Supplementary Information For

Large-scale screening of metal hydrides for hydrogen storage from first-principles calculations based on equilibrium reaction thermodynamics

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DFT optimization of crystal structures

We updated our original database by including 147 new crystal compounds whose structures are currently available. The compounds are listed in Table S1 and the detailed structural information is shown in Table S2.

Table S1. List of 359 compounds included in our database.

One-element compounds				
Al	B	C	Ca	K
Li	Mg	Na	Sc	Si
Ti	V			
Two-element compounds				
AlB_2	AlB_{12}	Al_4C_3	Al_2Ca	Al_4Ca
AlH_3	AlLi	Al_2Li_3	Al_3Li	Al_4Li_9
$Al_{12}Mg_{17}$	$Al_{14}Mg_{13}$	$Al_{30}Mg_{23}$	AlN	AlSc
$AlSc_2$	Al_2Sc	Al_3Sc	AlTi	$AlTi_3$
Al_2Ti	Al_3Ti	AlV	AlV_3	Al_3V
$Al_{10}V$	$Al_{23}V_4$	$Al_{45}V_7$	B_4C	$B_{13}C_2$
$B_3Ca_4LiN_6$	$(B_{10}H_{13})_2$	BN	$B_{13}N_2$	C_2Ca
C_2N_2	C_3N_4	C_5N_4	$C_{12}N_6$	CaB_4
CaB_6	CaH_2	$CaLi_2$	$CaMg_2$	CaN_6
Ca_2N	Ca_3N_2	$Ca_{11}N_8$	CaSi	$CaSi_2$
Ca_2Si	Ca_5Si_3	KB_6	KC_8	K_2C_2
KH	KN_3	K_3N	KSi	K_4Si_4
K_8Si_{46}	LiB	Li_5B_4	Li_2C_2	LiH

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LiMg	LiN_3	Li_3N	LiSi	Li_2Si
Li_7Si_2	$Li_{12}Si_7$	$Li_{13}Si_4$	$Li_{15}Si_4$	MgB_2
MgB_4	MgB_7	MgC_2	Mg_2C_3	MgH_2
Mg_3N_2	MgSc	Mg_2Si	Mg_5Si_6	N_4Si_3
NaB_{15}	Na_3B_{20}	Na_2C_2	NaH	NaN_3
Na_3N	NaSi	Na_4Si_4	Na_8Si_{46}	ScB_2
ScB_{12}	ScC	Sc_2C	Sc_2C_3	Sc_3C_4
Sc_4C_3	$Sc_{15}C_{19}$	ScH_2	ScN	ScSi
$ScSi_2$	Sc_5Si_3	SiB_3	SiB_6	SiC
SiH	TiB	TiB_2	TiC	Ti_2C
Ti_8C_5	TiH	TiH_2	TiN	Ti_2N
TiSi	$TiSi_2$	Ti_5Si_3	Ti_5Si_4	TiV
VB	VB_2	V_2B_3	V_3B_2	VC
V_2C	V_6C_5	V_8C_7	VH_2	V_2H
VN	V_2N	VSi_2	V_3Si	V_5Si_3
V_6Si_5				
Three-element compounds				
$Al(BH_4)_3$	Al_5C_3N	$Al_6C_3N_2$	$Al_7C_3N_3$	$Al_8C_3N_4$
$Al_2Ca_3N_4$	Al_2CaSi_2	$Al_2Ca_3Si_2$	$AlLi_3N_2$	AlLiSi
$Al_3Li_8Si_5$	$Al_3Li_{12}Si_4$	Al_2MgC_2	$AlMg_4Si_6$	$Al_{18}Mg_3Ti_2$
$AlSc_2Si_2$	BC_2N	$C_2H_4N_4$	$C_2H_{18}N_{18}$	$C_2N_2(NH)$
$CaAlH_5$	$Ca(AlH_4)_2$	Ca_4Al_3Mg	Ca_3AlN_3	CaAlSi
CaB_2C_2	$Ca(BH_4)_2$	$CaB_{12}H_{12}$	Ca_3BN_3	$CaCN_2$
CaC_4N_6	Ca_2HN	CaLiN	$CaLiSi_2$	Ca_2LiSi_3
$CaMg_2N_2$	CaMgSi	$Ca_4N_2(CN_2)$	$Ca_{11}N_6(CN_2)_2$	Ca_2N_3V
Ca_3N_3V	$CaSiN_2$	$Ca_2Si_5N_8$	$Ca_5(Si_2N_6)$	Ca_4TiN_4
H_9CN_9	$KAlH_4$	K_3AlH_6	KBH_4	$KB_{21}H_{18}$
$K_2B_6H_6$	$K_2(B_{10}H_{10})$	$K_2B_{12}H_{12}$	KCN	KC_2N_3
KC_4N_3	K_2CN_2	$K_3C_6N_9$	K_3LiSi_4	K_7LiSi_8
$KMgH_3$	K_2MgH_4	KNH_2	$(K(NH_2))(NH_3)_2$	$KSiH_3$
$LiAlB_{14}$	$LiAlH_4$	Li_3AlH_6	LiBC	$LiB_{13}C_2$
$Li_2B_{12}C_2$	LiBH	$Li(BH_2)$	$LiBH_4$	$Li_2B_{12}H_{12}$
$Li_3(BH_6)$	Li_3BN_2	$Li_2B_{12}Si_2$	LiCN	Li_2CN_2
$LiMgH_3$	LiMgN	Li_2MgSi	$Li_{12}Mg_3Si_4$	$LiNH_2$
Li_2NH	Li_4NH	LiN_3Si_2	Li_5N_3Si	Li_7N_4V
$LiNa_2N$	$LiNa_5N_2$	$Li_2Na_4N_2$	Li_2NaN	$Li_3Na_3N_2$
$Li_4Na_2N_2$	Li_5NaN_2	Li_3NaSi_6	Li_3ScN_2	$MgAlH_5$
$Mg(AlH_4)_2$	MgAlSi	$MgAl_2Si_2$	MgB_2C_2	$MgB_{12}C_2$

$Mg_2B_{24}C$	$Mg(BH_4)_2$	$MgB_{12}H_{12}$	MgB_9N	Mg_3BN_3
$MgB_{12}Si_2$	MgC_4N_6	Mg(NCN)	$Mg(NH_2)_2$	$MgSiN_2$
Mg_7TiH_{16}	N_2BH_7	$N_2B_{10}H_{18}$	$N_3B_3H_6$	$N_3B_3H_{12}$
$N_4B_9H_{11}$	$N_4B_{10}H_8$	$N_4B_{10}H_{22}$	NH_3BH_3	$(NH_4)_2B_{12}H_{12}$
$(NH_2)CN$	NH_4HCN_2	$N(SiH_3)_3$	$NaAlH_4$	Na_3AlH_6
$Na_5Al_3H_{14}$	NaAlSi	$NaAlSi_4$	$NaBH_4$	$Na_2(B_{10}H_{10})$
$Na_3(BN_2)$	NaCN	NaC_4N_3	Na_2CN_2	$Na_3C_6N_9$
$NaMgH_3$	NaN_3C_2	$NaNH_2$	$ScAl_3C_3$	Sc_2AlC
Sc_3AlC	ScB_2C	ScB_2C_2	Sc_2BC_2	$Sc_2V_3Si_4$
SiCN	SiC_2N_4	Si_2CN_4	Ti_2AlC	Ti_3AlC
Ti_3AlC_2	Ti_2AlN	Ti_3AlN	Ti_4AlN_3	Ti_6Si_2B
Ti_3SiC_2	$V_{12}Al_3C_8$	V_5SiB_2		
Four-element compounds				
$AlNC_3H_{10}$	BCH_5N_2	$B_{10}C_6H_{30}N_2$	$B_{20}C_3H_{30}N_2$	BC_4KN_4
$CH_3NH_2BH_3$	Ca_2N_2BH	$Ca(NH_2BH_3)_2$	$KAl(NH_2)_4$	$K_5C_2HN_4$
$KCaN_3H_6$	$K(HCN_2)$	K_2LiAlH_6	$KLi_3(NH_2)_4$	$KLi_7N_8H_{16}$
$K_2Li(NH_2)_3$	$K_2Mg(NH_2)_4$	K_2NaAlH_6	$K_2Na(NH_2)_3$	$K_3Si_6N_{11}H_6$
$LiAlMg_{10}H_{24}$	$LiAl(NH_2)_4$	$Li(B(CN)_4)$	$Li_4BN_3H_{10}$	$Li_2Ca(NH)_2$
$LiK(BH_4)_2$	$Li_2Mg(NH)_2$	$Li(NH_2BH_3)$	$(Li(NH_3)_4)_2(B_6H_6)(NH_3)_2$	$LiNa_2AlH_6$
$LiNa_2(NH_2)_3$	$Li_3Na(NH_2)_4$	$LiSc(BH_4)_4$	$Mg(BH_4)_2(NH_3)_2$	$(NH_4)B(CN)_4$
$NaAl(NH_2)_4$	$NaB(CN)_4$	NaN_2CH	$Si_2C_7H_{18}N_2$	$VC_8H_{24}N_4$
Five-element compounds				
$LiAlC_4H_{16}N_4$	$LiSi_3C_9H_{27}N_2$	$Si_2B_2C_{12}H_{37}N_5$		

