Investigation and Evaluation of Thermal Neutron Scattering in Nuclear Graphite

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Advanced Nuclear Reactors

DOE-NE

Lead	Reactor Name	Reactor Type	Neutron Spectrum	Fuel Type	Power	Enrichment (wt% ^{235}U)	Moderator	Reflector	Coolant
TerraPower	Natrium	SFR	Fast	Sodium-bonded Metallic Alloy U-10Zr Pins	345 MWe	19.75	N/A	—	Salt
X-energy	Xe-100	Pebble Bed HTGR	Thermal	UCO TRISO Particle Spherical Graphite Compacts	80 MWe	15.5	Graphite	Graphite	Helium
Kairos Power	KP-FHR	Pebble Bed FHR	Thermal	UCO TRISO Particle Annular Spherical Graphite Compacts with Low-Density Graphite Cores	140 MWe	19.55	Pyrolytic Graphite, FLiBe	Graphite	FLiBe
Westinghouse Nuclear	eVinci	Heat-pipe Microreactor	Thermal	UCO TRISO Particle Cylindrical Graphite Compacts	5 MWe	19.75	Graphite	—	Sodium Heat Pipes
Southern Company and TerraPower	MCFR	MSR	Fast	Dissolved Uranium in Salt ($NaCl\text{-}UCl_3$)	800 MWe	HALEU	N/A	—	Salt
BWXT	BANR	HTGR	Thermal	UN TRISO in SiC/Carbon Matrix Compact, Additively Manufactured	50 MWth	19.75 (Baseline Design)	Graphite	—	Helium
ARC	ARC-100	SFR	Fast	Sodium-bonded U-10Zr pins	100 MWe	20 Max.; 13.1 Avg.	N/A	Stainless Steel	Sodium
GA-EMS	FMR	GFR	Fast	UO_2 Pellets	44 MWe	19.75	N/A	Zr_3Si_2 and Graphite	Helium
MIT	HC-HTGR	HTGR	Thermal	TRISO Particle Graphite Compact	~58 MWth	—	Graphite	—	Helium

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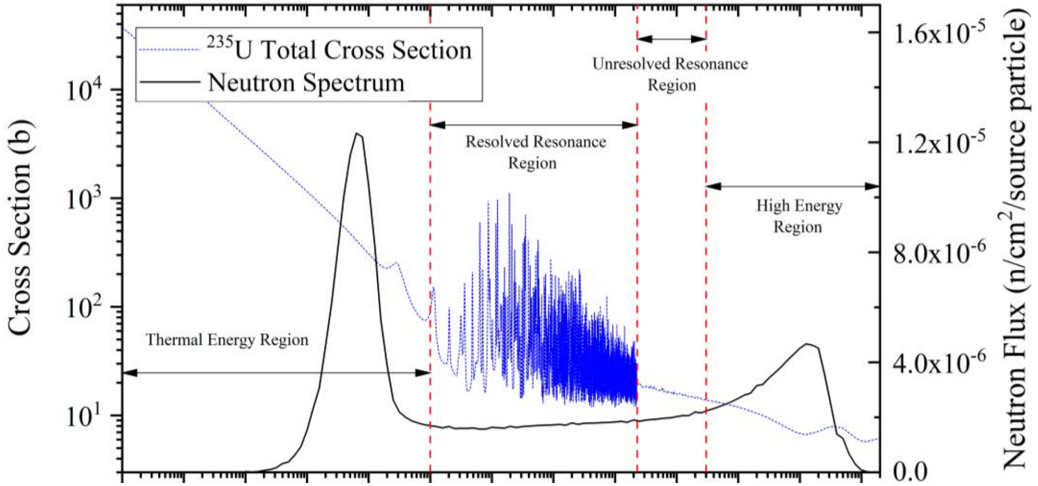
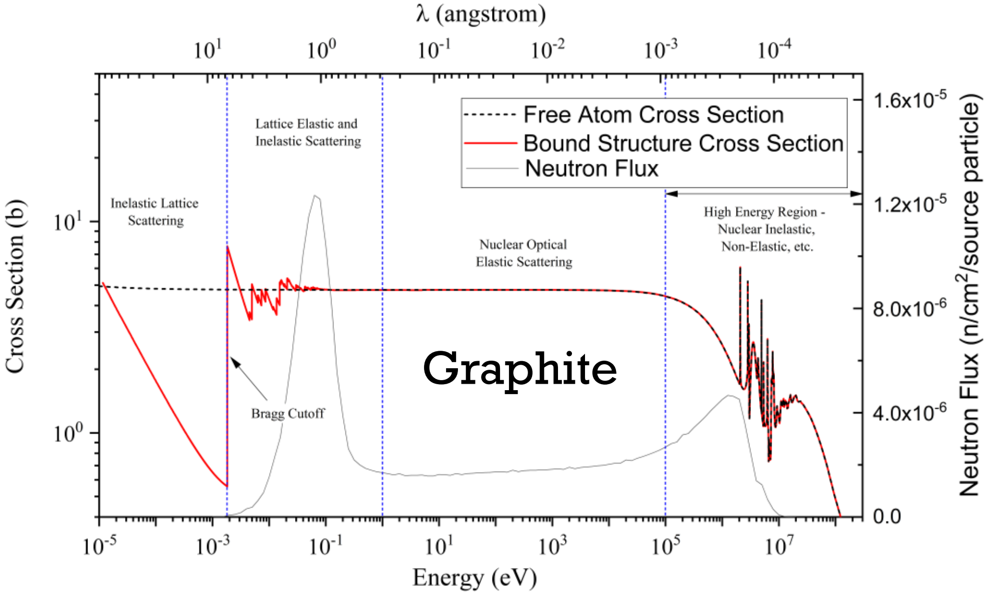
Thermal Nuclear Reactor





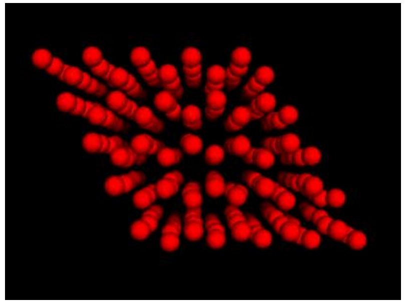
Fission

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

Thermalization

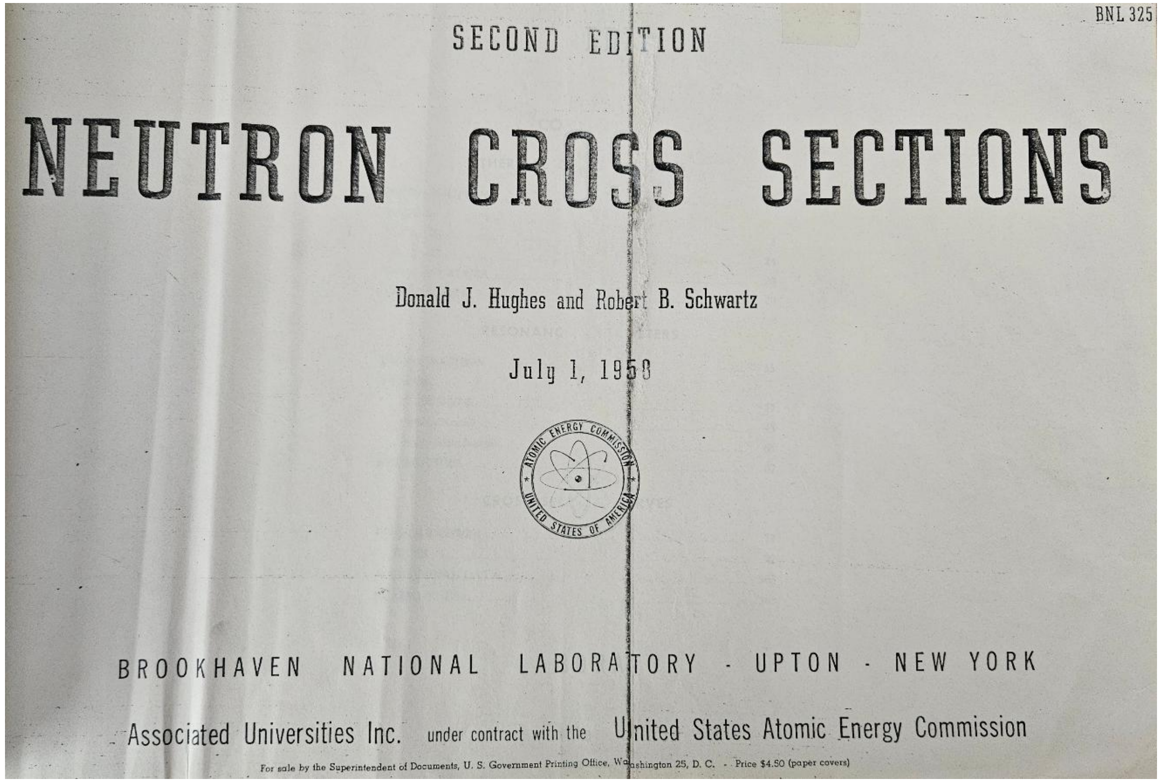
$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{1}{4\pi}\sqrt{\frac{E'}{E}}\left[\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)\right]$$

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



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

Graphite – What's the Problem (1950s)?



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SECOND EDITION

NEUTRON CROSS SECTIONS

Donald J. Hughes and Robert B. Schwartz

July 1, 1958

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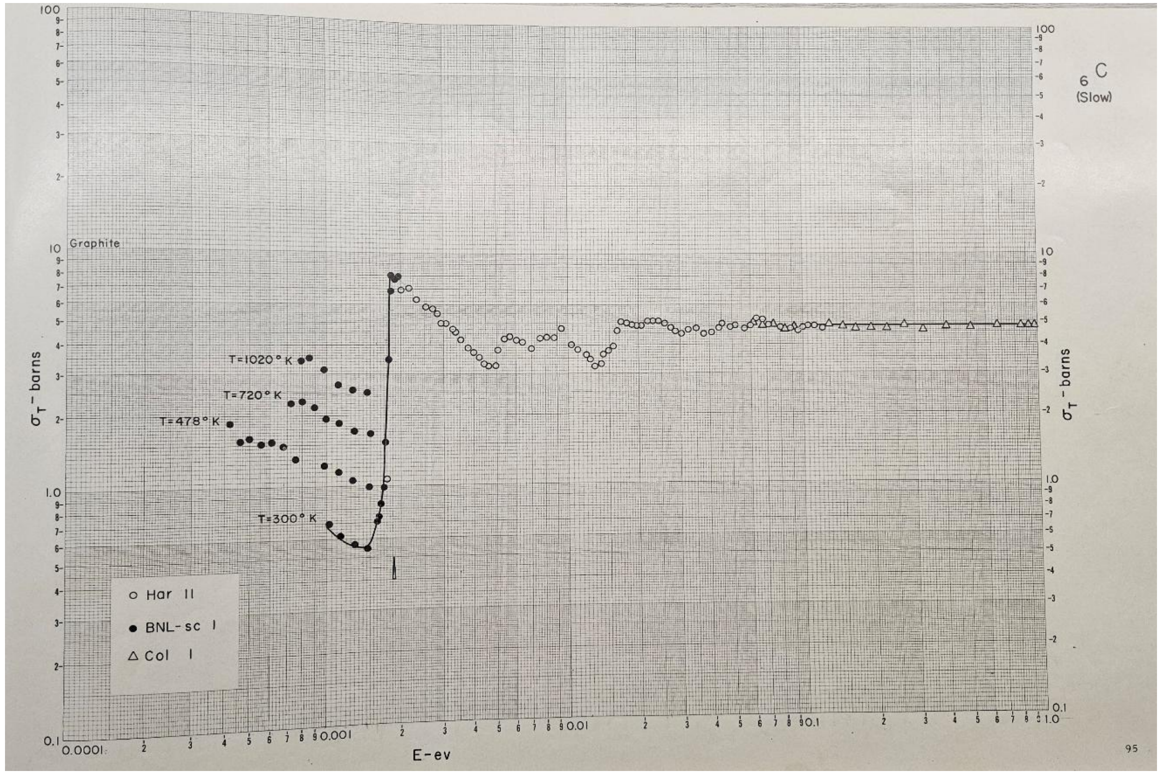
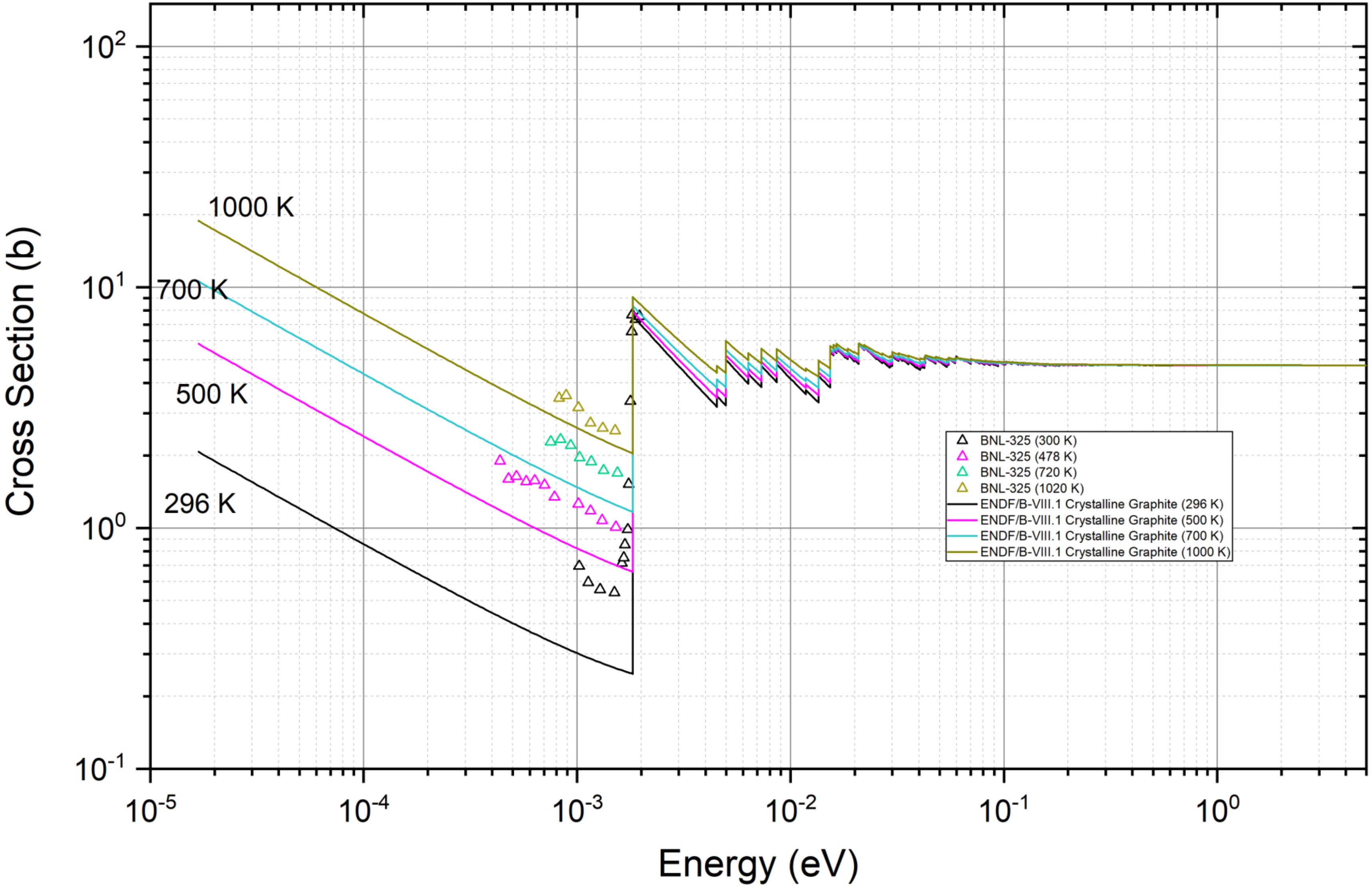


Graphite – What's the Problem (1950s)?



Graphite – What's the Problem (1980s)?

- Graphite reactor behavior
  - Criticality
  - Fission rates
  - Feedback
- Solution (ad hoc)
  - Assume crystalline (ideal) graphite structure (i.e., ideal S(α, β), but use "nuclear graphite" density (1.6-1.8 g/cm^3)
  - Mix crystalline graphite and free atom libraries to improve results.

CONF-880911--23

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DISCLAIMER

THE EFFECT OF CARBON CRYSTAL STRUCTURE ON
TREAT REACTOR PHYSICS CALCULATIONS

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1988 International Reactor Physics Conference
Jackson Hole, Wyoming
18-21 September 1988

Table II

Calculated Values of k-effective

Calculational Description	k-effective
Loading 1341 100% Graphite	0.9724 ± 0.0021
Loading 1341 59% Graphite	0.9921 ± 0.0012
Loading 1343 100% Graphite	0.9707 ± 0.0024
Loading 1343 59% Graphite	0.9922 ± 0.0017

Table III

Calculated Fission Density Ratios

Calculational Description	Calculated Fission Density Ratio	Calc.-Exp. / Exp.
100% Graphite	43.2 ± 1.4	+6.9%
59% Graphite	39.4 ± 1.2	-2.5%

Graphite – What's the Problem (2020s)?

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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

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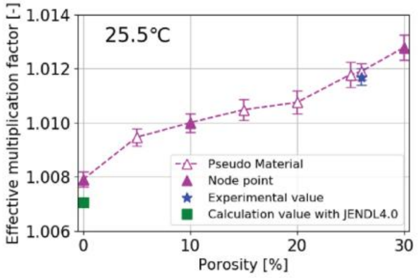

Figure 6. Calculation results of the k_{eff} values at 25.5 □ using graphite with each porosity.

