Thermal Neutron Scattering Cross Sections of Molybdenum Isotopes

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INTRODUCTION

Molybdenum and its alloys have found extensive applications in the nuclear industry. Besides existing in nuclear reactors as a product of 235U fission, molybdenum has been used in cladding and, most significantly, in advanced nuclear fuels in the form of U–Mo alloys such as U-10Mo [1–3].

U–Mo alloys are based on the solid solution of molybdenum in γ-uranium (the high-temperature bcc phase). Alloying pure γ-uranium with 6–12 wt% molybdenum produces a high-density γ-U–Mo alloy that exhibits high thermal conductivity, expands isotropically under irradiation, and maintains mechanical integrity at high burnup [4, 5]. Due to these characteristics, U–Mo alloys are under consideration by the Global Threat Reduction Initiative as low-enrichment replacement fuels for U–Mo alloys are under consideration for trans-uranium–burning advanced research and test reactors, which are currently operating with highly-enriched uranium fuels. They are also being considered as potential fuels for trans-uranium–burning advanced fast-neutron nuclear reactors [6].

Despite the existence of numerous studies concerning the mechanical and thermal properties of U–Mo alloys and how these properties change under irradiation [4, 5, 7–9], the effect of molybdenum on thermal neutron transport within the fuel is not sufficiently known. Of particular concern is the effect on the thermal neutron spectrum, which drives the fission rate and is used to calculate reactor physics parameters such as reactivity coefficients [10]. Since the thermal neutron scattering properties of the materials within the fuel matrix affect the neutron spectrum, thermal neutron scattering cross section libraries of U–Mo alloys are needed for use within neutron transport models.

A first step towards the calculation of thermal scattering data for U–Mo alloys is the calculation of thermal neutron scattering cross sections for pure molybdenum. In this work, we have generated the thermal neutron scattering cross sections of molybdenum isotopes (92Mo, 95Mo, 96Mo, and 100Mo) using NJOY [11]. The phonon density of states (PDOS), which is required for the cross section calculation, is calculated from molecular dynamics (MD) simulations. The resulting cross sections are assessed by comparing the results obtained for 96Mo with an experimentally-measured phonon density of states.

THEORY

There are two types of scattering when neutrons collide in solids, incoherent and coherent scattering. Incoherent scattering arises when the incident neutron wave interacts independently with each nucleus and the scattered waves do not interfere with each other. Coherent scattering, in contrast, arises when the incident waves interact with the sample as a whole and the scattered waves interfere with each other. Both coherent and incoherent scattering can be divided into elastic and inelastic scattering. Elastic scattering is the case in which there is no change in the kinetic energy of the incident neutron. Inelastic scattering results in an exchange of energy between the neutron and the sample, thereby producing or absorbing phonons. With this in mind, thermal scattering cross sections are usually divided into inelastic and elastic cross sections, and both of them contain coherent and incoherent parts.

To calculate the inelastic cross section in a crystal, NJOY [11] uses the incoherent approximation [12], which assumes that coherent interference can be neglected. Within this approximation, the inelastic scattering cross section for a neutron of initial energy $E$ to be scattered at an angle $\theta$ with final energy $E'$ is

$$\sigma(E \rightarrow E', \mu) = \frac{\sigma_b}{2k_bT} \sqrt{\frac{E'}{E}} S(\alpha, \beta),$$

where $\mu = \cos \theta$, $\sigma_b$ is the total bound scattering cross section, $T$ is the temperature, and $k_b$ is the Boltzmann constant. $S(\alpha, \beta)$ is the scattering law, which is directly dependent on the phonon density of states (also called phonon frequency distributions). The variables $\alpha$ and $\beta$ represent the momentum transfer and energy transfer, respectively, given by

$$\alpha = \frac{E' + E - 2\mu V E \sqrt{E'E'}}{A k_b T}, \quad \beta = \frac{E' - E}{k_b T},$$

where $A$ is the ratio of the mass of the scattered atom to the neutron mass and $\mu$ is the cosine of the scattering angle in the laboratory frame.

For coherent scatterers such as molybdenum, the incoherent elastic effect is negligible [11]. The coherent elastic cross section for a material containing a single atomic species is

$$\sigma(E, \mu) = \frac{2\pi \hbar^2 \sigma_{coh}}{4mNVE} \sum_{i=E/V}^{N} \sum_{r_i} |F(\tau)|^2 e^{-4W_e},$$

where $N$ is the number of atoms in a primitive unit cell of volume $V$, $m$ is the mass of the neutron, $\hbar$ is the reduced Planck constant, $\sigma_{coh}$ is the coherent bound scattering cross section, and $F(\tau)$ is the unit cell structure factor. The factor $W$ is the Debye–Waller factor, which is calculated using the phonon density of states of molybdenum, and $E_i$ are the Bragg edges, which correspond to the reciprocal lattice vectors $r_i$ according to the equation

$$E_i = \frac{\hbar^2 r_i^2}{8m}.$$

COMPUTATIONAL DETAILS

The phonon density of states, which is the most important input required to calculate the thermal scattering cross section, was obtained for each isotope through molecular dynamics simulations using the Phonon fix of Kong [13] as implemented in LAMMPS [14]. The interatomic potential used was the embedded atom method (EAM) potential of Smirnova et al. [15] for the ternary system U–Mo–Xe.

