hex) for 2.75 < H/Pu ≤ 3.0
    - Single phase solid solution for 3-ε < H/Pu < 3
- Only PuH_2 and PuH_{2+x} of practical interest for NCS
- Initial NNL work concentrates on PuH_2
  - PuH_{2+x} to be evaluated later



Phase diagram for the plutonium-hydrogen system. From R. N. R. Mulford and G. E. Sturdy, *J. Am. Chem. Soc.*, **78**, 3899 (1956).

H-PuH_2

- PuH_2 has a CaF_2 type FCC structure
  - 12 atoms per unit cell
  - 4 Pu atoms (blue) at vertices and faces of unit cell
  - 8 H atoms (grey) in tetrahedral holes between Pu atoms
  - Mass density of 10.40 g/cm^3
- Measured lattice parameter agree (X-ray diffraction)
  - *a* = 5.359 ± 0.002 Å, Mulford and Sturdy (1955)
  - *a* = 5.359 ± 0.001 Å, Coffinberry and Ellinger (1956)
  - *a* = 5.359 ± 0.002 Å, Muromura et al. (1972)
  - *a* = 5.3593 Å, Willis et al. (1985)
- H-PuH_2 TSL developed using AILD approach
  - VASP to calculate interatomic Hellman-Feynman forces for crystal structure using GGA+U
  - PHONON to determine dispersion relations and phonon density of states (DOS)
  - H-PuH_2 TSL evaluated in incoherent approximation using NJOY/LEAPR



PuH_2 Unit Cell

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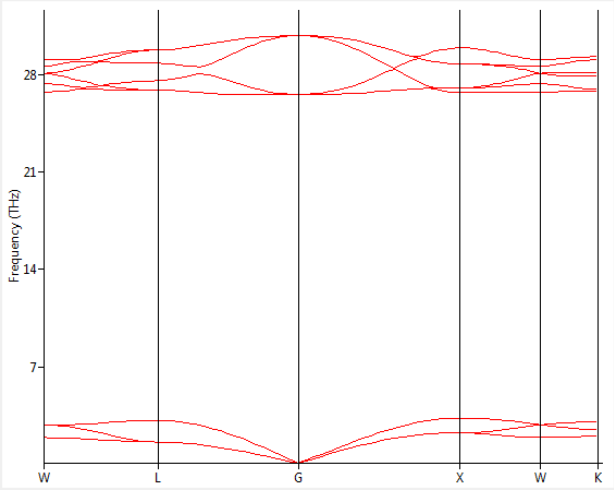
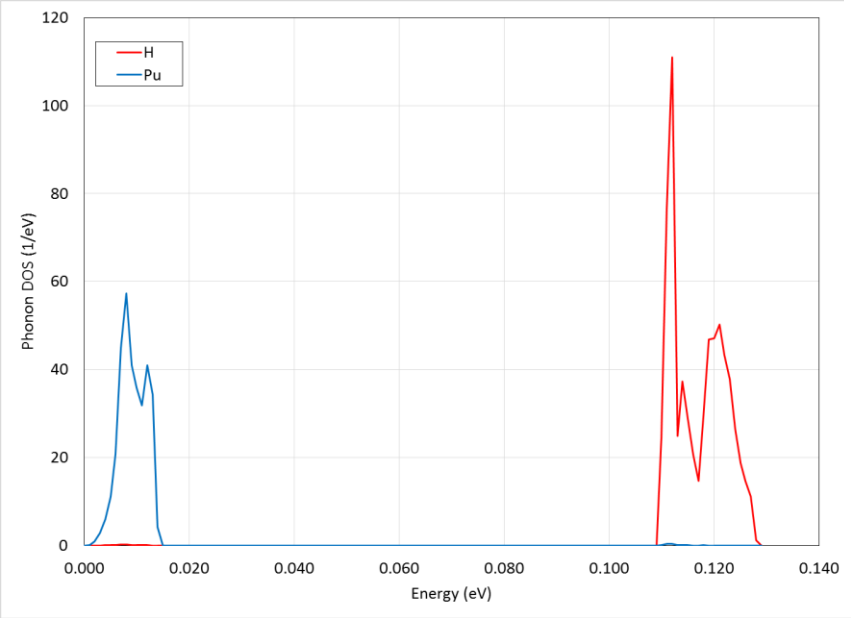
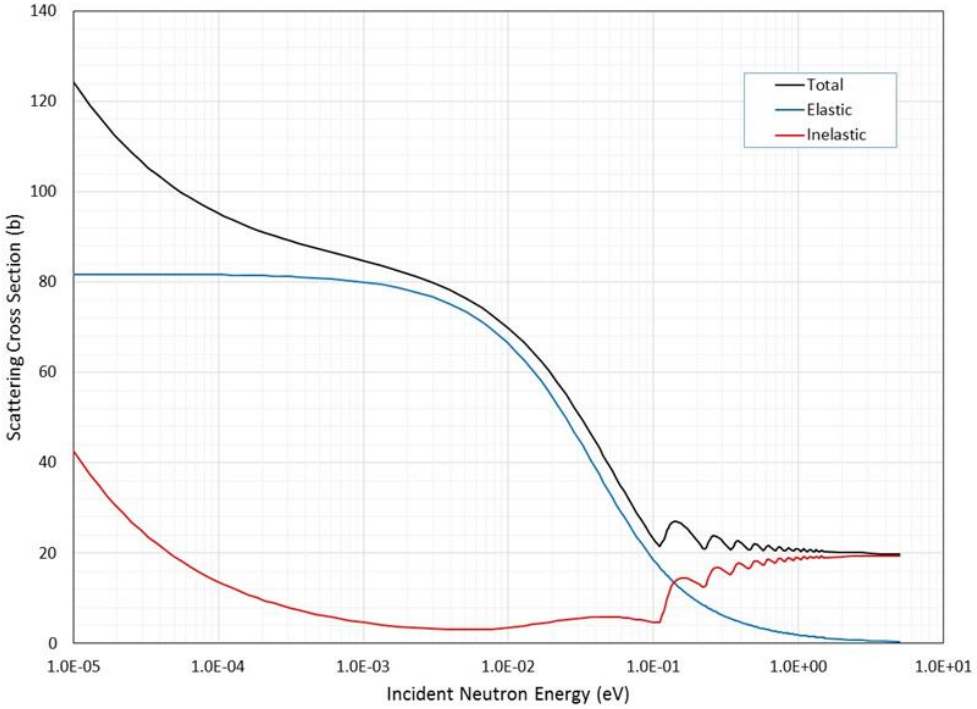
H-PuH_2