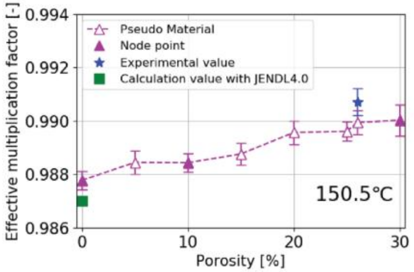

Figure 9. Calculation results of the k_{eff} values at 150.5 ℃ using graphite with each porosity.

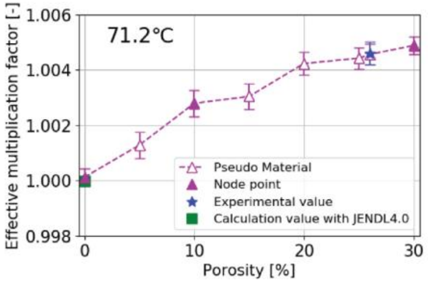

Figure 7. Calculation results of the k_{eff} values at 71.2 ℃ using graphite with each porosity.

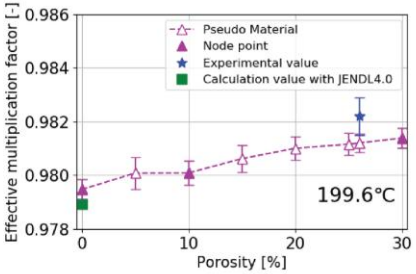

Figure 10. Calculation results of the k_{eff} values at 199.6℃ using graphite with each porosity.

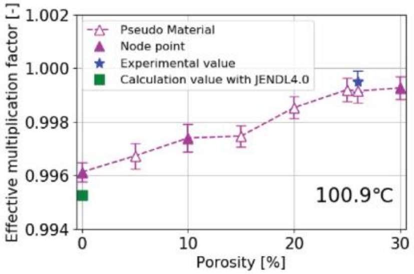

Figure 8. Calculation results of the k_{eff} values at 100.9 ℃ using graphite with each porosity.

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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Thermalization – Modern History (2000-2012)

U.S. DEPARTMENT OF ENERGY
NUCLEAR ENERGY RESEARCH INITIATIVE
ABSTRACT

PI: Ayman I. Hawari, Ph. D. **Proposal No.: 01-140**

Institution: University of Cincinnati

Title: Development and Validation of Temperature Dependent Thermal Neutron Scattering Laws for Applications and Safety Implications in Generation IV Nuclear Reactor Designs

The University of Cincinnati (UC), Oak Ridge National Laboratory (ORNL), and the Institute Balseiro (IB) propose to perform theoretical, computational, and experimental investigations on temperature dependent neutron thermalization in moderating materials that are of major importance to the safety of nuclear systems. The objectives of this work are: to critically review the currently used thermal neutron scattering laws for various moderators and fuel cells as a function of temperature, to use the review as a guide in examining and updating the various computational approaches in establishing the scattering law, to understand the implications of the obtained results on the ability to accurately define the operating and safety characteristics (e.g. the moderator temperature coefficient) of a given reactor design -- that is, to know not only the reactivity coefficients but also their errors, sensitivity coefficients and covariance matrices, to develop and generate new sets of temperature dependent thermal neutron scattering laws, $S(\alpha,\beta)$, either by an evolutionary process or by changing the models entirely (e.g., introducing the coherent part of the inelastic scattering or using the synthetic kernel approach), and finally to test and benchmark the developed models within the framework of a neutron slowing down experiment. In particular, the studies will concentrate on investigating the latest ENDF/B thermal neutron cross sections for reactor grade graphite, beryllium, beryllium oxide, zirconium hydride, high purity light water, and polyethylene at temperatures greater than or equal to room temperature. These materials are neutron moderators/reflectors that will be used in the development of Generation IV nuclear power reactors and in many applications in the nuclear science and engineering field. Of major importance is graphite, which is the moderator in the modular pebble bed reactor (MPBR) that is being examined internationally as a possible Generation IV power reactor, as the subcritical reactor in accelerator driven concepts, and as the incinerator of radioactive waste and weapon's plutonium. Furthermore, a newly developed highly conductive form of graphite, known as graphite foam, is currently under study as a reactor material. Added to that, these materials of interest in research reactors such as zirconium hydride (i.e., TRIGA), and in Nerva derivative power sources for space applications (e.g., zirconium hydride, Be and BeO reflectors).

To begin this work, we will perform a critical analysis of the models that are the basis of the present ENDF/B evaluation of the scattering law for a given moderator, and determine the sensitivity of calculated thermal neutron spectra to the details and parameters of the models. Furthermore, we will consider the impact of model parameters on the behavior of the neutron flux around nearby neutron absorption resonances that are going to define global quantities such as the asymptotic neutron spectra and, consequently, reactivity coefficients. We will also study the impact of model parameters on other measured observables such as neutron pulse and wave propagation parameters, and decay constants as a function of size and transuranium buildup and depletion. In addition, we will examine the latest developments in

Continued **01-140**

thermalization theory and condensed matter physics. Experiments, like the direct measurement of the double differential cross sections and specific heats, and theoretical developments (e.g., new phonon distribution in graphite) will be evaluated to define the degrees of freedom of the scattering media as well as the mechanisms for the transference of energy between the media and the neutrons. All this information will be included as input for new calculations of the scattering matrices with an updated version (to be developed as part of this research) of the present "state of the art" computer codes (used for ENDF/B compilations). As a result, thermal scattering laws, $S(\alpha,\beta)$, will be regenerated using basic input data and modern computational methods. Using these new sets of $S(\alpha,\beta)$, we will analyze the computational anomalies in the thermal scattering data that is in general use today and can cause "strange" behavior in the computational determination of the temperature coefficient of reactivity in nuclear reactors. Discrepancies of up to 150% have been encountered, which may have important safety implications [1]. Consequently, the impact of condensed matter models, and their respective input parameters, on the temperature coefficient of reactivity in nuclear reactors will be determined. Moreover, the computationally regenerated scattering data for the moderators of interest will be used in neutronics calculations of the temperature coefficients of reactivity for several Generation IV nuclear power reactors (e.g., MPBR).

Finally, since graphite is the moderator in various Generation IV reactor concepts, we will benchmark the developed $S(\alpha,\beta)$ model for graphite by performing a neutron thermalization experiment in a graphite (and if available in graphite foam) moderator that is driven by a pulsed neutron source. This measurement approach has the ability to observe the neutron behavior in a moderator as it passes through the slowing down and thermalization energy ranges before its diffusion and escape. Therefore, it is applicable to measurements in the energy range below 1 eV, which is not accessible using the traditional out-of-pile leakage spectrum measurements. The experiment will take place using the Oak Ridge Electron Linear Accelerator (ORELA) facility, and will be performed to obtain the integral time dependent reaction rate of a neutron detector that is placed within the moderator at various temperatures greater than or equal to room temperature (including temperatures encountered in normal operations and during reactor accident conditions). In addition, a beam will be extracted from the moderator to perform temperature dependent measurements (using the ORELA time of flight facilities) of the thermal neutron energy spectrum in the moderator. We will also introduce the new graphite $S(\alpha,\beta)$ data into time and energy dependent 3-D Monte Carlo (e.g., MCNP) computer simulations of the experiments. This will provide computational predictions of the experimental data and will enable validation of the nuclear data libraries for graphite in the thermal energy region.

Major outcomes

1) Modern condensed matter physics methods (AILD and MD)
2) Modern scattering theory (removed many approximations)
3) Several experiments including Pulsed neutron slowing down benchmark methods

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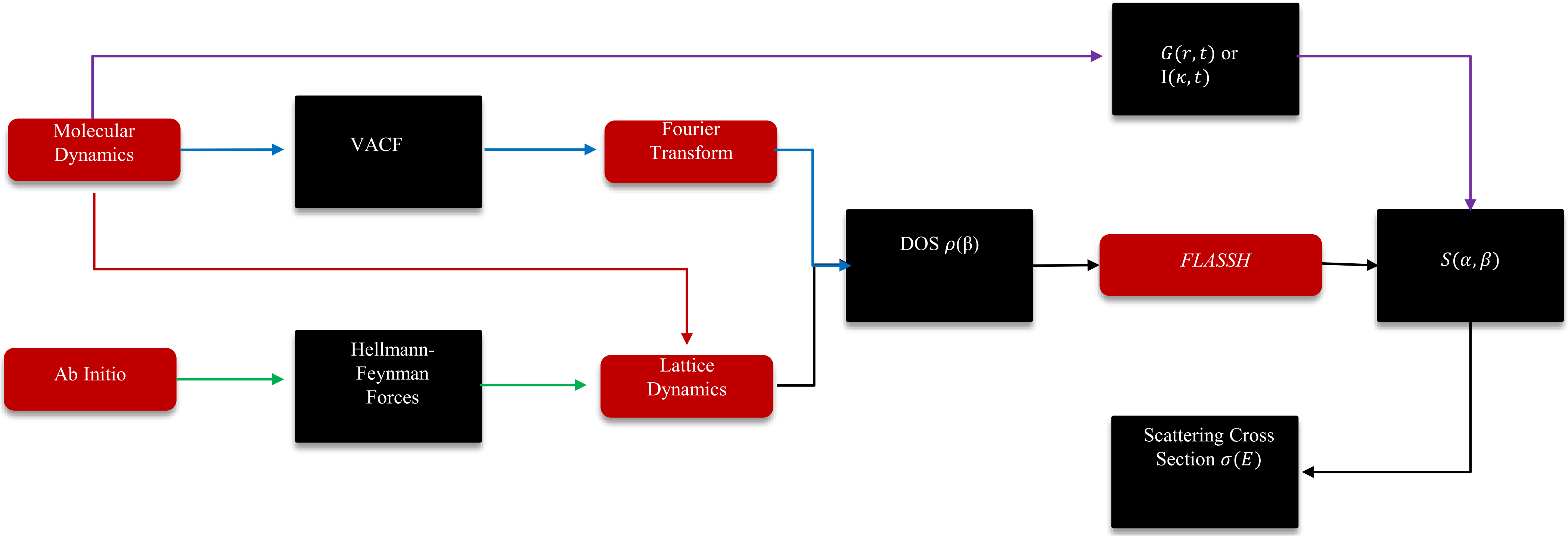
Thermalization – Modern History (2000-2012)

- **A selection of computational routes**
  - Somewhat material dependent



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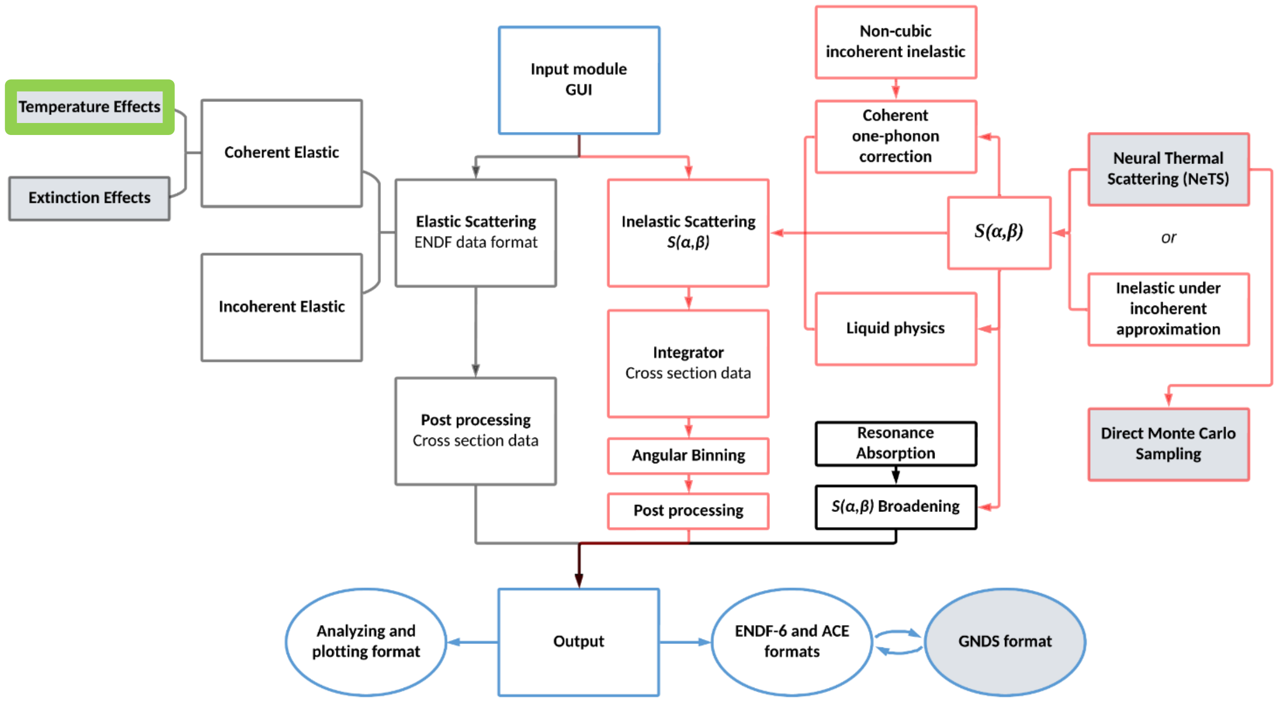
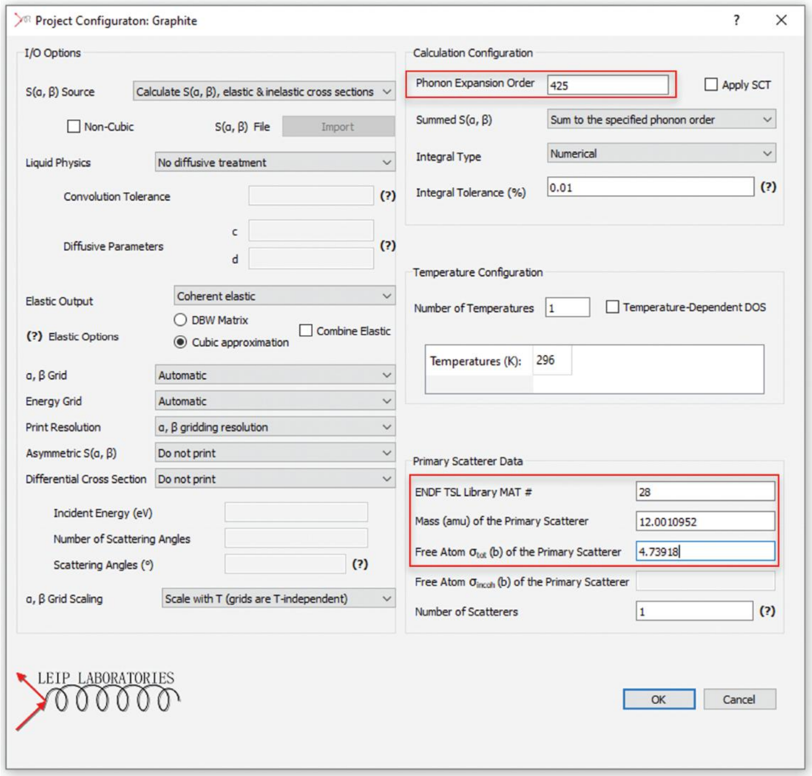


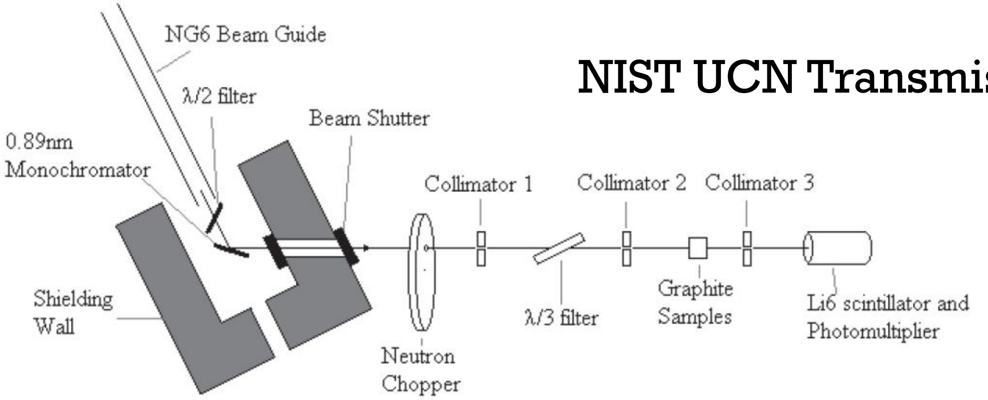


- Advanced features including
  - Temperature dependent phonon spectra
  - Coherent elastic scattering extinction effects
  - NeTS modules for key moderators
  - Improved liquid physics (addressing high viscosity liquids)
  - Enhanced Data formatting capabilities

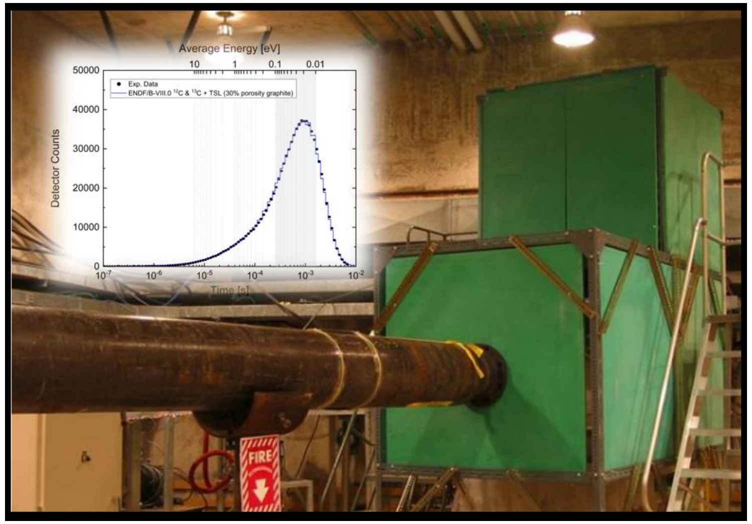
Thermalization – Modern History (2000-2012)