Table S2. Comparison of the experimental and the DFT calculated structural parameters for the 147 compounds listed in Table 1, with all distances in Å and angles in degrees.

Compound	Space group	Structural parameters (Å, degree)	
		Experimental	Calculated
AlB_{12}	$P4_12_12$	*a* = 10.17 *c* = 14.28	*a* = 11.41 *c* = 13.86
Al_4C_3	R-3mH	*a* = 3.335 *c* = 24.967 γ = 120.0	*a* = 3.349 *c* = 25.109 γ = 120.0
Al_4Ca	I4/mmm	*a* = 4.354 *c* = 11.18	*a* = 4.368 *c* = 11.19
$Al_{14}Mg_{13}$	Im-3m	*a* = 10.44	*a* = 10.2
$Al_{30}Mg_{23}$	R-3H	*a* = 12.825	*a* = 12.787
$Al_{23}V_4$	P63/mmc	*a* = 7.693 *c* = 17.04 γ = 120	*a* = 7.681 *c* = 17.04 γ = 120
$Al_{45}V_7$	C2/m	*a* = 25.604 *b* = 7.621 *c* = 11.081 β = 128.92	*a* = 25.655 *b* = 7.608 *c* = 11.086 β = 128.88
B_4C	R-3mH	*a* = 5.60 *c* = 12.12 γ = 120	*a* = 7.40 *c* = 8.77 γ = 120
$B_{13}C_2$	R-3mH	*a* = 5.633 *c* = 12.164 γ = 120	*a* = 5.656 *c* = 12.12 γ = 120
$(B_{10}H_{13})_2$	Pbca	*a* = 10.66 *b* = 10.55 *c* = 14.56	*a* = 10.83 *b* = 10.69 *c* = 14.78
$B_{13}N_2$	R-3m	*a* = 5.45 *c* = 12.26	*a* = 5.49 *c* = 12.41
C_2Ca	C2/m	*a* = 7.208 *b* = 3.828 *c* = 7.368 β = 107.193	*a* = 7.170 *b* = 3.833 *c* = 7.416 β = 106.961
C_2N_2	Pcab	*a* = 6.31 *b* = 7.08 *c* = 6.19	*a* = 6.72 *b* = 6.38 *c* = 6.06

C_3N_4	P-6m2	*a* = 4.742 *c* = 6.720 γ = 120	*a* = 4.766 *c* = 6.400 γ = 120
C_5N_4	R3cH	*a* = 9.062 *c* = 11.625 γ = 120	*a* = 8.912 *c* = 11.416 γ = 120
$C_{12}N_6$	Pa-3	*a* = 10.781	*a* = 10.746
CaB_4	P4/mbm	*a* = 7.10 *c* = 4.14	*a* = 7.17 *c* = 4.1
Ca_2Si	Pnma	*a* = 7.69 *b* = 4.82 *c* = 9.05	*a* = 7.65 *b* = 4.83 *c* = 9.09
KC_8	Fddds	*a* = 4.92 *b* = 8.51 *c* = 21.40	*a* = 4.97 *b* = 8.61 *c* = 21.37
KSi	P-43n	*a* = 12.62	*a* = 12.72
K_8Si_{46}	Pm-3n	*a* = 10.30	*a* = 10.36
LiB	PNMA	*a* = 6.40 *b* = 3.00 *c* = 5.6	*a* = 6.225 *b* = 3.073 *c* = 5.589
LiMg	Im-3m	*a* = 3.484	*a* = 3.434
$Li_{12}Si_7$	Pnma	*a* = 8.6 *b* = 19.755 *c* = 14.336	*a* = 8.54 *b* = 19.631 *c* = 14.32
$Li_{13}Si_4$	Pbam	*a* = 7.99 *b* = 15.21 *c* = 4.43	*a* = 7.902 *b* = 15.022 *c* = 4.432
$Li_{15}Si_4$	I-43d	*a* = 10.69	*a* = 10.6
NaB_{15}	Imam	*a* = 5.847 *b* = 8.415 *c* = 10.298	*a* = 5.848 *b* = 8.426 *c* = 10.295
Na_3B_{20}	Cmmm	*a* = 18.695 *b* = 5.701 *c* = 4.151	*a* = 18.636 *b* = 5.693 *c* = 4.158
Na_4Si_4	C2/c	*a* = 12.153 *b* = 6.545 *c* = 11.132 β = 118.9	*a* = 12.151 *b* = 6.563 *c* = 11.109 β = 90.0

Na_8Si_{46}	Pm-3n	a = 10.19	a = 12.31
$Sc_{15}C_{19}$	P-42_1c	a = 7.5 c = 15.0	a = 7.51 c = 14.612
SiB_3	Imma	a = 8.392 b = 12.568 c = 6.213	a = 8.381 b = 12.588 c = 6.223
SiC	F-43m	a = 4.36	a = 4.38
TiV	Im-3m	a = 3.159	a = 3.103
V_2N	P-31m	a = 4.917 c = 4.568 γ = 120.0	a = 4.899 c = 4.522 γ = 120.0
V_5Si_3	I4/mcm	a = 9.429 c = 4.756	a = 9.393 c = 4.715
$Al(BH_4)_3$	Pna2_1	a = 18.02 b = 6.14 c = 6.20	a = 17.99 b = 6.12 c = 6.20
Al_2MgC_2	P-3m	a = 3.377 c = 5.817 γ = 120.0	a = 3.385 c = 5.82 γ = 120.0
$Al_{18}Mg_3Ti_2$	Fd-3ms	a = 14.788	a = 14.775
BC_2N	Pmm2	a = 2.528 b = 2.502 c = 3.587	a = 2.56 b = 2.533 c = 3.637
$C_2H_4N_4$	P21/c	a = 3.791 b = 12.412 c = 9.113 β = 91.49	a = 3.651 b = 12.012 c = 9.189 β = 91.32
$C_2H_{18}N_{18}$	P-1	a = 4.621 b = 8.585 c = 9.271 α = 108.49 β = 95.29 γ = 102.99	a = 4.501 b = 8.415 c = 9.174 α = 109.353 β = 95.17 γ = 103.09
$C_2N_2(NH)$	Cmc2_1	a = 7.57 b = 4.44 c = 4.0	a = 7.63 b = 4.48 c = 4.04
Ca_4Al_3Mg	Pbcm	a = 6.179 b = 24.211	a = 6.191 b = 24.248

