Generation of Thermal Scattering Laws for YH$_2$ using Ab Initio Methods

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INTRODUCTION

Metal hydrides such as zirconium hydride (ZrH$_x$) have been used as a moderator/reflector material in nuclear reactors because of their ability to retain hydrogen at elevated temperatures. Yttrium hydride (YH$_2$) is of interest as a potential high temperature moderator/ reflector material because of its superior hydrogen density at elevated temperatures. The *ab initio* approach [1] is used to generate thermal neutron scattering laws (TSLs) for hydrogen in yttrium hydride (H-YH$_2$) and yttrium in yttrium hydride (Y-YH$_2$). The *ab initio* code VASP (Vienna ab initio Simulation Package) [2] and lattice dynamics code PHONON [3] were used to generate the dispersion relations and phonon frequency distributions for YH$_2$. This information was then used to prepare H-YH$_2$ and Y-YH$_2$ thermal scattering laws using the LEAPR module of the NJOY code. VASP/PHONON calculations have previously been used to generate TSL data for Be [1], high density graphite [1,4], ThH$_2$ [5], α-SiO$_2$ [6], and were used to prepare the ENDF/B-VII.1 TSL evaluation for α-SiO$_2$ [7].

CRYSTAL STRUCTURE AND LATTICE DYNAMICS

YH$_2$ has a CaF$_2$ type face-center cubic (FCC) structure. The unit cell is composed of 12 atoms with the hydrogen atoms located in the tetrahedral holes between the Y atoms. The structure of the YH$_2$ unit cell is shown in Fig. 1 with the Y atoms colored blue and the H atoms colored light grey. As optimized by VASP, this structure has a lattice constant of a = 5.2032 Å, which agrees with the average experimental value of a = 5.204 Å [8].
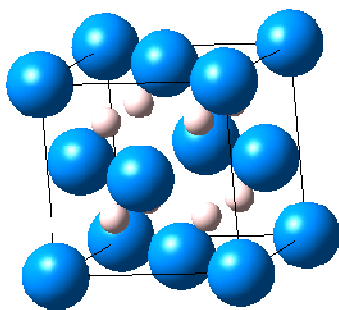
Fig. 1. The YH$_2$ unit cell.

The VASP code performed first principles quantum mechanics simulations of the YH$_2$ lattice structure using density functional theory with a generalized gradient approximation (GGA) and a projector augmented wave (PAW) pseudopotential. The YH$_2$ lattice structure was optimized to a total energy threshold of 0.0001 eV using the MedeA automated convergence tool [9]. The dispersion relations and density of state were calculated using the MedeA-PHONON tool. A 2×2×2 supercell (96 atoms) with a 250 eV planewave cutoff energy was used to calculate the dispersion relations. The phonon frequency distribution (density of states) was calculated using a ± 0.02 Å displacement.

Fig. 2 shows the dispersion relations along the highest symmetry points of the Brillioun zone derived from the PHONON calculation. The lower branches are acoustical modes which are mainly due to the heavy Y atom vibrations. The higher branches are optical modes mainly due to the H atom vibrations.
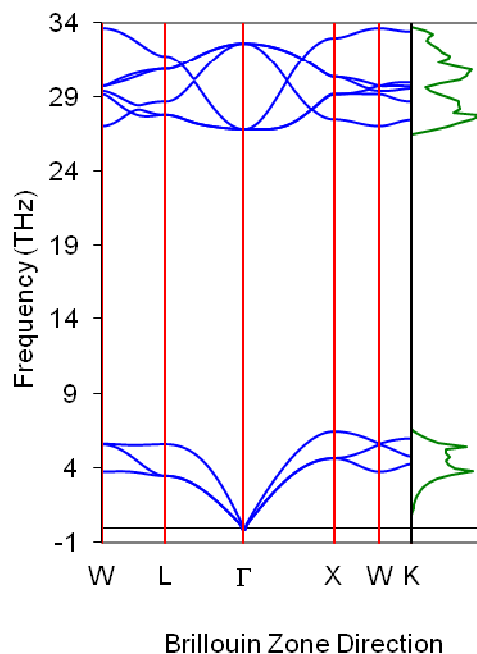

