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Evaluation of thermal neutron scattering law of nuclear-grade isotropic graphite

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Introduction

✓ We have started a 3-year project entitled "Development of Nuclear Data Evaluation Framework for Innovative Reactor" in 2021.

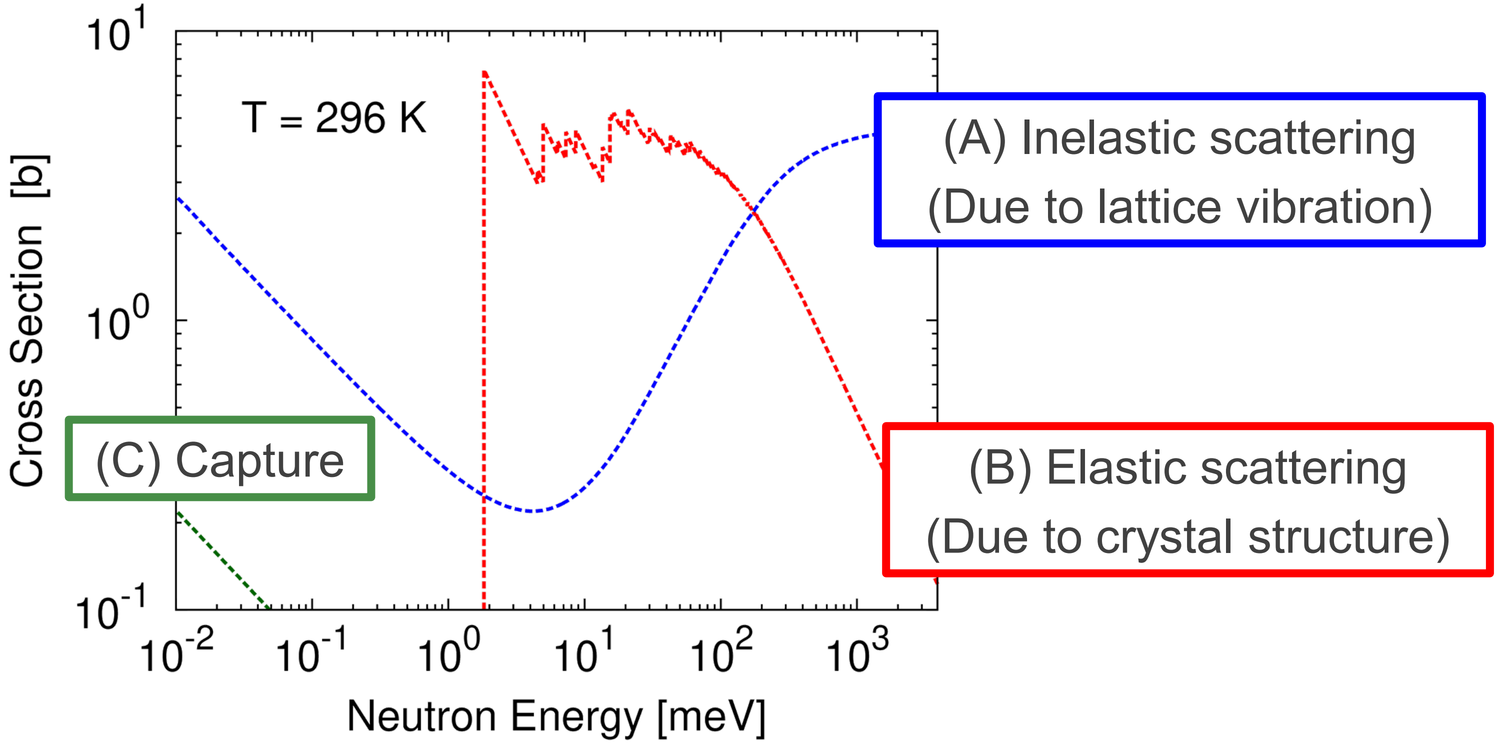
→ The objective is to establish a scheme to improve the accuracy of nuclear data required in the development of innovative nuclear reactors within a short time period through collaboration between experiments and evaluations.

✓ Scattering and transmission experiments on nuclear-grade isotropic graphite in the meV region were conducted by Kimura et al. (presented at 3 p.m. Monday).

✓ Thermal neutron scattering law (TSL) data of isotropic graphite is evaluated considering the above experimental results.

