

Porosity in nuclear graphite and its impact on nuclear reactor science and criticality safety applications[★]

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Abstract

Porosity in nuclear-grade graphite significantly influences its low-energy neutron scattering, yet its effect on underlying phonon properties remains debated. This work integrates inelastic and small-angle neutron scattering (INS/SANS) experiments, advanced atomistic simulations with a novel machine-learned potential (DeepMD), total cross-section measurements, and neutronics calculations (SCALE, MCNP, OpenMC) to investigate porosity's impact on neutron thermalization. INS measurements on diverse graphite grades reveal no discernible porosity effect on phonon spectra, which align with crystalline graphite. Conversely, total cross-section data below ≈ 10 meV show increased scattering attributable to SANS. Our DeepMD simulations demonstrate that realistic micropores do not distort phonon spectra, challenging the assumptions in current ENDF/B-VIII.1 porosity thermal scattering laws (TSLs). These TSLs, based on random atom removal, produce unphysical phonon spectra and inflate inelastic cross-sections. Augmenting a crystalline TSL with a SANS component accurately captures experimental total cross-sections. Neutronics benchmarks (ICS-BEP/IRPhE) show ENDF porosity TSLs unphysically increase neutron multiplication factor, k_{eff} . Crucially, incorporating SANS physics (NCrystal/OpenMC) indicates accurately modeled porosity negligibly affects k_{eff} , reactor physics, or criticality safety.

Keywords: graphite, porosity, nuclear data, thermal scattering law, inelastic neutron scattering, small-angle neutron scattering, critical benchmarks

1. Introduction

Graphite has played a pivotal role in nuclear reactor technology since the very first reactor (Chicago Pile-1 in 1942) used graphite blocks as a neutron moderator. Since then, many reactors worldwide have relied on graphite as a moderator or reflector, from early plutonium production piles and gas-cooled power reactors to research reactors [1, 2]. Today, graphite remains a key material in several operating reactors, such as the British advanced gas-cooled reactor (AGR) and Russian RBMK (Reaktor Bolshoy Moshchnosty Kanalny, high-power channel reactor) designs. It is also crucial for next-generation systems like very high-temperature reactors (VHTRs) and molten salt reactors [3]. High-purity graphite (<50 ppm impurities) efficiently slows down neutrons due to its high elastic scattering

and low absorption cross-sections, with its low atomic mass allowing for effective moderation and enabling neutrons to lose energy with relatively small number of elastic collisions and reach thermal equilibrium. Additionally, graphite's outstanding thermomechanical properties—including high thermal conductivity, low thermal expansion, and exceptional thermal shock resistance—make it both a robust neutron moderator and a durable structural material in reactor cores. Structurally, it is strong yet lightweight, and it can be readily fabricated into large, complex shapes required for core components (e.g., moderator blocks, fuel channel bricks, reflectors).

Nuclear graphite is an artificial, polycrystalline form of carbon with a complex microstructure. During production, filler particles (calcined petroleum or pitch coke) are bonded with carbonized pitch and then graphitized at high temperatures. Two primary mechanisms create porosity in this process: gas evolution pores in the binder phase and Mrozowski cracks in the filler grains and binder phase. Volatile gases released from the pitch binder during baking create open pores as they escape, leaving voids in the carbon matrix [4]. Meanwhile, the graphite crystallites (especially within coke filler particles) undergo anisotropic thermal contraction upon cooling, leading to internal stress cracks known as Mrozowski cracks [4]. These

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cracks form predominantly along the graphite *c*-axis, where thermal contraction is greatest, and are a key source of microscopic porosity in the filler phase [5].

The overall porosity of nuclear graphite is typically on the order of 15–25% by volume [2], although it varies with the graphite grade and manufacturing route. Fine-grained, isostatically pressed graphite grades (such as IG-110) typically have uniformly distributed and predominantly open porosity. In contrast, coarser-grained molded grades generally exhibit a higher proportion of closed porosity within larger filler particles [2]. Arregui-Mena et al. [2] noted that porosity must be characterized across multiple length scales in filler and binder regions to understand graphite’s bulk properties. In essence, the pore structure is a hierarchically organized network: large voids (tens to hundreds of microns) often reside in the binder-rich regions, whereas many microcracks and micropores (down to nanometers) permeate the graphite crystallites. Small-angle neutron scattering (SANS) experiments reveal that graphite’s porosity follows a fractal-like distribution over at least ≈ 0.2 nm to 300 nm, indicating a continuous population of nanometer-scale pores (primarily Mrozowski cracks) within that range [4]. Notably, the graphite matrix around these pores remains predominantly crystalline. This observation is consistent with the fact that nuclear graphite is typically fully graphitized during manufacturing. It is widely accepted that porosity has minimal or negligible impact on the vibrational properties (phonon spectra) of nuclear graphite, given that the carbon atoms remain crystallographically ordered around pore surfaces, maintaining their vibrational modes close to crystalline graphite [6, 7].

Despite its minimal impact on phonon spectra, porosity substantially influences neutron scattering at thermal energies via the SANS effect. SANS primarily arises from elastic neutron scattering at small angles off numerous interfaces between the graphite matrix and pores. In total cross section measurements, this interaction removes neutrons from the transmitted beam without significant absorption or large-angle scattering [8, 9, 10]. Consequently, total neutron cross sections measured in porous nuclear graphite are significantly higher at thermal and sub-thermal neutron energies compared to predictions based on ideal crystalline models that include only elastic and inelastic scattering [11, 12].

Current thermal scattering laws (TSLs) for nuclear graphite in the ENDF/B-VIII.1 evaluated nuclear data library have attempted to incorporate porosity effects by randomly removing atoms from an ideal crystalline structure (large supercell) in molecular dynamics (MD) simulations [13]. This simplified approach introduces nonphysical disruptions (point and small cluster defects) to the atomic arrangement, resulting in distorted phonon spectra and, subsequently, incorrect thermal neutron scattering cross sections. Our experimental measurements and computational analyses, documented herein, demonstrate that this approach misrepresents the true physical impact of porosity on neutron scattering behavior.

In this paper, we present an experimental and computational investigation to accurately quantify the influence of porosity on the neutron scattering properties of nuclear graphite. We report inelastic neutron scattering (INS) measurements performed

using the ARCS and VISION spectrometers; these measurements reveal consistent phonon spectra across graphite samples with varying porosities, thereby confirming that porosity does not appreciably alter atomic vibrational dynamics. Additionally, we used SANS data obtained from the general-purpose small-angle neutron scattering (GP-SANS) instrument at the High Flux Isotope Reactor (HFIR) to characterize nano-pore-size distributions across multiple graphite grades. To complement findings from INS measurements, we developed a machine-learned potential based on *ab initio* molecular dynamics (AIMD) simulations utilizing VASP [14, 15, 16], LAMMPS [17], and DeepMD [18, 19] frameworks. This model is shown to accurately capture the graphite structure and to enable realistic porosity modeling via microporous configurations. Moreover, existing total cross section (transmission) measurements were analyzed, and new ones were measured in this work for some of the samples used in the INS measurements. Finally, we investigated the consequences of porosity-induced inaccuracies in current TSLs, employing a combination of integral benchmark evaluations from criticality safety benchmarks (ICSBEP) and reactor physics experiments (IRPhE). We demonstrate that improperly modeled porosity significantly biases criticality calculations. This bias artificially overestimates the effective neutron multiplication factor (k_{eff}) and can thereby affect reactor safety and design assessments.

2. Experimental and Computational Details

2.1. Inelastic neutron scattering measurements

Thermal neutron INS experiments were carried out to probe the phonon spectra of various nuclear graphite samples with different porosities. We utilized two types of time-of-flight neutron spectrometers at ORNL: (a) the ARCS and SEQUOIA direct-geometry spectrometers [20, 21, 22], and (b) the VISION indirect-geometry spectrometer [23].

For direct-geometry spectrometers, choppers select the incident neutron energy, and time-of-flight methods—based on the known detector distance—measure the final neutron energy, allowing the energy transfer to be determined. The ARCS instrument is optimized for experiments that require high neutron flux over a wide Q -range and offers a moderate energy resolution but extensive angular coverage. ARCS directly measures $S(Q, \omega)$, the dynamic structure factor that describes the probability of scattering events transferring momentum Q and energy ω . Owing to its large Q and angular coverage, ARCS is especially well suited for deriving phonon spectra under the *incoherent* approximation. Using this framework, the phonon spectra can be extracted from $S(Q, \omega)$ using the following relation [24]:

$$S(Q, \omega) = e^{-2W(Q)} \frac{Q^2 \hbar}{2M\omega} \left[n(\omega) + 1 \right] g(\omega), \quad (1)$$

where $e^{-2W(Q)}$ is the Debye–Waller factor, Q is the momentum transfer, M is the atomic mass, ω is the phonon frequency, $n(\omega)$ is the Bose–Einstein occupation factor (equal to $\frac{1}{e^{\hbar\omega/k_B T} - 1}$), $\hbar$ is the reduced Planck’s constant, k_B is the Boltzmann constant,

and $g(\omega)$ is the phonon density of states (DOS, phonon spectra). SEQUOIA provides higher energy resolution than ARCS, but ARCS's broader Q coverage makes it advantageous for efficiently collecting phonon DOS data.

In indirect-geometry spectrometers, the incident neutron beam is “white”, that is, it covers a broad energy range, up to the maximum set by a T0 and bandwidth chopper. After scattering from the sample, neutrons enter crystal analyzers—pyrolytic graphite, in the case of VISION—which fix the final neutron energy by Bragg reflection. The neutrons subsequently pass through a 20 K beryllium filter that removes higher-order reflections from the graphite analyzers. This step often produces a sharp feature at around 11–12 meV (the so-called “ λ_2 peak”) in VISION measurements. The time-of-flight method, along with known flight path distances, enables the determination of the incident energy and thus energy transfer. With this setup, instruments like VISION maintain an approximately constant fractional energy resolution (1–1.5%) across the 2–1000 meV energy range. The analyzers are arranged in forward (low Q) and backward (high Q) scattering geometries. A comparison of the Q – ω trajectories between direct- and indirect-geometry spectrometers is shown in Figure 1.

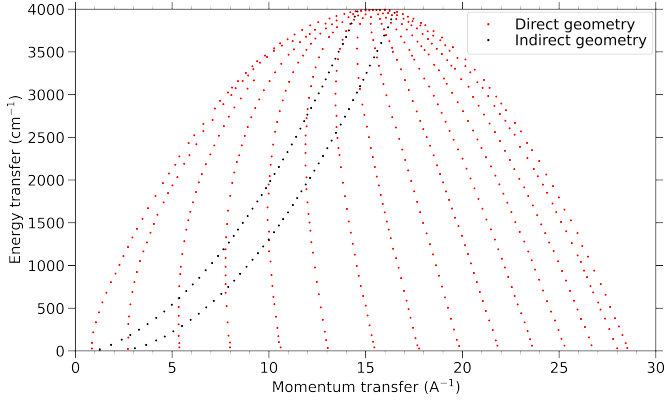


Figure 1: The comparison of the trajectories in Q and ω space between direct- and indirect-geometry spectrometers.

As stated previously, the measured quantity at the VISION instrument is $S(Q, \omega)$ along two trajectories in Q – ω space, albeit with higher energy resolution than that at ARCS. This improved resolution is essential for validating computed quantities such as $S(Q, \omega)$. In the coherent one-phonon approximation, $S(Q, \omega)$ can be calculated as [25]

$$S_{\text{coh}\pm 1}(\mathbf{Q}, \omega) = \frac{1}{2N} \sum_s \sum_{\tau} \frac{1}{\omega_s} \left| \sum_d \frac{b_d}{\sqrt{M_d}} \exp(-W_d) \exp(i\mathbf{Q} \cdot \mathbf{r}_d) (\mathbf{Q} \cdot \mathbf{e}_{ds}) \right|^2 \times \left(n_s + \frac{1}{2} \pm \frac{1}{2} \right) \delta(\omega \pm \omega_s) \delta(\mathbf{Q} \pm \mathbf{q} - \tau), \quad (2)$$

where N is the number of atoms in the system, ω_s is the phonon frequency of mode s , M_d is the mass of atom d , b_d is the scattering length, $\exp(-W_d)$ is the Debye–Waller factor, $\mathbf{r}_d$ is the atom position, $\mathbf{e}_{ds}$ is the polarization vector, and n_s is the Bose–Einstein occupation factor of mode s . The wave vector of the

phonon mode is $\mathbf{q}$, and τ is the corresponding reciprocal lattice vector. Polarization vectors and other necessary quantities, including the phonon spectra (in the incoherent approximation), can be computed via the finite-displacement method as implemented in the Phonopy code [26, 27]. VASP code was utilized for calculating inter-atomic forces known as the Hellmann–Feynman forces [28]. The coherent one-phonon approximation for $S(Q, \omega)$, using the same Q – ω trajectory as that at VISION and convolved with its instrumental resolution, can be carried out using OCLIMAX [29], which essentially serves as a digital twin of the VISION instrument.

2.2. Atomistic modeling and simulations

Our computational approach aimed to rigorously test the effect of porosity on graphite’s phonon spectra using realistic atomic models. We developed a machine-learned inter-atomic potential for carbon that can accurately describe both crystalline graphite and configurations with micropores. The workflow consisted of several steps: (1) first-principles calculation of graphite lattice dynamics over a range of volumes to capture thermal expansion (quasi-harmonic approximation, QHA), (2) generation of an extensive *ab initio* molecular dynamics (AIMD) dataset at various temperatures, (3) training of a Deep Potential Molecular Dynamics (DeePMD) model on the AIMD data, and (4) classical MD simulations with the trained potential on large supercells of graphite with and without engineered pores, followed by phonon spectrum calculations from the MD trajectories.

2.2.1. Determination of temperature-dependent lattice constants

Graphite crystallizes in the hexagonal system (space group $P6_3/mmc$). At room temperature, its lattice constants are approximately $a = 2.462 \text{ \AA}$ and $c = 6.708 \text{ \AA}$ [30], with four atoms in the unit cell. To determine the temperature-dependent lattice constants for graphite, we employed the quasi-harmonic approximation with Phonopy, which calculates the Helmholtz vibrational free energy. The QHA refines the harmonic model by allowing phonon frequencies to vary with volume, thus introducing limited anharmonic effects at non-zero temperatures. In practice, the method involves summing vibrational free energy contributions from all phonon modes over a range of volumes and temperatures. Determining the volume that minimizes the free energy at each temperature reveals how the lattice parameters vary with temperature. We used VASP for the force and static energy calculations, following the procedure detailed below.

The first step was to relax the graphite unit cell and determine an appropriate functional for these calculations. We tested several functionals and found that GGA-PBESol, coupled with a DFT-D3 zero-damping scheme to account for long-range van der Waals (vdW) forces in layered materials like graphite, yielded the most accurate lattice constants. The relaxed 0 K lattice constants ($2.598 \text{ \AA}$ for a and $6.606 \text{ \AA}$ for c) were in good agreement with experimental room-temperature values. A $20 \times 20 \times 6$ Monkhorst–Pack k -point grid was used

for the relaxation, while other parameters appear in Table 1—a summary of all VASP parameters used in this work for various calculations.

