

Generation of Thermal Scattering Laws for YH₂ using Ab Initio Methods

Michael L. Zerkle Bettis Atomic Power Laboratory

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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)

 $N_{\rm H}$ 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 + δ-YH₂ phases
 - δ -YH₂ phonon frequency
- δ-YH₂ phonon frequency distribution needed to develop thermal scattering law (TSL) for YH_{2-x}



FIG. 10.2 Partial phase diagram of the yttrium-hydrogen system.

(Source: Metal Hydrides, Academic Press, p. 443, 1968)



YH₂ Crystal Structure

- YH_2 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 lattice constant compares **extremely well** with recent X-ray diffraction measurements by Daou and Vajda [1992]
 - a = 5.2032(3) Å (90 K)
 - a = 5.2085(3) Å (295 K)





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 Covergence 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



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Optimization of the YH₂ 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 $\sim 0.05 \text{ } 1/\text{\AA}$
 - 42×42×42 final k-mesh
 - Optimized lattice constant a = 5.20319995 Å
- Optimized lattice structure then used in PHONON calculations

		Con	vergen	ce Job	48 (fini	shed) o	n serve	r local			
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E Display onl	ly stops from	Final	Phes	e							
Rafresh Step	Job Columns	Display									
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35	18 0	PV/C only	475	0.697	44.4	3.938 s	45.3118	0.0003	-5.10893	0.0001	
	19 0	PV/C only	487.5	0.697	44.4	3.875 s	45.3125	0.0007	-5.10017	0.00024	
	20 0	PV/C only	500	0.697	44.4	3.859 s	45.3125	0	-5.10417	0	
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	21.18	Panel	512.5	0.0617747	34 34 34	3 m 2,516 s	45.3378	0.0003	-0.11253	0.0001	
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			1								



YH₂ Dispersion Relations

- PHONON calculation use
 - 2×2×2 supercell (96 atoms)
 - Planewave cutoff energy of 250 eV
 - k-spacing of $0.5/\text{\AA}$ (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







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Thermodynamic Properties Agree with Measurements

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LEAPR Models for YH₂ TSLs

- H-YH₂ and Y-YH₂ 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 ¹H and ⁸⁹Y evaluations
- α and β mesh borrowed from ENDF/B-VII.0 ZrH TSLs
- 10 temperatures (293.6, 400, 500, 600, 800, 1000, 1200, 1400, 1600 K)

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Y-YH2 Incoherent Elastic Scattering Cross Section 10¹ 293.6 K 400 K 500 K Cross section (barns) 600 K 700 K 10⁰ 800K 1000 K 1200 K <mark>1400 K</mark> 1600 K 10 10⁻² -10⁻⁵ 10⁻³ 10⁰ 10⁻⁴ 10⁻² 10⁻¹ Energy (eV)

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Conclusions

- *ab initio* tools like MedeA (VASP, PHONON) have sufficiently matured and can be used to generate TSLs for new moderator materials
- H-YH₂ and Y-YH₂ TSLs produced using the *ab initio* approach
- Results consistent with available measurements
- Currently reviewing α and β mesh to ensure it adequately resolves the S(α , β ,T) functions for H-YH₂ and Y-YH₂
- Working to submit H- YH_2 and Y- YH_2 evaluations to CSEWG