Cross section of JENDL-5 for graphite (crystalline)

* TSL data are taken from ENDF/B-VIII.0



✓ Inelastic and (coherent) elastic components are the targets of evaluation.

→ Small-angle neutron scattering (SANS) due to structures larger than the crystal structure (pores and inter-grain voids) is discussed later.

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Evaluation method of inelastic scattering

✓ Inelastic scattering component was evaluated by the theoretical calculation.

1. Compute the phonon density of states (DOS) with the first-principles lattice dynamics simulations.
2. Obtain TSL for inelastic scattering by processing the above DOS with the LEAPR module of NJOY-2016.

✓ Quantum ESPRESSO [1] was used for the first-principles calculation code.

→ Perform first-principles simulations with pseudopotentials and plane-wave basis.

[1] P. Giannozzi *et al.,* J. Chem. Phys. **152**, 154105 (2020).

Conditions of first-principles simulation

✓ Although the target of evaluation is graphite with pores, the simulations were performed for ideal crystalline graphite.

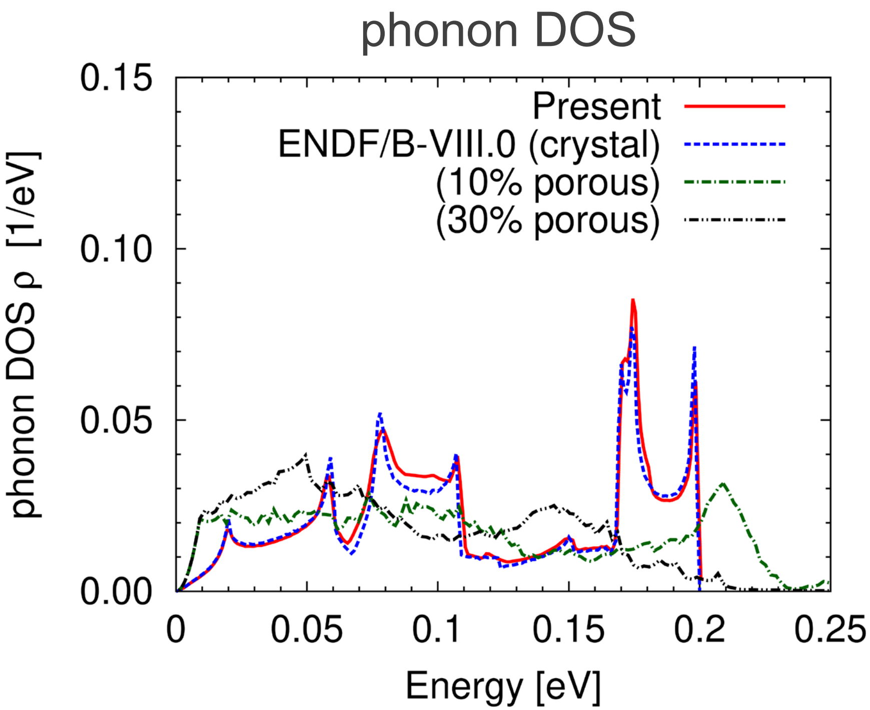
→ The vacancies in graphite are larger than the crystals and other non-vacant region (where inelastic scattering occurs) are highly crystalline [1].

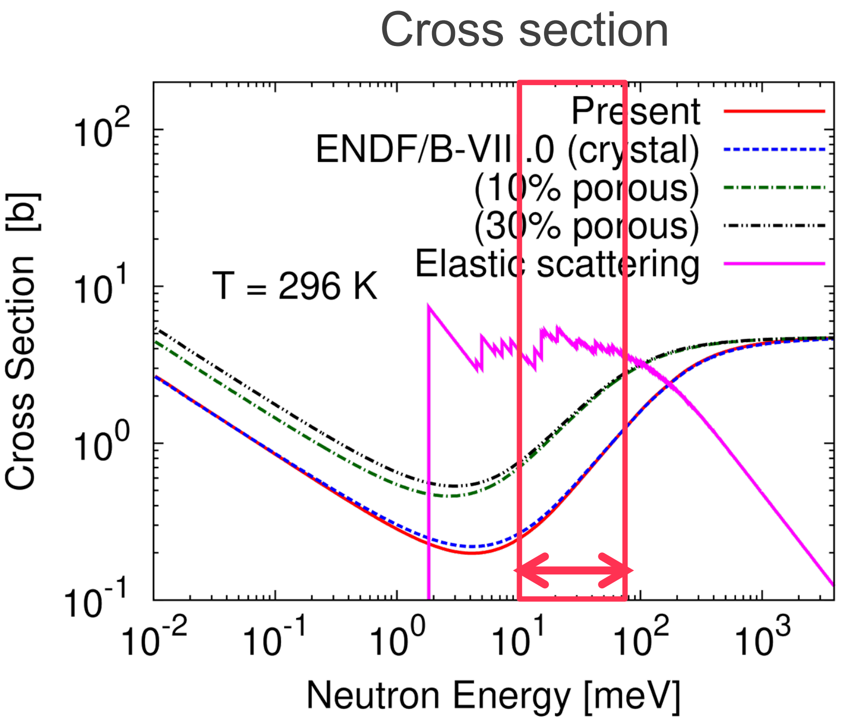
→ This contrasts with the modelling in ENDF/B-III.0 evaluation for nuclear-grade graphite (carbon atoms are randomly removed from crystalline graphite).

