Supplementary Material

For

Pressure-induced high-temperature superconductivity in ternary Y-Zr-H compounds

Wendi Zhao,[1] Hao Song*,[1] Mingyang Du[2], Qiwen Jiang[2], Tiancheng Ma[2], Ming Xu[1], Defang Duan,[2,*] and Tian Cui[1,2,*]

[1]Institute of High Pressure Physics, School of Physical Science and Technology, Ningbo University, Ningbo 315211, China

[2]State Key Laboratory of Superhard Materials, College of Physics, Jilin University, Changchun 130012, China
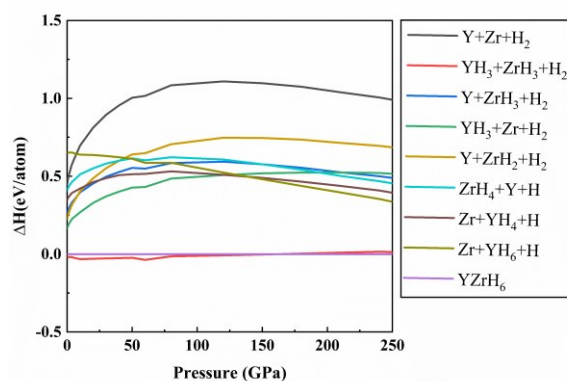
Fig.S1 The enthalpy difference curves with respect to YZrH$_6$ from divergent synthetic routes as a function of pressure.
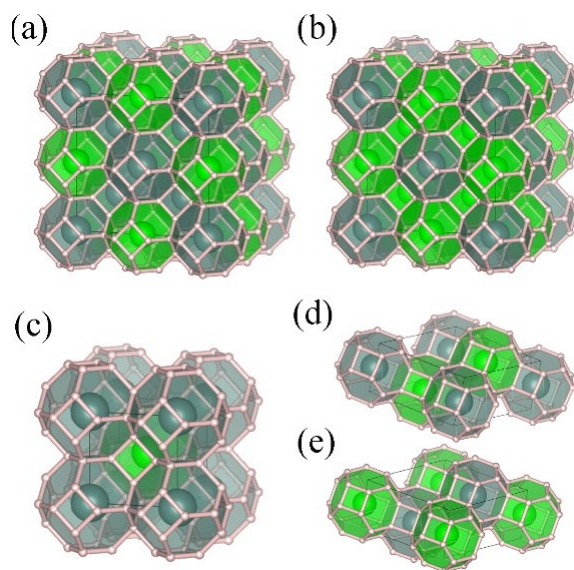
Fig.S2 Predicted crystal structure of clathrate hexahydrides at 200 GPa. (a) $Fm\bar{3}m$-Y$_3$ZrH$_{24}$, (b) $Fm\bar{3}m$-YZr$_3$H$_{24}$, (c) $Pm\bar{3}m$-YZrH$_{12}$, (d) $P\bar{3}m1$-YZr$_2$H$_{18}$ and (e) $P\bar{3}m1$-Y$_2$ZrH$_{18}$. The gray, green and pink spheres represent Y, Zr and H atoms, respectively.
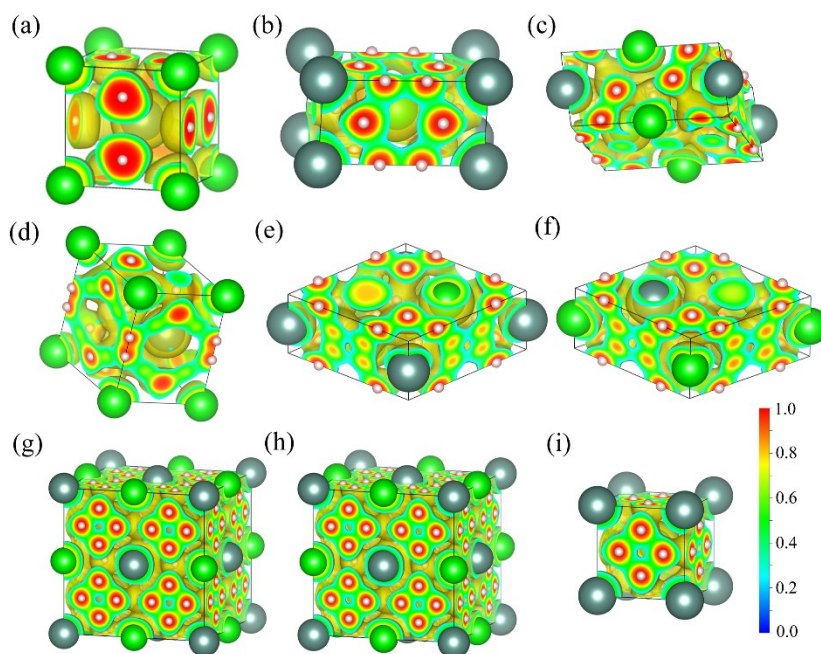

