US National Nuclear Data Week 2015
November 2 – 6, 2015 • Brookhaven National Laboratory, Upton, NY, USA

Thermal Scattering Law Data Generation and Validation In the 21st Century

Ayman I. Hawari

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

Acknowledgement

- Past and current undergraduate and graduate students and postdocs at North Carolina State University

- Collaboration with LLNL and Bettis labs

- Funding by the NCSP and NR programs

Vision

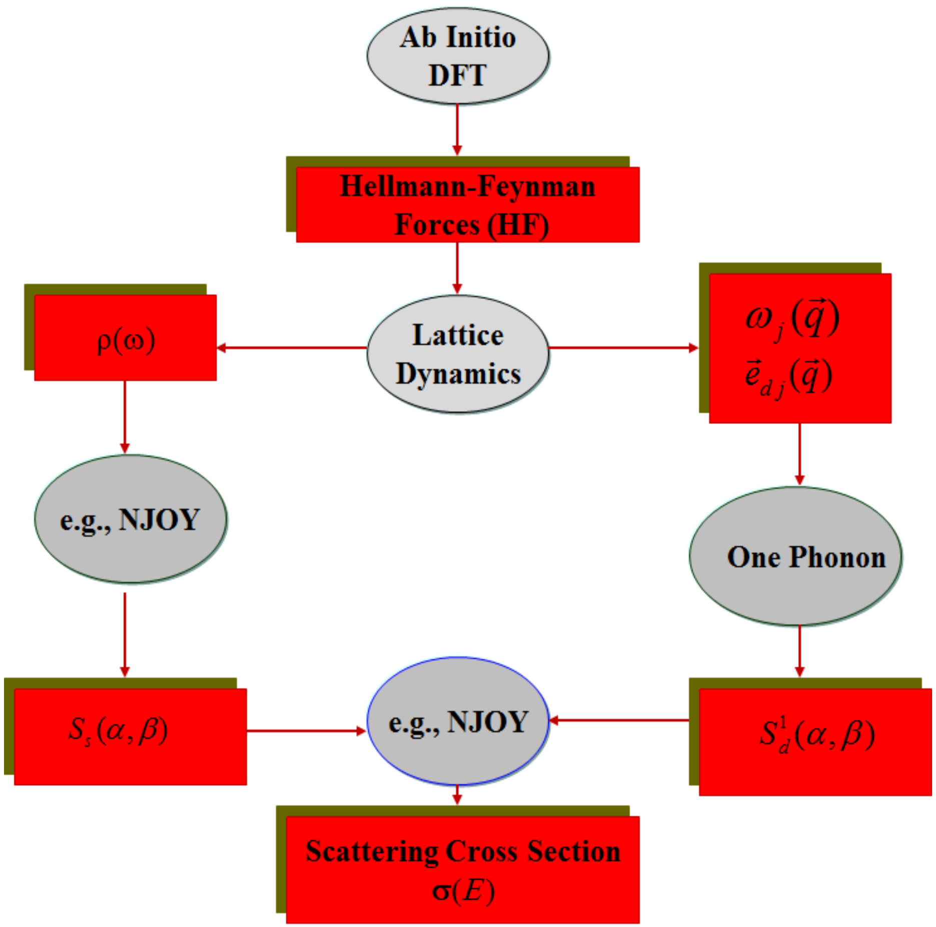
- Establish a predictive and approximation free approach for generating the needed data (i.e., TSL) to describe the energy exchange of thermal neutrons in matter
  - Initiated in 2000/2001

- Various applications:
  - Nuclear criticality safety
  - Nuclear reactor design
  - Neutron beam spectral shaping (i.e., filtering)
  - Neutron source (cold, ultracold, etc.) characterization

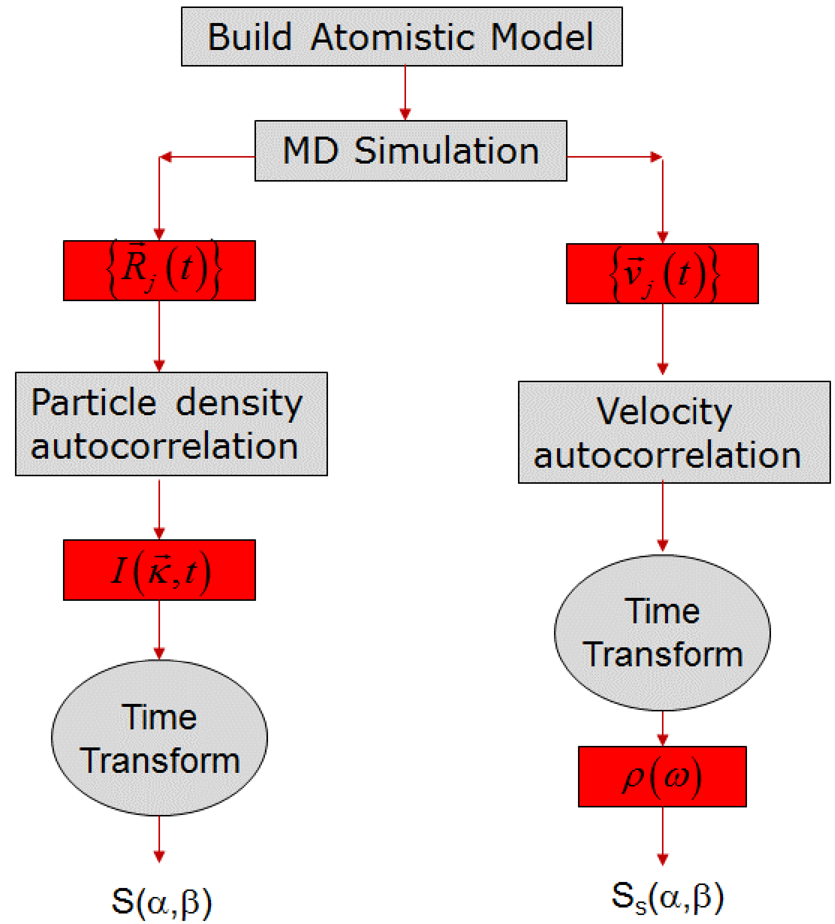
Methods

- S(α,β) is a material property
  - can be calculated very accurately!
- Several approaches can be used to extract the fundamental information for calculating the scattering law and eventually the cross sections
  - Empirical atomic force analysis combined with dynamical matrix calculations
    - Basis of current ENDF/B libraries
  - Ab initio Quantum (DFT) methods combined with dynamical matrix calculations
  - Molecular Dynamics (classical MD or ab initio) methods combined with correlation function analysis