✓ Parameters having significant impact the results (e.g., pseudopotential, k-points mesh, plane wave cutoff energy) were determined with references the previous study on crystalline graphite [2].

[1] I. Al-Qair et al., Ann. Nucl. Ene. **161**, 108437 (2021). [2] I. Al-Qair, PhD thesis, NCSU (2007).

Results of simulation



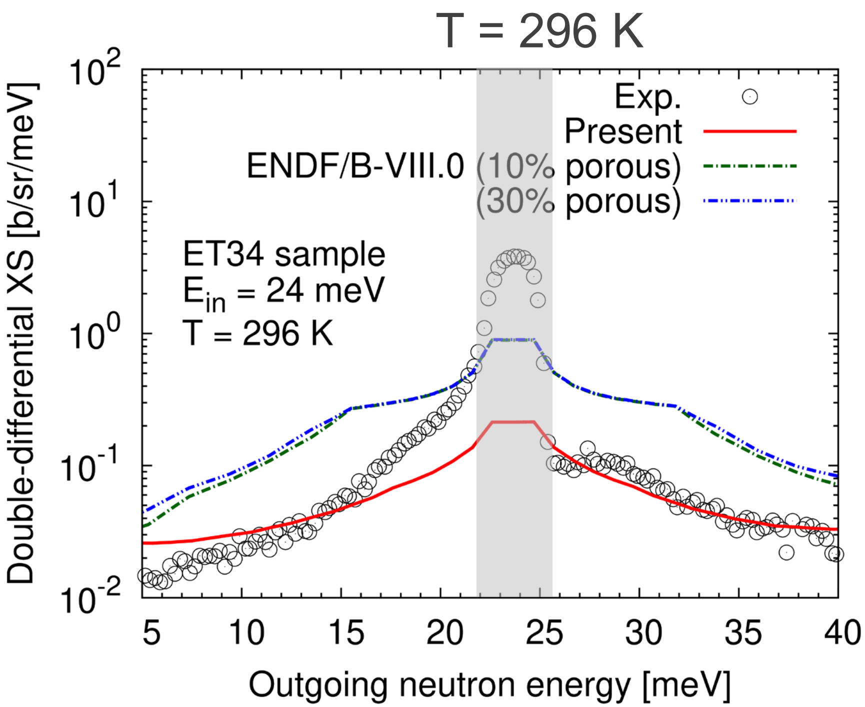


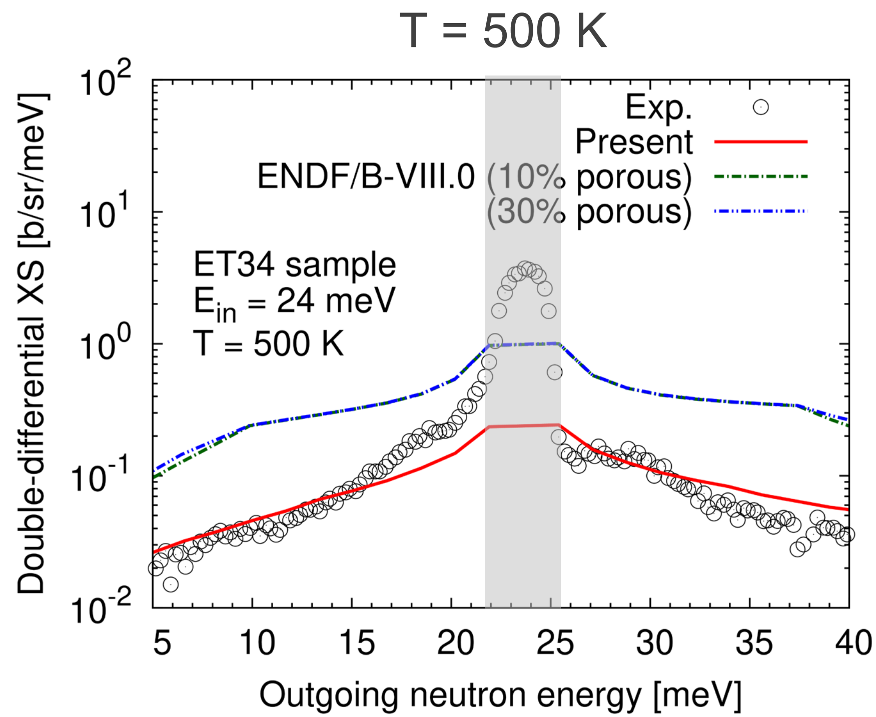
✓ The present results are close to the evaluated values for crystalline graphite of ENDF/B-VIII.0 (evaluated based on the first-principles simulations).

✓ The present results are largely different from the values for porous graphite.

→ It is difficult to judge which is better in the tens of meV region by comparison with the experimental total cross section.

Double-differential cross sections (E_{in} = 24 meV)





* The porosity of the sample in the experiment is approximately 11%.

