Benchmark and Validation of Thermal Neutron Scattering in Nuclear Graphite

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Acknowledgement

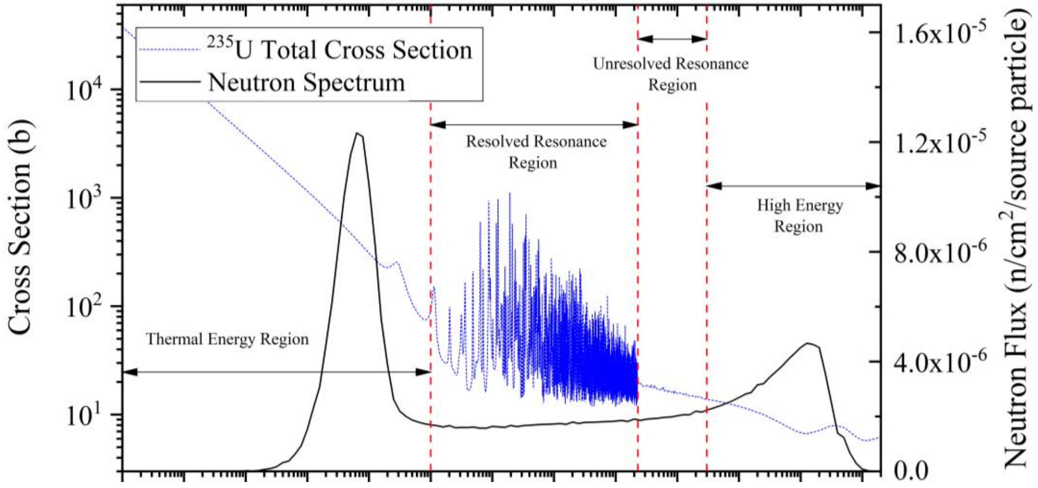
- The LEIP Team

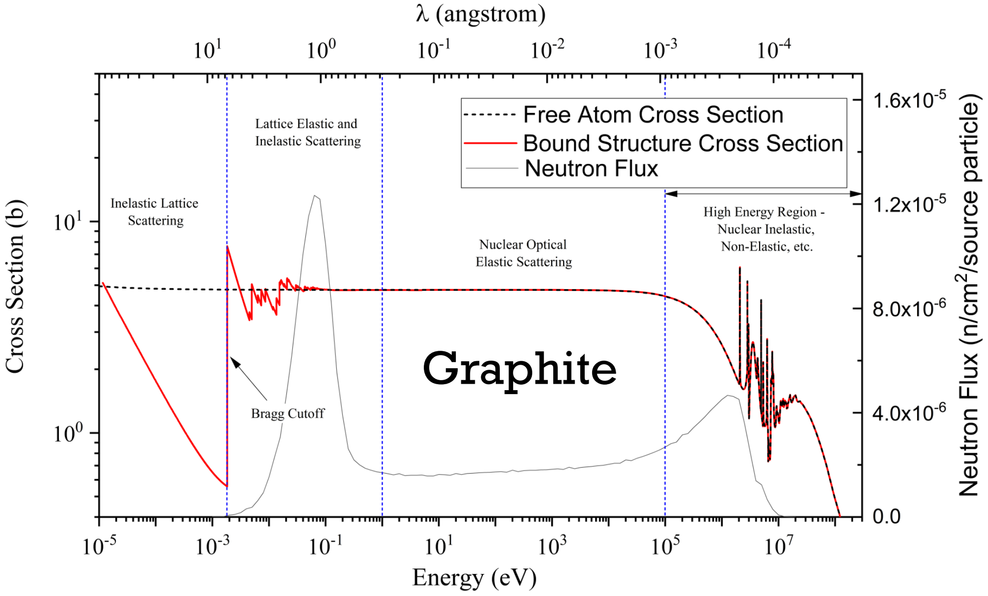
  

- Funding by the various US agencies including DOE-NE, NSF, NNSA
- Collaborations with colleagues at many universities, national laboratories, and industry

Thermal Nuclear Reactor





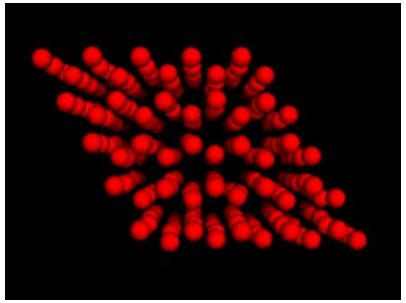
Fission

$$R_f = N\int \sigma_f(E)\phi(E)dE$$

Thermalization

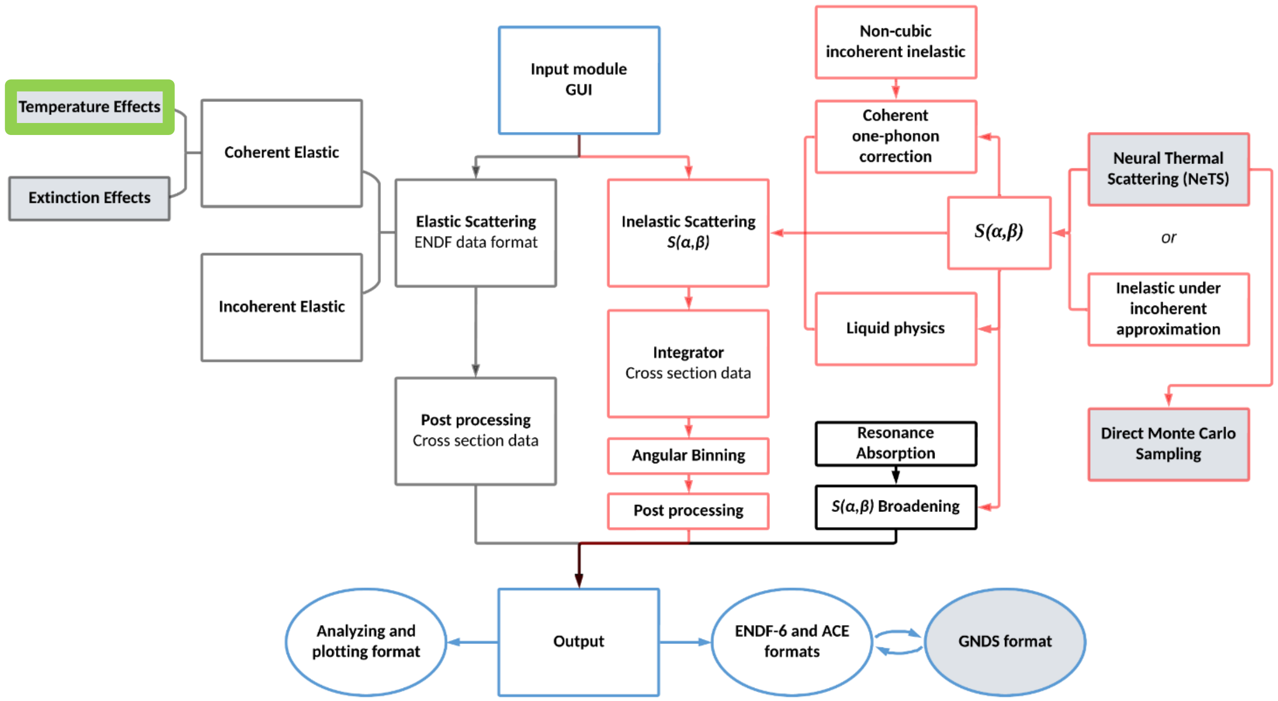
$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{1}{4\pi}\sqrt{\frac{E'}{E}}\left[\sigma_{coh}S(\alpha,\beta)+\sigma_{inc}S_s(\alpha,\beta)\right]$$

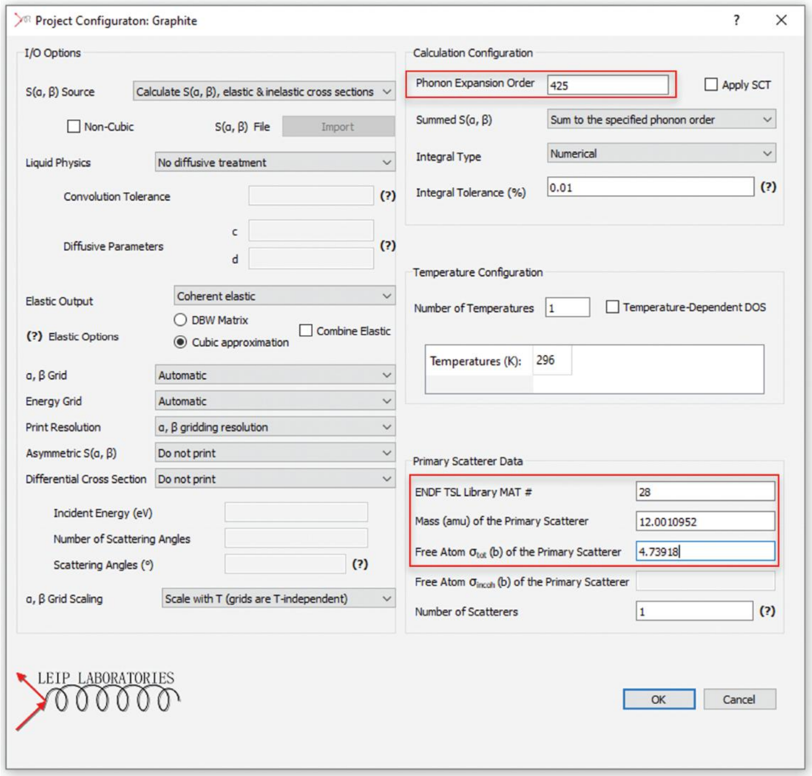
$$S(\alpha,\beta) = \frac{1}{k_B T}\frac{1}{2\pi\hbar}\int e^{i(\vec{\kappa}\cdot\vec{r}-\omega t)}G(\vec{r},t)d\vec{r}dt$$



Moderator microstructure dictates the physics, operations, and safety of the reactor

FLASSH





- Advanced features including
  - Temperature dependent phonon spectra
  - Coherent elastic scattering extinction effects
  - NeTS modules for key moderators
  - Improved liquid physics (addressing high viscosity liquids)
  - Enhanced Data formatting capabilities

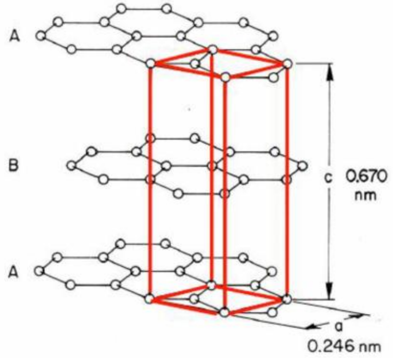
Graphite in ENDF/B-VIII.1

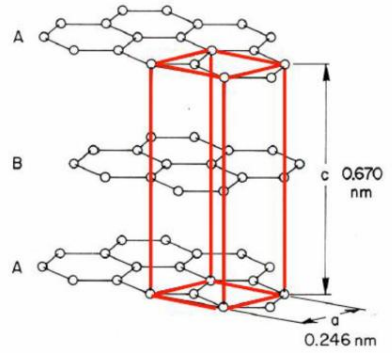
- ENDF/B-VIII.0 and ENDF/B-VIII.1 evaluations produced for different types of graphite crystalline and nuclear (i.e., porous)

  - **MT=2** block holds the coherent elastic scattering component

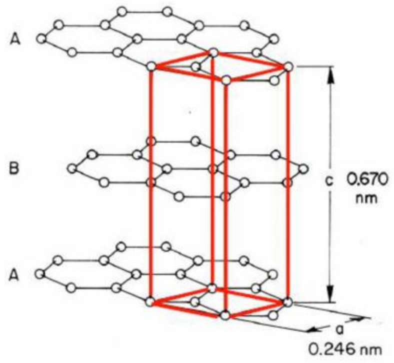
  - **MT=4** S(α,β) block accounts for either ideal crystalline or porous (nuclear) graphite through the use of the appropriate graphite phonon DOS

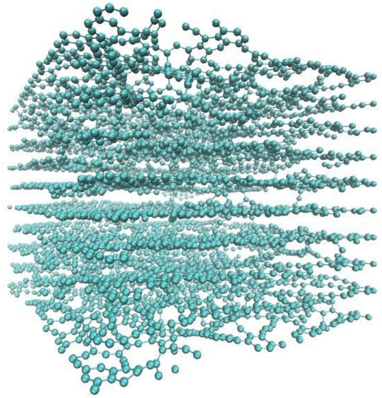
Crystalline





Nuclear





Graphite in ENDF/B-VIII.1

- Five (5) variations of graphite in ENDF/B-VIII.1
  - (all evaluated by LEIP group)

1. Crystalline graphite (incoherent approximation, 0% porosity)
2. Crystalline graphite (+Sd correction, 0% porosity)
3. Nuclear graphite (incoherent approximation, 10% porosity)
4. Nuclear graphite (incoherent approximation, 20% porosity)
5. Nuclear graphite (incoherent approximation, 30% porosity)

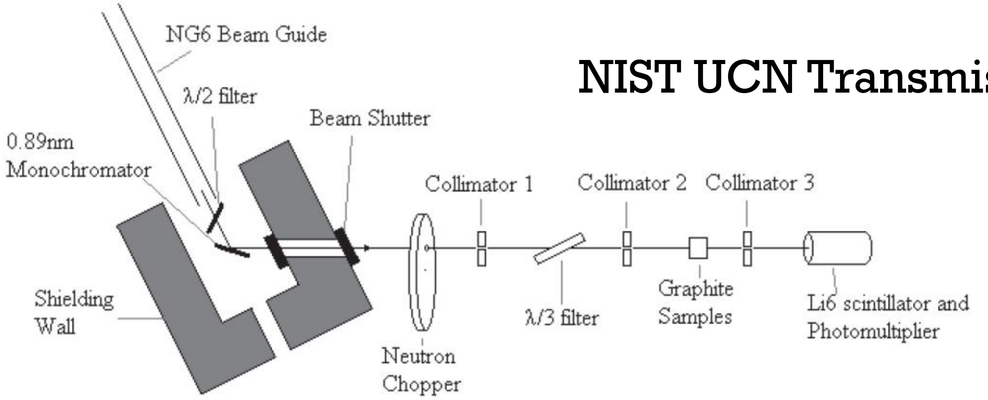
$$\mathbf{Porosity}(\%) = \left(\mathbf{1} - \frac{\boldsymbol{\rho_{nuclear\ graphite}}}{\boldsymbol{\rho_{crystalline\ graphite}}}\right) \times \mathbf{100\%}$$

Density (g/cm^3)	Library
2.26	0% porous
1.9-2.0	10% porous
1.7-1.9	20% porous
< 1.7	30% porous