DFT Based

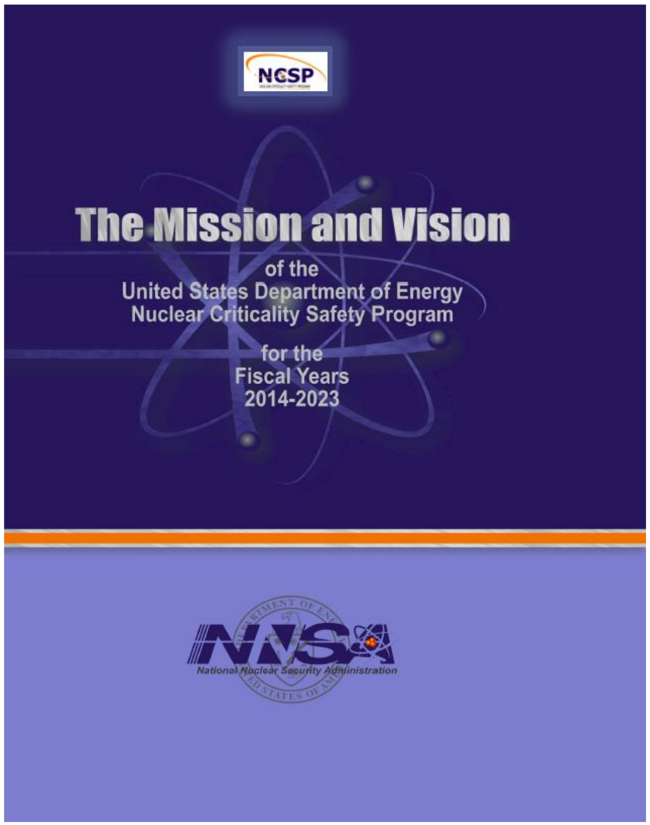


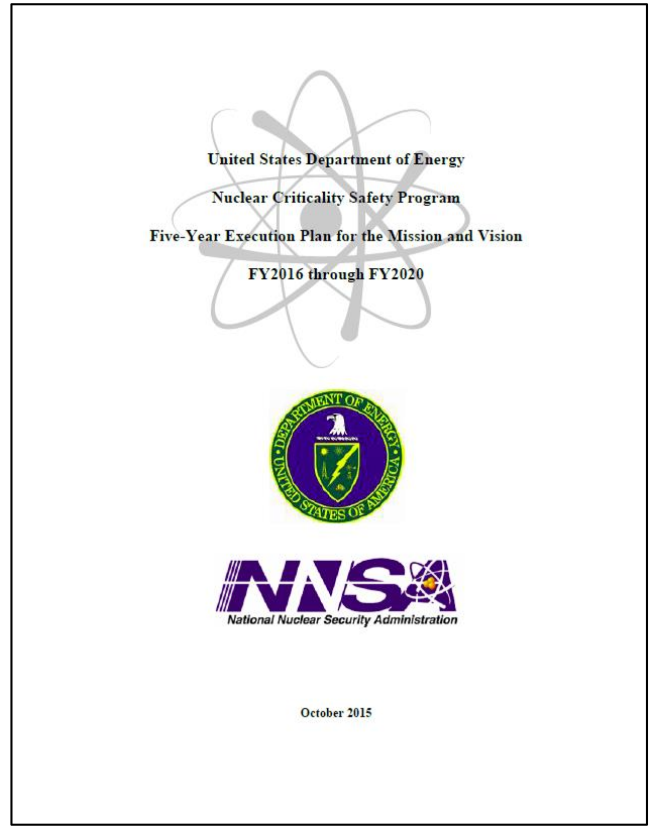
MD Based



Merge both into the DFT based Ab Initio MD approach

NCSP Program (with NR)





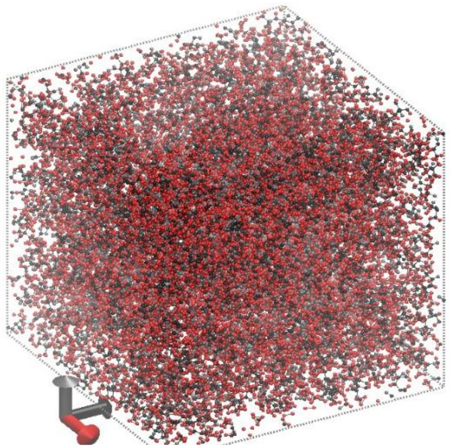
Appendix B
Nuclear Data

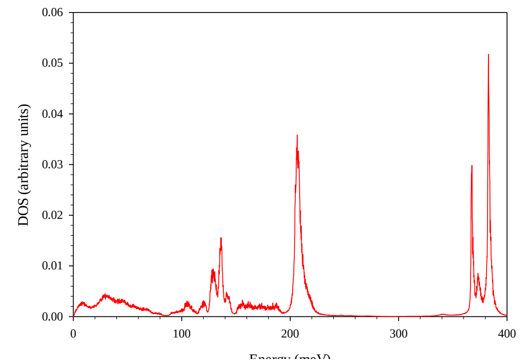
Priority Needs */ **Additional Needs**	Thermal scattering (BeO, HF, D_2O, SiO_2, CH_2, C_2F_4, $C_5O_2H_8$, etc.), ^{239}Pu, Cr, ^{237}Np, Pb, ^{55}Mn, Ti, ^{240}Pu / ^{233}U, Th, Be, ^{51}V, Zr, F, K, Ca, Mo, Na, La
Completed Evaluations (FY)	Minor Actinides (13), SiO_2 (12), ^{55}Mn (12), $^{180,128,183,184,186}W$ (14)

