Graphite: cautionary tale on the importance of inelastic neutron scattering and transmission measurements

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TPR2024 meeting, February 20–23, 2024

Atomistic calculations of phonon spectrum

- Molecular dynamics (MD), ab-initio molecular dynamics (AIMD), and density functional theory (DFT) calculations are used typically for phonon spectrum calculations. DFT calculations are basis for coherent one-phonon inelastic scattering physics for strong coherent scatterers like carbon in graphite.
- The basis for these calculations are inter-atomic potentials.
- The potentials are only approximations to actual solution of Schrodingers equation, and are tailored to reproduce specific experimental quantities.
- Reproducing specific experimental quantities (i.e. lattice constants) does not mean other quantities will be reproduced well.
- The initial structure for non-crystaline materials is relatively unknown, and it is usually dependent on the evaluators choice of inter-atomic potential and evaluators understanding of the material.
- **Surely with so many unknowns measurements need to be part of the evaluation and validation process?**

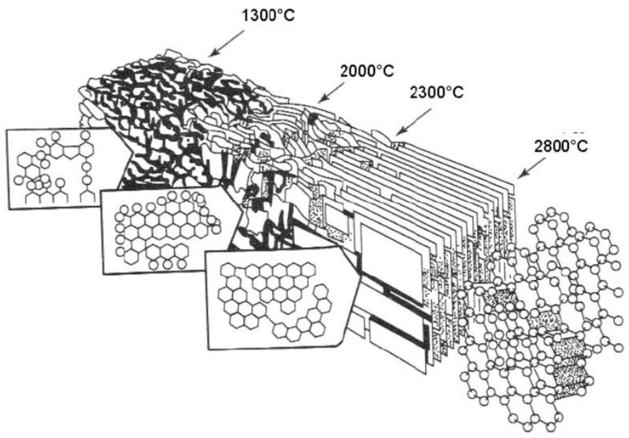
Are only atomistic calculations enough?

- ENDF/B-VIII.1.b2 has 5 different graphite TSL evaluations: crystalline, Sd-graphite, 10%, 20%, and 30% porosity reactor graphite

o *What is graphitization process?*

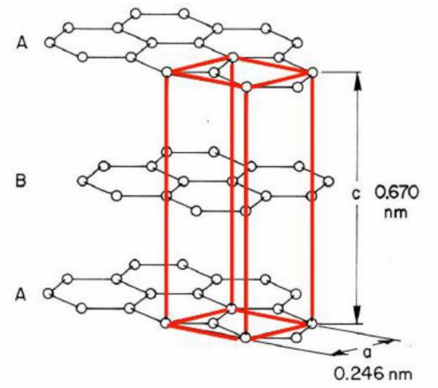
 - Graphitization is the process of heating amorphous carbon for a prolonged period of time, **rearranging the atomic structure to achieve an ordered crystalline structure** that is typical of solids.

Evolution of graphitization process, reproduced from [1]:

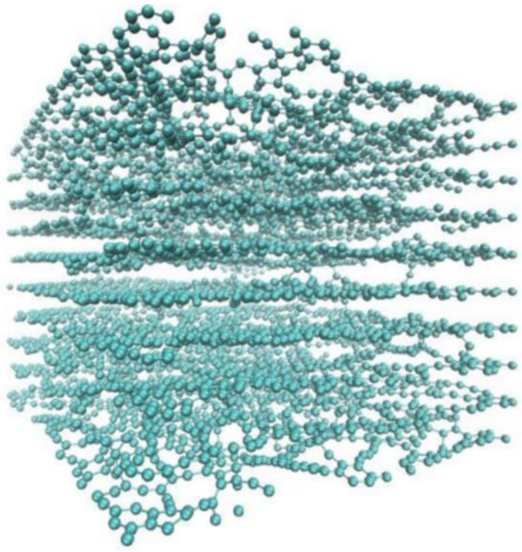


- **How does all this manifest itself in inelastic scattering measurements?**

- Crystal structure for Crystalline and Sd graphite [2]:



- Porous structure for 30% porosity graphite [2]:



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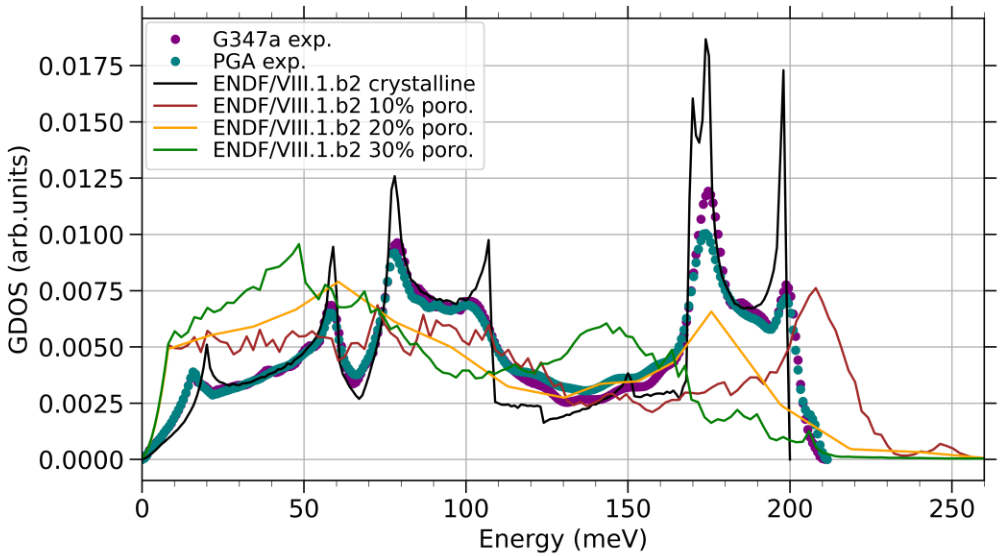
[1] Delhaes, P., ``Carbon Science and Technology - From Energy To Materials", Hoboken, NJ, John Wiley & Sons, Inc., pp.35-38., 2012

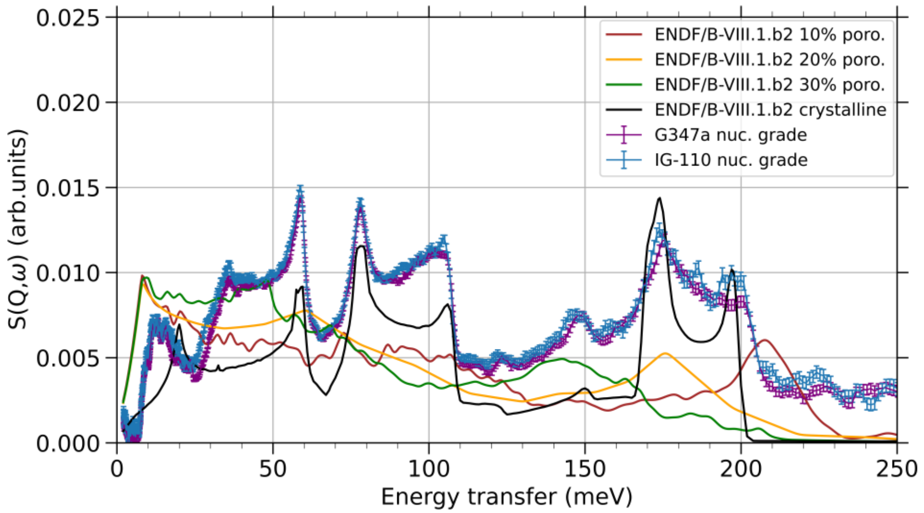
[2] A. Hawari and V. Gillete. ``Inelastic Thermal Neutron Scattering Cross Sections for Reactor-grade Graphite", Nuc. Data Sheets, 118:176–178, 2014

Inelastic neutron scattering (INS) measurements

- Phonon spectrum (GDOS) measurements at ARCS instrument at SNS ORNL:



- S(Q,W) measurements at VISION instrument at SNS ORNL:



In the incoherent and Gaussian approximation, the $S(\alpha,\beta)$, as expressed in NJOY LEAPR module, in terms of phonon expansion can be written as:

$$S(\alpha,\beta) = e^{-\alpha\lambda}\sum_{n=0}^{\infty}\frac{1}{n!}\alpha^n\frac{1}{2\pi}\times\int_{-\infty}^{\infty}e^{i\beta\hat{t}}\left[\int_{-\infty}^{\infty}P(\beta')e^{i\beta'\hat{t}}e^{-\beta'/2}d\beta'\right]^n d\hat{t}$$

where:

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)} \quad \text{and} \quad W = \frac{\int_{-\infty}^{\infty}P(\beta)e^{-\beta/2}d\beta}{AkT}$$