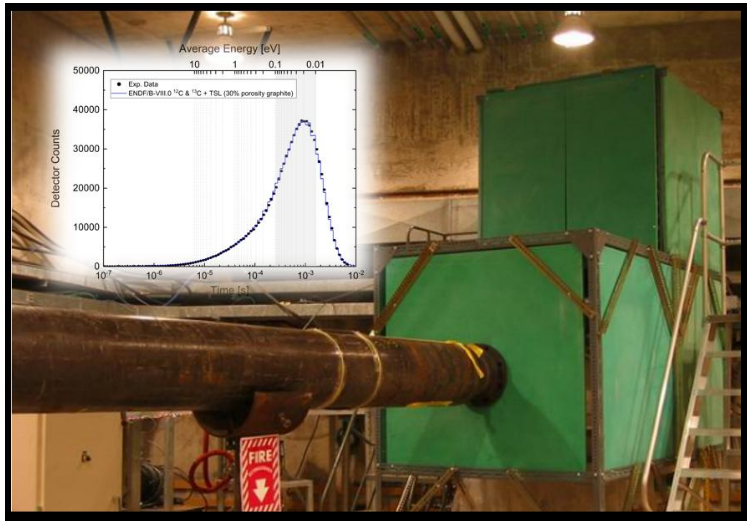
TSL Validation Approach

- Experimental **HOLISTIC** approach

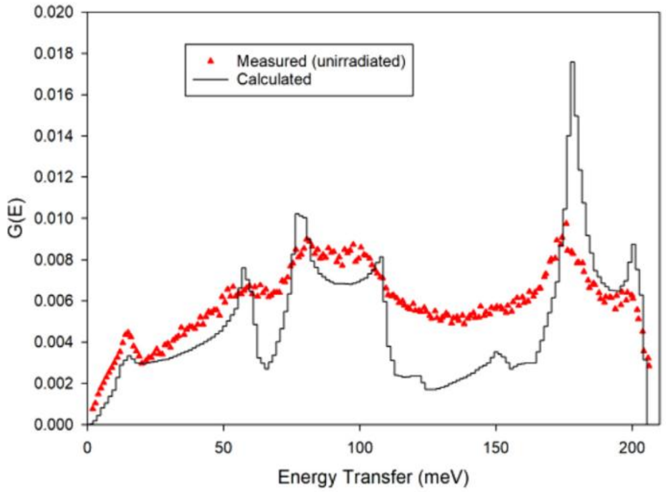
NIST UCN Transmission



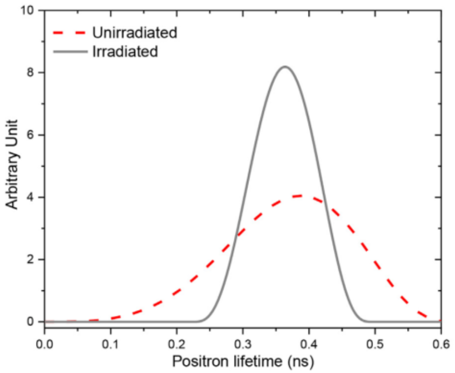
ORELA Benchmark

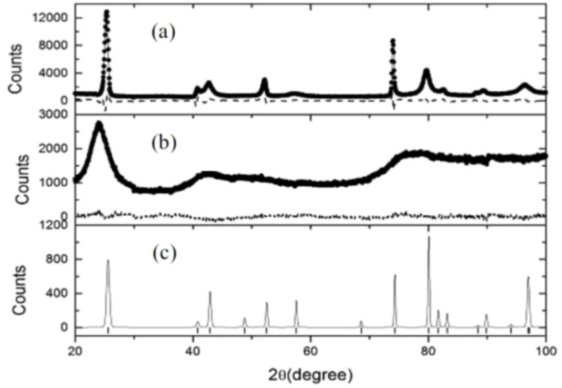


INS SNS



PAS and NPD





TSL Validation Approach

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On a measurement approach to support evaluation of thermal scattering law data

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A R T I C L E I N F O

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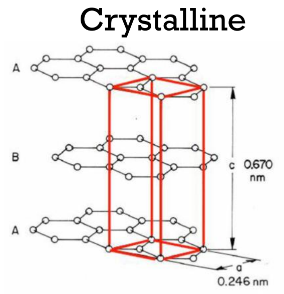
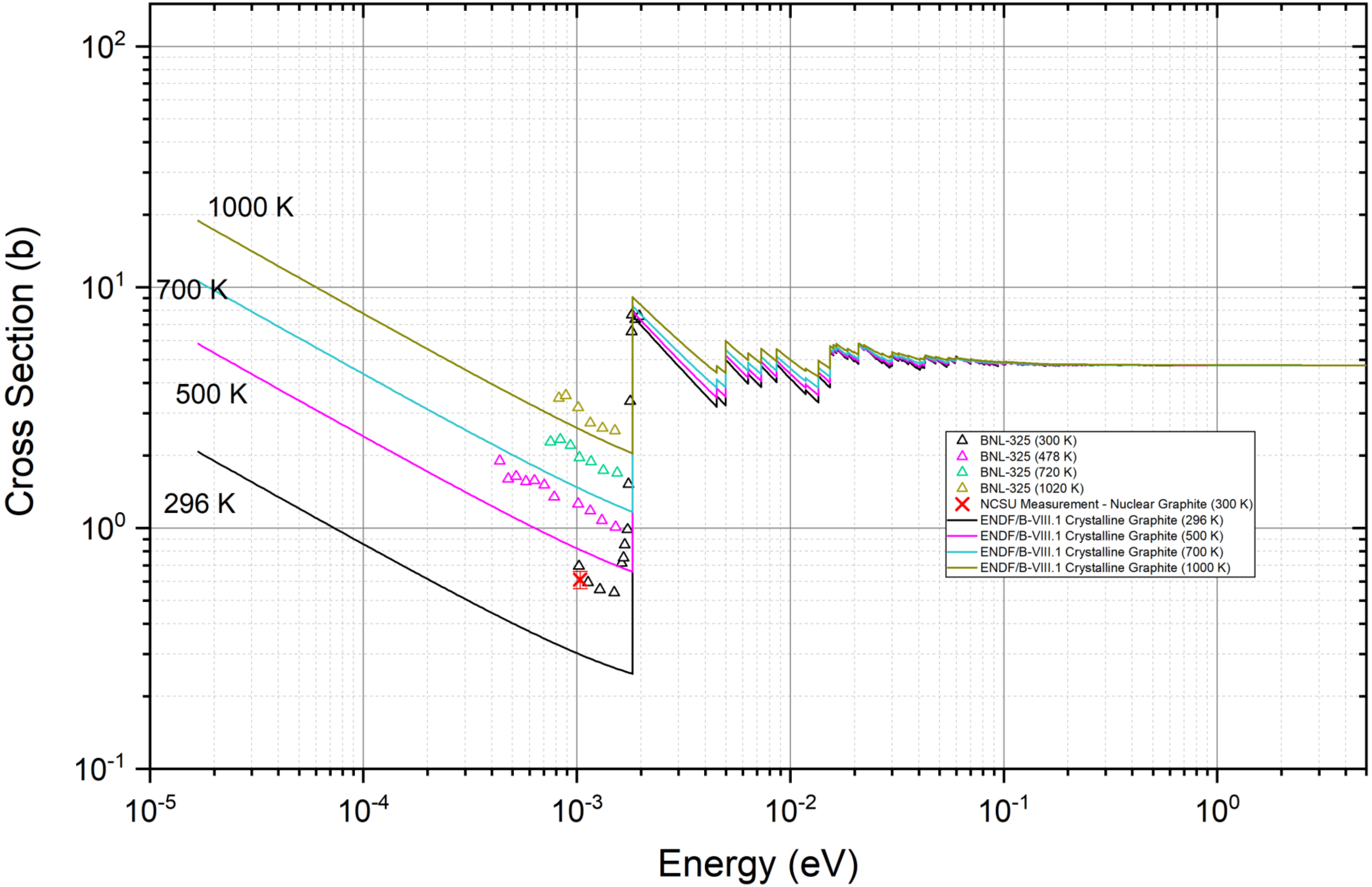
Cross section
Measurement
ENDF

A B S T R A C T

Inelastic thermal neutron scattering in materials that act as neutron moderators, reflectors, and filters results in shaping the neutron spectrum at low energies. This phenomenon is described using differential scattering cross sections calculated from three components including the bound atom (i.e., nuclear) scattering cross section of the neutron, the ratio of the outgoing and incoming neutron energy, and the thermal scattering law (TSL), i.e., $S(\alpha,\beta)$, where α and β represent dimensionless momentum and energy exchange variables, respectively. To date, no TSL libraries are generated using measured data. However, valuable information may be derived from measurements and "targeted" experiments that can validate TSL data and the related inelastic scattering cross sections. As a demonstration, a suite of coordinated measurements and experiments is described that was designed and used to support the evaluation of the TSL for "nuclear" graphite. This experimental suite includes neutron powder diffraction (for structure analysis), positron annihilation (for nano porosity assessment), inelastic neutron scattering measurements using a chopper spectrometer, transmission experiments using neutrons with energy below the Bragg cutoff thereby accessing the total (inelastic) cross section, and a slowing-down-time experiment to observe the developing neutron spectrum in the material. This experimental suite was key to understanding the difference in TSL between "nuclear" and "ideal" graphite and for the inclusion of "nuclear" graphite in the ENDF/B-VIII.0 nuclear data library release.