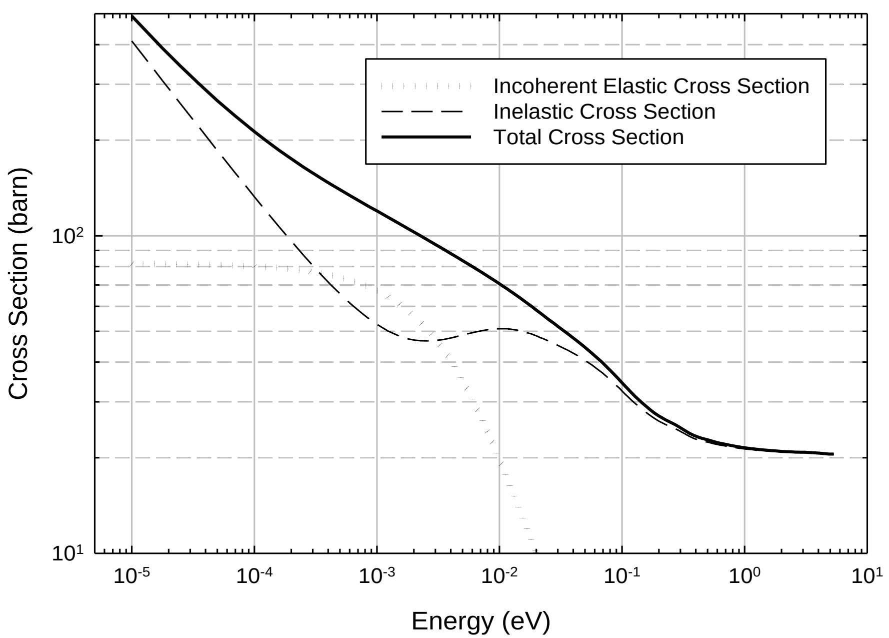
Examples - Materials Studied at NCSU

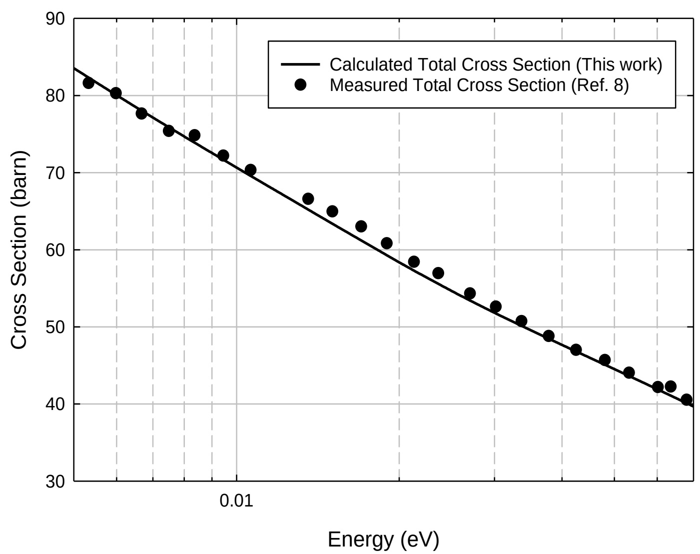
- Lucite ($C_5O_2H_8$, contributed to NNDC/ENDF, ANS 2015)
- Polyethylene (CH_2)
  - Support criticality safety applications
- Silicon dioxide (contributed to NNDC/ENDF, PHYSOR 2008, ANS 2011, NDS 2014)
  - Support criticality safety applications
- Silicon carbide (contributed to NNDC/ENDF, ANS 2013)
  - Support advanced fuel cycle applications
- Graphite, Beryllium (PHYSOR 2004 & 2008, NDS 2014, PHYSOR 2016), BeO (unpublished)
  - Treatment of nuclear graphite (porous system)
  - Including coherent inelastic for both 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 ($C_5O_2H_8$)





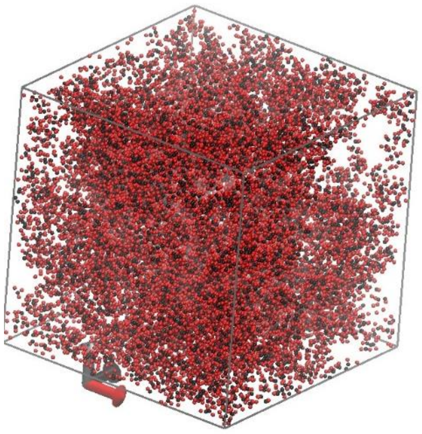


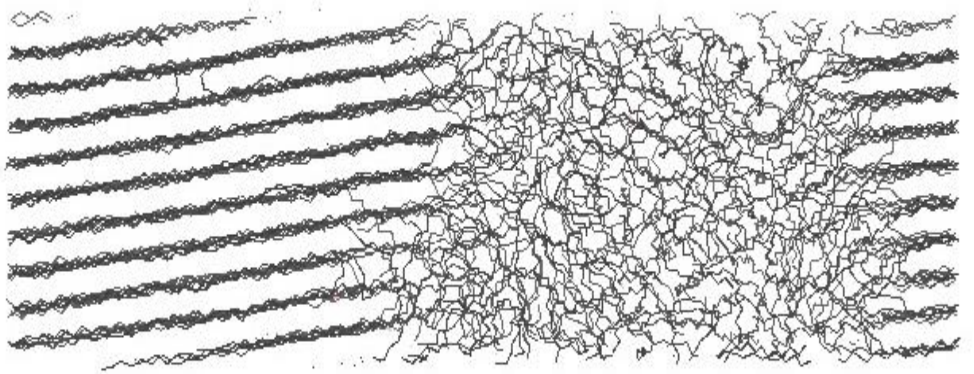


Exp. Data: G. SIBONA et al., Anna. Nucl. Energy, 18, 689 (1991).

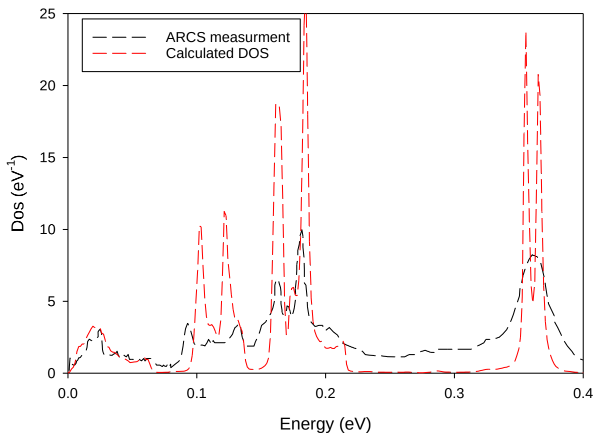
Polyethylene (CH_2) System

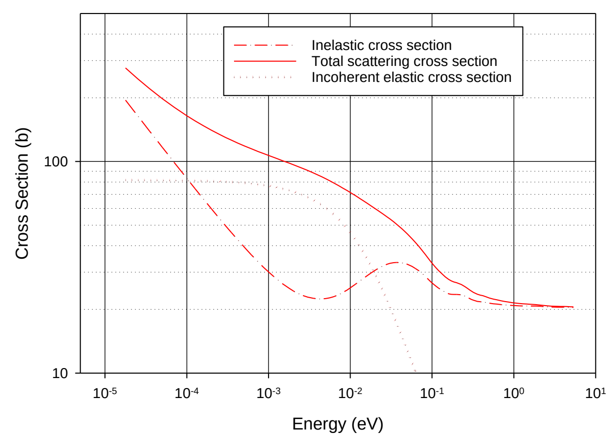
- Systems of multiple polymer chains are needed to account for inter-chain interactions.
- Simulated 20 polymer chains, each 200 monomers long, 24,000 atoms.

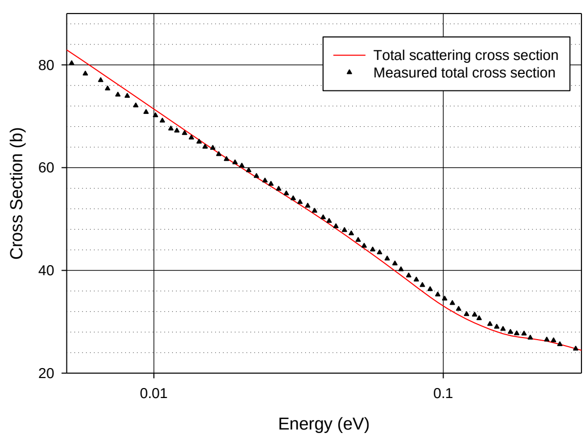


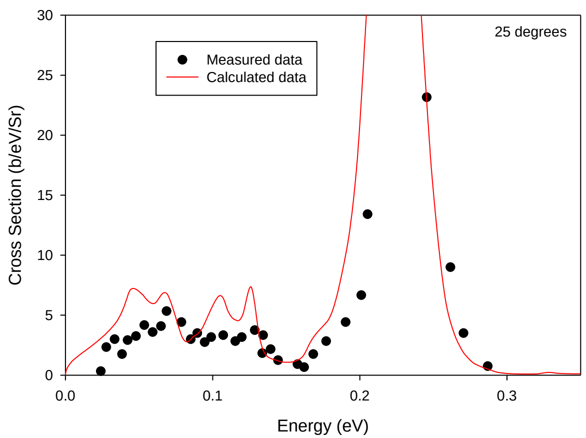


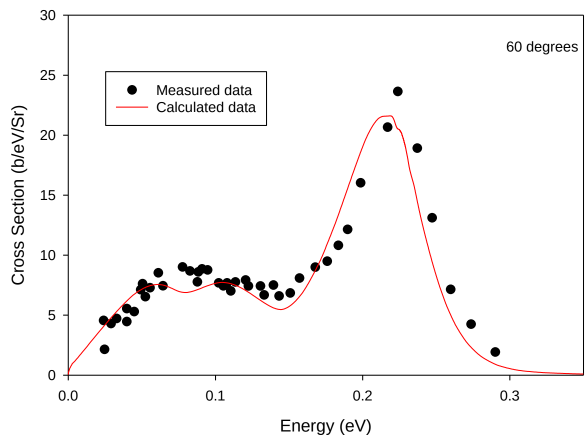
Polyethylene (CH_2)











