OECD/NEA Meeting: WPEC SG42
"Thermal Scattering Kernel S(α,β): Measurement, Evaluation and Application"
May 10 – 11, 2016 • Paris, France

Thermal Scattering Law (TSL) Data Generation Activities at North Carolina State University

Ayman I. Hawari

Nuclear Reactor Program
Department of Nuclear Engineering
North Carolina State University
Raleigh, North Carolina, USA

Acknowledgement

- The many graduate students and Postdocs, and research staff at North Carolina State University

Acknowledgement

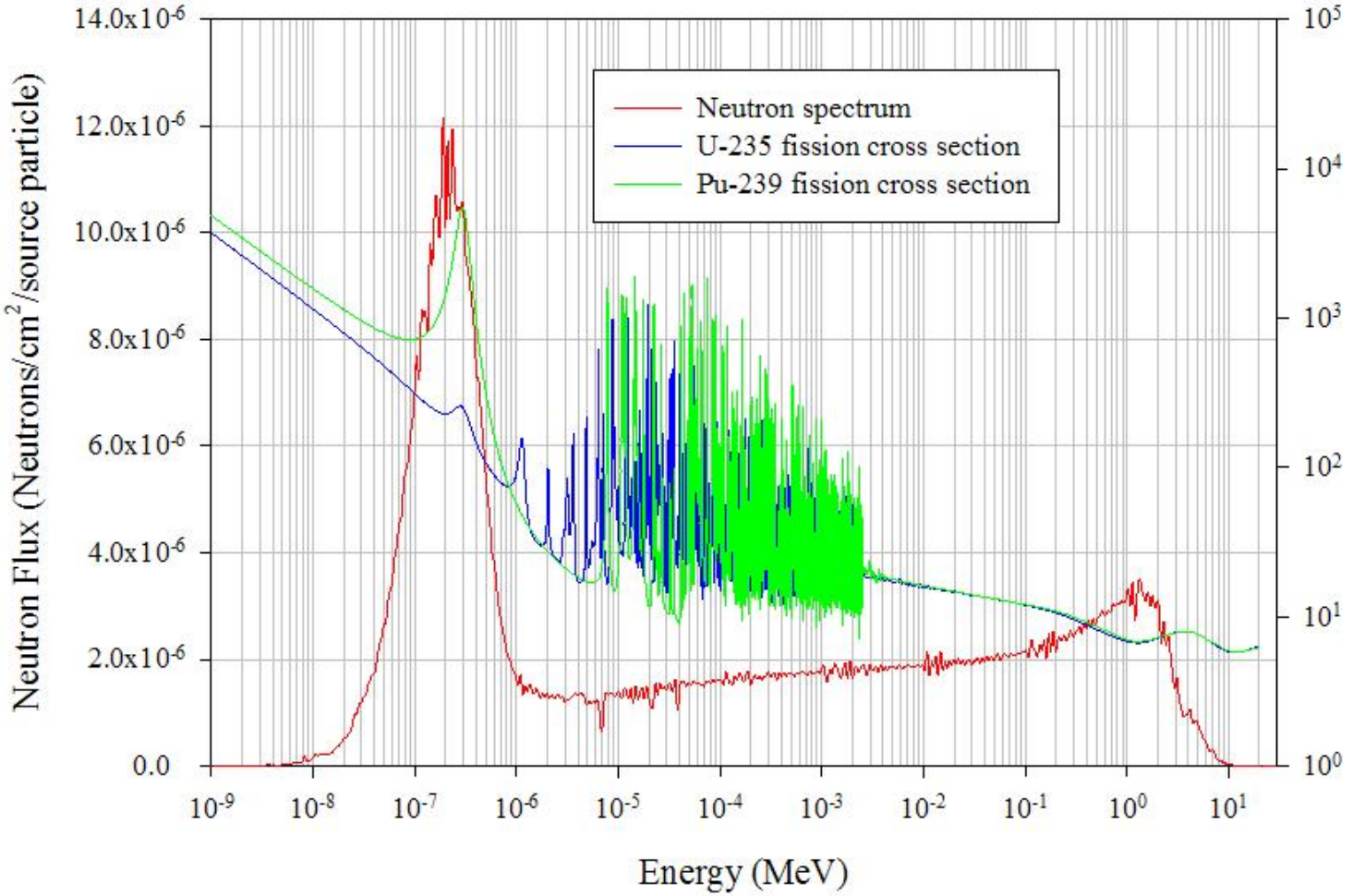
- Collaboration with LLNL and Bettis labs

- Funding by
  - US DOE NNSA Nuclear Criticality Safety program
  - US Naval Reactors program
  - US DOE office of Nuclear Energy (NE) through NERI and NEUP programs

Vision

- Establish a predictive and approximation free approach for generating the needed data (i.e., TSL) to describe neutron thermalization in matter
- Various applications:
  - Nuclear criticality safety
  - Nuclear reactor design
  - Neutron beam spectral shaping (i.e., filtering)
  - Neutron source (cold, ultracold, etc.) characterization

Motivation


Neutron spectrum
U-235 fission cross section
Pu-239 fission cross section
Neutron Flux (Neutrons/cm^2/source particle)
14.0x10^{-6}
12.0x10^{-6}
10.0x10^{-6}
8.0x10^{-6}
6.0x10^{-6}
4.0x10^{-6}
2.0x10^{-6}
0.0
10^5
10^4
10^3
10^2
10^1
10^0
10^{-9} 10^{-8} 10^{-7} 10^{-6} 10^{-5} 10^{-4} 10^{-3} 10^{-2} 10^{-1} 10^0 10^1
Energy (MeV)

Thermal Neutron Scattering

Using Born approximation, it can be shown that the double differential scattering cross section has the form

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{1}{4\pi}\frac{k'}{k}\left\{\sigma_{coh}S(\vec{\kappa},\omega) + \sigma_{incoh}S_s(\vec{\kappa},\omega)\right\}$$

The scattering law $S(\vec{\kappa},\omega)$ is composed of two parts

$$S(\vec{\kappa},\omega) = S_s(\vec{\kappa},\omega) + S_d(\vec{\kappa},\omega)$$

Van Hove's formulation

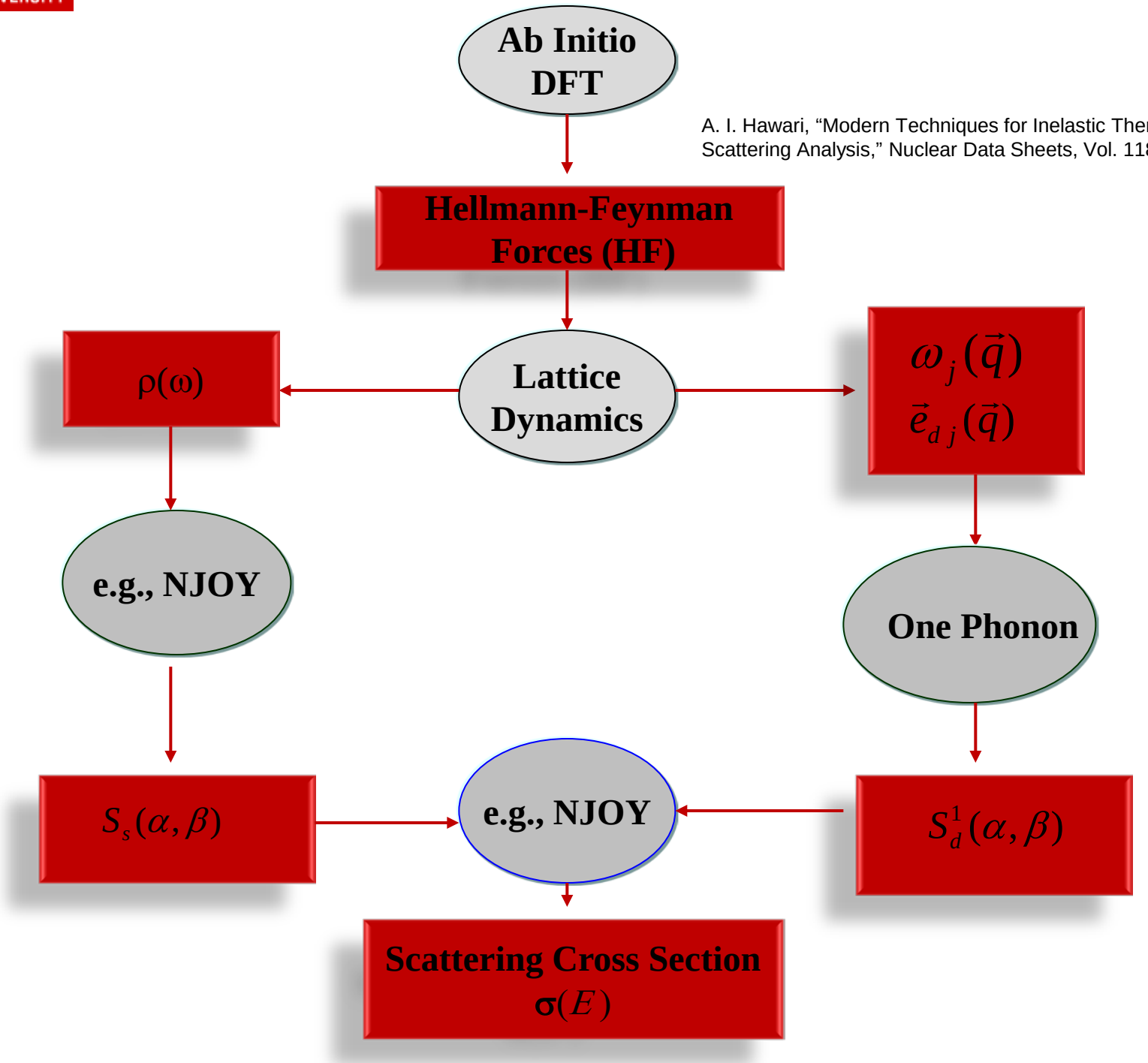
$$I(\vec{\kappa},t) = \int G(\vec{r},t)\exp(i\vec{\kappa}\cdot\vec{r})d\vec{r}$$

$$S(\vec{\kappa},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} G(\vec{r},t)e^{i(\vec{\kappa}\cdot\vec{r}-\omega t)}d\vec{r}dt$$