Next, we systematically distorted the relaxed unit cell by uniformly scaling a and c lattice constants over a range (e.g., from 95% to 115% of their relaxed values), generating a grid of slightly larger and smaller unit cells. This step produced a large number of calculations and was facilitated by computational resources at the National Energy Research Scientific Computing Center (NERSC) and Rensselaer Polytechnic Institute’s Center for Computation Innovations (CCI). For each scaled structure, we performed an additional relaxation with ISIF=2 in VASP to relax the internal atomic positions at the fixed lattice dimensions. Next, for each relaxed, scaled structure, we constructed a $5 \times 5 \times 2$ supercell using Phonopy. The unperturbed version of this supercell was used to compute the static total energy in a single-point VASP force calculation. Then, with Phonopy’s finite-displacement supercells, we ran single-point VASP force calculations to extract forces on each atom for each distortion. Phonopy’s post-processing returns vibrational free energy, $F_{\text{phonon}}(T)$, as a function of temperature for each scaled structure. The static energy and vibrational free-energy terms combine into the total free energy,

$$F_{\text{QHA}}(T, a, c) = E_{\text{static}}(a, c) + F_{\text{phonon}}(T, a, c), \quad (3)$$

so that at each temperature, the equilibrium lattice parameters $a(T)$ and $c(T)$ are determined by minimizing a polynomial fit to a continuous $F_{\text{QHA}}(T, a, c)$ surface across the sampled (a, c) grid. An example of such a polynomial fit to the total free energy surface at 300 K is shown in Figure 2. The temperature-dependent values for $a(T)$ and $c(T)$ appear in Figure 3. ChatGPT o1 [31] was used to set up a series of bash and Python scripts that automated much of this workflow.

At 300 K, the calculated lattice constant a is 2.4671 Å, which is within 0.2% of the experimental value (2.462 Å). More importantly, the predicted negative thermal expansion at low temperature for the a -axis (in-plane direction) [32] is reasonably reproduced by our calculations. The calculated c value at 300 K is 6.7229 Å, also within 0.2% of the reported experimental 6.708 Å.

2.2.2. DeePMD potential training and AIMD dataset

The training data for our DeePMD potential were generated by performing AIMD NVT simulations (i.e., constant number of particles N , volume V , and temperature T around an equilibrium value) under a Nosé–Hoover thermostat. Temperatures used to generate the training dataset were 15, 50, 100, 150, 200, 250, 275, 300, 325, 350, 400, 450, 500, 600, 700, 800, 900, 1000, 1100, 1200, 1300, 1400, 1500, 1600, 1700, 1800, 1900, and 2000 K. As in the QHA calculations, vdW forces were modeled via the DFT-D3 zero-damping method, and a $5 \times 5 \times 2$ supercell was adopted. All AIMD calculations were done at the gamma point, which is standard for AIMD when large supercell is used. Lattice constants for each temperature were taken from the QHA values in Section 2.2.1. At each temperature, the structure was equilibrated for 1 ps with a 1 fs time

Table 1: Summary of VASP parameters used throughout this work. The top section lists common settings for all calculations, and the bottom section details calculation-specific parameters.

Common Parameters (all calculations)	
LREAL	.FALSE.
PREC	Accurate
ALGO	Normal
EDIFF	1×10^{-8}
EDIFFG	-1×10^{-8}
NSW	100
ENCUT	900 eV
ISMear	-1
SIGMA	0.2 eV
GGA	PS
IVDW	11
ADDGRID	.TRUE.
Calculation-Specific Overrides	
Initial Cell Relaxation	
	• ISIF=3 (relax cell <i>and</i> atoms) • IBRION=2 (Conjugate Gradient) • KPOINTS: $20 \times 20 \times 6$
Cell Relaxation (QHA)	
	• ISIF=2 (fix cell, relax atoms only) • IBRION=2 • NSW=100 • KPOINTS: $20 \times 20 \times 6$
Single-Point Force (QHA)	
	• IBRION=-1 (no relaxation) • KPOINTS: $3 \times 3 \times 4$ • Supercell size: $5 \times 5 \times 2$
AIMD	
	• PREC=Normal • ISIF=2 (fix cell, relax atoms only) • EDIFF=1×10^{-6} • ENCUT=500 eV • NELMIN=4 • IBRION=0 • ISYM=0 • SMASS=0 • Γ-point only • Supercell size: $5 \times 5 \times 2$ • Nose–Hoover NVT at 15–2000 K • Timestep=1 fs; 1 ps equil. + 4 ps production

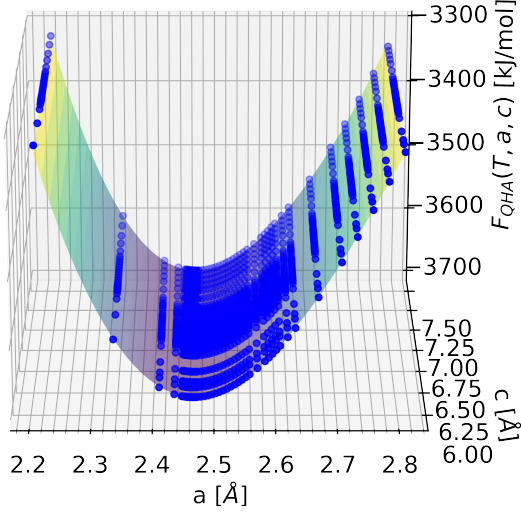


Figure 2: An example polynomial fit to the total free energy surface at 300 K.

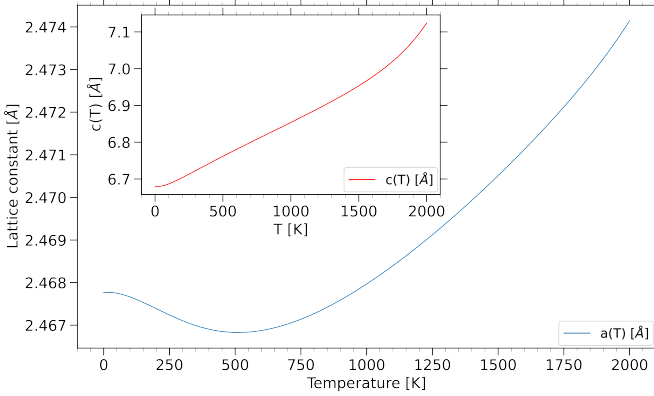


Figure 3: Calculated $a(T)$ values for graphite, with $c(T)$ shown in the inset.

step, followed by a 4 ps production run at 1 fs/time step. This yielded 4000 frames per temperature and 112,000 frames in total (112 ps). The other parameters are found in Table 1.

The dataset was split so that 87.5% of the frames were used for training, 10% for validation, and 2.5% for final testing. We employed the `se_e2_a` descriptor in DeePMD [18, 19]; this descriptor is designed to capture local atomic environments by encoding pairwise interactions and higher-order correlations in a rotationally invariant form. Such a descriptor is vital for layered materials like graphite, where strong in-plane covalent bonding contrasts with weaker interlayer vdW forces. Our potential used three layers in the descriptor network (64, 128, 256 neurons) and three layers of 240 neurons each in the fitting network. An exponentially decaying learning rate, dropping every 5000 steps from 0.001 to 3.51×10^{-8} , was employed. The loss function carefully weights energies, forces, and virial components; the virial prefactor ensures realistic stress–strain responses and me-

chanical properties. The force prefactor (hyperparameter that determines the weight or importance given to force predictions during the training process) starts higher (1000) than the energy (0.02) and virial (0.01) factors, compelling the model to prioritize accurate force predictions, which is crucial for correctly computing phonon dispersions and phonon spectra.

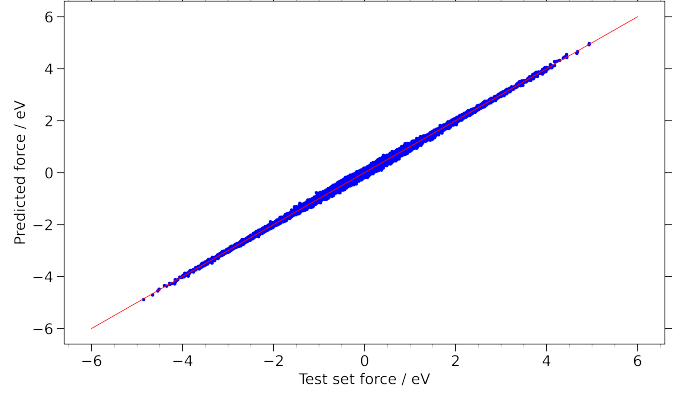


Figure 4: Test results for the force predictions of the trained potential against 300 K AIMD test data.

According to `dpctest` metrics, the trained DeePMD potential exhibits excellent accuracy. Against the 300 K AIMD data, the energy mean absolute error (MAE) is 0.04 eV, with a root mean square error (RMSE) of 0.05 eV (about 2×10^{-4} eV/atom). The forces show an MAE of 0.035 eV/Å and an RMSE of 0.047 eV/Å. Virial errors are also well-controlled, with a total MAE of 0.30 eV and an RMSE of 0.41 eV, corresponding to 1.5×10^{-3} and 2.04×10^{-3} eV/atom, respectively. These results, as illustrated in Figure 4, confirm the potential’s suitability for accurate energy and force calculations, which are critical for phonon dispersion and vibrational property predictions.

2.2.3. Simulation of microporous graphite and phonon spectra

To investigate how pores influence phonon spectra, we used the trained DeePMD potential in LAMMPS [17] to simulate several large graphite supercells, both porous and non-porous. Typical pore sizes in nuclear graphite range from a few micrometers up to hundreds of micrometers [2]. Such sizes are far beyond direct atomistic simulation; however, our INS results and prior studies [6, 7] indicate that these typical pore sizes have no observable effect on phonon spectra. We therefore focus on micropore length scales (up to a few nm) in our models. We employed a custom Python script to generate porous supercell structures as follows: starting from a perfect crystalline supercell of graphite, spherical “voids” were carved out by removing all atoms within a chosen radius of randomly selected center points. We enforced a minimum spacing between pore centers and from pores to cell boundaries to avoid overlap or truncated pores. This algorithm allows controlled introduction of a specified porosity (fraction of atoms removed) while roughly mimicking a realistic random pore distribution. Because of computational cost constraints, the largest systems we could simulate were on the order of 10^5 atoms, limiting pore sizes to tens of Å (nanometers) and porosities up to 30%. Nonethe-

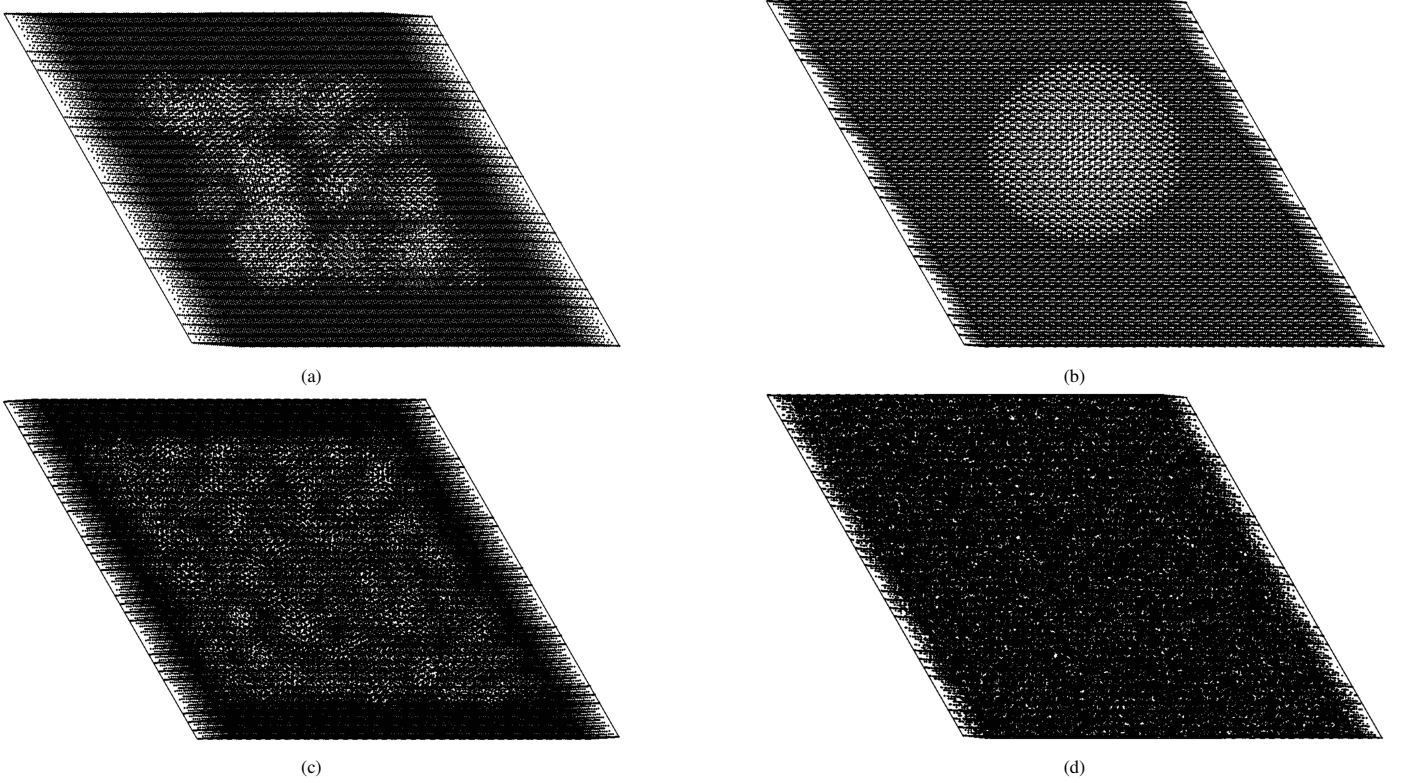


Figure 5: Visualization of four simulated graphite structures: (a) a $35 \times 35 \times 12$ supercell with 62 randomly placed micropores (radii 5–10 Å, 10.2% porosity), (b) a single spherical pore (22 Å radius, 7.5% porosity) in an equivalent cell, (c) numerous tiny pores (3.5–6 Å radii, 16.9% porosity in a $40 \times 40 \times 16$ cell), and (d) a 30% porous structure generated by random removal of atoms (no defined pore geometry, ~89k atoms). Carbon atoms are shown in gray, and void regions are transparent.

less, these models enable a direct test of whether micropores alter the phonon spectrum.

We prepared five representative graphite structures: one non-porous crystalline reference and four porous cases with different pore characteristics. Figure 5 illustrates four of these structures. Structure (a) is a $35 \times 35 \times 12$ supercell (approximately 52,000 atoms before any removal) in which 62 spherical pores of radii 5–10 Å were inserted, yielding a porosity of 10.2%. This case, denoted “realistic micropores,” reflects the pore size distribution gleaned from our SANS data (dominant pore radii in the 5–10 Å range, discussed in Section 3.2). Structure (b) is a $35 \times 35 \times 12$ cell with a single large spherical pore of radius 22 Å (porosity 7.5%). Structure (c) is a larger $40 \times 40 \times 16$ cell with many very small pores (radius 3.5–6 Å, 52 pores, 16.9% porosity). Structure (d) is a $40 \times 40 \times 20$ cell where 30% of atoms were removed completely at random (no defined pore shapes), emulating the method used in ENDF to generate 30% porosity TSLs. We also prepared a 10% random removal structure (not pictured) in a $35 \times 35 \times 12$ cell for comparison. Table 2 summarizes the parameters of these configurations, including cell dimensions, nominal porosity, pore size, number of pores, and remaining atom count.

Each structure was simulated at 300K using LAMMPS with the DeePMD potential. We first equilibrated the system in the *NVT* ensemble at 300K for 200ps (2fs time step) to ensure a steady state. Then, a production run of 500ps (1fs time step) was carried out, collecting atomic velocities for phonon anal-

Table 2: Summary of the graphite structures used in MD simulations. Nominal porosity is the fraction of atoms removed to create pores. Pore radii given as a range indicate multiple pores of varying size; ‘—’ indicates random atom removal (no well-defined pore radii).