		c = 5.886	c = 5.905
CaAlSi	P-6m2	a = 4.2 c = 4.4	a = 4.21 c = 4.41
$Ca(BH_4)_2$	F2dd	a = 8.78 b = 13.02 c = 7.41	a = 8.75 b = 12.94 c = 7.37
$CaB_{12}H_{12}$	C2/c	a = 14.328 b = 7.164 c = 11.017 α = β = 89.84 γ = 122.07	a = 14.307 b = 7.152 c = 11.001 α = β = 89.85 γ = 122.07
CaC_4N_6	C2/c	a = 12.446 b = 6.08 c = 7.898 β = 145.2	a = 12.855 b = 6.261 c = 7.674 β = 149.86
$Ca_4N_2(CN_2)$	Pnma	a = 11.44 b = 3.58 c = 13.84	a = 11.51 b = 3.58 c = 13.92
$Ca_{11}N_6(CN_2)_2$	P42/MNM	a = 14.523 c = 3.608	a = 14.551 c = 3.6221
$CaSiN_2$	Pbca	a = 5.123 b = 10.207 c = 14.823	a = 5.163 b = 10.279 c = 14.933
$Ca_2Si_5N_8$	Pbca	a = 10.584 b = 9.652 c = 13.663	a = 10.616 b = 9.675 c = 13.669
$Ca_5(Si_2N_6)$	C12/C1	a = 9.836 b = 6.0519 c = 12.757 β = 100.20	a = 9.899 b = 6.094 c = 14.736 β = 121.155
Ca_4TiN_4	P-1	a = 5.98 b = 6.01 c = 8.99 α = 71.57 β = 79.47 γ = 68.26	a = 6.01 b = 6.04 c = 9.02 α = 71.62 β = 79.32 γ = 68.07
H_9CN_9	$P2_1/c$	a = 6.679 b = 7.722	a = 6.555 b = 7.546

		c = 13.143 β = 95.44	*c* = 12.901 β = 95.71
KBH_4	Fm-3m	*a* = 6.71	*a* = 6.69
KBH_4	$P4_2/nmc$	*a* = 4.68 *c* = 6.57	*a* = 4.71 *c* = 6.61
$KB_{21}H_{18}$	C2	*a* = 12.49 *b* = 7.11 *c* = 16.94 β = 93.81	*a* = 12.71 *b* = 7.22 *c* = 17.04 β = 94.1
$K_2B_6H_6$	Fm-3m	*a* = 8.839	*a* = 8.897
$K_2(B_{10}H_{10})$	P121/n1	*a* = 12.855 *b* = 11.178 *c* = 6.823 β = 93.357	*a* = 11.993 *b* = 9.748 *c* = 9.028 β = 91.93
$K_2B_{12}H_{12}$	Fm-3	*a* = 10.629	*a* = 10.639
KC_4N_3	P-1	*a* = 8.665 *b* = 8.873 *c* = 3.89 α = 86.7 β = 90.1 γ = 105	*a* = 8.827 *b* = 9.296 *c* = 4.009 α = 83.8 β = 90.9 γ = 104.3
KNH_2	$P2_1/m$	*a* = 4.586 *b* = 3.904 *c* = 6.223 β = 95.8	*a* = 4.458 *b* = 3.745 *c* = 6.111 β = 94.958
$(K(NH_2))(NH_3)_2$	$C222_1$	*a* = 6.839 *b* = 9.953 *c* = 6.590	*a* = 6.834 *b* = 9.681 *c* = 6.5711
$LiAlB_{14}$	Imam	*a* = 5.847 *b* = 8.143 *c* = 10.354	*a* = 5.852 *b* = 8.142 *c* = 10.353
$LiB_{13}C_2$	Imma	*a* = 5.668 *b* = 10.820 *c* = 8.040	*a* = 5.842 *b* = 9.661 *c* = 8.923
$Li_2B_{12}C_2$	AMM2	*a* = 4.706 *b* = 5.318 *c* = 5.318 α = 115.798	*a* = 4.663 *b* = 5.553 *c* = 5.553 α = 100.47

LiBH	PNMA	*a* = 6.2 *b* = 3.0 *c* = 6.3	*a* = 5.646 *b* = 3.076 *c* = 6.505
$Li(BH_2)$	PNMA	*a* = 8.1 *b* = 3.0 *c* = 5.9	*a* = 8.322 *b* = 3.037 *c* = 5.485
$Li_2B_{12}H_{12}$	Pa3	*a* = 9.577	*a* = 9.580
$Li_3(BH_6)$	R3-H	*a* = 5.182 α = β = γ = 91.11	*a* = 5.356 α = β = γ = 94.16
$Li_2B_{12}Si_2$	Cmce	*a* = 6.106 *b* = 10.979 *c* = 8.405	*a* = 6.118 *b* = 11.012 *c* = 8.43
$LiMgH_3$	R3c	*a* = 4.96 *c* = 13.34	*a* = 4.94 *c* = 13.24
Li_2MgSi	Fm-3m	*a* = 12.83	*a* = 12.748
LiN_3Si_2	$Cmc2_1$	*a* = 9.222 *b* = 5.296 *c* = 4.78	*a* = 9.277 *b* = 5.329 *c* = 4.812
$LiNa_2N$	P6/mmm	*a* = 4 *c* = 4.2 γ = 120	*a* = 4.37 *c* = 3.838 γ = 120
$LiNa_5N_2$	C_2	*a* = 6.731 *b* = 5.944 *c* = 6.383 β = 91.18	*a* = 6.735 *b* = 5.949 *c* = 6.389 β = 91.15
$Li_2Na_4N_2$	P4/nmm	*a* = 3.895 *c* = 6.114	*a* = 4.066 *c* = 6.099
Li_2NaN	P6/nmm	*a* = 3.65 *c* = 4.6 γ = 120	*a* = 3.62 *c* = 4.716 γ = 120
$Li_3Na_3N_2$	Pm	*a* = 3.854 *b* = 3.676 *c* = 6.32 β = 90.31	*a* = 3.853 *b* = 4.208 *c* = 7.272 β = 89.85
$Li_4Na_2N_2$	Fm-3m	*a* = 5.265	*a* = 5.404
Li_5NaN_2	P4/mmm	*a* = 3.965 *c* = 5.504	*a* = 3.705 *c* = 5.186
Li_3NaSi_6	Pnma	*a* = 17.972	*a* = 18.021

		b = 3.788 *c* = 10.299	*b* = 3.804 *c* = 10.331
$MgAl_2Si_2$	P-3m1	*a* = 4.05 *c* = 6.74	*a* = 4.08 *c* = 6.69
$MgB_{12}C_2$	C2/c	*a* = 7.27 *b* = 8.78 *c* = 7.28 β = 105.33	*a* = 7.26 *b* = 8.77 *c* = 7.25 β = 105.32
$Mg_2B_{24}C$	P-4n2	*a* = 8.94 *c* = 5.07	*a* = 8.96 *c* = 5.09
$MgB_{12}H_{12}$	C2/m	*a* = 11.689 *b* = 8.712 *c* = 6.907 β = 122.47	*a* = 11.687 *b* = 8.711 *c* = 6.905 β = 122.5
$MgB_{12}Si_2$	Pnma	*a* = 10.98 *b* = 6.11 *c* = 8.36	*a* = 11.03 *b* = 6.13 *c* = 8.39
MgC_4N_6	Pnnm	*a* = 6.171 *b* = 7.17 *c* = 7.404	*a* = 6.443 *b* = 7.289 *c* = 7.429
Mg_7TiH_{16}	Fm3m	*a* = 9.564	*a* = 9.341
N_2BH_7	Pbcn	*a* = 9.53 *b* = 5.12 *c* = 13.01	*a* = 9.768 *b* = 5.237 *c* = 12.672
$N_2B_{10}H_{18}$	Pnma	*a* = 18.096 *b* = 7.373 *c* = 7.223	*a* = 18.237 *b* = 7.528 *c* = 7.284
$N_3B_3H_6$	$P4_32_12$	*a* = 5.428 *c* = 16.279	*a* = 5.63 *c* = 17.223
$N_3B_3H_{12}$	Pbcm	*a* = 4.403 *b* = 12.21 *c* = 11.227	*a* = 4.442 *b* = 12.382 *c* = 11.272
$N_4B_9H_{11}$	$P2_1/c$	*a* = 8.318 *b* = 5.951 *c* = 19.265 β = 95.3	*a* = 8.611 *b* = 6.263 *c* = 20.044 β = 94.6
$N_4B_{10}H_8$	C2/c	*a* = 11.411 *b* = 6.658	*a* = 11.945 *b* = 7.373