Calculated dispersion relation for PuH_2





Total, elastic, and inelastic scattering cross section for H-PuH_2 at 293.6 K generated by NDEX

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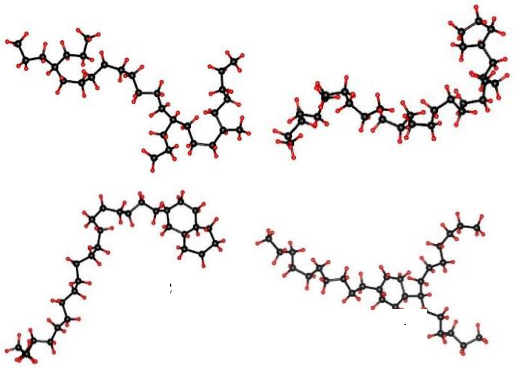
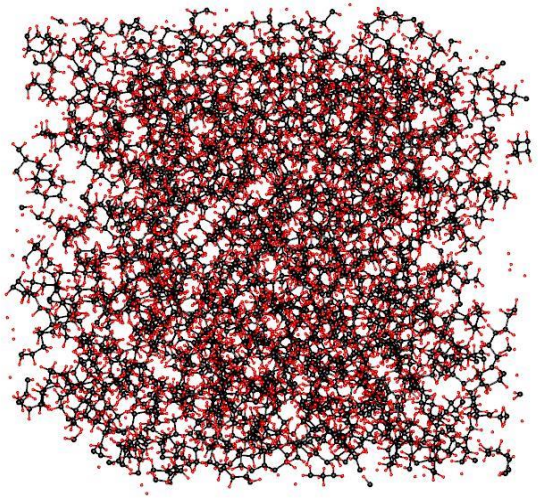
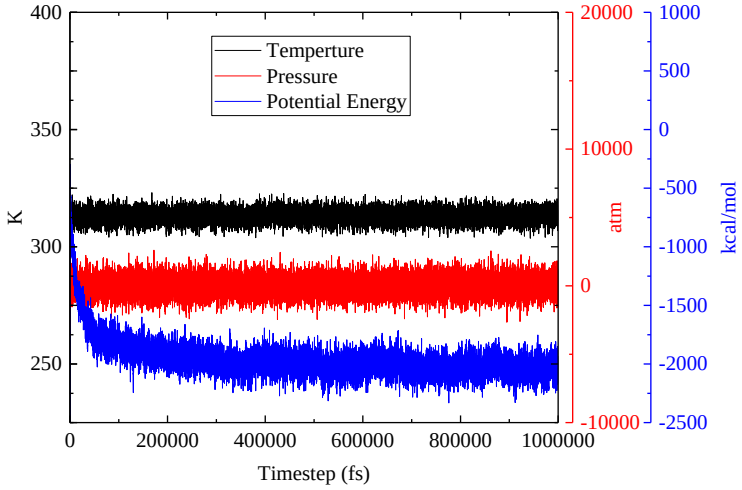
Heavy-Paraffinic Oil