Structure	Cell Size	Nom. Porosity (%)	Pore Radius (Å)	# Pores	# Atoms
Crystalline	$35 \times 35 \times 12$	0.0	—	0	59k
(1) Realistic Nanopores	$35 \times 35 \times 12$	10.2	5–10	62	52k
(2) Single Large Pore	$35 \times 35 \times 12$	7.5	22.1	1	54k
(3) Many Nanopores	$40 \times 40 \times 16$	16.9	3.5–6	52	85k
(4) 30% Random Removal	$40 \times 40 \times 20$	30.0	—	—	89k
(5) 10% Random Removal	$35 \times 35 \times 12$	10.0	—	—	53k

ysis. We applied periodic boundary conditions in all directions and maintained the overall system at constant temperature using a Nosé–Hoover thermostat. The velocity autocorrelation function (VACF) for all carbon atoms was computed during the production run (averaged over time using LAMMPS’s `fix ave/time`). The phonon density of states (DOS) was obtained by Fourier-transforming the normalized VACF, yielding $g(\omega)$, often called the generalized density of states (GDOS). In practice, we post-processed the VACF data: a Fast Fourier Transform (FFT) converted the time-domain VACF to a raw frequency spectrum, which we then rebinned into 1 meV intervals and convolved with a small Gaussian broadening to obtain a smooth phonon DOS curve.

3. Results and Discussion

3.1. INS results

The inelastic neutron scattering results demonstrate that graphite's phonon spectrum is essentially insensitive to porosity. Previously reported phonon spectra derived from ARCS measurements at room temperature cover various nuclear graphite grades, including irradiated samples [6, 7]. ENDF/B-VIII.0 [33] and the newly released ENDF/B-VIII.1 nuclear data libraries each contain five nuclear graphite TSL files: a crystalline version (based on phonon spectra in the incoherent approximation), an Sd version (including the coherent one-phonon contribution), and three porosity variants at 10%, 20%, and 30%. These porosity TSLs were calculated in the incoherent approximation using classical MD and with random removal of atoms from a crystalline structure to achieve the desired pore fraction. Figure 6 compares the derived phonon spectra from the ARCS measurements [6, 7] with the calculated crystalline and porosity TSL spectra.

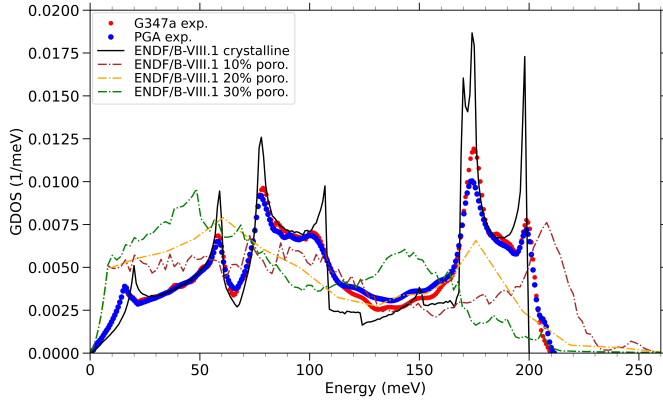


Figure 6: Comparison of phonon spectra derived from ARCS measurements [6, 7] and the calculated phonon spectra for crystalline, 10%, 20%, and 30% porosity TSLs.

Pile grade A (PGA) graphite has an apparent porosity of about 25% and a grain size of roughly 800 μm , whereas G347a-grade graphite has about 17.8% porosity and a grain size of about 50 μm . Figure 6 shows that these two graphite grades exhibit nearly identical phonon spectra, as porosity does not significantly affect atomic thermal motion (vibrations), which the phonon spectra represent. Around the pores, the atoms remain in a crystallized state (i.e., strongly bonded carbon atoms) and exhibit vibrational modes very similar to crystalline graphite; this observation is consistent with comparisons to the crystalline TSL phonon spectra. Measured and calculated crystalline TSL phonon spectra show fairly good agreement. However, introducing porosity by randomly removing atoms in the MD simulations—and using a potentially inadequate MD potential—produces large distortions in the calculated phonon spectra. Significant random atomic removal disrupts the graphite's local crystalline structure, causing significant variations in the atomic bonds and the atomic force constants and consequently, the phonon frequencies.

To further illustrate that porosity does not influence graphite's vibrational spectra, we measured six different nuclear

graphite grades using VISION at 5 K. Typically, INS spectra for TSL validation are obtained at cryogenic temperatures to minimize Debye–Waller effects. Phonon calculations via finite-displacement (frozen-phonon) methods effectively correspond to 0 K, and these phonon spectra are typically used for different temperatures in TSL evaluations. Additionally, we measured two graphite grades at 293.6 K. Figures 7 and 8 compare the measured $S(Q, \omega)$ spectra at 5 and 293.6 K (the higher temperature data are scaled up to reduce plot overcrowding), in both backward (high Q) and forward (low Q) trajectories in Q – ω space.

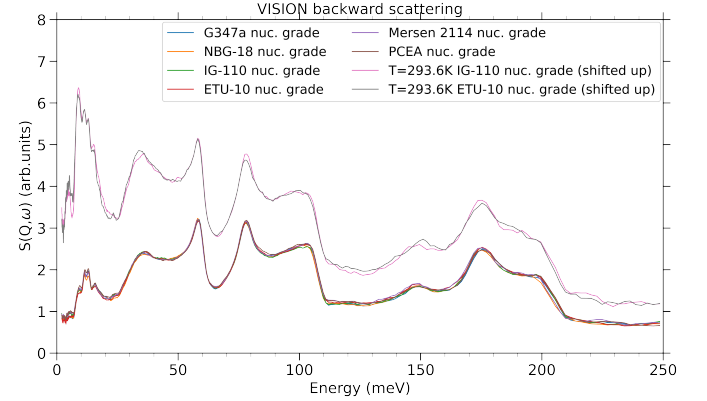


Figure 7: Comparison of the measured $S(Q, \omega)$ spectra at 5 and 293.6 K in the backward trajectory (Q – ω space). The higher temperature data are scaled up to reduce plot overcrowding.

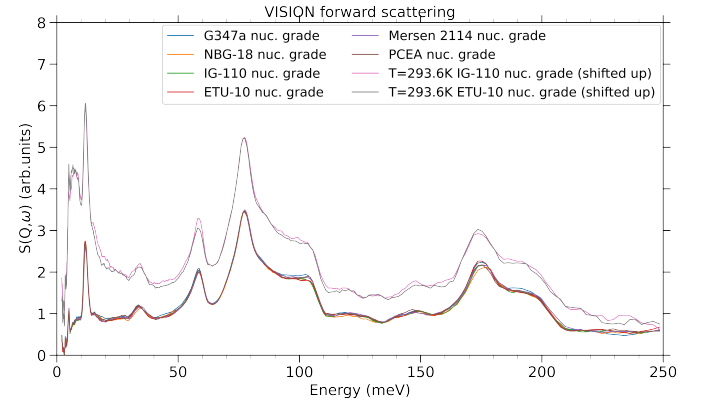


Figure 8: Comparison of the measured $S(Q, \omega)$ spectra at 5 and 293.6 K in the forward trajectory (Q – ω space). The higher temperature data are scaled up to reduce plot overcrowding.

Figures 7 and 8 display only minor differences among the measured $S(Q, \omega)$ spectra of these graphite grades, despite significant variations in density, porosity (10% to 25%), and grain sizes (13 to 1600 μm), as shown in Table 3 [34]. The differences between 5 K and 293.6 K spectra at low energies stem from the Debye–Waller effect: higher temperatures amplify atomic vibrations, broadening features near the elastic peak at $\omega = 0$ and slightly redistributing intensity into inelastic wings. Although not shown, VISION $S(Q, \omega)$ spectra include an elastic peak (typically from about -2 to 2 meV in the energy transfer).

Graphite	Grain Size [μm]	Density [g/cm^3]	Porosity [%]
PGA	800	1.70	25
G347a	50	1.85	17.8
IG-110	20	1.77	21.6
NBG-18	1600	1.85	17.8
PCEA	360	1.83	18
Mersen 2114	13	1.81	10
ETU-10	40	1.77	20

Table 3: Properties of selected nuclear graphite grades.

The importance of VISION measurements for nuclear data validation (as well as general phonon and $S(Q, \omega)$ calculations validation) is in the simplicity with which calculated TSLs can be compared with the measurements. It is straight-forward to extract $S(Q, \omega)$ —at the same trajectory in Q – ω space as in VISION measurements—from an ENDF TSL file, apply the pre-determined VISION instrumental resolution function, and then compare it to VISION measured data. The stored quantity in ENDF TSL files for INS cross sections is $S(\alpha, \beta)$, but the transformation from $S(\alpha, \beta)$ to $S(Q, \omega)$ is straightforward:

$$S(\alpha, \beta) = k_B T \exp\left(\frac{-\hbar\omega}{2k_B T}\right) S(Q, \omega), \quad (4)$$

where T is the temperature, and k_B is the Boltzmann constant. It is possible to compare an ENDF TSL with a $S(Q, \omega)$ measured using ARCS or SEQUOIA instruments, but doing so involves the use of Monte Carlo neutronics codes such as MCNP [35] or McViNE [36], which is a methodology that has been utilized in other works [37, 38, 39].

Figures 9 and 10 compare the measured and calculated $S(Q, \omega)$ at 5 K in both backward and forward scattering trajectories. The TSLs for crystalline, 10%, 20%, and 30% porosity from ENDF/B-VIII.1 are shown. The Sd TSL (coherent one-phonon approximation) was evaluated using a private code unavailable to the public ??, and because no 5 K TSL was provided, we employed OCLIMAX to demonstrate the effect of coherent one-phonon contribution.

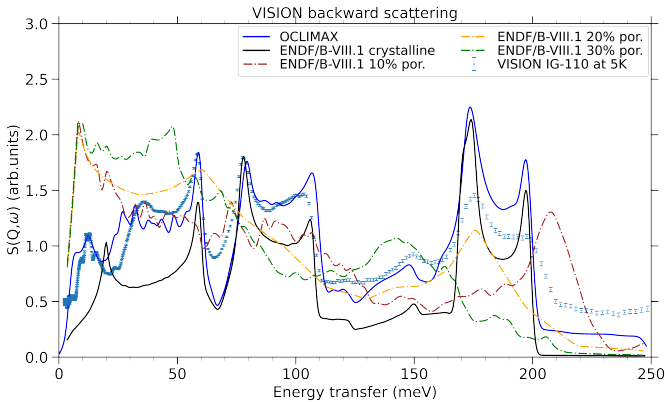


Figure 9: Comparison of calculated and measured $S(Q, \omega)$ at 5 K in backward scattering (Q – ω space).

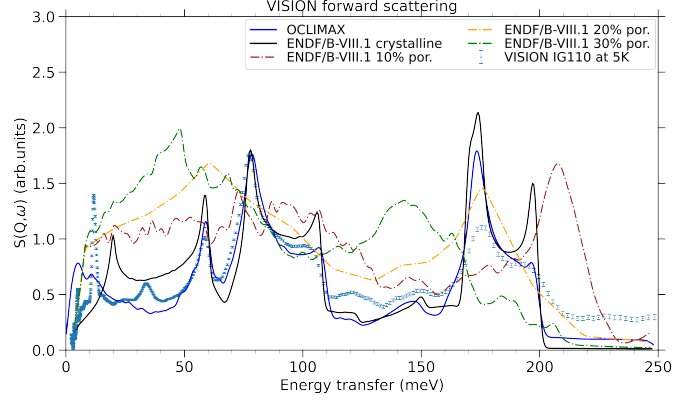


Figure 10: Comparison of calculated and measured $S(Q, \omega)$ at 5 K in forward scattering (Q – ω space).

Figures 9 and 10 show that the large distortions in the calculated phonon spectra for the 10%, 20%, and 30% porosity TSLs (Figure 6) carry over to the computed $S(Q, \omega)$, leading to significant disagreements with measured data. By contrast, the crystalline TSL (calculated using the incoherent approximation) yields better agreement with the experiment. At energies above 50 meV, there is good agreement in the shape of the $S(Q, \omega)$ calculated from crystalline phonon spectra and the measurement. However, below 50 meV, where the coherent inelastic effects are dominant, there is a disagreement with the measured $S(Q, \omega)$ spectra. The peak at ≈ 35 meV, observed in both forward and backward trajectories in Q and ω space, is missing, whereas the peak at ≈ 20 meV in the calculation should be shifted to lower energies. Caution is necessary here when talking about the measured VISION $S(Q, \omega)$ spectra because the $\lambda/2$ peak usually manifests itself in VISION data around 11–12 meV as a well-isolated sharp peak. Based on backward scattering measurement, we can see a significant structure in that energy region, so the contribution from $\lambda/2$ cannot be easily isolated and identified. When looking at the OCLIMAX coherent one-phonon calculation of $S(Q, \omega)$, we can see that it is in better agreement with the shape of the measured data, although there are still some minor inconsistencies. Ultimately, VISION’s higher energy resolution makes coherent inelastic features more evident and reaffirms VISION measurements as essential for validating our understanding of phonon and $S(Q, \omega)$ calculations.

Overall, both the phonon spectra derived from ARCS and the $S(Q, \omega)$ data from VISION (shown in Figures 6, 9, and 10) confirm that the inherent porosity of nuclear graphite does not affect its atomic vibrational properties.

3.2. Small-angle neutron scattering measurements results

SANS is an experimental technique used to probe nanoscale inhomogeneities in materials by measuring the intensity of neutrons scattered at very small angles from a collimated beam. In these experiments, the scattered intensity $I(Q)$ is recorded as a function of momentum transfer Q , which is inversely related to the real-space dimensions of the scattering features. By analyzing the $I(Q)$ profile, information about the size, shape, and

distribution of pores or other inhomogeneities can be extracted.

The GP-SANS instrument at Oak Ridge National Laboratory’s HFIR is specifically designed for high-resolution SANS measurements. Leveraging a high-flux reactor source and advanced detector technology, the instrument covers a broad Q range, enabling detailed characterization of nanoscale structures—a crucial capability for studying nuclear graphite, where pore structure significantly affects material performance. For our measurements, small circular samples with a radius of 6 mm and a thickness of 1 mm were used to minimize multiple scattering. Three instrument settings were employed to probe a scattering vector range of 0.0007 to 1 $\text{\AA}^{-1}$. The configurations are shown in Table 4.

Table 4: Scattering vector (Q) settings and corresponding instrument parameters.

Scattering Vector Setting ($\text{\AA}^{-1}$)	Wavelength ($\text{\AA}$)	Source Collimation (m)	Sample–Detector Distance (m)
0.004–0.02	12	17.5	19.0
0.007–0.12	4.75	7.35	7.5
0.07–0.8	4.75	7.35	1.0

The measured $I(Q)$ spectra of six different grades of nuclear graphite can be seen in Figure 11.

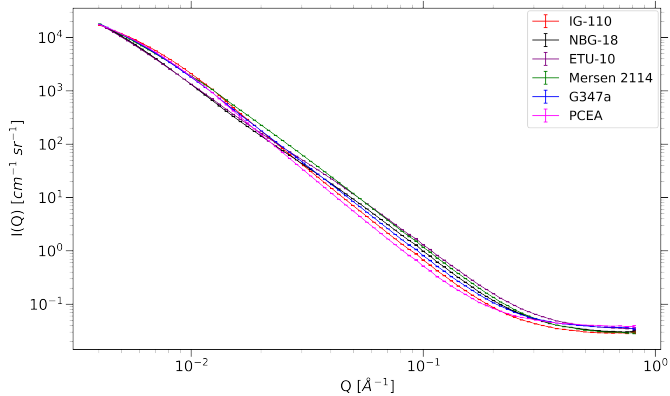


Figure 11: The measured $I(Q)$ spectra of six different grades of nuclear graphite from the GP-SANS instrument at HFIR.