		c = 13.058	*c* = 15.268 α = 91.09
$N_4B_{10}H_{22}$	C2/c	*a* = 7.7 *b* = 7.7 *c* = 9.772 α = 83.872 β = 83.872 γ = 82.307	*a* = 7.813 *b* = 7.229 *c* = 9.473 α = 77.29 β = 76.99 γ = 82.3
NH_3BH_3	Pmn21	*a* = 5.395 *b* = 4.887 *c* = 4.986	*a* = 5.356 *b* = 4.796 *c* = 4.921
$(NH_4)_2B_{12}H_{12}$	Fm-3	*a* = 10.88	*a* = 10.79
$(NH_2)CN$	Pbca	*a* = 6.856 *b* = 6.628 *c* = 9.147	*a* = 6.726 *b* = 6.597 *c* = 8.916
NH_4HCN_2	$P2_12_12$	*a* = 6.44 *b* = 6.58 *c* = 7.4	*a* = 6.38 *b* = 6.5 *c* = 7.3
$Na_5Al_3H_{14}$	P4/mnc	*a* = 6.769 *c* = 10.289	*a* = 6.7 *c* = 10.2
$NaBH_4$	Fm-3m	*a* = 6.15	*a* = 6.02
$NaBH_4$	$P\text{-}42_1c$	*a* = 4.35 *c* = 5.86	*a* = 4.31 *c* = 5.82
$Na_2(B_{10}H_{10})$	P121/n1	*a* = 10.283 *b* = 13.022 *c* = 6.673 β = 93.754	*a* = 9.846 *b* = 12.153 *c* = 8.104 β = 93.074
$Na_3(BN_2)$	$P2_1/c$	*a* = 5.717 *b* = 7.931 *c* = 7.883 β = 111.32	*a* = 5.737 *b* = 7.966 *c* = 7.9 β = 111.29
$Na_3C_6N_9$	$P2_1/c$	*a* = 11.048 *b* = 23.381 *c* = 3.516 β = 97.913	*a* = 11.397 *b* = 24.101 *c* = 3.937 β = 97.913
Sc_2AlC	P63/MMC	*a* = 3.228 *c* = 14.873 γ = 120.0	*a* = 3.285 *c* = 15.043 γ = 120.0

Ti_2AlC	$P6_3/mmc$	a = 2.97 c = 13.22	a = 3.07 c = 13.71
Ti_6Si_2B	P-62m	a = 6.802 c = 3.338 γ = 120	a = 6.777 c = 3.312 γ = 120
$V_{12}Al_3C_8$	P63/MCM	a = 5.088 c = 22.983 γ = 120	a = 5.065 c = 22.638 γ = 120
V_5SiB_2	I4/mcm	a = 5.81 c = 10.79	a = 5.774 c = 10.762
$AlNC_3H_{10}$	$P2_1/c$	a = 5.428 b = 9.908 c = 9.963 β = 99.254	a = 5.379 b = 11.302 c = 10.271 β = 99.2
BCH_5N_2	$Pna2_1$	a = 7.973 b = 6.445 c = 6.976	a = 7.986 b = 6.515 c = 7.103
$B_{10}C_6H_{30}N_2$	$P2_1/c$	a = 8.369 b = 16.663 c = 11.989 β = 100.34	a = 8.586 b = 17.002 c = 12.249 β = 100.67
$B_{20}C_3H_{30}N_2$	$P2_12_12_1$	a = 10.334 b = 10.873 c = 17.524	a = 10.449 b = 11.199 c = 17.78
BC_4KN_4	$I4_1/a$	a = 6.976 c = 14.21	a = 7.151 c = 14.563
$CH_3NH_2BH_3$	Pnma	a = 11.10 b = 6.58 c = 4.92	a = 11.07 b = 6.35 c = 4.88
$Ca(NH_2BH_3)_2$	C121	a = 9.10 b = 4.37 c = 6.44 β = 93.19	a = 9.12 b = 4.29 c = 6.34 β = 93.1
$KAl(NH_2)_4$	$C222_1$	a = 10 b = 5.8 c = 10.14	a = 10.2 b = 5.82 c = 10.142
$K_5C_2HN_4$	P4/ncc	a = 9.095 c = 11.029	a = 9.225 c = 11.202

$KCaN_3H_6$	$P2_1/c$	a = 6.767 b = 11.68 c = 6.624 β = 106.7	a = 6.797 b = 11.834 c = 6.797 β = 106.82
$K(HCN_2)$	$P2_12_12_1$	a = 7.087 b = 9.09 c = 9.014	a = 7.229 b = 9.172 c = 9.158
K_2LiAlH_6	R-3m	a = 5.62 c = 27.4	a = 5.62 c = 27.31
$KLi_3(NH_2)_4$	$I4_1/amd$	a = 7.238 c = 23.956	a = 8.208 c = 23.699
$KLi_7N_8H_{16}$	$I4_1/a$	a = 7.18 c = 44.39	a = 7.678 c = 46.545
$K_2Li(NH_2)_3$	P42/m	a = 6.872 c = 11.706	a = 6.866 c = 11.726
$K_2Mg(NH_2)_4$	$P2_1/c$	a = 7.455 b = 7.024 c = 13.545 β = 105.6	a = 7.255 b = 7.255 c = 13.626 β = 105.25
K_2NaAlH_6	$P2_1/c$	a = 5.733 b = 5.754 c = 8.128 β = 89.97	a = 5.743 b = 5.7492 c = 8.0934 β = 89.99
$K_2Na(NH_2)_3$	P42/m	a = 7.351 c = 13.129	a = 7.514 c = 13.314
$K_3Si_6N_{11}H_6$	$P4_332$	a = 10.789	a = 10.797
$LiAlMg_{10}H_{24}$	P121	a = 8.989 b = 8.985 c = 4.485 β = 89.655	a = 8.915 b = 8.942 c = 4.449 β = 89.65
$Li(B(CN)_4)$	P43m	a = 7.849 α = β = γ = 60.0	a = 7.7822 α = β = γ = 60.0
$Li_2Ca(NH)_2$	P-3m1	a = 3.57 c = 5.95	a = 3.58 c = 5.84
$LiK(BH_4)_2$	Pnma	a = 7.91 b = 4.49 c = 13.84	a = 7.78 b = 4.43 c = 13.72
$Li(NH_2BH_3)$	Pbca	a = 7.11	a = 6.92