- Experimental **HOLISTIC** approach

NIST UCN Transmission



ORELA Benchmark



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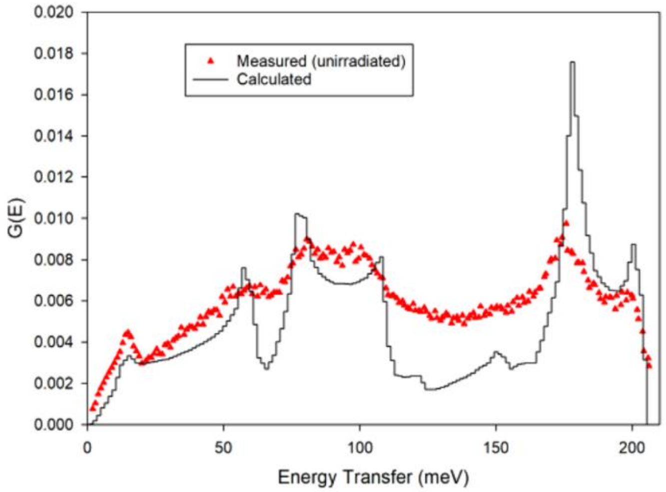

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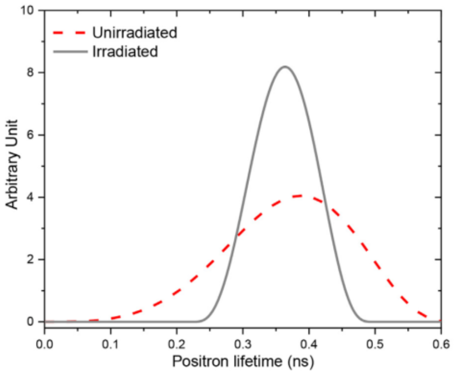

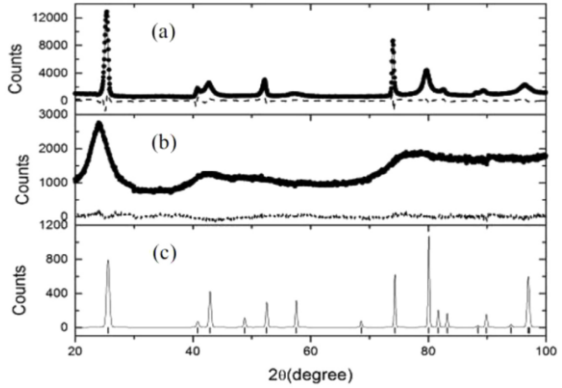

TSL – Modern History (2000-2012)

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Modern Techniques for Inelastic Thermal Neutron Scattering Analysis

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A predictive approach based on ab initio quantum mechanics and/or classical molecular dynamics simulations has been formulated to calculate the scattering law, $S(\vec{\kappa},\omega)$, and the thermal neutron scattering cross sections of materials. In principle, these atomistic methods make it possible to generate the inelastic thermal neutron scattering cross sections of any material and to accurately reflect the physical conditions of the medium (i.e, temperature, pressure, etc.). In addition, the generated cross sections are free from assumptions such as the incoherent approximation of scattering theory and, in the case of solids, crystalline perfection. As a result, new and improved thermal neutron scattering data libraries have been generated for a variety of materials. Among these are materials used for reactor moderators and reflectors such as reactor-grade graphite and beryllium (including the coherent inelastic scattering component), silicon carbide, cold neutron media such as solid methane, and neutron beam filters such as sapphire and bismuth. Consequently, it is anticipated that the above approach will play a major role in providing the nuclear science and engineering community with its needs of thermal neutron scattering data especially when considering new materials where experimental information may be scarce or nonexistent.

I. INTRODUCTION

Low energy or "thermal" neutrons are characterized by energies that are on the order of the excitation (vibration, rotation etc.) energy in the medium in which they interact. Furthermore, their de Broglie wavelength is on the order of the separation distance in solids. Consequently, such neutrons are highly sensitized to the atomic binding details of the system that surrounds them including its structure and dynamics. In fact, the structural and dynamic properties of the atomic system are sampled through scattering interactions between the system's atoms and molecules and the neutrons. The scattering of low energy neutrons in an atomic system is generally described using thermal neutron scattering cross sections. Traditionally, the cross sections are quantified based on Born scattering theory combined with Fermi's Golden rule and the assumption of an extremely short range (delta function) nuclear potential [1]. The outcome of this approach, is an expression for the double differential scattering cross section given by

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

where $S(\vec{\kappa},\omega)$ is known as the scattering law, $\vec{\kappa}$ is the scattering vector, ω is the frequency, k and k' represent the magnitude of the wave vector of the incident and scattered neutron respectively, σ_{coh} is the bound atom coherent scattering cross section, and σ_{inc} is the bound atom incoherent scattering cross section. In general, S is composed of two terms as follows

$$S\left(\vec{\kappa},\omega\right) = S_s\left(\vec{\kappa},\omega\right) + S_d\left(\vec{\kappa},\omega\right), \tag{2}$$

where S_s is known as the self-scattering law, which accounts for non-interference (incoherent) effects, while S_d is the distinct scattering law and accounts for interference (coherent) effects. Examination of Eq. 1 shows that the thermal neutron scattering cross section depends on two factors: first, the neutron-nucleus interaction as represented by the bound atom cross sections, and second, a factor that represents the dynamics of the scattering system (i.e., the collection of atoms) as represented by the scattering law.

Frequently, the calculations of the thermal scattering cross section invoke the incoherent approximation where S_d is set equal to zero in Eq. 2. Based on this assumption, Eq. 1 is developed to give (e.g., see Ref. [2])

$$\frac{d^2\sigma}{d\Omega dE} = \frac{1}{4\pi}\frac{k'}{k}\left[\sigma_{coh} + \sigma_{incoh}\right] S_s\left(\vec{\kappa},\omega\right). \tag{3}$$

However, for some important neutronic materials such as graphite and beryllium, this assumption can introduce

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Inelastic Thermal Neutron Scattering Cross Sections for Reactor-grade Graphite

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Current calculations of the inelastic thermal neutron scattering cross sections of graphite are based on representing the material using ideal single crystal models. However, the density of reactor-grade graphite is usually in the range of 1.5 g/cm^3 to approximately 1.8 g/cm^3, while ideal graphite is characterized by a density of nearly 2.25 g/cm^3. This difference in density is manifested as a significant fraction of porosity in the structure of reactor-grade graphite. To account for the porosity effect on the cross sections, classical molecular dynamics (MD) techniques were employed to simulate graphite structures with porosity concentrations of 10% and 30%, which are taken to be representative of reactor-grade graphite. The phonon density of states for the porous systems were generated as the power spectrum of the MD velocity autocorrelation functions. The analysis revealed that for porous graphite the phonon density of states exhibit a rise in the lower frequency region that is relevant to neutron thermalization. Using the generated phonon density of states, the inelastic thermal neutron scattering cross sections were calculated using the NJOY code system. While marked discrepancies exist between measurements and calculations based on ideal graphite models, favorable agreement is found between the calculations based on the porous graphite models and measured data.

I. INTRODUCTION

Traditionally, ENDF/B graphite evaluations provide what is known as $S(\alpha,\beta)$ libraries to describe neutron thermalization. These evaluations are based on representing the material using ideal single crystal models [1]. Furthermore, the libraries are generated within the incoherent approximation of thermal neutron scattering theory. Past work showed that this approach introduced noticeable inaccuracies in the generated cross sections relative to measured data [2]. While the deficiencies due to the incoherent approximation were remedied by accounting for coherent inelastic scattering, it remained clear that the discrepancies between the measured cross sections for reactor-grade graphite and the estimates based on ideal single crystal models require a review of the atomisitic models from which these estimates were derived.

Ideal graphite consists of planes (sheets) of carbon atoms arranged in a hexagonal lattice with 4 atoms per unit cell. Strong covalent bonding exits between intraplaner atoms, while the interplaner bonding (i.e., between the carbon sheets) is of the weak Van der Waals type. The planes are stacked in an "ABAB" sequence. Fig. 1 shows a representation of ideal crystalline graphite.

Alternatively, reactor-grade graphite represents a multi-phase material where graphite ideal crystals are embedded in a carbon binder matrix. Most strikingly, the density of reactor-grade graphite is usually in the range of 1.5 g/cm^3 to approximately 1.8 g/cm^3, while ideal graphite is characterized by a density of nearly 2.25 g/cm^3. This difference in density is manifested as a significant fraction of porosity in the structure of reactor-grade graphite. However, this structural feature of graphite is not captured in the process of generating the inelastic thermal neutron scattering cross sections.

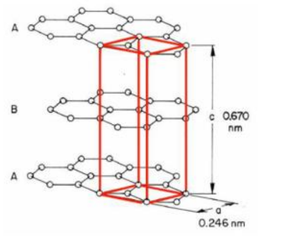

FIG. 1. The crystal structure of ideal graphite including its unit cell (bold lines).

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

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ABSTRACT

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.

1. Introduction

Over the past 15 years, a general methodology has been developed to generate thermal neutron scattering law (TSL) data, and calculate the inelastic thermal neutron scattering cross sections, for materials under various conditions and while relaxing many of the traditional approximations (e.g., the incoherent approximation) (Hawari, 2014, 2004). The methodology is based on using molecular dynamics (MD) and density functional theory (DFT) atomistic simulation methods to derive the fundamental input needed for TSL calculations, such as the atomic and molecular system's excitation density of states. Alternatively, the TSL may be directly accessed from the atomistic simulations and the resulting atomic correlations. In this case, corrections are needed to account for missing quantum effects such as detailed balance (Hehr, 2010). This methodology has resulted in the largest contribution (in the last 50 years) of TSL data to the recently released ENDF/B-VIII.0 nuclear data libraries (Brown, 2018).

In general, the formulation that would be the subject of the evaluation process originates from the fundamental equations for the double differential thermal scattering cross section. This formulation is derived using the first Born approximation of scattering theory and assumes a highly localized nuclear potential known as the Fermi pseudopotential [e.g., see Ref. Squires, 1978]. The outcome of this formulation is the following expression for the double differential thermal scattering cross section

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{1}{4\pi k_B T}\sqrt{\frac{E'}{E}}[\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)] \tag{1}$$

$$S(\alpha,\beta) = S_s(\alpha,\beta) + S_d(\alpha,\beta), \tag{2}$$

where, $S(\alpha,\beta)$ is the thermal scattering law, $S_s(\alpha,\beta)$ is the self scattering law, $S_d(\alpha,\beta)$ is the distinct scattering law, α and β are dimensionless momentum and energy exchange variables, σ_{coh} is the coherent bound atom cross section, σ_{inc} is the incoherent bound atom cross section, E in the incoming neutron energy, E' is the scattered neutron energy, Ω is the scattering solid angel, k_B is Boltzmann's constant, and T is the temperature of the scattering medium. In this case, S_s is related to the Fourier transform in space and time of the density correlation function for an atom at a given initial location in the atomic system with its location at time t. S_d is related to the Fourier transform in space and time of the density

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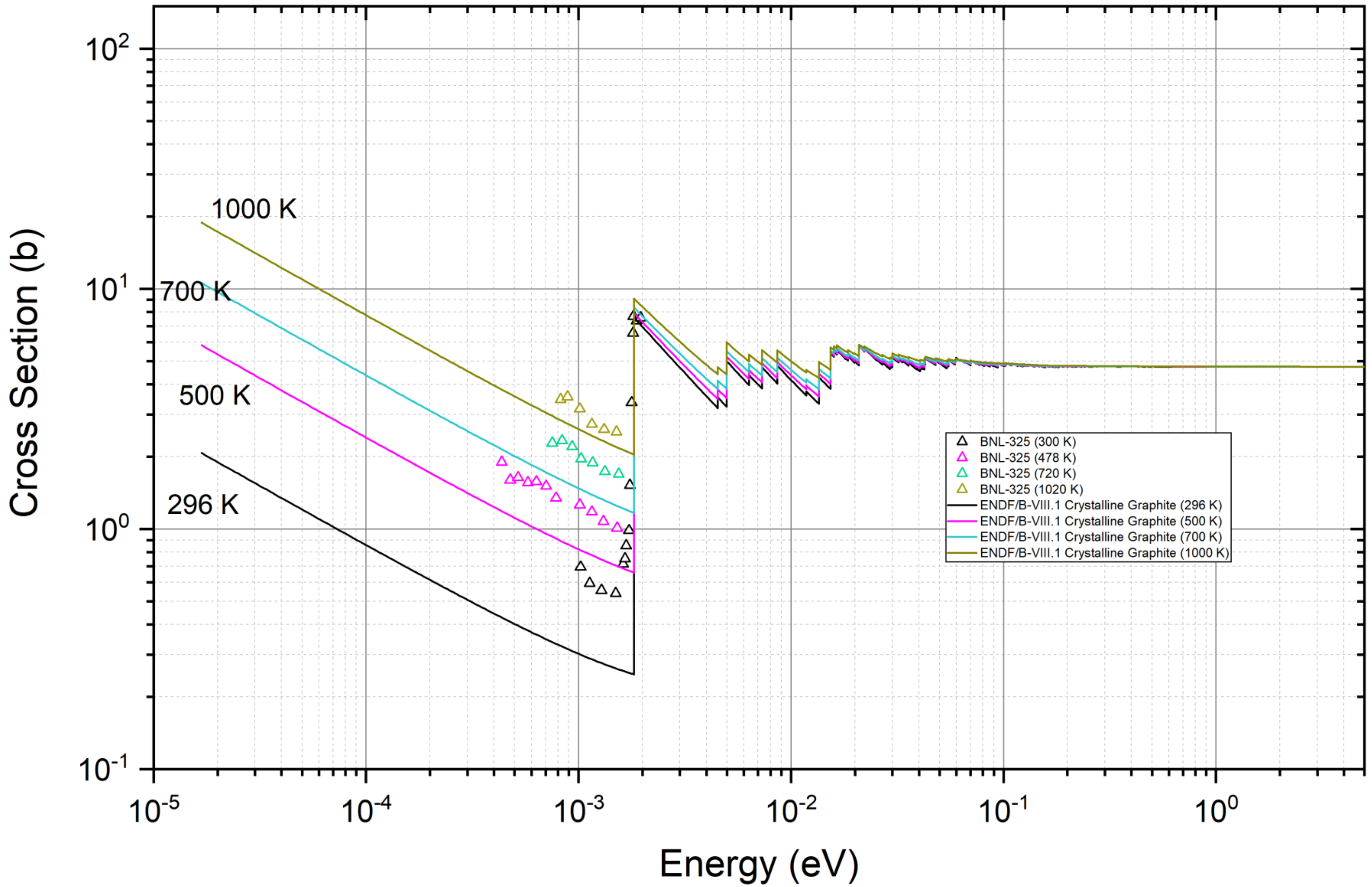
Graphite – Still a Problem!



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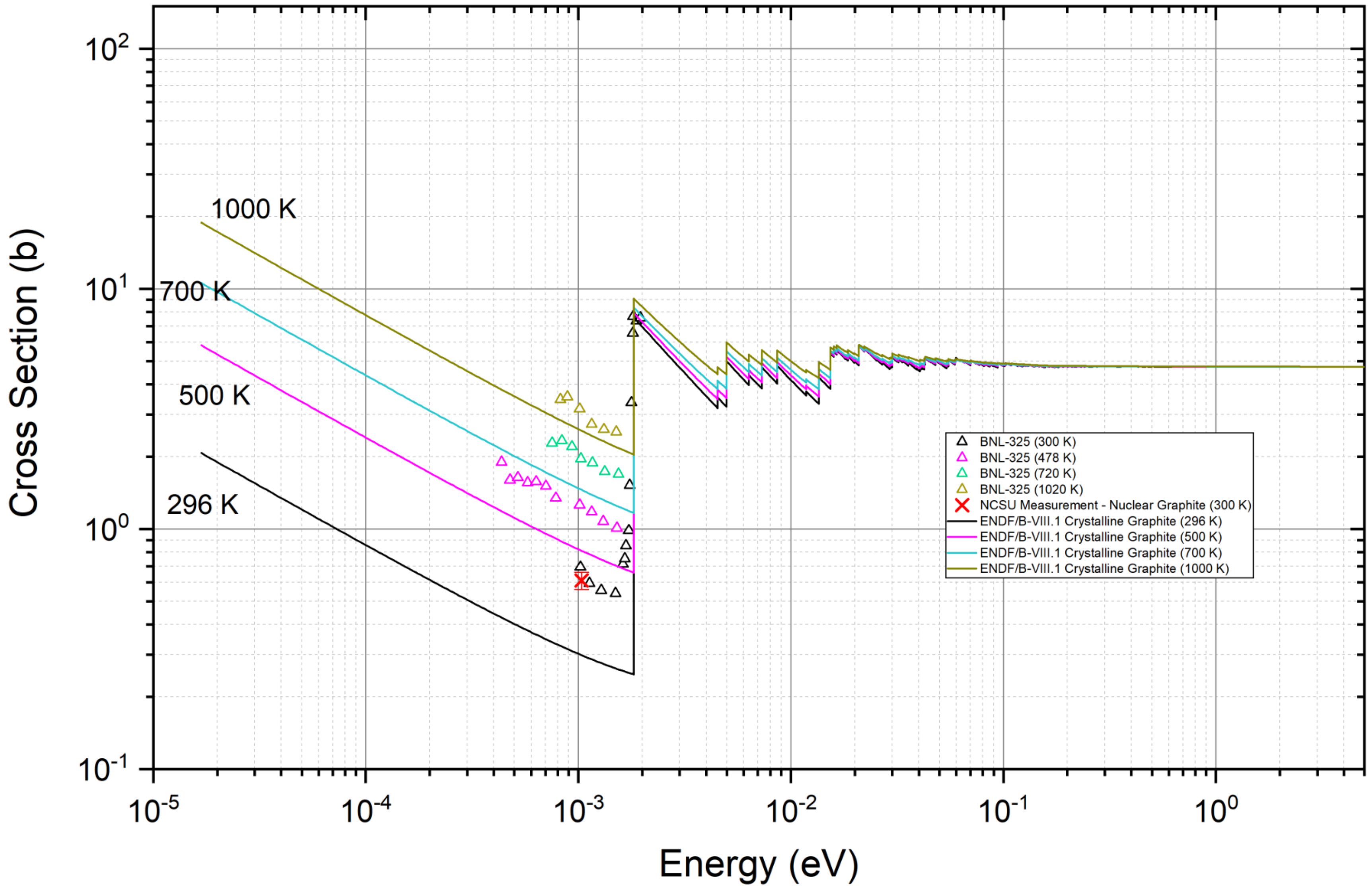

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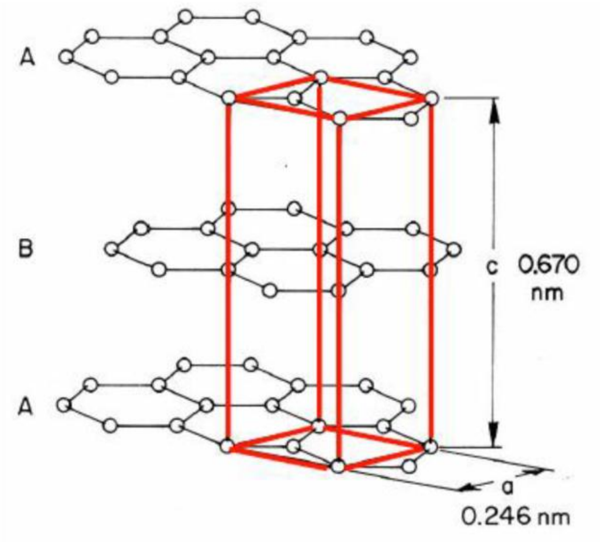

- Model used since the 1960s and until ENDF/B-VII.1 (released in 2011).