All simulations were performed on a triclinic cell of $10 \times 11 \times 12$ primitive cells (1320 atoms). The system was equilibrated at 300 K/0 bar using a Nosé–Hoover [16, 17] thermostat under Parrinello–Rahman dynamics [18, 19]. Following equilibration, the thermo/barostats were discontinued and the instantaneous positions of the atoms were computed under ordinary velocity Verlet integration for 100 ns. Eight independent runs with different initial velocity seeds were used to obtain better averages. The phonon density of states was generated via the post-processing software PHANA [13] using a Monkhorst–Pack [20] mesh of $80 \times 80 \times 80$ $q$-points.

The phonon density of states was then run through the LEAPR and THERMR module of the NJOY code [11] to calculate the elastic and inelastic scattering cross sections at 300 K.

RESULTS AND ANALYSIS

The phonon density of states of $^{96}$Mo is presented in Figure 1, along with data calculated by Powell et al. [21] from experimental dispersion curves measured by inelastic neutron scattering at 296 K. The MD results mimicked the two-peak feature of the PDOS derived from experimental dispersion relations, but failed to reproduce the location and intensity of the peaks. As can be seen, the intensity of the low energy peak is overestimated, and the peaks are shifted to lower energies by as much as 3 meV.

The calculated phonon densities of states for different isotopes are shown in Figure 2. As expected, the phonon densities of states for all the isotopes have the same characteristics, but the curves are shifted to higher frequencies for lighter atoms. This is due to the fact that the isotopes with lower mass vibrate at higher frequencies ($\omega \propto m^{-1/2}$).

Figure 3 shows the elastic and inelastic thermal scattering cross section of $^{96}$Mo evaluated from the phonon density of states as calculated from molecular dynamics simulations. The results are compared to the cross sections obtained from the frequency distribution calculated by Powell et al. [21]. As can be seen, the calculated inelastic cross sections are in good agreement with the data derived from the experimental PDOS, especially in the high-energy region. The deviations below 0.3 eV result from the shift of the peaks in the PDOS to lower energy values. Likewise, the calculated elastic cross sections are in excellent agreement with the results obtained from the available experimental estimates of the density of states.
cross sections have the same features for all isotopes. The values of the cross sections, however, are different for each isotope. These variations arise from the dependence of the cross section not only on the phonon density of states, but also on the bound scattering cross section and the nuclear spin.

Fig. 4. Calculated elastic and inelastic thermal neutron scattering cross sections for four isotopes of molybdenum at $T = 300$ K.

The total scattering cross section of $^{96}$Mo compared to the scattering cross section for a free nucleus from the ENDF/B-VII.1 evaluation [22] are shown in Figure 5. As can be seen, the free nucleus cross sections are larger than the calculated values for the bound nucleus at lower energies but approximate the bound cross sections at higher energies. Similar results were obtained for the other isotopes studied.

Fig. 5. Total thermal neutron scattering cross sections of $^{96}$Mo at $T = 300$ K compared to the free nucleus ENDF/B-VII.1 data [22].

CONCLUSIONS

In this work, the phonon densities of states for different isotopes of molybdenum were calculated based on molecular dynamics simulations. The results were assessed by comparing the results for $^{96}$Mo with available frequency distributions calculated from experimental dispersion relations. The generated phonon density of states, along with the available experimental data, was then used to calculate the inelastic and elastic thermal scattering cross sections at 300 K using NJOY [11].

The simulations provided similar results for all isotopes of molybdenum studied. When comparing the results of $^{96}$Mo to the data of Powell et al. [21], the simulations succeeded in reproducing the characteristics expected of the phonon density of states, but did not manage to reproduce quantitatively the intensities and locations of the peaks. Despite these differences, the MD simulations generate cross sections that are in excellent agreement with the results calculated from the experimental phonon density of states of Powell et al. [21]. This is due to the fact that the cross section is an integrated quantity, and as such the minor inaccuracies in the density of states have relatively little impact on the cross section.

Comparisons with ENDF/B-VII.1 data show how the free atom model misestimates the cross section below 0.01 eV, which indicates that the obtained thermal cross sections should be better over the free atom cross sections actually used.

The generated cross sections have been compiled into ACE format and are available to use in neutron transport software such as MCNP [23]. The cross sections calculated in this work will allow for more accurate modeling of neutron interactions within the nuclear fuel cycle in the thermal energy range. These calculations are also an important first step in obtaining cross sections relevant to U-10Mo and similar alloy fuels.

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REFERENCES