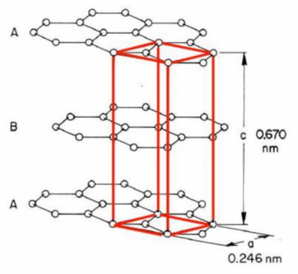
Transmission Measurement

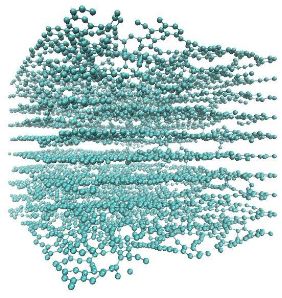


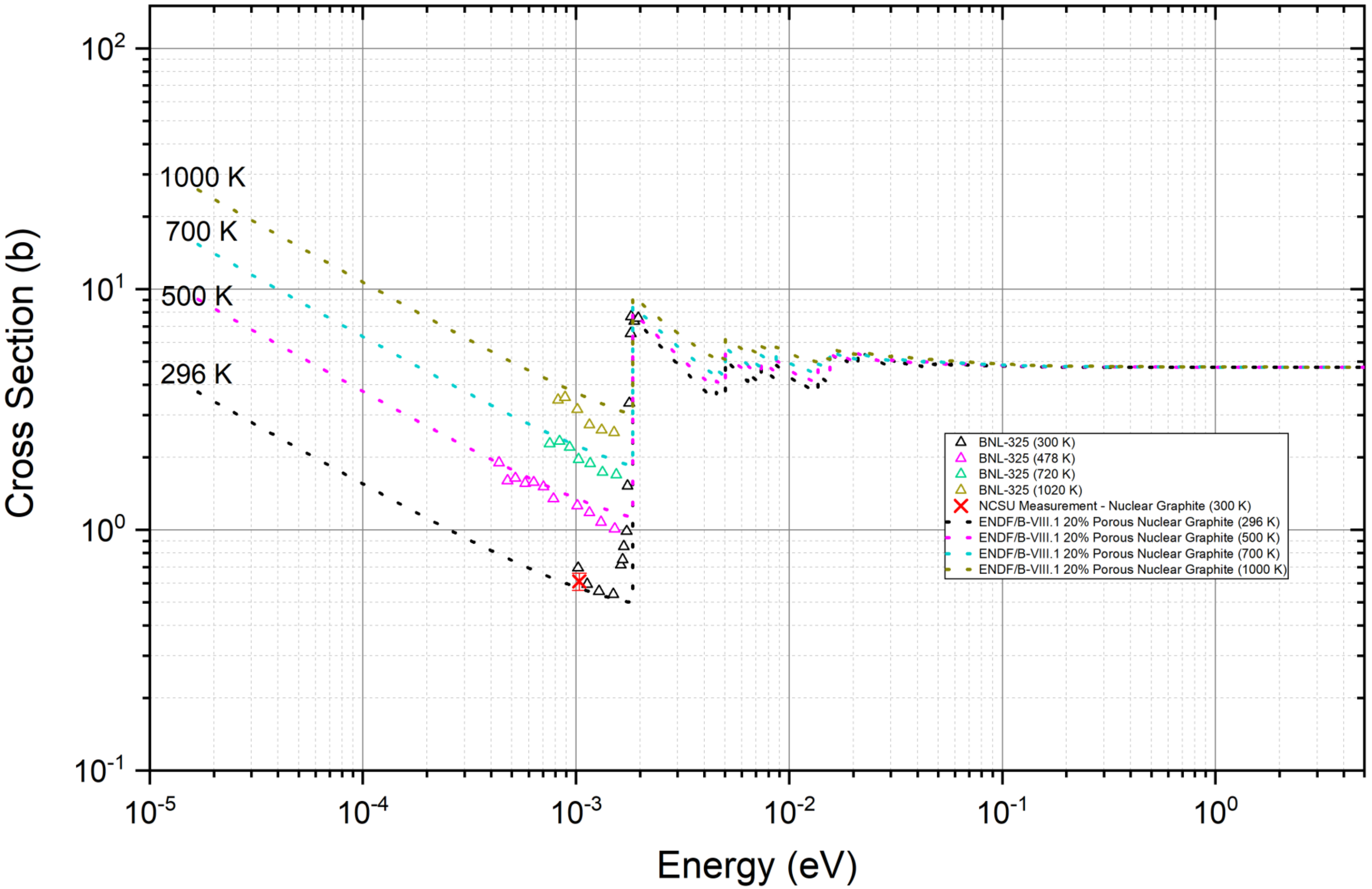


Transmission Measurement

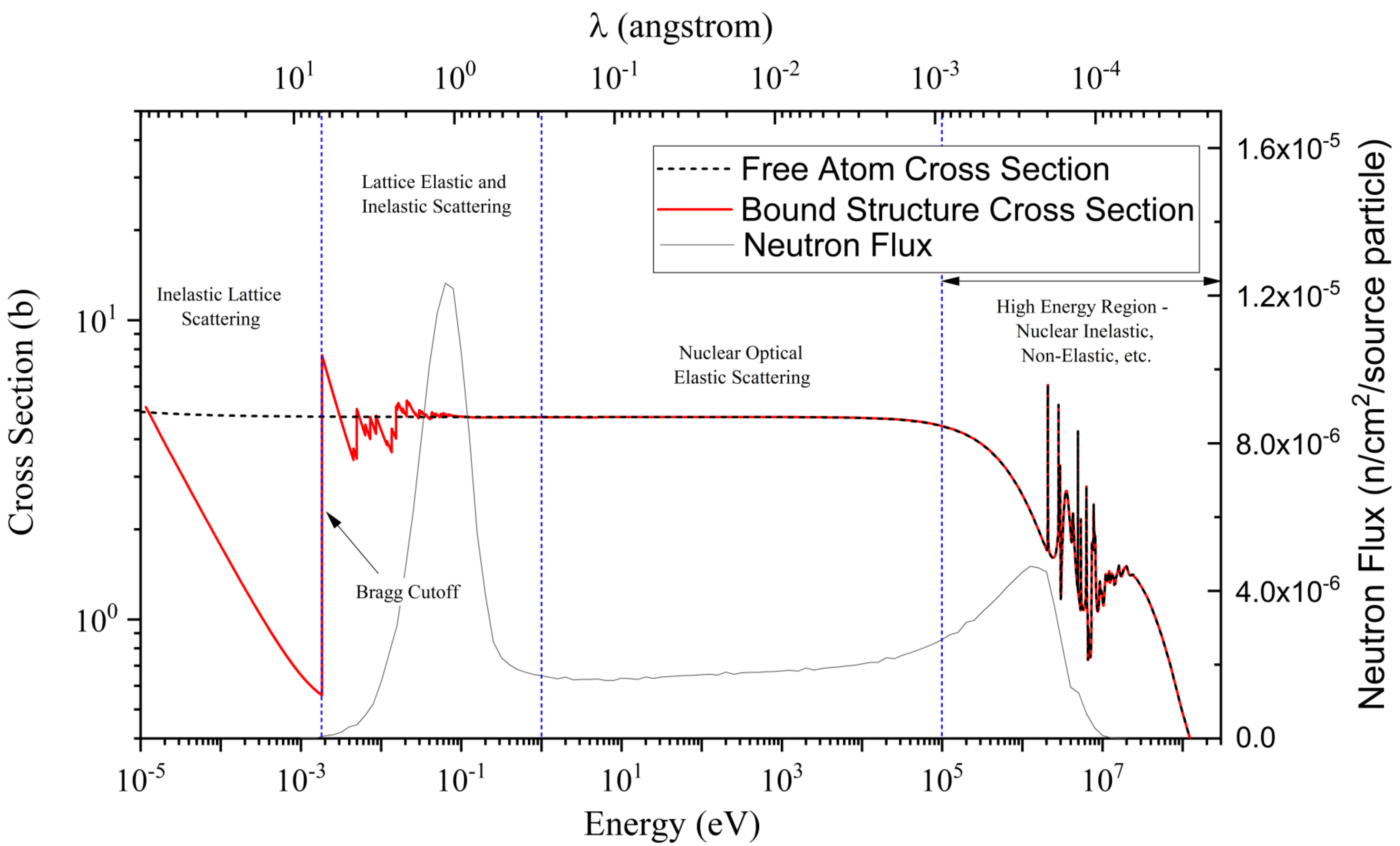
Nuclear

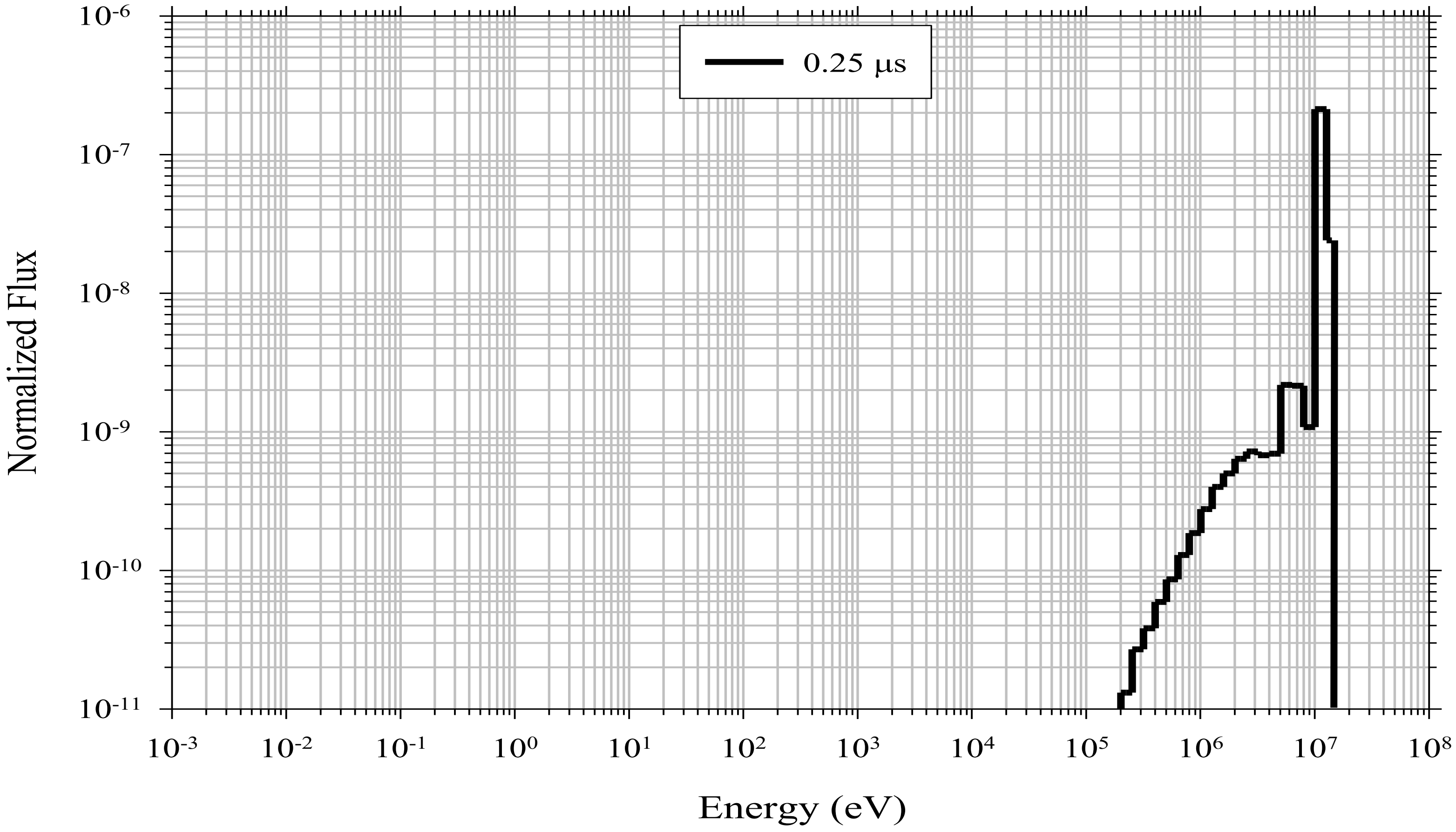




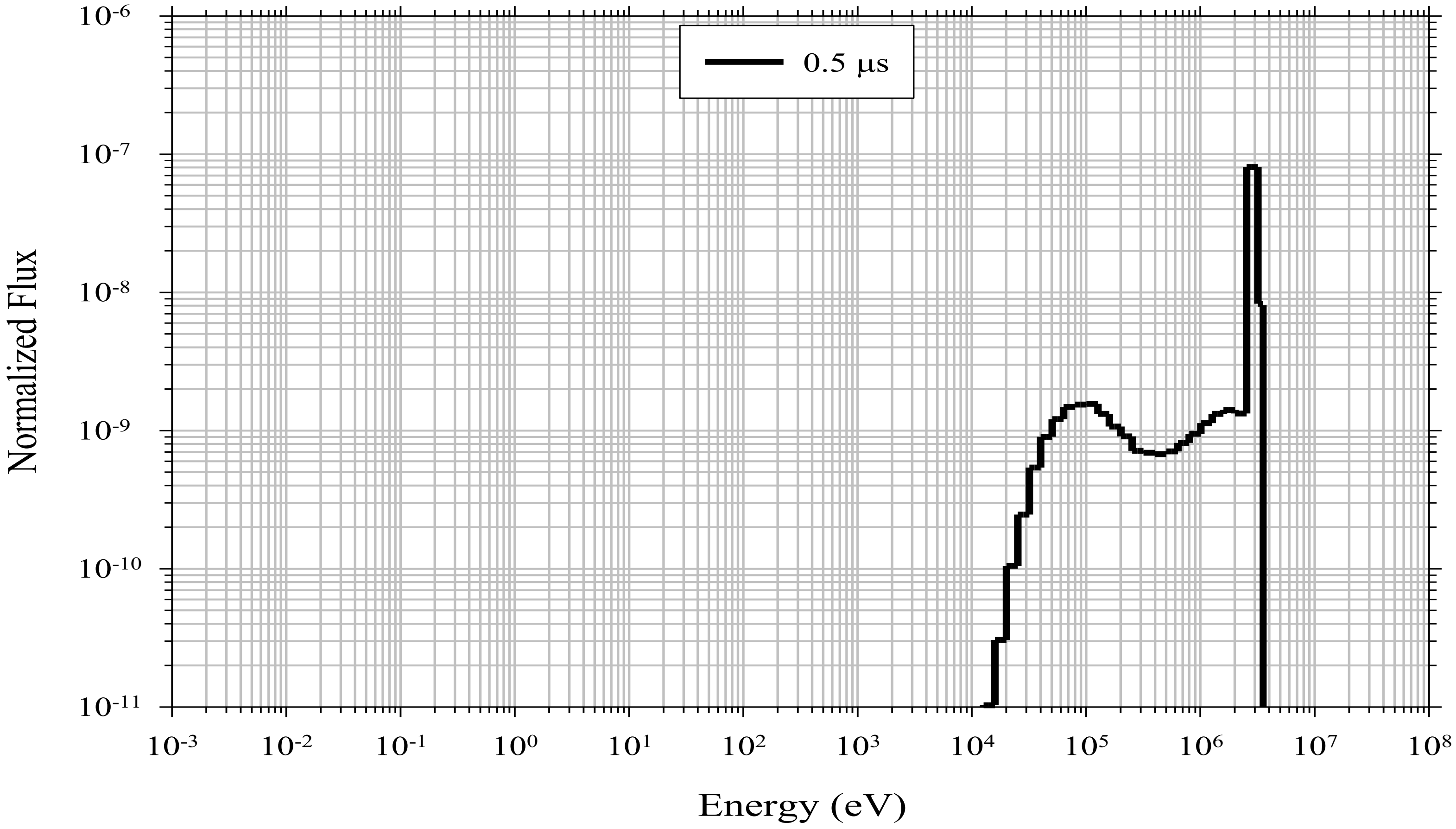


Slowing Down in Graphite





0.25 μs
Normalized Flux
10-6
10-7
10-8
10-9
10-10
10-11
10-3
10-2
10-1
100
101
102
103
104
105
106
107
108
Energy (eV)



