Supporting Information for

First-Principles Computational Screening of Perovskite Hydrides for Hydrogen Release

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TABLE OF CONTENTS

Table S1

Optimized Lattice Parameters of Perovskite Hydrides (ABH_3)

material	space group	lattice parameter (Å)				references
		a	b	c	β (deg.)	
$LiBeH_3$	*Pnma* (No. 62)	4.517	6.273	4.387		This work
		4.424(2)	6.217(2)	4.639(2)		Expt.[1]
		4.526	6.293	4.404		Calc.[2]
		4.536(1)	6.299(2)	4.410(4)		Calc.[3]
$NaBeH_3$	$Pm\bar{3}m$ (No. 221)	3.346				This work
		3.350				Calc.[2]
		3.352				Calc.[4]
$KBeH_3$	$P2_1/c$ (No. 14)	7.052	5.459	8.692	108.31	This work
		7.085	5.562(1)	8.948(3)	107.45	Calc.[3]
		7.362	5.779	9.297	106.70	Calc.[5]
$RbBeH_3$	$P2_1/c$ (No. 14)	7.502	5.685	9.175	109.43	This work
		7.439	5.789(9)	9.457(9)	108.05	Calc.[3]
		7.150	5.613	9.030	107.40	Calc.[5]
$CsBeH_3$	$P2_1/m1$ (No. 11)	5.098	5.937	7.463	108.01	This work

		5.096(9)	5.935(9)	7.845(1)	107.97	Calc.[3]
$LiMgH_3$	*R3c* (No. 161)	4.945		13.293		This work
		4.958		13.337		Calc.[6]
		4.9226		13.210(6)		Calc.[7]
$NaMgH_3$	*Pnma* (No. 62)	5.446	7.643	5.365		This work
		5.463(4)	7.703	5.410(8)		Expt.[8]
		5.456(8)	7.696(8)	5.377(9)		Calc.[9]
		5.452(5)	7.695(2)	5.368(3)		Calc.[6]
$KMgH_3$	$Pm\bar{3}m$ (No. 221)	4.018				This work
		4.023				Expt.[10]
		4.029(5)				Calc.[6]
		4.010				Calc.[11]
$RbMgH_3$	$P6_3/mmc$ (No. 194)	5.932		14.377		This work
		5.903		14.315(8)		Expt.[12]
		5.906(8)		14.326(1)		Calc.[6]
$CsMgH_3$	*Pmmn* (No. 59)	9.994	6.148	8.584		This work
		9.995(8)	6.132(7)	8.573(6)		Expt.[13]
		9.997(6)	6.133(1)	8.574(1)		Calc.[14]
		9.992(2)	6.140(5)	8.576(8)		Calc.[6]
$LiCaH_3$	*R3c* (No. 161)	5.218		12.461		This work
$NaCaH_3$	$P2_1/c$ (No. 14)	6.805		7.191	90.136	This work
$KCaH_3$	*Pnma* (No. 62)	6.325	8.929	6.299		This work
$RbCaH_3$	$Pm\bar{3}m$ (No. 221)	4.547				This work
		4.547				Expt.[15]
		4.542(7)				Calc.[16]
$CsCaH_3$	$Pm\bar{3}m$ (No. 221)	4.631				This work
		4.6170(2)				Expt.[12]
		4.629(7)				Calc.[16]
$LiSrH_3$	$Pm\bar{3}m$ (No. 221)	4.644				This work
		3.835(7)				Expt.[17]
		3.833				Expt.[18]
$NaSrH_3$	$P2_1/c$ (No. 14)	6.853	6.151	7.739	90.716	This work
$KSrH_3$	*Pnma* (No. 62)	6.744	9.394	6.588		This work
$RbSrH_3$	*Pnma* (No. 62)	6.833	9.607	6.785		This work
$CsSrH_3$	$Pm\bar{3}m$ (No. 221)	4.888				This work
$LiBaH_3$	$Pm\bar{3}m$ (No. 221)	4.020				This work
		4.023(9)				Expt.[17]
		4.023				Expt.[18]
$NaBaH_3$	*R3c* (No. 161)	6.207		14.600		This work
$KBaH_3$	*R3c* (No. 161)	6.937		17.459		This work
$RbBaH_3$	*R3c* (No. 161)	7.158		17.856		This work
$CsBaH_3$	*Pnma* (No. 62)	7.390	10.329	7.275		This work