CONF-880911--23

DE89 003625

THE EFFECT OF CARBON CRYSTAL STRUCTURE ON TREAT REACTOR PHYSICS CALCULATIONS

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submitted to:

1988 International Reactor Physics Conference
Jackson Hole, Wyoming
18-21 September 1988

Table II

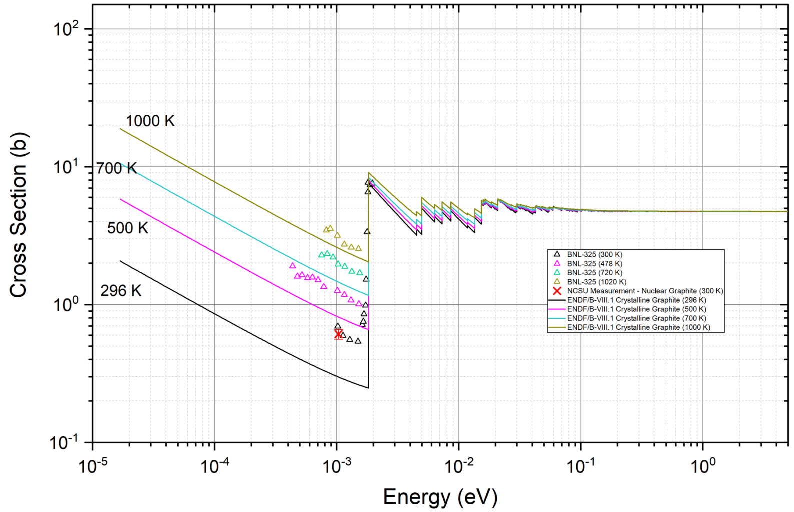
Calculated Values of k-effective

Calculational Description	k-effective
Loading 1341 100% Graphite	0.9724 $\pm$ 0.0021
Loading 1341 59% Graphite	0.9921 $\pm$ 0.0012
Loading 1343 100% Graphite	0.9707 $\pm$ 0.0024
Loading 1343 59% Graphite	0.9922 $\pm$ 0.0017

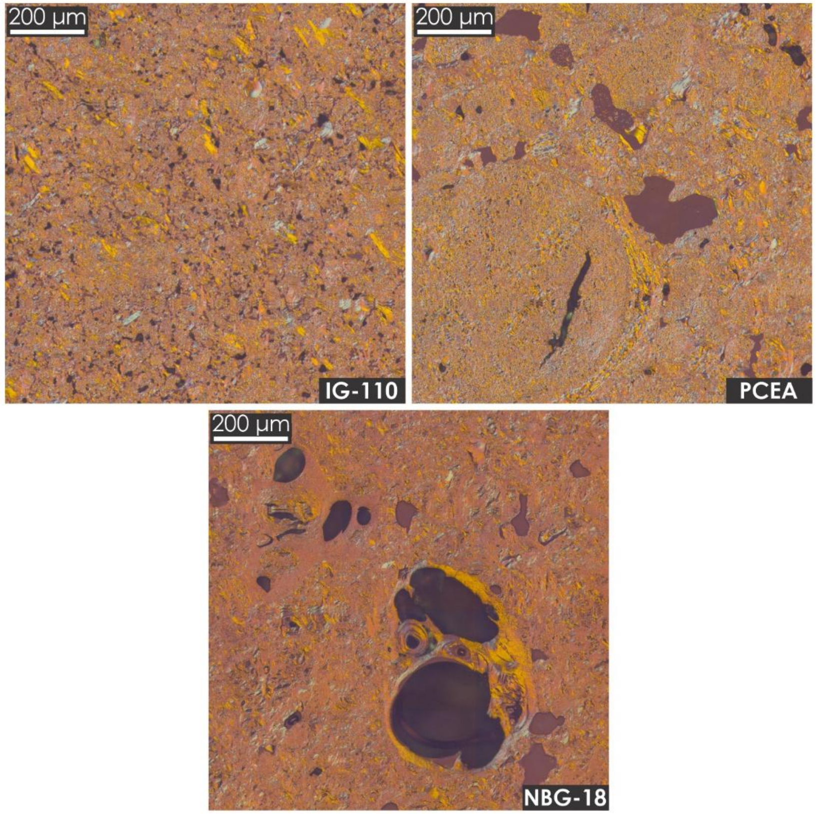
Table III

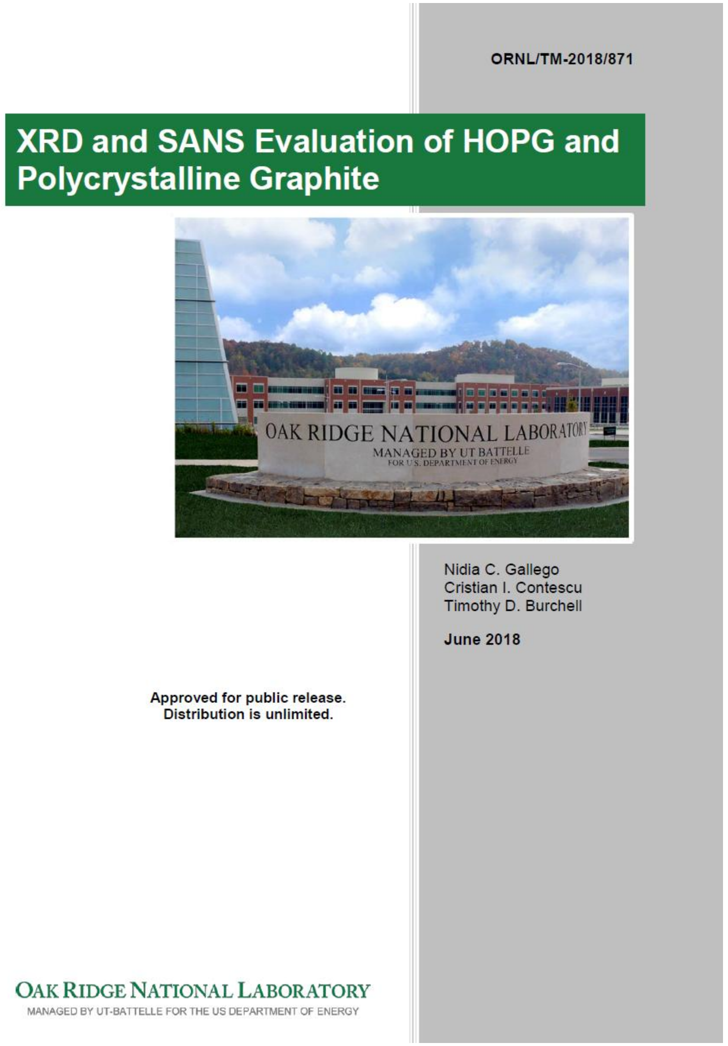
Calculated Fission Density Ratios

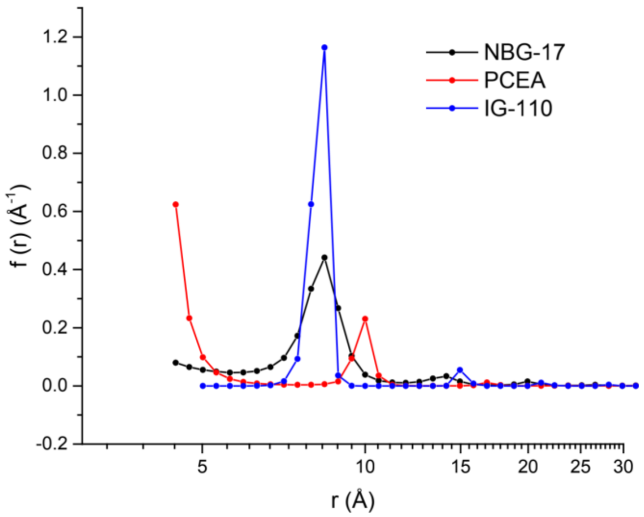
Calculational Description	Calculated Fission Density Ratio	Calc.-Exp. Exp.
100% Graphite	43.2 $\pm$ 1.4	+6.9%
59% Graphite	39.4 $\pm$ 1.2	-2.5%



What is Nuclear Graphite?







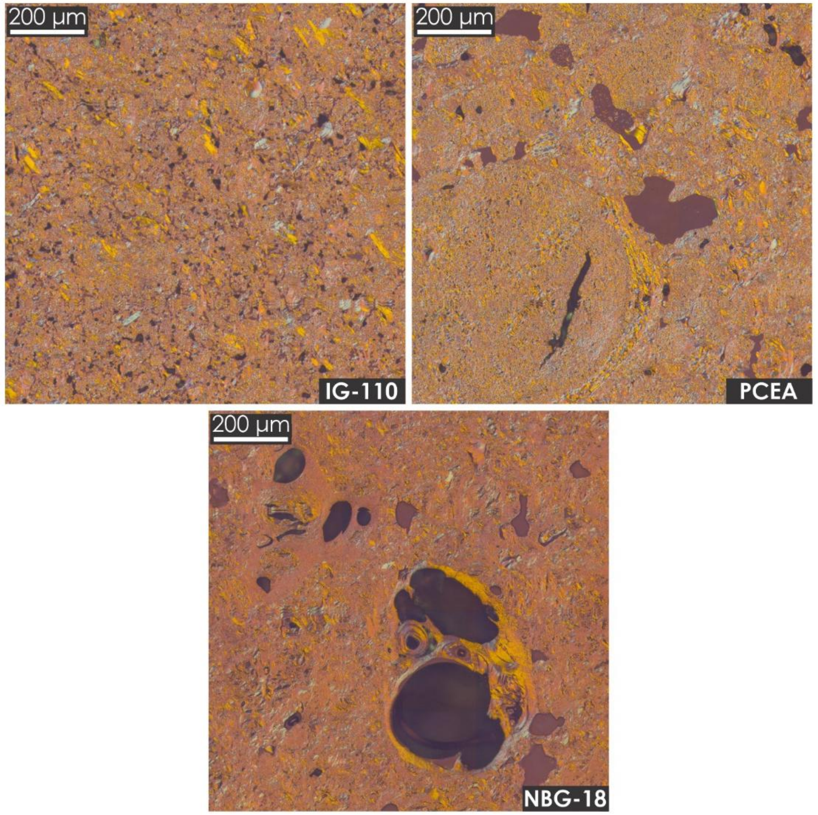
What Does A Neutron See?

What Does A Neutron See?

- Assume a nuclear graphite bulk (or pile)

What Does A Neutron See?

- Assume a nuclear graphite bulk (or pile)



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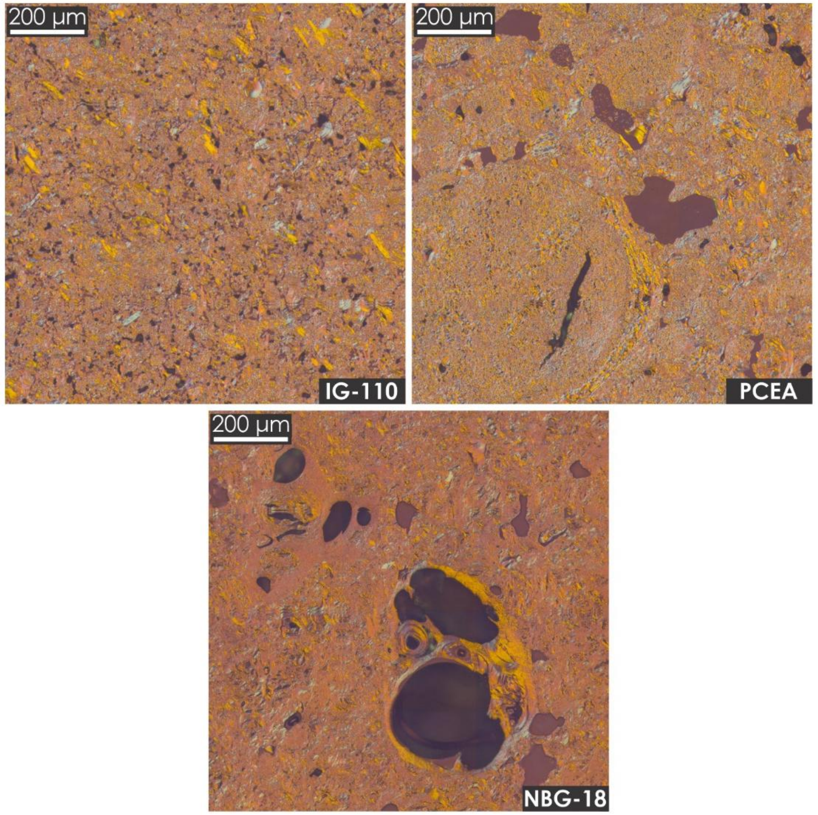
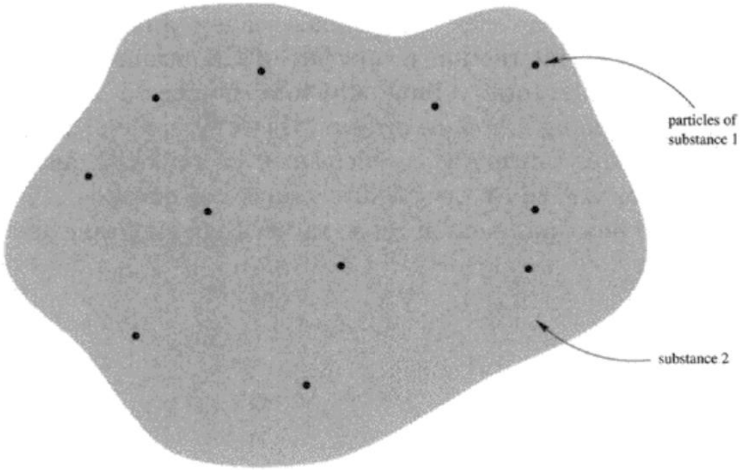
What Does A Neutron See?