		b = 13.93 *c* = 5.15	*b* = 13.52 *c* = 5.07
$(Li(NH_3)_4)_2(B_6H_6)(NH_3)_2$	P21/c	*a* = 7.483 *b* = 11.871 *c* = 10.605 β = 95.371	*a* = 7.397 *b* = 11.649 *c* = 10.449 β = 95.21
$LiNa_2AlH_6$	P21/c	*a* = 5.165 *b* = 5.251 *c* = 7.339	*a* = 4.777 *b* = 4.715 *c* = 6.613
$LiNa_2(NH_2)_3$	$P4_2/m$	*a* = 6.28 *c* = 11.15	*a* = 6.17 *c* = 10.90
$LiSc(BH_4)_4$	P-42c	*a* = 6.08 *c* = 12.03	*a* = 6.45 *c* = 11.95
$Mg(BH_4)_2(NH_3)_2$	Pbca	*a* = 17.49 *b* = 9.41 *c* = 8.73	*a* = 17.73 *b* = 9.35 *c* = 8.68
$(NH_4)B(CN)_4$	I41/a	*a* = 7.132 *c* = 14.745	*a* = 7.453 *c* = 14.617
$NaAl(NH_2)_4$	$P2_1/c$	*a* = 7.328 *b* = 6.047 *c* = 13.151 β = 94.04	*a* = 6.565 *b* = 6.353 *c* = 15.362 β = 94.3
$NaB(CN)_4$	Fd-3mZ	*a* = 11.68	*a* = 11.874
$Si_2C_7H_{18}N_2$	$P2_1/c$	*a* = 9.71 *b* = 11.11 *c* = 11.88 β = 102.3	*a* = 10.143 *b* = 11.599 *c* = 12.422 β = 103.1
$VC_8H_{24}N_4$	P-1	*a* = 8.29 *b* = 12.016 *c* = 13.835 α = 75.662 β = 79.404 γ = 84.966	*a* = 8.637 *b* = 12.503 *c* = 14.479 α = 75.89 β = 79.47 γ = 85.3
$LiAlC_4H_{16}N_4$	$I4_1$	*a* = 14 *c* = 9.275	*a* = 14.128 *c* = 9.571
$LiSi_3C_9H_{27}N_2$	P-1	*a* = 8.776 *b* = 9.579 *c* = 21.949	*a* = 9.077 *b* = 9.875 *c* = 22.544

		α = 100.84 β = 92.18 γ = 115.67	α = 101.16 β = 91.95 γ = 115
$Si_2B_2C_{12}H_{37}N_5$	$P2_1/c$	*a* = 15.785 *b* = 11.966 *c* = 11.804 β = 102.19	*a* = 16.2 *b* = 12.212 *c* = 12.205 β = 102.3

Table S3: Multi-step reactions in which individual reactions are independent (not linked via intermediate compounds). ΔG_{max} - ΔG_{min} is the difference between ΔG for the final step and the first step in a multi-step reaction. The $T\Delta S_{conf}$ term is given only for compounds known to have partial occupancy.

Class I: (reactions having (ΔG_{max}- ΔG_{min}) ≤ 10 kJ/mol H_2)				
No.	**Reaction**	**wt.%**	**ΔU_0 ($T\Delta S_{conf}$) (kJ/mol H_2)**	**ΔG_{max}- ΔG_{min} (kJ/mol H_2)**
1	Entire reaction $6LiBH_4+15Mg(BH_4)_2+MgH_2+8Si \rightarrow 8Mg_2Si+3Li_2B_{12}H_{12}+55H_2$	9.31		1.6
	1^{st} step $MgH_2+0.5Si \rightarrow 0.5Mg_2Si+H_2$	0.169	37.6	
	2^{nd} step $6LiBH_4+15Mg(BH_4)_2+7.5Si \rightarrow 7.5Mg_2Si+3Li_2B_{12}H_{12}+54H_2$	9.136	39.2	
2	Entire reaction $18.33Si+40Mg(BH_4)_2+4Ca(BH_4)_2 \rightarrow 18.33Mg_2Si+3.33MgB_{12}H_{12}$ $+4CaB_{12}H_{12}+132H_2$	9.01		2.4
	1^{st} step $10Si+20Mg(BH_4)_2+4Ca(BH_4)_2 \rightarrow 10Mg_2Si+4CaB_{12}H_{12}+72H_2$	4.914	41.2	
	2^{nd} step $8.33Si+20Mg(BH_4)_2 \rightarrow 8.33Mg_2Si+3.33MgB_{12}H_{12}+60H_2$	4.095	43.6	
3	Entire reaction $5MgH_2+10Si+15Mg(BH_4)_2+6KBH_4 \rightarrow 10Mg_2Si+3K_2B_{12}H_{12}+59H_2$	7.69		2.6
	1^{st} step $5MgH_2+2.5Si \rightarrow 2.5Mg_2Si+5H_2$	0.652	37.6	
	2^{nd} step $7.5Si+15Mg(BH_4)_2+6KBH_4 \rightarrow 7.5Mg_2Si+3K_2B_{12}H_{12}+54H_2$	7.042	37.3 (-2.87)	
4	Entire reaction $35Si+80Mg(BH_4)_2+8KBH_4 \rightarrow 35Mg_2Si+10MgB_{12}H_{12}+4K_2B_{12}H_{12}+252H_2$	8.86		3.5
	1^{st} step $10Si+20Mg(BH_4)_2+8KBH_4 \rightarrow 10Mg_2Si+4K_2B_{12}H_{12}+72H_2$	2.532	37.3 (-2.9)	
	2^{nd} step $25Si+60Mg(BH_4)_2 \rightarrow 25Mg_2Si+10MgB_{12}H_{12}+180H_2$	6.329	43.6	

5	Entire reaction $16LiBH_4+28.33Si+60Mg(BH_4)_2 \rightarrow 28.33Mg_2Si+8Li_2B_{12}H_{12}+3.33MgB_{12}H_{12}+204H_2$	9.38		4.5
	1^{st} step $16LiBH_4+20Si+40Mg(BH_4)_2 \rightarrow 20Mg_2Si+8Li_2B_{12}H_{12}+144H_2$	6.622	39.2	
	2^{nd} step $8.33Si+20Mg(BH_4)_2 \rightarrow 8.33Mg_2Si+3.33MgB_{12}H_{12}+60H_2$	2.759	43.62	
6	Entire reaction $25MgH_2+NaMgH_3+13Mg(NH_2)_2 \rightarrow NaH+13Mg_3N_2+52H_2$	7.28		5.3
	1^{st} step $25MgH_2+12.5Mg(NH_2)_2 \rightarrow 12.5Mg_3N_2+50H_2$	6.996	26	
	2^{nd} step $NaMgH_3+0.5Mg(NH_2)_2 \rightarrow NaH+0.5Mg_3N_2+2H_2$	0.28	31.3	
7	Entire reaction $4MgH_2+47Si+108Mg(BH_4)_2 \rightarrow 47Mg_2Si+18MgB_{12}H_{12}+328H_2$	9.11		6.1
	1^{st} step $4MgH_2+2Si \rightarrow 2Mg_2Si+4H_2$	0.111	37.6	
	2^{nd} step $45Si+108Mg(BH_4)_2 \rightarrow 45Mg_2Si+18MgB_{12}H_{12}+324H_2$	9.002	43.6	
8	Entire reaction $2LiBH_4+MgH_2+7Mg(BH_4)_2+3.83Si \rightarrow 3.83Mg_2Si+Li_2B_{12}H_{12}+0.33MgB_{12}H_{12}+25H_2$	9.07		6.1
	1^{st} step $MgH_2+0.5Si \rightarrow 0.5Mg_2Si+H_2$	0.363	37.6	
	2^{nd} step $2LiBH_4+5Mg(BH_4)_2+2.5Si \rightarrow 2.5Mg_2Si+Li_2B_{12}H_{12}+18H_2$	6.533	39.2	
	3^{rd} step $2Mg(BH_4)_2+0.83Si \rightarrow 0.83Mg_2Si+0.33MgB_{12}H_{12}+6H_2$	2.178	43.6	
9	Entire reaction $MgH_2+6Mg(BH_4)_2+Ca(BH_4)_2+3.42Si \rightarrow 3.42Mg_2Si+0.17MgB_{12}H_{12}+CaB_{12}H_{12}+22H_2$	8.6		6.1
	1^{st} step $MgH_2+0.5Si \rightarrow 0.5Mg_2Si+H_2$	0.391	37.6	