In Gallego et al. [40], SANS $I(Q)$ data at high Q values (i.e., $Q > 0.1 \text{ \AA}^{-1}$) were fitted using a polydisperse spherical model to extract the pore size distribution for narrow pores (radius $< 30 \text{ \AA}$). The largest pore sizes accessible via conventional SANS are dictated by the lowest measurable scattering vector, with the maximum accessible radius approximately given by

$$r_{\max} \approx \frac{\pi}{Q_{\min}}. \quad (5)$$

Since our $I(Q)$ data ranges in Q from 0.004 $\text{\AA}^{-1}$ to 0.81 $\text{\AA}^{-1}$, the maximum accessible pore radius is around 75 $\text{\AA}$, and the minimum accessible radius is about 4 $\text{\AA}$. As shown in the work by Arregui-Mena et al. [2], most pores in nuclear graphite exceed these sizes, and, as discussed in Section 2.1 for $S(Q, \omega)$ measurements, these pores do not affect the phonon spectra. It is worth mentioning that, ultra SANS or other techniques are required to characterize larger pore sizes.

To demonstrate computationally—see Section 2.2—that even the smallest realistic pores have no effect on the phonon spectra (contrary to the ENDF/B-VIII.1 porosity TSLs), we fitted the $I(Q)$ data over 0.08–0.8 $\text{\AA}^{-1}$, a range that captures the narrow-pore distribution. We utilized the McSAS software [41]—a form-free, Monte Carlo–based regression tool—to analyze SANS data collected from different grades of nuclear graphite. By fitting the measured $I(Q)$ using a spherical pore model, McSAS enabled us to retrieve a pore size distribution spanning radii from approximately 1 to 30 $\text{\AA}$. The $I(Q)$ fits for the PCEA and ETU-10 grades are shown in Figure 12, and the corresponding pore radii distributions for all six graphite grades appear in Figure 13. Notably, the highest relative volume fraction of the smallest pores occur in the radius range of about 5–10 $\text{\AA}$, in good agreement with results reported in Gallego et al. [40].

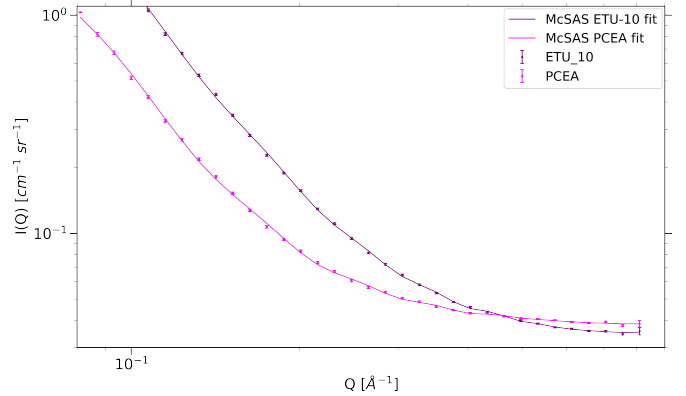


Figure 12: The McSAS-obtained $I(Q)$ fits in the Q range from 0.08 to 0.8 $\text{\AA}^{-1}$ for PCEA and ETU-10 graphite grades.

3.3. Atomistic simulation results

As discussed in Section 3.1, the probable causes of the distortions observed in the phonon spectra for the 10%, 20%, and 30% porosity TSLs are twofold, with a strong interplay between them. The first factor is the inadequate potential used in MD calculations. This shortcoming can be clearly seen in Hawari [13, p. 26] when comparing the phonon spectrum of ideal (i.e., crystalline) graphite with those of 10% and 30% porosity, all computed via MD. A second comparison on that page contrasts an *ab initio* phonon spectrum for crystalline graphite and the measured phonon spectrum; both exhibit fairly good overall agreement in shape. However, although the MD spectrum for crystalline graphite matches the *ab initio* result up to about 60 meV, substantial discrepancies arise at higher energies. In particular, the *ab initio* spectrum shows two distinct peaks around ≈ 175 and 200 meV, whereas the MD spectrum has a single peak near 210 meV. These discrepancies indicate that the chosen MD potential does not adequately describe atomic thermal motion in graphite.

The second factor that contributes to the distortions in the 10%, 20%, and 30% porosity TSL phonon spectra relates to

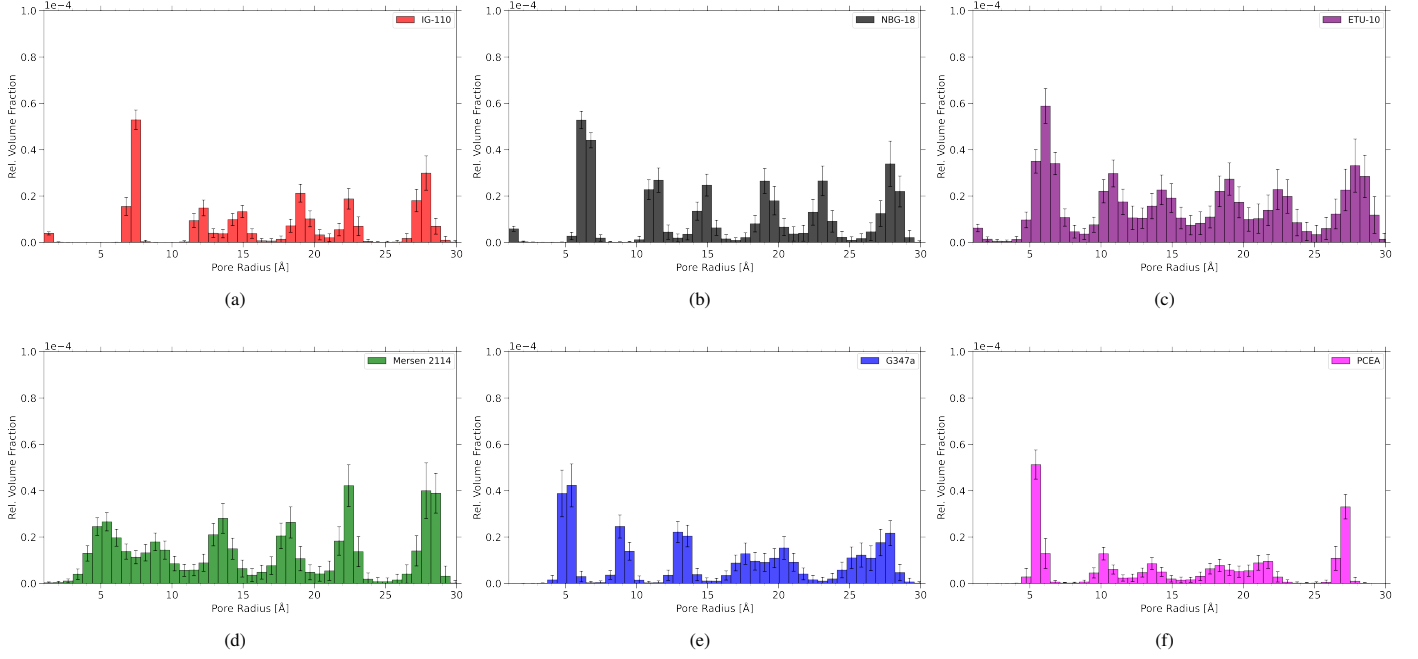


Figure 13: McSAS-calculated pore radii distributions for (a) IG-110, (b) NBG-18, (c) ETU-10, (d) Mersen 2114, (e) G347a, and (f) PCEA grades of nuclear graphite.

how porosity is modeled, namely, through random atom removal until reaching the desired porosity fraction. To confirm that these two factors indeed explain the phonon spectrum distortions in the 10%, 20%, and 30% porosity TSLs, we performed a detailed study to assess the nuclear graphite porosity effect on vibrational properties using a machine-learned potential generated via DeepMD and LAMMPS.

Our DeepMD-based simulations address both issues: we use a DFT-accurate potential and introduce pores as actual voids (removing clusters of atoms to form a cavity, rather than isolated vacancies spread throughout). Figure 14 establishes a baseline by comparing the phonon spectra for *crystalline* graphite from experiment, ENDF/B-VIII.1 (which uses a DFT phonon spectrum), the previous classical MD [13] from the evaluators of ENDF/B-VIII.1 crystalline TSL, and our DeepMD simulation. The experimental phonon spectra (e.g. from INS measurements) and ENDF/B-VIII.1 phonon spectra show the characteristic split optic peaks near 170–200 meV. The classical MD phonon spectra for crystalline graphite (digitized from [13]) deviates above 60 meV, merging those peaks into one and slightly misplacing others. Our DeepMD-based phonon spectra aligns much more closely with the ENDF (DFT) and experimental curves, capturing both high-energy peaks and generally following the experimental spectrum within small discrepancies (a slight intensity mismatch in parts of the acoustic band below 60 meV). This confirms that the DeepMD potential provides a good representation of graphite’s vibrational dynamics in the absence of defects. It should be emphasized that our trained DeepMD potential is not intended to perfectly match the experiment or serve as the basis for a new ENDF TSL evaluation. Rather, our goal is simply to demonstrate that pore presence does not alter phonon spectra. Other DeepMD descriptors

offer higher accuracy than `se_e2_a`, but `se_e2_a` was chosen here due to lower computational cost.

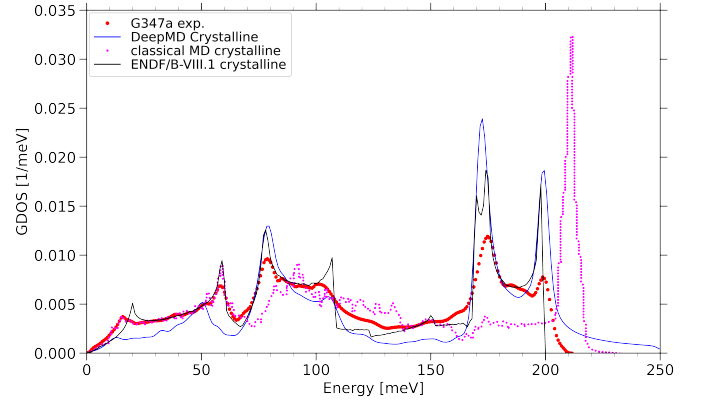


Figure 14: Comparison of the experimental, ENDF/B-VIII.1 crystalline, classical MD crystalline, and DeepMD-based calculated crystalline phonon spectra.

Having validated the potential, we computed phonon DOS for the various porous structures described in Table 2. Figure 15 compares the calculated phonon spectra for various microporous structures. It is evident that porosity does not affect the phonon spectra in the manner suggested by the 10%, 20%, and 30% ENDF/B-VIII.1 porosity TSLs. Instead, the overall spectral shape remains in line with both calculated crystalline and measured phonon spectra. That is, regardless of the pore size, the main phonon spectra peaks are well pronounced and located at the same phonon energies, in excellent agreement with INS measurements and DFT calculations.

Next, we explicitly verify that the problematic ENDF approach—random vacancies—indeed causes distortions even

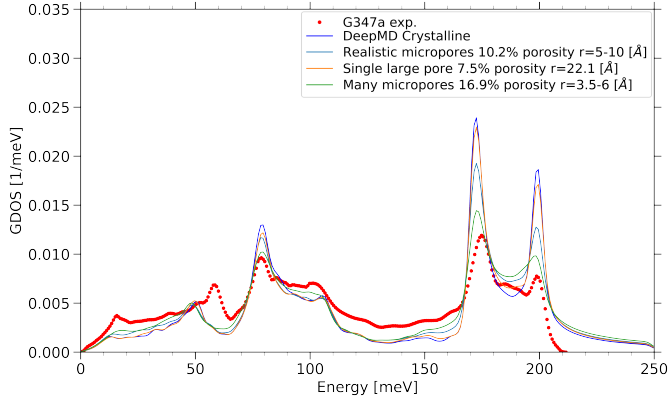


Figure 15: Comparison between the experimental and DeePMD-based calculated phonon spectra for crystalline graphite, realistic micropores, a single large pore, and a structure with many micropores.

under our improved potential (albeit less severe). We created structures by randomly deleting 10% and 30% of the atoms from a crystalline supercell (cases 5 and 4 in Table 2). Figure 16 shows the phonon spectra for the 10% and 30% random-removal cases compared to the crystalline phonon spectra. As expected, distortions appear: the 30% vacancy case shows a noticeable shift and broadening of the high-frequency optic band; the two-peak structure is smeared into one broader hump. The 10% vacancy case shows a milder distortion but still some broadening and a slight increase in low-energy modes relative to crystalline. These trends mirror the ENDF TSL spectra—though our DeePMD potential somewhat mitigates the severity (the peaks are not as completely merged as in the classical MD potential case). Still, it is clear that random removal (point defects) produces an artificial softening of the lattice. In reality, porosity is not point defects but extended voids, which, as we’ve seen, leave the phonons spectrum intact. Thus, our simulations conclusively demonstrate that the ENDF porosity strategy introduces spurious phonon effects, whereas actual porosity does not. The distortions in the ENDF 10–30% TSLs stem from the modeling approach, not from actual physics of porous graphite.

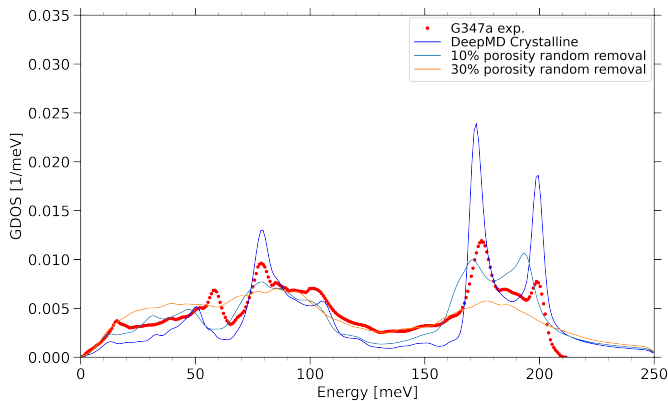


Figure 16: Comparison between the experimental and DeePMD-based calculated phonon spectra for the crystalline structure and those created by randomly removing atoms to achieve 10% and 30% porosity.

In summary, our combined INS measurements and atomistic simulations firmly establish that nuclear graphite’s inherent porosity does *not* alter its vibrational (phonon) spectrum. All major phonon peaks remain unchanged in energy and intensity with porosity. The increases in inelastic neutron scattering cross sections seen in ENDF’s 10–30% TSL are not due to any real phonon effect of pores, but rather due to the nonphysical random defect model and the limitations of the potential used. Our work shows that a crystalline graphite TSL (with a coherent one-phonon addition, as in the Sd variant) is adequate to represent the true inelastic scattering, and porosity should instead be accounted for via its separate small-angle scattering contribution (discussed next). We now turn to the total cross-section (transmission) measurements to quantify that SANS contribution and evaluate how to incorporate it properly.

3.4. Total cross section measurements

The quality of nuclear data used by neutronics codes is crucial for accurately predicting reactor behavior under both steady-state and transient conditions to support safety evaluations, accident analyses, and the development of advanced reactor concepts.

For thermal neutrons (energies below ~ 10 eV), TSL data are utilized by codes like SCALE [42] and MCNP [35] to model inelastic and elastic scattering processes that occur when neutrons interact with bound atoms. In practice, TSL data in ENDF format provide the dynamic structure factor, $S(\alpha, \beta)$, which describes the probability of inelastic scattering at a given energy and momentum transfer via phonon excitation or de-excitation in the material. SCALE and MCNP read these TSL libraries and sample angles and energy transfer during neutron transport, thereby incorporating thermal motion and lattice vibrations into the neutron scattering behavior. Along with inelastic scattering data, TSL files also include coherent and incoherent elastic cross sections: the coherent elastic cross section reflects interference from the ordered atomic lattice, whereas the incoherent elastic cross section corresponds to random variations in the nuclei’s scattering lengths. Integrating the differential scattering cross sections over all angles and energies yields the total scattering cross section; adding the absorption cross section then gives the total cross section. In neutron-transport simulations, codes such as SCALE and MCNP use this total cross section to set the mean free path of thermal neutrons in the material, thus determining how often they scatter or are absorbed. Thus, whereas $S(\alpha, \beta)$ describes in detail the energy and angular distributions of scattered neutrons, the total scattering cross section normalizes how often those interactions occur, thereby fixing the overall moderation and thermalization rates.