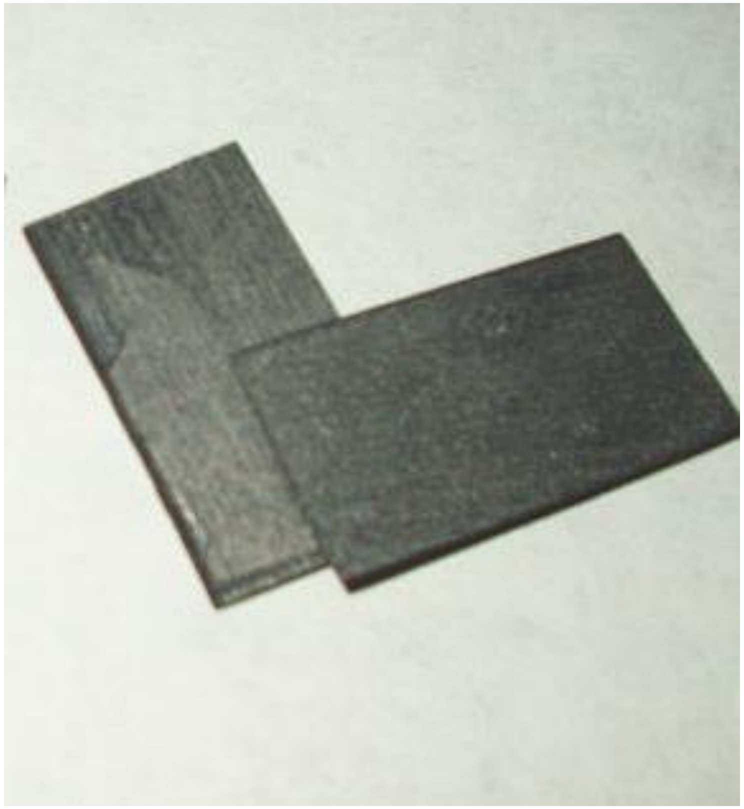
Exp. Data:

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).

TSL Data Evaluation Path Forward

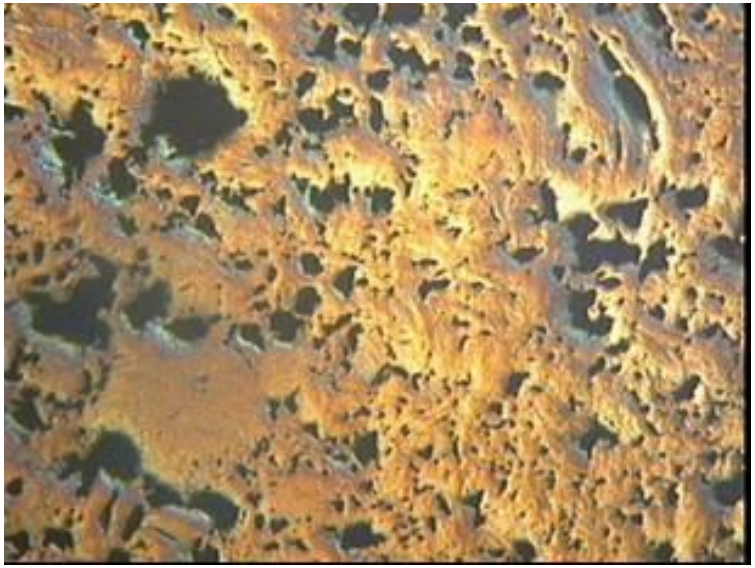
- **Simulate/Calculate**
  - **Generate TSL libraries (very efficient)**
    - Support applications
    - Support design and interpretation of validation and benchmark experiments
    - Capture variations in system conditions
- **Validate**
  - Selected experiments
- **Create next generation tools**
  - **TSL and cross section calculations**
    - Take advantage of the wealth of information available through modern atomistic modeling techniques
  - **Uncertainty quantification**