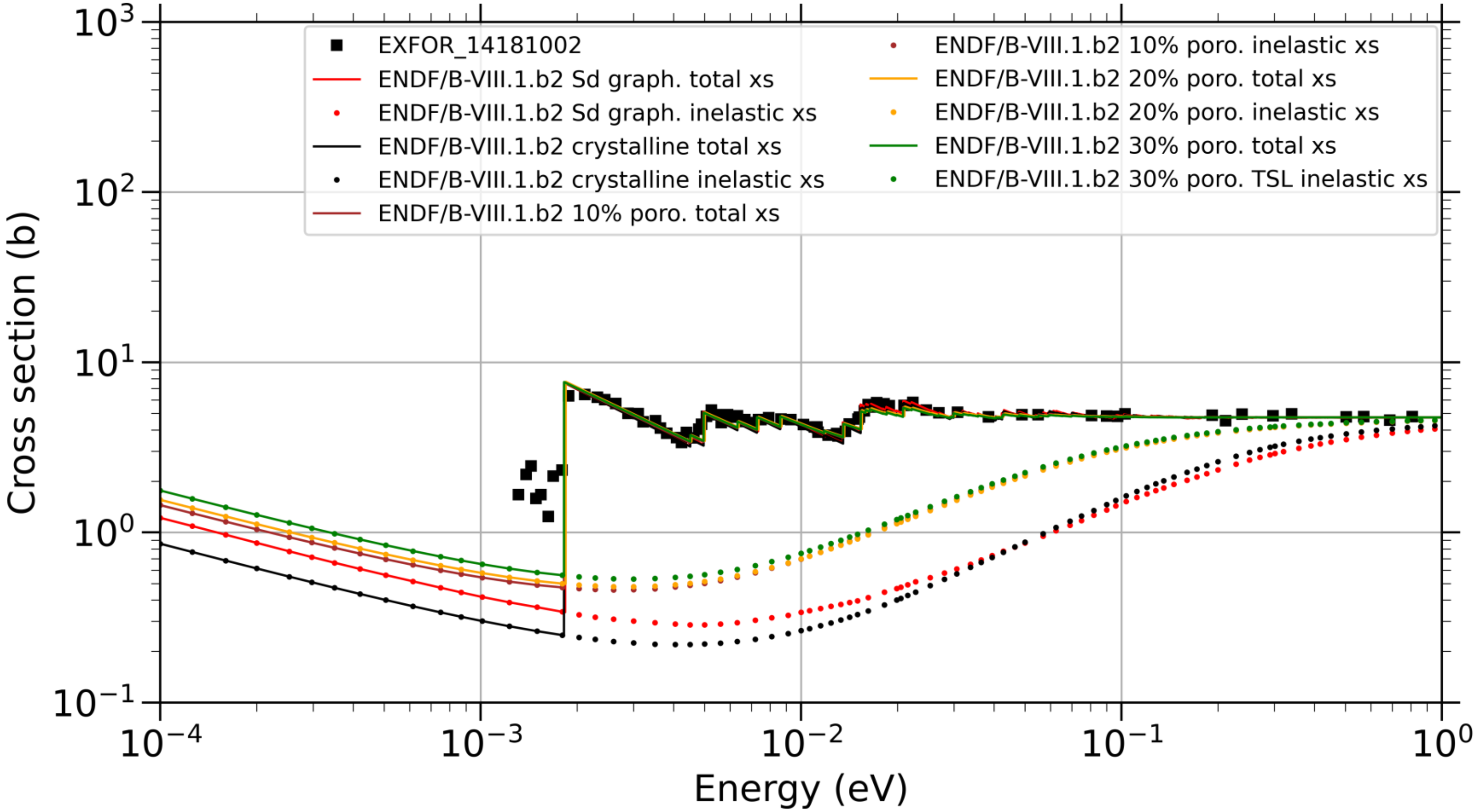
with $\rho(\beta)$ as the phonon spectrum.

Measured quantity $S(Q,\omega)$ is directly related to $S(\alpha,\beta)$:

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

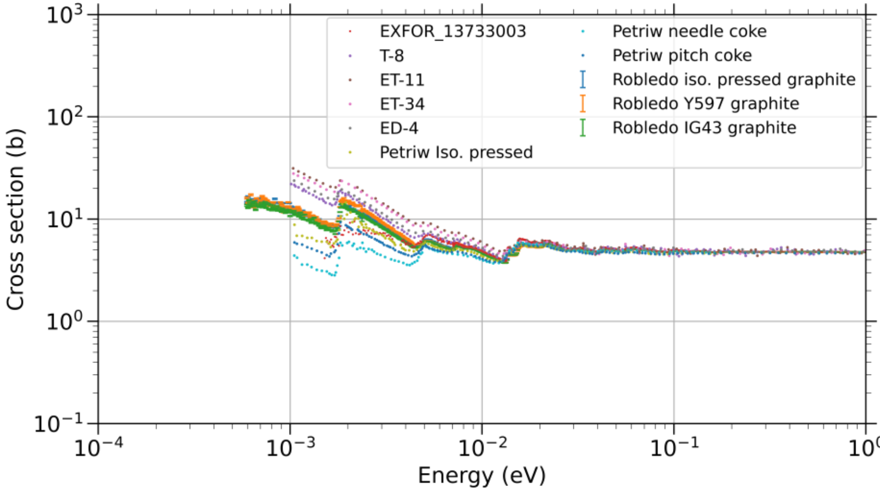
Graphite thermal transmission (total cross section) measurements

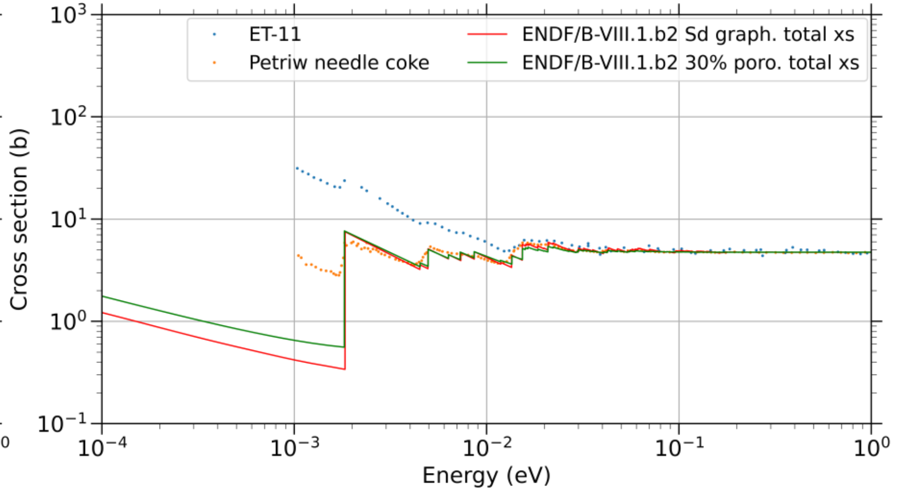
- ***Sd-graphite*** is the most physically accurate TSL from differential level
- The effect of porosity in 10%, 20%, and 30% TSLs was inaccurately modeled, which resulted in **increase of the inelastic scattering cross section**
- The actual effect of porosity is seen in **Small Angle Neutron Scattering (SANS)** cross section, and not in the inelastic cross section

