Annals of Nuclear Energy 87 (2016) 242–251

Contents lists available at ScienceDirect

Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

Thermal neutron scattering cross sections of beryllium and magnesium oxides

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ARTICLE INFO

Article history:
Received 25 May 2015
Received in revised form 8 September 2015
Accepted 10 September 2015

Keywords:
BeO
MgO
Phonons
Thermal neutron scattering cross section
Moderators
Filter efficiency

ABSTRACT

Alkaline-earth beryllium and magnesium oxides are fundamental materials in nuclear industry and thermal neutron scattering applications. The calculation of the thermal neutron scattering cross sections requires a detailed knowledge of the lattice dynamics of the scattering medium. The vibrational properties of BeO and MgO are studied using first-principles calculations within the frame work of the density functional perturbation theory. Excellent agreement between the calculated phonon dispersion relations and the experimental data have been obtained. The phonon densities of states are utilized to calculate the scattering laws using the incoherent approximation. For BeO, there are concerns about the accuracy of the phonon density of states used to generate the current ENDF/B-VII.1 libraries. These concerns are identified, and their influences on the scattering law and inelastic scattering cross section are analyzed. For MgO, no up to date thermal neutron scattering cross section ENDF library is available, and our results represent a potential one for use in different applications. Moreover, the BeO and MgO efficiencies as neutron filters at different temperatures are investigated. BeO is found to be a better filter than MgO, especially when cooled down, and cooling MgO below 77 K does not significantly improve the filter's efficiency.

1. Introduction

Alkaline earth oxides are an important class of materials used in wide range of industrial applications. In particular, beryllium and magnesium oxides (BeO and MgO) are fundamental materials in nuclear industry and neutron scattering applications (Holmryd and Connor, 1969; Carpenter et al., 1989; Thiyagarajan et al., 1998; Adib et al., 2011, 2014). They are used as thermal neutron monochromators and filters. This is due to the high coherent scattering cross sections, and low absorption and low incoherent scattering cross sections of Be, Mg, and O atoms, as well as the availability of oxides crystals with a small mosaic spread. In addition, because of its high thermal conductivity, BeO was one of the earliest materials considered to be used as a neutron moderator (Manly, 1964; Konings, 2012).

Monochromator crystals select a very narrow wavelength band from a broad incident thermal neutron spectrum using Bragg diffraction. Filters are needed to effectively transmit thermal neutrons while attenuating the unavoidable higher-order neutrons and gamma rays to reduce the radiation background. Moderators are used for slowing down and thermalizing neutrons to achieve higher fission rates. The design, setup and performance characteristics of neutron facilities to implement such applications are guided by computational simulations. The reliability of such simulations depends on the accuracy of the modeling of thermal neutrons interactions with the scattering medium, which requires a detailed knowledge of the lattice dynamics of the scattering medium (Kobayashi et al., 1996; Shirane et al., 2002). Therefore, appropriate neutron scattering cross section libraries for the material of interest are generated and used for various neutron scattering applications (Hawari et al., 2006).

The first BeO thermal neutron scattering cross sections library was generated in the late 1960s (Koppel and Houston, 1968), based on the empirical lattice dynamics data of Borgonovi (1968), calculated using the shell model. The adjustable parameters were obtained by fitting to the available elastic constants (Bentle, 1966), Raman scattering (Loh, 1968), and phonon dispersion relations measured by Brugger et al. (1967) using the inelastic neutron scattering technique. However, this data is quite limited: the measured phonon dispersion relations are only along the (ΓM) direction, and don't include all phonon branches. More importantly,

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http://dx.doi.org/10.1016/j.anucene.2015.09.006
0306-4549/

unphysical extra low-lying optical phonon branches (below 20 meV) are reported (Brugger et al., 1967). One year after this work, measurements of the BeO phonon dispersion relations were repeated (Ostheller et al., 1968), using the same single crystal. The measurements are extended over two high-symmetry directions (ΓM and ΓA), and the quality of the data was greatly improved. It was also found that the low lying phonon branches observed by Brugger et al. (1967) are not genuine branches of BeO, but instead resulted from multi-phonon processes (Ostheller et al., 1968). Surprisingly, the lattice dynamical model of Borgonovi (1968) and the thermal neutron scattering library were not updated according to these more accurate measurements of the phonon dispersion relations (Ostheller et al., 1968). On the contrary, the BeO library (ENDF/B-VII.1) was also recalculated and released (MacFarlane, 1994) in 1994 by Los Alamos National Laboratory, using the same old lattice dynamical data (Brugger et al., 1967).

On the other hand, the thermal neutron scattering cross section of MgO was modeled using a semi-empirical method originally proposed by Steyerl (1977) and Freund (1983), to study neutron transmission in Be, Si, Cu, Bi and Al_2O_3. In this model, the Debye temperature is used as the only fitting parameter (Thiyagarajan et al., 1998). This modeling was facilitated by the transmission measurements of thermal neutrons at different temperatures. Thiyagarajan et al. (1998), modeled their data using a Debye temperature of 938 K, while the same data were modeled again by Adib et al. (2011) and led to a different Debye temperature of 700 K. To date, no thermal neutron scattering library in the ENDF format is available for MgO.