Total cross sections are typically measured through transmission experiments, where the measured transmission T relates to the cross section by

$$T = e^{-n\sigma_t}, \quad (6)$$

where n is the areal density of the sample, and σ_t is the total microscopic cross section. In such experiments, a well-collimated neutron beam is directed onto the sample, and a detector—usually shielded by collimators that block scattered

neutrons—counts only transmitted (unscattered) neutrons. The experimental transmission is given by

$$T = \frac{C_s - B_s}{C_o - B_o}, \quad (7)$$

where C_s is the normalized counts with the sample in place, B_s is the background counts with the sample in, C_o is the counts with the sample out, and B_o is the background counts with the sample out.

EXFOR (*EXchange FORmat*) is a comprehensive database [43] that compiles experimental nuclear reaction data from laboratories worldwide. Managed by the International Network of Nuclear Reaction Data Centres (NRDC), EXFOR contains measured cross sections, angular distributions, and other nuclear reaction observables. It serves as a vital reference for both fundamental nuclear physics and practical reactor safety evaluations. Figure 17 compares historical graphite data [44, 45]—including total cross section measurements on highly ordered pyrolytic graphite (HOPG)—with the calculated total cross section, as well as the inelastic and coherent elastic cross sections from all graphite ENDF/B-VIII.1 TSLs.

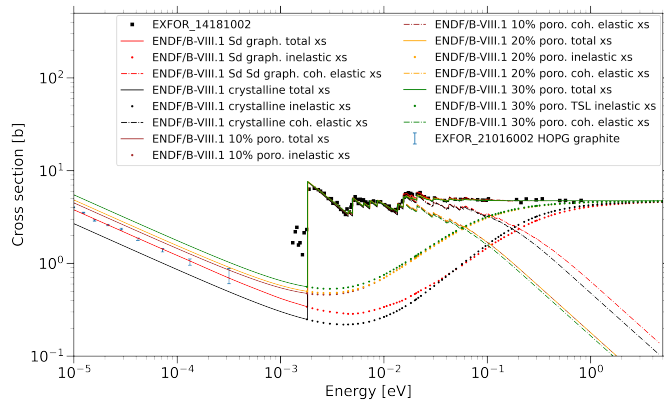


Figure 17: Experimental vs. calculated total cross sections, alongside inelastic and coherent elastic cross sections, for all graphite ENDF/B-VIII.1 TSLs.

Several observations can be drawn from Figure 17. Above the first Bragg edge (1.82 meV), all calculated total cross sections show nearly identical behavior—agreeing with EXFOR 14181002 data—even though their inelastic scattering cross sections differ. A strong interplay among the coherent elastic, incoherent elastic, and inelastic scattering cross sections, mediated by the Debye–Waller factor (which depends on the phonon spectra), yields a compensating effect: any increase in inelastic scattering is offset by a decrease in coherent elastic scattering, keeping the total cross section nearly constant. This effect is evident in Figure 17.

Another key observation is the inelastic scattering cross section, which significantly influences the results presented in Section 3.5. Figure 17 reveals a systematic rise in inelastic scattering cross section when transitioning from the crystalline to the 30% porosity TSL. This increase can be attributed to the unphysical phonon spectrum distortions identified in Sections 2.1 and 2.2. In contrast, the modest inelastic cross section increase

for the Sd graphite TSL is a result of incorporating the coherent one-phonon approximation, aligning its cross section with HOPG data below the first Bragg edge. These observations suggest that among the five ENDF/B-VIII.1 TSLs, the Sd graphite TSL reproduces crystalline graphite scattering most accurately; thus, it is the best candidate for describing inelastic, coherent, and incoherent elastic scattering in nuclear graphite.

Contrary to the assumption in the ENDF/B-VIII.1 porosity TSLs—that is, that porosity manifests solely as altered inelastic scattering—porosity actually introduces an additional scattering mechanism. Decades of experiments (see Figure 18) show that polycrystalline nuclear graphite’s total cross section at sub-thermal energies can be substantially larger—often by an order of magnitude—than that predicted by perfect-crystal models based only on phonons plus absorption. For instance, whereas theory suggests a total cross section of roughly 0.4 b for graphite at 0.001 eV (including absorption and elastic and inelastic scattering), measurements on nuclear-grade graphite typically exceed 4 b [9, 10], reaching up to 31 b for T8, ET-11, ET-34, and ED-4 grades digitized from [11]. The lone exception is nearly fully dense, highly oriented pyrolytic graphite (with $\sim 0\%$ porosity), whose cross section is close to the theoretical Sd TSL value. This large discrepancy, which has been documented since the 1950s, is now recognized as stemming from SANS within the porous microstructure.

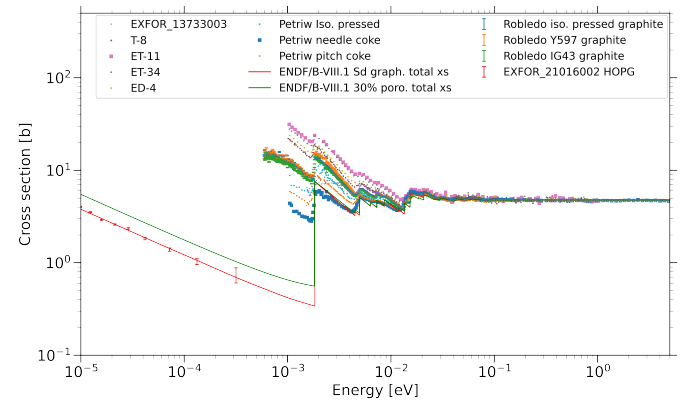


Figure 18: Comparison of experimental and calculated total cross sections for ENDF/B-VIII.1 Sd and 30% porosity TSLs.

Early investigators such as Egelstaff [8] proposed that neutrons undergo gentle refraction and slight angular deviations when traversing the numerous air–solid interfaces in porous graphite. These small-angle deflections broaden the transmitted beam—an effect Egelstaff observed directly as increased divergence behind a graphite slab—and appear as additional attenuation in transmission measurements. The findings of Egelstaff were criticized by Bowman et al., [46], but as Gould, Hawari, and Sharapov [47] showed, low-energy neutron scattering in graphite is dominated by such small-angle scattering events. More recent work [9, 10] confirms that small-angle scattering events remove neutrons from the direct beam without large-angle scattering or absorption: the extra cross section observed in transmission experiments arises from SANS in the void structure, not from phonon interactions. An ex-

ample of how SANS removes neutrons from the direct beam (i.e., reduces transmission) is shown in Figure 19, where a 1 mm graphite sample’s transmission is computed via Equation 6 using the ENDF/B-VIII.1 Sd TSL and then compared to 1 mm transmission computed from IG43 graphite’s total cross section [12].

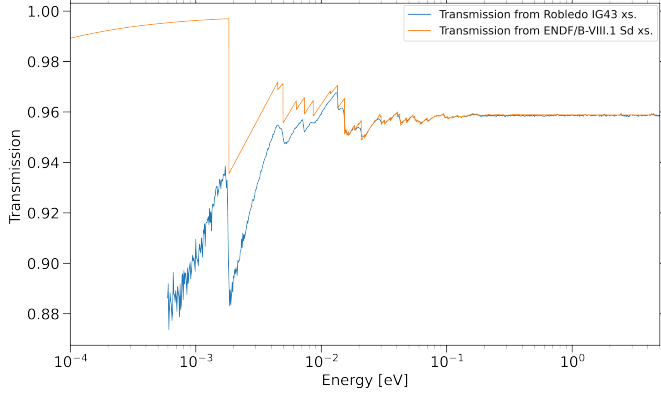


Figure 19: Calculated transmission for a 1 mm thick graphite sample (using ENDF/B-VIII.1 Sd TSL), compared with 1 mm transmission computed from IG43 graphite total cross section.

Petriw [9] conducted a systematic experimental SANS study in nuclear graphite, measuring multiple samples of varying thicknesses, orientations, and sample-to-detector distances. He showed that porosity-induced SANS is primarily responsible for the increased total cross section observed below ≈ 11 meV in nuclear graphite. A critical conclusion from Petriw is that the SANS cross section changes only in thinner samples, remaining essentially constant for thicker specimens. This finding is particularly relevant to nuclear reactor applications, where graphite blocks far exceed the thickness of typical transmission samples.

The term “SANS cross section” refers to the contribution of SANS to the overall total cross section and should not be confused with the SANS experimental technique itself. In a typical SANS experiment, a well-collimated neutron beam is directed at a sample, and the intensity of neutrons scattered at very small angles (i.e., close to the incident beam direction) is measured. This scattered intensity is highly sensitive to structural inhomogeneities—such as pores, voids, or other density fluctuations—allowing details such as pore size distributions and surface roughness to be extracted. In contrast, when discussing SANS cross sections in transmission measurements, we refer to the effect of these small-angle scattering events have on the measured transmission. In such experiments, detectors count only the unscattered (directly transmitted) neutrons, while collimators block those scattered out of the direct beam. Consequently, the removal of neutrons via SANS results in an apparent increase in the total cross section (or equivalently, a decrease in transmission). Furthermore, although neutrons may undergo multiple small-angle scattering events within the sample, one should not assume that every neutron scattered at a small angle and inadvertently counted as transmitted (due to the inherent imperfections in transmission setups) directly contributes to the SANS cross section.

Along with the more recent measurements of various Japanese nuclear graphite grades [11], total cross section (i.e., transmission) experiments were also performed at the VESUVIO instrument at the ISIS Neutron and Muon Source [12]. Figure 18 shows these alongside ENDF Sd and 30% porosity TSL results. VESUVIO [48, 49] is a state-of-the-art inverse-geometry neutron spectrometer that was originally designed to measure nuclear momentum distributions for light elements such as hydrogen and helium using epithermal neutrons. Initially equipped with ^6Li -doped detectors, it was upgraded in 2008 to use YAP-doped gamma-ray detectors in a foil cycling configuration, which enhances count stability, resolution, and the separation between signals from light and heavier nuclei. Uniquely, VESUVIO employs a noninvasive final-energy filter that allows neutrons of all energies to reach the sample, enabling concurrent measurements of neutron Compton scattering, diffraction, transmission, and gamma spectroscopy. In this work we report transmission data from VESUVIO for three nuclear graphite grades, IG-110, NBG-18, and PCEA. Figure 20 compares our results to those from Robledo [12].

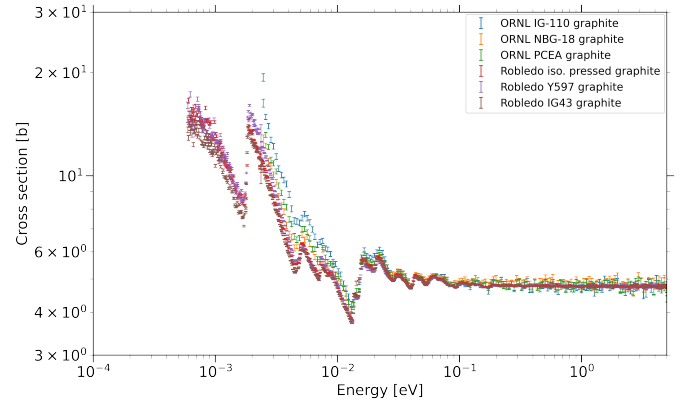


Figure 20: Comparison of IG-110, NBG-18, and PCEA graphite total cross sections with Robledo’s [12] measurements.

Our Xpress proposal allocation at ISIS imposed limited beam time, so we used 2 mm-thick samples rather than the 2.5 cm-thick ones in Robledo’s experiment. Although our data do not extend to the lowest energies reached by Robledo *et al.*, the overall agreement is encouraging. To corroborate Petriw’s finding that the SANS cross section converges in thicker specimens, we plan to repeat these measurements with larger sample thicknesses.

Notably, Robledo *et al.* [12] and Galván Josa *et al.* [10] introduced analytical models for extracting the SANS cross section. According to Robledo’s formula,

$$\sigma_{\text{SANS}}(k_0) = \frac{\sigma_0}{2k_0^2} \int_0^{2k_0} q I(q) dq, \quad (8)$$

where k_0 is the neutron wavevector, σ_0 is the bound coherent scattering cross section, and $I(q)$ is the SANS structure factor. The SANS approach has already been integrated into MCNP [50] and OpenMC [51] (for nanodiamonds [52, 53]) via NCrysal [54, 55].

Galván Josa et al. [10] refined Robledo’s model to determine the analytical form of the SANS cross section by fitting to experimental data. This refined model was applied in [56] to calculate the SANS cross section for ET-34 graphite based on measurements from Kimura et al. [11]. Moreover, the formalism given in Galván Josa et al. [10] enables one to derive the scattering intensity $I(q)$ from Equation 8. The resulting $I(q)$ can then be incorporated into the NCrystal SANSND plugin—which was originally developed to account for small-angle neutron scattering in nanodiamonds [53]—thereby effectively including the SANS cross section in neutron scattering simulations. We have repeated the fitting procedure for the same data. The SANS cross section was calculated from the fitted data. The comparison can be seen in Figure 21, which shows that the combination of the ENDF/B-VIII.1 crystalline TSL and analytical SANS fit produces a good agreement with the measurement—the ENDF/B-VIII.1 crystalline TSL was used instead of Sd TSL due to Sd TSLs’ unavailability in NCrystal. When an $I(Q)$ curve—scaled from one of the $I(Q)$ curves from Figure 11 to match the corresponding total cross section of ET-34 graphite—is used with NCrystal SANSND plugin, the agreement is good overall. There is a slight overestimation of the SANS cross section at energies above 2 meV, caused by the $1/E$ (where E is the energy) shape of the SANS cross section.

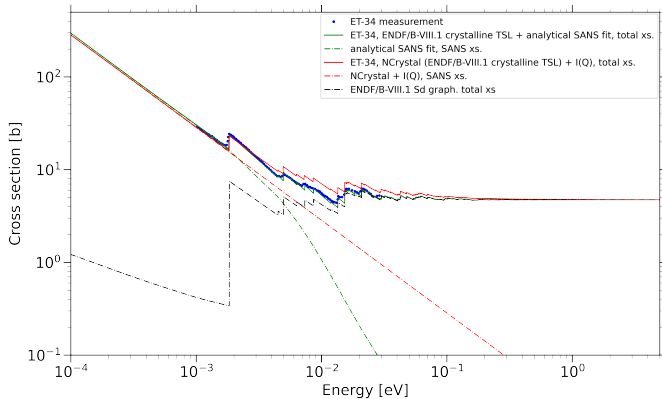


Figure 21: The comparison of the experimental ET-34 cross section and the calculated total cross sections using analytical fit for SANS cross section and NCrystal calculated SANS cross section.

ET-34, IG-43, and Petriw’s needle coke graphite measurements represent the upper, middle, and lower bounds of the measured cross sections, as shown in Figure 18. The total cross sections incorporating the NCrystal SANS cross section, which was calculated by scaling the measured $I(Q)$ curves from Figure 11 to match the corresponding total cross sections, are presented in Figure 22. Overall, the agreement is good despite an overestimation of energies above the first Bragg edge. When it comes to the needle coke graphite overestimation, similar behavior was seen by Petriw as well, which he attributed to the texture effects in the needle coke graphite [12, p. 187]. In [57], the authors argue that the transmission data measured at VESUVIO for nanodiamonds, which exhibit a noticeable SANS cross section, require corrections for in-scattering effects: that is, neutrons that are scattered yet still detected as

transmitted. Once these corrections are applied, the incorporation of the SANS cross section (maintaining the characteristic $1/E$ behavior, as computed by NCrystal) brings the calculated total cross section into good agreement with experimental measurements. This finding implies that the current experimental graphite total cross sections, particularly those obtained by Robledo at VESUVIO, should be adjusted to account for in-scattering. Such an adjustment supports the validity of SANS implementation in NCrystal. Moreover, a similar overestimation that can be attributed to the $1/E$ SANS shape is observed when employing the SANS implementation in MCNP [50]; this can be seen as well in Figure 22 for ET-34 graphite.