Fig. 2. Ab initio calculated YH$_2$ dispersion relations

Fig. 3 shows the partial phonon frequency distributions calculated for YH$_2$. The phonon density of states has two well-separated regions (due to the large

mass ratio between Y and H): an acoustical region (0-0.027 eV) which is the preferred region for Y atom vibrations and an optical region (0.110-0.140 eV) which is the preferred region for H atom vibrations. The optical region is centered at ~0.125 eV which compares well with the 0.128±0.004 eV estimate by Flotow [10] derived from heat capacity measurements and the 0.127±0.007 eV value recommended by Rush [11] derived from neutron scattering measurements. The YH_2 optical mode in more recent neutron scattering measurements by Udovic [12] displayed a similar bimodal structure centered at ~0.125 eV.

Thermodynamic properties of YH_2 can also be calculated from the phonon density of states. Fig. 4 show the calculated heat capacity for YH_2 which compare well with measurements by Flotow [10].
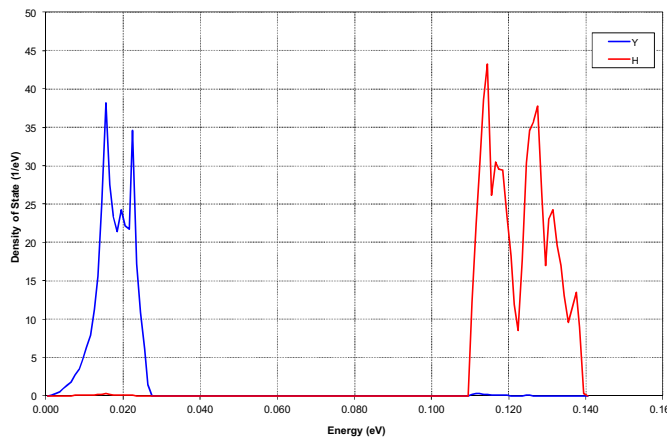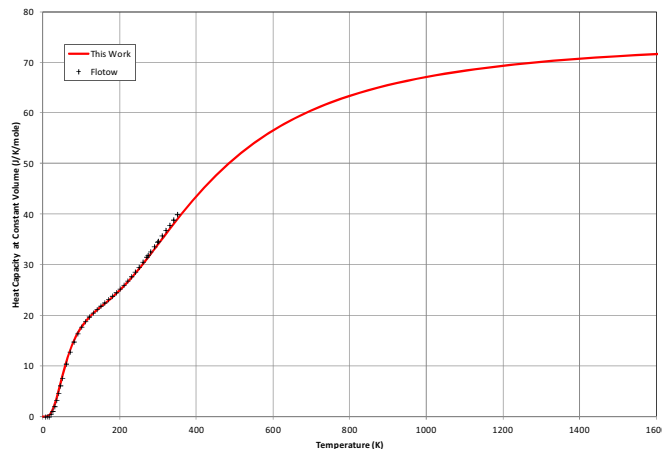

Fig. 3. Phonon frequency distribution for YH_2.

Fig. 4. Molar heat capacity for YH_2.

THERMAL SCATTERING LAWS

TSLs for H-YH_2 and Y-YH_2 were developed from the phonon frequency distributions calculated by MedeA-PHONON using the LEAPR module of NJOY [13].

ENDF/B-VII.1 atomic mass ratio and free atoms scattering cross section data for ^1H and ^{89}Y were used to develop the TSLs. The α and β mesh were borrowed from the ENDF/B-VII.0 H-ZrH and Zr-ZrH evaluations. TSLs were generated at 10 temperatures: 293.6, 400, 500, 600, 700, 800, 1000, 1200, 1400 and 1600 K. Fig. 5 shows the inelastic scattering and incoherent elastic scattering cross sections for H-YH_2. The inelastic scattering and incoherent elastic scattering cross sections for Y-YH_2 given in Fig. 6.