Graphite Types



Ideal Graphite

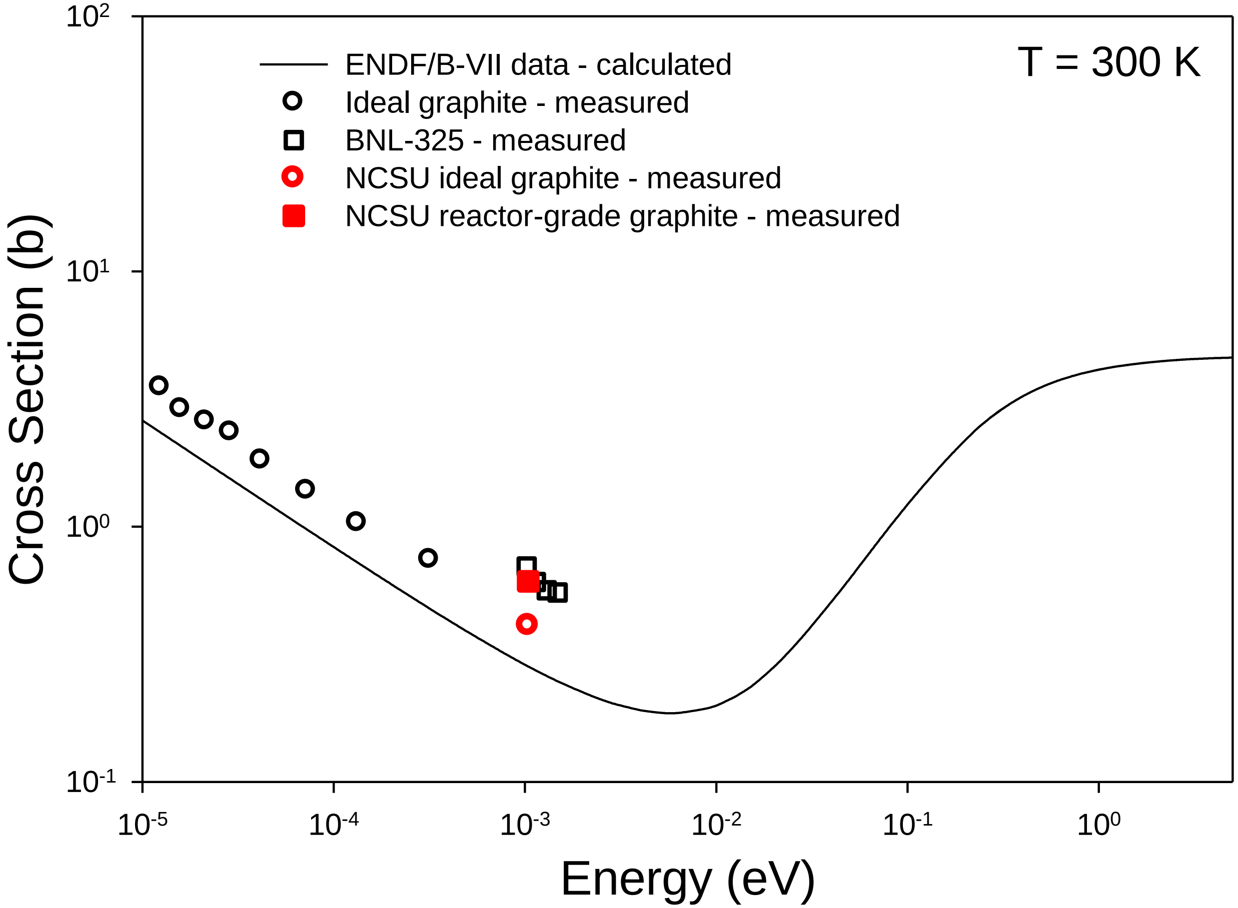
Density = 2.25 g/cm^3



Nuclear Graphite

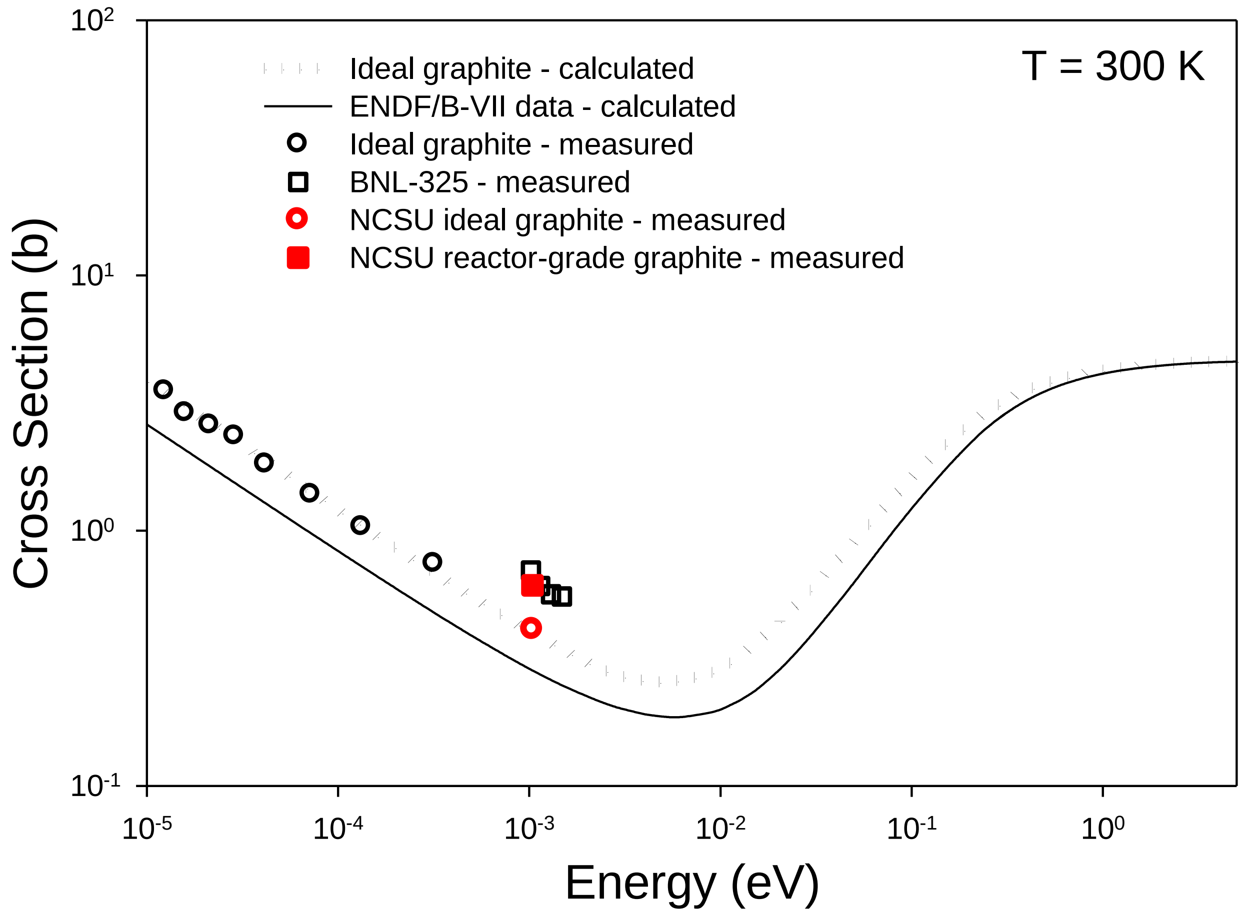
Density = 1.5 – 1.8 g/cm^3

Graphite



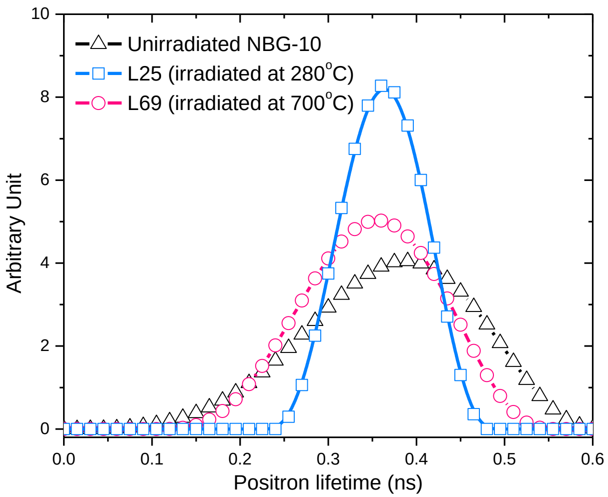
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
T = 300 K
Cross Section (b)
Energy (eV)
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-5}
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}