- Heavy paraffinic oil is generally used as a liquid lubricant in circulating oil systems
- COMPASS potential used to model organic compounds
- Non-equilibrium and Equilibrium MD (NEMD and EMD) employed during the simulation process
- 9020 atoms
- 100 molecules
- 45x45x45 $Å^3$ supercell
- Simulation times in the ns range with 1 fs timesteps
- Model Validation:
  - Density
  - Viscosity
  - Diffusivity





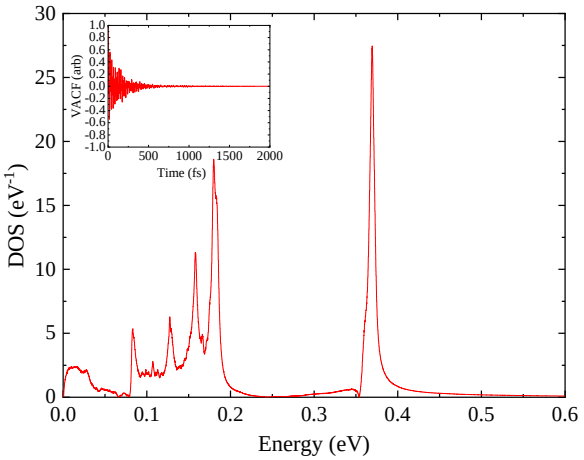


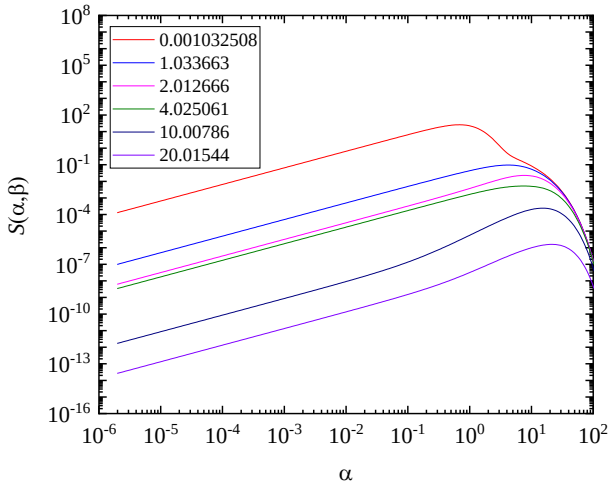
Heavy-Paraffinic Oil

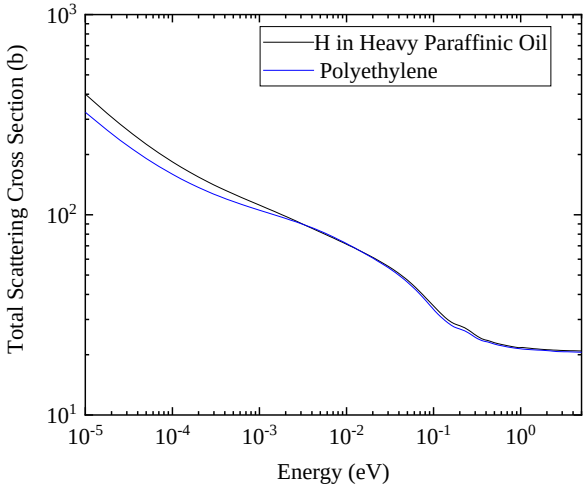
Property	Target Value	Simulation Results
Density (300K) [g/cm^3]	0.86	0.829 ± 0.004

