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

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Motivation

- Metal hydrides (ZrH$_x$) used as high temperature moderator/reflector in nuclear reactors
  - TRIGA
  - SNAP-10
  - TOPAZ
- Yttrium hydride (YH$_x$) has superior hydrogen density ($N_H$) at elevated temperatures
- Test improvements to MedeA software, use *ab initio* approach to generate TSLs for new moderator materials

Hydrogen in metallic Zr, Ce, Y and Ca in equilibrium with 1 atm $H_2$ at various temperatures. (Source: *Metal Hydrides*, Academic Press, p. 442, 1968)

$N_H$ is number of hydrogen atoms/cc x $10^{-22}$
Yttrium – Hydrogen Phase Diagram

- H has limited solubility in α-Y at low temperatures.
- H/Y < 2.0 is region of practical interest for nuclear applications
  - Mixture of α-Y + YH₂ phases at lower temperatures (see next slide)
  - YH₂-x at elevated temperatures
- YH₂ phonon frequency distribution needed to develop thermal scattering law (TSL) for YH₂-x

(Source: Metal Hydrides, Academic Press, p. 443, 1968)
Microstructure of Yttrium Hydride

Y-1.29% H alloy (H/Y = 1.15). Structure consists of 50% α-yttrium solid solution (dark regions) and 50% hydride phase (light regions)

Y-2.17% H alloy (H/Y = 1.96). The structure is nearly all hydride phase.

(Source: Metal Hydrides, Academic Press, pp. 447-448, 1968)
**YH₂ Crystal Structure**

- YH₂ has a CaF₂ type FCC structure
  - 12 atoms
  - 4 Y atoms (blue) at vertices and faces of cubic unit cell
  - 8 H atoms (grey) located in tetragonal holes between Y atoms
- Lattice constant a = 5.2032 Å
- MedeA calculated value compares well with average experimental value of a = 5.204 Å from X-ray diffraction measurements.
Ab Initio Approach Using MedeA

- MedeA software used to perform the *ab initio* calculations
- Informatica tool used to select the initial unit cell structure from the ICSD (crystal structure) database
- Automated Convergence tool used to optimize unit cell structure
  - Converge k-mesh
  - Converge planewave cutoff energy
  - Converge supercell size
  - Iterative VASP *ab initio* calculations
- PHONON tool performs the lattice dynamics calculations on optimized structure
  - Dispersion relations
  - Phonon density of states
  - Thermodynamic properties
- NJOY/LEAPR used to generate TSL from phonon frequency distribution
Optimization of the YH$_2$ Structure

- **VASP calculations**
  - Detailed quantum mechanical first principle simulation of crystal lattice using density functional theory
  - Generalized Gradient Approximation (GGA)
  - Projector Augmented Wave (PAW) pseudopotential

- **Optimized structure to a total energy threshold of 0.1 meV**
  - Required 42 VASP calculations
  - 537.5 eV planewave cutoff energy
  - k-spacing of ~0.05 1/Å
  - 42×42×42 final supercell size
  - Optimized lattice constant $a = 5.20319995$ Å

- **Optimized lattice structure then used in PHONON calculations**
YH₂ Dispersion Relations

- PHONON calculation use
  - 2×2×2 supercell (96 atoms)
  - Planewave cutoff energy of 250 eV
  - k-spacing of 0.5/Å (5×5×5 k-mesh)
  - ± 0.02 Å displacement

- Dispersion relations (at right)
  - Well separated acoustical and optical modes (as expected)
  - Lower branches are acoustical modes mainly due to heavy Y atom vibrations
  - Higher branches are optical modes mainly due to light H atom vibrations
YH$_2$ Phonon Frequency Distribution

Density of State (1/eV)

Energy (eV)

- Y-YH$_2$
- H-YH$_2$
Thermodynamic Properties Agree with Measurements

YH₂ Molar Heat Capacity

- This Work
- Flotow

Heat Capacity at Constant Volume (J/K/mole)

Temperature (K)

Heat Capacity at Constant Volume (J/K/mole)

Temperature (K)
Thermodynamic Properties Agree with Measurements

YH₂ Molar Entropy

- This Work
- Flatow

Entropy (J/K/mole) vs Temperature (K)
Thermodynamic Properties Agree with Measurements

YH₂ Enthalpy Increment

- This Work
- Flotow
LEAPR Models for YH$_2$ TSLs

- H-YH$_2$ and Y-YH$_2$ TSL generated using LEAPR
- Phonon frequency distributions from PHONON calculation
- Incoherent approximation
- Atomic mass ratios and free atom scattering cross sections from ENDF/B-VII.1 $^1$H and $^{89}$Y evaluations
- $\alpha$ and $\beta$ mesh borrowed from ENDF/B-VII.0 ZrH TSLs
- 10 temperatures (293.6, 400, 500, 600, 800, 1000, 1200, 1400, 1600 K)
H-YH₂
Incoherent Elastic Scattering Cross Section

Cross section (barns)

Energy (eV)
Y-YH2
Incoherent Elastic Scattering Cross Section

Cross section (barns)

Energy (eV)
Conclusions

- *ab initio* tools like MedeA have sufficiently matured and can be used to generate TSLs for new moderator materials
- H-YH$_2$ and Y-YH$_2$ TSLs produced using the *ab initio* approach
- Results consistent with available measurements
- Need to review $\alpha$ and $\beta$ mesh to confirm it adequately resolves the $S(\alpha,\beta,T)$ functions for H-YH$_2$ and Y-YH$_2$
- Cross section measurements would be of use to more completely validate TSLs for ENDF/B