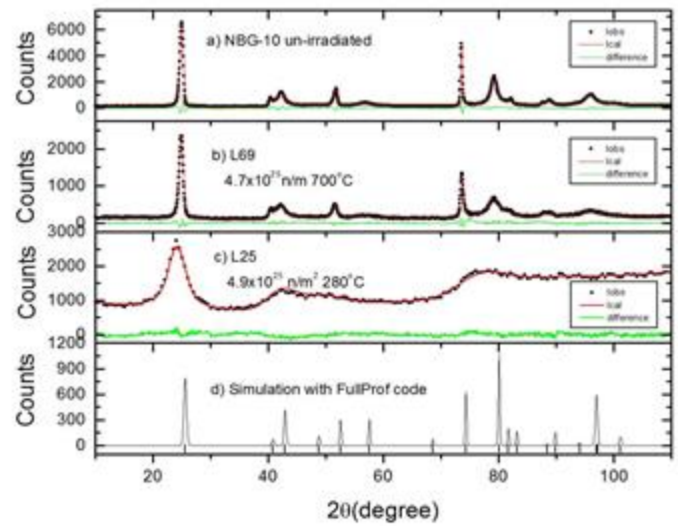
Graphite

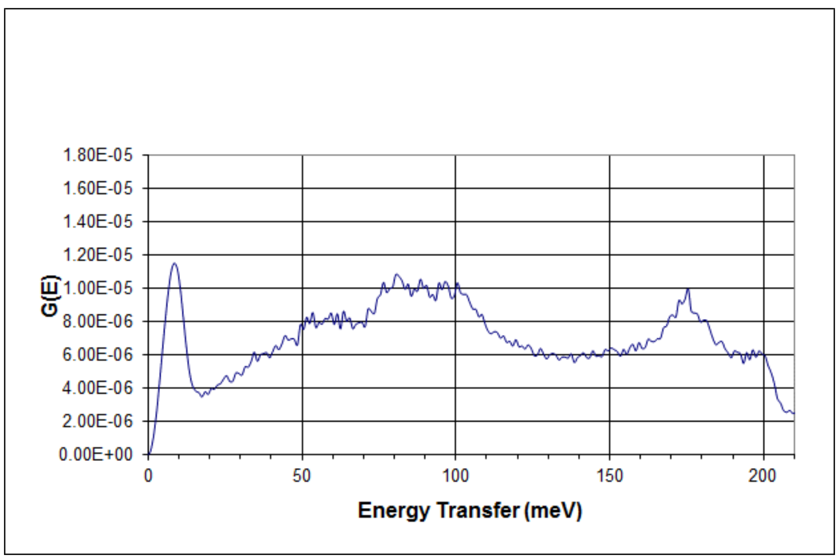


T = 300 K
Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

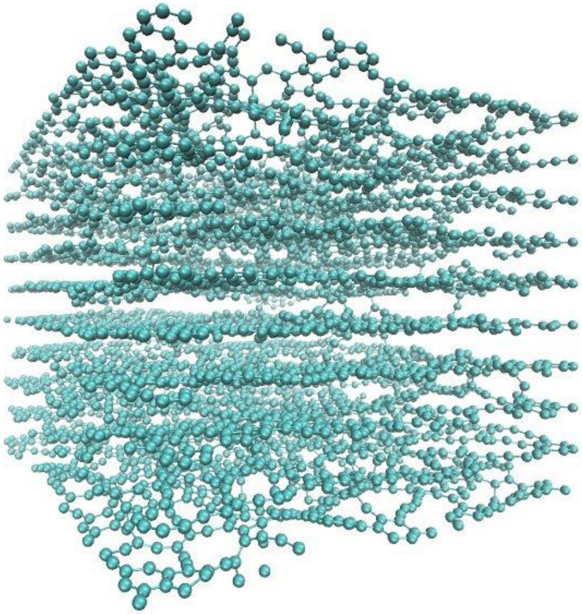
Targeted Experiments

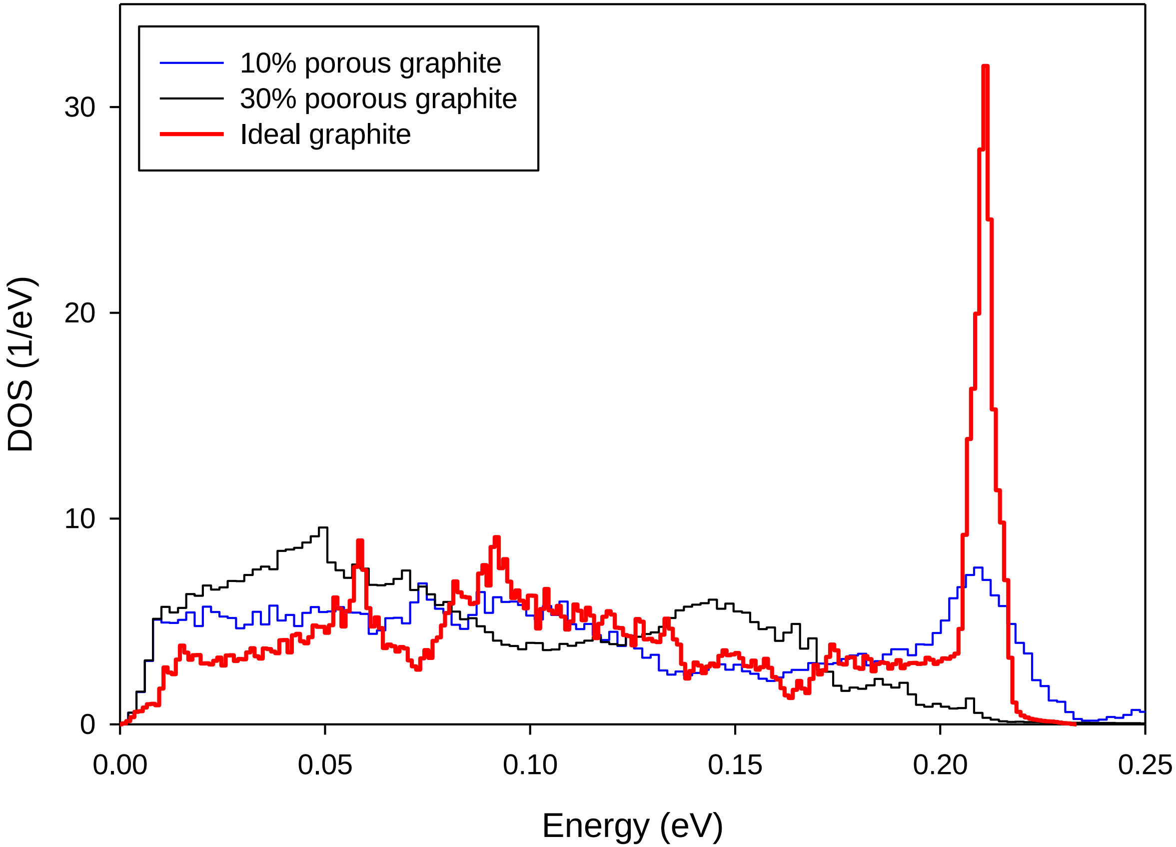




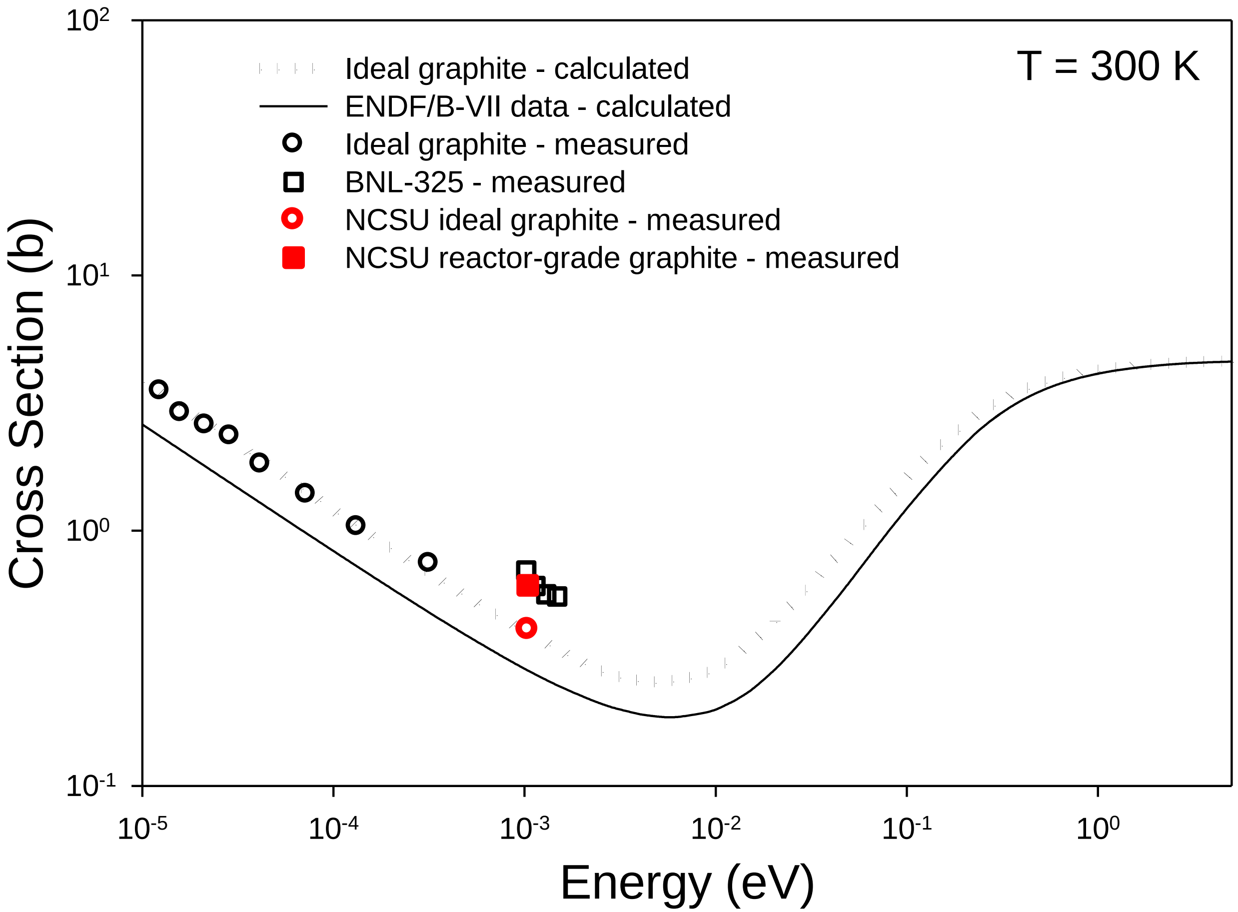


Molecular Dynamics Models

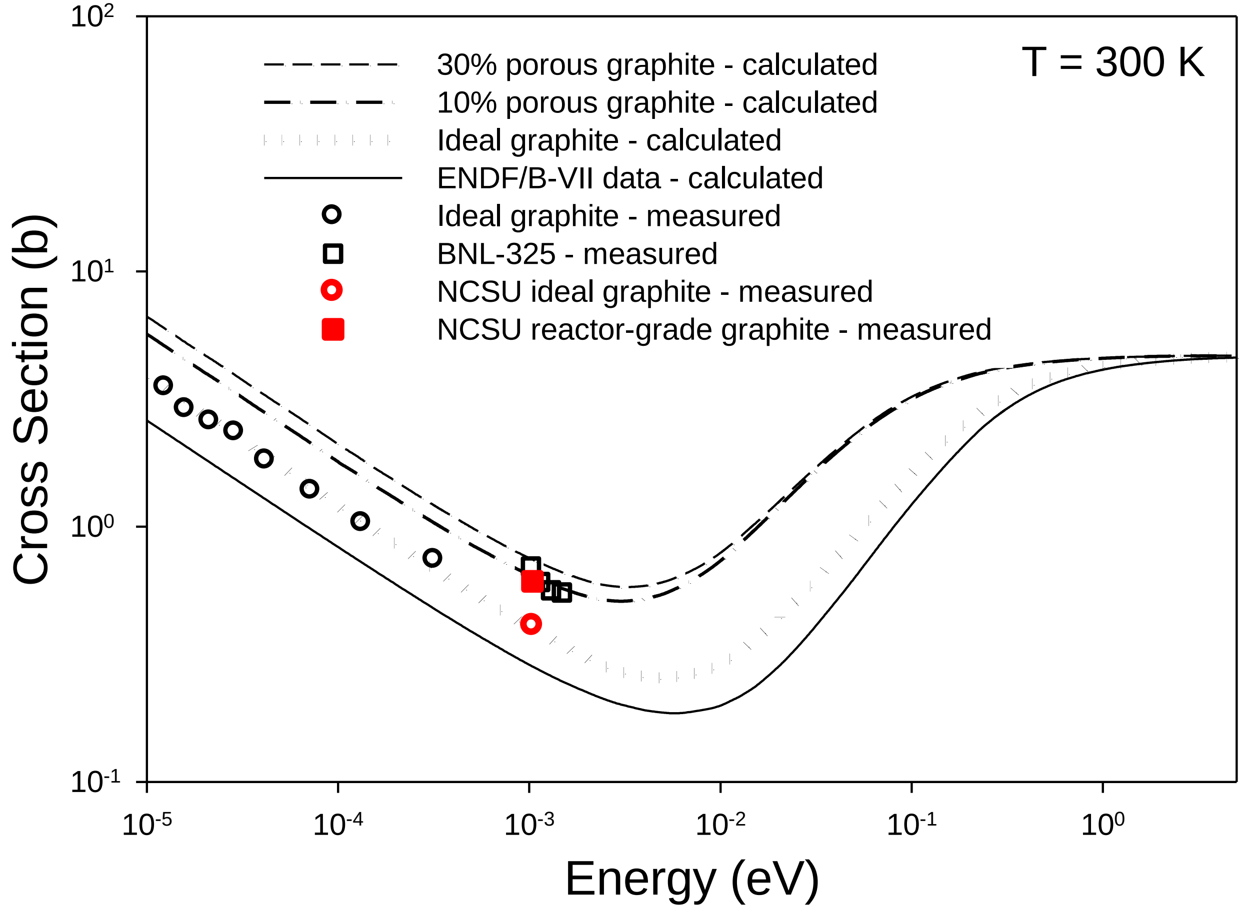




Graphite

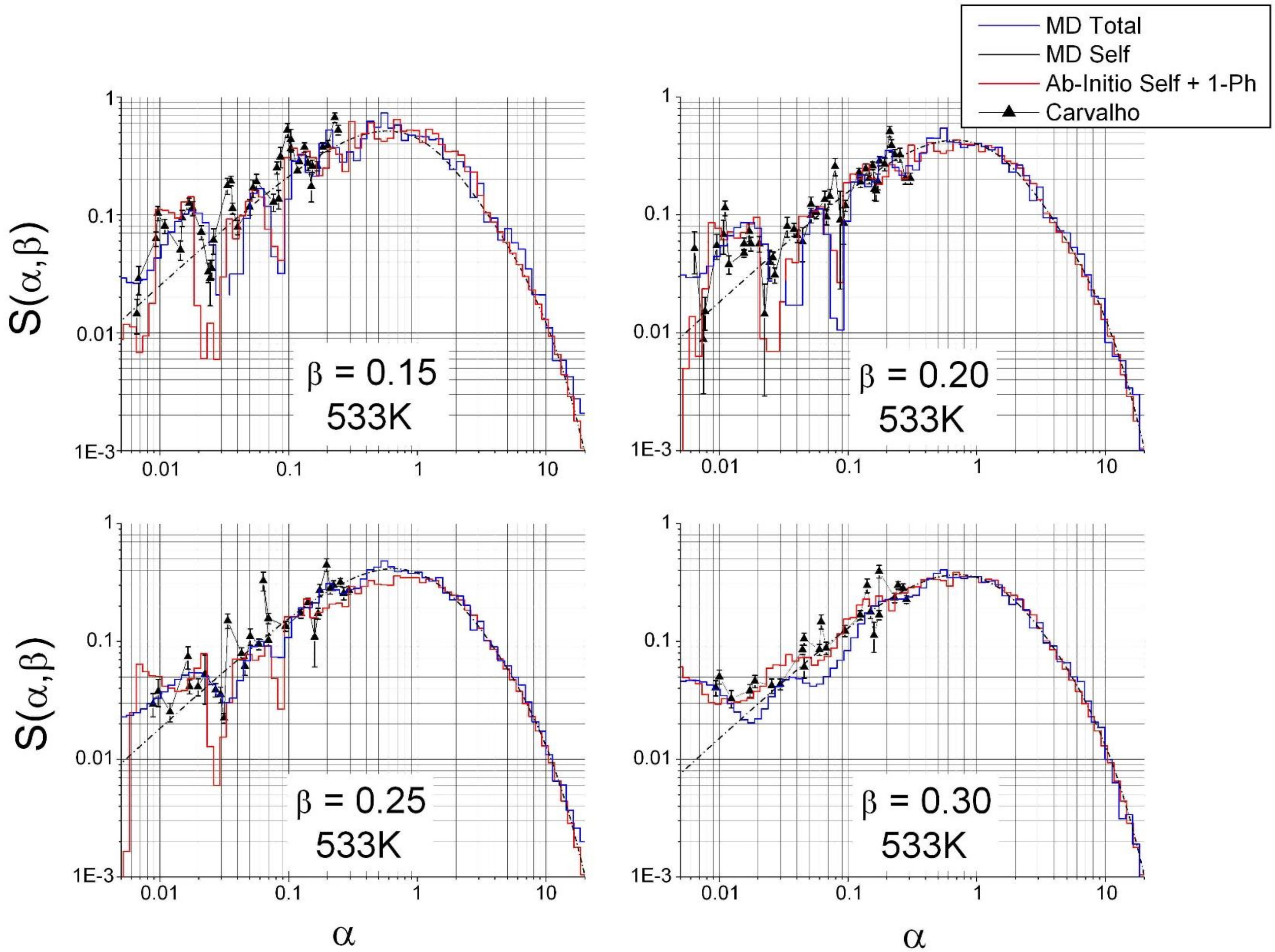

T = 300 K
Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
Cross Section (b)
Energy (eV)
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-5}