Fig.S3 The electron localization functions. (a) $Pm\bar{3}$-YZrH$_6$ at 1 atm and (b) $P4/mmm$-YZrH$_8$, (c) $P2/m$-YZr$_3$H$_{16}$ (d) $P\bar{6}m2$-YZrH$_{18}$ (e) $P\bar{3}m1$-YZr$_2$H$_{18}$ (f) $P\bar{3}m1$-Y$_2$ZrH$_{18}$ (g) $Fm\bar{3}m$-Y$_3$ZrH$_{24}$ (h) $Fm\bar{3}m$-YZr$_3$H$_{24}$ (i) $Pm\bar{3}m$-YZrH$_{12}$ at 200 GPa. Isosurface values are all set to 0.3.
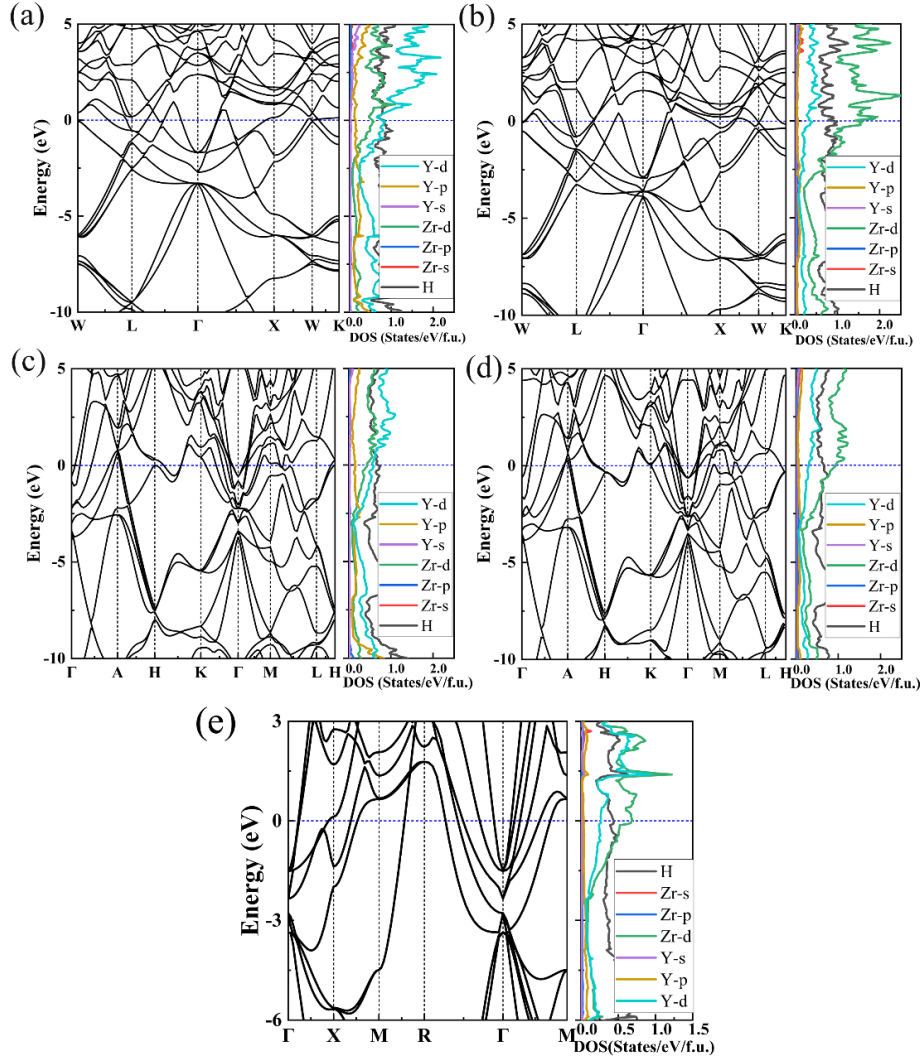
Fig. S4 Calculated electronic band structures and atom-projected density of states for (a) $Fm\overline{3}m$-Y_3ZrH_{24}, (b) $Fm\overline{3}m$-YZr_3H_{24}, (c) $P\overline{3}m1$-Y_2ZrH_{18}, (d) $P\overline{3}m1$-YZr_2H_{18} at 200 GPa, and (e) $Pm\overline{3}m$-$YZrH_{12}$ at 160 GPa. The Fermi level is set to zero.