	2nd step $5Mg(BH_4)_2+Ca(BH_4)_2+2.5Si \rightarrow 2.5Mg_2Si+CaB_{12}H_{12}+18H_2$	7.033	41.2	
	3rd step $Mg(BH_4)_2+0.42Si \rightarrow 0.42Mg_2Si+0.17MgB_{12}H_{12}+3H_2$	1.172	43.62	
10	Entire reaction $46LiBH_4+10Mg(BH_4)_2+5Si \rightarrow 35LiH+5Mg_2Si+5.5Li_2B_{12}H_{12}+81.5H_2$	9.77		8
	1st step $4LiBH_4+10Mg(BH_4)_2+5Si \rightarrow 5Mg_2Si+2Li_2B_{12}H_{12}+36H_2$	4.314	39.2	
	2nd step $42LiBH_4 \rightarrow 35LiH+3.5Li_2B_{12}H_{12}+45.5H_2$	5.452	47.2	
11	Entire reaction $12LiBH_4+MgH_2+Mg(BH_4)_2+Si \rightarrow 9.67LiH+Mg_2Si+1.17Li_2B_{12}H_{12}+17.17H_2$	9.36		9.6
	1st step $MgH_2+0.5Si \rightarrow 0.5Mg_2Si+H_2$	0.545	37.6	
	2nd step $0.4LiBH_4+Mg(BH_4)_2+0.5Si \rightarrow 0.5Mg_2Si+0.2Li_2B_{12}H_{12}+3.6H_2$	1.963	39.2	
	3rd step $11.6LiBH_4 \rightarrow 9.67LiH+0.97Li_2B_{12}H_{12}+12.57H_2$	6.851	47.16	
12	Entire reaction $10LiBH_4+2MgH_2+Si \rightarrow 8.33LiH+Mg_2Si+0.83Li_2B_{12}H_{12}+12.83H_2$	8.67		9.6
	1st step $2MgH_2+Si \rightarrow Mg_2Si+2H_2$	1.35	37.6	
	2nd step $10LiBH_4 \rightarrow 8.33LiH+0.83Li_2B_{12}H_{12}+10.83H_2$	7.315	47.2	
Class II: (reactions having 10 kJ/mol $H_2 \leq (\Delta G_{max}- \Delta G_{min}) \leq 20$ kJ/mol H_2)				
1	Entire reaction $33MgH_2+17Mg(NH_2)_2+2KMgH_3 \rightarrow 17Mg_3N_2+K_2MgH_4+68H_2$	7		12.6
	1st step $33MgH_2+16.5Mg(NH_2)_2 \rightarrow 16.5Mg_3N_2+66H_2$	6.791	26	
	2nd step $0.5Mg(NH_2)_2+2KMgH_3 \rightarrow 0.5Mg_3N_2+K_2MgH_4+2H_2$	0.206	38.6	
2	Entire reaction $10MgH_2+NaMgH_3+6Mg(NH_2)_2+2KMgH_3 \rightarrow NaH+6Mg_3N_2+K_2MgH_4+24H_2$	6.17		12.6

	1^{st} step $10MgH_2+5Mg(NH_2)_2 \rightarrow 5Mg_3N_2+20H_2$	5.14	26	
	2^{nd} step $NaMgH_3+0.5Mg(NH_2)_2 \rightarrow NaH+0.5Mg_3N_2+2H_2$	0.514	31.3	
	3^{rd} step $0.5Mg(NH_2)_2+2KMgH_3 \rightarrow 0.5Mg_3N_2+K_2MgH_4+2H_2$	0.514	38.6	

Estimated reaction temperatures of metal hydride mixtures

We estimated the reaction temperatures of the 72 of the single-step reactions listed in Table S2 and every step associated with the 23 multi-step reactions in Tables 3 and S3 as described in Eq. (3) of the text. The reaction temperatures of $MgH_2/Mg(NH_2)_2$ and $LiNH_2/LiH/KBH_4$ mixtures for a H_2 pressure ofno 1 bar were taken from the van't Hoff plots in Figs. 2 and 3. Figures S1—S4 show the estimated temperature needed to generate a partial pressure of H_2 of 1 bar for each reaction (T_{est}) as a function of a H_2 capacity for the single-step reactions in each category listed in Table 2. Figures S5–S7 show the cumulative H_2 capacity of each reaction as a function of T_{est} for the multi-step reactions where the relevant steps are independent without any connection between the steps. Figures S5 and S6 show the reactions included in Class I of Table S3 and Fig. S7 shows the reactions included in Class II of Table S3.

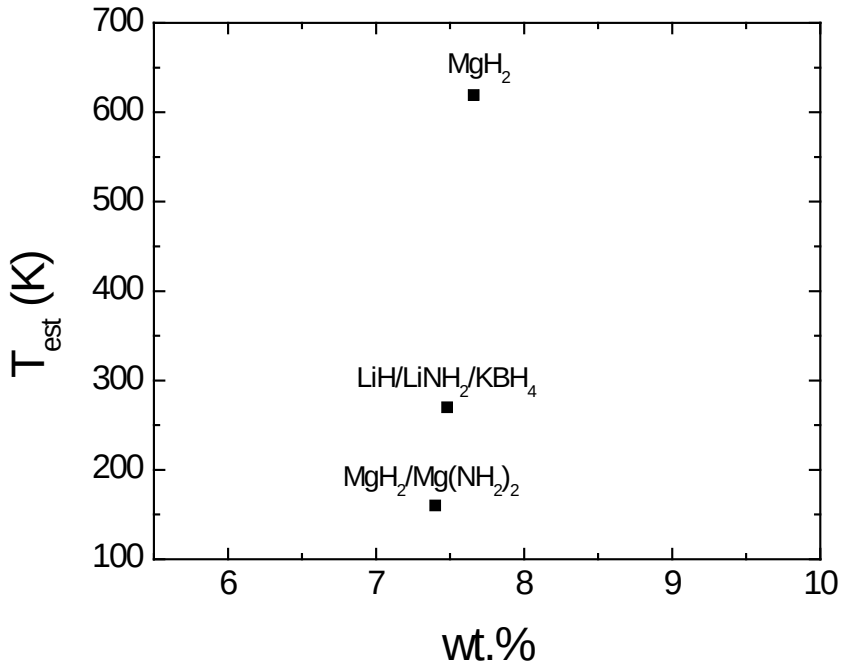

Figure S1. The estimated temperature for generating a partial pressure of H_2 of 1 bar (T_{est}) as a function of H_2 capacity (wt.%) for the "interesting reactions" in Table 2.