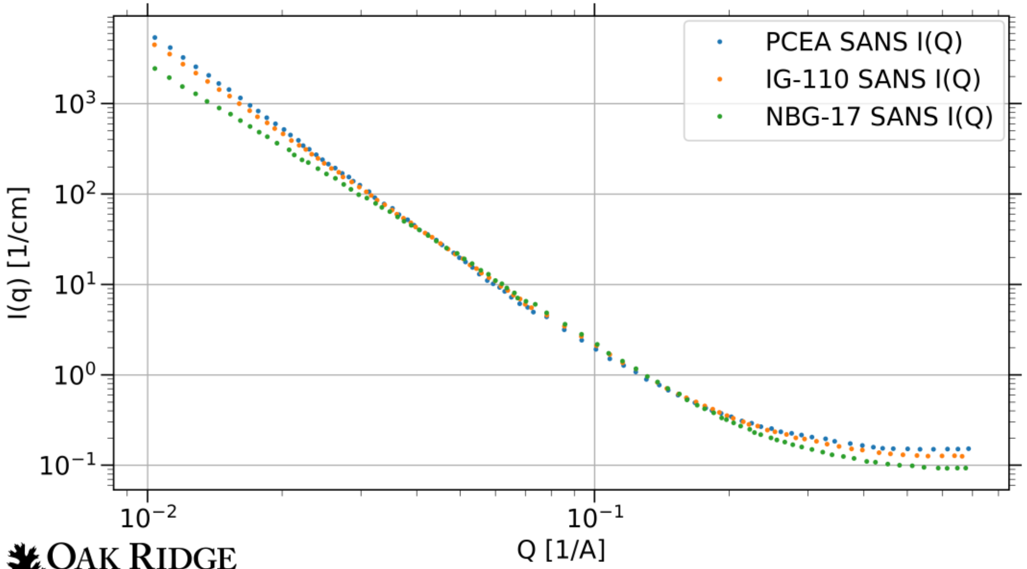


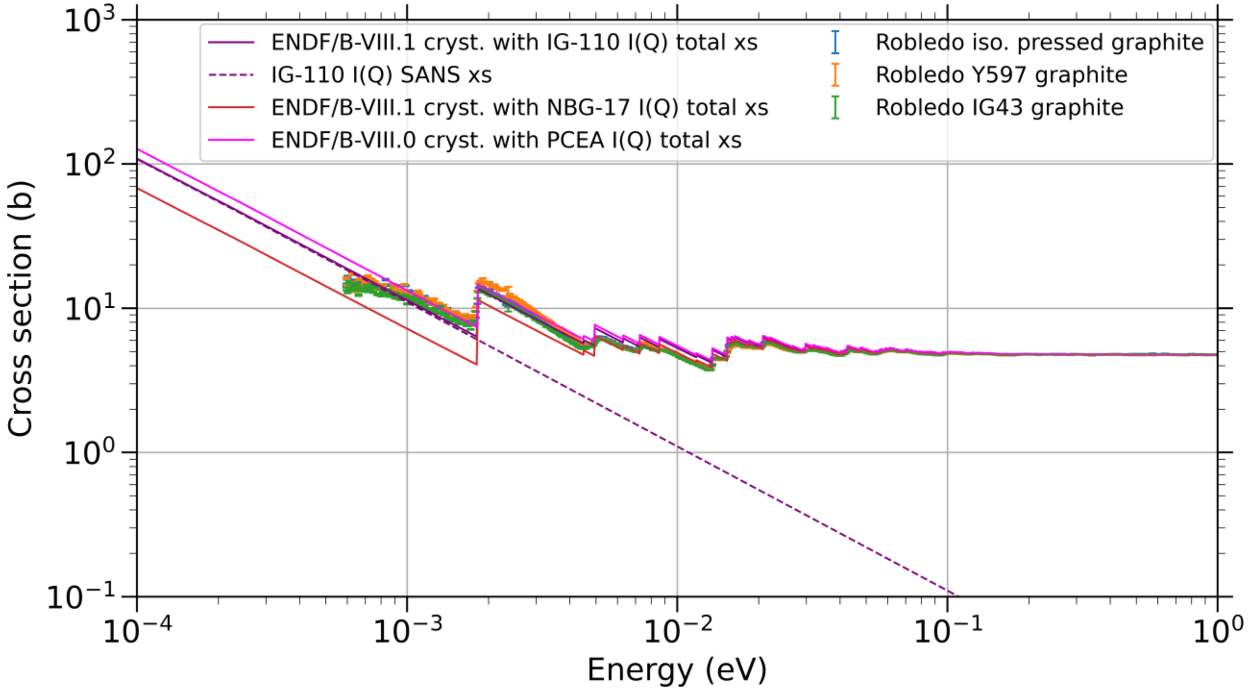
Graphite thermal transmission (total cross section) measurements

- ***There are multiple of transmission measurements on different grades of nuclear graphite that show impact of SANS, from Harvey in 1982, Petriw in 2010, Robledo in 2020, and Japanese measurements in 2022.***
- SANS is an elastic scattering (only change of direction)
- By measuring SANS of different grades of nuclear graphite we can reproduce their transmission.









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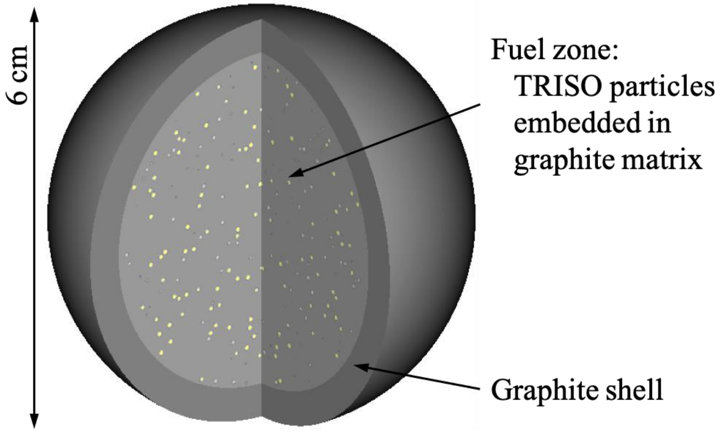
Tools

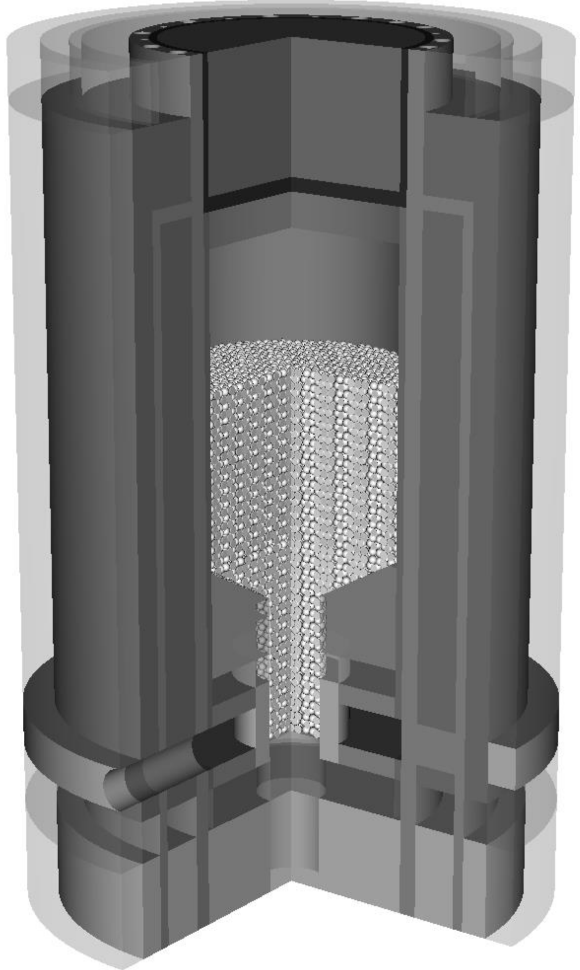
- Neutron transport using a recent beta version of SCALE 7:
  - KENO-VI Monte Carlo
  - Shift Monte Carlo
- MCNP6.2 with small angle neutron scattering physics
- Nuclear data libraries:
  - Continuous-energy library ENDF/B-VIII.0
  - ENDF/B-VIII.1.b2 TSL data:
    - Crystalline
    - Crystalline + Sd
    - 10% porosity
    - 20% porosity
    - 30% porosity
    - No TSL data (Carbon free gas)

Model 1: HTR-10

- 10 MWth Pebble-bed High Temperature Gas-cooled Reactor
- Relevant characteristics:
  - UO_2 fuel density: 10.4 g/cm^3
  - ^{235}U enrichment: 17 wt.%
  - TRISO packing fraction: ~9%
  - Number of particles per pebble: 8,385
  - Pebble radius: 3 cm (fuel zone: 2.5 cm)
  - Graphite densities indicate porosities between **19–30%**
    - Dummy pebbles: 18.6%
    - Fuel pebbles (matrix, shell): 23.5%
    - Reflector and carbon brick: up to 30%
- HTR-10 initial criticality:
  - 9,627 fuel pebbles
  - 7,263 dummy pebbles
  - 61% packing fraction
  - Room temperature
  - Fresh fuel