- Assume a nuclear graphite bulk (or pile)





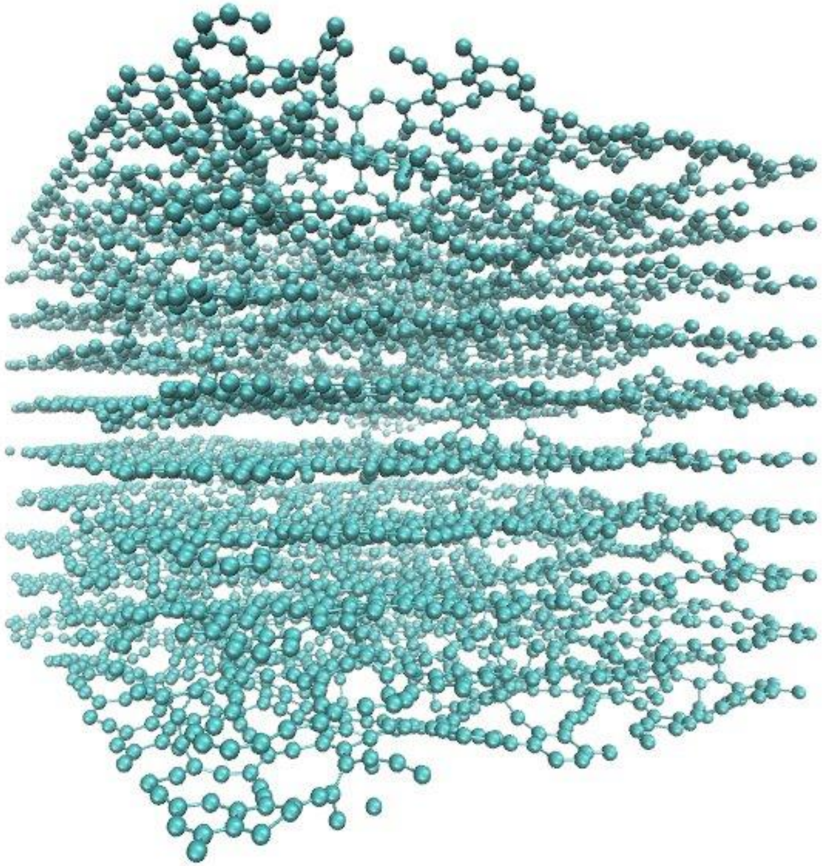
neutronic scale

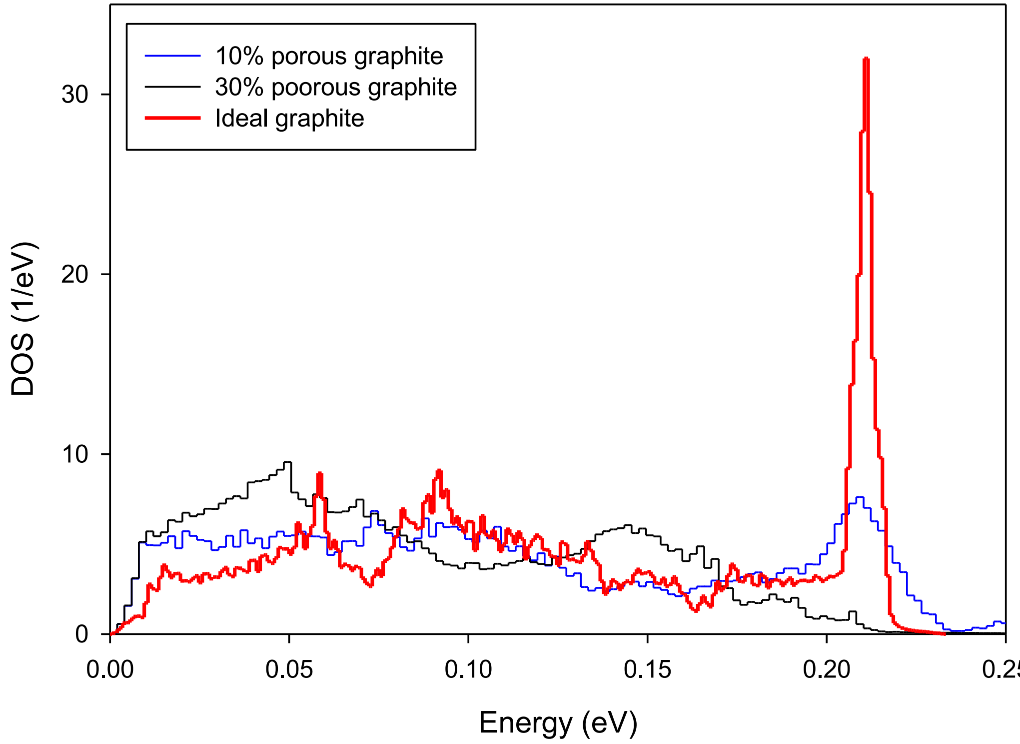
$$\mathbf{mfp} = \frac{\mathbf{1}}{\boldsymbol{\Sigma}} \approx \mathbf{2.5\ cm}$$

Thermalization – Modern History (2000-2012)

- **Porous** homogeneous and uniform model of nuclear graphite model
- **Conserve scattering reaction rate**

Classical MD





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

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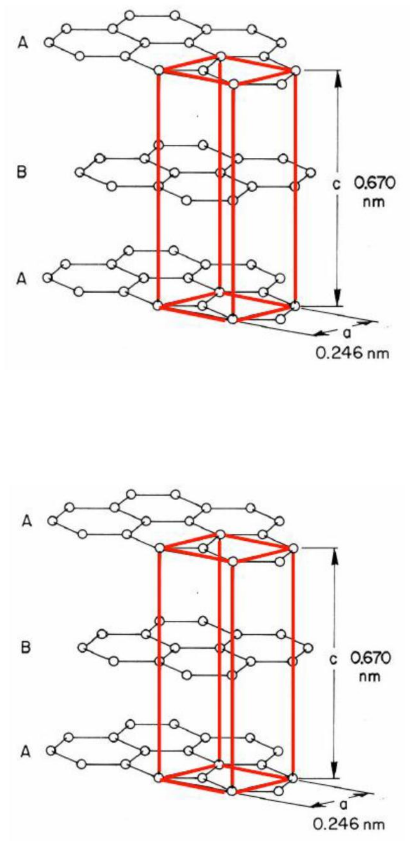
Graphite in ENDF/B-VIII.1

- 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



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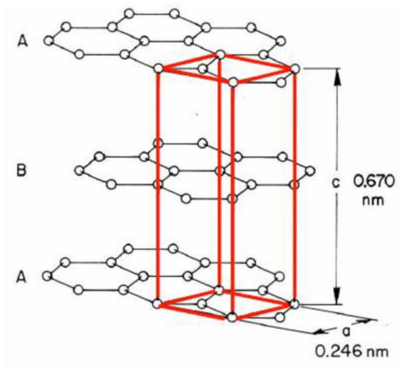
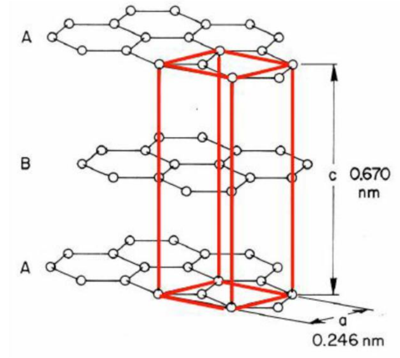
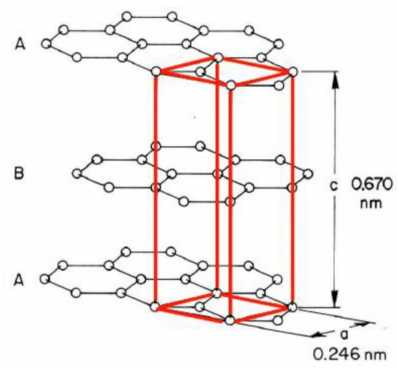
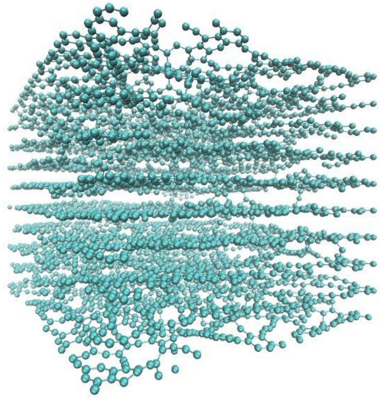
Crystalline





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

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

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

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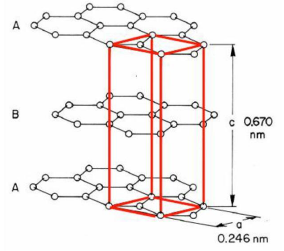
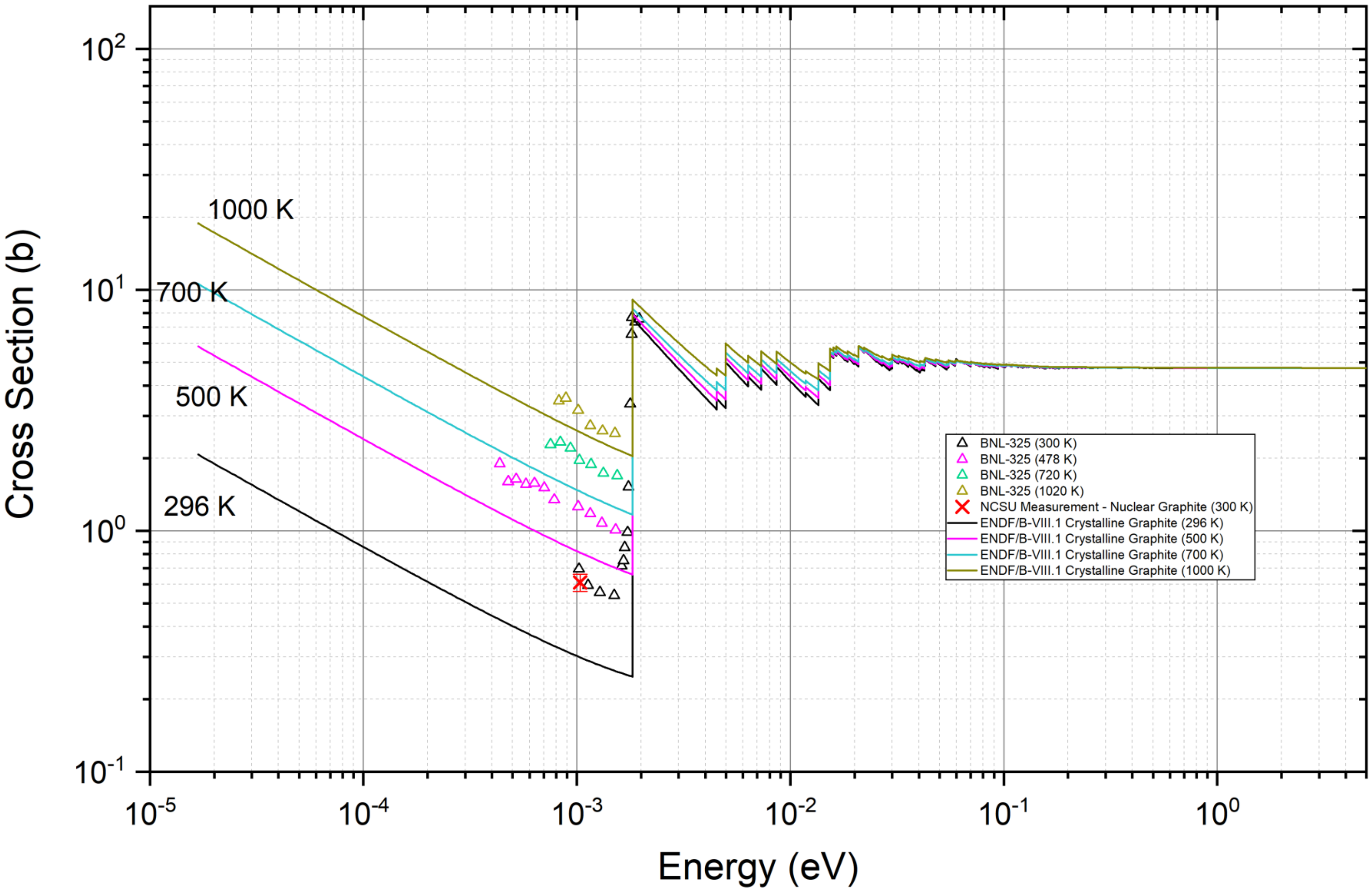
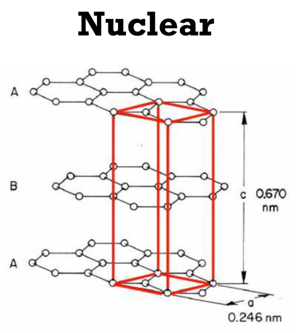
Graphite – Still a Problem!

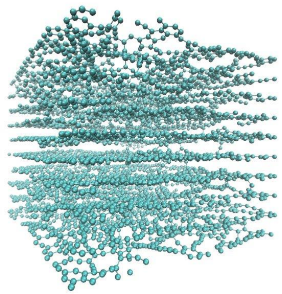
Crystalline

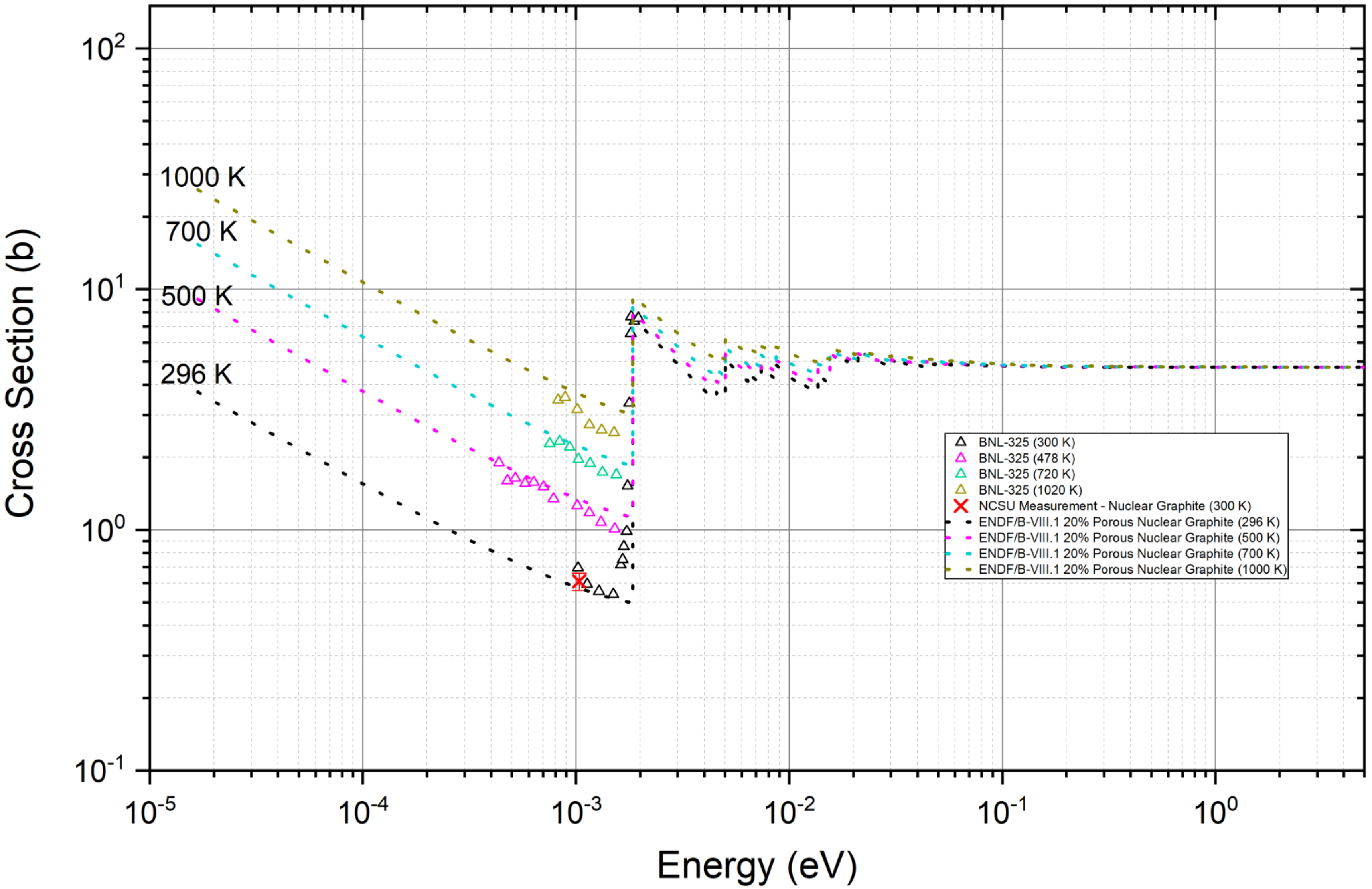




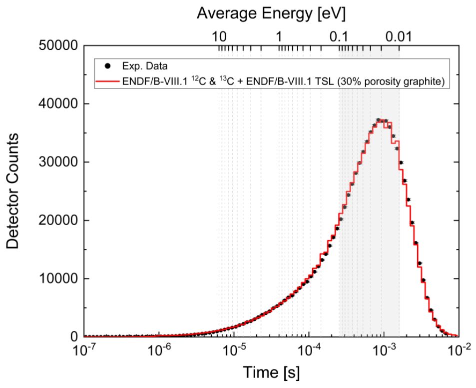
Nuclear Graphite – Much Improved!







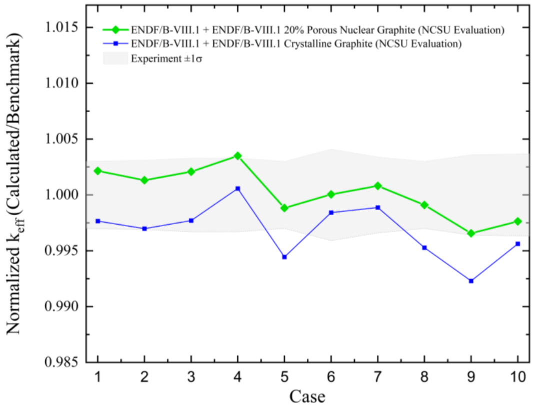
Nuclear Graphite – Much Improved!