Property	Target Value	Simulation Results
Viscosity (313K) [mm^2/s (cSt)]	19.7	20.50 ± 0.84
Viscosity (373K) [mm^2/s (cSt)]	4.0	3.65 ± 0.29

Component	Diffusivity [cm^2/s]
Hydrogen in Paraffinic Oil	2.78 x 10^{-7} [300K] [This Work]
Hydrogen in Water	2.3 x 10^{-5} [298K] [12]



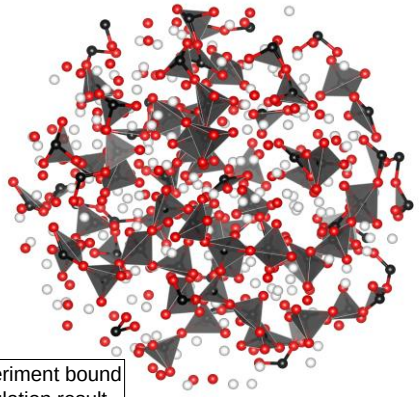


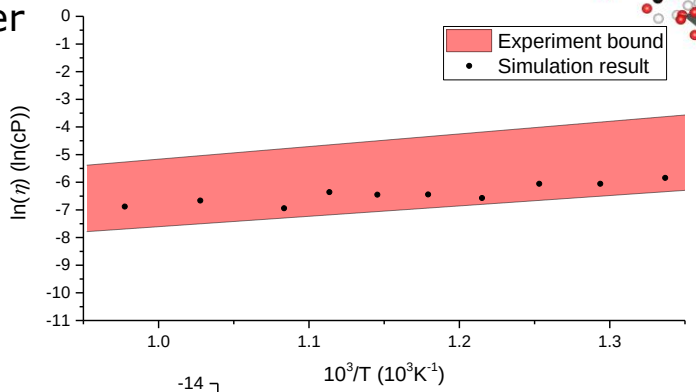


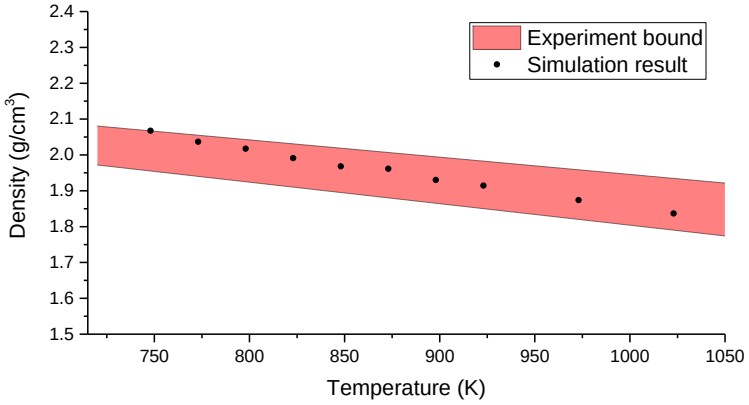
Molten Salt FLiBe

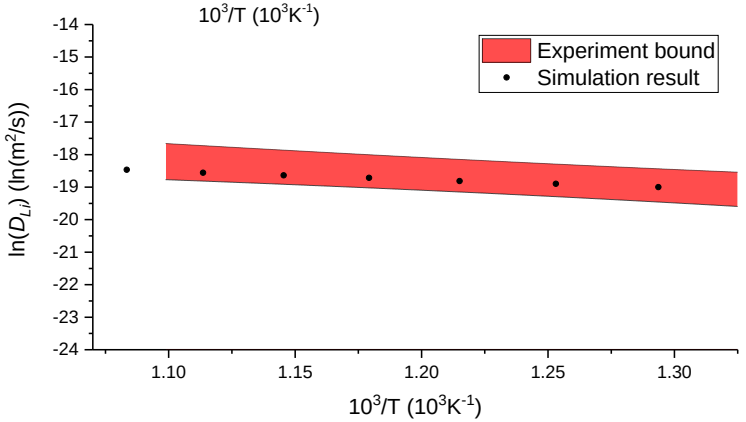
- Molten Salt FLiBe (Li_2BeF_4) has been proposed as a coolant, moderator, and heat storage medium in thermal neutron driven nuclear reactors.
- 7,000 atoms in 43.67 Å × 43.67 Å× 43.67 Å cube with periodic boundaries
- Timestep 1 fs equilibrated for 20 ps under NPT condition
- Model validation
  - Density
  - Viscosity
  - Diffusion coefficient
  - Heat Capacity