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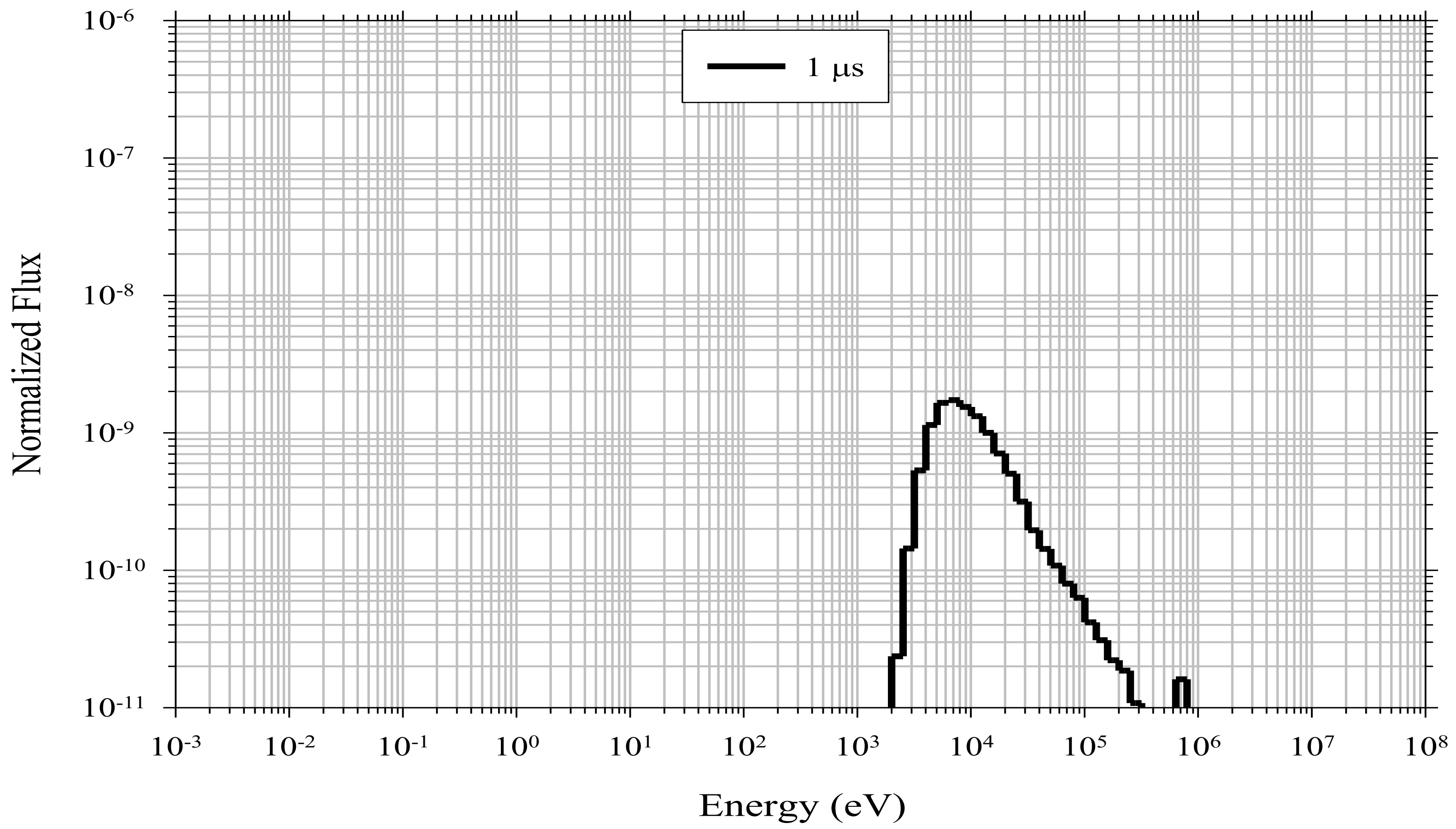
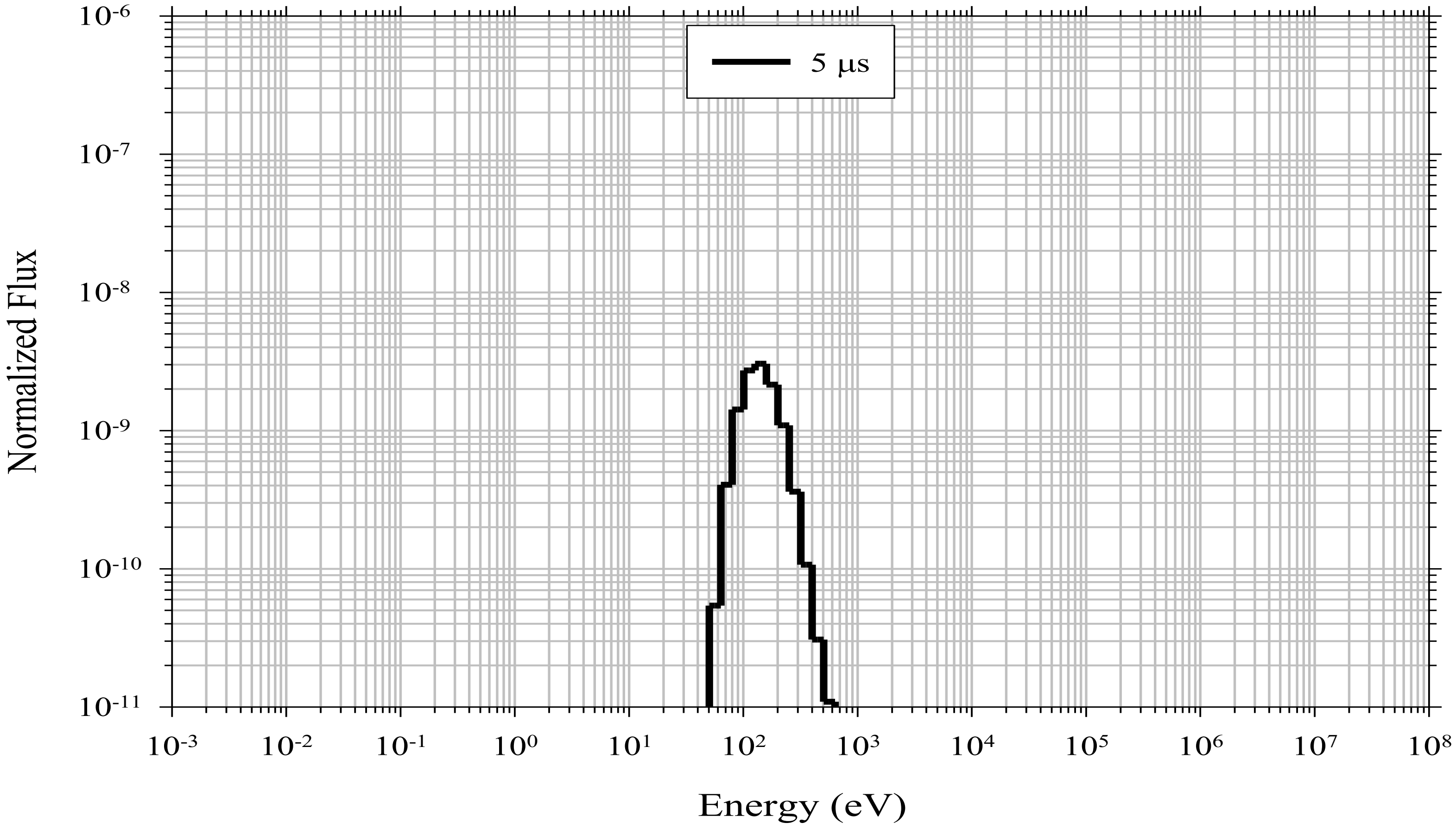
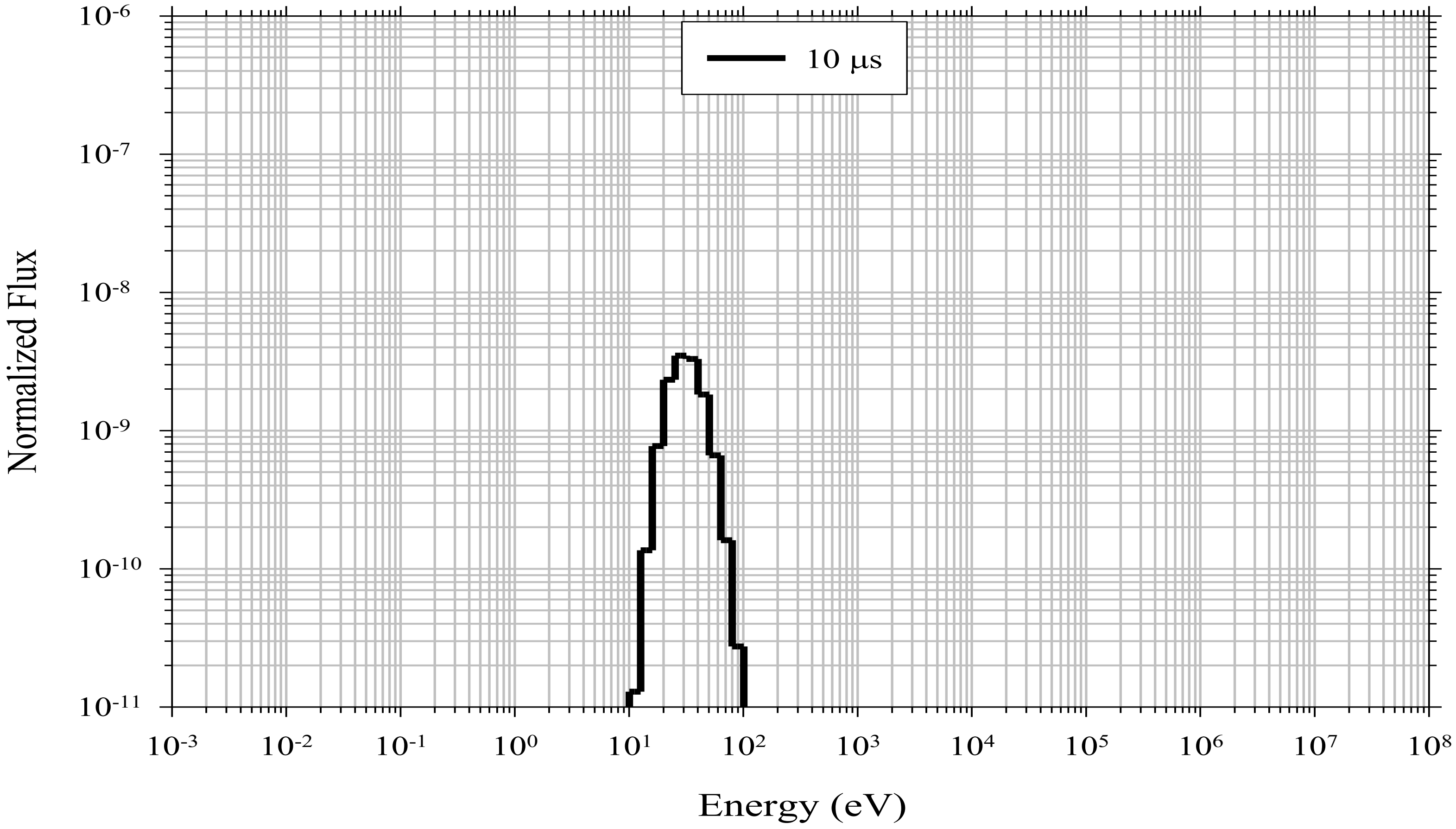
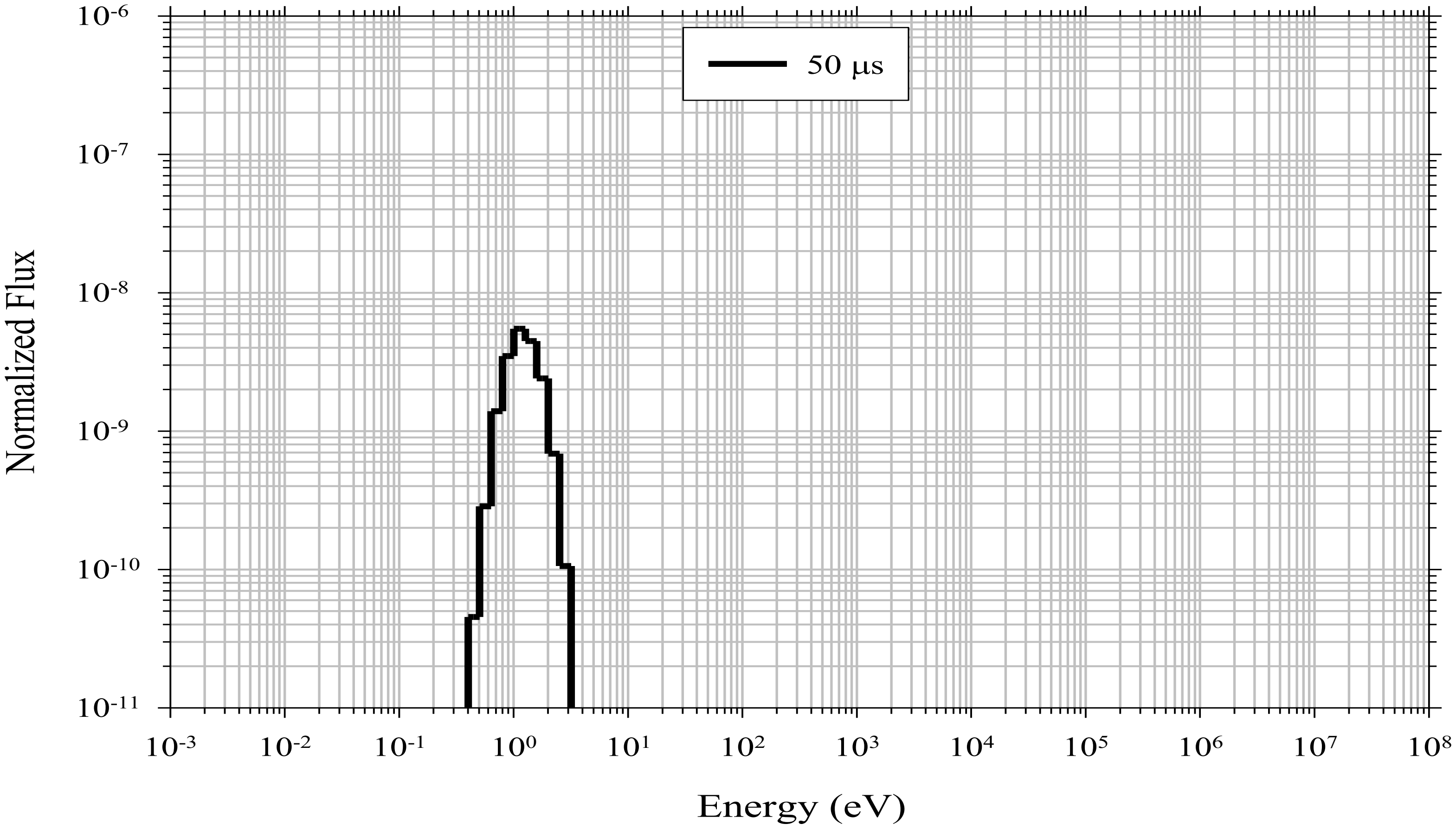

1 μs
Normalized Flux
Energy (eV)
10-6
10-7
10-8
10-9
10-10
10-11
10-3
10-2
10-1
100
101
102
103
104
105
106
107
108

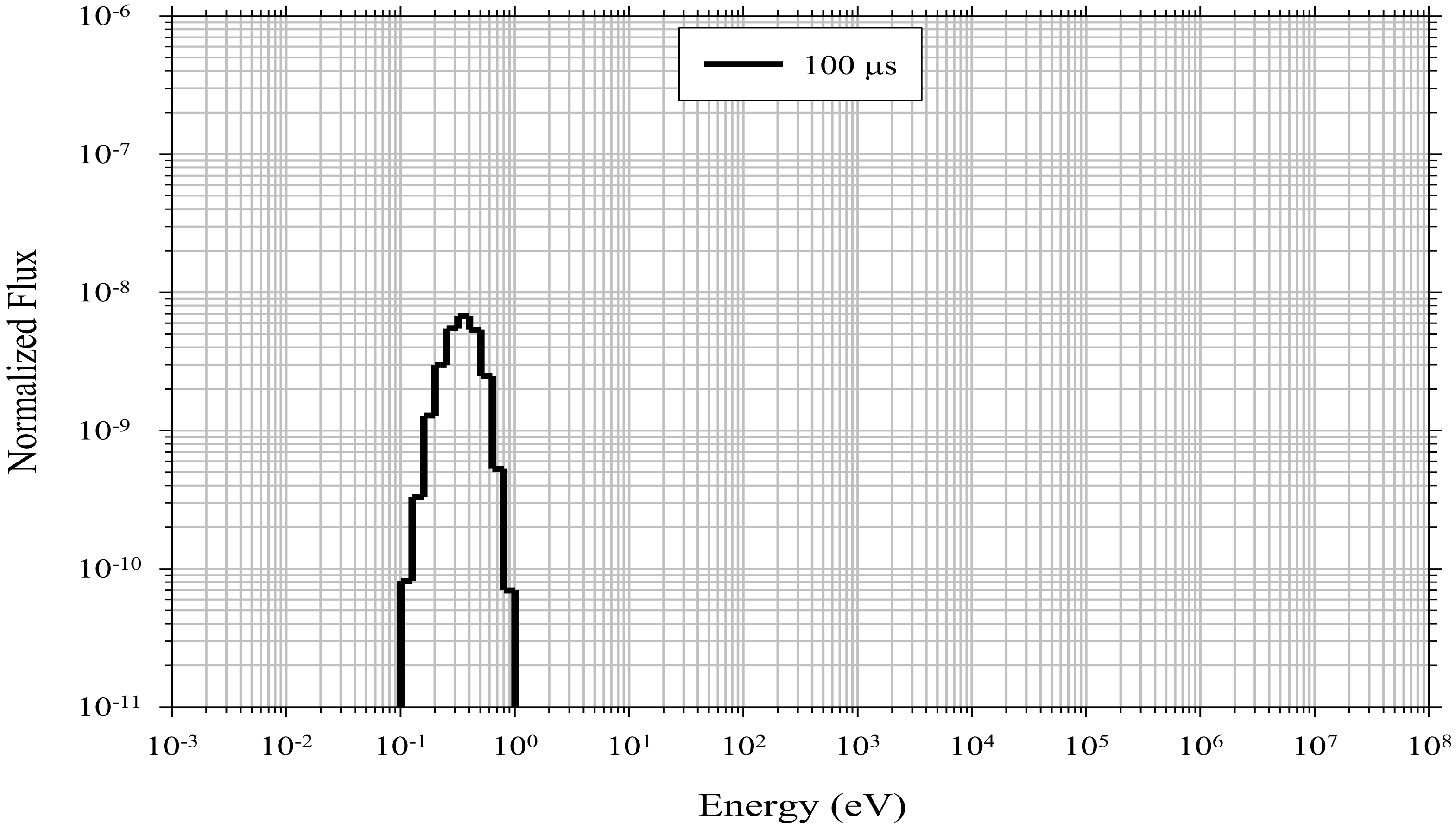

5 μs
Normalized Flux
10-6
10-7
10-8
10-9
10-10
10-11
10-3
10-2
10-1
100
101
102
103
104
105
106
107
108
Energy (eV)



10 μs
Normalized Flux
Energy (eV)
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-3} 10^{-2} 10^{-1} 10^{0} 10^{1} 10^{2} 10^{3} 10^{4} 10^{5} 10^{6} 10^{7} 10^{8}



50 μs
Normalized Flux
10-6
10-7
10-8
10-9
10-10
10-11
10-3
10-2
10-1
100
101
102
103
104
105
106
107
108
Energy (eV)



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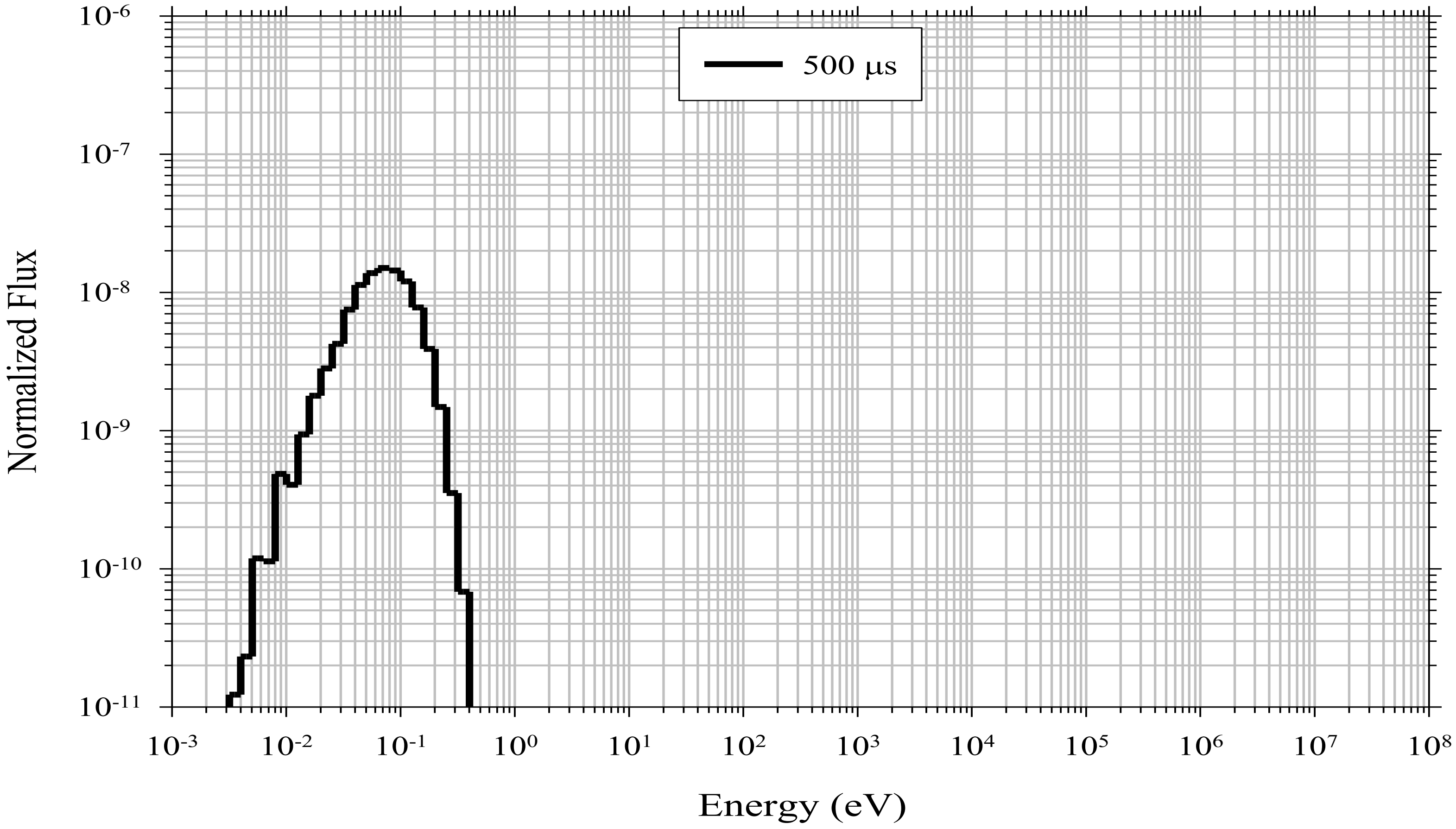