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

Density = 1.66 g/cm^3
Porosity ≈ 30%

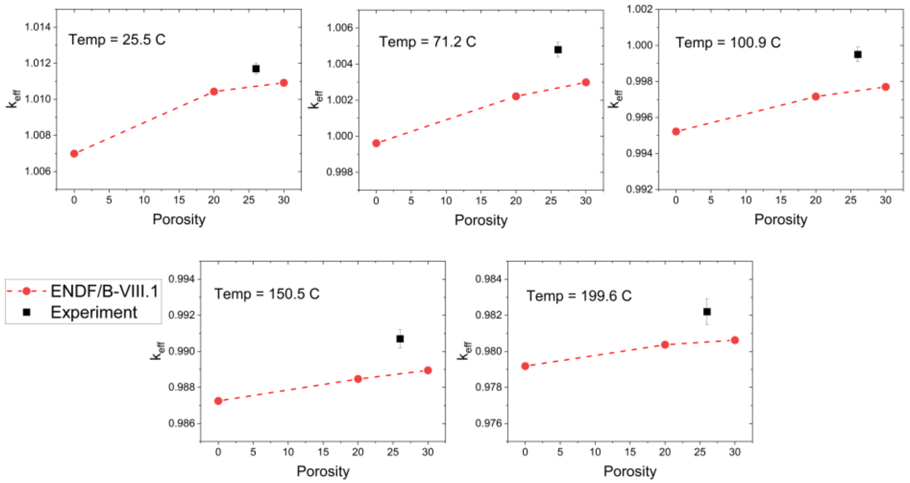
Cross Sections	Mean Absolute Deviation (%)
ENDF/B-VIII.0 + Cry	4.14%
ENDF/B-VIII.1 + Cry	4.09%
ENDF/B-VIII.1 + Cry +S_d	5.01%
ENDF/B-VIII.1+30%	1.79%



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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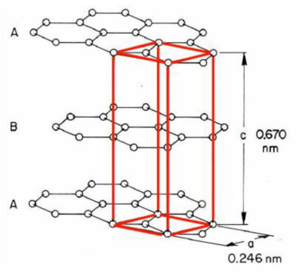
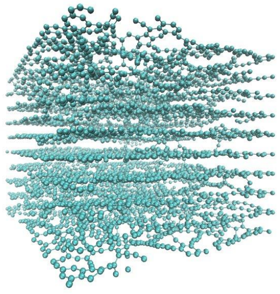
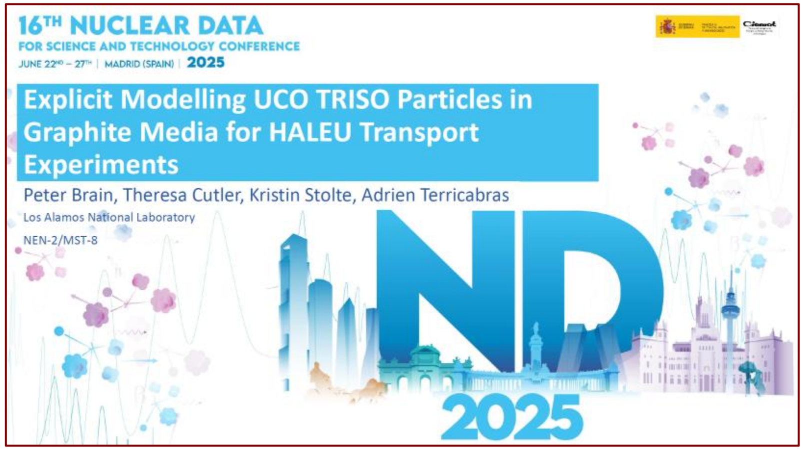
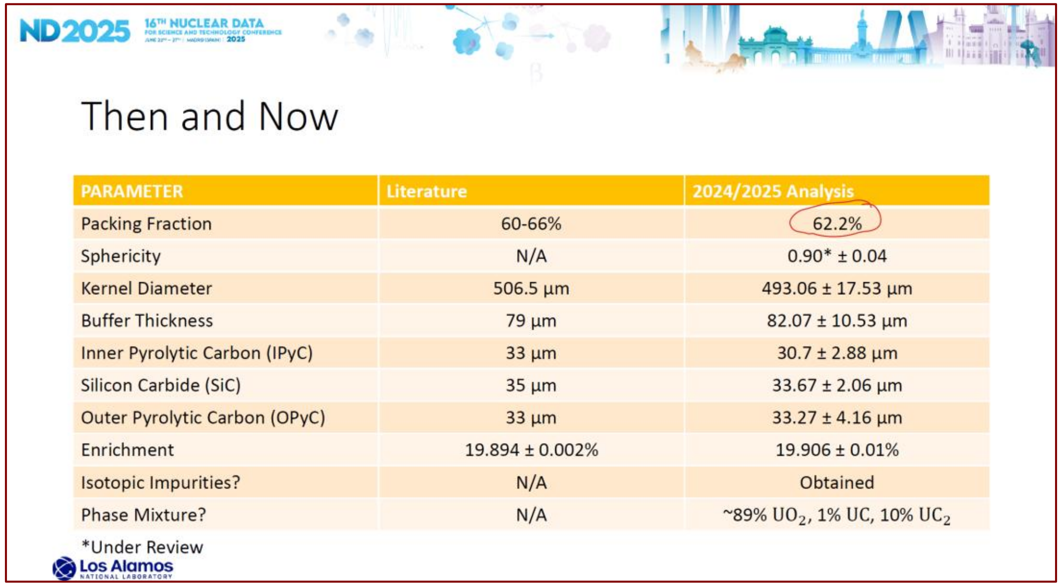
Nuclear Graphite – Current Testing

Nuclear

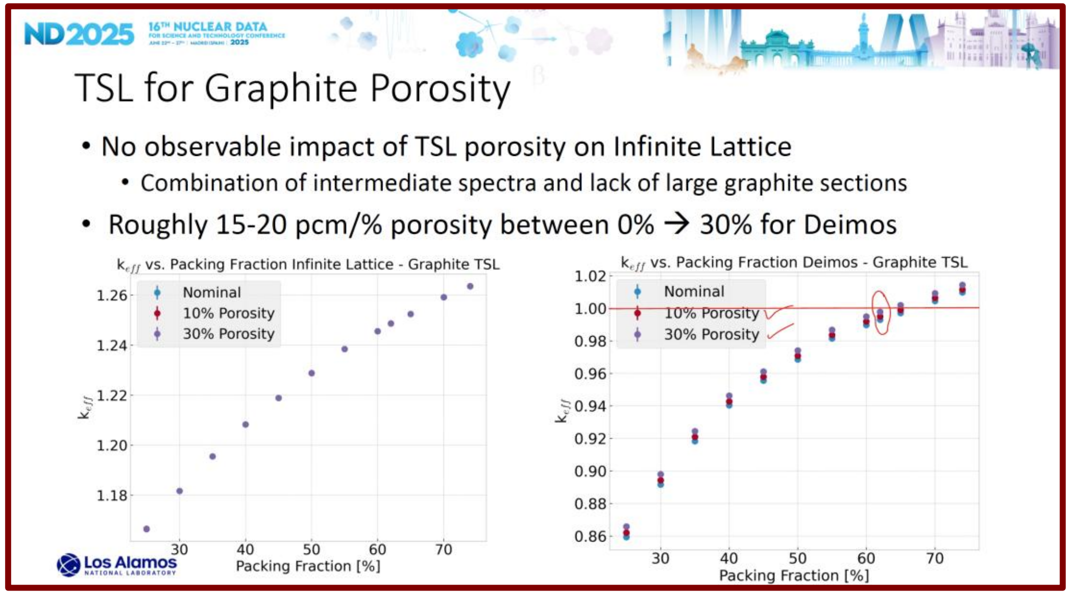








PARAMETER	Literature	2024/2025 Analysis
Packing Fraction	60-66%	62.2%
Sphericity	N/A	0.90* ± 0.04
Kernel Diameter	506.5 μm	493.06 ± 17.53 μm
Buffer Thickness	79 μm	82.07 ± 10.53 μm
Inner Pyrolytic Carbon (IPyC)	33 μm	30.7 ± 2.88 μm
Silicon Carbide (SiC)	35 μm	33.67 ± 2.06 μm
Outer Pyrolytic Carbon (OPyC)	33 μm	33.27 ± 4.16 μm
Enrichment	19.894 ± 0.002%	19.906 ± 0.01%
Isotopic Impurities?	N/A	Obtained
Phase Mixture?	N/A	~89% UO_2, 1% UC, 10% UC_2



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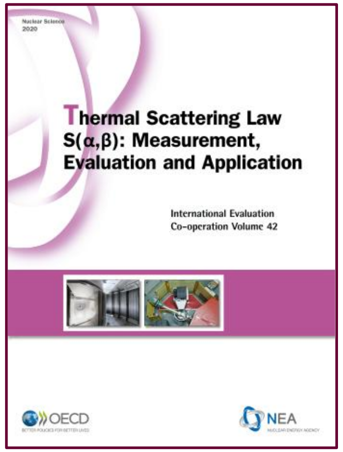
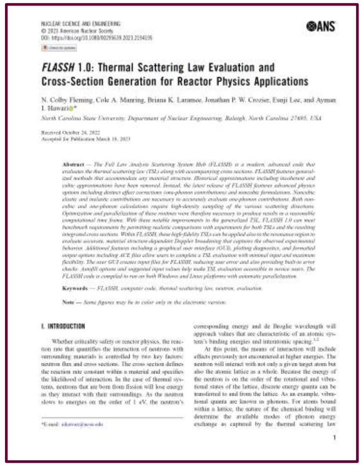
Outcome



ICSBEP Benchmark

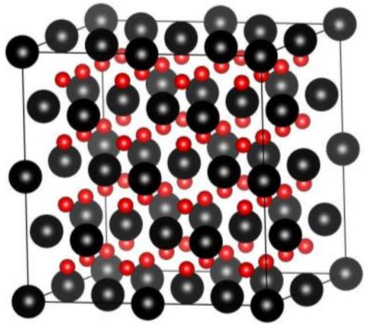


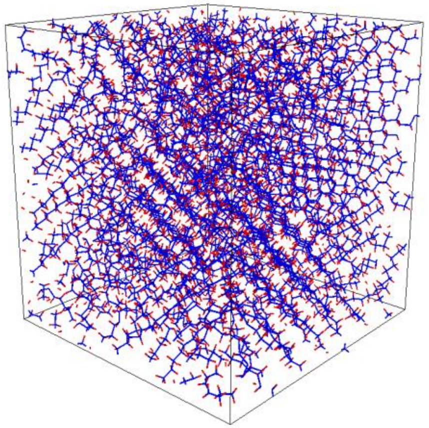
ENDF/B-VIII.1 Paper

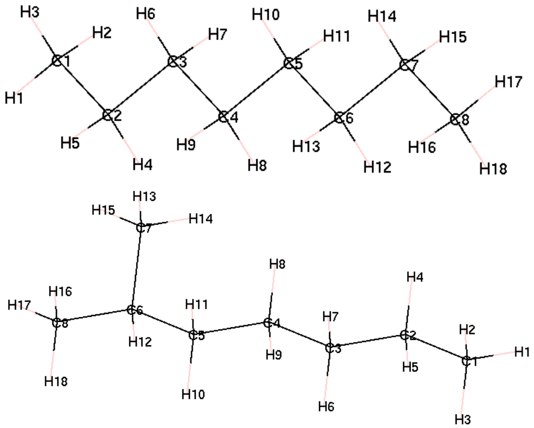
- 66 TSL evaluations were <u>accepted</u> for ENDF/B-VIII.1 for the following materials
  - **Al2O3**, Be-metal, **Be-metal+Sd**, BeO, **FLiBe, CaH2**, CH2, SiC, **UC**, **HF**, **Heavy Paraffinic Oil**, UN, **PuO2**, SiO2, UO2, **U-metal**, Grph-10, **Grph-20P**, Grph-30p, Grph-cryst, **Grph+Sd, Enrichment dependent fuel libraries**
  - **FY 25 : Paraffin, U3Si2, UMo, light Paraffinic oil**

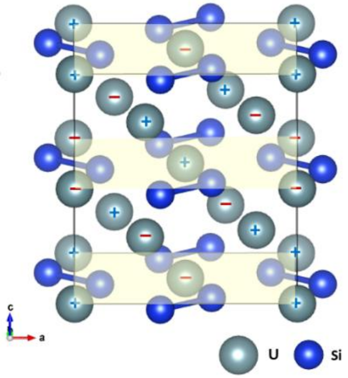
TSLs & Beyond

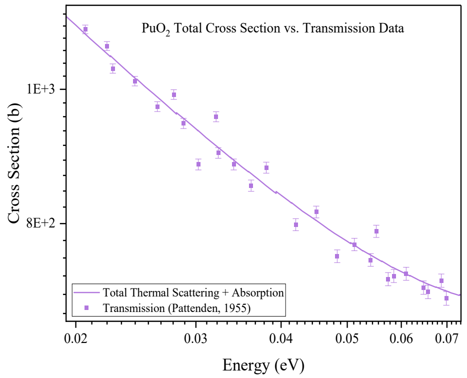
Thermalization – Modern History (2012-Present)

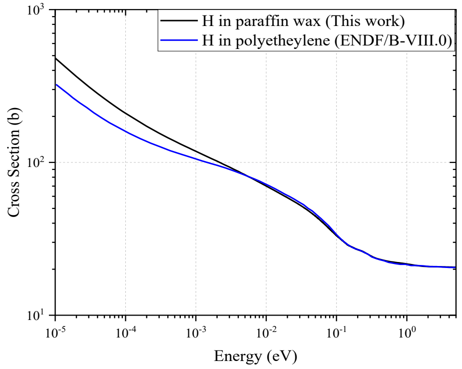


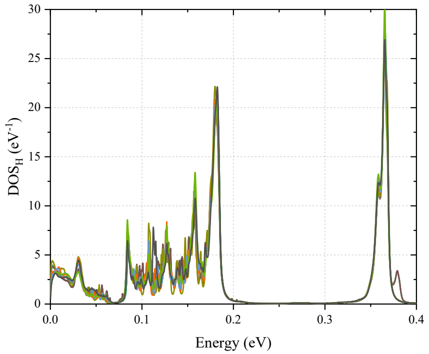


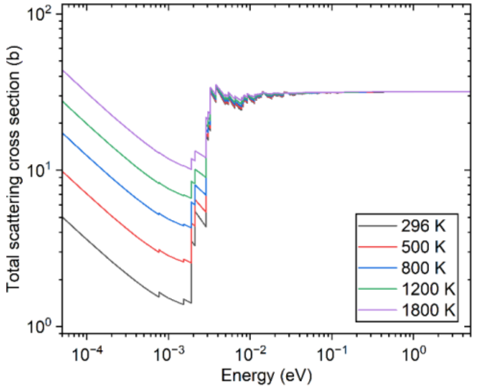












Plutonium dioxide

Paraffin

Paraffinic oil

Uranium Silicide

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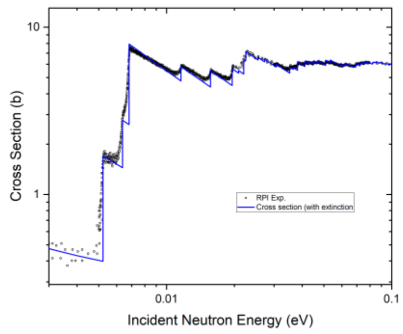
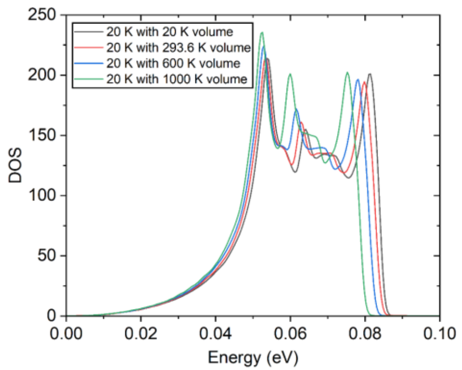
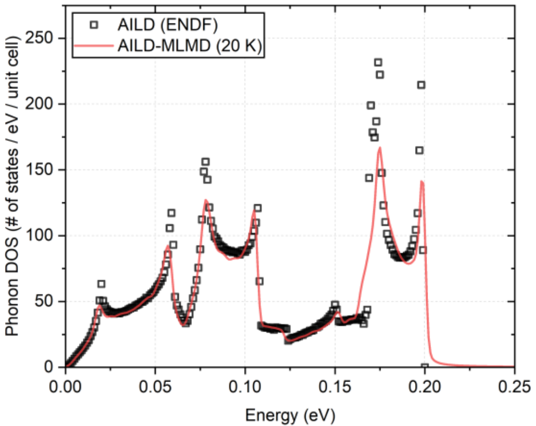
TSL State-of-the-Art

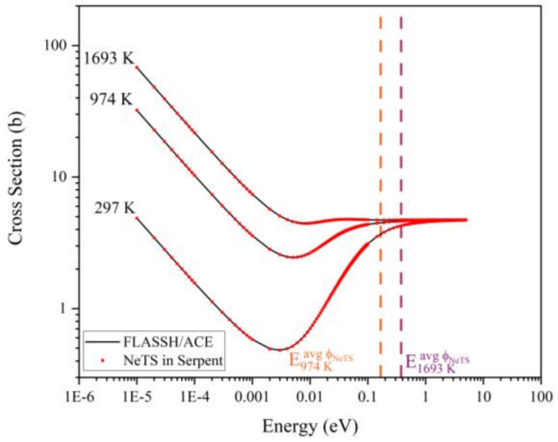
- Everything developed for graphite applies to all other materials
  - Atomistic simulation methods (AILD and MD)
    - Continue to evolve
    - Enabled ML
  - Advanced TSL evaluation methods
    - ***FLASSH***
  - Holistic experimental/measurement approach
    - don't pick and choose!

