FIG. S1 Calculated formation energy for $V_{H}$ and $H_{i}$ with different charge states in (a) NaBH$_4$ and (b) KBH$_4$. 
FIG S2: H₂ diffusion pathway in LiBH₄. (a) initial state of H₂ viewed from [010] direction; (b) initial state of H₂ viewed from [100] direction; (c) transition state of H₂ viewed from [010] direction; (d) transition state of H₂ viewed from [100] direction. For the initial state, the distance of the center of mass of H₂ to the nearby Li atoms is 3.12 Å; for the transition state, the distance of center of mass of H₂ to the nearby Li (B) atoms is 2.8 Å (2.9 Å).
FIG S3: H\textsubscript{2} diffusion pathway in NaBH\textsubscript{4}. (a) initial state of H\textsubscript{2}; (b) transition state of H\textsubscript{2}; (c) final state of H\textsubscript{2}. For the initial and final state of H\textsubscript{2}, the distance of center of mass of H\textsubscript{2} to the nearby corner of the tetrahedron shown in the figure is 2.62 Å. For the transition state of H\textsubscript{2}, the distance between the center of mass of H\textsubscript{2} and corners of the cube defined by the nearby NA atoms is 2.32 Å.