✓ The present results for crystallin graphite reproduce the experimental data better than the evaluated values for porous graphite of ENDF/B-VIII.0.

→ The regions in graphite where inelastic scattering occurs are highly crystalline.

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Evaluation method of elastic scattering

✓ The elastic scattering component is evaluated by the positions and intensities of the Bragg peaks.

→ Cross section and angular distribution are uniquely determined from the peak position and intensity.

$$\sigma(E,T) = \frac{1}{E}\sum_{i=1}^{E_i<E} s_i(T), \quad \frac{d\sigma}{d\Omega}(E,\mu,T) = \frac{1}{2\pi}\frac{1}{E}\sum_{i=1}^{E_i<E} s_i(T)\,\delta(\mu-\mu_i)$$

$$\left(\mu_i = 1 - \frac{2E_i}{E}\right)$$

E incident energy

T temperature

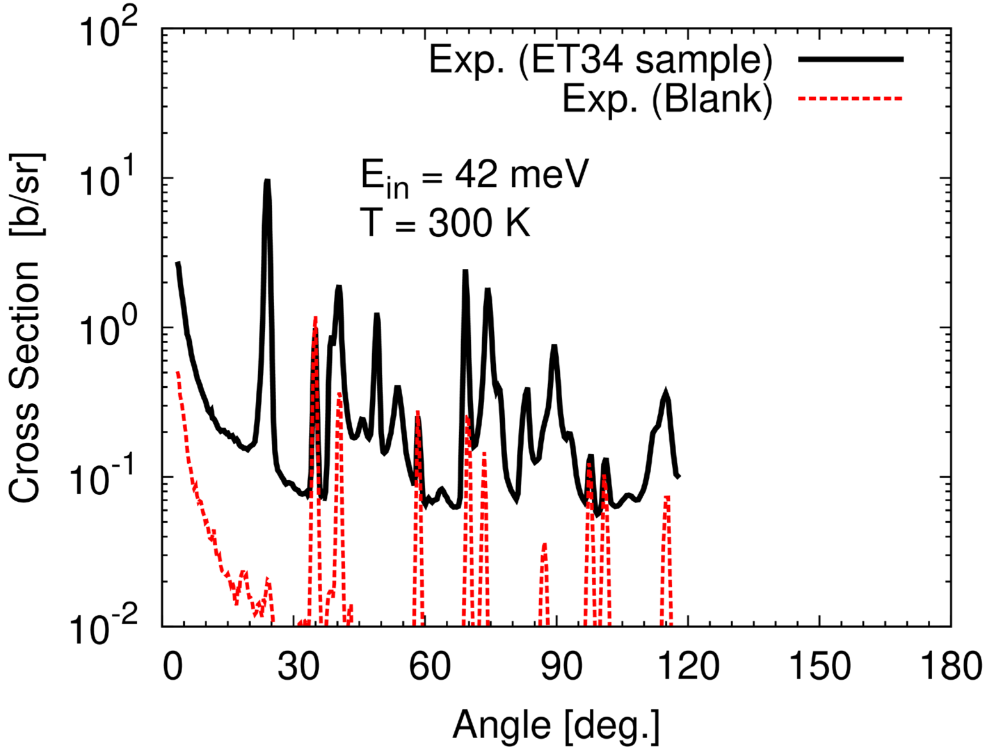
E_i *i*-th peak position (eV)

s_i *i-th* peak intensity (eV b)

