Supplementary Information

# Li-Na Ternary Amidoborane for Hydrogen Storage: Experimental and First-principles Study

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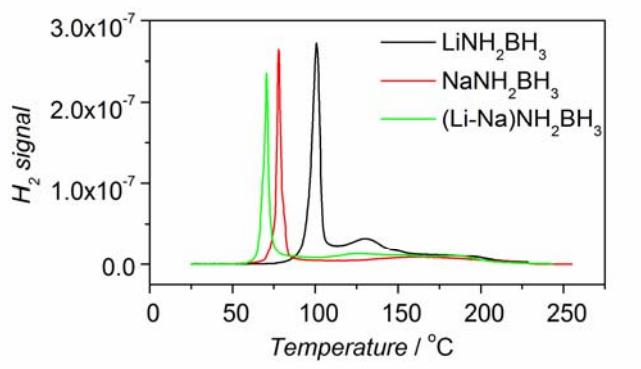


**Fig. S1** Temperature-programmed desorption (TPD) spectra of $LiNH_2BH_3$ (LiAB), $NaNH_2BH_3$ (NaAB) and (Li-Na)$NH_2BH_3$.

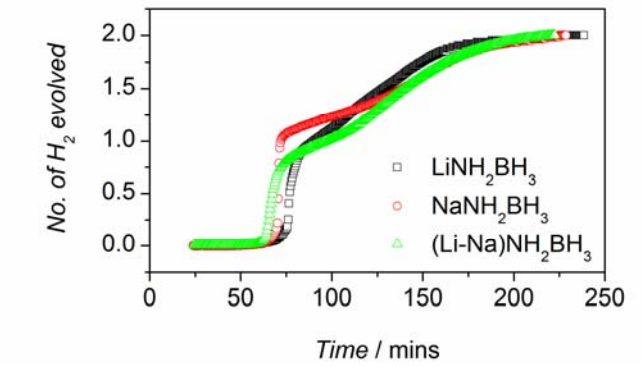


**Fig. S2** $H_2$ evolution from $LiNH_2BH_3$ (LiAB), $NaNH_2BH_3$ (NaAB) and Li-Na)$NH_2BH_3$ during dehydrogenation process. *ca.* 2 *equiv.* $H_2$ (~ 8.9 wt. % $H_2$) are detached via a two-step process at temperature below 200 ºC, which is similar to the H-desorption processes of LiAB and NaAB. Quantitative measurement of hydrogen desorption at elevated temperatures was carried out on a commercial gas reactor controller provided by the Advanced Materials Corporation. Sample was ramp-heated in argon at 1 K $min^{-1}$ each time.

**Table S1** 14 possible symmetrically non-equivalent $Li_4/Na_4$ arrangements on 8-cation sites (A, B, C…H are corresponding sites in Fig. S3) in $Li_4Na_4AB_8$.

| No. | $Li_4/Na_4$ arrangements | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | A | B | C | D | E | F | G | H |
| 01 | Li | Na | Li | Li | Na | Li | Na | Na |
| 02 | Li | Li | Na | Li | Na | Li | Na | Na |
| 03 | Li | Na | Li | Li | Na | Na | Na | Li |
| 04 | Li | Li | Na | Li | Na | Na | Na | Li |
| 05 | Li | Na | Na | Li | Li | Na | Li | Na |
| 06 | Na | Li | Li | Na | Li | Na | Na | Li |
| 07 | Na | Li | Li | Na | Na | Li | Li | Na |
| 08 | Na | Na | Li | Li | Na | Na | Li | Li |
| 09 | Li | Na | Li | Na | Li | Na | Li | Na |
| 10 | Li | Na | Li | Na | Na | Li | Na | Li |
| 11 | Na | Na | Li | Li | Na | Li | Li | Na |
| 12 | Na | Li | Na | Li | Na | Na | Li | Li |
| 13 | Na | Na | Li | Li | Li | Li | Na | Na |
| 14 | Li | Li | Li | Li | Na | Na | Na | Na |

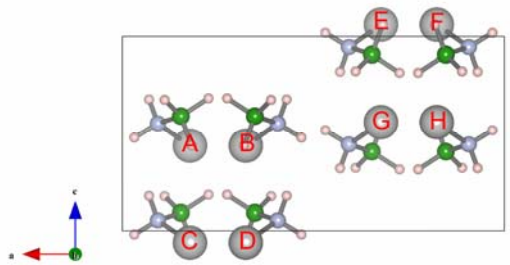


**Fig. S3** Labeled 8-cation sites in unit cell of $Li_{1-x}Na_xAB$. The possibilities of $Li_4/Na_4$ occupation on 8-cation sites are $C_8^4 = 70$, we calculated the pair correlation functions of all possible configurations, and categorized them into 14 symmetrically non-equivalent Li/Na arrangements (Table S1), as the symmetrically equivalent arrangements possess the equal pair correlation functions. The optimized structures are displayed in Fig. S4.