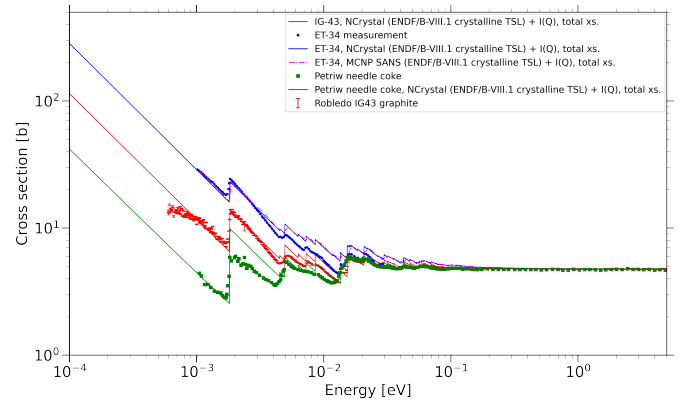


Figure 22: Comparison of experimental ET-34, IG-43, and Petriw’s needle-coke cross sections with total cross sections computed by incorporating the NCrystal SANS cross section.

The takeaway from the total cross-section analysis is that the long-standing discrepancy between measured and calculated neutron attenuation in graphite is fully explained by SANS. The ENDF approach of simulating porosity via altered phonon spectra only partially addressed the issue and in an incorrect way—by increasing σ_{inel} instead of adding a new scattering component. A better approach, as demonstrated, is to keep the graphite TSL as crystalline (or crystalline+coherent, i.e. Sd) and add a SANS cross section term to account for porosity. This approach is now feasible in transport codes via tools like NCrystal. Our measurements and modeling indicate that once in-scatter corrections are applied, the SANS models (which have a characteristic $1/E$ low-energy behavior) will match experiments extremely well. Any slight residual discrepancies (like Petriw’s data overshoot at certain energies) are likely due to specific microstructural texture or experimental geometry, not fundamental physics omissions.

In practical terms, a reactor physics calculation that needs to account for graphite porosity should include a SANS scattering kernel (with parameters fitted to the graphite grade’s pore structure) in addition to the standard thermal scattering law for graphite. This would replace the current practice of selecting a “porous” TSL that inherently modifies phonon scattering. Next, we will examine how these different treatments (crystalline vs porous TSL, with or without SANS) impact integral parameters like k_{eff} in reactor and criticality benchmarks.

3.5. Neutronics benchmark calculations

In critical benchmark calculations, the effective multiplication factor, k_{eff} , quantifies a system’s neutron economy by representing the average ratio of neutrons produced to those lost by absorption and leakage. In an ideal critical system, k_{eff} is equal to 1, and even slight deviations from unity can reveal discrepancies in the underlying nuclear data.

Integral benchmarks such as those compiled in the *International Handbook of Evaluated Criticality Safety Benchmark Experiments* (ICSBEP Handbook) [58] or the *International Handbook of Reactor Physics Experiments* [59] (IRPhE Handbook) are crucial for system-level nuclear data validation, as they compare simulation outputs from neutronics codes (e.g., SCALE, MCNP) with experimental data from well-characterized critical assemblies. However, because k_{eff} is influenced by the full energy spectrum of all nuclear interactions, these benchmarks effectively function as a “black box,” where k_{eff} is not only influenced by the TSL data, but also fast neutron cross sections, resonance parameters, and other high-energy nuclear data.

TSL data provided in ENDF format offer detailed probability distributions for both inelastic scattering—characterized by the dynamic structure factor $S(\alpha, \beta)$ —and elastic scattering (including both coherent and incoherent contributions). Neutronics codes combine these contributions to compute the total cross section, from which the neutron mean free path is derived. Because the mean free path affects neutron moderation and overall system behavior, direct experimental techniques such as INS and transmission (i.e., total cross section) measurements are essential for validating the TSL data.

Although ICSBEP integral benchmarks are indispensable for confirming that evaluated nuclear data yield realistic reactor behavior in full-system simulations, they should be regarded as a secondary validation tool. Direct microscopic measurements, such as INS and transmission experiments, are necessary to isolate and verify individual contributions (e.g., TSL data and fast neutron interactions) before testing the data in an integral context. This complementary approach reinforces overall confidence in both the microscopic nuclear data and the predictive capability of reactor physics models.

In this section, we discuss the impact of different ENDF/B-VIII.1 TSLs on the selection of ICSBEP and IRPhE benchmarks that have been found to be sensitive to graphite TSLs. The benchmarks selected for this study include HCT-016, ICT-008, HTR-10, HTRR, PROTEUS, and LCT-060. Calculations were performed using both SCALE6.3.2 and MCNP6.2. In SCALE, we ran simulations using both a modified ENDF/B-VIII.0 library—adjusted to include ENDF/B-VIII.1 20% porosity and Sd TSLs—and the standard ENDF/B-VIII.1 library. It is important to note that the SHIFT module in SCALE6.3.2 does not support the format of the Sd TSL, which affects some benchmark calculations. In MCNP6.2, calculations were conducted with the modified ENDF/B-VIII.0 library (incorporating the ENDF/B-VIII.1 20% porosity and Sd TSLs). This dual-code approach not only facilitates cross-code comparison but also compensates for cases where input files were available in

only one of the codes, thereby ensuring comprehensive coverage of the selected benchmarks. Note that the SCALE and MCNP models can differ based on how they were constructed. In this study, the MCNP models were taken as-is from the ICSBEP benchmarks, with only the nuclear data library version (ENDF/B-VIII.0) altered.

3.5.1. HTR-10 benchmark

HTR-10 is a 10 MWth (megawatt thermal) pebble-bed reactor at the Institute of Nuclear and New Energy Technology (INET), Tsinghua University, designed to demonstrate and further develop high-temperature reactor (HTR) technology. For its first critical trial, operators loaded a total of 16,890 spherical pebbles—9,627 fueled and 7,263 “dummy” graphite-only—into a cylindrical cavity surrounded by graphite reflectors. Although ultimately intended for helium coolant, the reactor’s cold startup experiment took place in air at near-ambient pressure. Each 6 cm diameter fuel pebble has a 5 cm core of tri-structural isotropic (TRISO)-coated UO_2 particles within a graphite matrix at 17 wt% enrichment. Roughly 8,335 TRISO particles appear in each fuel sphere, whereas the dummy pebbles of the same overall size are pure graphite of higher mass density. Surrounding the active region are graphite reflectors of about 1.76 g/cm³ density, plus an outer boronated carbon layer (≈ 1.59 g/cm³) along the vessel boundary to reduce neutron leakage. From these stated densities and by adopting 2.26 g/cm³ as the nominal crystalline graphite baseline, the bulk porosities of each graphite component can be approximated. For example, the fuel pebble matrix at 1.73 g/cm³ ends up at around 23.5% porosity (assuming perfect crystalline density of 2.26 g/cm³), whereas the dummy pebble graphite (1.84 g/cm³) is closer to 18.6%. The larger reflector blocks, at 1.76 g/cm³, imply about 22% porosity, and the boronated carbon bricks at 1.59 g/cm³ approach nearly 30%. The graphical representation of the SCALE model for HTR-10 can be seen in Figure 23.

Neutronics calculations were performed with SCALE6.3.2—employing both ENDF/B-VIII.0 and ENDF/B-VIII.1 libraries—and with MCNP6.2 using the ENDF/B-VIII.0 library. A summary of the k_{eff} calculation results is provided in Table 5.

Table 5: Comparison of calculated k_{eff} values for the HTR-10 benchmark.

TSL	SCALE ENDF/B-VIII.0		SCALE ENDF/B-VIII.1		MCNP ENDF/B-VIII.0	
	k_{eff}	Unc.	k_{eff}	Unc.	k_{eff}	Unc.
Sd	1.00650	0.00024	1.00572	0.00024	1.00722	0.00008
Crystalline	1.00607	0.00024	1.00586	0.00024	1.00678	0.00008
10% poro.	1.00987	0.00024	1.00965	0.00024	1.01018	0.00008
20% poro.	1.01129	0.00024	1.01070	0.00024	1.01181	0.00008
30% poro.	1.01203	0.00024	1.01220	0.00024	1.01321	0.00008
exp.	1.00000	0.00370				

Several observations can be made from Table 5. First, the performance using ENDF/B-VIII.0 and ENDF/B-VIII.1 libraries (with the same TSL) is generally consistent, with differences on the order of approximately 50 pcm (per cent mille, where 1 pcm = 1×10^{-5}). The agreement between SCALE and MCNP models is also relatively good, which is in line with expected differences arising from variations in nuclear data pro-

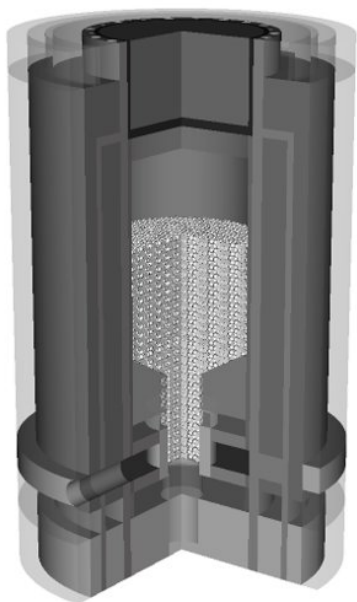


Figure 23: The SCALE model of HTR-10.

cessing, the implementation of physics, and model construction.

Another notable observation is that switching from crystalline TSL to a 30% porosity TSL results in an increase in the calculated k_{eff} values. Figure 17 indicates that the main differences between these TSLs lie in the inelastic scattering cross section, which is influenced by the underlying phonon spectra (i.e., $S(\alpha, \beta)$). The INS measurements show that the calculated $S(\alpha, \beta)$ values for porosity TSLs are overestimated and result in the increased inelastic scattering cross section, which in turn increases the calculated k_{eff} values. With the increased inelastic scattering cross section, as more neutrons thermalize (i.e. slow down or lose energy), their probability of being absorbed in the fuel and causing fission increases, thus yielding higher k_{eff} values. For example, the calculated k_{eff} for the 30% porosity TSL is 1.01203, which is 1203 pcm above the measured value of 1 (with an uncertainty of 370 pcm). These increases in the inelastic scattering cross section, and the resulting increased k_{eff} values, are unphysical. The typical slight increase in the k_{eff} values for Sd graphite (higher inelastic scattering cross section), when compared to crystalline, is physical due to Sd TSLs' better representation of INS and total cross section measurements.

3.5.2. HTTR benchmark

The High-Temperature Engineering Test Reactor (HTTR) at the Japan Atomic Energy Agency is a 30 MWth, prismatic-block, graphite-moderated, helium-cooled reactor. It was built to push forward high-temperature gas-cooled reactor (HTGR) technology and to host a variety of irradiation tests. About half the size of a future commercial HTGR, it maintains a large enough reactivity margin to handle temperature, xenon, and burnup effects during power operation.

During startup core physics tests, JAEA formed annular (partial) cores with 19, 21, 24 (2 configurations), and 27 columns

loaded from the radial periphery inward. These configurations helped measure the approach to critical, control-rod positions, reactivity increments, and reaction-rate profiles. Eventually, they moved to a fully loaded core with 30 columns of hexagonal blocks, enabling measurements of overall critical parameters, temperature coefficients, rod worths, subcritical margins, and axial flux distributions. Each column stacks hexagonal graphite blocks about 58 cm tall and 36 cm across the flats, some carrying 31 or 33 cylindrical fuel rods, others serving as reflectors or control channels. Each fuel rod holds 14 annular fuel compacts, and each compact has thousands of TRISO-coated UO_2 particles embedded in a graphite matrix. Typical fuel enrichments range from 3.4 wt% ^{235}U up to 9.9 wt% in different radial/axial zones to flatten power. The density of the graphite matrix is around 1.75 g/cm³—about 22–23% porous. The TRISO particles themselves are roughly 920 μm in outer diameter, each with a 600 μm UO_2 kernel surrounded by multiple carbon and silicon-carbide layers. The fully loaded core is surrounded by replaceable reflectors (IG-110 graphite) and an outer permanent reflector (PGX graphite at 1.73 g/cm³, 23.5% porosity). The graphical representation of SCALE model for fully loaded HTTR core can be seen in Figure 24.

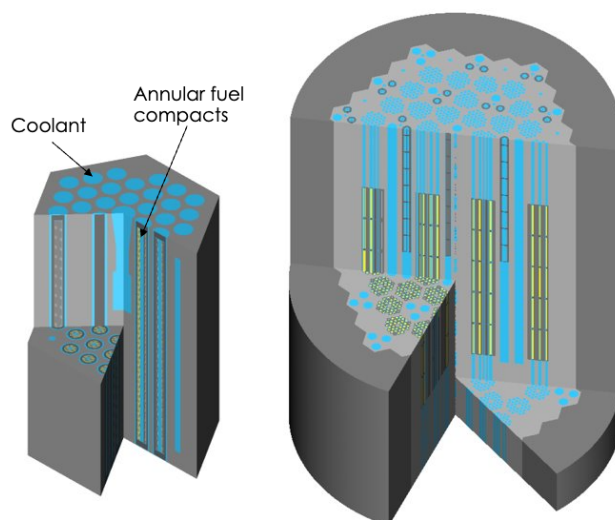


Figure 24: The SCALE model of fully loaded HTTR core (right) and HTTR fuel block (left).

Neutronics calculations of critical fully loaded core were performed with SCALE6.3.2 – employing both ENDF/B-VIII.0 and ENDF/B-VIII.1 libraries – and with MCNP6.2 using the ENDF/B-VIII.0 library. A summary of the k_{eff} calculation results is provided in Table 6.

As with the HTR-10 results, only minor differences appear between ENDF/B-VIII.0 and ENDF/B-VIII.1 nuclear data libraries. Consequently, for the remaining benchmarks, we focus on ENDF/B-VIII.0 results. We do note a slightly larger discrepancy between the SCALE and MCNP models in the fully loaded core case. As with HTR-10, moving from a crystalline to 30% porosity TSL raises k_{eff} ; the jump between crystalline and Sd TSL is relatively small, whereas the 30% porosity TSL

Table 6: Comparison of calculated keff values for the full loaded critical HTTR core configuration.

TSL	SCALE ENDF/B-VIII.0		SCALE ENDF/B-VIII.1		MCNP ENDF/B-VIII.0	
	keff	Unc.	keff	Unc.	keff	Unc.
Sd	1.01102	0.00024	1.01052	0.00024	1.01784	0.00011
Crystalline	1.01093	0.00024	1.01016	0.00024	1.01758	0.00011
10% poro.	1.01419	0.00024	1.01384	0.00024	1.02065	0.00011
20% poro.	1.01558	0.00024	1.01575	0.00024	1.02257	0.00011
30% poro.	1.01699	0.00024	1.01726	0.00024	1.02396	0.00011
exp.	1.0025	0.00710				

is roughly 600 pcm above the crystalline one. Even though the calculated keff values deviate from the experimental values, similar to HTR-10 benchmark, it is clear that there is a clear negative trend when using porosity TSLs, and it is also clear that the miss-prediction of the calculated values cannot be solely attributed to the TSLs.

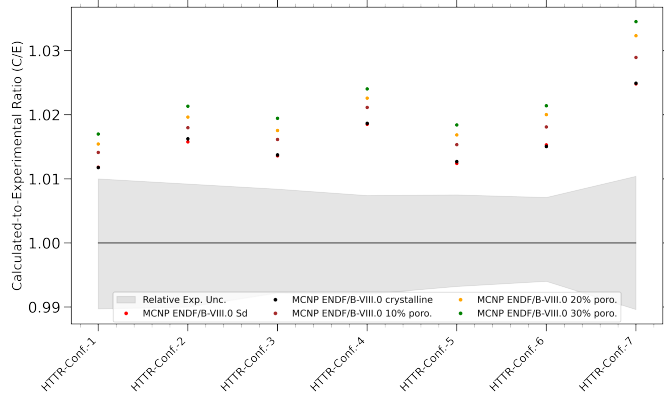


Figure 25: Calculated (using MCNP6.2) over experimental (C/E) keff values for different HTTR configurations.