Fig. S5 Calculated phonon dispersion curves, projected phonon density of states (PHDOS), and Eliashberg spectral function $\alpha^2F(\omega)$ together with the electron-phonon integral $\lambda(\omega)$ for (a) $Fm\bar{3}m$-Y_3ZrH_{24} (b) $Fm\bar{3}m$-YZr_3H_{24}, (c) $P\bar{3}m1$-Y_2ZrH_{18}, (d) $P\bar{3}m1$-YZr_2H_{18} at 200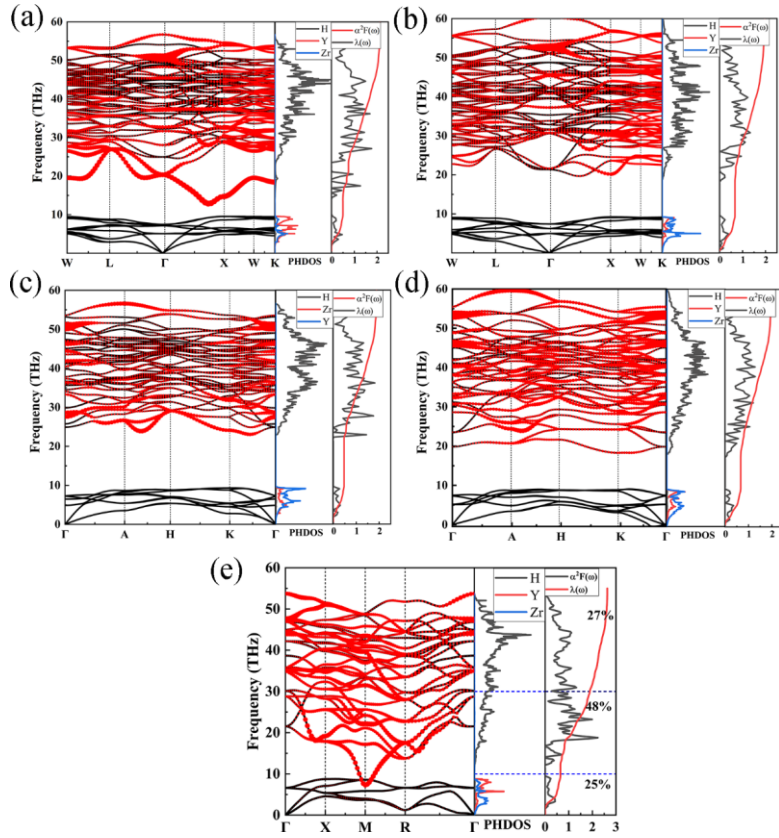 GPa and (e) $Pm\bar{3}m$ $YZrH_{12}$ at 160 GPa. The size of the red solid dots in the phonon spectra is proportional to the strength of electron-phonon coupling.

Table S1. Structural information of predicted hydrides.

Structure	Lattice parameters (Å)	Atomic coordinates				Sites
$Pm\bar{3}$		H1	0.2367	0.0000	0.5000	6f
YZrH$_6$	a=b=c=3.9768	Y1	0.5000	0.5000	0.5000	1b
1atm	α = β = γ = 90	Zr1	0.0000	0.0000	0.0000	1a
P4/mmm		H1	0.0000	0.5000	0.2588	4i
YZrH$_8$	a=b= 2.6855	H2	0.0000	0.0000	0.3730	2g
200 GPa	c=5.0677	H3	0.5000	0.5000	0.1310	2h
	α = β = γ = 90	Y1	0.0000	0.0000	0.0000	1a
		Zr1	0.5000	0.5000	0.5000	1d
	a=3.1472	H1	0.2460	0.2558	0.7449	4o
	b=3.8042	H2	0.0000	0.25814	0.0000	2i
$P2/m$	c=6.1948	H3	0.0638	0.0000	0.1928	2m
YZr$_3$H$_{16}$	α =90	H4	0.4422	0.0000	0.3181	2m
200 GPa	β = 103.34	H5	0.1913	0.5000	0.5587	2n
	γ= 90	H6	0.3095	0.5000	0.9324	2n
		Zr1	0.2525	0.5000	0.2514	2n
		H7	0.5000	0.2501	0.5000	2l
		Y1	0.5000	0.0000	0.0000	1d
		Zr2	0.0000	0.0000	0.5000	1c
	a=3.3872	H1	0.8226	0.6451	0.3019	6n
	b=3.3872	H2	0.0000	0.0000	0.4123	2g
	c=5.1842	H3	0.6667	0.3333	0.5000	1f
$P\bar{6}m2$	α =90	H4	0.9782	0.4891	0.8170	6n
YZrH$_{18}$	β = 90	H5	0.3333	0.6667	0.8908	2h
200 GPa	γ= 120	H6	0.6667	0.3333	0.0000	1e
		Y1	0.3333	0.6667	0.5000	1d
		Zr1	0.0000	0.0000	0.0000	1a
$Fm\bar{3}m$		H1	0.2504	-0.5000	-0.3741	96j
Y$_3$ZrH$_{24}$	a=b=c=6.9907	Zr1	0.5000	-0.5000	-0.5000	4b
200 GPa	α = β = γ = 90	Y1	0.2500	-0.2500	-0.2500	8c
		Y2	0.0000	0.0000	0.0000	4a
$Fm\bar{3}m$		H1	0.5000	0.2507	0.3754	96j
YZr$_3$H$_{24}$	a=b=c=6.6703	Zr1	0.2500	0.2500	0.2500	8c
200 GPa	α = β = γ = 90	Zr2	0.5000	0.5000	0.5000	4b
		Y1	0.0000	0.0000	0.0000	4a
$P\bar{3}m1$	a=4.6782	H1	0.5864	0.9196	0.6713	12j
Y$_2$ZrH$_{18}$	b=4.6782	H2	0.7534	0.7534	-0.0000	6g
200 GPa	c=2.8418	Zr1	-0.000	0.0000	0.5000	1b
	α =β = 90	Y1	0.6667	0.3333	0.8273	2d
	γ= 120					