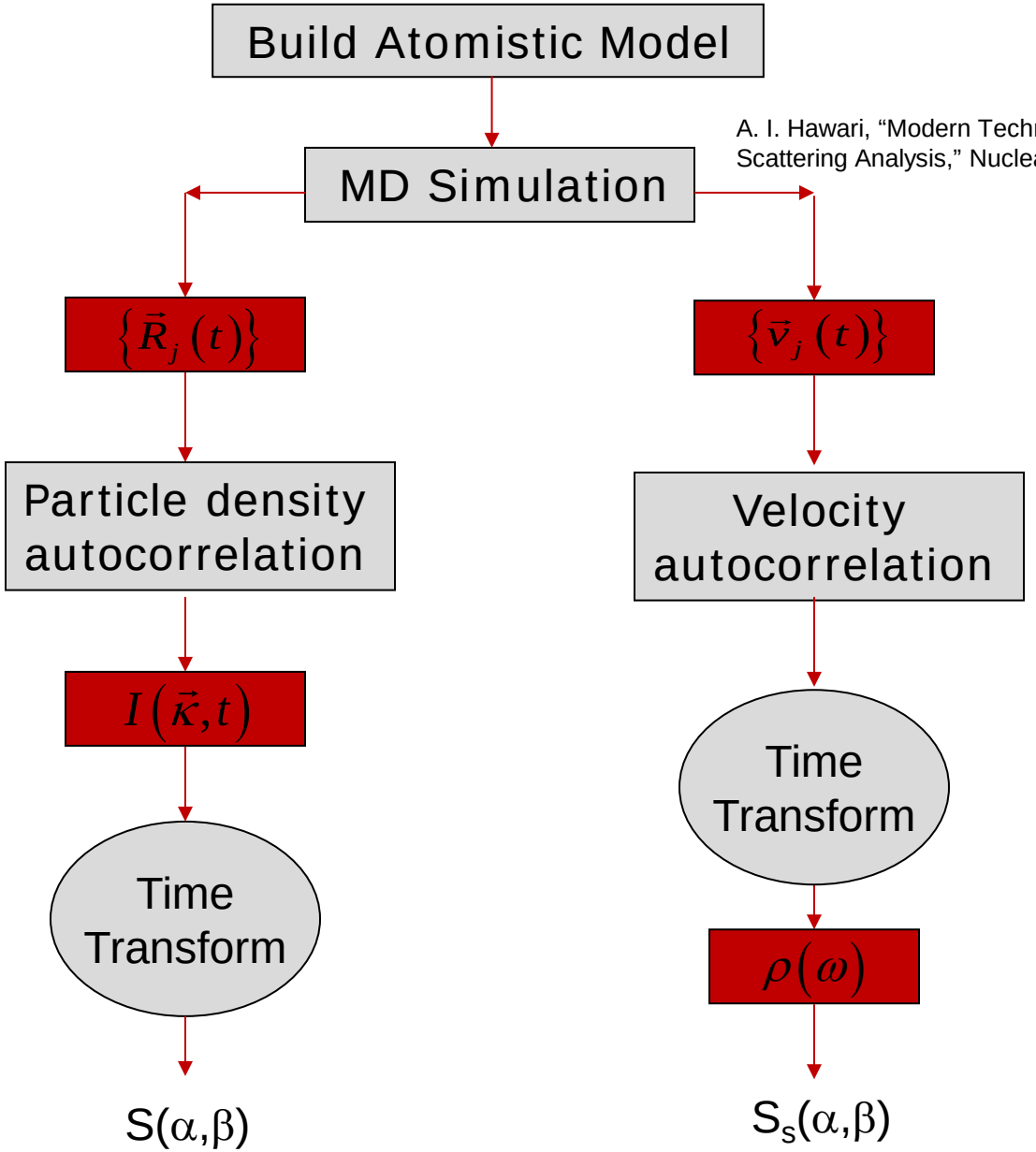
where $G(\vec{r},t)$ is the *dynamic pair correlation function* and is expressible in terms of the atomic positions.

Methods

- $S(\vec{\kappa},\omega)$ is a material property
  - can be calculated accurately!
- Implemented several approaches can be used to extract the fundamental information for calculating the TSL and eventually the cross sections
  - Density Functional Theory (DFT) based methods
    - Ab Initio Lattice dynamics
    - Ab Initio Molecular Dynamics
  - Classical Molecular Dynamics (MD) methods
- Considered modifications to alleviate existing assumptions
  - Incoherent approximation for inelastic scattering analysis
  - Coherent elastic scattering (approximations)



A. I. Hawari, "Modern Techniques for Inelastic Thermal Neutron Scattering Analysis," Nuclear Data Sheets, Vol. 118, 172, 2014.



A. I. Hawari, "Modern Techniques for Inelastic Thermal Neutron Scattering Analysis," Nuclear Data Sheets, Vol. 118, 172, 2014.

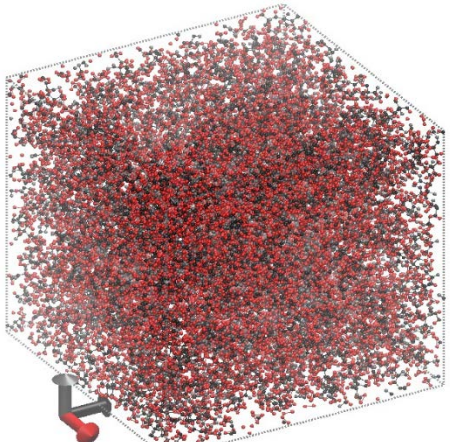
Evaluation Approach

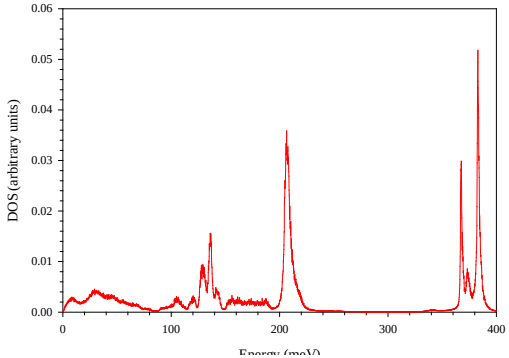
- Construct atomistic model of a material
- Verify ability of model to reproduce physical properties of the material (equilibrium conditions)
  - Density, thermal expansion, thermal conductivity,...
  - Ergodic behavior, correlations,...
- Calculate TSL and produce thermal scattering cross sections
  - Check consistency of results with computational assumptions/models
  - If possible, compare to experimental data

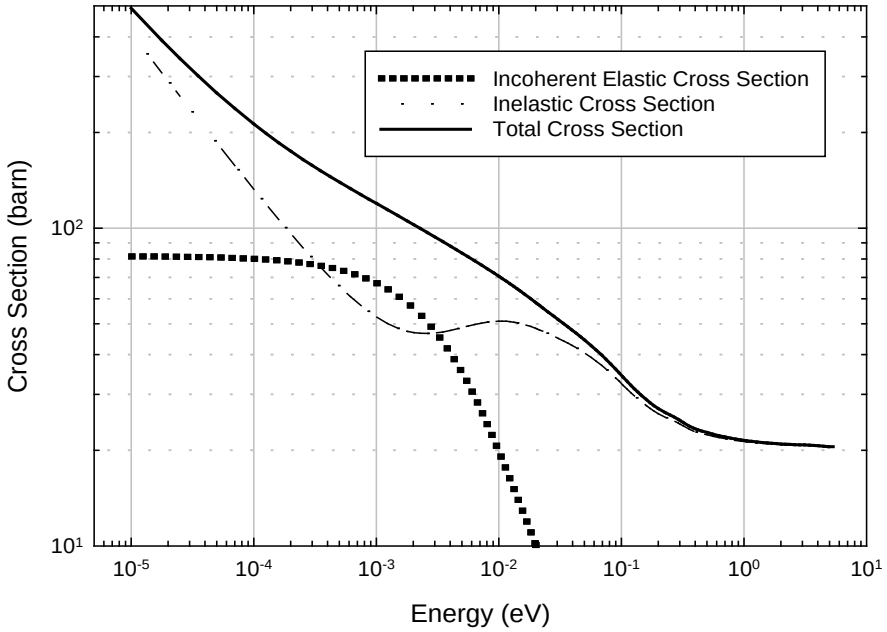
Examples - Materials Studied at NCSU

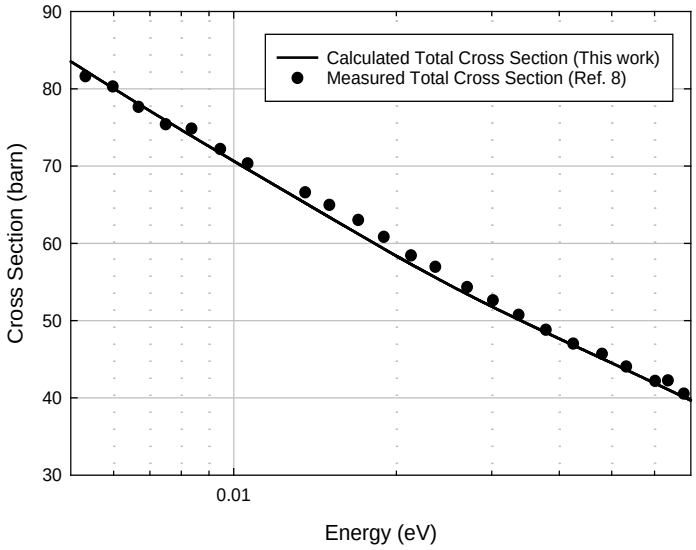
- Lucite (C5O2H8, contributed to NNDC,/ENDF, ANS 2015)
- Polyethylene (C2H4)
  - Support criticality safety applications
- Silicon dioxide (contributed to NNDC/ENDF, PHYSOR 2008, ANS, 2011, ND 2013)
  - Support criticality safety applications
- Silicon carbide (contributed to NNDC/ENDF, ANS 2013)
  - Support advanced fuel cycle applications
- Graphite, Beryllium (PHYSOR 2004 & 2008, ND 2013, PHYSOR 2016), BeO (unpublished), FLiBe (PHYSOR 2016)
  - Treatment of nuclear graphite (porous system)
  - Including coherent inelastic for graphite and beryllium
- uranium-zirconium hydride, calcium hydride, thorium hydride (PHYSOR 2004), uranium dioxide, uranium silicide
- Sapphire and bismuth (PHYSOR 2006)
  - Thermal neutron filters
- Solid methane (predictive analysis – AccApp 2011)

