Generation of the Thermal Neutron Scattering Library for Crystalline Graphite

Haelee Hyun[a,b*], Do Heon Kim[b], Ser Gi Hong [a]
[a]*Department of Nuclear Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul, 04763, Republic of Korea*
[b]*Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Republic of Korea*
[*]*Corresponding author: hyunhl@kaeri.re.kr*

Keywords : thermal neutron scattering, graphite, moderator, nuclear data, NJOY, ab-initio

1. Introduction

Graphite has been considered as an efficient moderator in various types of nuclear reactors due to its properties such as the low neutron absorption and high neutron scattering. Besides, graphite has the high thermal stability and good heat conductivity, which also makes it beneficial to be used as a moderator. In this regard, the thermal neutron scattering library for graphite has been considered as an important component in reactor design and analyses.

Since the molecular dynamics (MD) and ab-initio simulations are introduced into the generation of thermal neutron scattering data, graphite data have been newly evaluated based on the methodologies. There are three types of the thermal neutron scattering data for graphite in the latest nuclear data libraries (e.g., ENDF/B-VIII.0 and JENDL-5), which are the crystalline graphite, reactor grade graphite with 10% porosity, and reactor grade graphite with 30% porosity. The data for crystalline graphite were generated based on the ab-initio simulation using VASP (Vienna Ab-initio Simulation Package) code[1], whereas the data for reactor grade graphite data were produced through the MD simulation using LAMMPS code[2].

In this study, the thermal neutron scattering libraries for crystalline graphite were generated based on VASP code simulation by employing both the local density approximation (LDA) and generalized gradient approximation (GGA) and compared with those of ENDF/B-VIII.0 and ENDF/B-VII.1.

2. Methods and Results

The phonon density of state (PDOS) of crystalline graphite was calculated using the VASP code. The PDOS is served as a crucial input parameter for the nuclear data processing code NJOY2016.

2.1 Thermal Scattering Law for Graphite

In practice, the thermal scattering cross section is divided into three different parts in the NJOY code: the coherent elastic, incoherent elastic, and incoherent inelastic (this category includes both incoherent and coherent parts). The scattering from carbon is almost completely coherent. Hence, the incoherent inelastic scattering and coherent elastic scattering cross sections should be handled to generate the thermal scattering cross section for graphite.

The inelastic double differential scattering cross section is given by Eq. (1).

$$\sigma(E, E', \mu) = \frac{\sigma_b}{4\pi kT}\sqrt{\frac{E'}{E}}e^{-\beta/2}S(\alpha, \beta), \qquad (1)$$

where E and E' are the incident and secondary neutron energies, σ_b is the characteristic bound cross section, k is the Boltzmann constant and T is the temperature of the material in Kelvin. $S(\alpha, \beta)$ is the thermal scattering law, where α and β are the momentum transfer $\alpha = (E' + E - 2\mu\sqrt{E'E})/AkT$ and energy transfer $\beta = (E' - E)/kT$, respectively. To calculate the thermal scattering law through NJOY code, the PDOS, $\rho(\beta)$ has to be provided as an input parameter, which gives information of the excitation state of the system[3].

In crystalline solid materials consisting of coherent scatterers, interference effects are caused due to the scattering between different planes of atoms of the crystalline materials. In this scattering process, the energy loss is not caused, which is called the coherent elastic scattering. The double differential coherent elastic scattering cross section is given by Eq. (2).

$$\sigma(E, E', \mu) = \frac{\sigma_c}{E}\sum_{E_i<E} f_i\, e^{-2WE_i}\delta(\mu - \mu_0)\delta(E - E'), \qquad (2)$$

where

$$\mu_0 = 1 - 2(E_i/E) \qquad (3)$$

In Eq. (2), E is the incident neutron energy, E' is the secondary neutron energy, μ is the scattering cosine in the laboratory reference system, σ_c is the characteristic coherent cross section for the material, W is the effective Debye-Waller coefficient, the E_i are the Bragg edges, and the f_i are related to the crystallographic structure factors. The Debye-Waller coefficient is given by Eq. (4).

$$W = \frac{\lambda}{AkT}, \qquad (4)$$

where A is the ratio of the scatterer mass to the neutron mass, λ is computed from the phonon density of state as shown by Eqs. (5) and (6).

$$\lambda = \int_{-\infty}^{\infty} P(\beta) e^{-\beta/2} d\beta \quad (5)$$

$$P(\beta) = \frac{\rho(\beta)}{2\beta \sinh(\beta/2)}, \quad (6)$$

where $\rho(\beta)$ is called as the PDOS. Consequentially, the PDOS is an indispensable parameter for calculating both inelastic and elastic scattering cross sections of graphite.