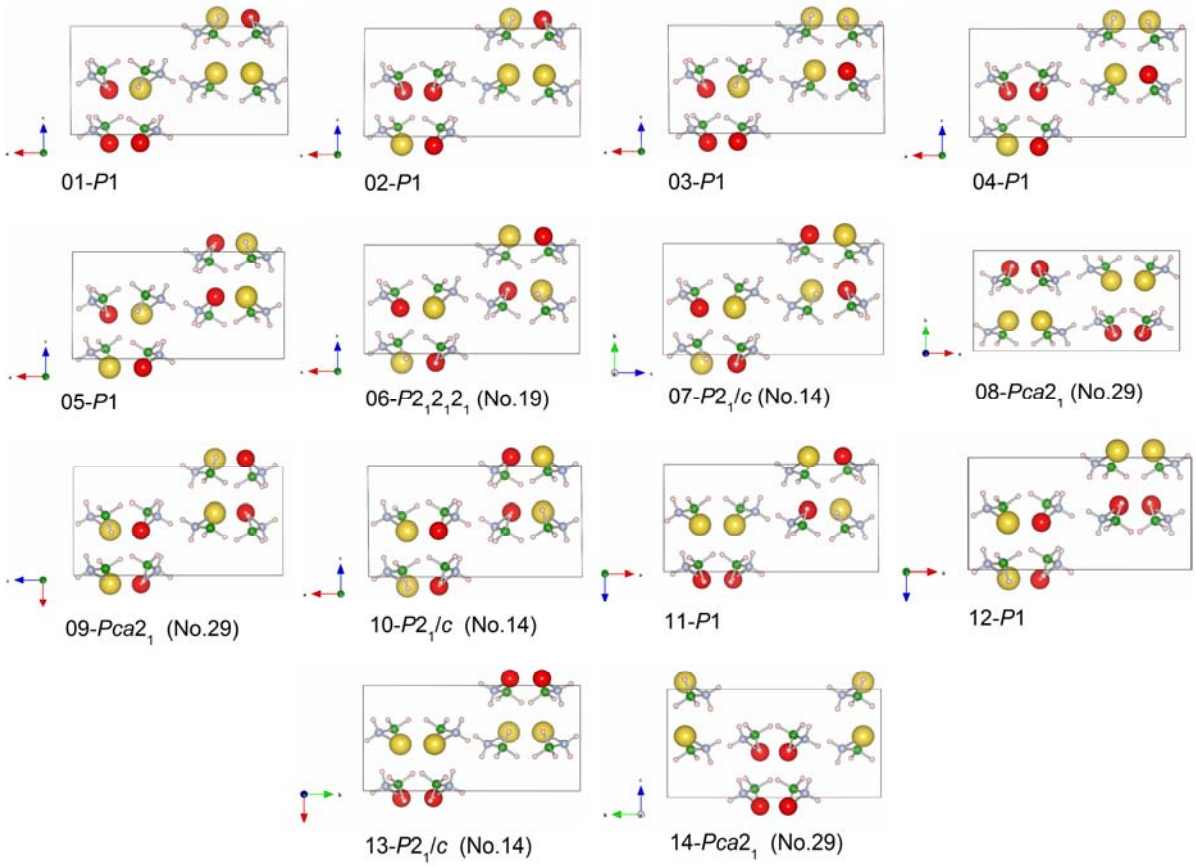


**Fig. S4** The optimized structures of 14 possible symmetrically non-equivalent configurations of $Li_4Na_4AB_8$.

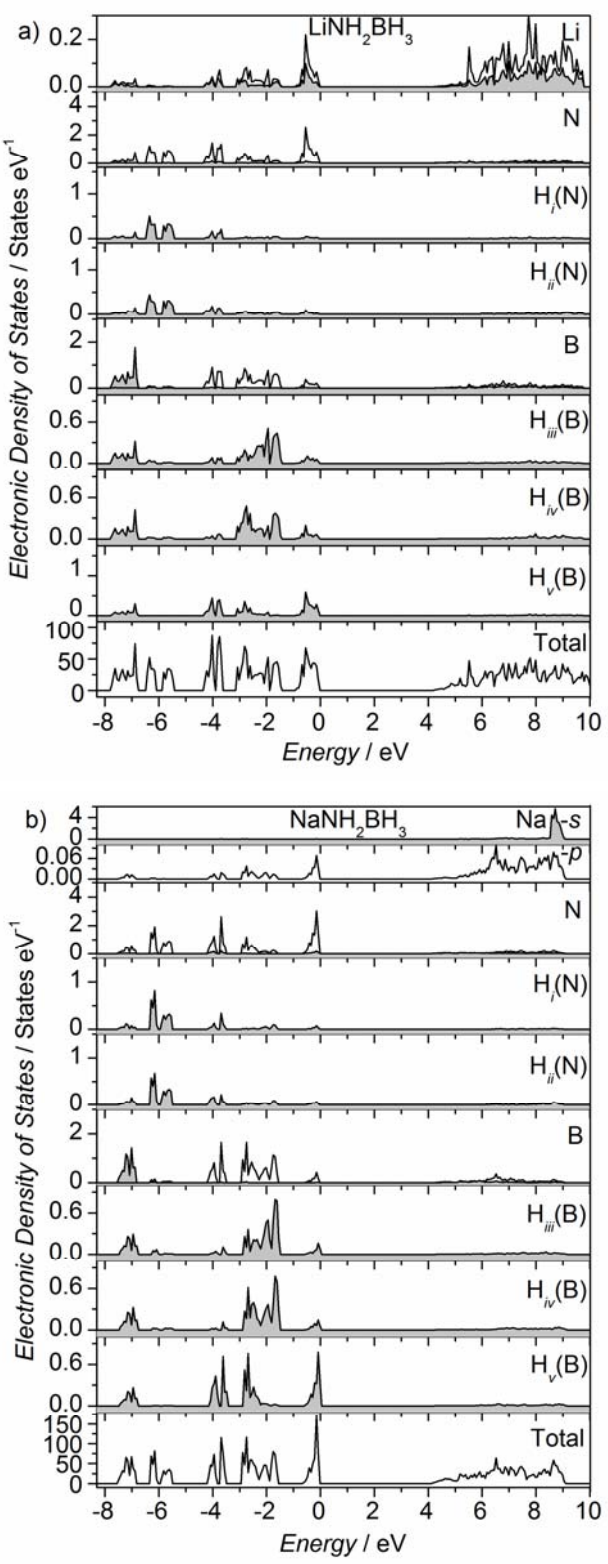


**Fig. S5** The partial and total electronic density of states of (a) $LiNH_2BH_3$ (LiAB) and (b) $NaNH_2BH_3$ (NaAB). The Fermi level is set at zero energy; *s*-electron contributions are depicted in gray.

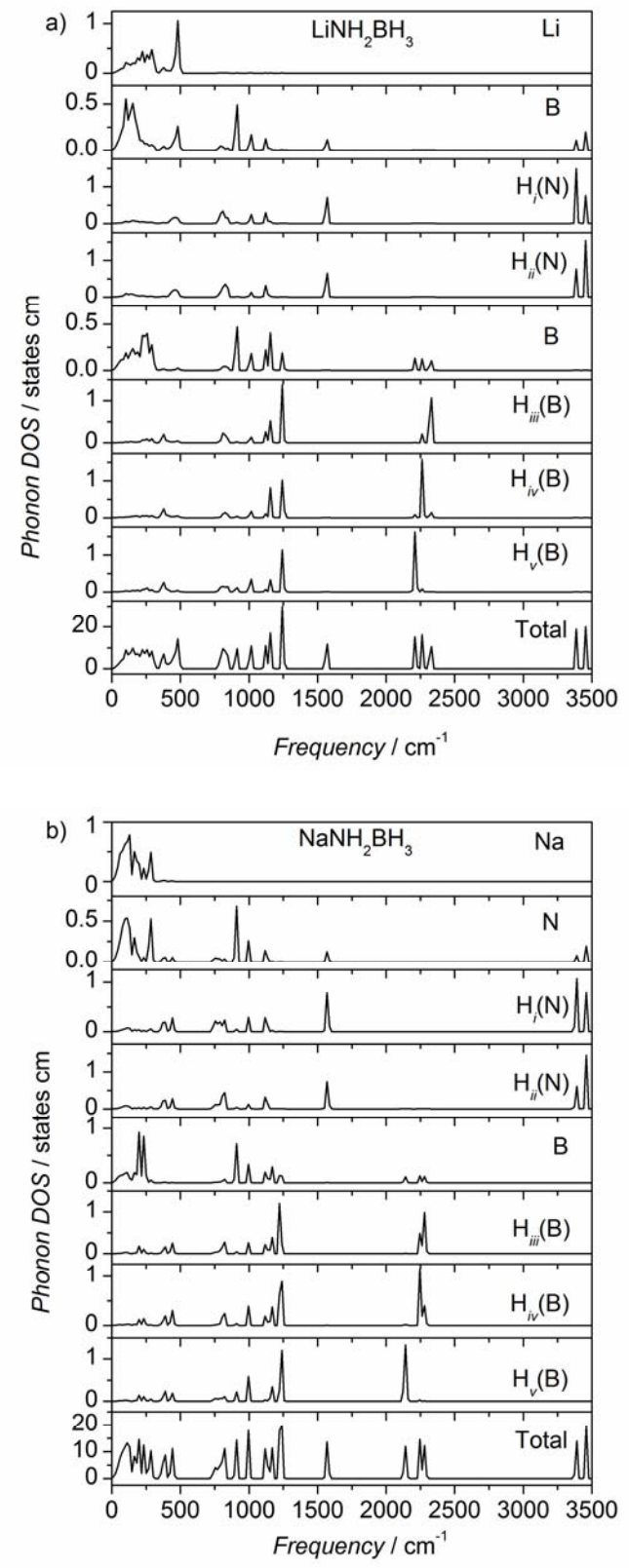


**Fig. S6** The calculated partial and total phonon density of states of (a) $LiNH_2BH_3$ (LiAB) and (b) $NaNH_2BH_3$ (NaAB).