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}

Graphite



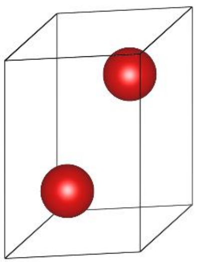
T = 300 K
30% porous graphite - calculated
10% porous graphite - calculated
Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
Cross Section (b)
Energy (eV)
10^{2}
10^{1}
10^{0}
10^{-1}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

Graphite Scattering Law

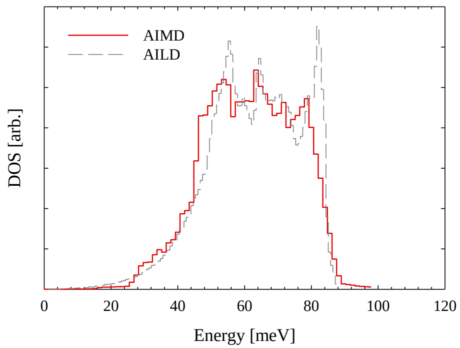


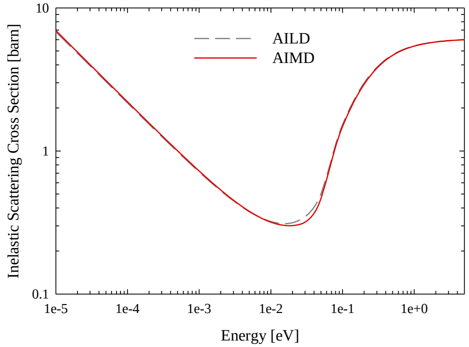
Holistic Approach
Ab Initio Molecular Dynamics

- Be – HCP ($P6_3/mmc$)