Lucite $(C_5O_2H_8)_n$



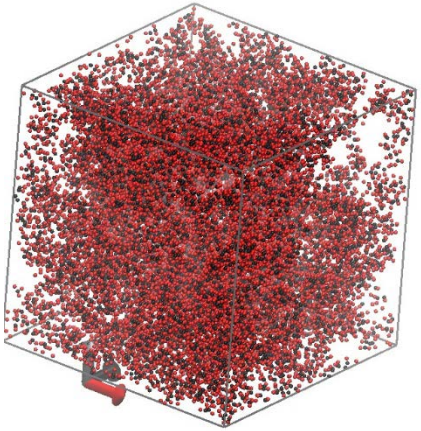


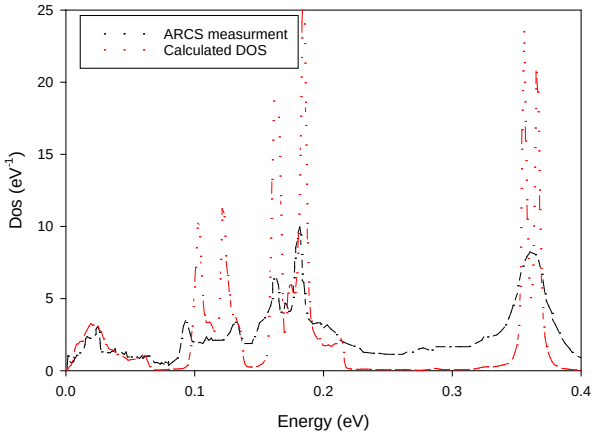


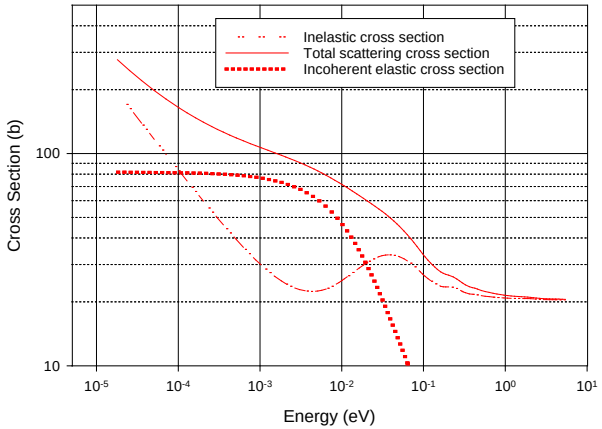


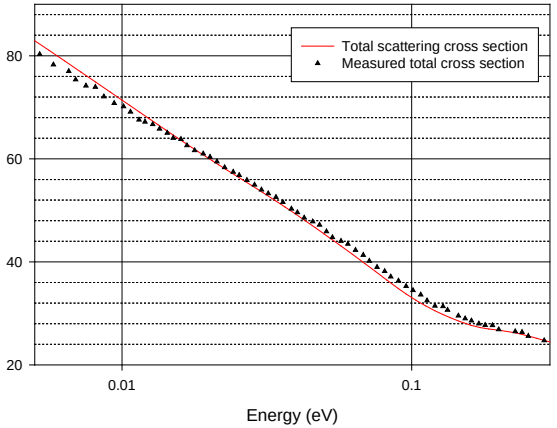
1) A. I. Hawari et al "Analysis of Thermal Neutron Scattering in Polymethyl Methacrylate (Lucite)," Transactions of the American Nuclear Society, 113, 2015.
2) Exp. Data: G. SIBONA et al., Anna. Nucl. Energy, 18, 689 (1991).

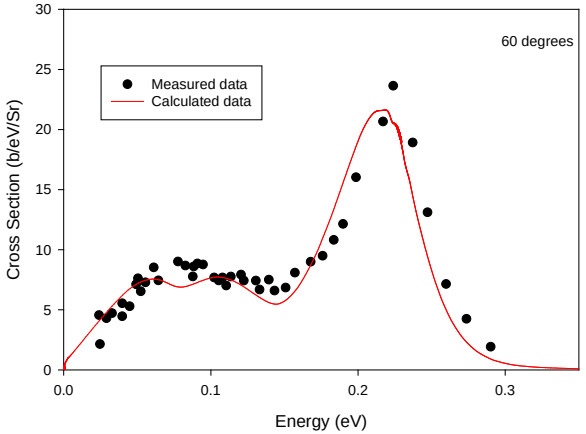
Polyethylene $(CH_2)_n$









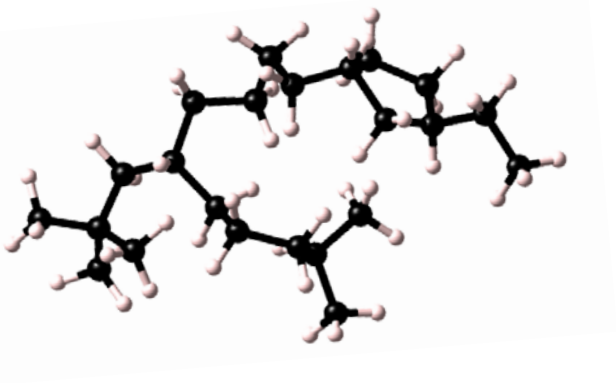


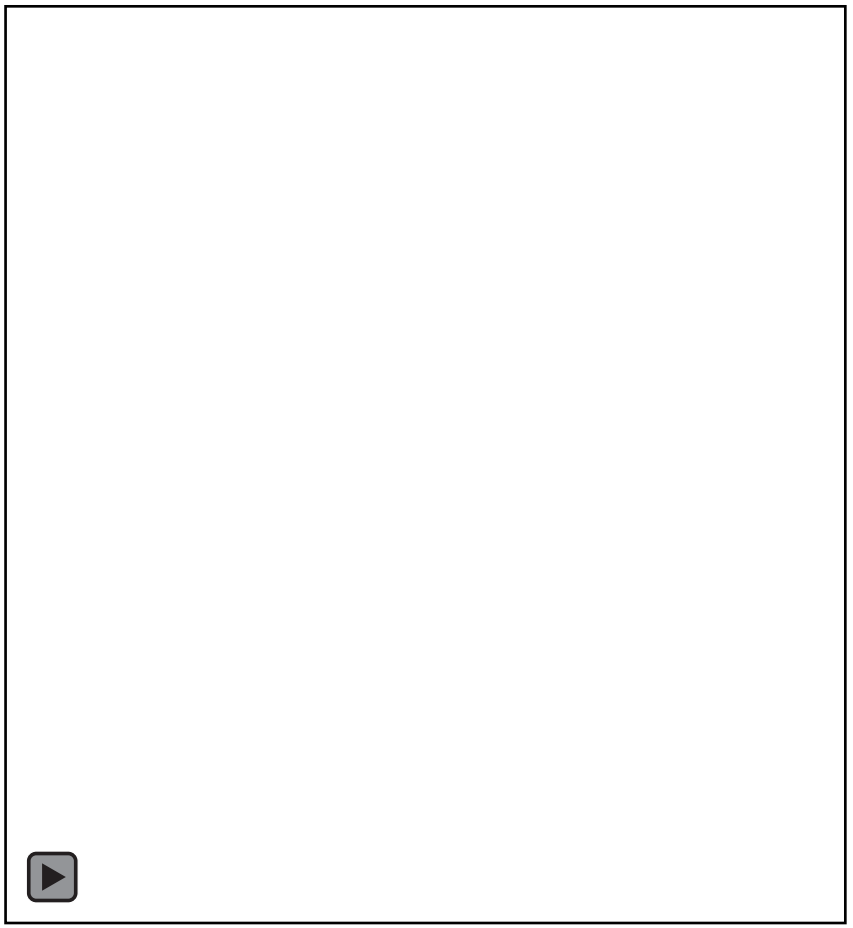
Exp. Data from
1) G. SIBONA et al., Anna. Nucl. Energy, 18, 689 (1991).
2) R. E. Hill et al., NIM A, 538, 686 (2005).
3) C. M. Lavelle et al. , NIM A, 711, 166 (2013).

Lubricant Oil MD Model

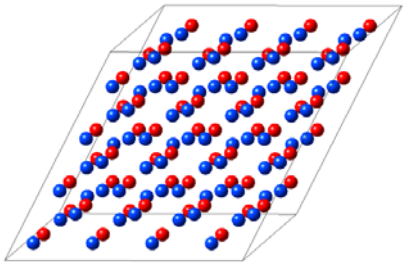
- Distilled and dewaxed heavy paraffinic mineral oil
- Example molecule

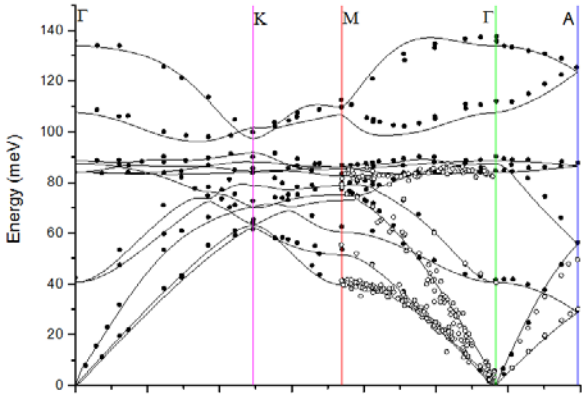
  $C_{23}H_{48}$
- Test model 100 molecules
- Model reproduces expected density nearly 0.8 g/cm^3

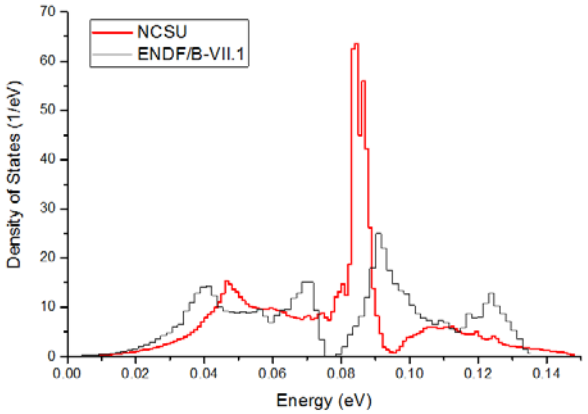




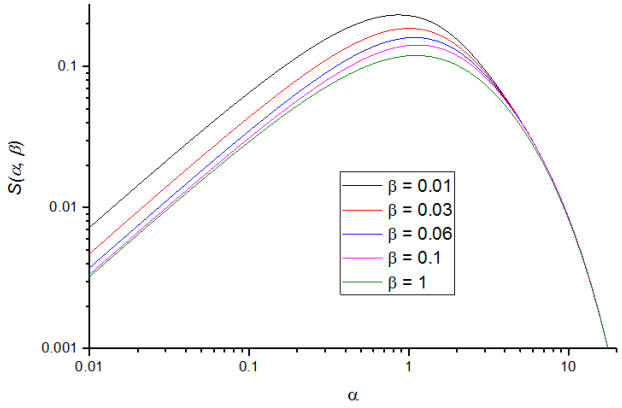
Beryllium Oxide (BeO)