HTR-10 fuel pebble

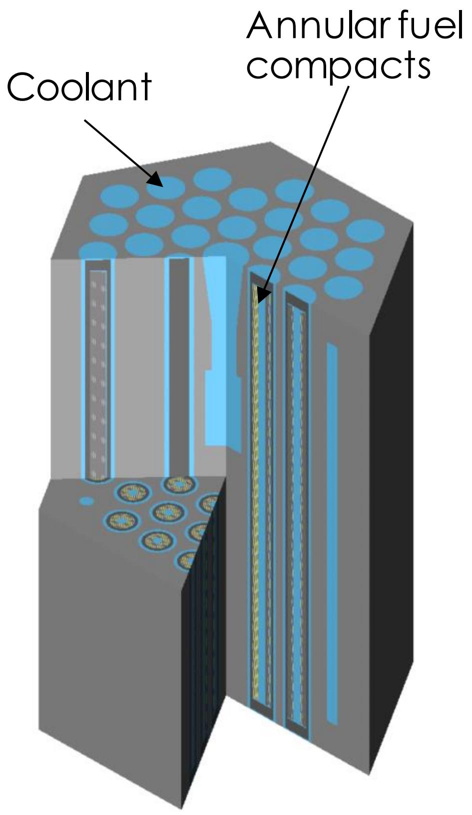


SCALE model of the HTR-10

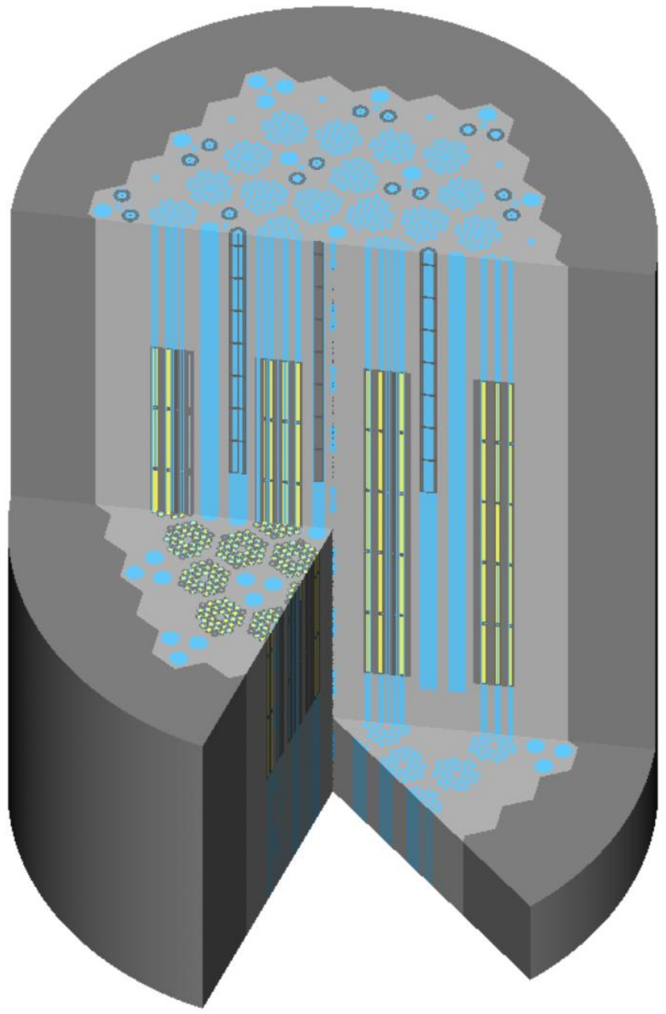
International Handbook of Reactor Physics Experiments, "Evaluation of the Initial Critical Configuration of the HTR-10 Pebble-Bed Reactor," HTR10-GCR-RESR-001, OECD/NEA, 2007.

Model 2: HTTR

- 30 MWth Prismatic High Temperature Gas-cooled Reactor
- Relevant characteristics:
  - UO_2 fuel density: 10.39 g/cm^3
  - ^{235}U enrichment: 3.4–9.9 wt.%
  - TRISO packing fraction: 30%
  - Number of particles per fuel compact: 12,987
  - Fuel compact inner radius/outer radius/length: 1 cm/2.3 cm/3.9 cm
  - Graphite densities indicate porosities between **22–25%**
    - Graphite overcoat and cladding: 24.8%
    - Graphite reflector around blocks: 24.0%
    - Graphite in blocks: 22.2%
- HTTR criticality experiment:
  - Configuration with fully loaded core (30 fuel blocks)
  - Room temperature
  - Fresh fuel



HTTR fuel block



SCALE model of the HTTR

International Handbook of Reactor Physics Experiments, "Evaluation of the Start-up Core Physics Tests at Japan's High Temperature Engineering Test Reactor (Fully-Loaded Core)," HTTR-GCR-RESR-001, OECD/NEA, 2010.

SCALE: Impact of graphite TSL evaluation on the HTR-10

TSL library	293 K, just structure		293 K, just pebbles		293 K, both	
Cross section data	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]
Crystalline+ Sd					1.00637 ± 0.00019	+637
Crystalline					1.00650 ± 0.00019	+650
10% porosity	1.00554 ± 0.00019	+554	1.01031 ± 0.00019	+1031	1.00960 ± 0.00019	+960
20% porosity	1.00504 ± 0.00019	+504	1.01195 ± 0.00019	+1195	1.01115 ± 0.00019	+1115
30% porosity	1.00469 ± 0.00019	+469	1.01402 ± 0.00019	+1402	1.01389 ± 0.00019	+1389
Carbon (free gas)	1.00333 ± 0.00019	+333	1.02390 ± 0.00019	+2390	1.02091 ± 0.00019	+2091
HTR-10 exp.					1.00000 ± 0.00370	

1. **In graphite structure** -> as **inelastic xs goes up** -> absorption and leakage in surrounding structure go up -> **k_{eff} goes down**
2. **In pebbles** -> as **inelastic xs goes up** -> neutrons thermalize and cause fission in fuel -> **k_{eff} goes up**
3. When used for all materials, two effects compete, but pebble effect dominates

SCALE: Impact of graphite TSL evaluation on the HTTR

TSL library	293 K, just structure		293 K, just pebbles		293 K, both	
Cross section data	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]
Crystalline+ Sd					1.01090 ± 0.00019	**+840**
Crystalline					1.01113 ± 0.00019	**+863**
10% porosity	1.01310 ± 0.00019	**+1060**	1.01223 ± 0.00019	**+973**	1.01433 ± 0.00019	**+1183**
20% porosity	1.01423 ± 0.00019	**+1173**	1.01332 ± 0.00019	**+1082**	1.01572 ± 0.00019	**+1322**
30% porosity	1.01489 ± 0.00019	**+1239**	1.01437 ± 0.00019	**+1187**	1.01709 ± 0.00019	**+1469**
Carbon (free gas)	1.02160 ± 0.00019	**+1910**	1.02035 ± 0.00019	**+1785**	1.02663 ± 0.00019	**+2413**
HTTR exp.					**1.0025 ± 0.00710**	

- **As the inelastic goes up, k_{eff} goes up for all cases.**

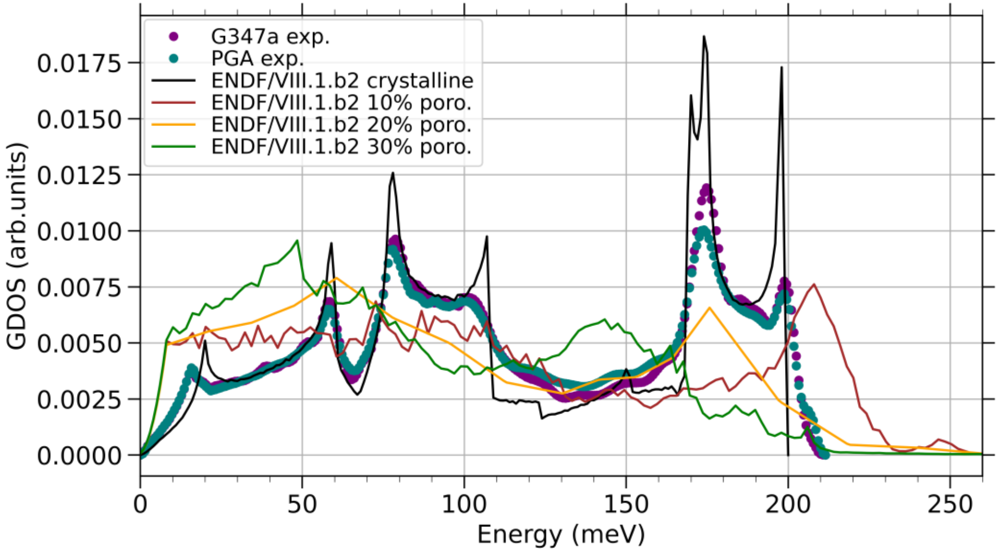
MCNP: Impact of graphite TSL evaluation on the HTR-10