  - a=2.2856 (2.27 AIMD)
  - c=3.5832 (3.55 AIMD)
- VASP - AIMD
  - 5x5x5 supercell
  - GGA-PAW
  - 3x3x3 k-mesh
  - Fermi 'smearing' of electronic structure occupation
  - 350eV Plane-wave cut-off
  - 300 K under NVE conditions



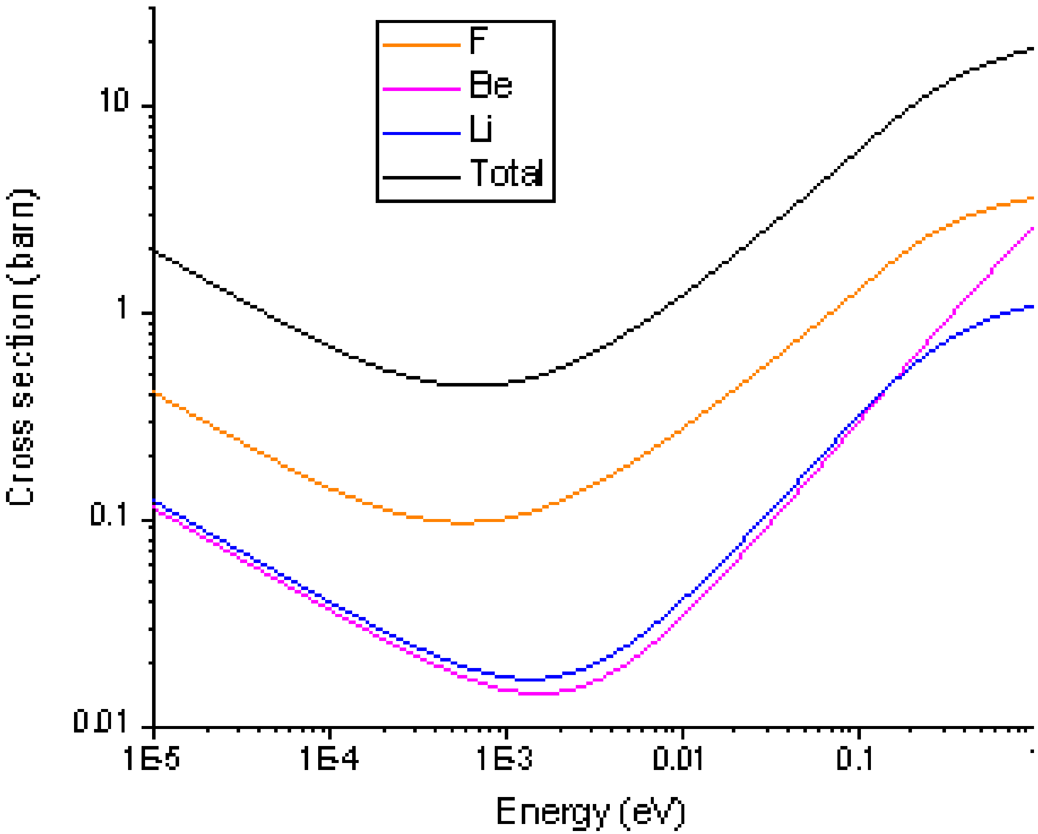
Accepted PHYSOR 2016





First Look
Liquid FLiBe

- MD simulations combined with analysis to extract of the TSL data



Accepted PHYSOR 2016

Summary

- Developed 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 targeted validation experiments should address all TSL data needs