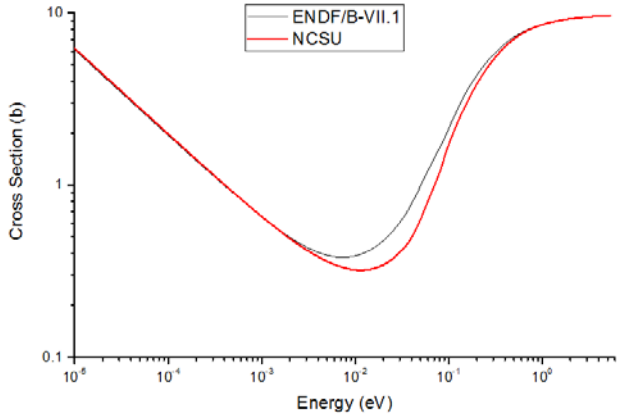




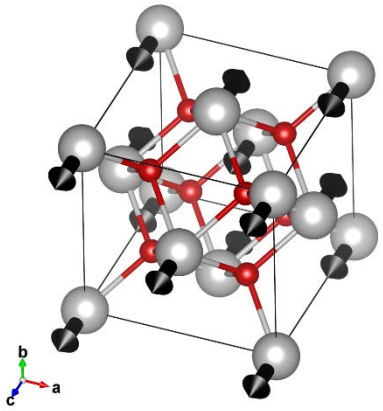


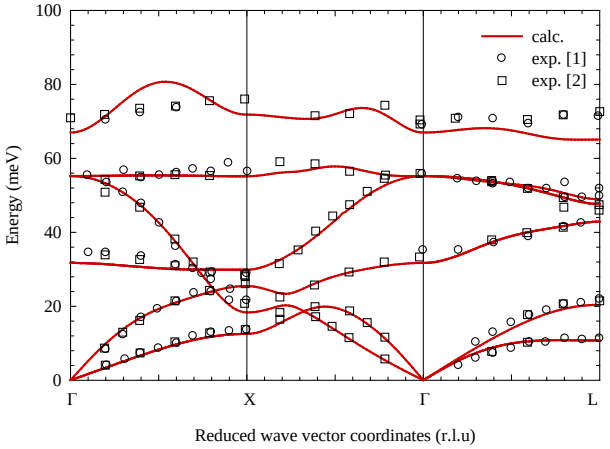
- Hexagonal structure
- 4x4x2 super-cell
- GGA



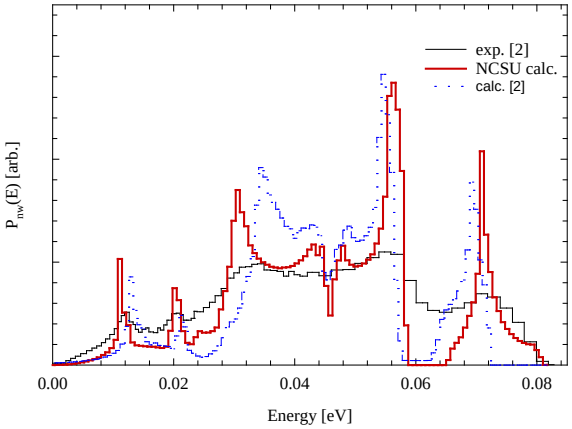


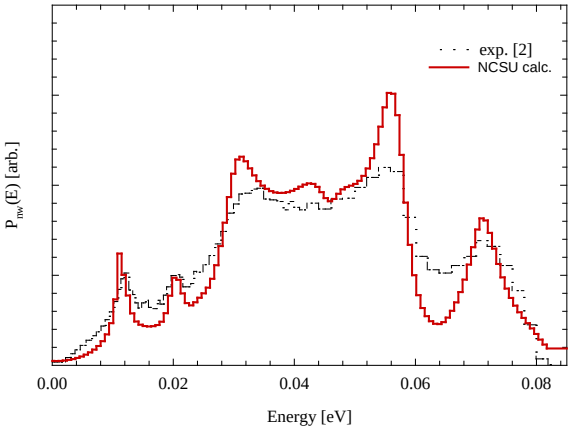
Uranium Dioxide (UO_2)





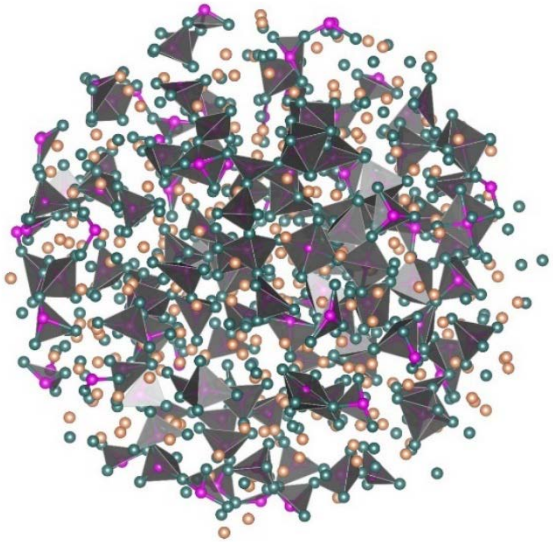
- Fluorite structure
- 2x2x2 super-cell
- GGA-PBE+U
- Predicted band gap 2 eV

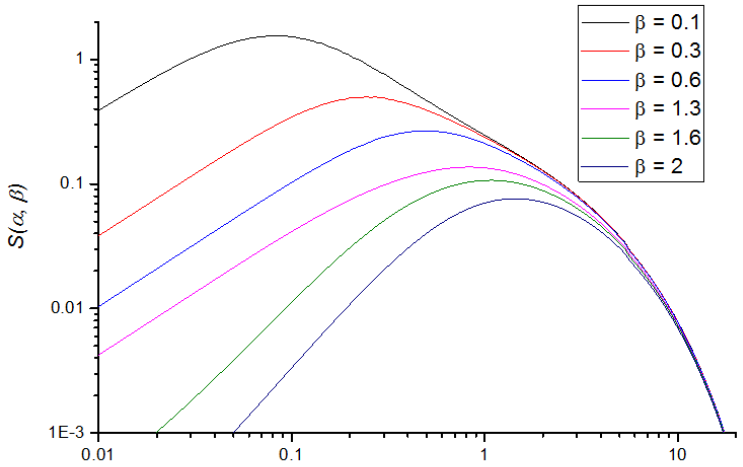


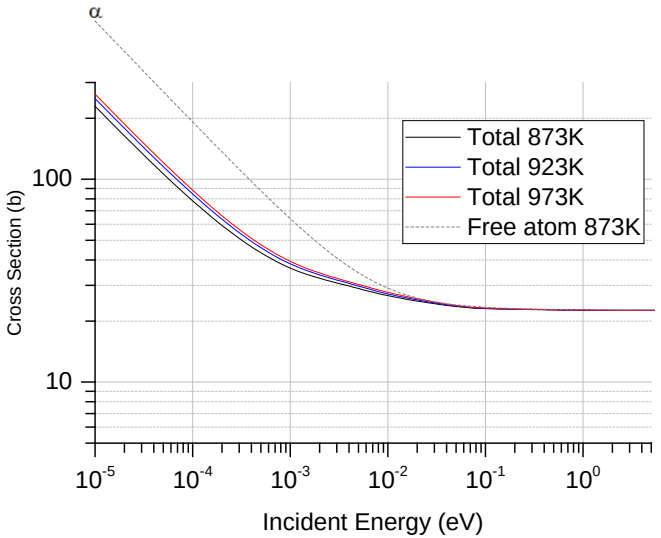


1) G. Dolling, R. A. Cowley, and A. D. B. Woods, "The crystal dynamics of Uranium Dioxide," Can. J. Phys., 43, 8 (1965) 1397
2) J. W. L. Pang, A. Chernatynskiy, B. C. Larson, W. J. L. Buyers, D. L. Abernathy, K. J. McClellan, and S. R. Phillpot, "Phonon density of states and anharmonicity in UO2," Phys. Rev. B, 89, (2014) 115132

Liquid FLiBe (Li_2BeF_4)

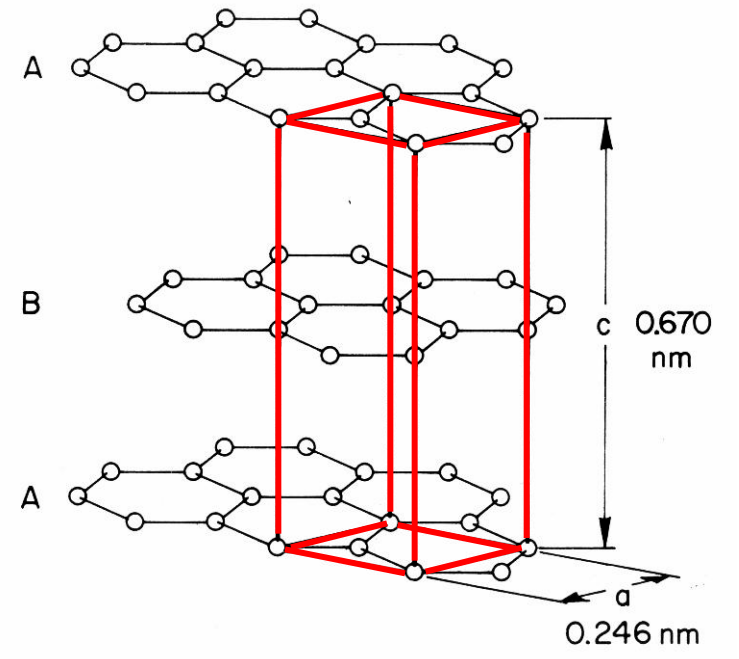






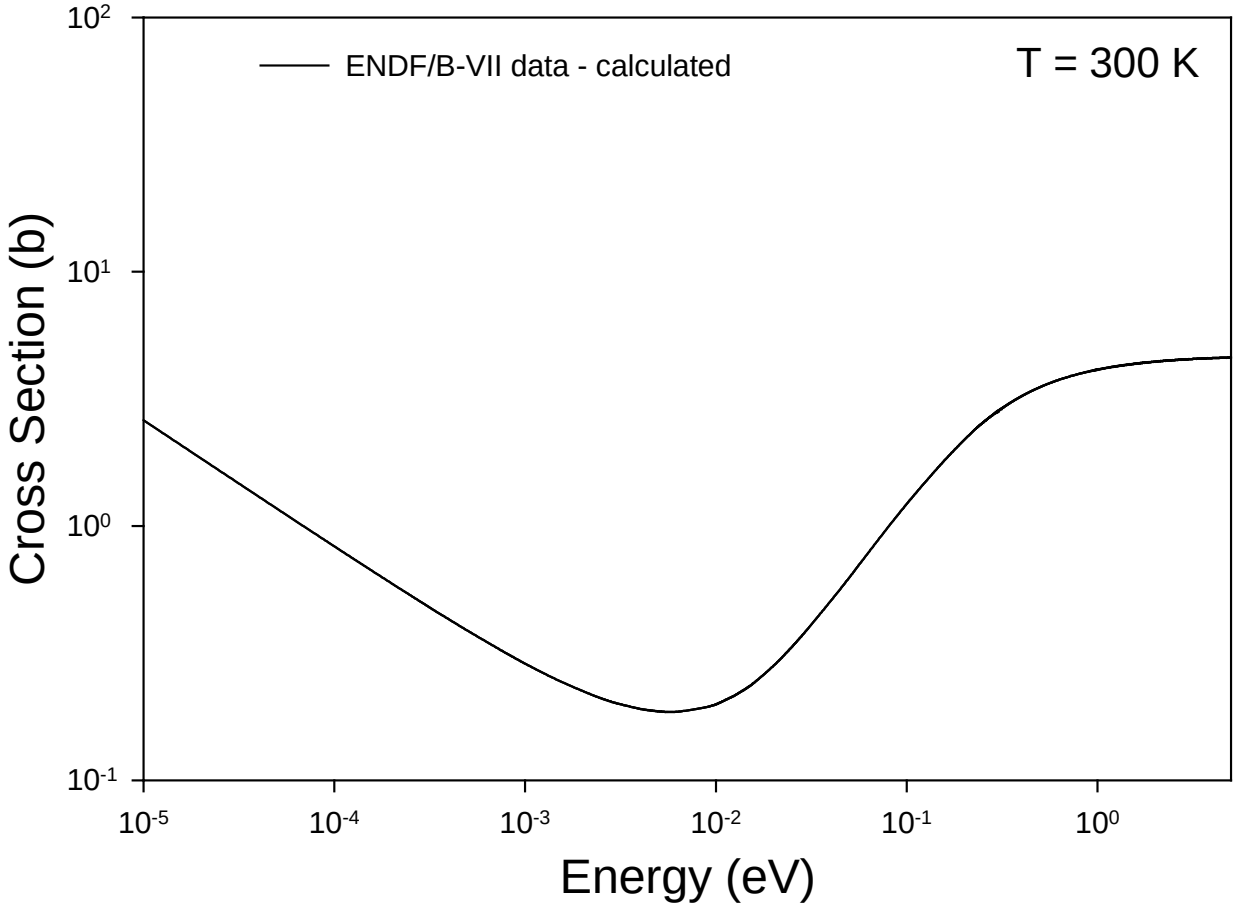
Graphite

Ideal graphite consists of planes (sheets) of carbon atoms arranged in a hexagonal lattice. Covalent bonding exits between intraplaner atoms, while the interplaner bonding is of the weak Van der Waals type. The planes are stacked in an "abab" sequence.