μ cosine of scattering angle

Differential cross section (E_{in} = 42 meV) (1)

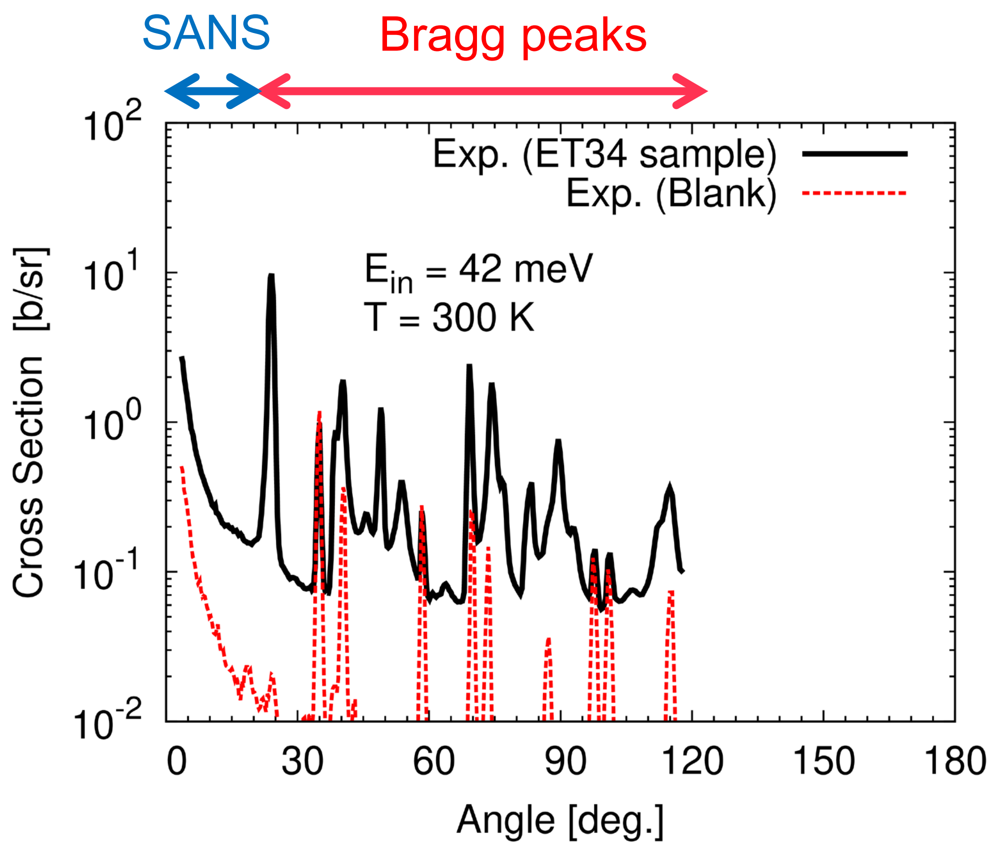
✓ The angular distributions of the neutron scattering experiments performed in this project were used to evaluate the Bragg peaks.



✓ Contributions from the aluminum container are seen in the blank measurement.

→ Not negligible in evaluating the Bragg peaks of graphite sample.

Differential cross section (E_{in} = 42 meV) (1)