	Heat Capacity $Cal/g \cdot K$
Simulation	0.551
Exp.	0.577

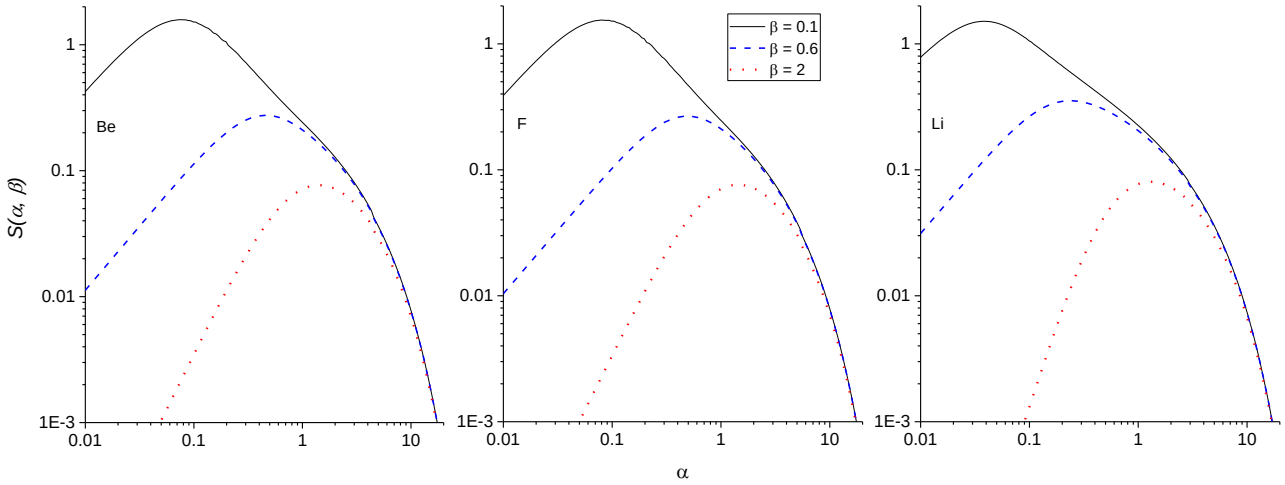


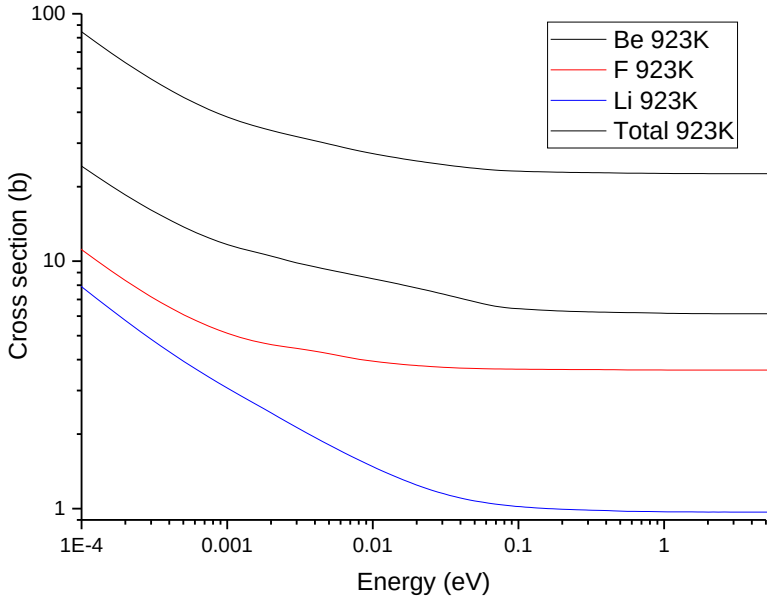






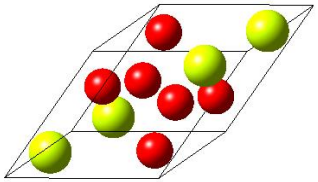
Molten Salt FLiBe



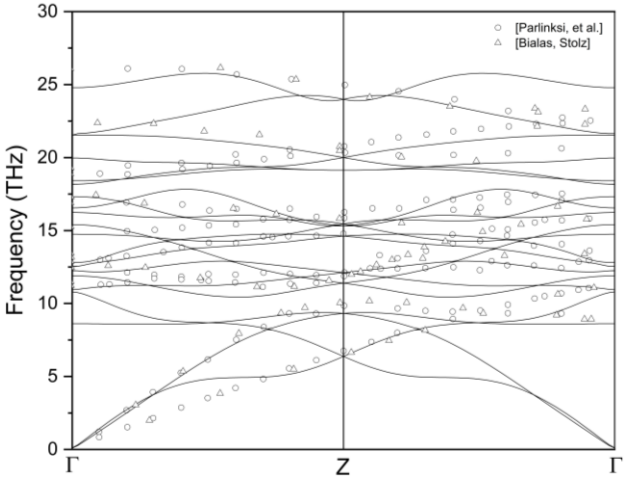


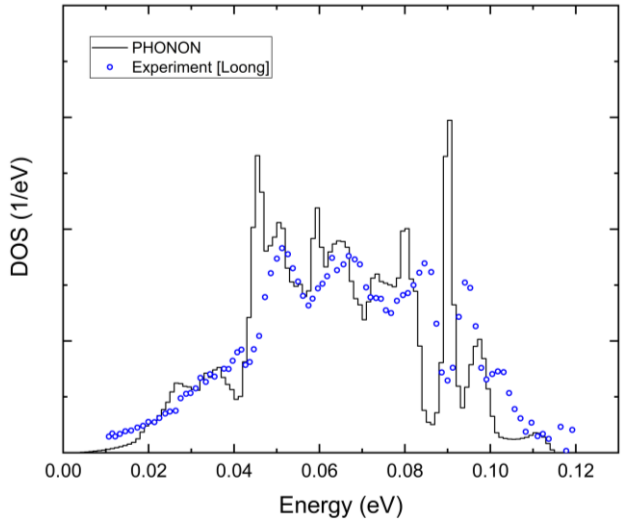
Sapphire (Al_2O_3)

- Sapphire (Al_2O_3) is a common crystalline material for thermal neutron filters
  - Large, high purity, single crystal