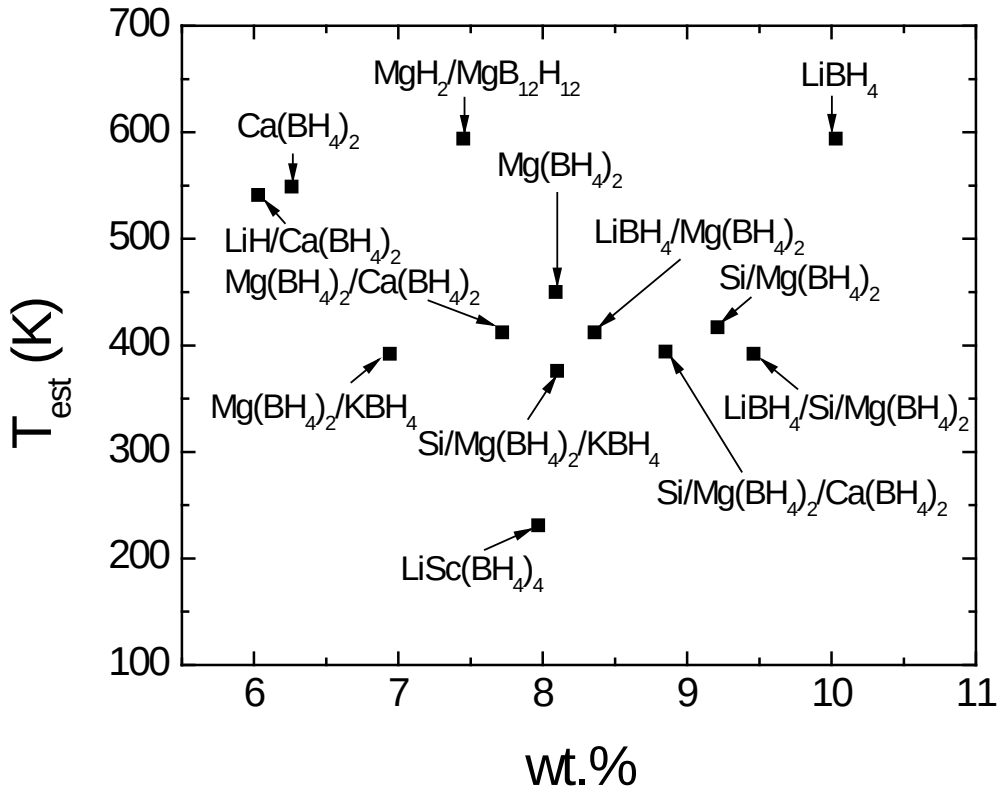

Figure S2. The estimated temperature for generating a partial pressure of H_2 of 1 bar (T_{est}) as a function of H_2 capacity (wt.%) for thirteen reactions involving $B_{12}H_{12}$ species in Table 2.

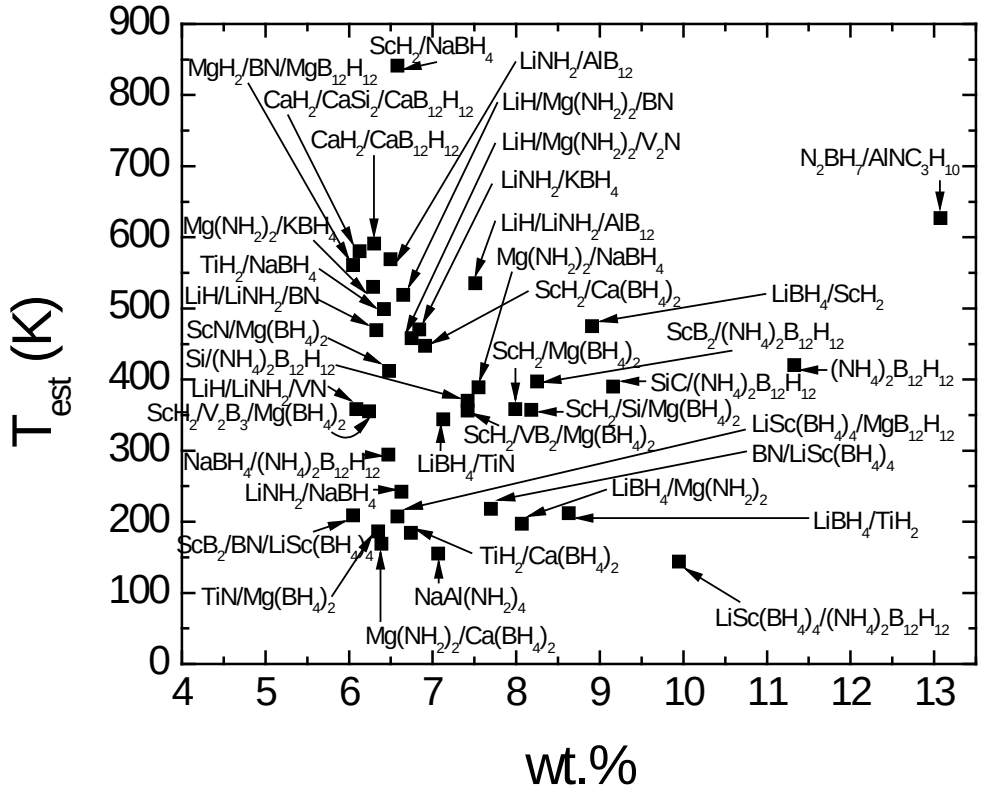

Figure S3. The estimated temperature for generating a partial pressure of H_2 of 1 bar (T_{est}) as a function of H_2 capacity (wt.%) for the 39 reactions involving refractory materials in Table 2.

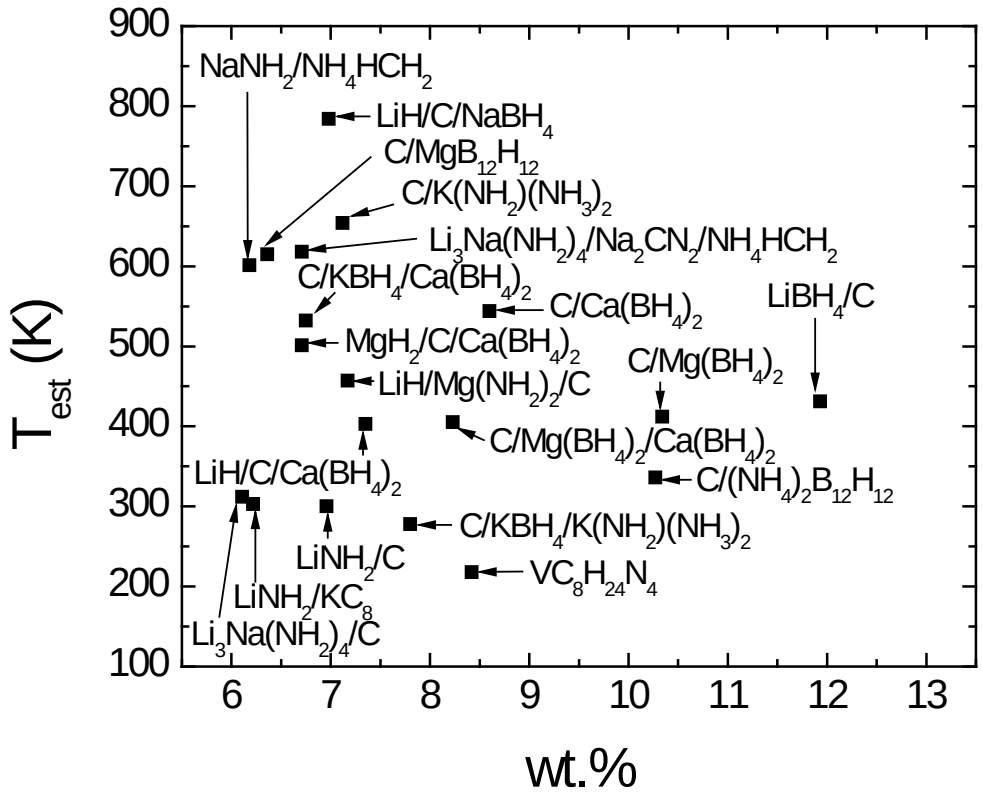

Figure S4. The estimated temperature for generating a partial pressure of H_2 of 1 bar (T_{est}) as a function of H_2 capacity (wt.%) for the nineteen reactions involving C in Table 2.