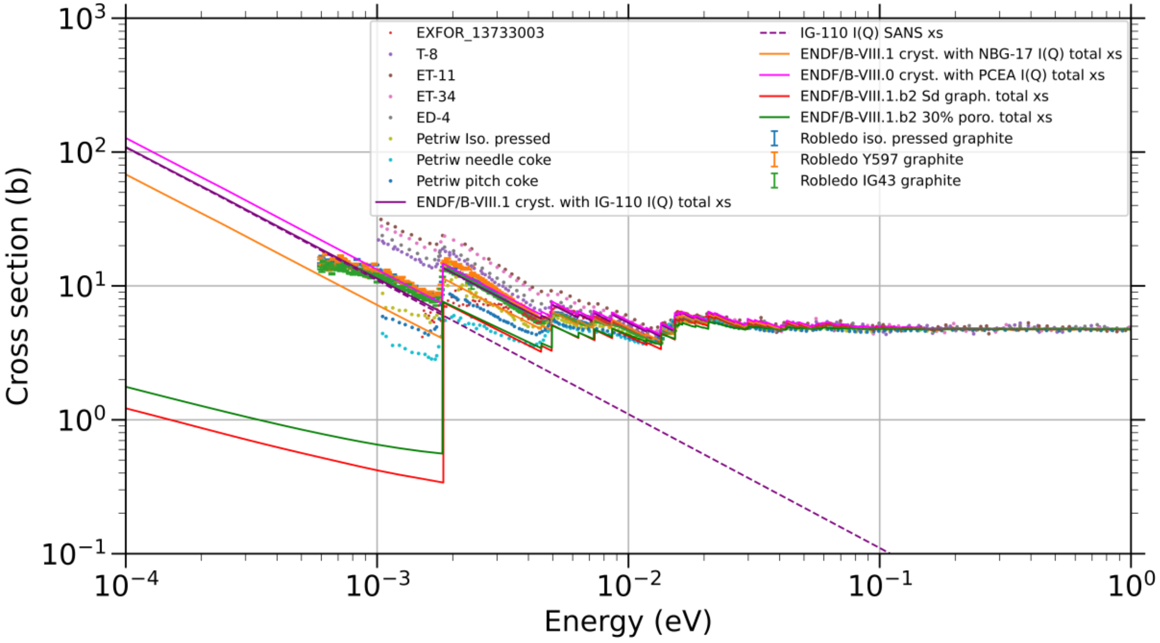
TSL library	293 K, just structure		293 K, just pebbles		293 K, both	
Cross section data	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]
Crystalline+ Sd					1.00722 ± 0.00008	+722
Crystalline					1.00678 ± 0.00008	+678
10% porosity	1.00663 ± 0.00008	+663	1.01045 ± 0.00008	+1045	1.01018 ± 0.00008	+1018
20% porosity	1.00639 ± 0.00008	+639	1.01261 ± 0.00008	+1261	1.01181 ± 0.00008	+1181
30% porosity	1.00579 ± 0.00008	+579	1.01480 ± 0.00008	+1480	1.01321 ± 0.00008	+1321
Sd + PCEA SANS	1.00765 ± 0.00008	+765	1.00653 ± 0.00008	+653	1.00708 ± 0.00008	+708
Sd + IG-110 SANS	1.00780 ± 0.00008	+780	1.00621 ± 0.00008	+621	1.00683 ± 0.00008	+683
Sd + NBG-17 SANS	1.00730 ± 0.00008	+730	1.00695 ± 0.00008	+695	1.00706 ± 0.00008	+706
HTR-10 exp.					1.00000 ± 0.00370	

1. **In graphite structure** -> **SANS reflects neutrons back into the core** -> fission goes up -> **k_{eff} goes up**
2. **In pebbles** -> **SANS reflects neutrons away from the fuel** -> less fission caused-> **k_{eff} goes down**
3. When used for all materials, two effects compete, but pebble effect wins out

Conclusions

- Compared three different benchmarks with (unknown graphite) using MCNP6.2 and SCALE ENDF/VIII.1 graphite TSLs.
- SCALE and MCNP results are consistent and comparable, within the expectations of model differences.
- HTR-10, and HTTR benchmarks show that the increase in the inelastic cross section of the porous graphite libraries lead to a significant increase in the k_{eff}.
- Addition of SANS cross sections in MCNP6.2 results in a slight improvement of k_{eff} compared to 'crystalline + Sd' TSL.
- Porosity in graphite manifests itself through SANS and not through increase in the inelastic cross section as represented in porous ENDF TSLs.
- **Graphite is a perfect example of why both INS and transmission measurements are need! Without INS measurements we would be misled by the atomistic modeling, and without transmission measurements we would not see the effects of SANS.**





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Acknowledgements

- The work presented here is sponsored by the NRC and DOE's NCSP.
- The model development and the presented calculations benefited from the discussion with many colleagues in the Nuclear Energy and Fuel Cycle Division at ORNL.

Model 3: PROTEUS

- Zero-power High Temperature Gas-cooled Reactor
- Relevant characteristics:
  - UO_2 fuel density: 10.88 g/cm^3
  - ^{235}U enrichment: 16.7 wt.%
  - Fuel compact inner radius/outer radius: 2.35 cm/3 cm
  - Graphite densities indicate porosities between **22–25%**
    - Majority of the system: 22.1%
    - Moderator pebbles: 25.66%
    - TRISO: 51.3% (buffer)-15.9% (IPyC and OPyC)

- Core configurations:

❑ PROTEUS-GCR-EXP-001
- ❖ Cores 1, 1A, 2, and 3
- ❖ **Hexagonal Close Packing**
- ❖ 1:2 Moderator-to-fuel Pebble Ratio

❑ PROTEUS-GCR-EXP-002
- ❖ Cores 4
- ❖ **Random Packing**
- ❖ 1:1 Moderator-to-fuel Pebble Ratio

❑ PROTEUS-GCR-EXP-003
- ❖ Cores 5,6,7, and 8
- ❖ **Columnar Hexagonal Point-on-Point Packing**
- ❖ 1:2 Moderator-to-fuel Pebble Ratio

❑ PROTEUS-GCR-EXP-004
- ❖ Cores 9 and 10
- ❖ **Columnar Hexagonal Point-on-Point Packing**
- ❖ 1:1 Moderator-to-fuel Pebble Ratio

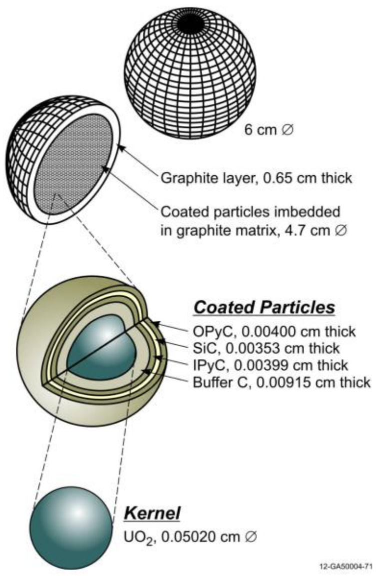


Figure 3.1-8. Fuel Pebble and TRISO Particle.

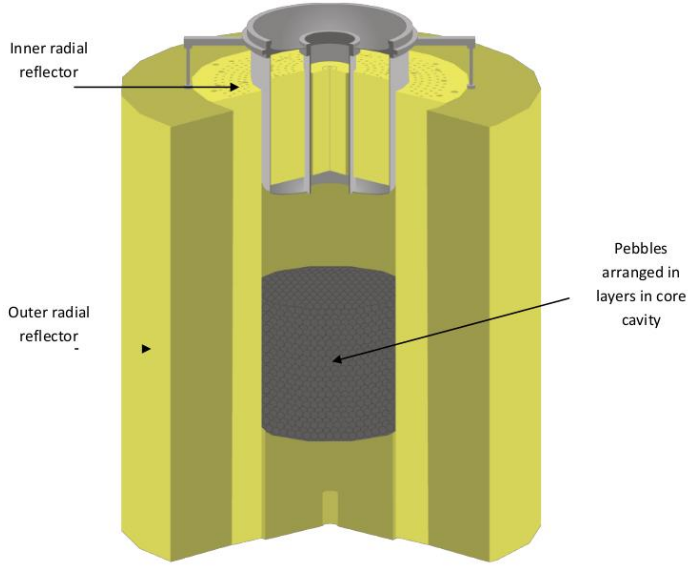


Figure 1.0-1. Generic HTR-PROTEUS configuration (derived from Ref. 2).