Structural Property of a $RbMgH_3$ System

The range of Rb–H (Mg–H) bond length is 2.964–3.049 (2.012–2.056 Å) in the optimized structure of $RbMgH_3$. These are consistent with the reported experimental results (i.e., d (Rb–H): 2.96–3.03 Å and d (Mg–H): 2.01–2.04 Å).[12]

Table S2

Formation Enthalpies (eV) of All Hydrides by Pathways 2–5

material	pathway 2	pathway 3	pathway 4	pathway 5
$LiBeH_3$ (*Pnma*)	**−0.630**	−0.475	0.208	0.363
$NaBeH_3$ ($Pm\bar{3}m$)	**−0.258**	−0.103	0.120	0.275
$KBeH_3$ ($P2_1/c$)	**−0.531**	−0.376	−0.131	0.024
$RbBeH_3$ ($P2_1/c$)	**−0.505**	−0.350	−0.196	−0.041
$CsBeH_3$ ($P2_1/m1$)	**−0.583**	−0.429	−0.261	−0.106
$LiMgH_3$ (*R3c*)	**−1.339**	−0.822	−0.501	0.017
$NaMgH_3$ (*Pnma*)	**−1.043**	−0.526	−0.665	−0.148
$KMgH_3$ ($Pm\bar{3}m$)	**−1.292**	−0.775	−0.892	−0.375
$RbMgH_3$ ($P6_3/mmc$)	**−1.142**	−0.625	−0.833	−0.316
$CsMgH_3$ (*Pmmn*)	**−1.194**	−0.676	−0.871	−0.354
$LiCaH_3$ (*R3c*)	**−2.424**	−0.740	−1.585	0.099
$NaCaH_3$ ($P2_1/c$)	**−1.755**	−0.071	−1.377	0.307
$KCaH_3$ (*Pnma*)	**−1.902**	−0.217	−1.502	0.182
$RbCaH_3$ ($Pm\bar{3}m$)	**−1.900**	−0.216	−1.591	0.093
$CsCaH_3$ ($Pm\bar{3}m$)	**−1.961**	−0.277	−1.638	0.046
$LiSrH_3$ ($Pm\bar{3}m$)	**−0.163**	1.455	0.676	2.294
$NaSrH_3$ ($P2_1/c$)	**−1.719**	−0.101	−1.341	0.277
$KSrH_3$ (*Pnma*)	**−1.691**	−0.073	−1.291	0.327
$RbSrH_3$ (*Pnma*)	**−1.676**	−0.058	−1.367	0.251
$CsSrH_3$ ($Pm\bar{3}m$)	**−1.809**	−0.191	−1.487	0.131
$LiBaH_3$ ($Pm\bar{3}m$)	**−2.554**	−1.082	−1.716	−0.243
$NaBaH_3$ (*R3c*)	**−1.723**	−0.251	−1.346	0.127
$KBaH_3$ (*R3c*)	**−1.449**	0.023	−1.049	0.423
$RbBaH_3$ (*R3c*)	**−1.371**	0.102	−1.062	0.410
$CsBaH_3$ (*Pnma*)	**−1.461**	0.011	−1.139	0.334

Table S3

Bond Orders of Dopant-free and Cs-doped $NaCaH_3$ Systems[a]

system	1	2	3	4	5	6	7	8	9	10
$NaCaH_3$	0.152	0.084	0.151	0.151	0.204	0.198	0.172	0.217	0.234	0.225
Cs-doped $NaCaH_3$	0.130	0.150	0.097	0.136	0.195	0.204	0.181	0.219	0.185	0.219

[a]All bonds of 1−10 are shown in Figure 3.

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