Calculated (using MCNP6.2) over experimental (C/E) keff values for different HTTR configurations can be seen in Figure 25. A similar negative trend of the increase of keff values for porosity TSLs can be observed.

3.5.3. HTR-PROTEUS benchmark

In HTR-PROTEUS, a zero-power test reactor at the Paul Scherrer Institut in Switzerland, the core region is a large 22-sided graphite annulus (≈ 3.3 m high, 3.26 m across) with a central cavity that can be loaded with pebble-type fuel and moderator elements. Over several years (1992–1996), ten major core configurations (Cores 1–10) were assembled in distinct ways, creating separate IRPhEP benchmarks. These include hexagonal close-packed lattices (Cores 1–3) at a 1:2 moderator-to-fuel pebble ratio, random-packing (Core 4) at 1:1, columnar hex lattices (Cores 5–8) again at 1:2, and finally columnar hex lattices (Cores 9–10) at 1:1. In hexagonal close-packed (HCP) pebble lattices, each new layer of spheres (fuel or moderator pebbles) nestles into the “grooves” formed by three adjacent spheres in the layer below. This leads to the densest possible stacking of uniform spheres, at about 74% packing fraction. By contrast, columnar hex (sometimes called “point-on-point”) packing places each new layer directly above the spheres in the layer

below, with no horizontal offset. Although this configuration is easier to hand-load in an experimental facility, it yields a lower packing fraction—typically around 60%—because each vertical column remains aligned straight upward with modest lateral gaps between columns. Many of the core configurations also inserted polyethylene rods of different sizes to simulate water ingress. Reactor shutdown and control relied on eight borated steel rods around the perimeter plus four fine “control rods” (stainless steel). Most of the graphite reflectors and pebbles in HTR-PROTEUS have densities in the 1.68–1.78 g/cm³ range, implying porosities between about 21% and 26%. MCNP input models for different core configurations were taken directly from IRPhEP handbook. The results for C/E keff values can be seen in Figure 26.

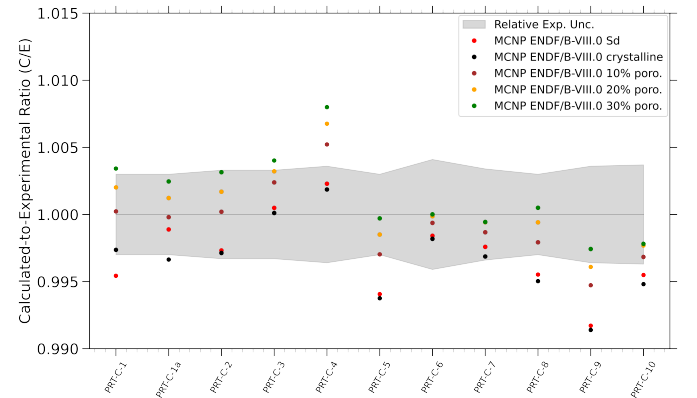


Figure 26: The resulting values for calculated over experimental (C/E) keff values for different HTR-PROTEUS core configurations using MCNP6.2.

A central observation from Figure 26 is that HTR-PROTEUS configurations show the same tendency of higher keff values when using porosity TSLs. Based on INS and total cross section measurements, however, this increase is not physically warranted. Indeed, if one were to calculate χ^2 values for different TSLs in the HTR-PROTEUS benchmark, the 10% porosity TSL might seem to agree best with experimental data, but this highlights the drawback of relying solely on integral benchmarks to decide which TSL is “better”. In the absence of differential (INS) or quasi-integral (total cross section) data, integral tests, while invaluable for flagging issues in nuclear data, should not serve as the ultimate judge of TSL accuracy—particularly when one TSL set aligns more strongly with differential and quasi-integral data.

Figure 26 also reveals an interesting trend involving polyethylene rods. In configurations 3, 6, 7, 8, and 10, where polyethylene rods simulate water ingress, the spread in keff results is narrower than in cores without polyethylene rods. Core 10, for instance, repeats Core 9’s geometry but adds 6.5 mm poly rods. Because hydrogen’s high incoherent inelastic scattering helps offset the effects of porosity TSLs, these setups exhibit reduced sensitivity to the problematic rise in inelastic scattering cross section in the porosity TSLs. At the same time, introducing hydrogen increases the overall keff values by improving neutron thermalization.

3.5.4. ICT-008 benchmark

In ICT-008 (IEU-COMP-THERM-008), the ASTRA facility experiments aimed to validate annular-core HTR physics using pebble-type TRISO fuel in a graphite-reflector system. The core is composed of a central “internal” graphite reflector, an annular ring of spherical fuel pebbles (each roughly 6 cm in diameter, at $\approx 21\%$ ^{235}U enrichment), and a thick “side” (external) graphite reflector. An optional top reflector, also formed of graphite spheres, can be installed above the fuel bed. These experiments—ranging from short to tall core loadings—charted how criticality conditions shift under variations in bed height, control rod positioning, poison rod insertions, and the presence or absence of the top reflector. Within the five benchmark configurations, the total number of pebbles changes from $\approx 16,897$ up to $\approx 30,432$, and the bed height rises from ≈ 180 cm to ≈ 320 cm. Configurations 1–4 have no graphite top reflector (thus air above the bed), whereas Configuration 5 adds a ≈ 100 cm-thick top reflector of graphite spheres. Each arrangement uses consistent graphite reflectors at densities of $\approx 1.66\text{--}1.69\text{ g/cm}^3$, corresponding to $\approx 25\text{--}27\%$ porosity.

The SCALE model inputs were generated according to section 3 of the benchmark report, which specifies how to reconstruct the official benchmark models. The resulting C/E keff values are shown in Figure 27. As in earlier benchmarks, the ICT-008 configurations display the same pattern of higher keff predictions when porosity TSLs are used. This increase stems from the elevated inelastic scattering cross sections associated with the porosity TSL files—an effect demonstrated here to be unphysical.

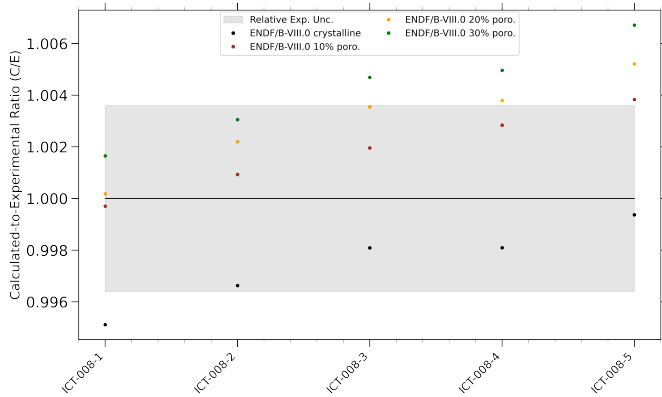


Figure 27: The resulting values for calculated over experimental (C/E) keff values for different ICT-008 core configurations using SCALE6.3.2.

3.5.5. HCT-016 benchmark

In the IGR (“Impulse Graphite Reactor”) reactor (HCT-016, HEU-COMP-THERM-016 benchmark), the core consists of graphite columns, many of which are impregnated with highly enriched uranium ($\approx 90\%$ ^{235}U) at about 3.1 g of U per kg of graphite. These fuel-impregnated columns, interspersed with unimpregnated (pure) graphite columns, are arranged in a roughly cubic stack ($\approx 140 \times 140$ cm cross section). A movable central portion can be raised or lowered to adjust reactivity in concert with several Gd_2O_3 -based control rods. Six critical

configurations are documented, and each is designated by a specific core temperature (from 13°C up to 794°C), rod insertions, and position of the movable stack. Because the graphite blocks have very little uranium by volume, even small changes in impurities (particularly boron) or geometry can significantly affect keff. Based on a theoretical graphite density of 2.26 g/cm^3 , the fuel-impregnated blocks at 1.71 g/cm^3 exhibit about 24.3% porosity, and the unimpregnated reflector blocks at 1.65 g/cm^3 imply roughly 27% porosity. A top cross-sectional view of the IGR core and reflector is given in Figure 28. As can be seen from the figure, there is a substantial amount of graphite, colored in gray, in the benchmark. A summary of the temperatures for each case configuration is given in Table 7.

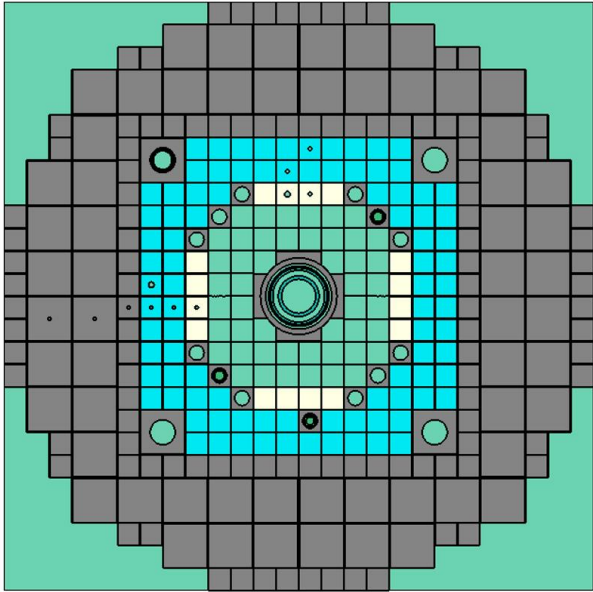


Figure 28: top cross sectional view of IGR core and reflector, graphite in the system was colored grey.

Critical Configuration	1	2	3	4	5	6
Temperature, $^\circ\text{C}$	18	13	50	27	476	794

Table 7: Temperature of uranium-graphite blocks in the HCT-016 benchmark model.

The SCALE model inputs were generated according to section 3 of the benchmark report, which specifies how to reconstruct the official benchmark models. The resulting C/E keff values are shown in Figure 29. As in previous benchmarks, the HCT-016 configurations at near-room temperature exhibit a trend of higher keff predictions when porosity TSLs are used. In contrast, for the high-temperature configurations, an inverse trend is observed, with a much narrower spread in keff values. At elevated temperatures, the inelastic scattering cross section increases considerably, so the relative impact of changes in this cross section is minimized.

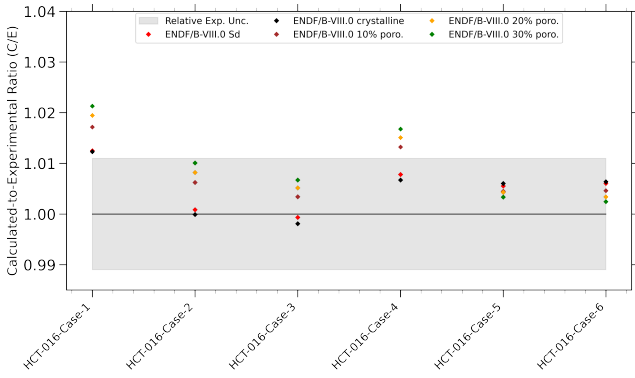


Figure 29: The resulting values for calculated over experimental (C/E) keff values for different HCT-016 core configurations using SCALE6.3.2.

3.5.6. LCT-060

In LEU-COMP-THERM-060 (LCT-060), the RBMK critical facility experiments at the Kurchatov Institute (1982–1988) created a half-height, zero-power version of the graphite-moderated RBMK reactor. The assembly includes an 18 x 18 grid of tall graphite columns (≈ 410 cm), each with a 114 mm bore, allowing insertion of fuel assemblies (at 1.8%, 2.0%, or 2.4% ^{235}U enrichment) or other materials (e.g., boron-steel rods, thorium absorber slugs, or water). Across the 28 configurations, investigators systematically varied (1) the fuel enrichments, (2) whether channels were left empty (air-filled) or flooded with water, and (3) the presence of absorbers (boron steel or thorium). These changes revealed how the reactivity responds to each factor. Over the course of achieving critical or near-critical states, reactivity data, control worths, and flux distributions were measured at room temperature ($\approx 20^\circ\text{C}$). Turning to the graphite properties, the moderator blocks in the RBMK critical facility typically have densities around $1.71\text{--}1.73\text{ g/cm}^3$, although each block's mass and dimensions can vary slightly. Using 2.26 g/cm^3 the densities correspond to roughly 23–25% porosity.

The SCALE model inputs were generated according to section 3 of the benchmark report, which specifies how to reconstruct the official benchmark models. The resulting C/E keff values are shown in Figure 30.

As shown in Figure 30, two distinct trends emerge. For cores without water in the channels, the expected trend is observed: keff increases with the use of porosity TSLs. Conversely, for cores with water-filled channels, an inverse trend appears, with keff decreasing as porosity TSLs are applied. Water in the channels further thermalizes the neutrons; when combined with the increased inelastic scattering cross section in the porosity TSLs, this extra thermalization leads to enhanced absorption in the surrounding materials, ultimately reducing keff.

3.5.7. Impact of SANS on criticality

In order to assess the impact of SANS physics on keff, we have utilized OpenMC with the NCrystal SANSND plugin. OpenMC as of version 0.15.2 supports the use of NCrystal thermal scattering materials. SANS components replicating ET-34,

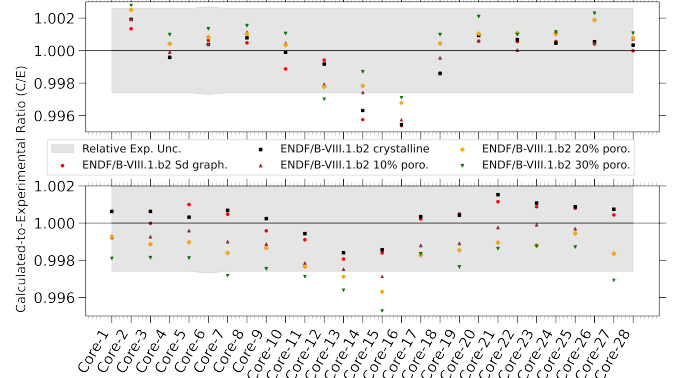


Figure 30: The resulting values for calculated over experimental (C/E) keff values for different LCT-008 core configurations using SCALE6.3.2. The bottom plot shows the core configurations with water-flooded channels, and the top plot shows core configurations without water.

IG-43, and Petriw needle coke graphite cross sections, as seen in Figure 22, were utilized. These cross sections represent the upper, middle, and lower bounds of the measured cross sections.

First, an analytical benchmark based on the GODIVA fast benchmark was employed. The HEU-MET-FAST-001 (Godiva) benchmark is a critical experiment conducted using a bare, highly enriched uranium (HEU) sphere, designed specifically to measure the critical mass of an unmoderated and unreflected uranium system. Commonly referred to as “Lady Godiva”, this configuration comprises nested hemispherical uranium shells forming a sphere approximately 6.883 inches (17.48 cm) in diameter, with a total mass of about 52.42 kg and a density near 18.74 g/cm^3 . The uranium employed is highly enriched, containing roughly 94 wt.% ^{235}U , along with small fractions of ^{234}U and ^{238}U . This benchmark has significant relevance in validating neutron transport codes and nuclear data libraries, as it offers an unmoderated, purely fast neutron spectrum. Two main computational models were used. One features a simplified solid uranium sphere and the other uses detailed nested shells, allowing thorough cross-verification of simulation results. Historically, the Godiva benchmark has been extensively used to verify and validate computational methods and nuclear data sets such as ENDF libraries, MCNP, SCALE, and other neutronics analysis codes. Its precise criticality data ($\text{keff} \approx 1.000 \pm 0.001$) makes it a cornerstone reference for criticality safety involving fast neutron systems.