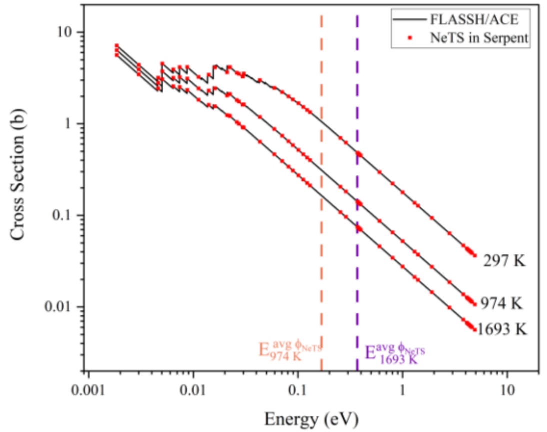


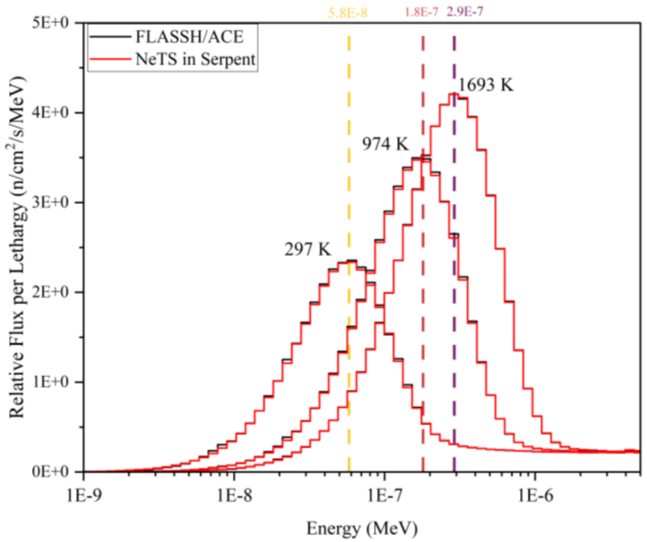




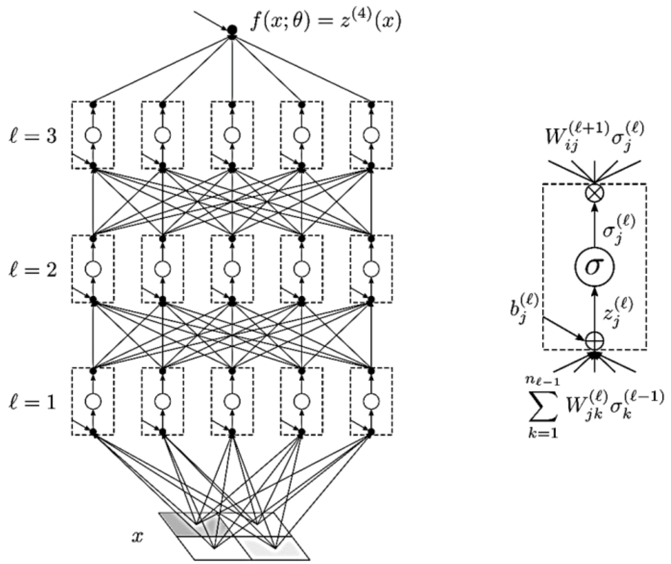
Thermalization – The Future

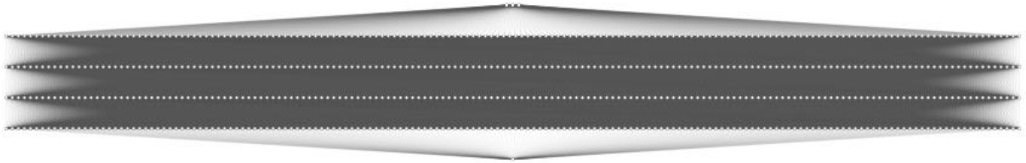






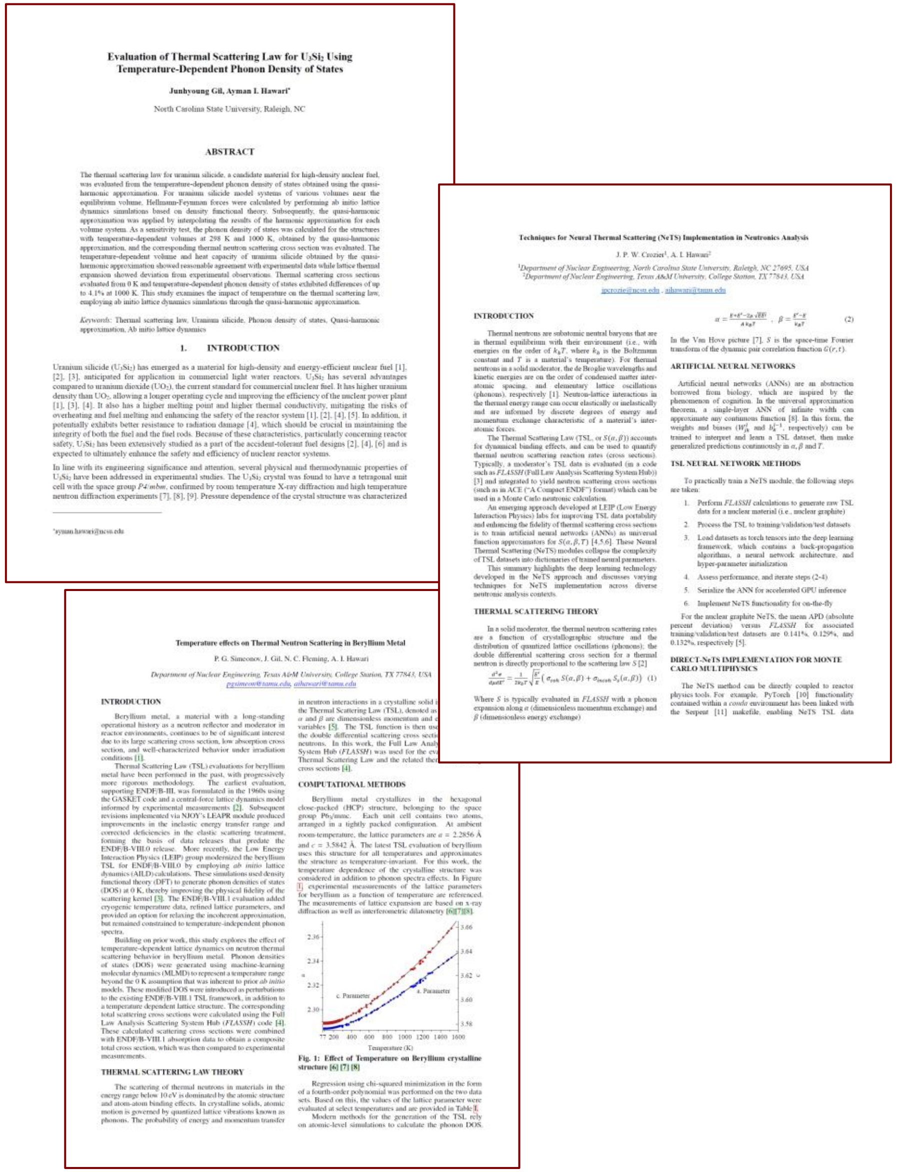
NeTS
working with
Monte Carlo-CFD
Multi-Physics
Neutron-by-neutron

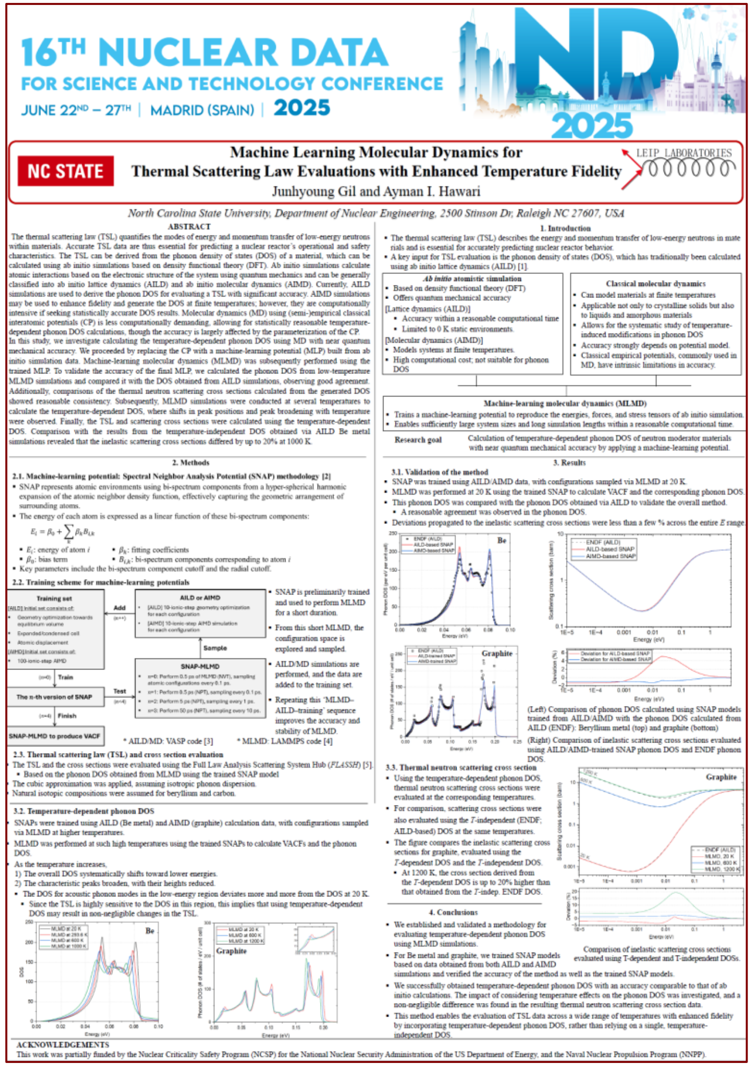


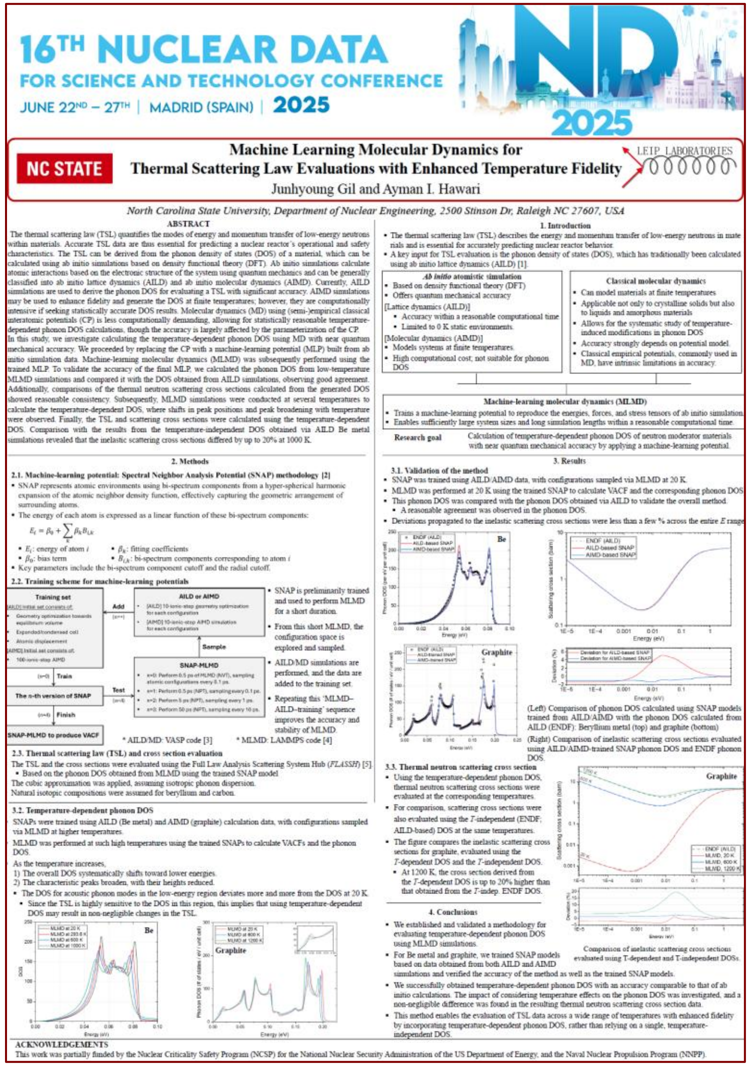


Framework	Data Storage
ACE	~ 30 MB / Temperature
NeTS	~ 340 KB for *200*+ Temperatures

Thermalization – The Future





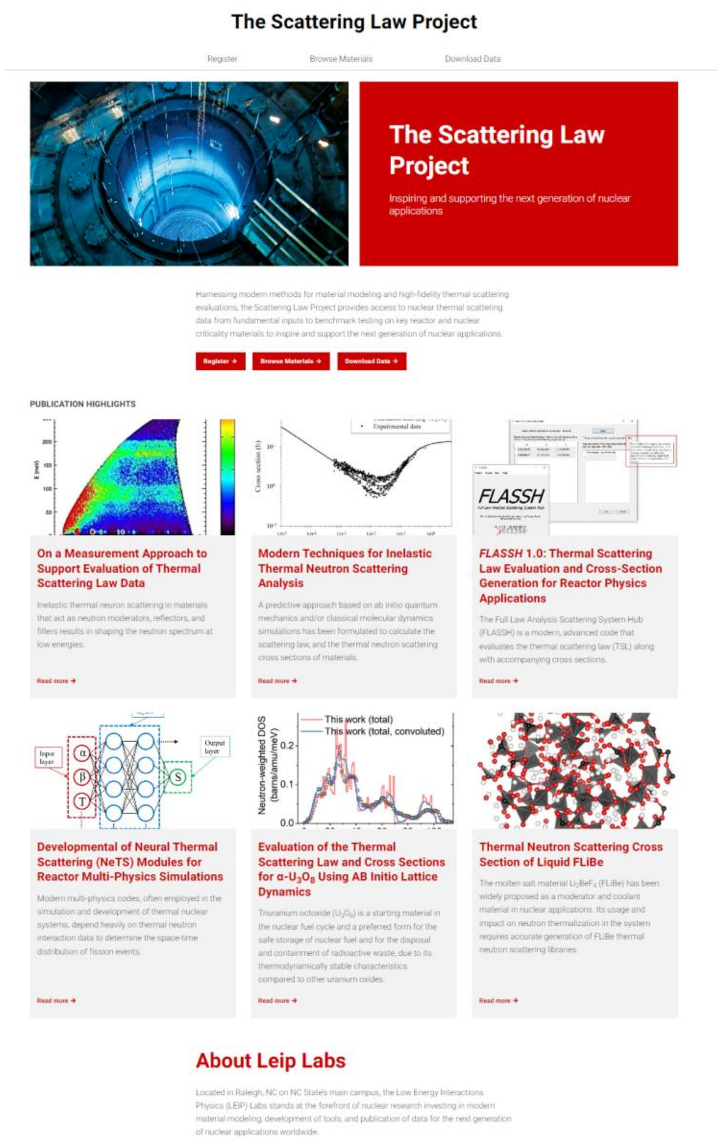


Summary

- Advanced nuclear reactors represent an excellent opportunity for innovation
  - Revived our knowledge of fundamental radiation interaction physics
  - Allowed the introduction of modern methods
    - computational and experimental
- Integration of computation and experimentation to see the unseen
  - Hybrid and adaptive
- Innovation is great for mentoring the next generation of nuclear engineering and science experts

TSL Project

- TSL Project website will be launched at **TAMU** during Fall 2025
- Will be include all LEIP work
  - TSL files (File 7)
  - ACE files
  - NeTS
  - Benchmark and validation work
  - All published work (papers, reports, PhD and MS theses, etc.)
- Will be open to all
  - Request feedback from users

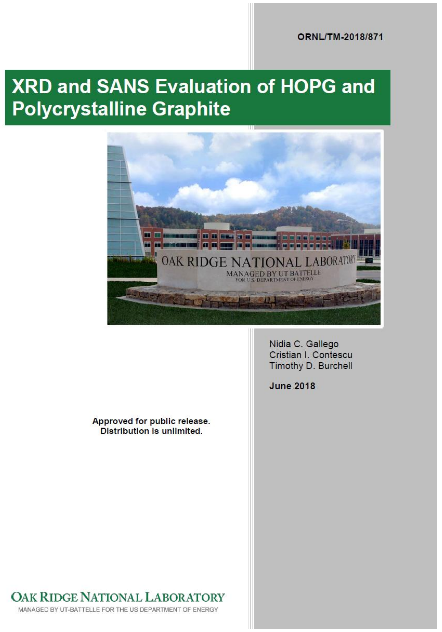


Thank You

?