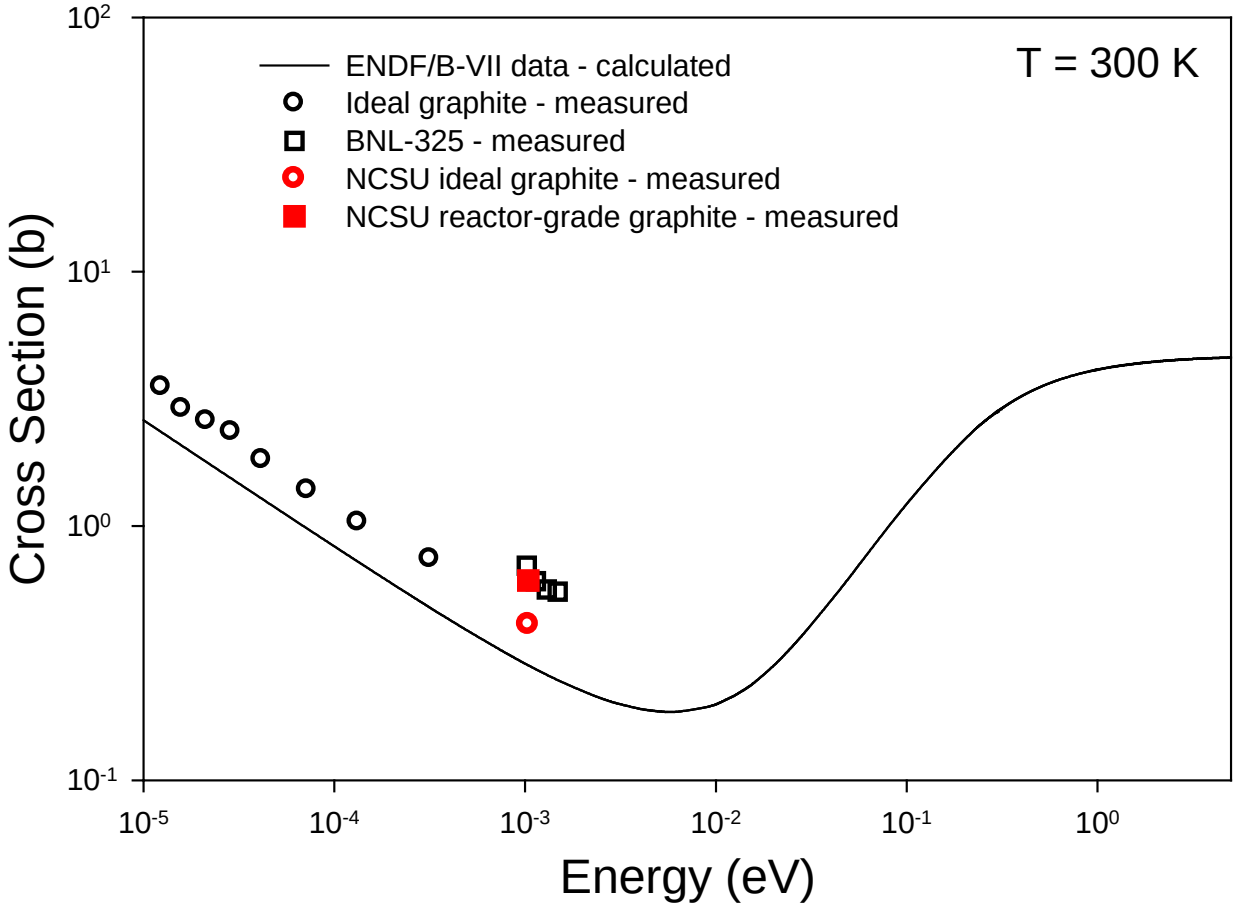


- Hexagonal Structure
- 4 atoms per unit cell
- a=b=2.46 Å
- c=6.7 Å

Graphite – 1

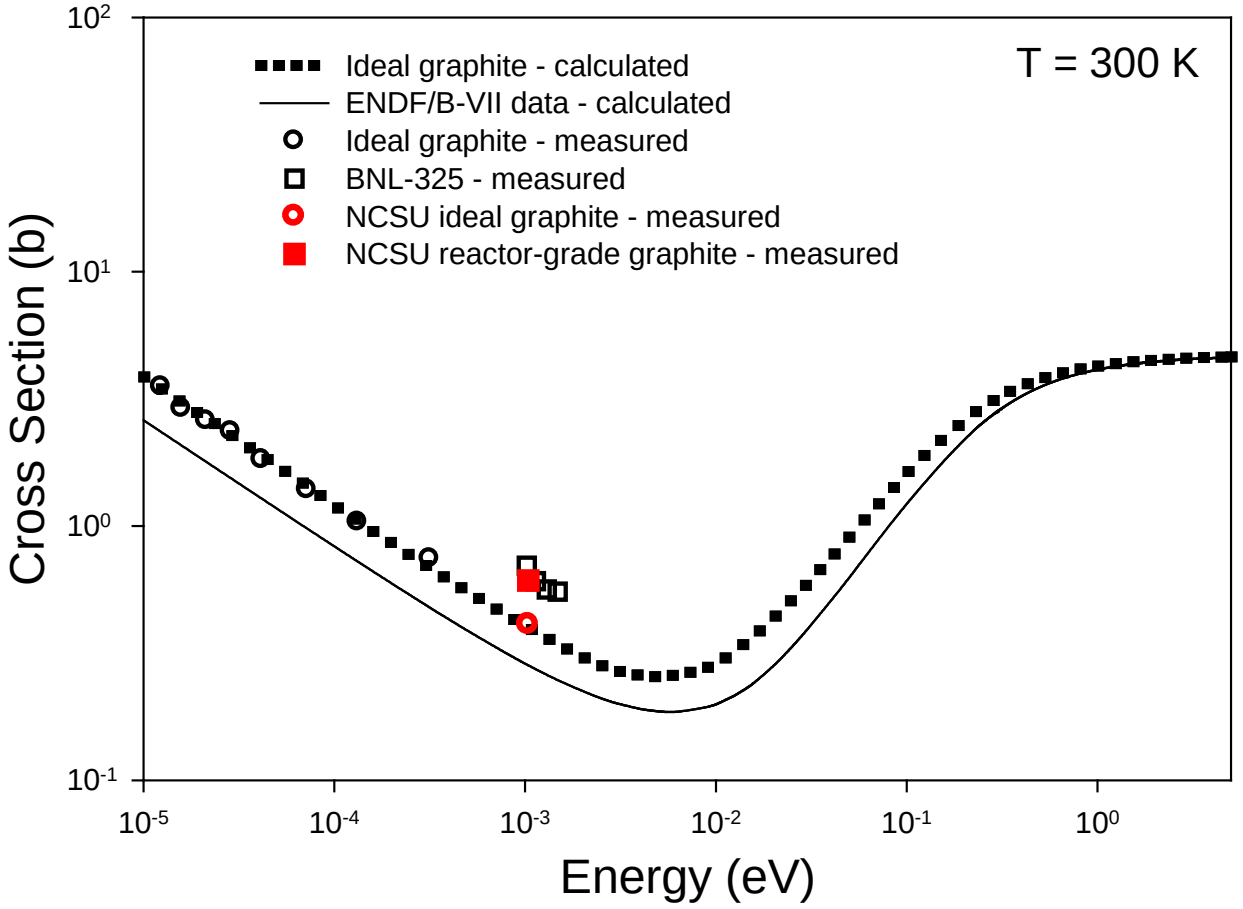

ENDF/B-VII data - calculated
T = 300 K
Cross Section (b)
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-5}
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}
Energy (eV)

Graphite – 2



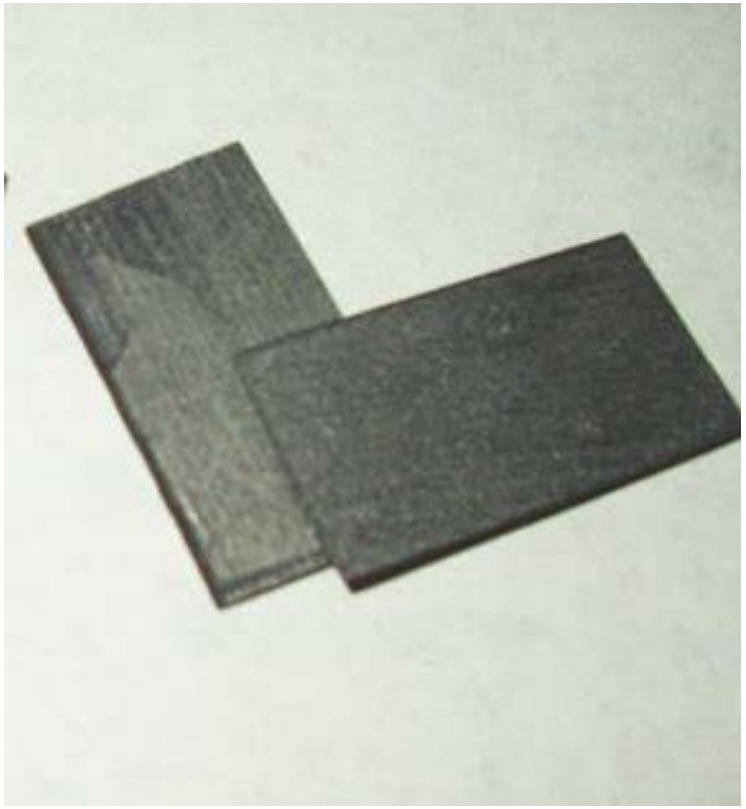
A. I. Hawari, T. Zhou, P. Huffman, "Measurement of the Total Inelastic Scattering Cross Section Of Graphite Using 9 Angstrom Neutrons," Trans. of the American Nuclear Society, v. 100, 2009