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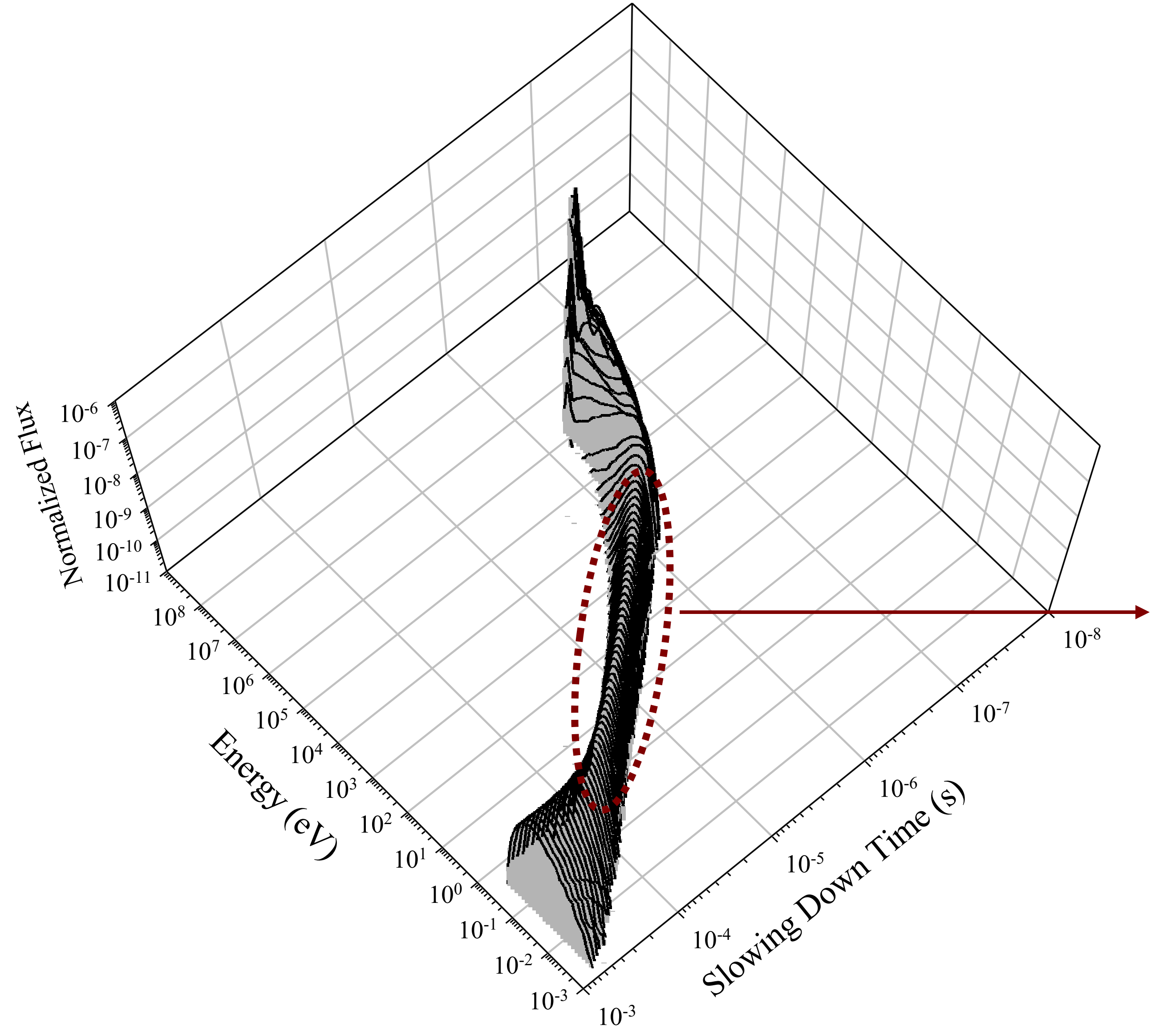

$$\overline{E} \approx \frac{k}{t^2}$$

FUND-ORELA-ACC-GRAPH-PNSDT-001

NEA/NSC/DOC(95)03

FUND-ORELA-ACC-GRAPH-PNSDT-001

Benchmark of Neutron Thermalization in Graphite Using the Slowing-Down-Time ORELA Experiment

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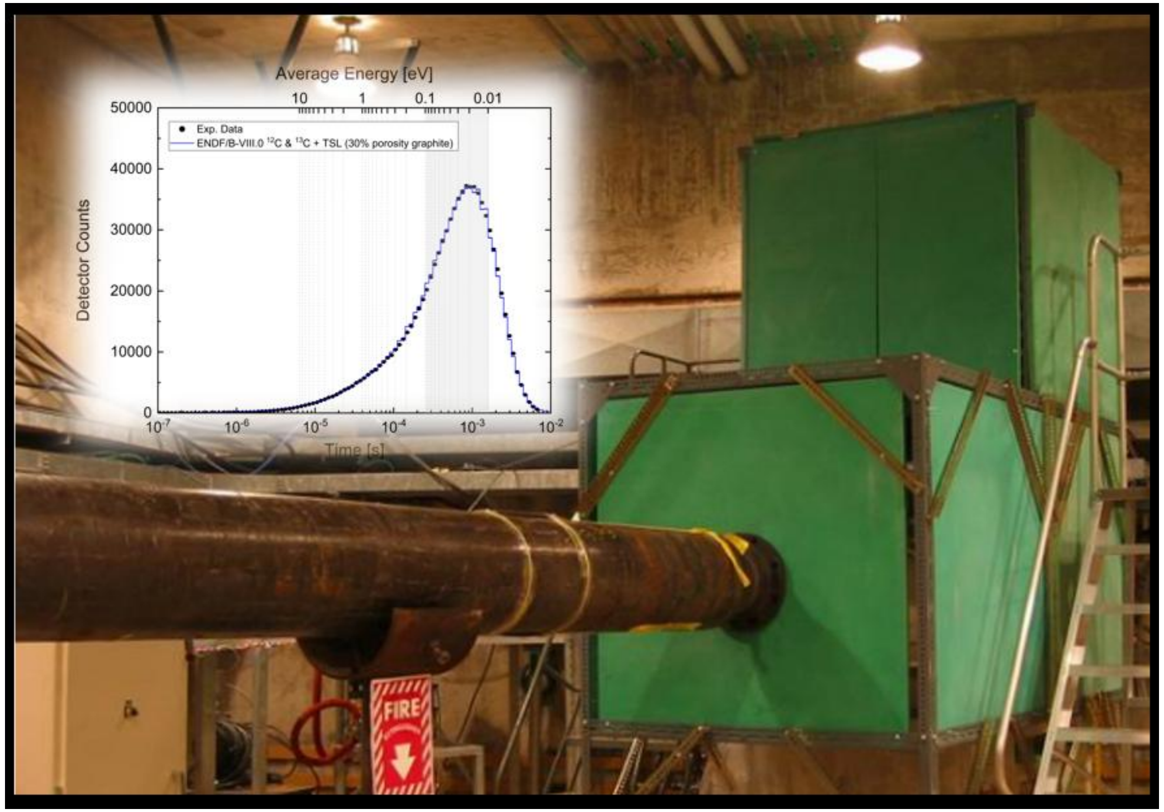
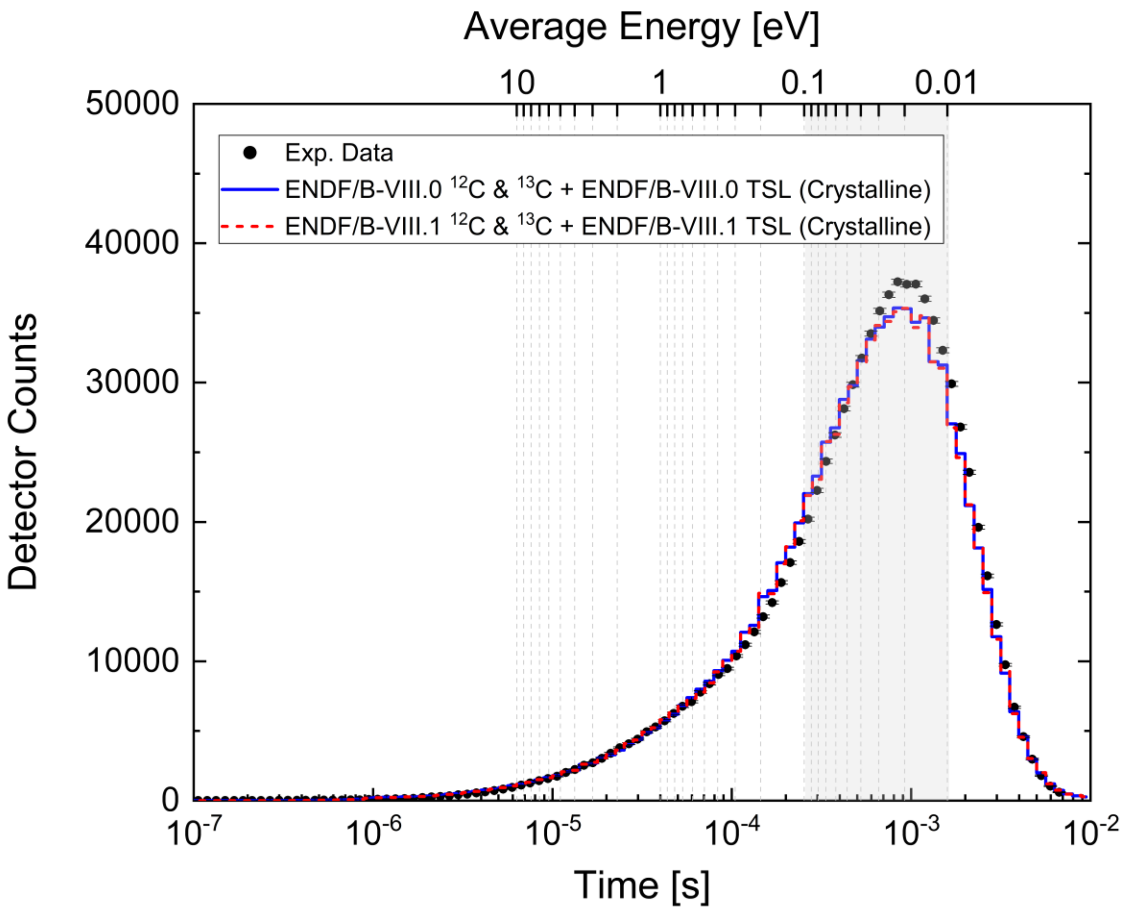


FUND-ORELA-ACC-GRAPH-PNSDT-001

- ENDF/B-VIII.0 comparison with ENDF/B-VIII.1

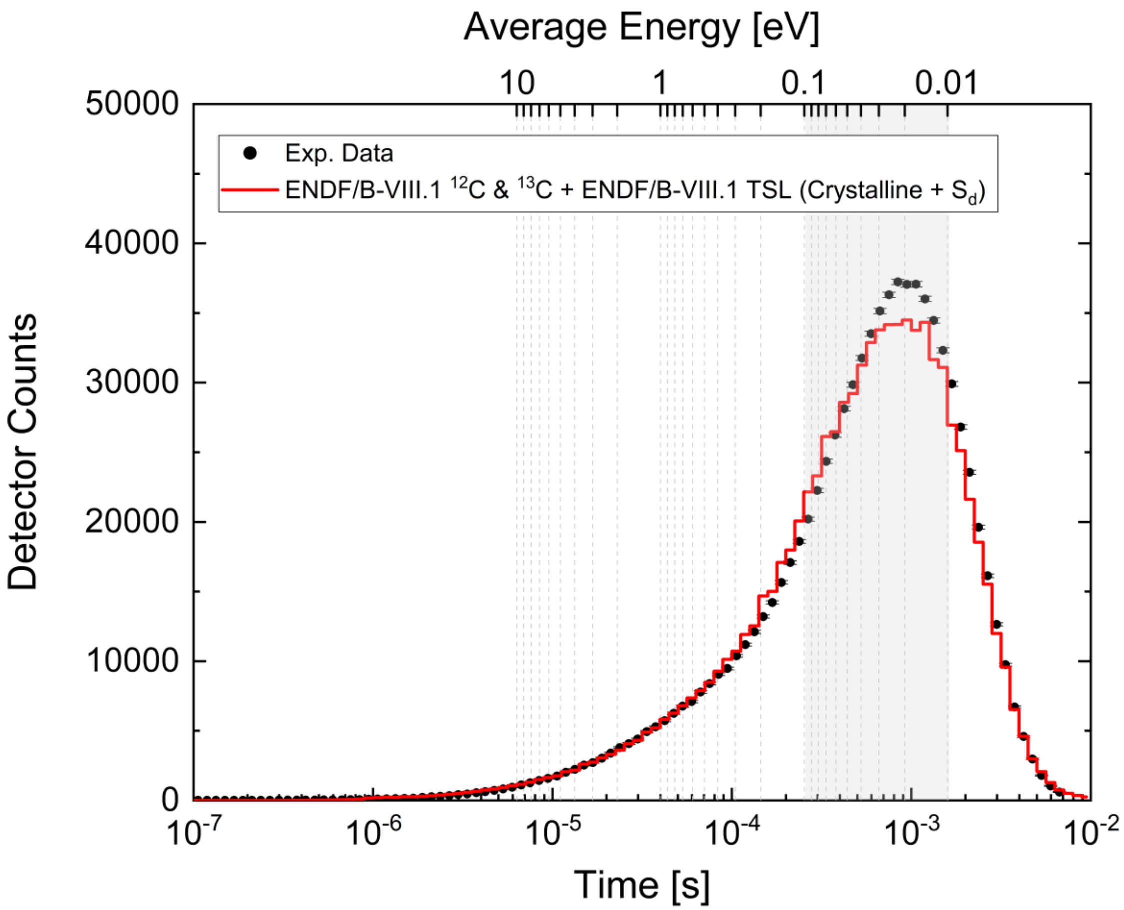


Density = 1.66 g/cm^3
Porosity ≈ 30%

Cross Sections	Mean Absolute Deviation (%)
ENDF/B-VIII.0 + Cry	4.14%
ENDF/B-VIII.1 + Cry	4.09%

FUND-ORELA-ACC-GRAPH-PNSDT-001

- ENDF/B-VIII.1 Crystalline Graphite + S_d does not capture the thermalization effects of nuclear graphite (nor was it intended to)



Density = 1.66 g/cm^3

Porosity ≈ 30%

Cross Sections	Mean Absolute Deviation (%)
ENDF/B-VIII.0 + Cry	4.14%
ENDF/B-VIII.1 + Cry	4.09%
ENDF/B-VIII.1 + Cry +S_d	5.01%