$P\bar{3}m1$	a=4.6681	H1	0.5813	0.9148	0.6569	12j
YZr$_2$H$_{18}$	b=4.6681	H2	0.7447	0.7447	0.0000	6g
200 GPa	c=2.8215	Zr1	0.6667	0.3333	0.8491	2d
	$\alpha = \beta = 90$	Y1	0.0000	0.0000	0.5000	1b
	$\gamma = 120$					
$Pm\bar{3}m$		H1	0.5000	-0.0000	0.7525	12h
YZrH$_{12}$	a=b=c=3.4831	Y1	0.0000	0.0000	0.0000	1a
200 GPa	$\alpha = \beta = \gamma = 90$	Zr1	0.5000	0.5000	0.5000	1b

Table S2. The average charge value (e) and number of electrons transferred $\delta(e)$ for different atoms in hexahydrides of the Y-Zr-H system at 200 GPa.

Structure	Atom	Charge Value	$\delta(e)$
	Y	9.611	1.389
$Fm\bar{3}m$ Y$_3$ZrH$_{24}$	Zr	9.023	0.977
	H	1.214	-0.214
	Y	9.622	1.378
$Fm\bar{3}m$ YZr$_3$H$_{24}$	Zr	10.483	1.517
	H	1.236	-0.236
	Y	9.776	1.224
$P\bar{3}m1$ Y$_2$ZrH$_{18}$	Zr	10.592	1.408
	H	1.213	-0.214
	Y	9.792	1.208
$P\bar{3}m1$ YZr$_2$H$_{18}$	Zr	10.656	1.344
	H	1.216	-0.216
	Y	9.638	1.362
$Pm\bar{3}m$ YZrH$_{12}$	Zr	10.550	1.450
	H	1.233	-0.234

Table S3. The calculated EPC parameter λ, logarithmic average phonon frequency ω_{log} (K), electronic density of states at Fermi level $N_{\varepsilon f}$ (states/spin/eV/f.u.). T_c values were estimated by the Allen-Dynes modified McMillan equation.

Structure	Space group	Pressure (GPa)	λ	ω_{log}	$N_{\varepsilon f}$	T_c (K) 0.1	T_c (K) 0.13
YZrH$_6$	$Pm\bar{3}$	1 atm	0.72	422.94	0.69	16.30	12.55
YZrH$_8$	$P4/mmm$	200	1.05	866.54	0.47	69.88	59.88
YZr$_3$H$_{16}$	$P2/m$	200	0.73	973.59	1.00	38.59	29.87
YZrH$_{18}$	$P\bar{6}m2$	200	2.21	794.923	0.58	156.23	142.12
Y$_3$ZrH$_{24}$	$Fm\bar{3}m$	200	2.13	992.67	1.42	184.92	168.62
Y$_2$ZrH$_{18}$	$P\bar{3}m1$	200	1.88	1025.28	1.11	169.70	154.04
YZrH$_{12}$	$Pm\bar{3}m$	160	2.63	803.46	0.85	182.67	167.06
		200	2.16	920.97	0.81	174.56	159.11
		300	1.70	1087.95	0.75	162.93	147.08
YZr$_2$H$_{18}$	$P\bar{3}m1$	200	1.98	775.73	1.24	136.68	123.97
YZr$_3$H$_{24}$	$Fm\bar{3}m$	200	1.95	748.76	1.73	130.52	118.31
		250	1.70	864.58	1.65	129.81	117.02
		300	1.57	924.35	1.59	127.57	114.47