In order to assess the impact of SANS as well as various TSL approaches, and to validate the NCrystal implementation in OpenMC, the composition of each layer in the detailed nested Godiva model was modified to include around 99.9wt.% graphite and reduced HEU until the effective multiplication factor, keff, converged to approximately 1.000 ± 0.001 . Although the corresponding material compositions do not reflect a real system, the modified configuration represents a simple system that is sensitive to graphite nuclear data. Figure 31 shows the resulting SCALE model of the Godiva benchmark, while Figure 32 plots the calculated flux per lethargy at the system's center.

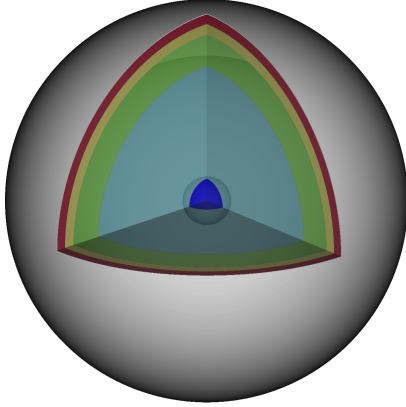


Figure 31: SCALE model of Godiva's detailed nested-shells configuration.

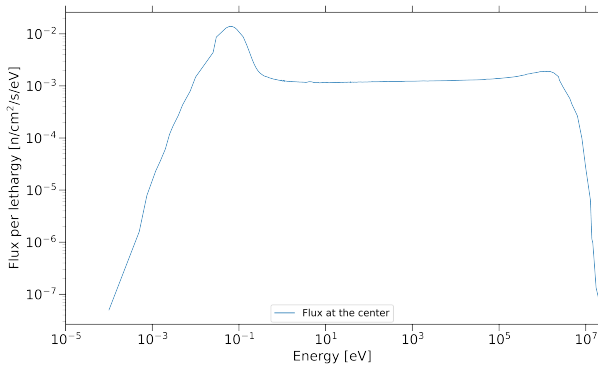


Figure 32: Flux per lethargy at the center of the Godiva configuration.

Table 8 summarizes the calculations that were performed in OpenMC with ENDF/B-VIII.0 data. The comparison includes the OpenMC standard ENDF library TSLs, which comprise the crystalline variant, porosity variants at 10%, 20%, and 30%, and the corresponding NCrystal representations of both crystalline and porosity TSLs. It also includes cases where small-angle neutron scattering (SANS) physics is added to the NCrystal crystalline model.

Table 8: Comparison of calculated k_{eff} values for the modified GODIVA benchmark.

TSL	$k_{eff} \pm \text{unc.}$
Crystalline	1.00080 ± 0.00032
10% poro.	1.00518 ± 0.00032
20% poro.	1.00793 ± 0.00031
30% poro.	1.00774 ± 0.00031
OpenMC+NCrystal Crystalline	1.00120 ± 0.00032
OpenMC+NCrystal 10% poro.	1.00728 ± 0.00032
OpenMC+NCrystal 20% poro.	1.00780 ± 0.00032
OpenMC+NCrystal 30% poro.	1.00944 ± 0.00031
OpenMC+NCrystal Crystalline + Petriw needle SANS	1.00120 ± 0.00032
OpenMC+NCrystal Crystalline + IG-43 SANS	1.00225 ± 0.00032
OpenMC+NCrystal Crystalline + ET-34 SANS	1.00287 ± 0.00032

Several observations arise from the data in Table 8. First, the ENDF/B-VIII.0 crystalline TSL and the NCrystal crystalline treatment produce very similar results for k_{eff} , differing by only about 40 pcm. Because both calculations feature statis-

tical uncertainties on the order of 30 pcm, this close agreement indicates that NCrystal adequately handles thermal scattering physics. A second key point is that the porosity-based TSLs significantly increase the reactivity, consistent with trends observed for the other benchmarks. Transitioning from a crystalline TSL to a 30% porosity variant raises k_{eff} by approximately 700–800 pcm in the standard ENDF data. The NCrystal porosity TSLs exhibit a similar effect. At 30% porosity, the increase is slightly higher, approaching 900 pcm above the crystalline baseline.

Lastly, when SANS physics is added to the baseline crystalline TSL, the effect on k_{eff} remains fairly modest. In the most extreme case (ET-34), SANS boosts k_{eff} by about 160 pcm relative to a purely crystalline baseline. For the IG-43 cross section, the increase is around 120 pcm. In contrast, for Petriw's needle-coke data, no appreciable change in k_{eff} is observed. Although SANS had a limited impact on k_{eff} in this theoretical case, such conditions are highly unlikely to be encountered in actual reactor or critical systems applications.

To assess the impact of SANS on realistic systems, we used the MCNP conversion tools for OpenMC [60] to translate MCNP input files into OpenMC format. The input files for the HCT-016 benchmark (cases 1, 2, 3, and 4), the HTR-PROTEUS benchmark case 4, and the HTR-10 benchmark were successfully translated.

As shown in Table 9, the calculated k_{eff} values from the MCNP calculations using the ENDF/B-VIII.0 crystalline TSL and the OpenMC calculations (converted from MCNP input files) using NCrystal to model the same system demonstrate fairly good agreement. The differences between these calculations are on the order of 200 pcm. More notably, the impact of SANS is generally around 50 pcm, with the Petriw needle SANS model showing negligible impact. Given the calculation uncertainty of approximately 30 pcm, these variations are not statistically significant. Generally, SANS is not expected to significantly affect nuclear and critical systems because the SANS cross section is dominant in a very low energy region, typically below 10 meV. The selected benchmarks, ranging from a highly theoretical modified Godiva case to graphite-moderated and reflected systems (such as HCT-016), and further to graphite-reflected systems with TRISO fuel (like HTR-10 and Proteus Case 4), provide a wide basis for studying the impact of SANS on the effective multiplication factor. Graphite moderated and reflected nuclear reactors and critical systems have a very low neutron population at these energies, resulting in a small SANS impact that is negligible in some cases. Therefore, it is reasonable to conclude that, at least for nuclear reactor and criticality safety operations, porosity in nuclear graphite does not strongly impact the neutron multiplication factor. While SANS could be relevant in neutron scattering applications, such as the design of neutron scattering guides or cold and very cold neutron sources, nuclear graphite is not typically used in these contexts.

3.5.8. Summary of benchmark calculations

This section examines the impact of different TSL evaluations for graphite in critical benchmark calculations drawn

Table 9: Comparison of calculated keff values for different benchmarks. Uncertainty is on the order of 30 pcm.

TSL	HCT-016 Case 1	HCT-016 Case 2	HCT-016 Case 3	HCT-016 Case 4	HTR-10	Proteus Case 4
MCNP Crystalline	1.00538	1.00104	1.00219	1.00587	1.00678	1.00577
OpenMC+NCrystal Crystalline	1.00778	1.00348	1.00360	1.00711	1.00753	1.00493
OpenMC+NCrystal Crystalline + Petriw needle SANS	1.00778	1.00348	1.00360	1.00711	1.00686	1.00493
OpenMC+NCrystal Crystalline + IG-43 SANS	1.00754	1.00355	1.00372	1.00695	1.00768	1.00500
OpenMC+NCrystal Crystalline + ET-34 SANS	1.00744	1.00294	1.00362	1.00707	1.00715	1.00515
exp.	1.00480	1.00400	1.00350	1.00320	1.00000	1.00390

from both the ICSBEP and IRPhE handbooks. The effective multiplication factor, keff, served as the primary metric for comparing simulation predictions (via SCALE6.3.2 and MCNP6.2) against measured criticality data. Each benchmark configuration—ranging from pebble-bed reactors (HTR-10, HTR-PROTEUS, HTTR, and ICT-008) to graphite stacks with varying enrichments (HCT-016, LCT-060)—includes substantial amounts of graphite moderator and reflector with densities typically in the 1.65–1.84 g/cm³ range (i.e., 18–27% porosity).

In most cases, substituting a porosity-based TSL for crystalline graphite systematically increases keff, often pushing calculated values well above experimental measurements. This behavior stems from an overestimation of the inelastic scattering cross section in the porosity TSL evaluations, leading to increased neutron thermalization and, consequently, a higher likelihood of fission. While the Sd TSL produces only a slight keff increase over crystalline graphite (in line with differential measurements), higher-porosity TSLs (10%, 20%, 30%) consistently drive larger, unphysical shifts in criticality. These trends are seen in both SCALE and MCNP calculations. Furthermore, configurations that include additional moderating elements—such as polyethylene rods or water-filled channels—reduce the relative impact of the porosity TSLs because hydrogen’s strong thermalization partially masks the overstated graphite inelastic cross section.

By using OpenMC and NCrystal, the impact of SANS on the effective multiplication factor has been studied, and it has been observed that the impact is minimal.

Critically, although integral benchmarks like those from ICSBEP and IRPhE are indispensable in validating full-system behavior, they cannot unambiguously pinpoint errors in specific nuclear data components, particularly in TSLs. Most of the time the results are inconclusive. Differential and quasi-integral experiments (e.g., INS and total cross section measurements) remain essential for isolating and validating individual scattering contributions.

4. Conclusion

This work demonstrates conclusively that the inherent porosity of nuclear graphite has no significant effect on its fundamental neutron moderation behavior, despite long-standing assumptions to the contrary. Our comprehensive study, integrating new inelastic neutron scattering measurements, SANS experiments, advanced atomistic simulations, and neutronics calculations, leads to the following key conclusions:

1. Porosity does not alter graphite phonon spectra: INS experiments on multiple graphite grades (spanning 10–25% porosity and diverse microstructures) revealed similar phonon DOS and scattering law $S(Q, \omega)$ compared to non-porous crystalline graphite. Even at cryogenic temperature (5 K) and high resolution, no additional vibrational modes or broadening attributable to pores were observed. Atomistic simulations using a DFT-based machine-learned potential confirmed that introducing realistic micropores leaves the phonon spectrum virtually unchanged from the crystalline case. These results directly contradict the assumptions built into the ENDF/B-VIII.1 “porosity” TSLs (10%, 20%, 30%), which were generated by randomly removing atoms in MD simulations and consequently exhibit unphysical distortions in the phonon spectrum. We find that those distortions are artifacts of the modeling procedure (vacancies and a less-accurate potential) rather than actual effects of porosity. Thus, the thermal scattering law for graphite should not be modified due to porosity insofar as vibrational dynamics are concerned.
2. Porosity introduces a separate SANS scattering component: While pores do not impact inelastic (phonon) scattering, they do give rise to significant elastic small-angle neutron scattering. Our transmission experiments and analysis of historical data show that porous nuclear graphite exhibits total cross sections at sub-thermal neutron energies an order of magnitude higher than perfect-crystal predictions. This excess is explained by neutrons undergoing numerous small-angle deflections on pore surfaces, effectively being removed from the forward beam (in transmission) without energy loss. We quantified the “SANS cross section” for various graphite grades and demonstrated that it can be accurately modeled by combining a crystalline graphite TSL with an additional elastic SANS scattering term. Using either analytical fits or direct SANS $I(Q)$ data (via NCrystal), we reproduced the large low-energy attenuation observed in graphite benchmarks with excellent accuracy. Importantly, this approach obviates any need to alter the phonon spectrum: a physically consistent description is one of a crystalline lattice (phonon scattering unchanged) plus a separate SANS contribution from the pore structure. This two-component model matches all measurements, whereas the ENDF porosity TSL approach (altering phonon scattering) fails to predict the magnitude of total cross sections and is not physically justifiable.

3. Criticality is largely insensitive to porosity (when modeled correctly): Our neutronics calculations on a wide range of critical benchmarks (pebble-bed reactors, prismatic reactors, graphite-moderated test lattices, etc.) indicate that properly modeled porosity has a small or negligible effect on the neutron multiplication factor k_{eff} . When porosity is represented via a SANS scattering kernel (added to a crystalline TSL), the change in k_{eff} for realistic systems is on the order of a few tens of pcm. This is because the SANS scattering predominantly affects very cold neutrons that contribute little to fission reactions in thermal systems. In stark contrast, the ENDF/B-VIII.1 porosity TSLs, by virtue of their altered inelastic scattering, artificially increased k_{eff} by several hundred pcm. These inflated values are non-physical and could lead to significant criticality safety or reactor design errors if used. For instance, our calculations showed spurious increases in k_{eff} for the HTR-10 and HTTR reactors, the PROTEUS experiments, and others when using the porosity-modified TSLs. Ultimately, when porosity is treated via SANS (which is the correct physical representation), its impact on reactor neutronics is negligible.
4. Recommendations for thermal scattering data and applications: In light of the above findings, we strongly recommend that thermal scattering law evaluations for graphite (such as future ENDF releases or JEFF/JENDL libraries) revert to using a crystalline Sd graphite TSL for all grades and avoid the introduction of “effective” porosity-induced modifications. Instead, if one needs to account for the influence of porosity, this should be done by including a small-angle scattering contribution. Modern Monte Carlo codes now have capabilities to handle such an added component. Our work provides validated SANS cross section models for several graphite types, which can serve as a basis. For typical reactor physics and criticality safety calculations, however, one may safely neglect SANS because its effect on k_{eff} is minimal. The priority should be to remove the unphysical phonon-based porosity effects from evaluated data to prevent potential biases. Doing so will ensure that simulations using these data (e.g., core eigenvalue predictions, flux spectra in graphite-moderated systems, etc.) remain accurate and aligned with reality.

In summary, we have shown that nuclear graphite can be treated as an effectively crystalline moderator with respect to its neutron thermal scattering behavior. The complex pore structure inherent to manufactured graphite, while highly relevant to mechanical properties and to neutron scattering experiments at very low energies, does not degrade or significantly change the material’s ability to thermalize neutrons in a reactor setting. This resolves a long-standing ambiguity and removes a potential source of conservatism or error in graphite-moderated reactor analyses. Going forward, if there is a case that is sensitive to SANS scattering, the modeling of reactor graphite should incorporate porosity in the form of SANS, but should retain the use of a physically sound crystalline thermal scattering law for all other aspects. By validating our nuclear data against both dif-

ferential measurements (INS, SANS) and integral experiments (critical benchmarks), we ensure a high degree of confidence in simulations of current and future graphite-moderated reactor systems. The findings here also highlight the value of machine-learned interatomic potentials in generating realistic materials data for nuclear applications, and the importance of revisiting legacy assumptions (like the impact of porosity) with modern experimental and computational tools.

Finally, while the effect of graphite porosity on neutron physics is largely negligible, our work does not diminish the importance of understanding graphite’s microstructure for other reasons. For example, porosity and crack structures play crucial roles in graphite’s mechanical strength, irradiation-induced dimensional changes, and thermal conductivity — factors that are vital for reactor core structural integrity and heat transfer. Those aspects must continue to be studied and managed in reactor applications. From a nuclear data perspective, however, we have untangled and resolved the porosity question, allowing future evaluations and applications to treat thermal neutron scattering in graphite with greater fidelity and confidence.

5. Acknowledgments

This work was performed under the framework of DOE / US Nuclear Regulatory Commission (NRC) collaboration for Criticality Safety support for the commercial-scale HALEU fuel cycles project (DNCSH).

This research used resources of the National Energy Research Scientific Computing Center (NERSC), a Department of Energy Office of Science User Facility using NERSC award ERCAP0033587.

This research used resources at the High Flux Isotope Reactor and the Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. The beam time was allocated to VISION on proposal number IPTS-30401.1, IPTS-31124.2, IPTS-32976.1, and IPTS-34177.

For the HTTR benchmark, John Bess, one of the benchmark evaluators, graciously provided the MCNP input models for the startup core configurations, as well as for the critical fully assembled core and the subcritical fully assembled core. We extend our thanks for his assistance.

The authors gratefully acknowledge the UK Science and Technology Facilities Council and the ISIS Facility for provision of neutron beam time via both direct access (Proposal ID: 2420073, VESUVIO) and Xpress access (Proposal ID: 2390171, VESUVIO) routes.

Statement: During the preparation of this work the author(s) used ChatGPT o1 in order to ensure clarity, coherence, and readability of content. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

6. Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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