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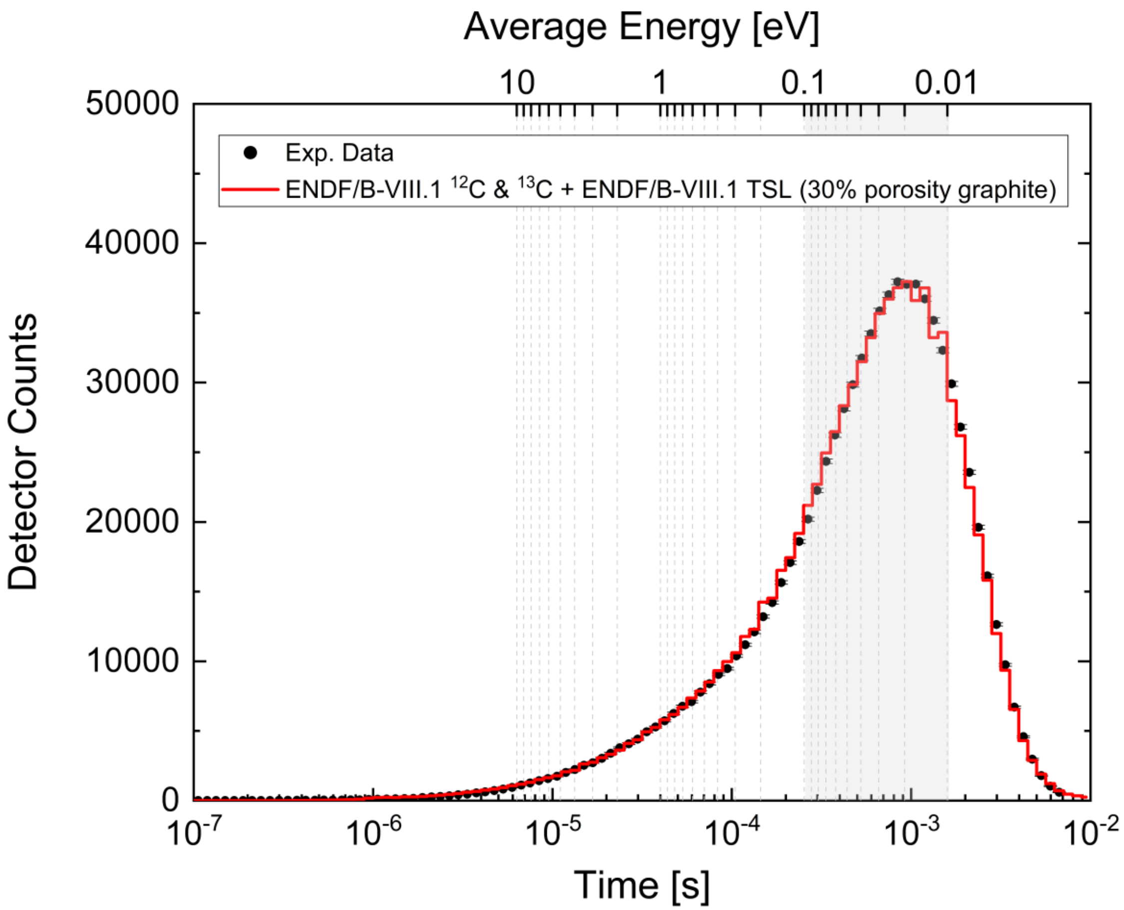
FUND-ORELA-ACC-GRAPH-PNSDT-001

- ENDF/B-VIII.1 Nuclear Graphite 30% Porous consistent with benchmark



Density = 1.66 g/cm^3
Porosity ≈ 30%

Cross Sections	Mean Absolute Deviation (%)
ENDF/B-VIII.0 + Cry	4.14%
ENDF/B-VIII.1 + Cry	4.09%
ENDF/B-VIII.1 + Cry +S_d	5.01%
ENDF/B-VIII.1+30%	1.79%

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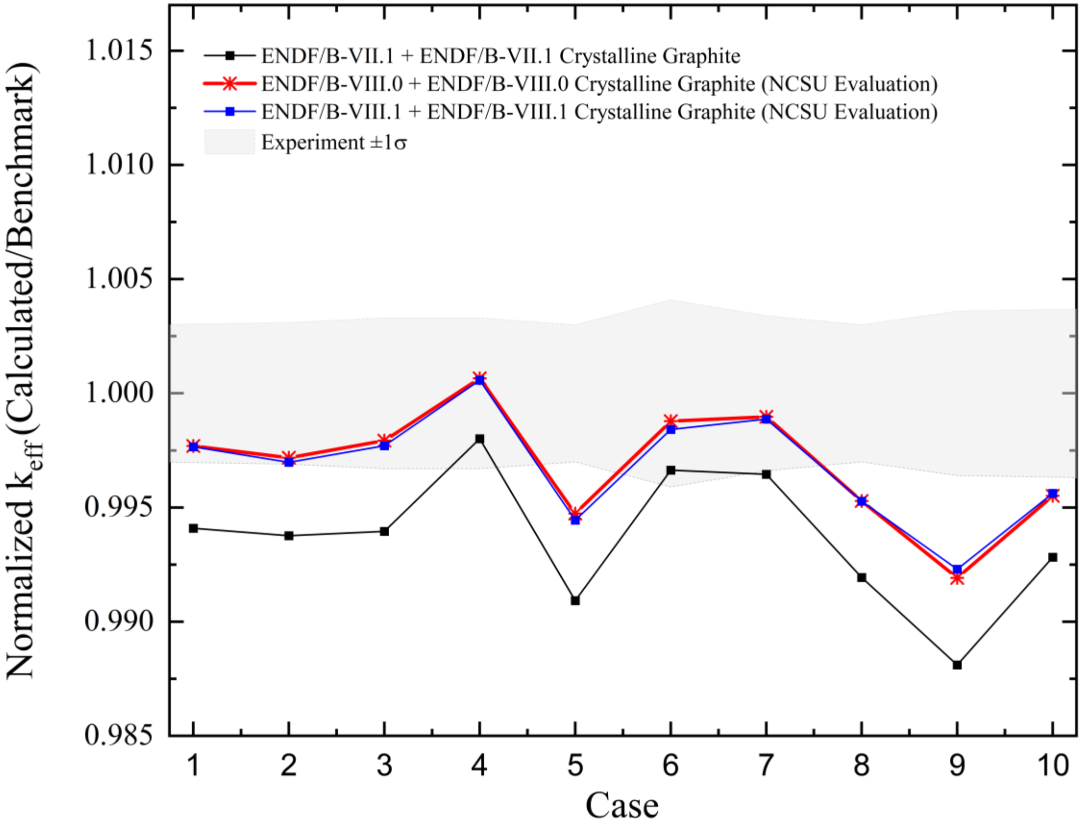
PROTEUS-GCR-EXP-001 to -004

- ENDF/B-VII.1 Crystalline Graphite vs. ENDF/B-VIII.1 Crystalline Graphite

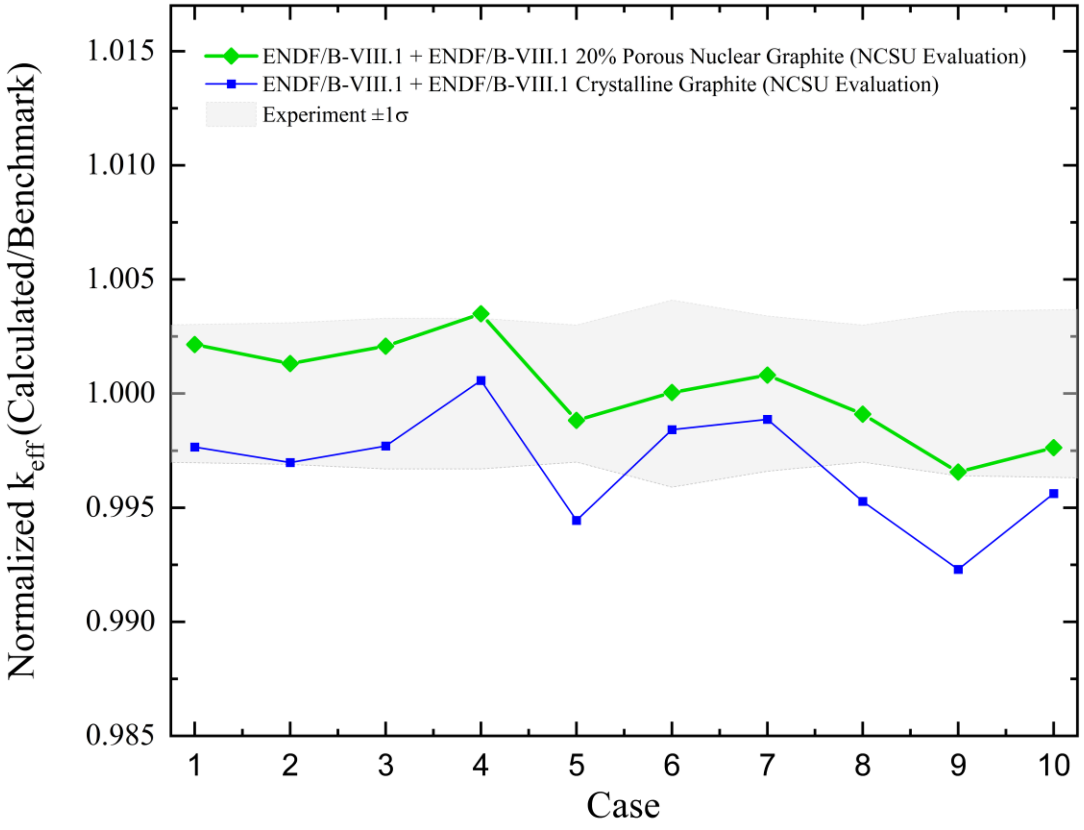


Density = 1.7 g/cm^3
Porosity ≈ 20-30%

Case	C-E (pcm) ENDF/B-VII.1 Crystalline	C-E (pcm) ENDF/B-VIII.0 Crystalline	C-E (pcm) ENDF/B-VIII.1 Crystalline
1	594	232	235
2	626	283	303
3	606	208	230
4	199	66	57
5	910	528	707
6	337	122	258
7	355	103	83
8	809	473	344
9	1193	811	782
10	719	450	529

PROPEUS-GCR-EXP-001 to -004

- ENDF/B-VIII.1 Crystalline Graphite vs. 20% Porous Nuclear Graphite



C-E (pcm)

Case	ENDF/B-VIII.1 Crystalline	ENDF/B-VIII.1 20% Porous
1	235	204
2	303	137
3	230	188
4	57	322
5	707	146
6	258	12
7	83	91
8	344	95
9	782	385
10	529	247
mean	350	183
χ^2	20	**5**

VHTRC-GCR-EXP-001

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ARTICLE

Check for updates

A pseudo-material method for graphite with arbitrary porosities in Monte Carlo criticality calculations

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ABSTRACT
The latest ENDF/B-VIII library adapted new porosity-dependent cross-section data of graphite. However, the porosity of the actual graphite does not necessarily correspond to the porosity given in the data. We have proposed a method to perform neutronic calculations at the desired porosity on the basis of the pseudo-material method. We have also compared the k_{eff} values calculated by the pseudo-material method with the experimental values for the VHTRC. In addition, we have investigated the temperature dependance of the calculation values obtained by this method. From these results, we have concluded that this method allows us to perform the neutronic calculations in which we can reflect detailed information on the porosity of graphite.

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KEYWORDS
ENDF/b-viii; graphite; porosity-dependent cross-section data; vhtrc; pseudo-material method

1. Introduction

In recent years, the high temperature gas-cooled reactor (HTGR) has attracted much interest as one of the Generation IV nuclear reactor systems. One of the notable features of the HTGR is that graphite is used not only as a moderator but also as a structural material owing to its excellent thermal and mechanical properties under the high temperature environment. A large amount of graphite is placed in the HTGR core so that the neutrons are sufficiently moderated. Therefore, the accuracy of neutronic calculation of the HTGR is highly dependent on the nuclear data of graphite. Especially, the thermal scattering law (TSL) data of graphite, which impacts the neutron energy spectrum, is fundamental to the detailed design and core analysis of the HTGR. In JENDL-4.0 [1], the TSL data of graphite is evaluated on the basis of the traditional evaluation model of Young and Koppel [2]. On the other hand, the latest ENDF/B-VIII.0 library adopted new TSL data of graphite [3]. It has the TSL data that depends on the porosities of graphite: crystalline graphite (i.e. graphite with a porosity of 0%), reactor graphite with a porosity of 10% and reactor graphite with a porosity of 30%. This is expected to result in more accurate neutronic calculations than before.

However, the actual graphite does not necessarily correspond to these porosities in the new TSL data. For instance, the density and porosity for some major graphite are shown in Table 1 [4–7], where the theoretical density of graphite is 2.25 g/cm^3 [8]. These values are neither 10% nor 30%. Currently, we can only handle neutronic calculations for three porosities of 0%, 10%, and 30%. In neutronic calculations for the cores containing graphite with other than the porosities mentioned above, the nearest TSL data will be selected for use. From the viewpoint of HTGR designers, it is desirable to be able to perform neutronic calculations using graphite with any porosity. Such calculations can eliminate the uncertainties associated with the difference between the porosity of actual graphite and the porosity given in the calculation input.

The purpose of this study is to propose a practical method for performing accurate neutronic calculations reflecting detailed information on the porosity of graphite. The proposed method allows us to perform neutronic calculations at any porosities between 0% and 30%. Using the Very High Temperature Reactor Critical Assembly (VHTRC) [9], criticality calculation results based on the proposed method are also described making comparisons with experimental results.

2. Methodology

The pseudo-material method is applied to a material containing graphite to perform neutronic calculations with the specified porosity. The pseudo-material method was originally developed by Conlin et al. in order to reduce the cross-section data for the huge temperature points [10]. In this method, a pseudo-material is defined such that a material at a certain

CONTACT Shoichiro Okita, okita.shoichiro@jaea.go.jp, Sector of Fast Reactor and Advanced Reactor Research and Development, Japan Atomic Energy Agency, Ibaraki, Japan.

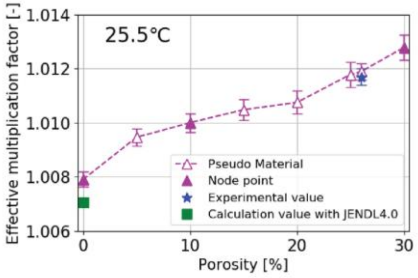

Figure 6. Calculation results of the k_{eff} values at 25.5 ℃ using graphite with each porosity.

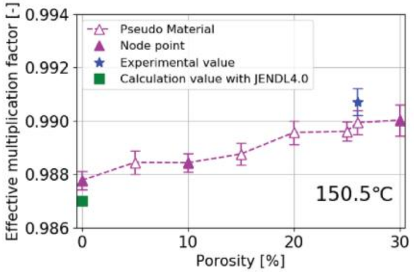

Figure 9. Calculation results of the k_{eff} values at 150.5 ℃ using graphite with each porosity.

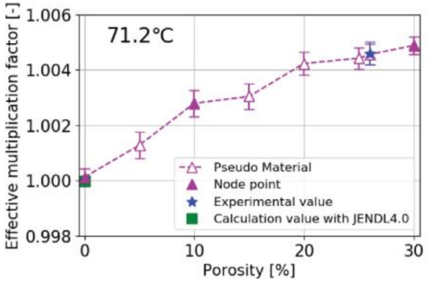

Figure 7. Calculation results of the k_{eff} values at 71.2 ℃ using graphite with each porosity.

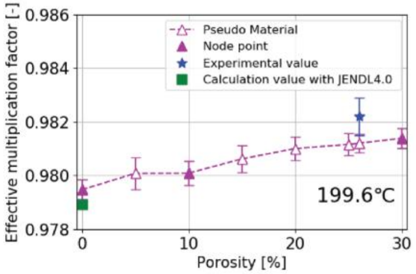

Figure 10. Calculation results of the k_{eff} values at 199.6℃ using graphite with each porosity.

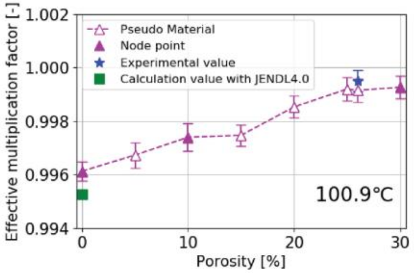

Figure 8. Calculation results of the k_{eff} values at 100.9 ℃ using graphite with each porosity.