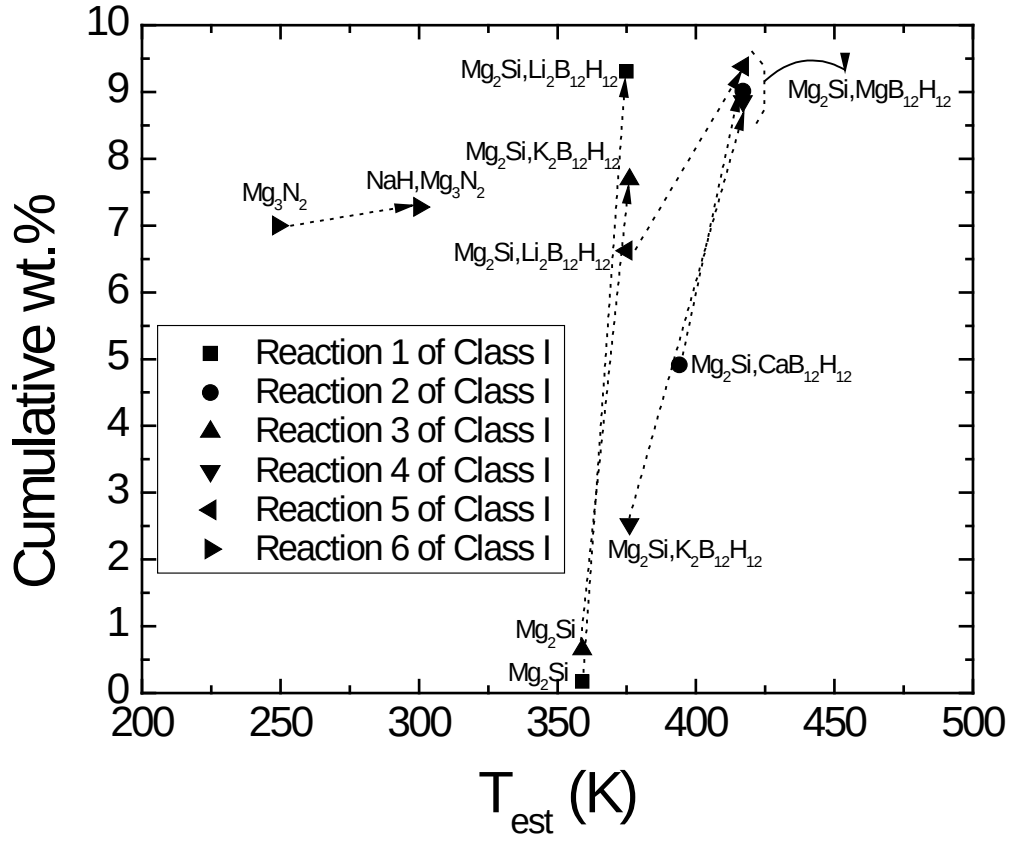

Figure S5. The cumulative H_2 capacity (wt.%) as a function of the estimated temperature for generating a partial pressure of H_2 of 1 bar (T_{est}) for the first six reactions in Class I of Table S3.

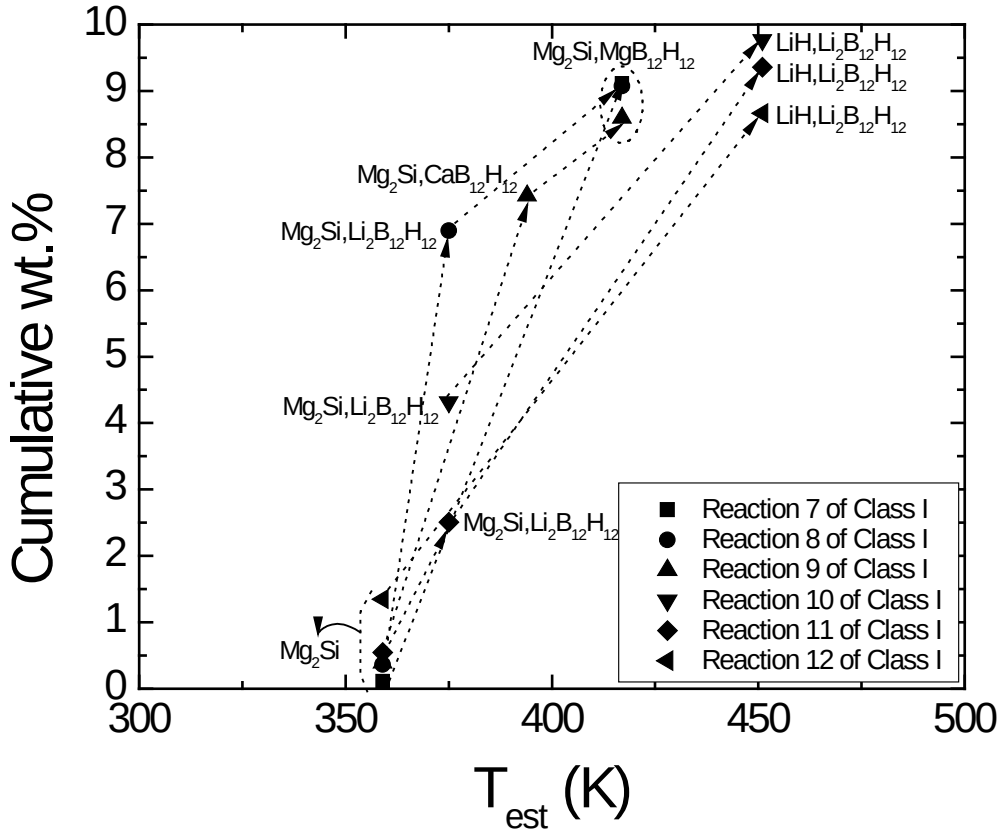

Figure S6. The cumulative H_2 capacity (wt.%) as a function of the estimated temperature for generating a partial pressure of H_2 of 1 bar (T_{est}) for the second six reactions in Class I of Table S3.

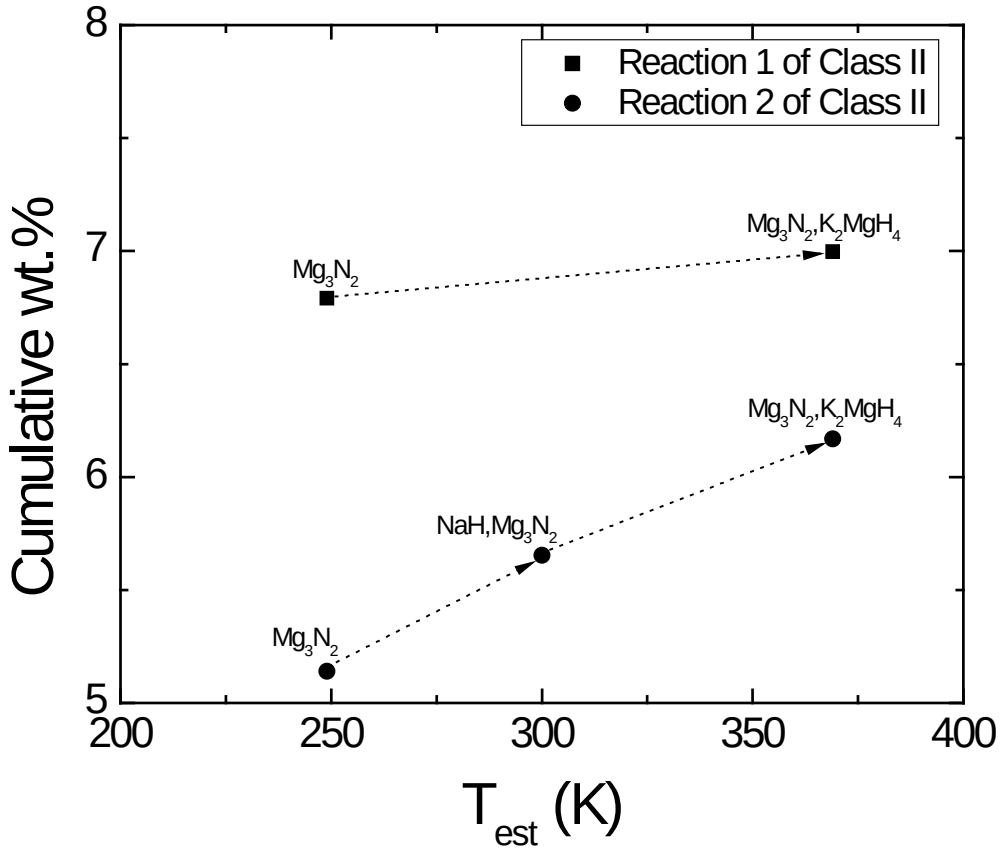

Figure S7. The cumulative H_2 capacity (wt.%) as a function of the estimated temperature for generating a partial pressure of H_2 of 1 bar (T_{est}) for the two reactions in Class II of Table S3.