Graphite – 3



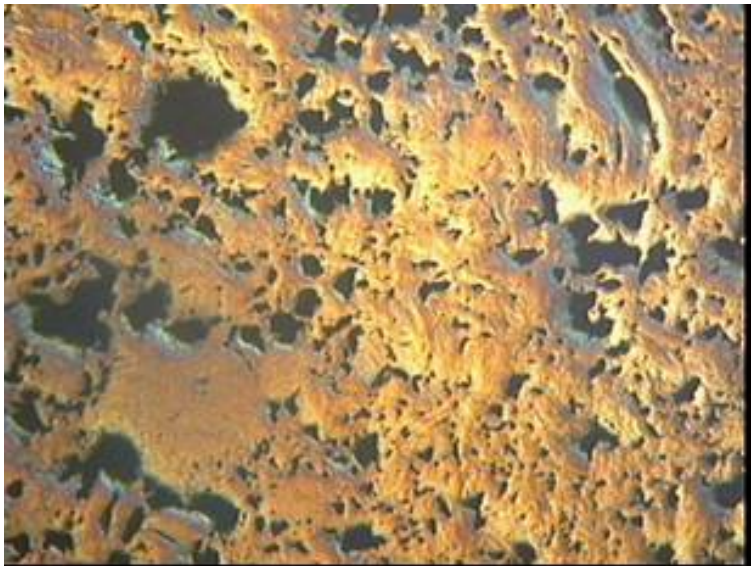
A. I. Hawari, T. Zhou, P. Huffman, "Measurement of the Total Inelastic Scattering Cross Section Of Graphite Using 9 Angstrom Neutrons," Trans. of the American Nuclear Society, v. 100, 2009

Graphite Types



Ideal Graphite

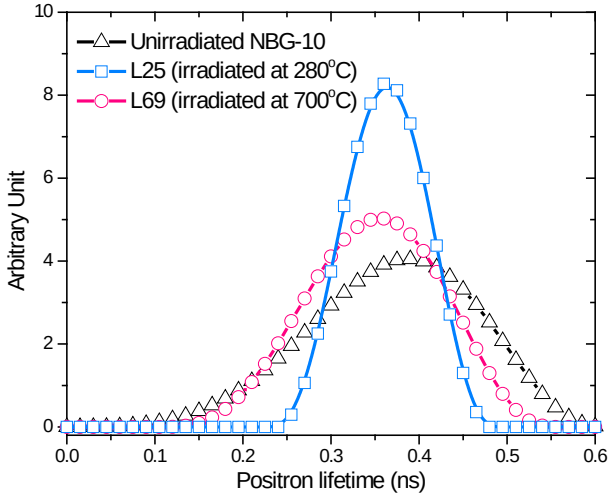
Density = 2.25 g/cm^3

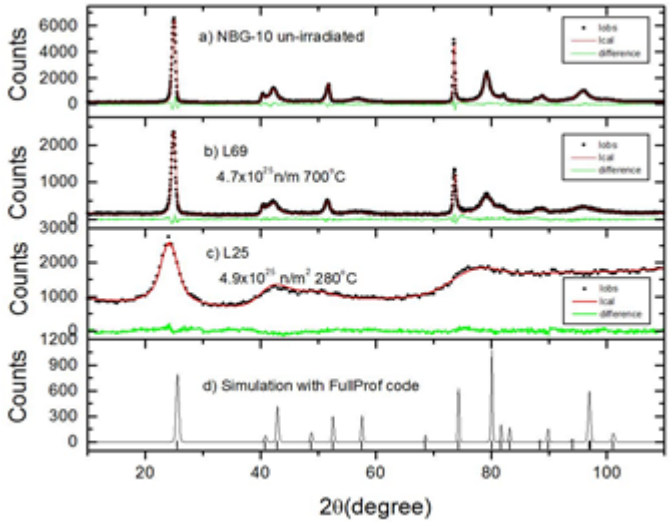


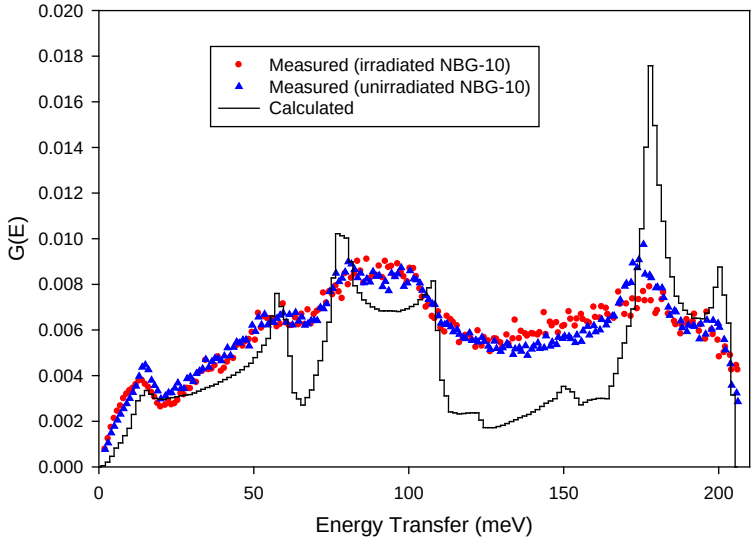
Nuclear Graphite

Density = 1.5 – 1.8 g/cm^3

Reactor-Grade (Nuclear) Graphite

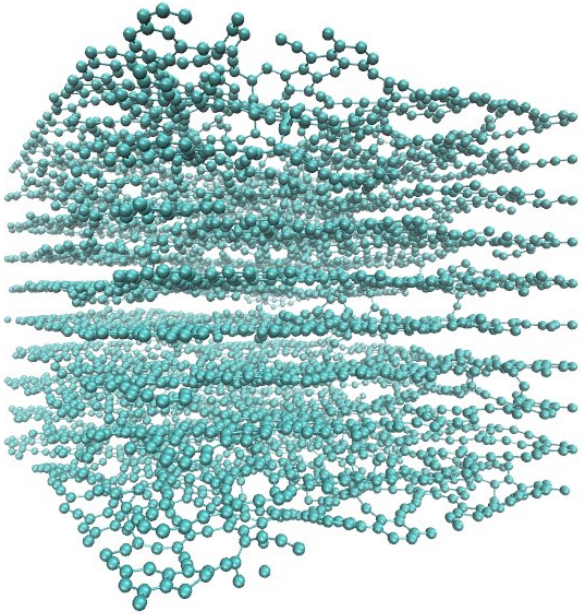




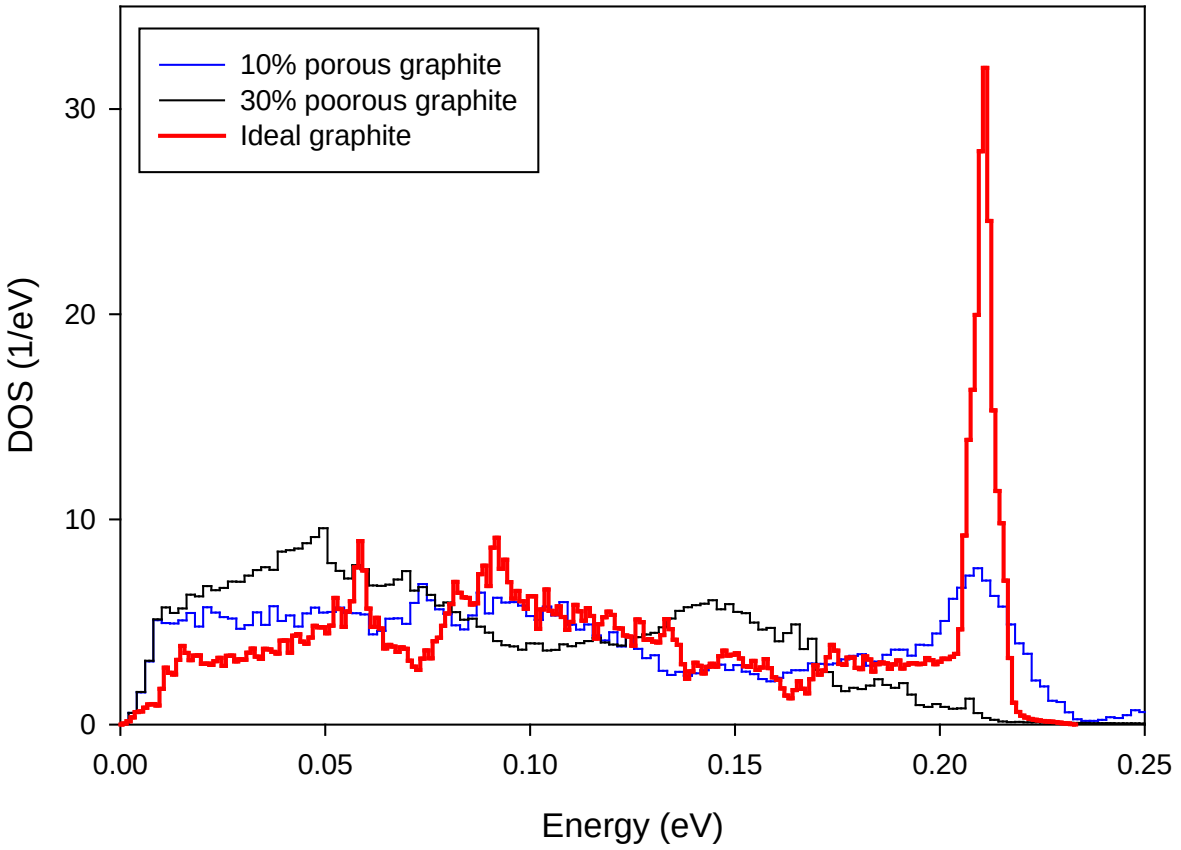


1) A. I. Hawari, A. I. Kolesnikov, Q. Cai, J. C. Holmes, P. D. Ferguson, "Inelastic Neutron Scattering Analysis of Reactor Grade Graphite," Trans. American Nuclear Societry, v. 110, 2014.
2) M. Liu, A. I. Hawari, "Positron Characterization of Neutron Irradiated Reactor Grade Graphite," Trans. American Nuclear Society, v. 110, 2014.
3) Q. Cai, A. I. Hawari, "Neutron Powder Diffraction Study of Reactor Grade Graphite," Trans. American Nuclear Society, v. 106, 2012.