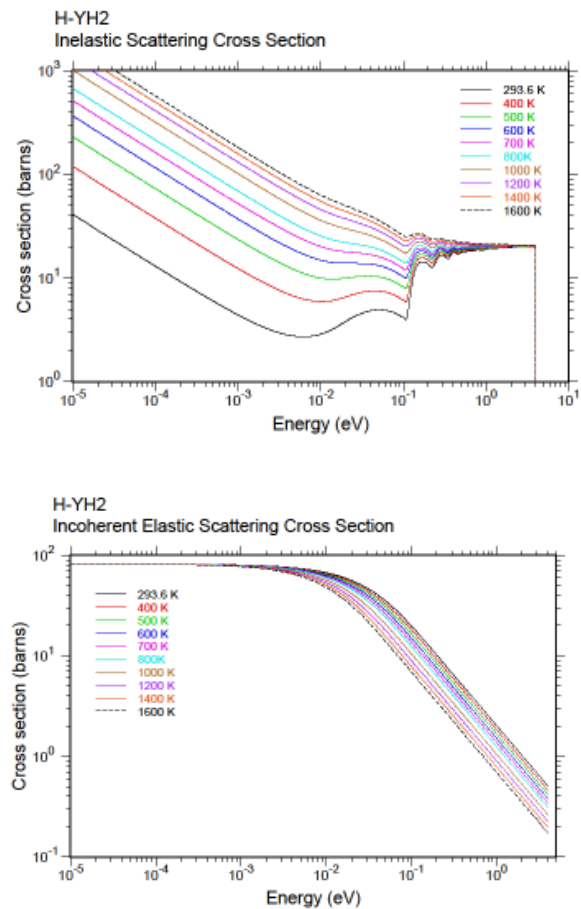Fig. 5. Inelastic scattering (top) and incoherent elastic scattering (bottom) cross sections for H-YH_2.

Y-YH2
Inelastic Scattering Cross Section

Y-YH2
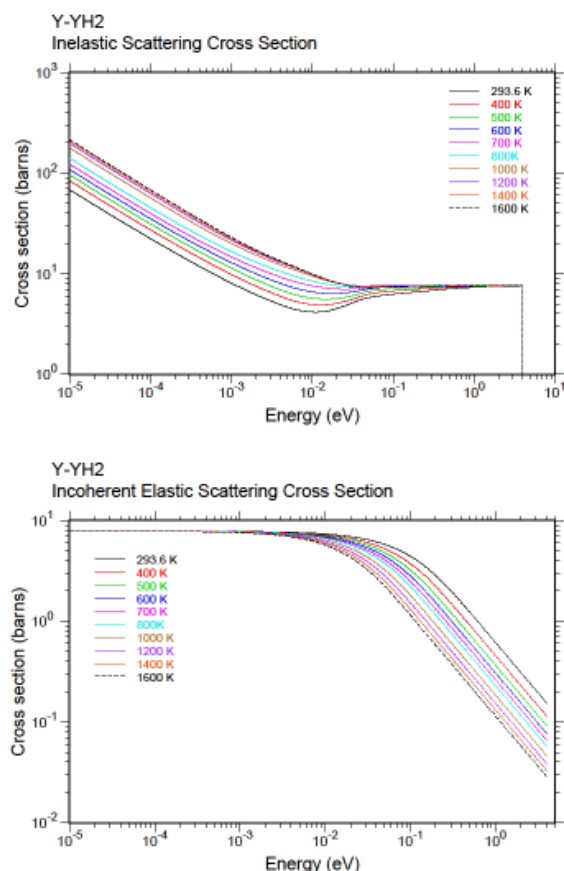Incoherent Elastic Scattering Cross Section

Fig. 6. Inelastic scattering (top) and incoherent elastic scattering (bottom) cross sections for Y-YH$_2$.

CONCLUSIONS

Computational material science tools such as VASP and PHONON appear to have matured to the point where they can be used to calculate TSLs for new moderator materials of interest in nuclear applications. TSLs for H-YH$_2$ and Y-YH$_2$ have been generated using the *ab initio* approach and compare well to the limited available measurements. Cross section measurements are needed to validate the YH$_2$ TSL evaluations.

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