INS and transmission measurements of different grades of nuclear graphite

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Thermal neutron scattering cross sections theory

- Inelastic scattering (coherent plus incoherent):

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:

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

where:

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

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

- Coherent elastic scattering:

$$\sigma^{coh} = \frac{\sigma_c}{E} \sum_{E_i > E} f_i e^{-2WE_i} \tag{3}$$

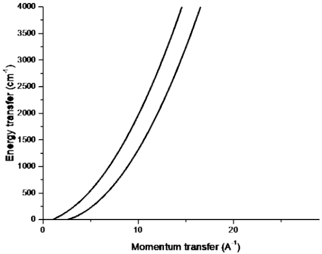
- Incoherent elastic scattering:

$$\sigma^{incoh} = \frac{\sigma_b}{2} \left(\frac{1 - e^{-4WE}}{2WE} \right) \tag{4}$$

- **As we can see all the scattering components are dependent on the inelastic physics through $\rho(\beta)$ and W (Debye-Waller factor)**

$S(\alpha, \beta)$ measurements

- INS indirect geometry spectrometers (VISION):

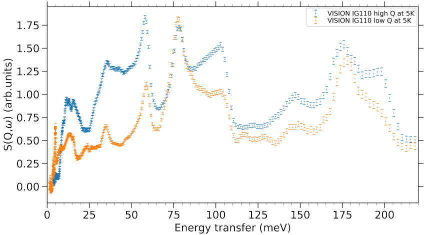


+ Measured quantity $S(Q, \omega)$ is directly related to $S(\alpha, \beta)$:

$$S(\alpha, \beta) = \frac{k_B T}{\hbar} \, exp\left(\frac{-\hbar\omega}{2k_B T}\right) S(Q, \omega) \qquad (5)$$

where T is the temperature, and k_B is the Boltzmann constant.

+ This means that we **can directly measure $S(\alpha, \beta)$**, which is what we store in ENDF TSL files.



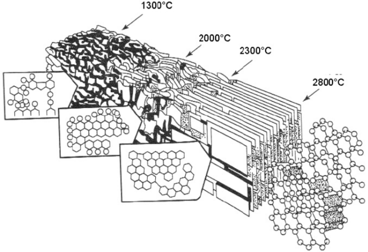
+ We can compare ENDF evaluations directly with these measurements by extracting $S(\alpha, \beta)$ along these trajectories (using **ENDFtk** now as well) and **applying well-known instrumental resolution**.
+ $S(Q, \omega)$ measurements are available to anyone through user proposal system at many facilities around the world (SNS at ORNL, NIST, ILL in France, ISIS UK, ESS Sweden, J-PARC in Japan).

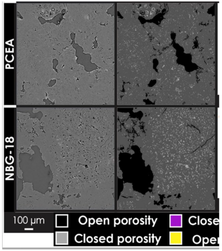
Thermalization of neutrons in nuclear graphite

- ENDF/B-VIII.1.b1 has 5 different graphite libraries: crystalline, Sd (crystalline), 10%, 20%, and 30% porosity reactor graphite

- 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.

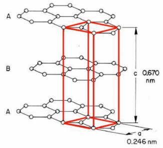
Evolution of graphitization process, reproduced from [1]:



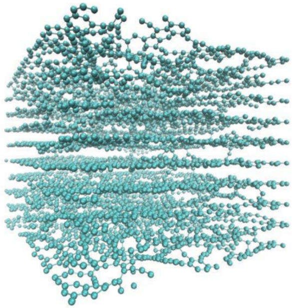


How does all this manifest itself in inelastic scattering measurements?

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



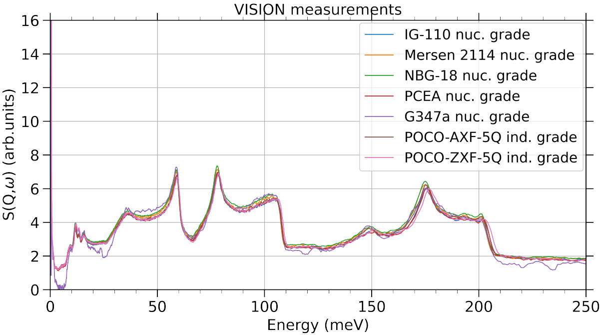
- Porous structure for 30% porosity graphite [2]:



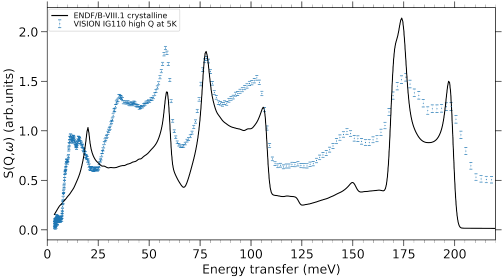
$S(Q, \omega)$ measurements at VISION instrument at SNS ORNL at 5K:

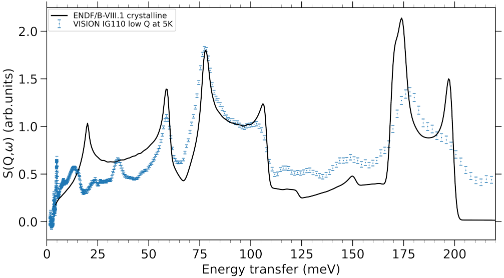
Graphite	Grain size [μm]	Density [g/cm^3]	Porosity [%]
PGA	800	1.70	25
G347a	50	1.85	17.8
IG-110	20	1.77	21.6
NBG-18	1600	1.85	17.8
PCEA	360	1.83	18
Mersen 2114	13	1.81	10
POCO-AXF-5Q	5	1.78	20
POCO-ZXF-5Q	1	1.78	20

Table 1: Properties of different types of graphite.