In this work, we report first-principles phonon dispersion relations and phonon density of states (PDOS) calculations for BeO and MgO, using density functional perturbation theory. For both systems, our calculated phonon dispersion relations are found to be in excellent agreement with experiment, which reflects the high accuracy and reliability of the calculated PDOS. The deficiencies of the empirical PDOS of Borgonovi (1968) for BeO are identified. The PDOSs are then used to calculate the scattering laws within the incoherent approximation, which, in turn, are employed to generate the elastic and inelastic thermal neutron scattering cross sections at different temperatures. The results are discussed in comparison with the available experimental data. For BeO, a detailed comparison between our results and those obtained using the empirical PDOS of Borgonovi (1968) is provided, which clearly shows the necessity of starting from reliable PDOS and, based on the calculated neutron scattering cross sections, the efficiencies of BeO and MgO as neutron filters are investigated, as a function of temperature.

The rest of the paper is organized as follows: the required theoretical background and mathematical formulations are briefly introduced in Section 2, the *ab initio* computational details are summarized in Section 3, the results obtained are presented and discussed in Section 4, and finally, a summary of our main results and conclusions is provided in Section 5.

2. Theoretical background

The total cross section of thermal neutrons as a function of neutron incident energy (E) and the temperature of the medium (T) is given by

$$\sigma_{tot}(E,T) = \sigma_{ab}(E) + \sigma_{el}(E,T) + \sigma_{in}(E,T), \tag{1}$$

Here, $\sigma_{ab}(E)$ is the temperature independent absorption cross section given by

$$\sigma_{ab}(E) = \sigma_o \sqrt{\frac{E_\circ}{E}}, \tag{2}$$

where σ_o is the absorption cross section measured at a reference incident neutron energy $E_\circ$, $\sigma_{el}(E,T)$ and $\sigma_{in}(E,T)$ are the elastic and inelastic scattering cross sections, respectively.

The elastic scattering cross section $\sigma_{el}(E,T)$ consists of coherent and incoherent parts. For coherent scattering atoms (like Be, Mg and O), the incoherent elastic scattering cross section is negligible. In the coherent elastic (or Bragg) scattering, neutrons experience a specular reflection by the atomic crystal planes, and thus $\sigma_{el}(E,T)$ is given as (MacFarlane, 1994; Squires, 1978)

$$\sigma_{el}(E,T) = \frac{2\pi\hbar^2}{4mNVE} \sum_{E_i<E} \left| \sum_{j=1}^{N} \sqrt{\sigma_{c,j}} e^{-W_j(T)E_i} e^{2\pi i \vec{\tau}_i \cdot \vec{d}_j} \right|^2. \tag{3}$$

Here, N and V are the number of basis atoms and the volume of the primitive unit cell, respectively, m is the mass of the neutron, ħ is the reduced Plank constant, $\sigma_{c,j}$ and $\vec{d}_j$ are the coherent bound scattering cross section and the atomic position of the jth atom, respectively. The exponential term, $e^{-2W_j(T)}$, is the Debye–Waller factor and E_i are called Bragg edges – correspond to the reciprocal lattice vectors $\vec{\tau}_i$ and – given by $E_i = \frac{\hbar^2 \tau_i^2}{8m}$. For non-Bravais lattices with cubic symmetry,

$$2W_j(T) = \frac{1}{M_j/m} \frac{1}{k_B T} \int_0^{\beta_{max}} \frac{\rho_j(\beta)}{\beta} coth\ (\beta/2) d\beta, \tag{4}$$

where M_j and $\rho_j(\beta)$ are the mass and partial PDOS of the jth atom, respectively. β is a dimensionless variable defined as $\beta = \hbar\omega/k_B T$. Here, ω is the phonon angular frequency and k_B is the Boltzmann constant.

The inelastic scattering cross section $\sigma_{in}(E,T)$ is related to the double differential scattering cross section, $\frac{d^2\sigma}{dE' d\Omega}$, by

$$\sigma_{in}(E,T) = \int \int \frac{d^2\sigma}{dE' d\Omega}\, dE' d\Omega, \tag{5}$$

which, in turn, is given by (Bell and Glasstone, 1970)

$$\frac{d^2\sigma}{dE' d\Omega} = \frac{1}{4\pi k_B T} \sqrt{\frac{E'}{E}}\, e^{-\beta/2} (\sigma_c S(\alpha,\beta) + \sigma_{Inc} S_s(\alpha,\beta)). \tag{6}$$

Here, σ_c and σ_{Inc} are the coherent and incoherent bound scattering cross sections, respectively, and E' is the scattered neutron energy. $S(\alpha,\beta)$ is a dimensionless function known as the scattering law, given by

$$S(\alpha,\beta) = S_s(\alpha,\beta) + S_d(\alpha,\beta), \tag{7}$$

where $S_s(\alpha,\beta)$ is the self-scattering law that accounts for non-interference (incoherent) effects, and $S_d(\alpha,\beta)$ is the distinct-scattering law that accounts for interference (coherent) effects. α represents the dimensionless momentum transfer variable, given by

$$\alpha = \frac{E' + E - 2\mu\sqrt{EE'}}{2Ak_B T}, \tag{8}$$

where μ is the cosine of the scattering angle and A is the ratio of nuclear to neutron masses.

In order to calculate the scattering law and to generate the thermal neutron scattering cross section as a function of temperature and incident neutron energy, the LEAPR and THERMR modules of NJOY code are employed (MacFarlane, 1994; MacFarlane and Muir, 1994). The formulation of these modules is based on the so-called incoherent approximation, where the S_d term is neglected (Bell and Glasstone, 1970; MacFarlane, 1994). That is, $\frac{d^2\sigma}{dE' d\Omega}$ given by Eq. (7) is rewritten as