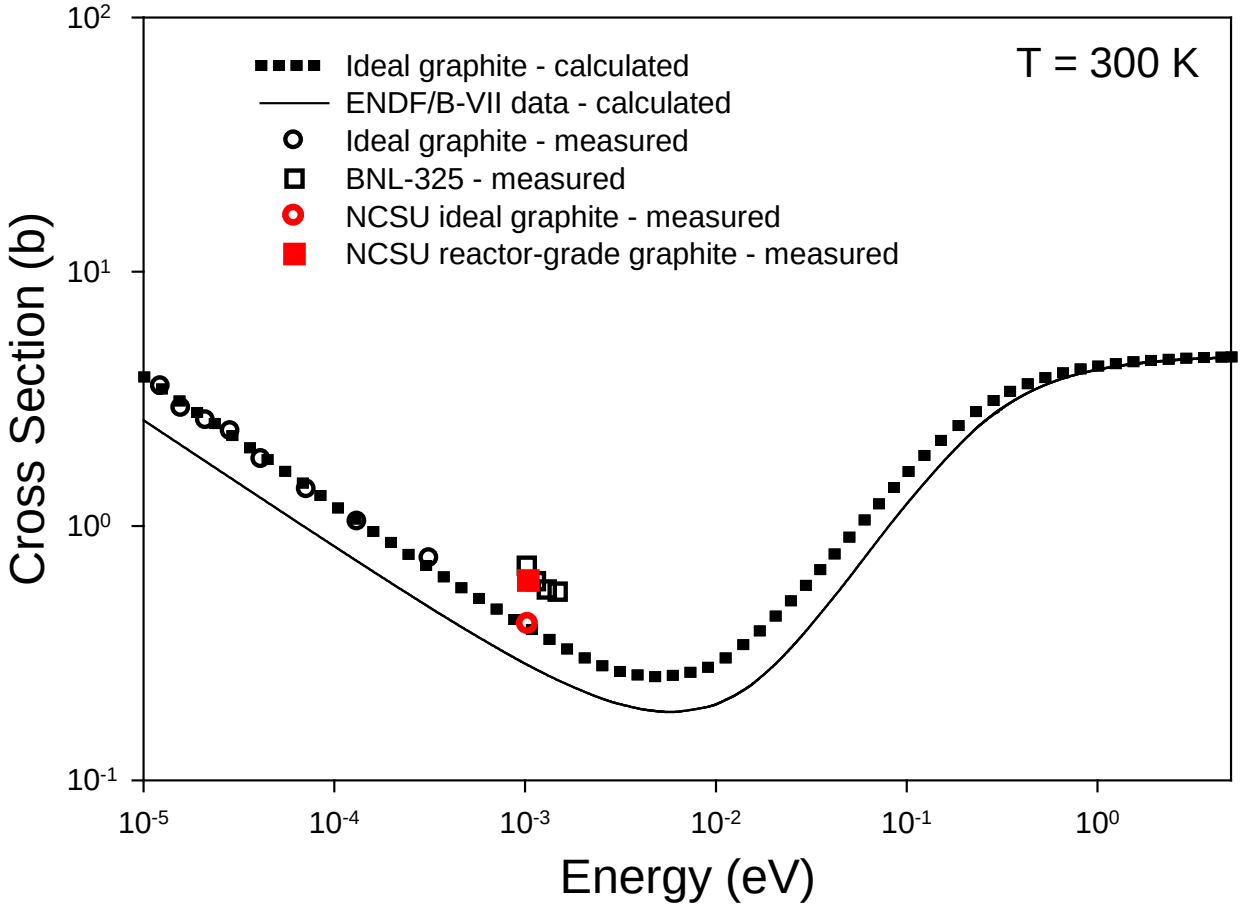
Molecular Dynamics Models



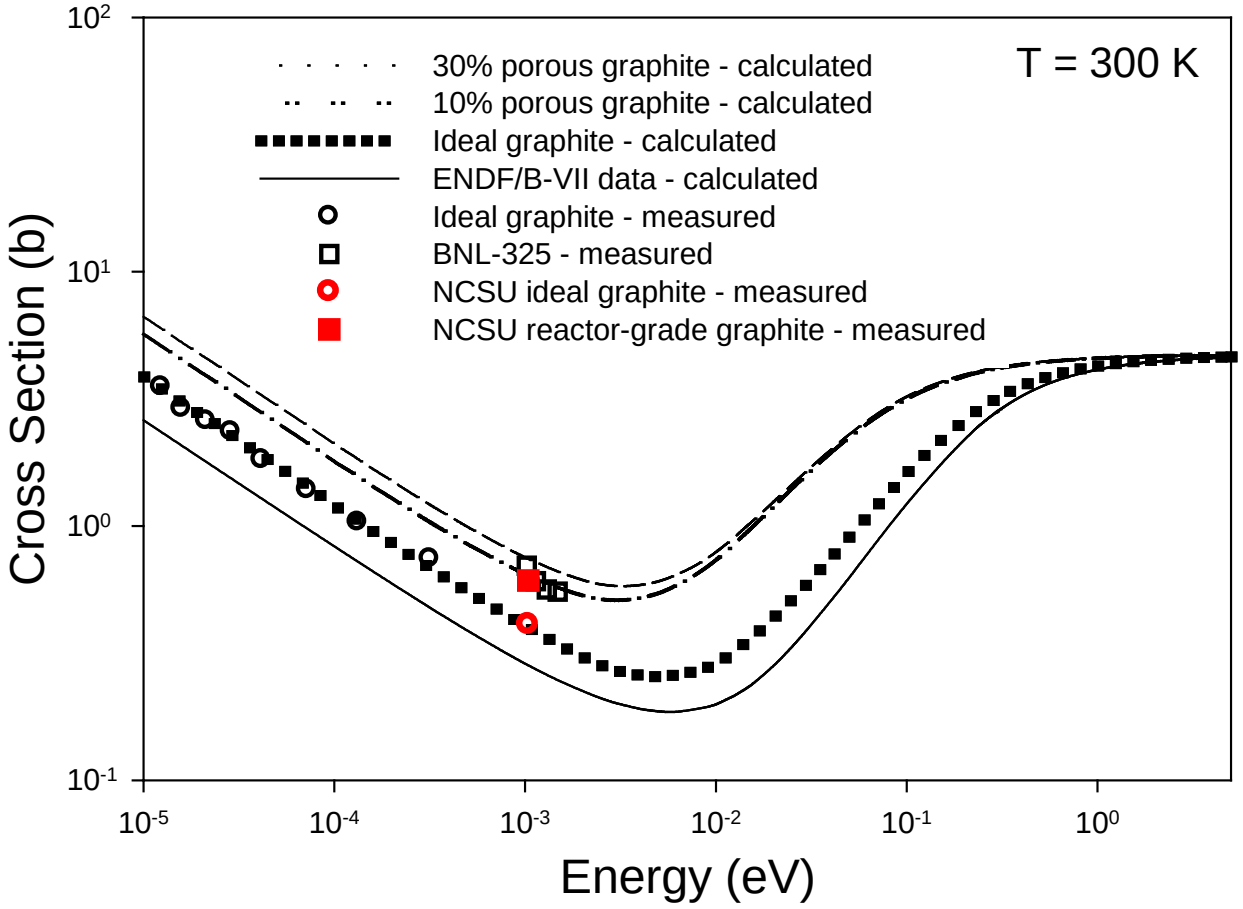
A. I. Hawari, V. H. Gillette, “Inelastic Thermal Neutron Scattering Cross Sections For Reactor-Grade Graphite,” Nuclear Data Sheets, Vol. 118, 176, 2014.



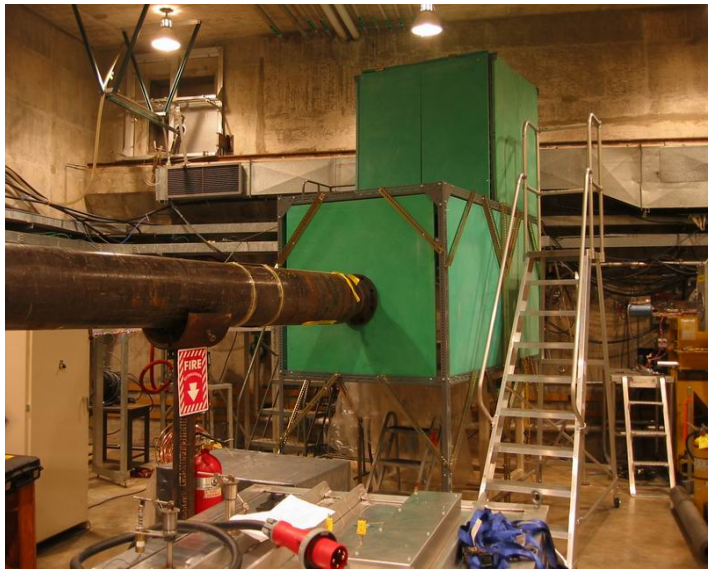
Graphite – 3

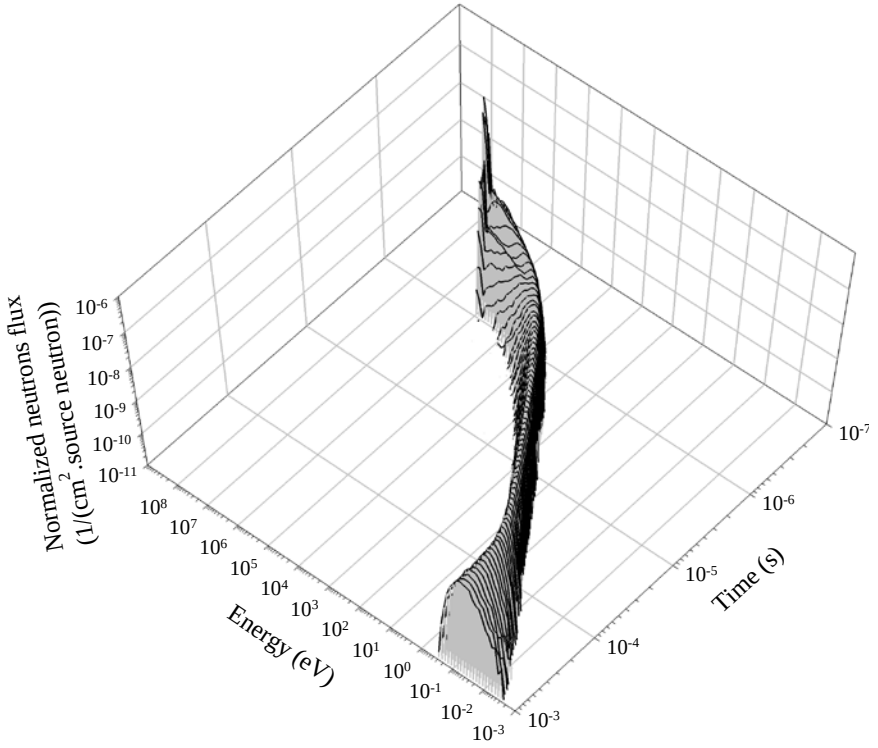

Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
T = 300 K
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