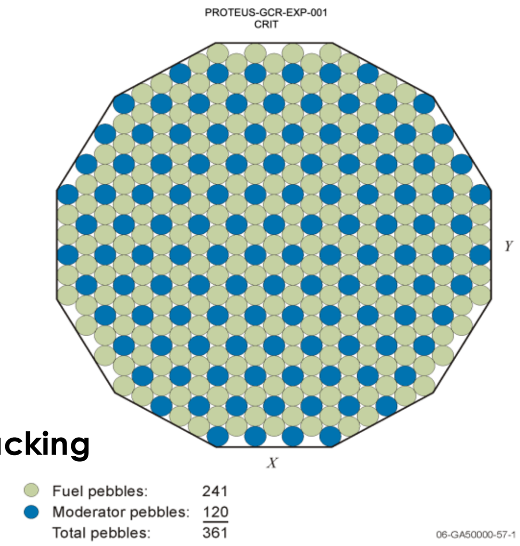


Figure 1.1-22. Odd fueled layers (1, 3, 5, 7, etc.) of Cores 1, 1A, and 2 (Ref. 1).

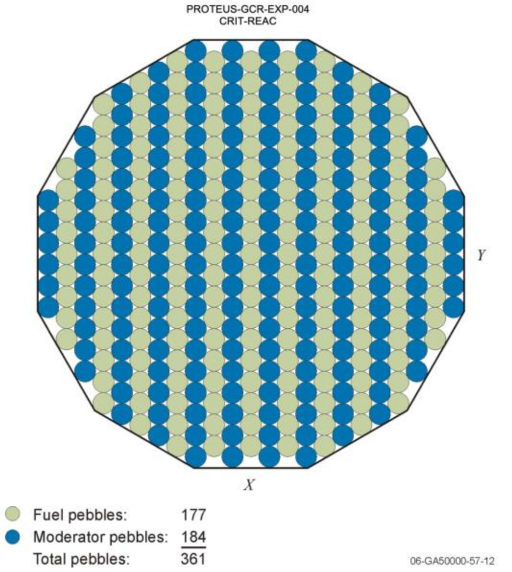
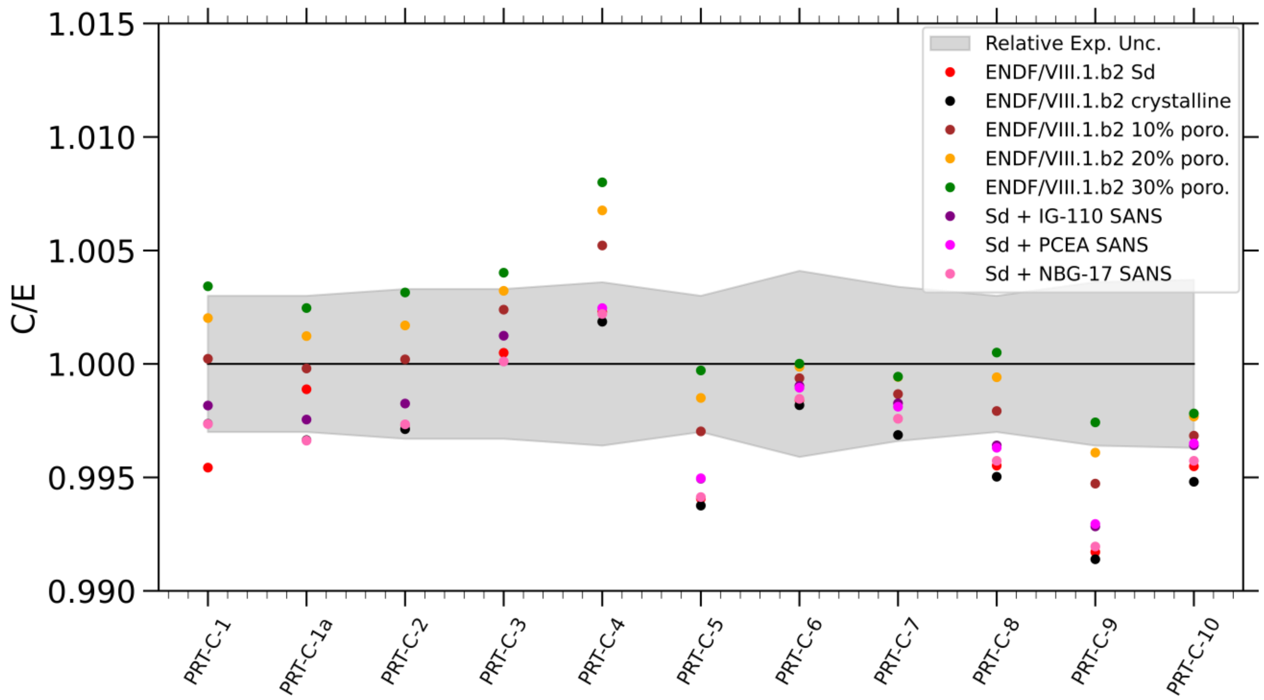


Figure 1.1-19. Layers 1, 7, 13, 19, and 25 of Core 9 (Ref. 1).

International Handbook of Reactor Physics Experiments, "HTR-PROTEUS PEBBLE BED EXPERIMENTAL PROGRAM CORES 1, 1A, 2, and 3: HEXAGONAL CLOSE PACKING WITH A 1:2 MODERATOR-TO-FUEL PEBBLE RATIO," *HTTR-GCR-RESR-001, OECD/NEA, 2006.*

MCNP: Impact of graphite TSL evaluation on the PROTEUS



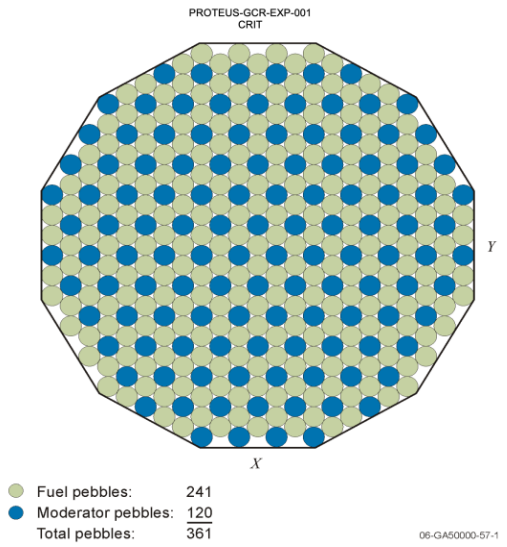


Figure 1.1-22. Odd fueled layers (1, 3, 5, 7, etc.) of Cores 1, 1A, and 2 (Ref. 1).

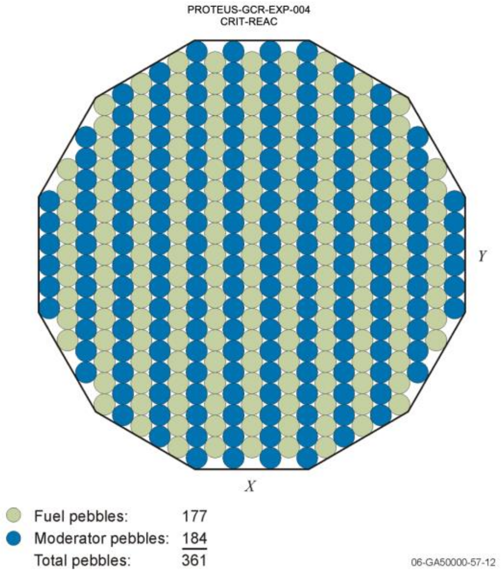


Figure 1.1-19. Layers 1, 7, 13, 19, and 25 of Core 9 (Ref. 1).

TSL	Chi^2
Crystalline+ Sd	1.57
Crystalline	1.72
10% porosity	0.654
20% porosity	0.664
30% porosity	0.936
Sd + PCEA SANS	1.15
Sd + IG-110 SANS	1.04
Sd + NBG-17 SANS	1.45

- Due to **increase in the inelastic xs** for porous TSLs, combined with the HCP pebble arrangements for Cores 1-3, which amplifies the effect due to decreased probability of leakage, porous TSLs seem like they provide a better k_{eff} values

Literature review of reactor benchmarks

Reactor Physics Experiment on a Graphite-Moderated Core to Construct Integral Experiment Database for HTGR

Shoichiro Okita, Yuji Fukaya, Atsushi Sakon, Tadafumi Sano, Yoshiyuki Takahashi & Hironobu Unesaki

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Figure 2 of 5

Fig. 2. Configuration of the core B7/4"G2/8"p8EU(3)+3/8"p38EU.