    - Oriented to minimized Bragg scattering
  - Wide-band-gap insulator
- Scattering largely from inelastic scattering
- DFT Parameters
  - GGA-PBE pseudopotential
  - 600 eV energy cutoff
  - 5x5x5 k-mesh
  - 2x2x2 supercell (80 atoms)
- Model validation
  - a lattice parameters for hexagonal unit cell
  - Shear modulus (B)
  - Young's modulus (Y)



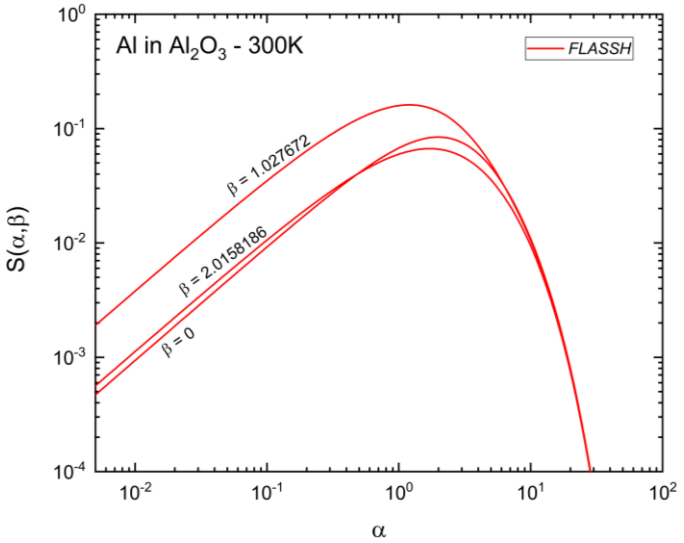
	a (Å)	B (GPa)	Y (GPa)
VASP	5.177	145.4	361
Exp.	5.128	145	345

