Supporting Information

Theoretical Insights into High-T_c Superconductivity of Structurally Ordered $YThH_{18}$: A First-Principles Study

Abdul Ghaffar,[*,†] Peng Song,[‡] Ryo Maezono,[¶] and Kenta Hongo[*,§]

†*School of Information Science, JAIST, 1-1 Asahidai, Nomi, 923-1211, Ishikawa, Japan* [a]

‡*School of Information Science, JAIST, 1-1 Asahidai, Nomi, 923-1211, Ishikawa, Japan* [b]

¶*School of Information Science, JAIST, 1-1 Asahidai, Nomi, 923-1211, Ishikawa, Japan*

§*Research Center for Advanced Computing Infrastructure, JAIST, 1-1 Asahidai, Nomi, 923-1211, Ishikawa, Japan*

E-mail: mwkabd2101@icloud.com; kenta_hongo@icloud.com

[a]Current Address: Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-2008, United States

[b]Current address: Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan

Table S1: Crystal structures of Y-Th-H predicted at each pressure (P). Lattice parameters (a, b and c) are given in unit of Å.

Compound	Space group	P (GPa)	Lattice parameters	Atomic coordinates (fractional)

Continued on next page

Table S1 – continued from previous page

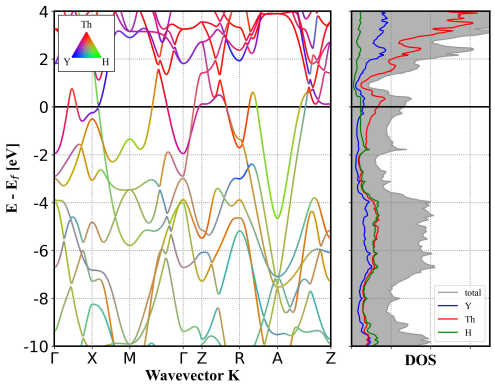
Compound	Space group	P (GPa)	Lattice parameters	Atomic coordinates (fractional)			
				Atoms	x	y	z
				Atoms	x	y	z
$YThH_8$	$P4/mmm$	200	a = 2.71221	Y(1b)	-0.00000	0.00000	0.50000
			b = 2.71221	Th(1c)	0.50000	0.50000	-0.00000
			c = 5.52173	H(4i)	-0.00000	0.50000	0.26329
			α = 90.0°	H(2g)	0.00000	0.00000	0.14890
			β = 90.0°	H(2h)	0.50000	0.50000	0.37151
			γ = 90.0°				
Y_2ThH_{12}	$I4/mmm$	200	a = 2.69993	Y(2e)	0.829360	0.829360	0.341270
			b = 2.69993	Th(1b)	0.500000	0.500000	0.000000
			c = 8.35315	H(4g)	0.910710	0.410710	0.178580
			α = 80.7°	H(2e)	0.948380	0.948380	0.103240
			β = 80.7°	H(2e)	0.713000	0.713000	0.574010
			γ = 90.0°	H(2e)	0.625680	0.625680	0.748630
				H(2d)	0.750000	0.250000	0.500000
$YThH_{18}$	$P\bar{6}m2$	200	a = 3.52410	Y(1e)	1.000000	0.000000	0.000000
			b = 3.52410	Th(1d)	0.666670	0.333330	0.500000
			c = 5.20468	H(6n)	0.022580	0.511290	0.172700
			α = 90.0°	H(6n)	0.181690	0.363370	0.305830
			β = 90.0°	H(2i)	0.000000	1.000000	0.397400
			γ = 120.0°	H(2h)	0.666670	0.333330	0.110920
				H(1a)	0.333330	0.666670	0.000000
				H(1b)	0.333330	0.666670	0.500000

Continued on next page

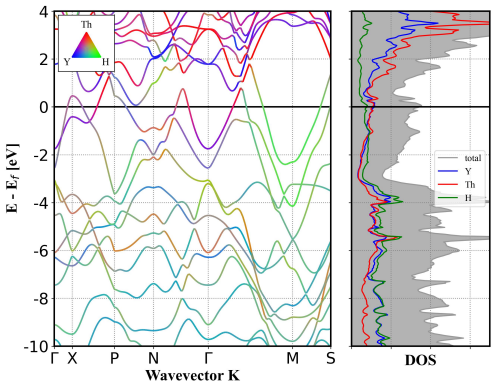
Table S1 – continued from previous page

Compound	Space group	P (GPa)	Lattice parameters	Atomic coordinates (fractional)			
				Atoms	x	y	z
$YThH_8$	$P4/mmm$	300	a = 2.57068	Y(1b)	0.00000	0.00000	0.50000
			b = 2.57068	Th(1c)	0.50000	0.50000	0.00000
			c = 5.40869	H(4i)	0.00000	0.50000	0.26272
			α = 90.0°	H(2g)	-0.00000	0.00000	0.15549
			β = 90.0°	H(2h)	0.50000	0.50000	0.36811
			γ = 90.0°				
Y_2ThH_{12}	$I4/mmm$	300	a = 2.56013	Y(2e)	0.829050	0.829050	0.341700
			b = 2.56013	Th(1b)	0.500000	0.500000	1.000000
			c = 8.17191	H(4g)	0.911260	0.410080	0.178960
			α = 81.0°	H(2e)	0.946170	0.946170	0.107740
			β = 81.0°	H(2e)	0.713880	0.713880	0.572580
			γ = 89.7°	H(2e)	0.624740	0.624740	0.750800
				H(2d)	0.749790	0.250210	0.500000