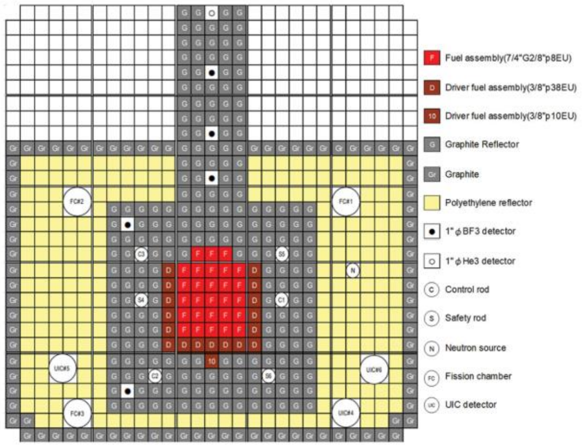


TABLE III C/E-1 Values for the k_{eff} Values

Nuclear Data Library	Critical State 1	Critical State 2	Critical State 3
JENDL-4.0	−0.09%	−0.07%	−0.08%
JEFF-3.2	+0.15%	+0.17%	+0.14%
ENDF/B-VII.1	+0.16%	+0.17%	+0.15%
ENDF/B-VIII.0 (30% porous graphite)	+0.17%	+0.18%	+0.17%
ENDF/B-VIII.0 (10% porous graphite)	+0.27%	+0.25%	+0.22%
ENDF/B-VIII.0 (ideal crystalline graphite)	+0.27%	+0.28%	+0.25%
JENDL-5 (30% porous graphite)	+0.01%	−0.01%	−0.02%
JENDL-5 (10% porous graphite)	+0.07%	+0.08%	+0.07%
JENDL-5 (ideal crystalline graphite)	+0.10%	+0.11%	+0.10%

Simulation-based studies on graphite absorption properties for ASTRA critical experiments

M.V. Shchurovskaya [a,*], A.E. Kruglikov [a,b], Yu.N. Volkov [a], N.I. Geraskin [a], V.A. Nevinitsa [b], P.A. Fomichenko [b], V.E. Jitarev [b]

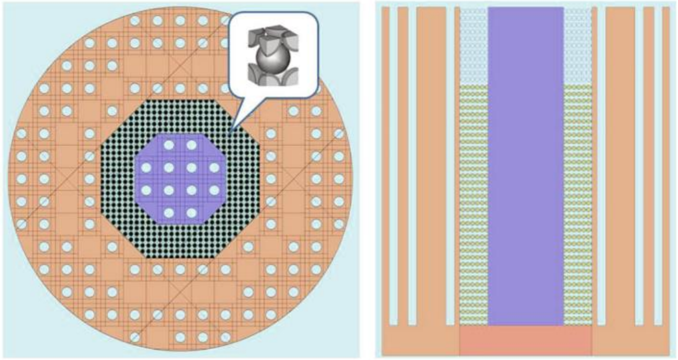


Fig. 4. Horizontal and vertical cross section of the Serpent model of the ASTRA facility.

Table 11
Serpent simulation results for ASTRA simplified core (model 2).

XS library	Reflector[(a)]		Fuel pebbles		k_{eff}	ρ, pcm
	EBC, ppm wt.	Porosity,%	EBC, ppm wt.	Porosity,%		
ENDF/B-VII.0	1	N/A	1	N/A	1.01370	1351
ENDF/B-VIII.0	0.4	0	1	0	1.01050	1039
	0.4	30	1	30	1.01677	1649
	0.4	30	1	10	1.01323	1306
	0.4	10	1	10	1.01419	1399
	0.4	30	1	0	1.01072	1061
	0.4	10	1	0	1.01145	1132
	0.54	0	1	0	1.00746	740
	1	0	1	0	0.99770	−231
	0	0	1	0	1.01945	1908

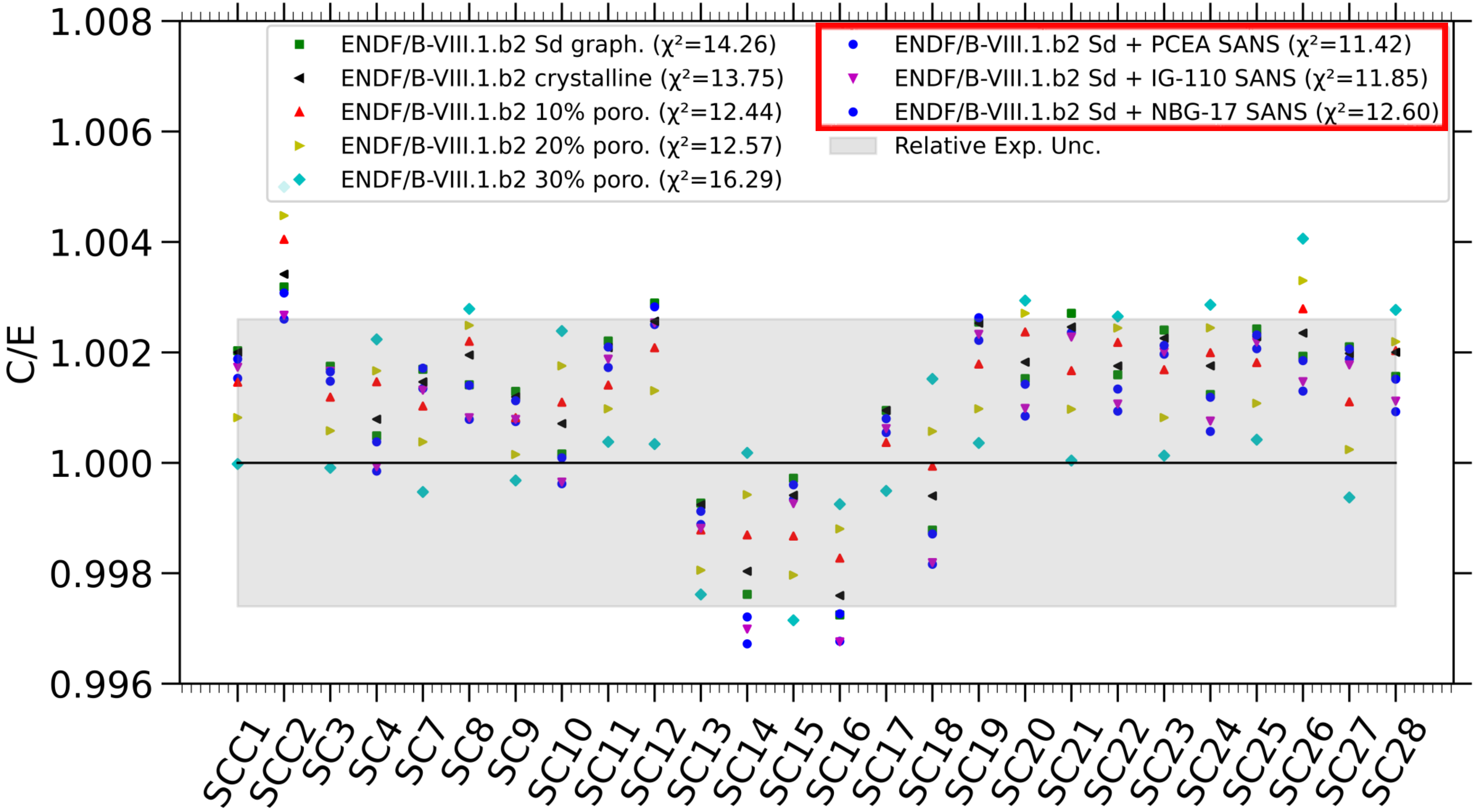
[(a)]Side, inner, and bottom reflector.

The EBC in graphite reflector for each cross-section library in case of the ASTRA facility should be estimated individually based on the value $\sigma_{eff}(0.0253\ eV) = 4.19$ mb reported in the benchmark specification. Therefore, when switching ENDF/B-VII.0 cross-section libraries to the libraries with increased carbon capture cross-section, the k_{eff} slightly decreases only due to the increase of absorption in matrix graphite of the fuel pebbles. As a result, the problem of overestimating the k_{eff} for the ASTRA benchmark configurations cannot be resolved only by using the nuclear data library with increased carbon capture cross-section. The introduction of graphite porosity to the calculation model will not give an improvement to the representation of the critical state. However, the reduction of the calculated reactivity bias between different ASTRA benchmark configurations is expected.