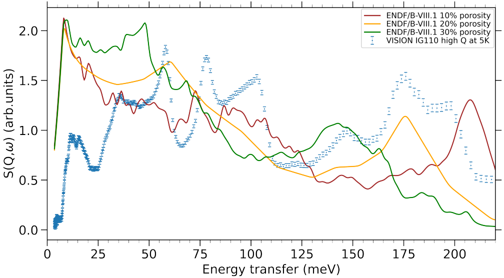


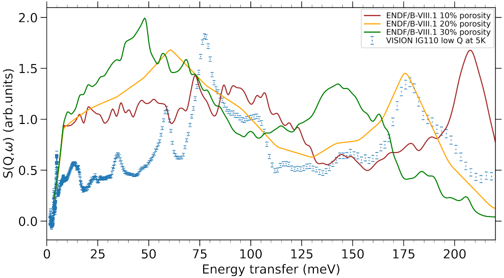
$S(Q,\omega)$ measurements at VISION instrument at SNS ORNL at 5 K:





$S(Q,\omega)$ measurements at VISION instrument at SNS ORNL at 5 K:





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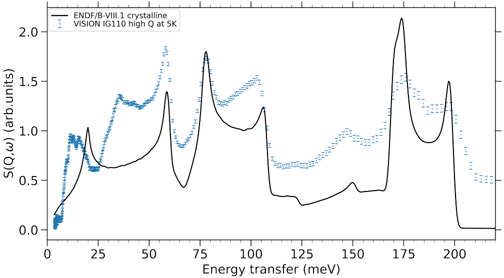
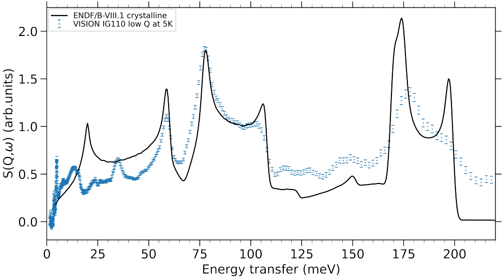
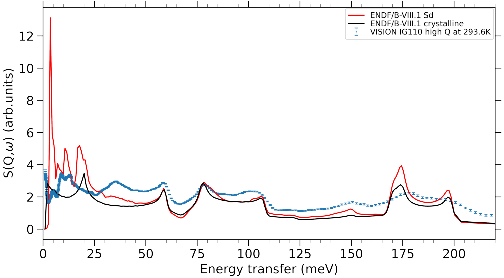
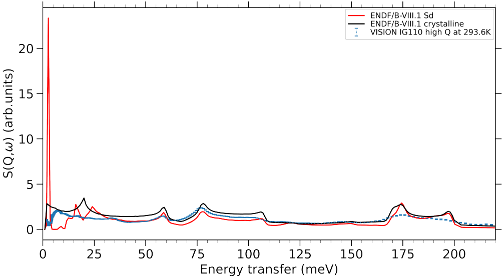
$S(Q, \omega)$ measurements at VISION instrument at SNS ORNL at room temperature:



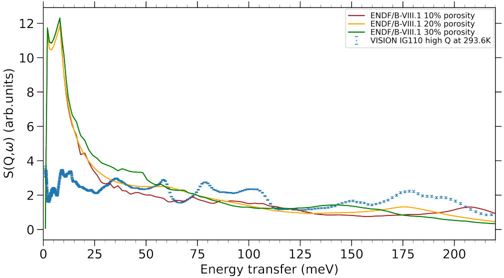


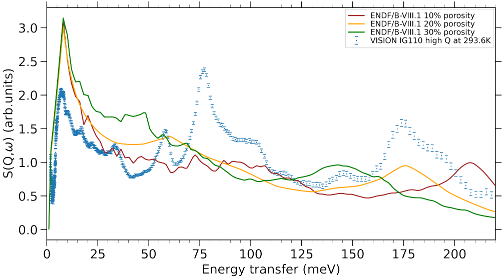
$S(Q,\omega)$ measurements at VISION instrument at SNS ORNL at room temperature:





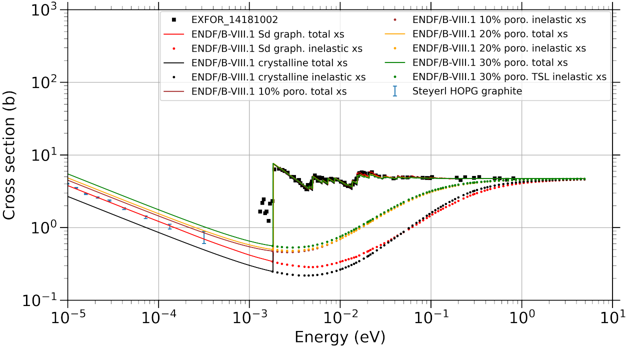
$S(Q,\omega)$ measurements at VISION instrument at SNS ORNL at room temperature:





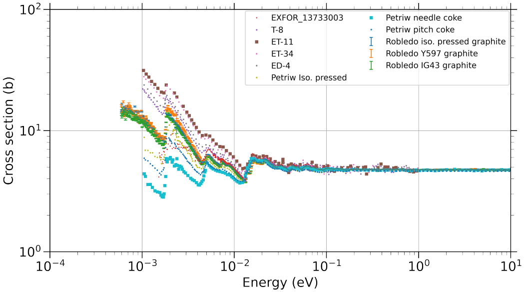
Graphite thermal transmission measurements

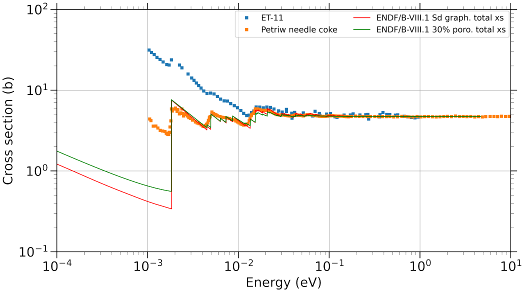
- **Sd-graphite** is the most physically accurate TSL from differential level
- The effect of porosity in 10%, 20%, and 30% TSLs was inaccurately modeled (by introducing defects on microscopic scale, when pores are a microscopic effect) which resulted in **increase of the inelastic scattering cross section**
- The actual effect of porosity is seen in **Small Angle Neutron Scattering (SANS)** cross section, and not in the inelastic cross section, and it is not quantifiable just by the percentage of the porosity, but it is a complex interplay of pore sizes and distributions



Graphite thermal transmission measurements

- **There are multiple of transmission measurements on different grades of nuclear graphite that show impact of SANS, and have been ignored in the validation of existing ENDF graphite TSLs.**





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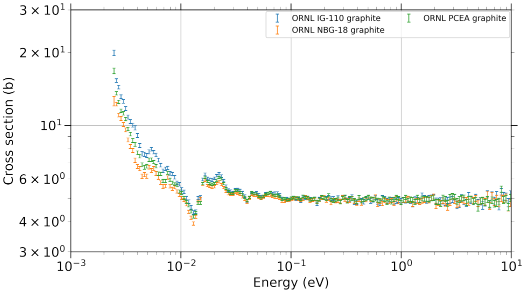
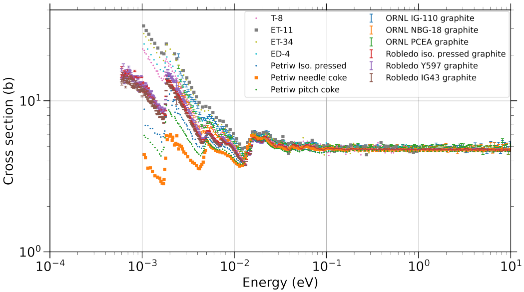
Graphite thermal transmission measurements

- **We have performed new transmission measurements of IG-110, NBG-18, and PCEA grades of nuc. graphite at VESUVIO instrument at ISIS in UK:**





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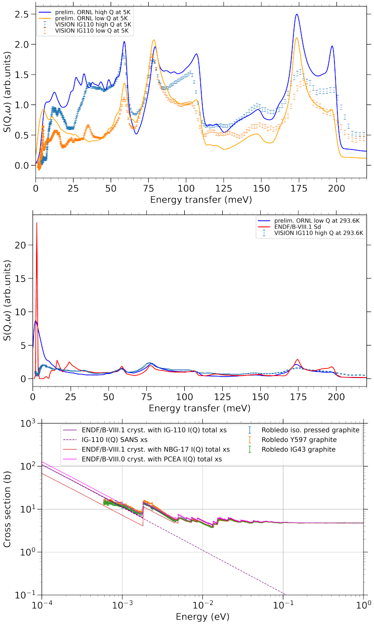
Summary & Conclusions

- New INS measurements of graphite at 5 K and room temperature.
- New transmission measurements of graphite at room temperature.
- All ENDF/B-VIII.1 graphite TSL fail to reproduce total cross section measurements.
- Porous graphite TSLs fail to reproduce both the differential and total cross section measurements.
- Porosity in graphite manifests itself through SANS (macroscopic structural effect) and not through increase in the inelastic cross section (microscopic structural effect) 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.**
- Current work at ORNL funded by DNCSH EAW1 to correctly model thermalization of neutrons in nuclear graphite.



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