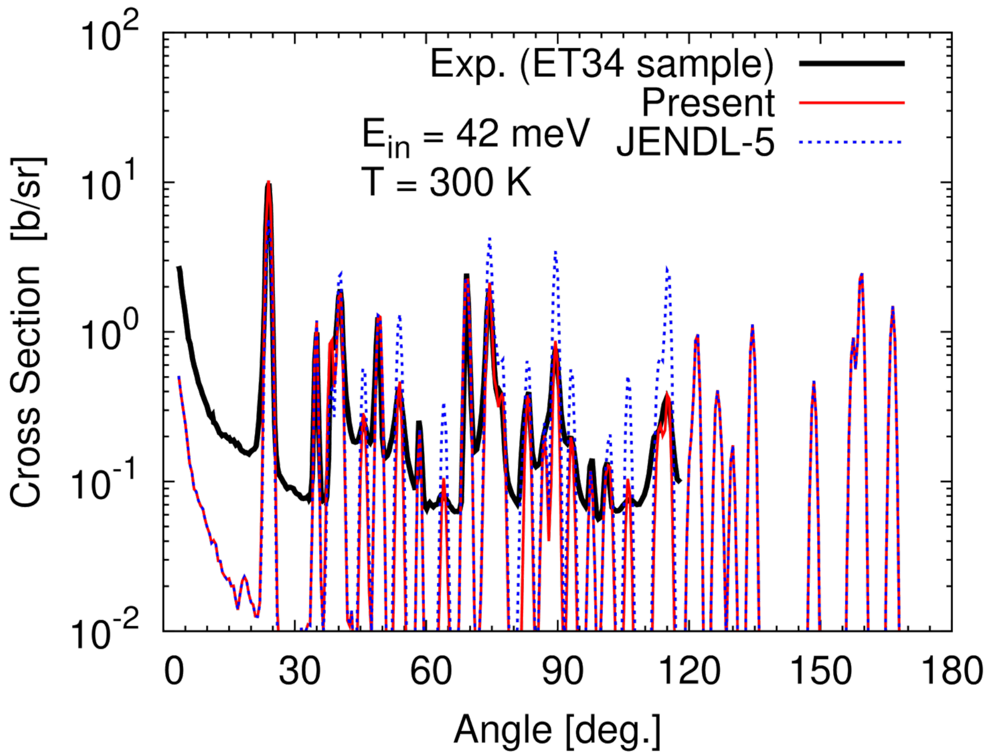


✓ Small angle neutron scattering (SANS) is seen in the most forward angles.

→ This does not affect the evaluation of the Bragg peaks.

Differential cross section (E_{in} = 42 meV) (2)

✓ We compare the evaluated values (+ contribution from the aluminum container) with the experimental values.