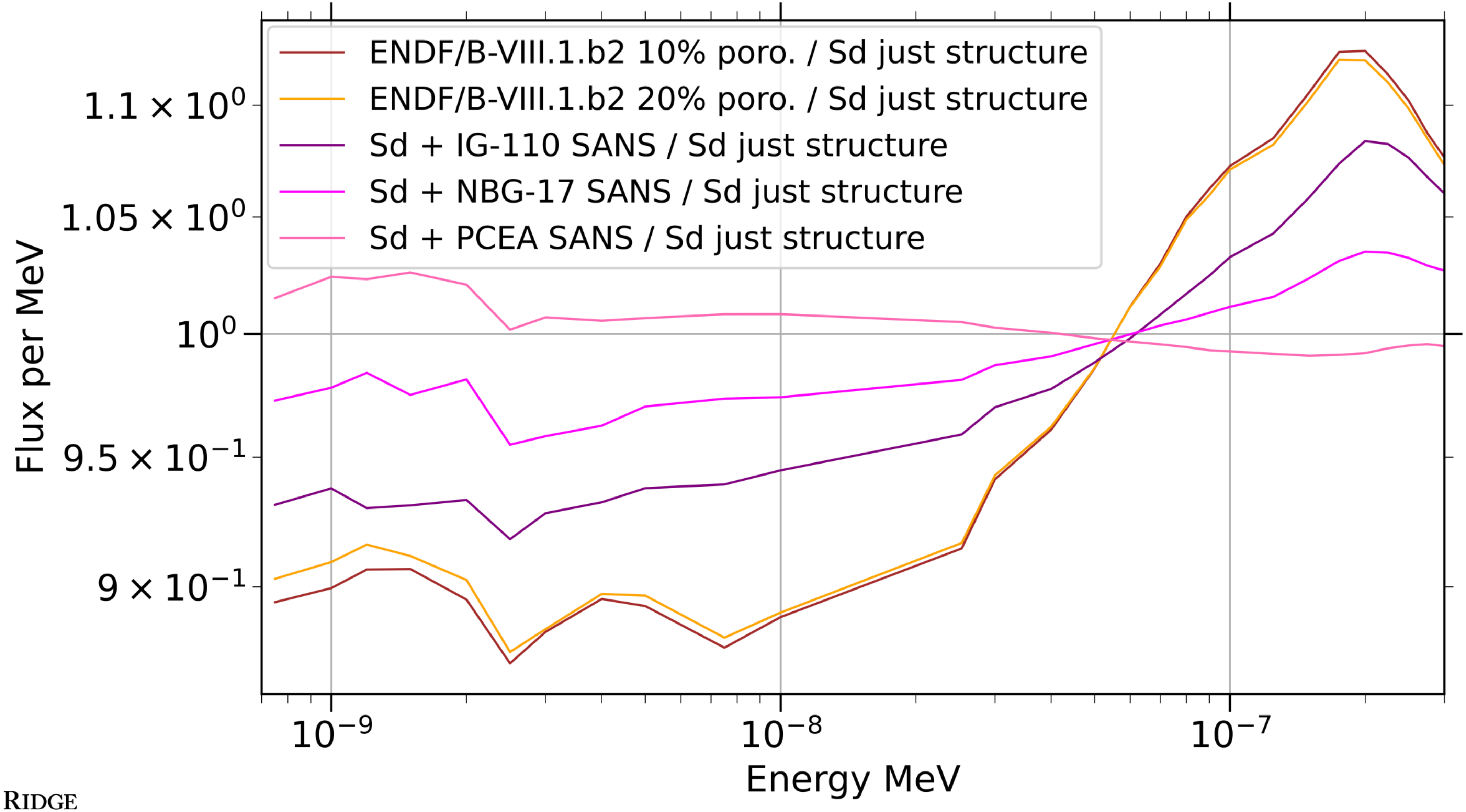
MCNP: Impact of graphite TSL evaluation on the HCT-016 (IGR reactor)

	Configuration 1		Configuration 2		Configuration 3		Configuration 4	
Cross section data	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]	k_{eff}	Δk [pcm]
Crystalline+ Sd	1.00538 ± 0.00008	**+538**	1.00104 ± 0.00008	**+104**	1.00219 ± 0.00008	**+219**	1.00587 ± 0.00008	**+587**
Crystalline	1.00463 ± 0.00008	**+463**	1.00017 ± 0.00008	**+17**	1.00183 ± 0.00008	**+183**	1.00521 ± 0.00008	**+521**
10% porosity	1.00924 ± 0.00008	**+924**	1.00551 ± 0.00008	**+551**	1.00647 ± 0.00008	**+647**	1.01043 ± 0.00008	**+1043**
20% porosity	1.01145 ± 0.00008	**+1145**	1.00800 ± 0.00008	**+800**	1.00906 ± 0.00008	**+906**	1.01283 ± 0.00008	**+1283**
30% porosity	1.01320 ± 0.00008	**+1320**	1.00986 ± 0.00008	**+986**	1.01095 ± 0.00008	**+1095**	1.01472 ± 0.00008	**+1472**
HCT-016 exp.	1.00000 ± 0.01100		1.00000 ± 0.01100		1.00000 ± 0.01100		1.00000 ± 0.01100	

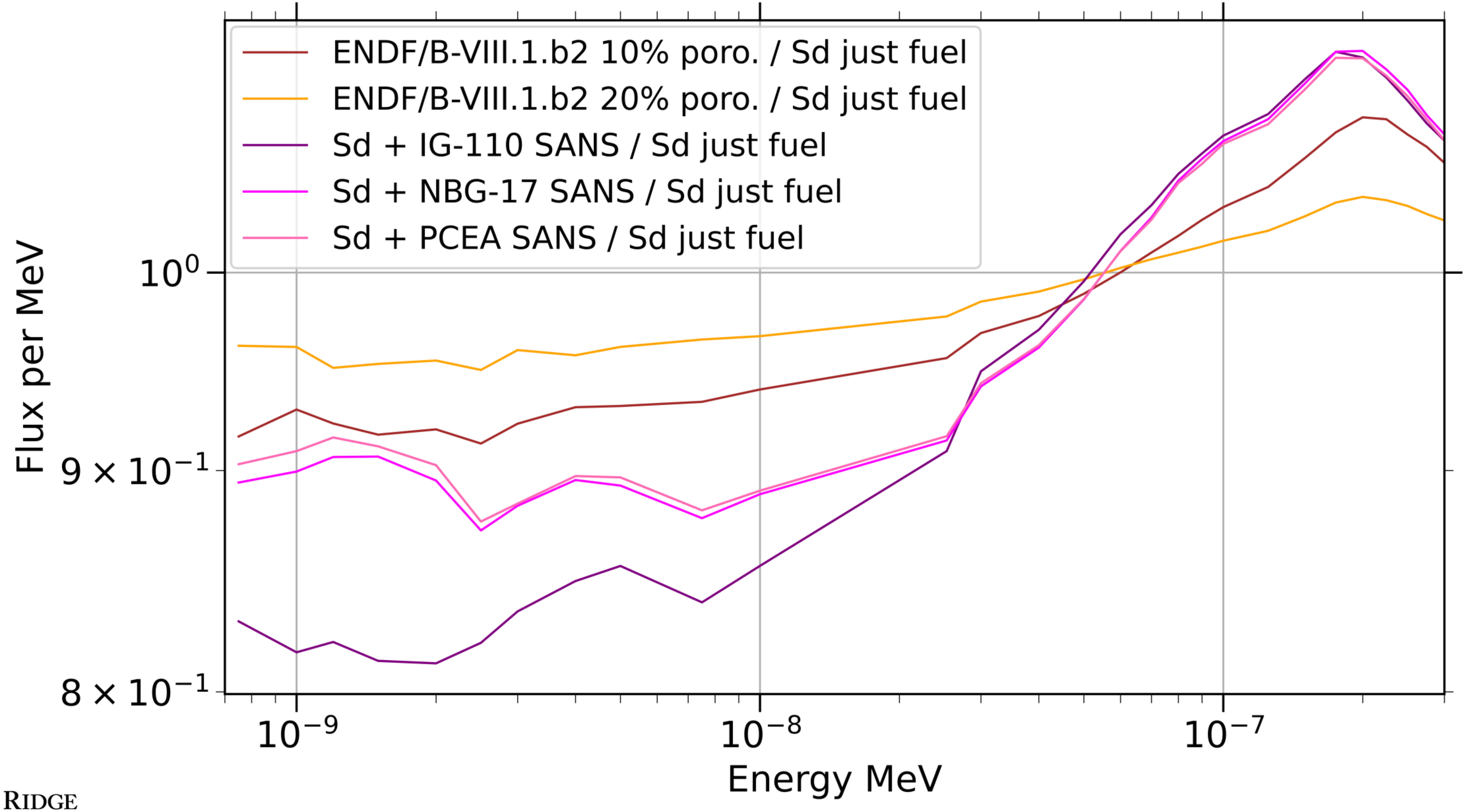
MCNP: Impact of graphite TSL evaluation on the LCT-060 benchmark



MCNP: Impact of graphite TSL evaluation on the HTR-10



MCNP: HTR-10 flux in TRISO right before UO2 kernel



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