Graphite – 4

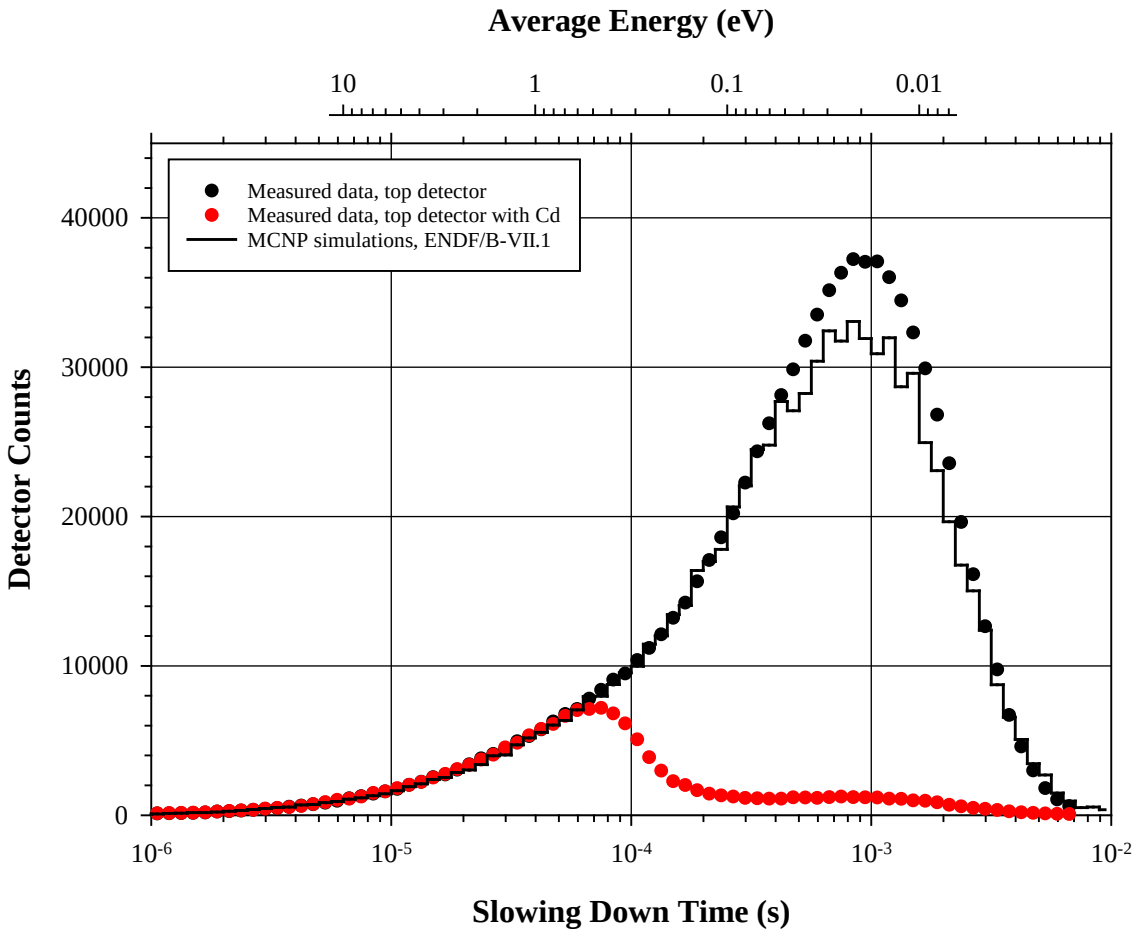

T = 300 K
30% porous graphite - calculated
10% porous graphite - calculated
Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

Slowing-Down Experiment

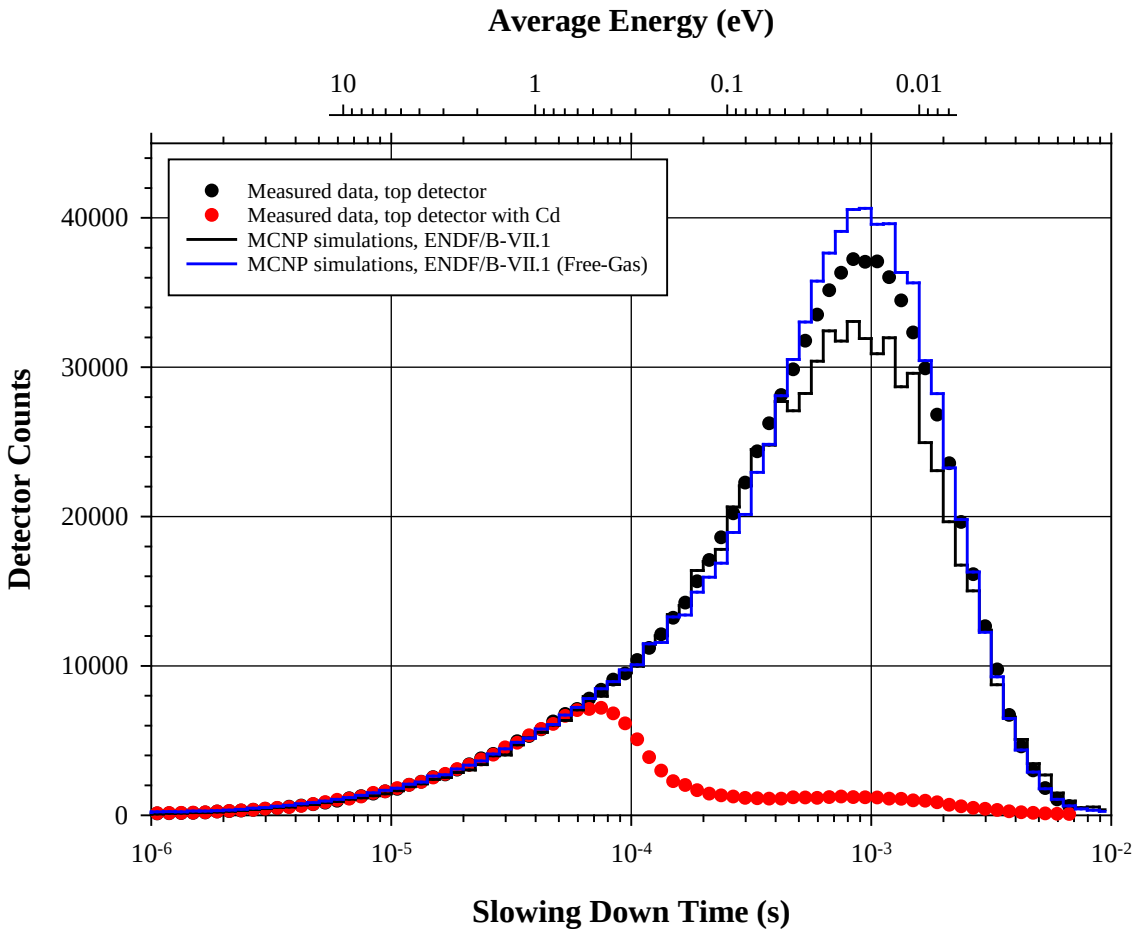




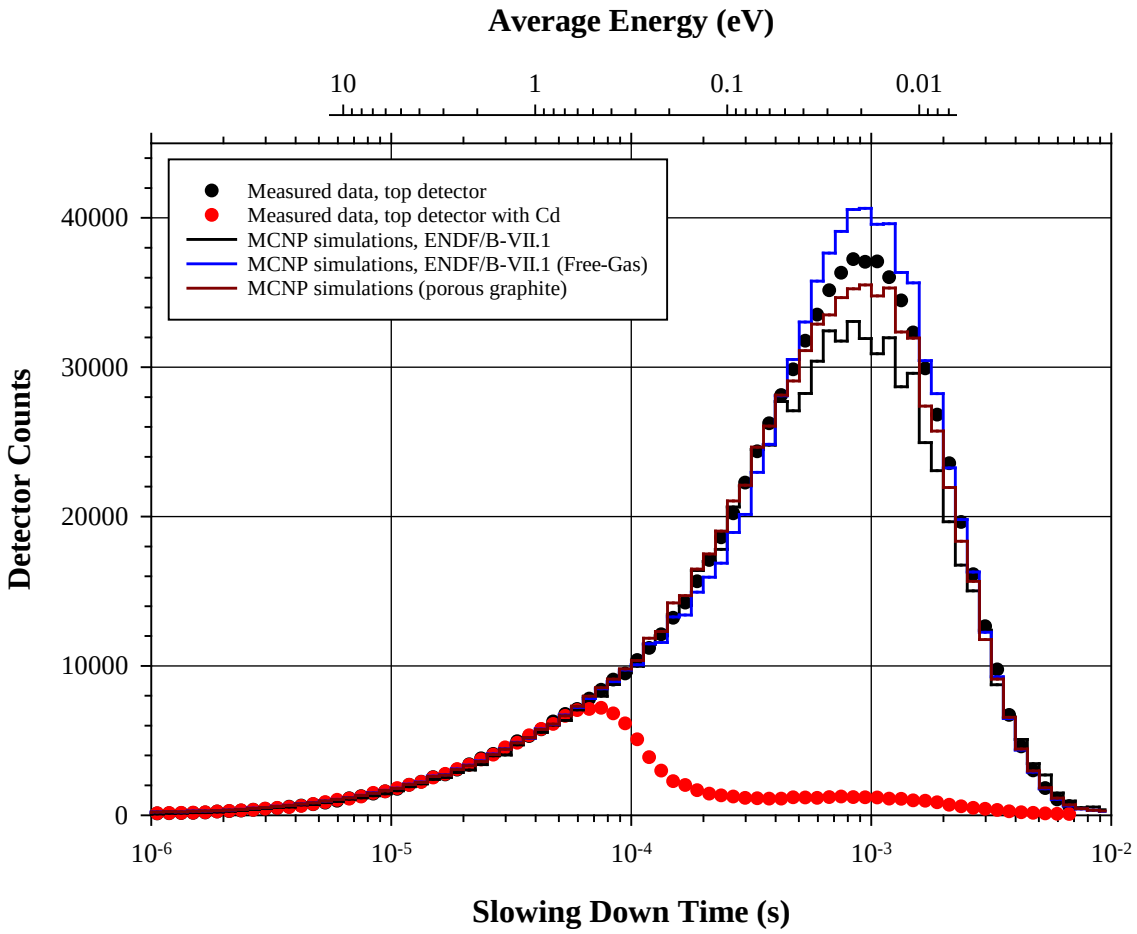
Top Detector



Top Detector



Top Detector



Evaluations 2015 – 2016

Material	Method	Comments
$(C_5H_8O_2)_n$	MD	Contributed to NNDC
$(CH_2)_n$	MD	ENDF File 7 ready
Lubricant Oil	MD	On going
BeO	DFT	ENDF File 7 ready
UO_2	DFT	ENDF File 7 under preparation
U_3Si_2	DFT	On going
FLiBe	MD	Near completion
Reactor-grade (nuclear) graphite	MD	File 7 ready (porous model)

Summary

- Developed and applied a modern approach for thermal neutron cross section calculations based on the use of atomistic simulations
  - Ab initio lattice dynamics
  - Molecular dynamics (ab initio and classical)
- The approach is predictive
  - New materials
  - All states of matter (solid, liquid, gas)
  - Imperfect structure
- Coupling modern computations with selected validation experiments should address all TSL data needs