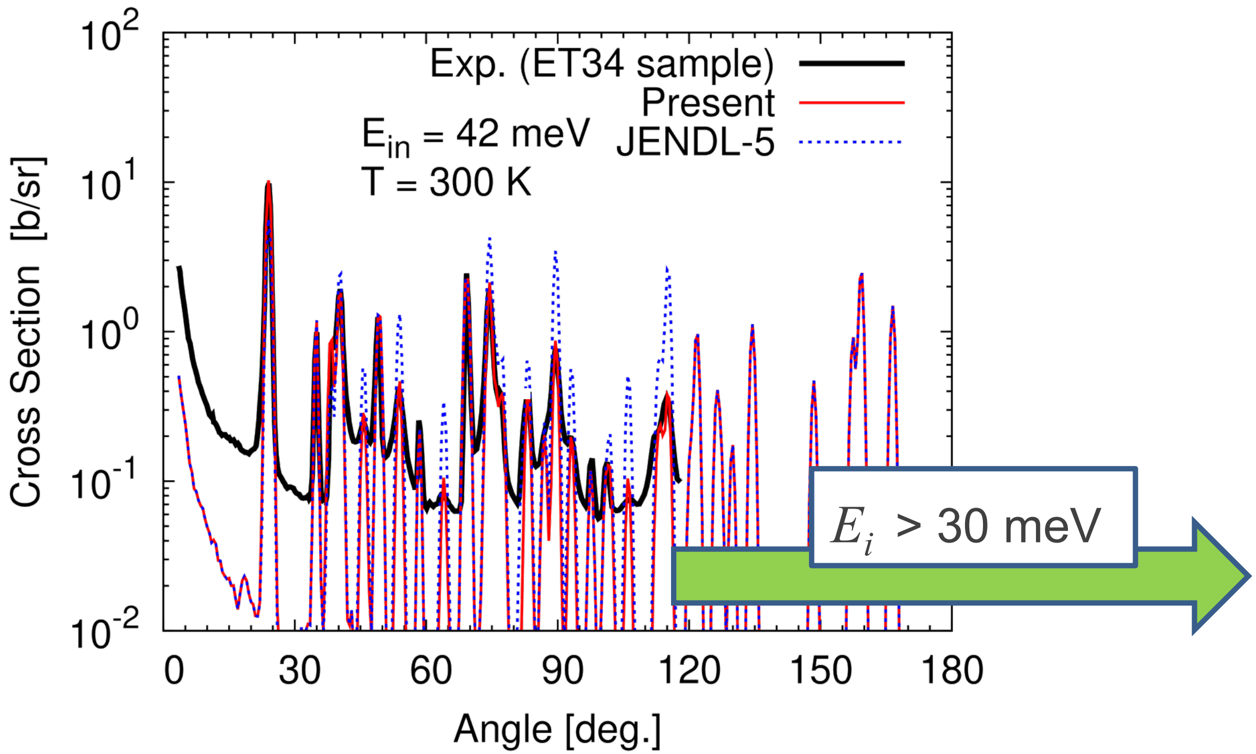


✓ JENDL-5 (=ENDF/B-VIII.0) data overestimate the present experimental data.

→ The intensities of the Bragg peaks were evaluated (positions were not changed).

Differential cross section (E_{in} = 42 meV) (2)

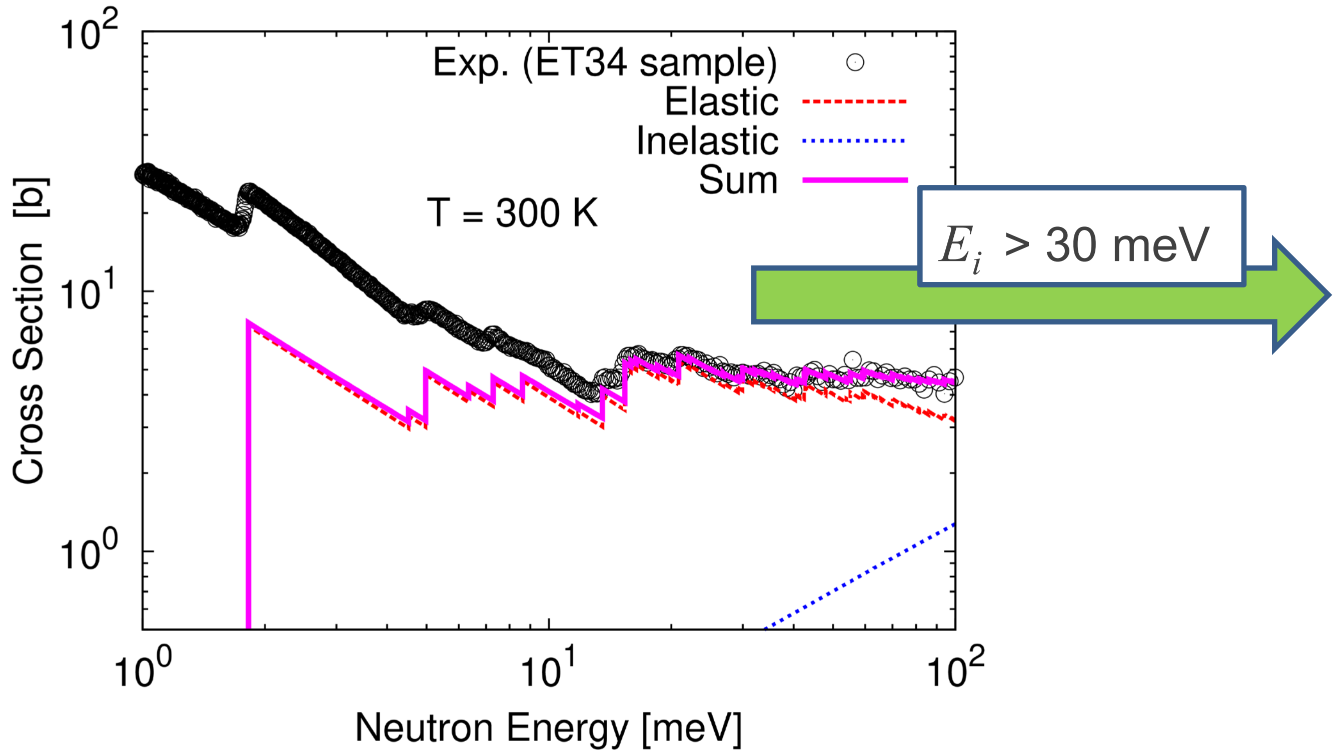
- ✓ In the present experiment, the data above 120° were not measured due to the limitation of equipment.



- ✓ Intensities at peak energies above 30 meV are unchanged at this stage.