2.2 Phonon Density Of State (PDOS) Calculations

To calculate the PDOS in crystalline graphite, the ab-initio simulation code VASP has been used. VASP utilizes the Density Functional Theory (DFT) to calculate the electronic structure of a system. The DFT is a computational quantum mechanical modeling method and uses exchange-correlation functionals to account for the interactions between electrons. The exchange-correlation functionals are mathematical expressions that approximate the exchange and correlation energies of a many-electron system.

In this study, the PDOS was calculated using the LDA and GGA. The LDA is the simplest form of exchange-correlation functional, which considers only electron density at a local point. Unlike the LDA, the GGA takes into account not only the electron density at a point but also its gradient. The GGA is designed to improve the accuracy of DFT calculations over LDA.

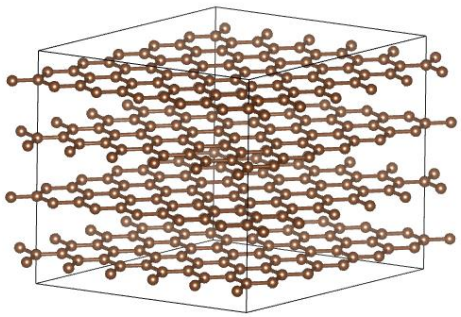


Fig. 1. Graphite $5 \times 5 \times 2$ super cell (336 atoms)

As shown in Fig. 1, the calculation was performed with 336 atoms using $5 \times 5 \times 2$ super cell. Also, an energy cutoff of 520 eV and the $6\times 6 \times 6$ k-point mesh were applied for the calculation. The PDOS is calculated by using PHONOPY software.

2.3 Calculation Results

In the ENDF/B-VII.1 library, the PDOS was processed using the GA (General Atomic) physics model in 1960s, referred to as the GA model[4]. The GA model was derived from fitting an experimental data (e.g., specific heat and compressibility) with the root sampling method. Recently, the ab-initio calculation has been introduced to generate the thermal scattering data for the solid materials including graphite. The ab-initio calculation-based thermal neutron scattering libraries have been released from the latest nuclear data libraries such as ENDF/B-VIII.0 and JENDL-5.

In this study, the newly calculated PDOSs were compared with those of the existing nuclear libraries such as ENDF/B-VII.1 and ENDF/B-VIII.0. As shown in Fig. 2, the GA model and ab-initio based PDOSs show noticeable discrepancies. In case of the experiment based PDOS, it might be inaccurate due to experimental errors, limitations of measurement, and incompleteness of data. Additionally, accurately separating contributions from multiple phonon excitations at lower energies cannot be corrected. On the other hand, the accuracy of the ab-initio calculation can be affected by the choice of computational model, the accuracy of exchange-correlation functionals, and the settings of computational parameters.

In the presented PDOS, the noticeable discrepancies were observed in the peak positions and intensities of the GA model when compared to those obtained using the VASP code. On the other hand, ab-initio-based PDOSs show high similarity to each other. Specifically, the peak positions of the PDOS calculated using LDA have better accordance with ENDF/B-VIII.0 than that of GGA at low energy range. The validity of the calculated PDOS is further corroborated by the comparison with the scattering cross sections.