MVP-3, JENDL4.0
Crystalline (0% porosity)
Graphite TSL

Temp.	Benchmark	C/E (pcm)
25.5°C	1.0115 ± 0.0032	-444
71.2°C	1.0046 ± 0.0033	-462
100.9°C	0.9994 ± 0.0035	-413
150.5°C	0.9906 ± 0.0035	-360
199.6°C	0.9820 ± 0.0037	-307

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VHTRC-GCR-EXP-001 - Reproduced

Original (MVP-3)

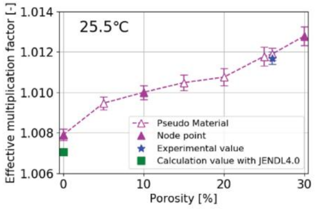


Figure 6. Calculation results of the k_{eff} values at 25.5 ℃ using graphite with each porosity.

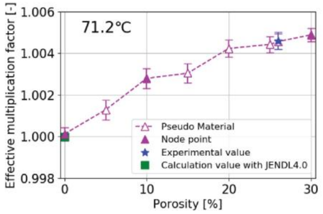


Figure 7. Calculation results of the k_{eff} values at 71.2 ℃ using graphite with each porosity.

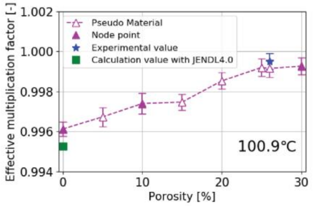


Figure 8. Calculation results of the k_{eff} values at 100.9 ℃ using graphite with each porosity.

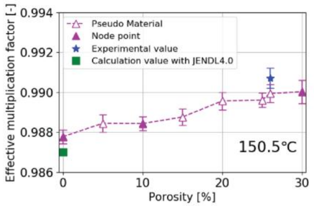


Figure 9. Calculation results of the k_{eff} values at 150.5 ℃ using graphite with each porosity.

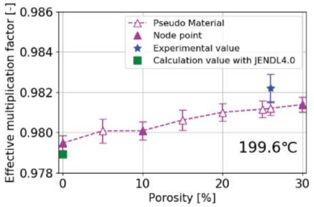
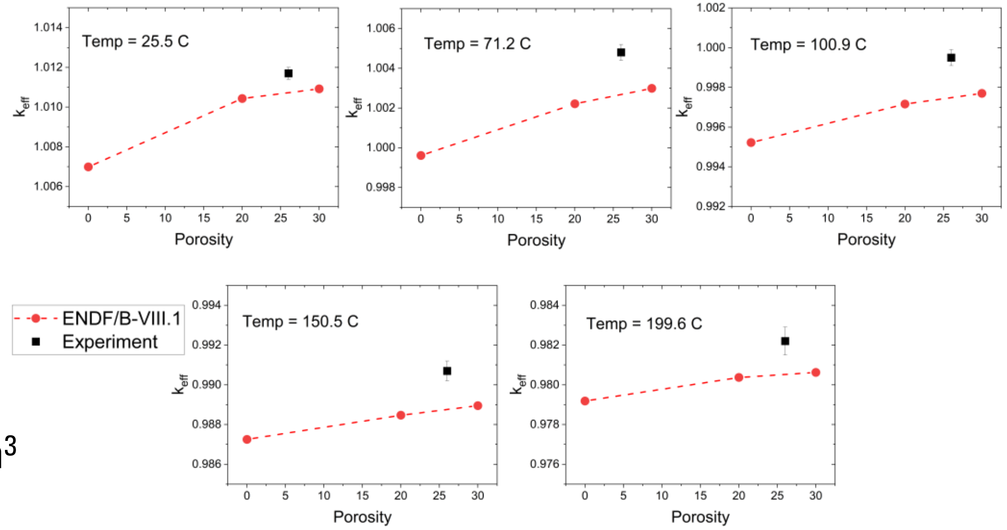


Figure 10. Calculation results of the k_{eff} values at 199.6℃ using graphite with each porosity.

JENDL 4.0	C-E (pcm)
Temp.	Crystalline Graphite
25.5°C	-444
71.2°C	-462
100.9°C	-413
150.5°C	-360
199.6°C	-307

Reproduced using MCNP



Density = 1.67 g/cm^3
Porosity ≈ 30%

ENDF/B-VIII.1	C-E (pcm)		
Temp.	0% Porous Crystalline Graphite	20% Porous Graphite	30% Porous Graphite
25.5°C	-462	-124	-76
71.2°C	-517	-257	-180
100.9°C	-430	-235	-181
150.5°C	-353	-228	-179
199.6°C	-314	-190	-164

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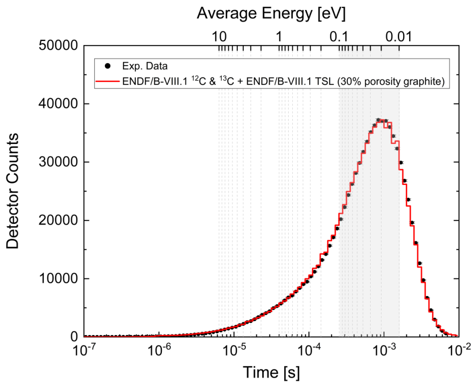
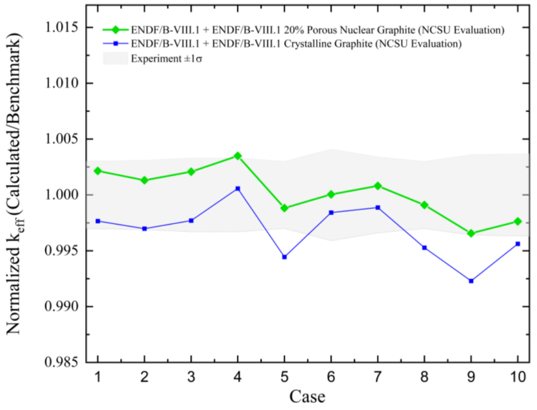
Nuclear Graphite – Much Improved!



FUND-ORELA-ACC-GRAPH-PNSDT-001
(Nuclear Graphite)

Density = 1.66 g/cm^3
Porosity ≈ 30%

Cross Sections	Mean Absolute Deviation (%)
ENDF/B-VIII.0 + Cry	4.14%
ENDF/B-VIII.1 + Cry	4.09%
ENDF/B-VIII.1 + Cry +S_d	5.01%
ENDF/B-VIII.1+30%	1.79%

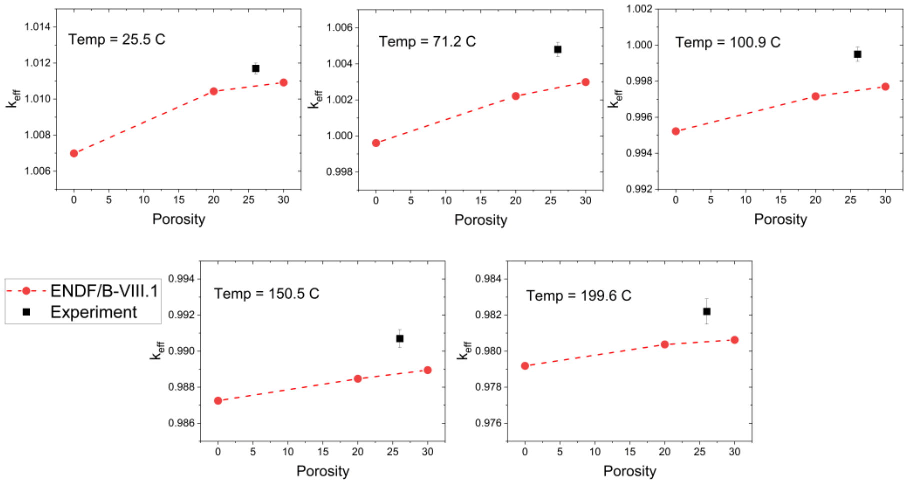


PROTEUS-GCR-EXP-001 to -004
(Nuclear Graphite)

Density = 1.7 g/cm^3
Porosity ≈ 20-30%

C-E (pcm)

Case	ENDF/B-VIII.1 Crystalline	ENDF/B-VIII.1 20% Porous
1	235	204
2	303	137
3	230	188
4	57	322
5	707	146
6	258	12
7	83	91
8	344	95
9	782	385
10	529	247
mean	350	183
χ^2	20	5



VHTRC-GCR-EXP-001
(Nuclear Graphite)

Density = 1.67 g/cm^3
Porosity ≈ 30%

ENDF/ B-VIII.1	C-E (pcm)		
Temp.	0% Porous Crystalline Graphite	20% Porous Graphite	30% Porous Graphite
25.5°C	-462	-124	-76
71.2°C	-517	-257	-180
100.9°C	-430	-235	-181
150.5°C	-353	-228	-179
199.6°C	-314	-190	-164

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Thank You

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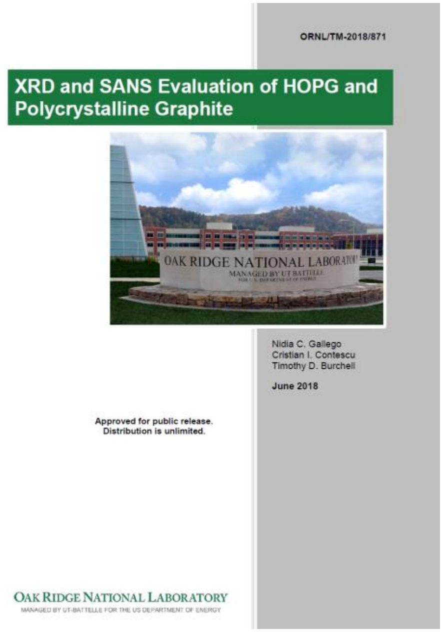
What Else?

SANS Observations


ORNL/TM-2018/871

XRD and SANS Evaluation of HOPG and Polycrystalline Graphite

Nidia C. Gallego
Cristian I. Contescu
Timothy D. Burchell

June 2018

Approved for public release.
Distribution is unlimited.

OAK RIDGE NATIONAL LABORATORY

- SANS contributions, based on data from the reference below, have a **negligible impact** on the total cross section and do not modify neutron thermalization in a reactor.

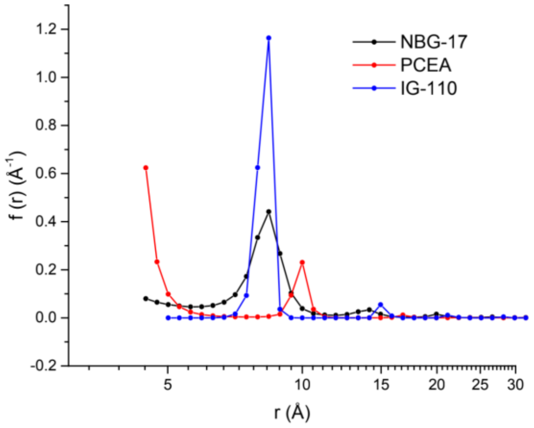
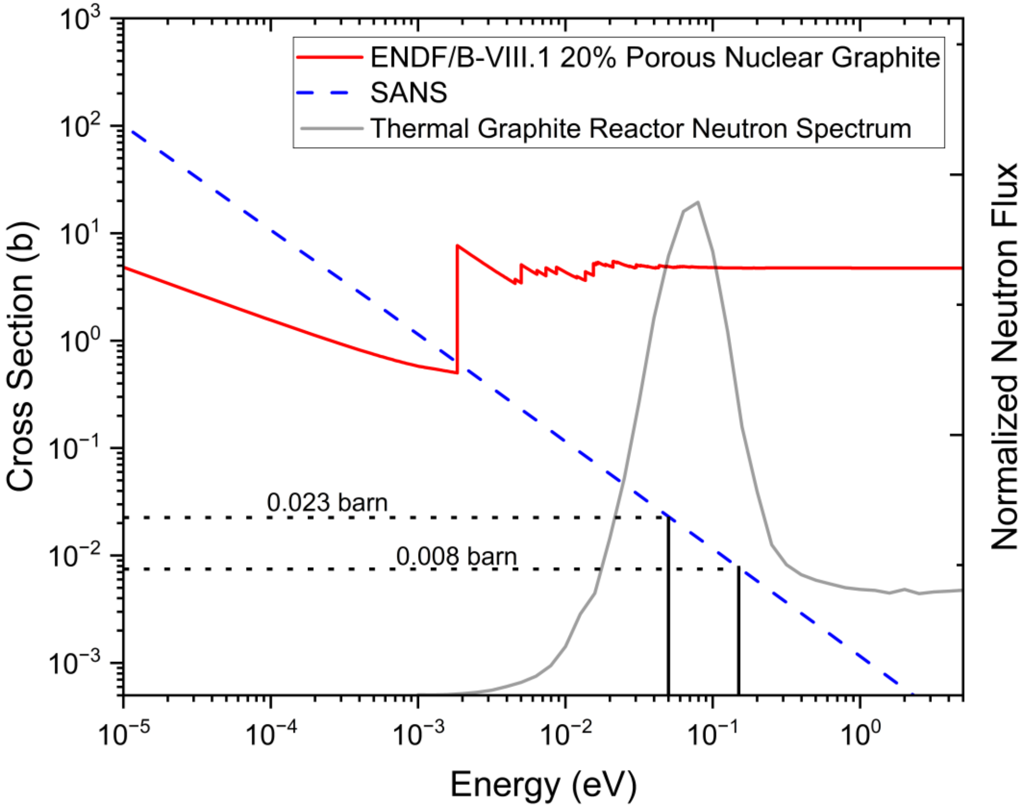


Figure 18 (b) from Reference

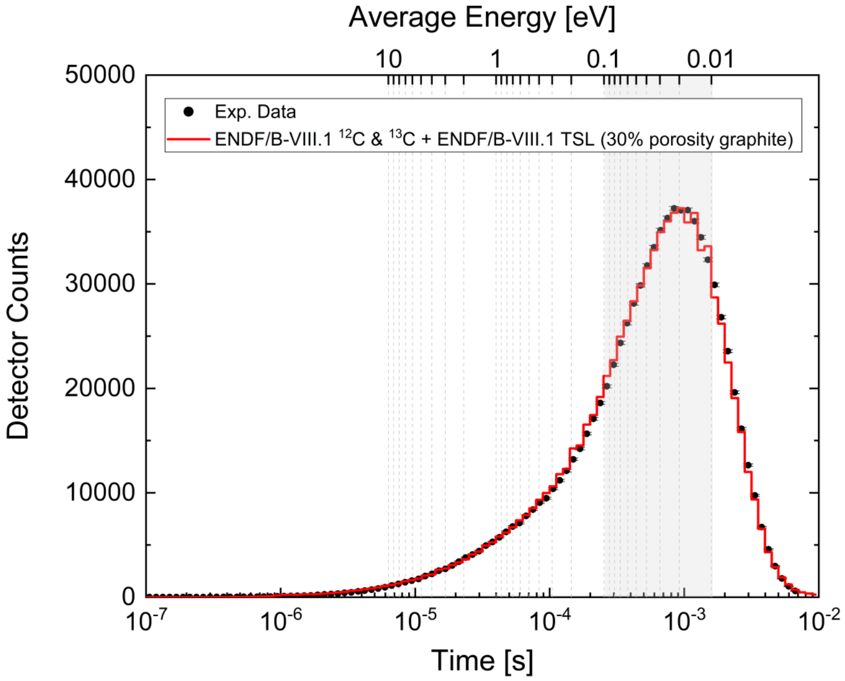


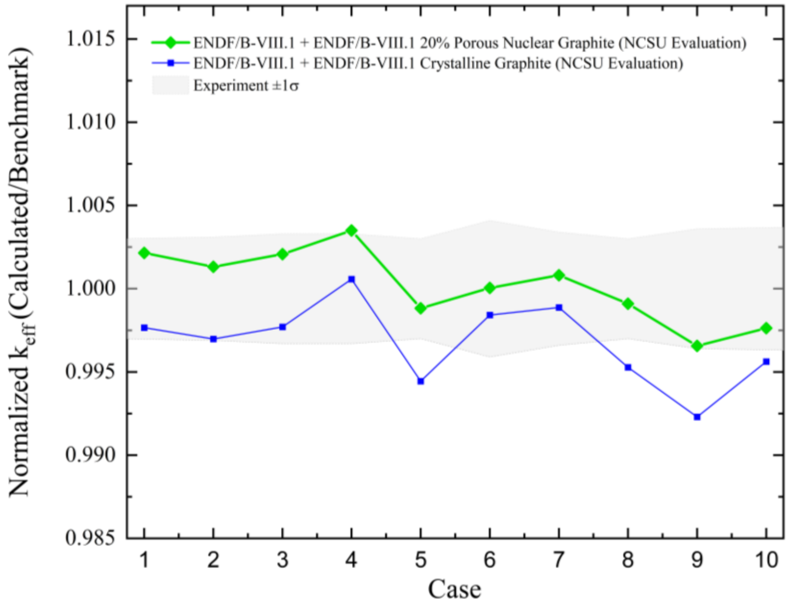
Ref. 2 - Petriw, et.al., “Porosity effects on the neutron total cross section of graphite”