Comparison with experimental total cross section

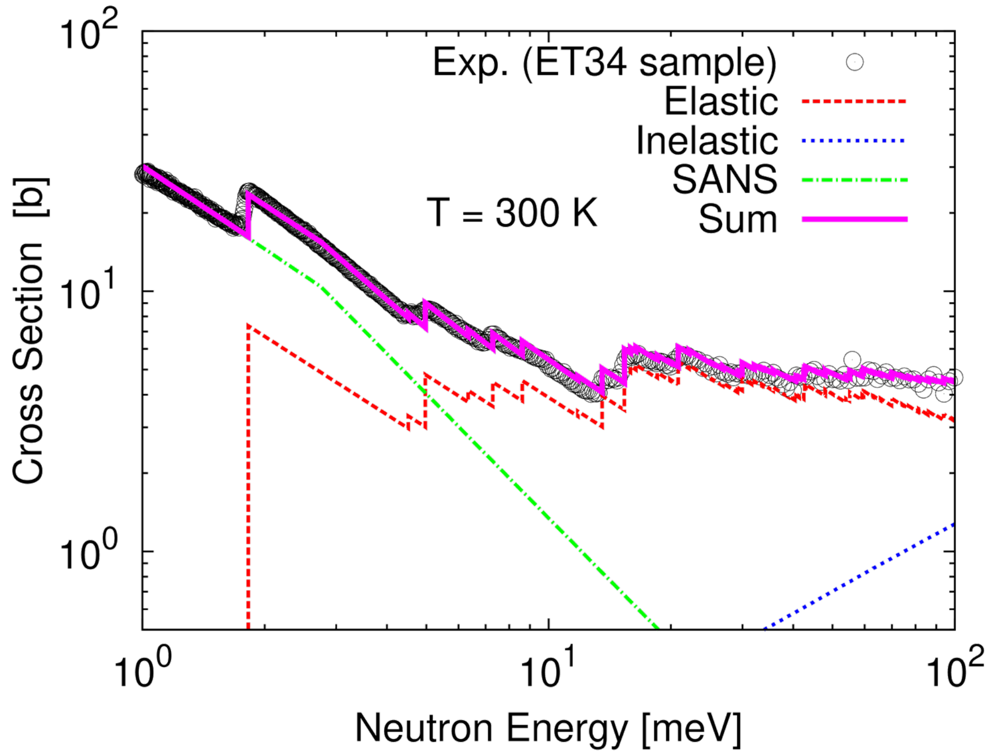
- ✓ Sum of evaluated inelastic and elastic scattering cross sections compared to the total cross sections measured in this project.



- ✓ The Bragg peak intensities above 30 meV were slightly adjusted.
- ✓ The evaluated values largely underestimate the experimental one below 10 meV.

Estimation of SANS component

✓ We estimate the small angle neutron scattering (SANS) components using the formulae proposed in the previous studies [1,2].



✓ By considering SANS, it is confirmed that the Bragg peak components below 30 meV are in good agreement also with the experimental total cross section.

[1] S. Petriw et al., J. Nucl. Mat. **396**, 181(2010). [2] K. Grammer et al., NIMA. **953**, 163226 (2020).

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Summary

- ✓ Thermal neutron scattering law (TSL) of isotropic graphite was evaluated considering the experimental results obtained in the project we have organized.

- ✓ Inelastic scattering was evaluated by the first-principles calculations for crystalline graphite, and the evaluated values reproduced the experimental data better than the ENDF/B-VIII.0 evaluation for reactor-grade (porous) graphite.

- ✓ Elastic scattering was evaluated based on the experimental data for angular distribution of outgoing neutron and total cross sections.

→ It was found that the quantification of small angle neutron scattering (SANS) is important in the comparison with total cross sections.

Outlook

✓ Compilation of the ENDF-6 formatted TSL file consisting of coherent elastic (Brag peaks) and inelastic scattering components.

→ SANS is not included since the official version of current nuclear data formats (ENDF-6, GNDS) have no place to store SANS.

✓ Derivation of inelastic scattering components from phonon density of states without incoherent approximation (assumed in NJOY).

→ We plan to use OCLIMAX [1] instead of NJOY.

→ Expected to improve data especially below a few meV.

✓ Evaluation of other samples used in the present experiment.

[1] Y.Q. Cheng et al., J. Chem. Theory Comput. **15**, 1974 (2019).