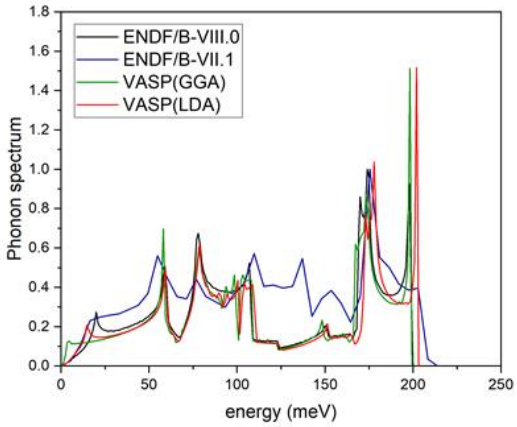
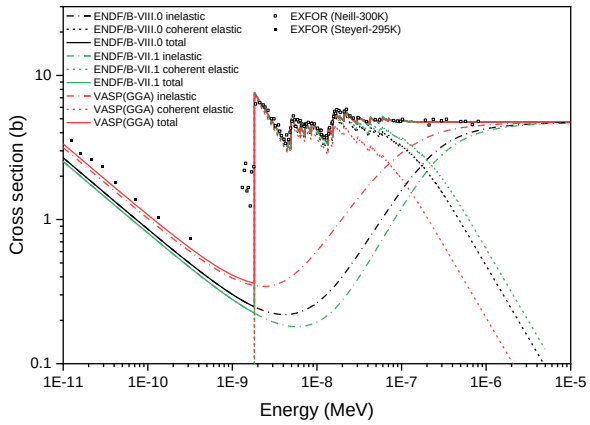


Fig. 2. Comparison of the phonon density of states

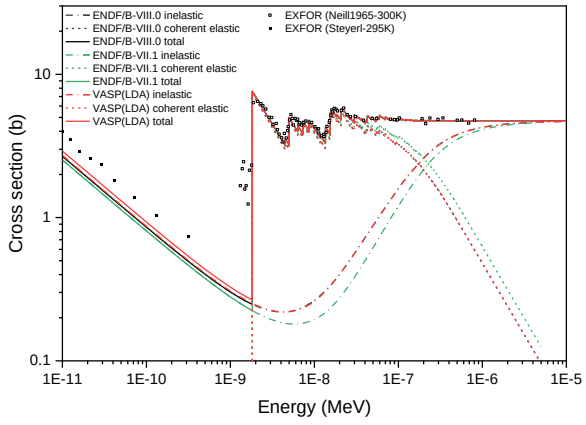
Figure 3 shows comparisons of scattering cross sections for graphite and experimental data[5,6]. Each scattering cross section was decomposed into two distinct contributions: the coherent elastic and inelastic components. As shown in Fig. 3, all total cross sections are in good agreement with the experimental data in the energy range above ~2×10^{-9} MeV. However, despite these agreements, each inelastic and elastic cross section exhibited different trends. Particularly, the calculated data using the GGA largely deviates from the other data. It is deduced that the observed discrepancy might be attributed to the peak of the PDOS at the low energy range ~5 meV. The PDOS at the small energy transfer range is important to calculate the inelastic scattering cross sections. According to Fig. 3. (a), the calculated

data using the GGA appear to most closely represent the experimental data in the low energy region. However, these results are inferred to be imprecise. Although it is known that the GGA is more accurate than the LDA, crystalline graphite comprises the strong intra-planar covalent bonding and weak inter-planar Van der Waals bonding. The GGA and LDA are effective in simulating intra-planar covalent bonds. However, it is known that the GGA is less successful in representing weak Van der Waals interactions, leading to the discrepancies observed in the calculations[7].

In fact, the interlayer distances calculated using the GGA and LDA were found to be 8.69 Å and 6.65 Å, respectively. When comparing the calculated results with the experimental interlayer distance of graphite, 6.678 Å[8], the LDA more accurately simulated the structure of graphite.



(a)



(b)

Fig. 3. Comparison of the scattering cross sections for graphite with experimental data: (a) the GGA applied and (b) the LDA applied

Additionally, the NJOY code applies the incoherent approximation in calculating the inelastic scattering cross section[2], which leads to inaccuracies in generating scattering cross sections. To mitigate these inaccuracies, it is known that an additional calculation of the coherent 1-phonon scattering law is necessary[9].

3. Conclusions

The thermal scattering cross sections for crystalline graphite were generated using ab-initio simulation code, VASP, and then compared with the existing library data. Notably, the scattering cross section data using the LDA exchange-correlation functional exhibited a good agreement with the ENDF/B-VIII.0 data.

For further work, the ICSBEP benchmark will be conducted to validate the thermal scattering data of graphite. Additionally, the reactor grade graphite has not been considered in this paper. The reactor grade graphite, characterized by its porosities, will be generated and analyzed in subsequent research.

ACKNOWLEDGEMENT

This work was supported by KAERI Institutional Program (Project No. 524560-24).

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