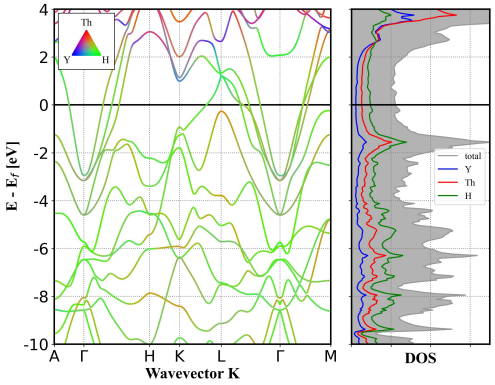
(a) *P4/mmm*-$YThH_8$ at 200GPa



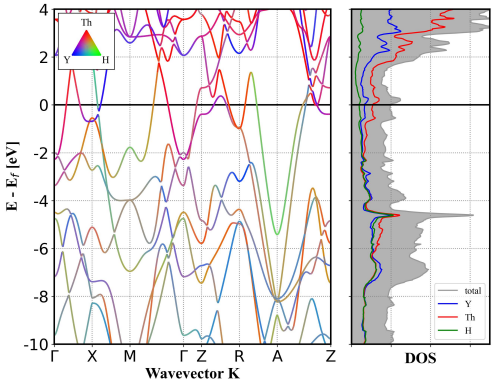
(b) *I4/mmm*-Y_2ThH_{12} at 200GPa



(c) *P*$\bar{6}$*m2*-$YThH_{18}$ at 200GPa



(d) *P4/mmm*-$YThH_8$ at 300GPa



(e) *I4/mmm*-Y_2ThH_{12} at 300GPa

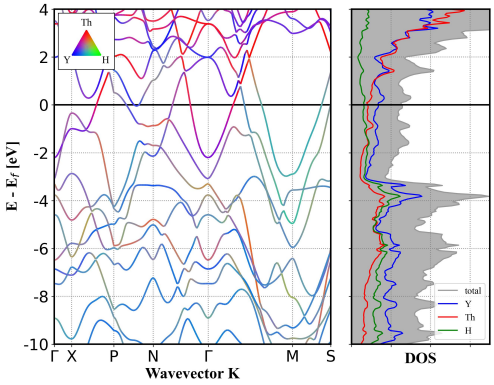


Figure S1: Electronic band structure together with projected density of states (PDOS) for all the stable structures.