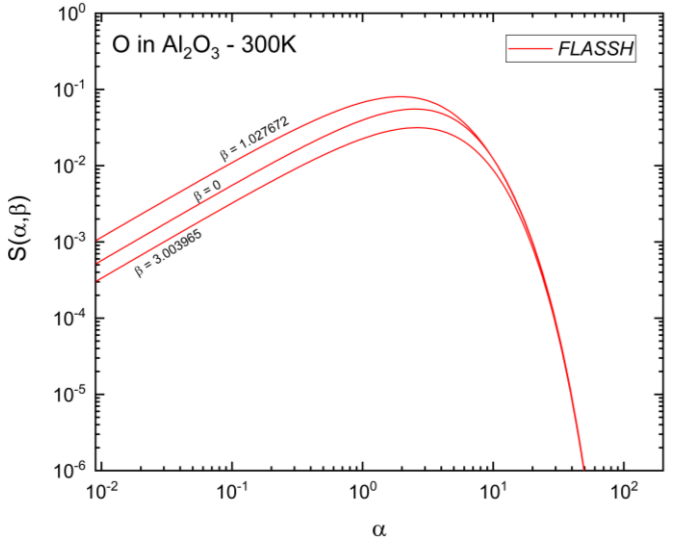


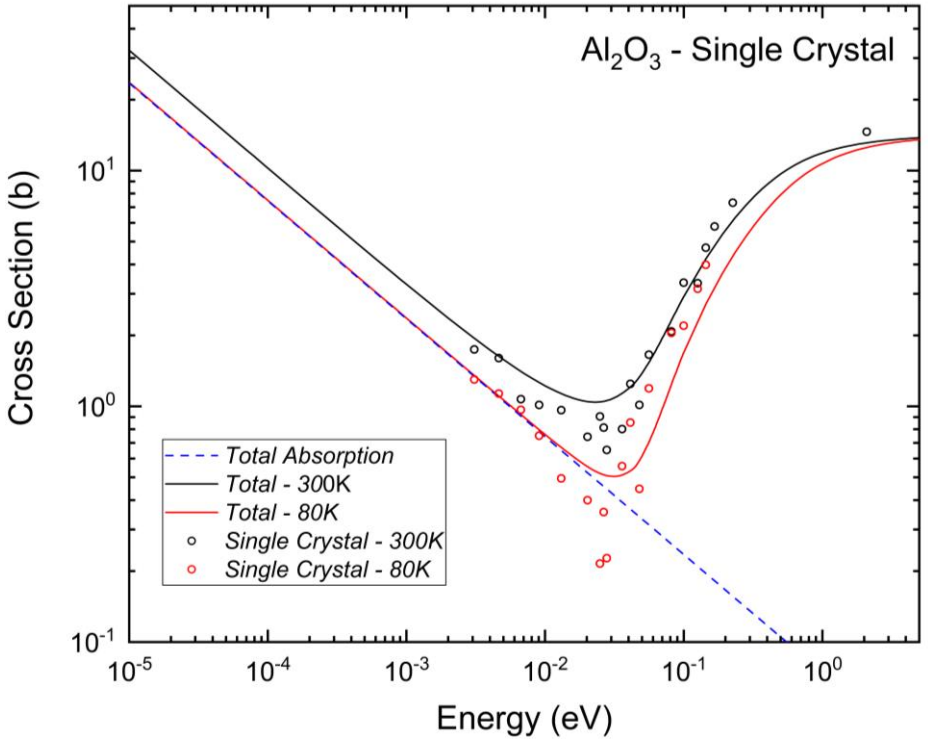


Neutron-weighted Total Al_2O_3 Phonon Density of States

Sapphire (Al_2O_3)







Summary

- NNL and NCSU will be contributing several TSL evaluations to upcoming release of ENDF/B
- Modern DFT and MD techniques continue to be primary tools in evaluation process
- For crystalline materials, a single crystal thermal neutron filter may be represented by ignoring MT=2 section in the TSL file.