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
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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

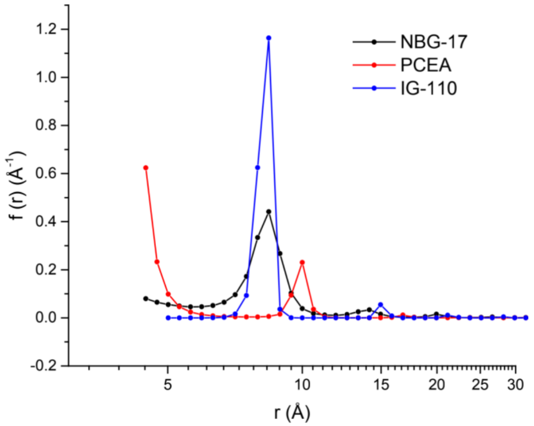
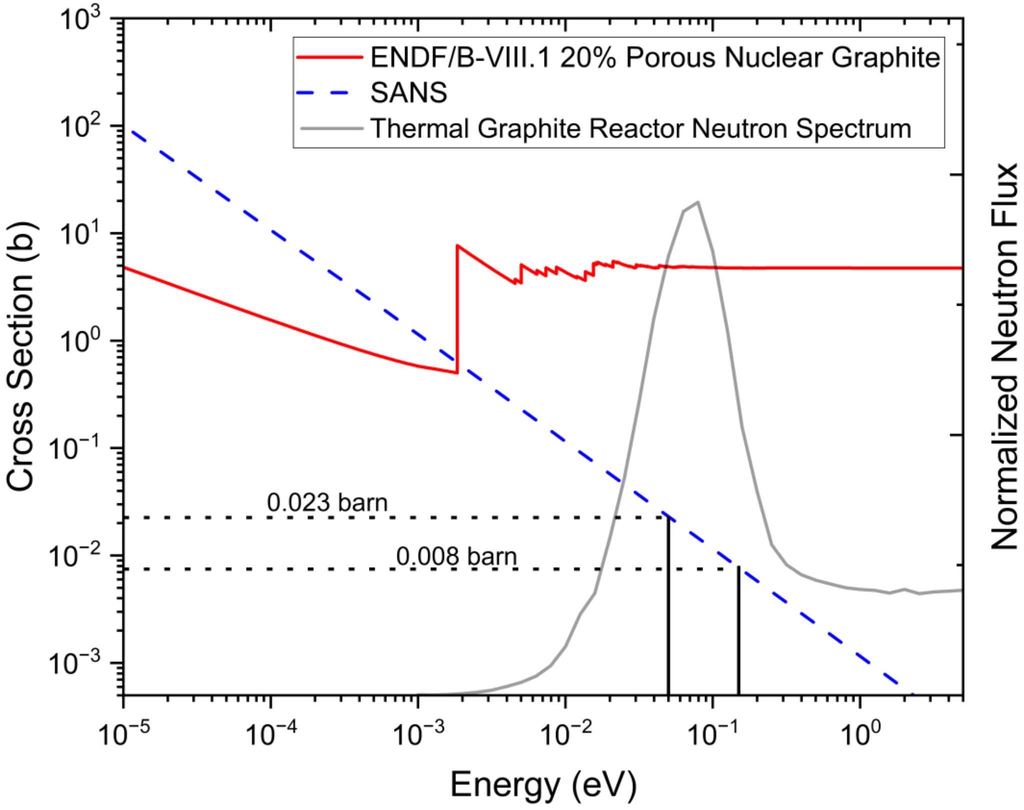


Figure 18 (b) from Reference

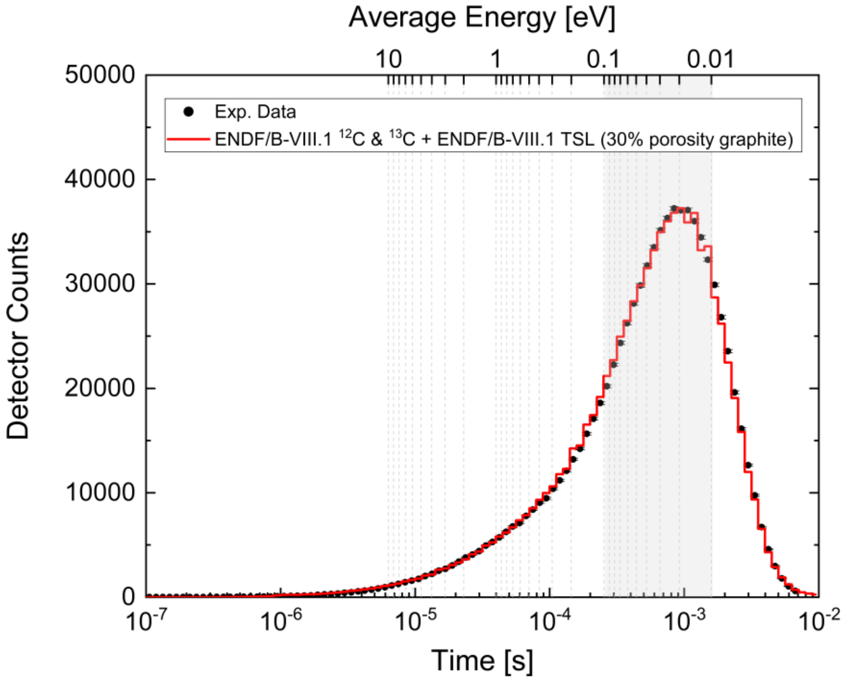


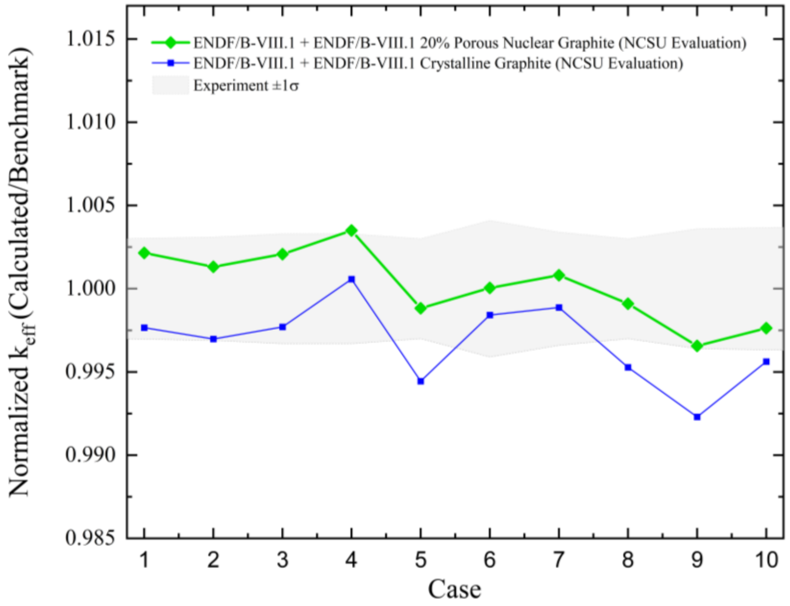
Ref. 2 - Petriw, et.al., “Porosity effects on the neutron total cross section of graphite”

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



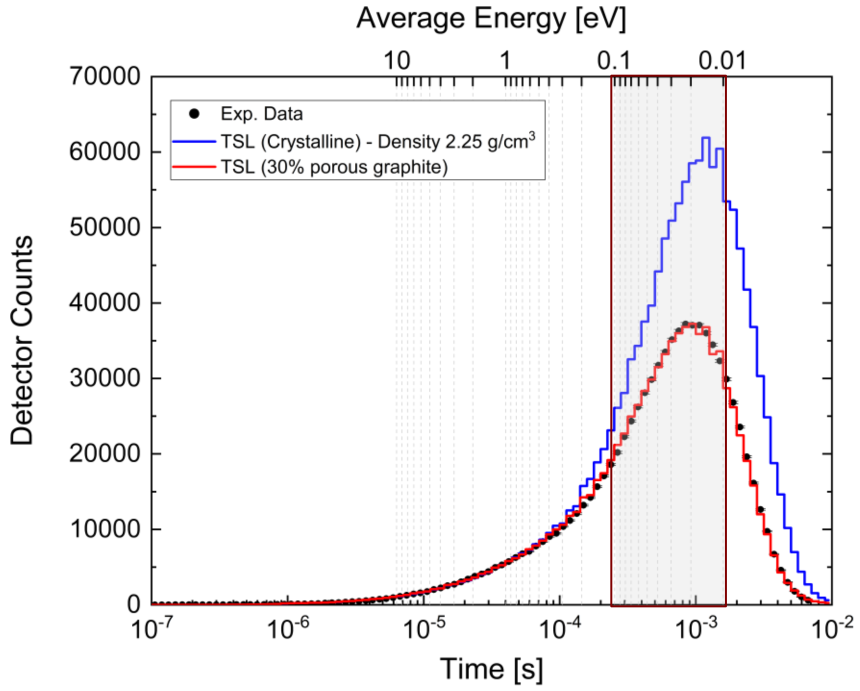


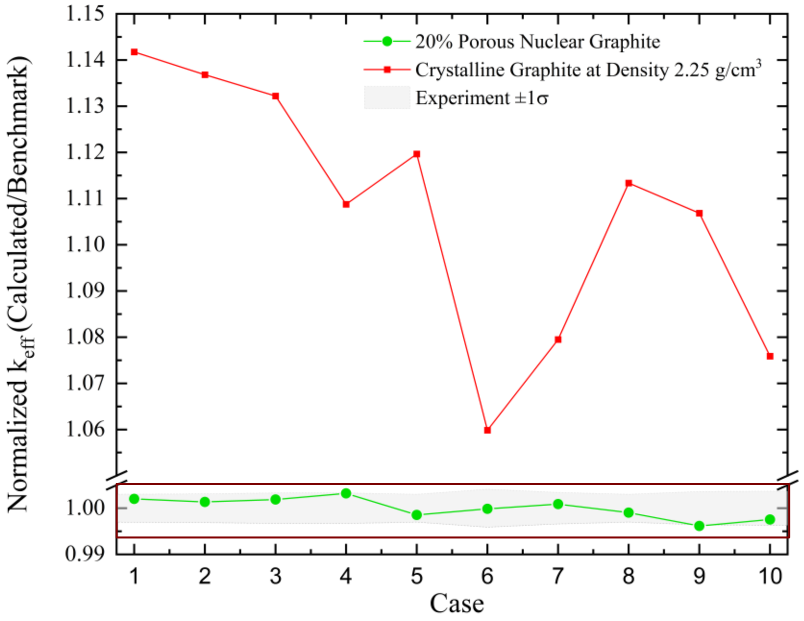
LEIP LABORATORIES

Graphite Density

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

<u>Self-consistent</u> phonon DOS and density

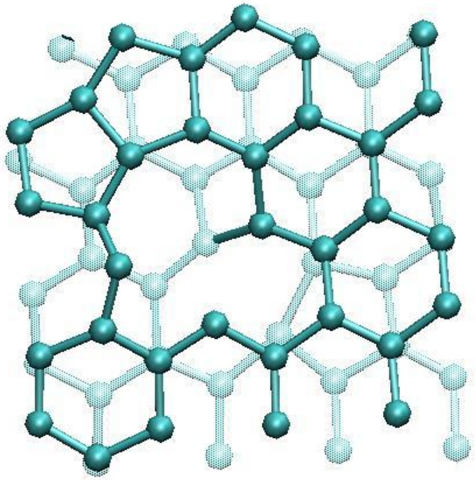




Radiation Damage
Interplanar Di-vacancy Defect

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

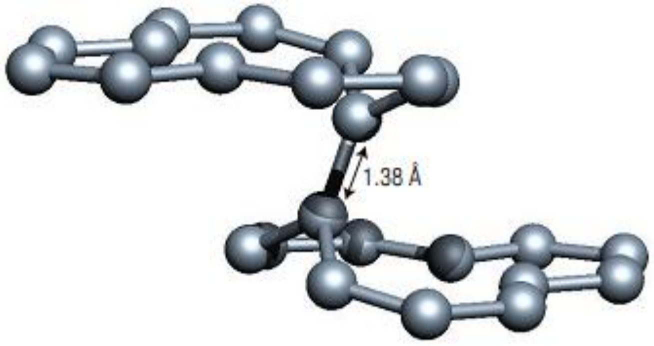
Nuclear Engineering

Raleigh, North Carolina

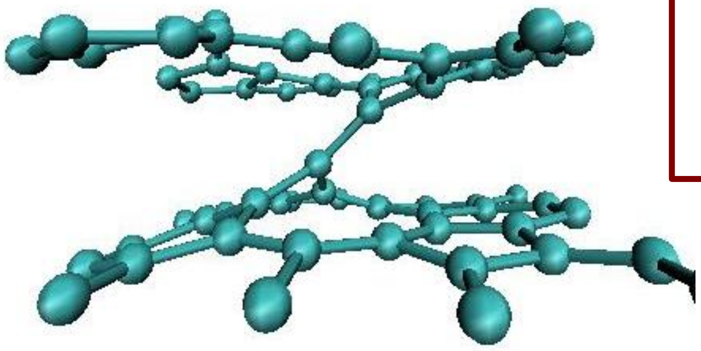
2010

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)

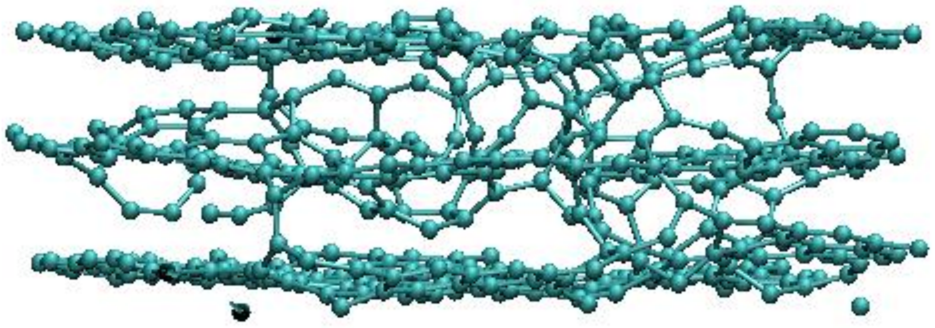


LEIP MD analysis

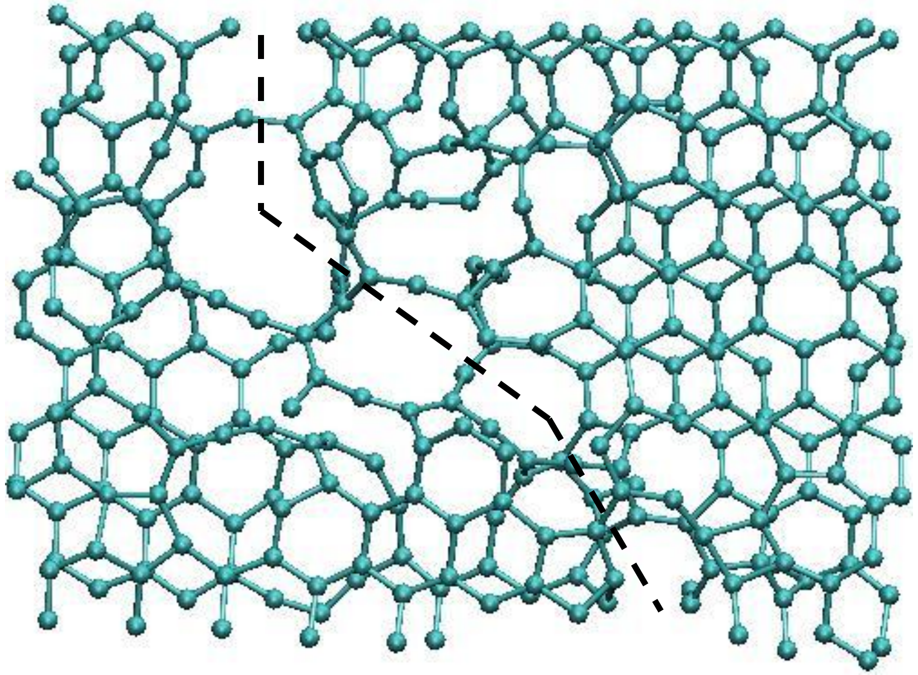
LEIP LABORATORIES

Radiation Damage
Interplanar Crosslinking

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



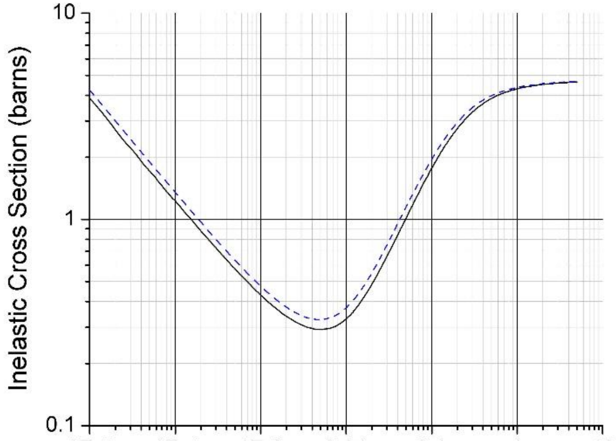
→ Individual point defects become less distinguishable



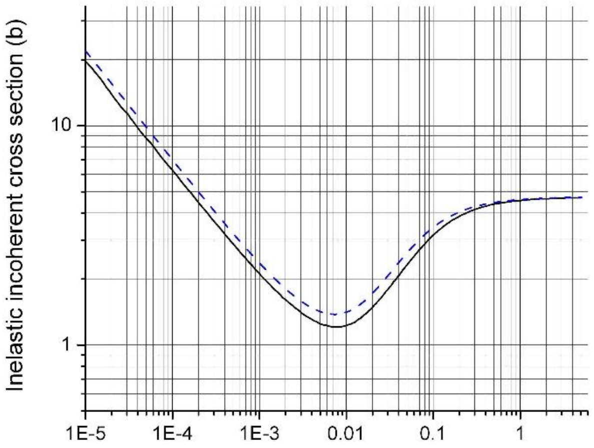
Radiation Damage
Effect of Temperature

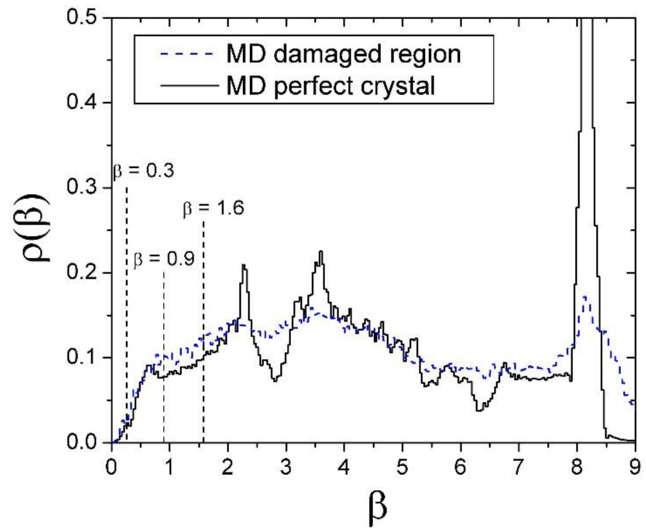
Cascade buildup

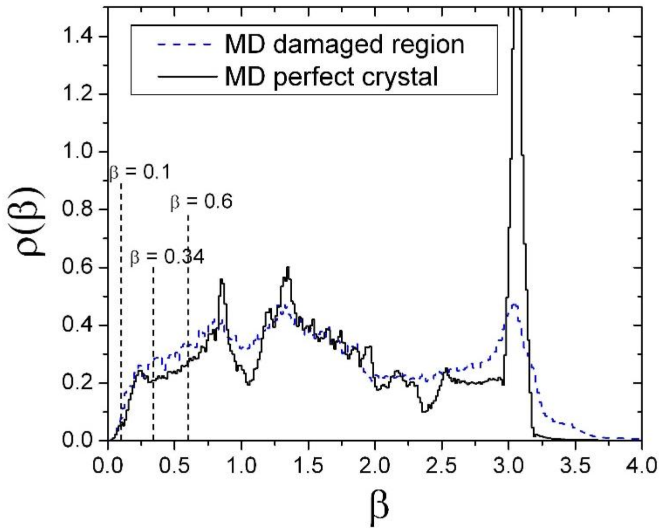
300K



800K







Porosity effect continues to dominate