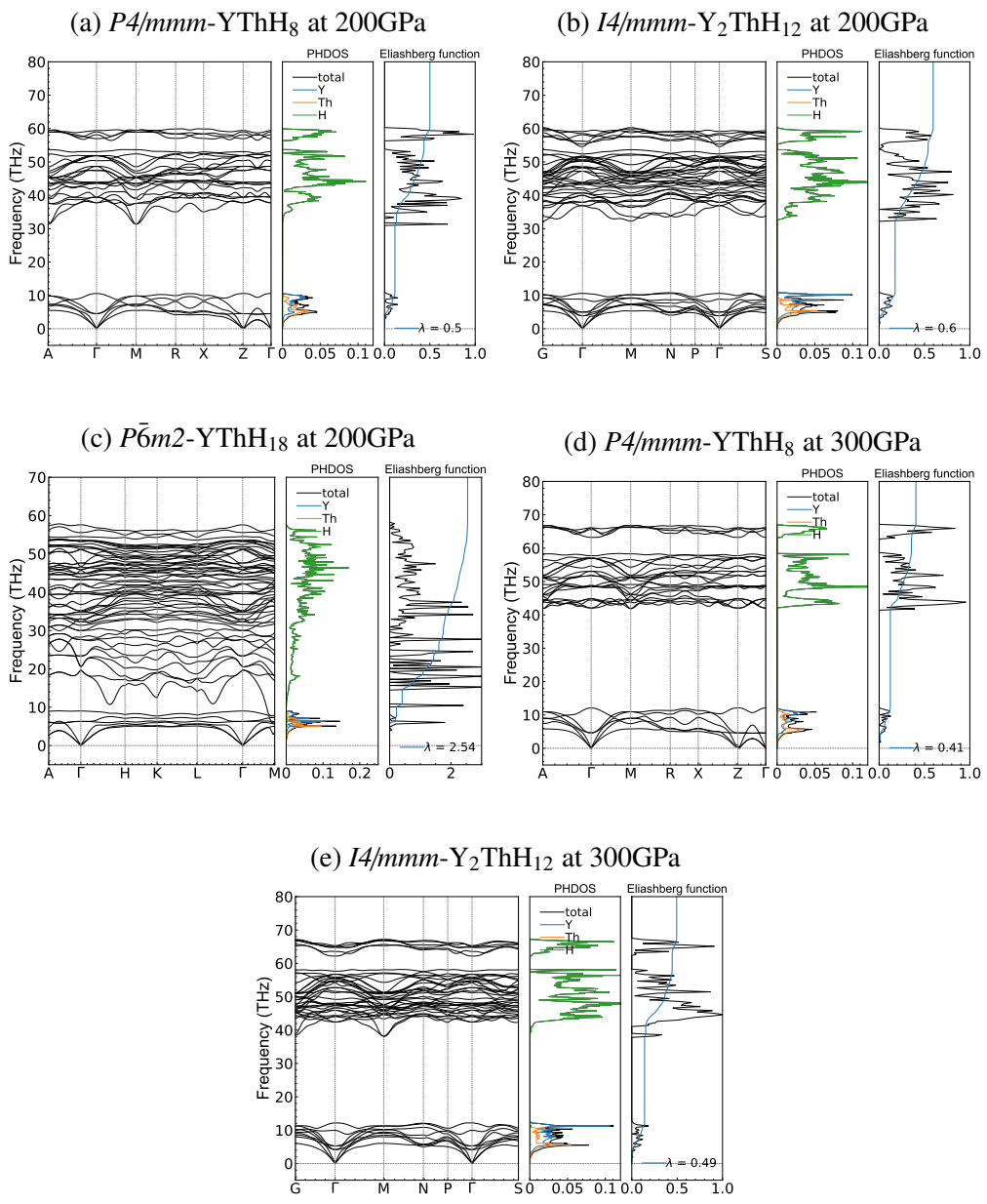


Figure S2: Phonon dispersion, projected phonon density of states (PHDOS), and Eliashberg spectral function α^2F(ω) associated with electron-phonon integral ($\lambda(\omega)$) for all the stable compounds. No imaginary modes suggest that all the structure in addition to their thermodynamic stability are dynamically stable.

Workflow for Novel Pressurized Superconducting Ternary Hydride Discovery and T_c Calculation

1. Set Up Problem Definition

- **Select Elements (Y-Th-H):** Choose the ternary hydride system (Y-Th-H) based on literature, known properties, and/or chemical intuition.
- **Define Pressure Conditions:** Specify the pressure range under which the hydride is expected to form, as high pressures often stabilize hydrogen-rich phases based on earlier work.

2. Evolutionary Algorithm (EA) Search for Stable Structures

- **Use of Evolutionary Algorithm (e.g., USPEX, CALYPSO):**
  - **Step 1: Generate Initial Population:** Start with random crystal structures in the desired composition system under the target pressure.
  - **Step 2: Energy and Structural Relaxation:** For each generated structure, perform initial energy minimization and structural relaxation using density functional theory (DFT) (e.g., with VASP or Quantum ESPRESSO).
  - **Step 3: Fitness Evaluation:** Rank structures by their formation energy (from DFT) and select the lowest energy structures for reproduction (crossover and mutation).
  - **Step 4: Iterative Evolution:** Continue the evolutionary search over several generations, with DFT energy and force evaluations guiding the algorithm to find the most stable structures.

3. Structure Optimization and Stability Check

- **Phonon Calculations (e.g., Using DFT or DFPT):**

- Perform phonon dispersion calculations to check the dynamical stability of the discovered hydrides. Dynamical stability is confirmed if there are no imaginary phonon frequencies across the Brillouin zone.

- **Convex Hull Analysis:**

  - Calculate the formation enthalpy of the ternary hydride relative to binary phases and elemental components. Check if the structure lies on the convex hull to confirm its thermodynamic stability.

4. Electron-Phonon Coupling (EPC) and Eliashberg Function

- **Electron-Phonon Coupling Calculation (e.g., Quantum ESPRESSO with EPW):**

  - **Step 1: DFT Ground State:** Calculate the electronic ground state using DFT with a high k-point mesh.
  - **Step 2: Phonon Dispersion:** Perform detailed phonon calculations using density functional perturbation theory (DFPT).
  - **Step 3: EPC Matrix Elements:** Calculate the electron-phonon coupling matrix elements for the system.
  - **Step 4: Eliashberg Function $\alpha^2F(\omega)$:** Use the EPC data to compute the Eliashberg spectral function $\alpha^2F(\omega)$, which provides insights into the strength of the electron-phonon interaction.

5. Superconducting Transition Temperature (T_c) Calculation

- **McMillan-Allen-Dynes Formula:**

  - Estimate the superconducting transition temperature T_c using the McMillan-Allen-Dynes formula:

$$T_c = \frac{\omega_{\log}}{1.2} \exp\left(\frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right)$$

where:

* $\omega_{\log}$: Logarithmic average phonon frequency
* λ: EPC constant
* μ^*: Coulomb pseudopotential (typically 0.1-0.13 for hydrides)