Graphite Density

- ORELA and PROTEUS with porous nuclear graphite density (i.e. use 1.6-1.7 g/cm^3)

Self-consistent phonon DOS and density

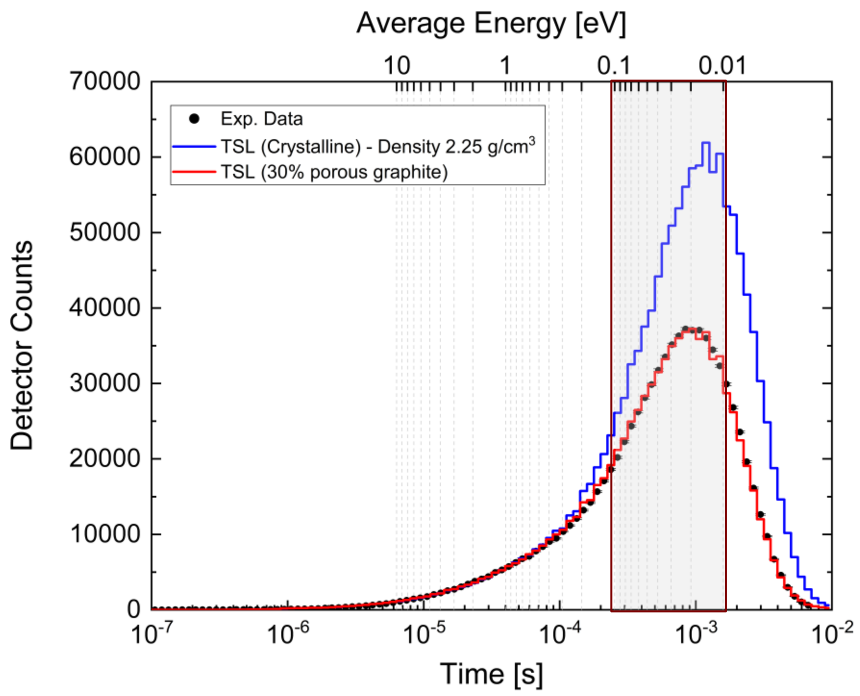


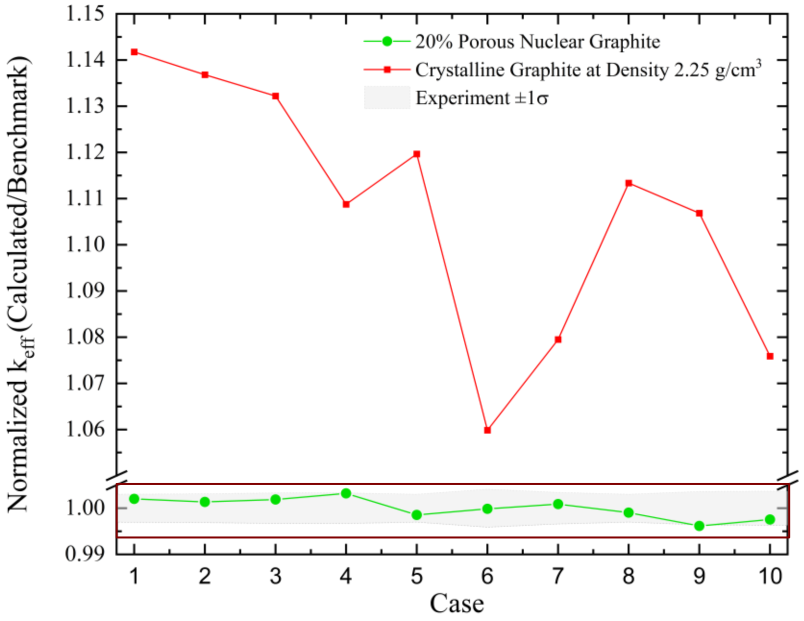


Graphite Density

- ORELA and PROTEUS without historical density approximation (i.e. use 2.25 g/cm^3)

Self-consistent phonon DOS and density

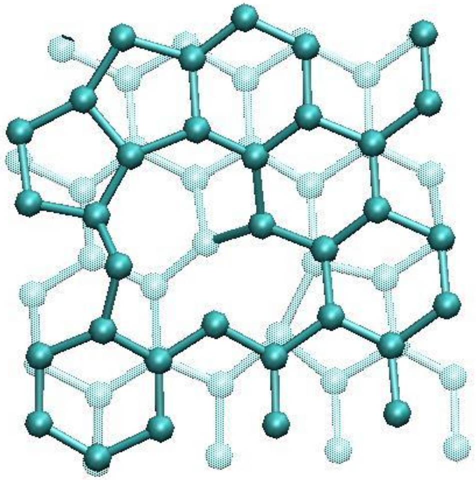




Radiation Damage
Interplanar Di-vacancy Defect

Observed in the MD system following cascade sequence. MD is predictive in this sense because no *a-priori* assumptions are necessary regarding the defect structure.

Static *ab-initio*, on the other hand, requires some initial guess that is then subject to optimization.



Development of the Thermal Neutron Scattering Cross Sections of Graphitic Systems using Classical Molecular Dynamics Simulations

by
Brian Douglas Hehr

A dissertation submitted to the Graduate Faculty of
North Carolina State University
in partial fulfillment of the
requirements for the Degree of
Doctor of Philosophy

Nuclear Engineering

Raleigh, North Carolina

2010

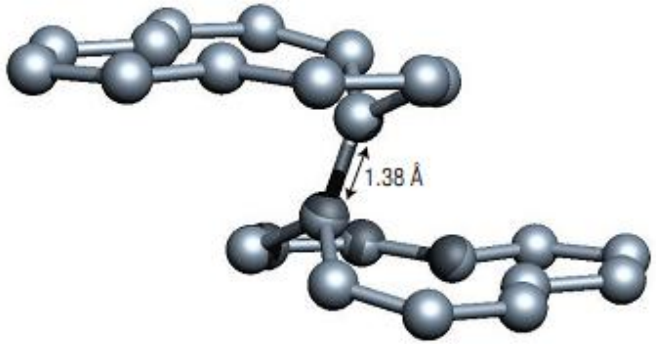
APPROVED BY:

Dr. Ayman I. Hawari,
Chair of Advisory Committee

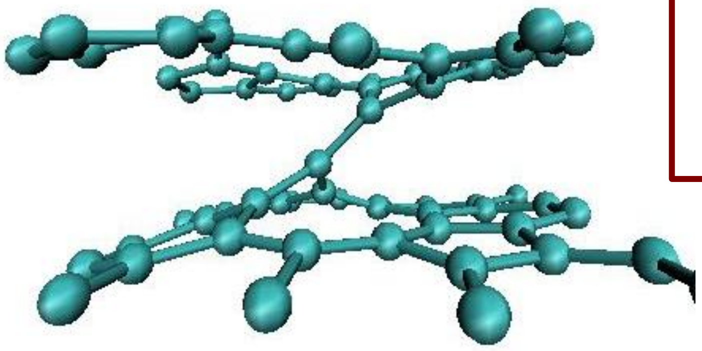
Dr. Mohamed A. Bourham

Dr. Bernard W. Wehring

Dr. Albert R. Young

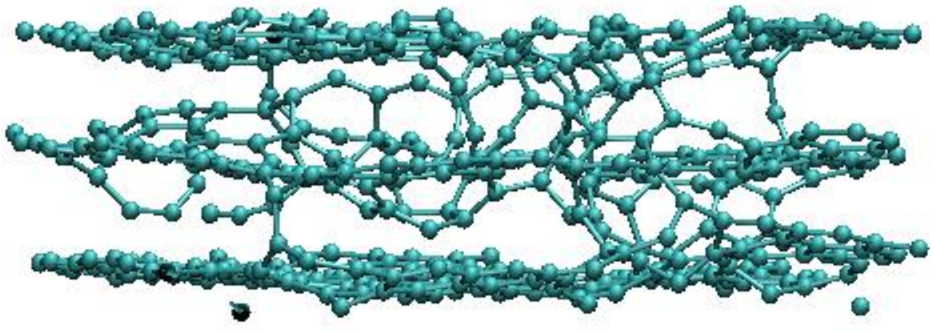


R. H. Telling, C. P. Ewels, A. A. El-Barbary and M. I. Heggie. *Nature Materials*. **2**, 333 (2003).
(ab-initio)

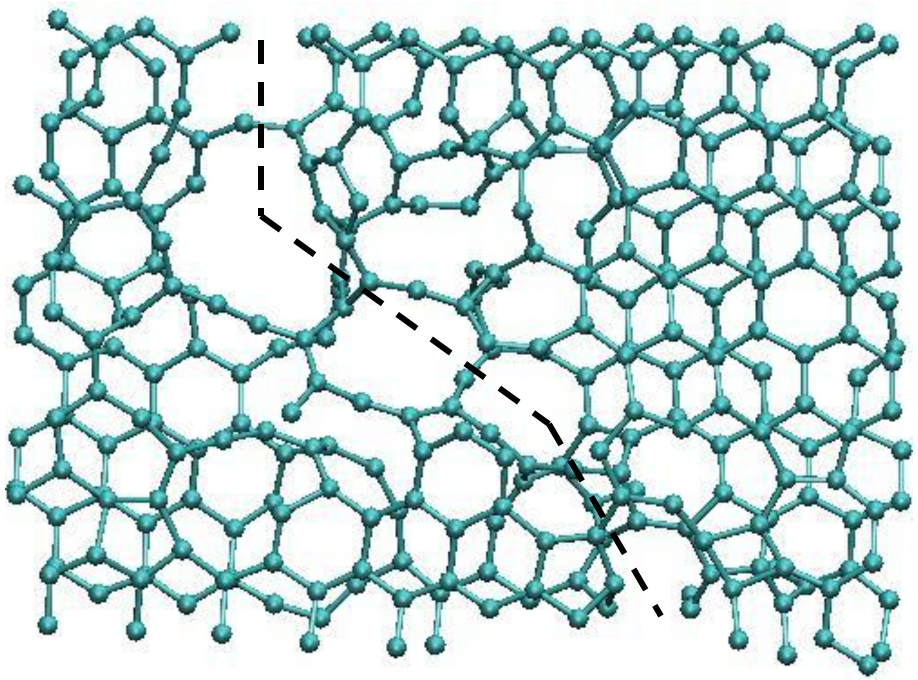


LEIP MD analysis

Radiation Damage
Interplanar Crosslinking



With increasing cascade buildup, the basal planes of graphite cross-link.



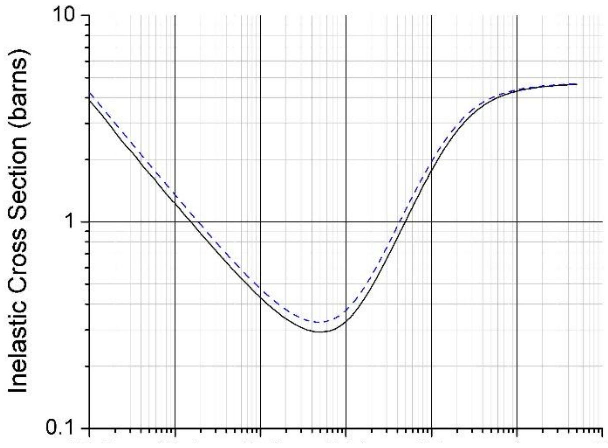
→ Individual point defects become less distinguishable

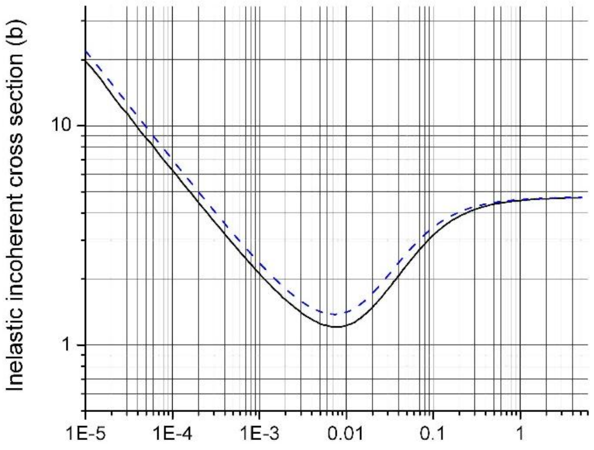
Radiation Damage
Effect of Temperature

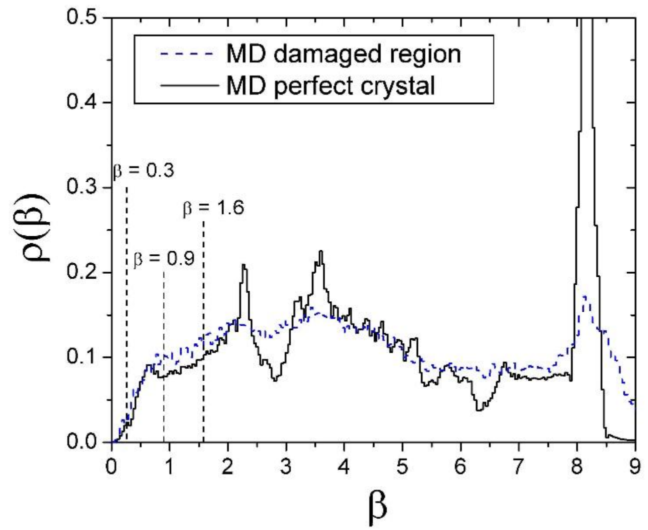
Cascade buildup

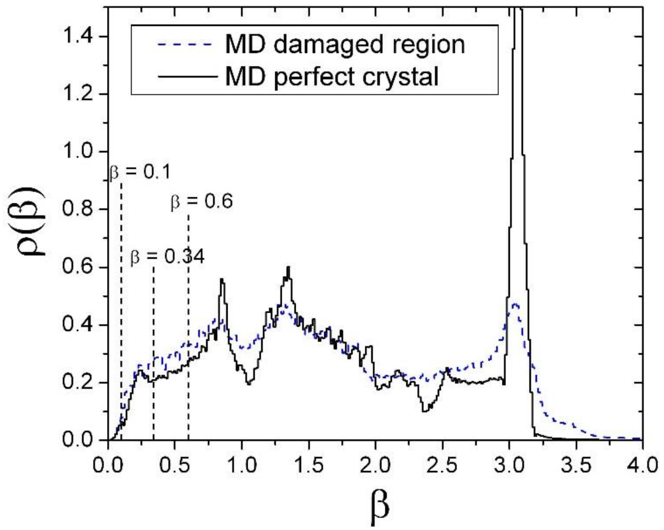
300K

800K









Porosity effect continues to dominate