

## Supporting information

### Effective Thermodynamic Alteration to $\text{Mg}(\text{NH}_2)_2\text{-LiH}$ System:

### Achieving near Ambient-Temperature Hydrogen Storage

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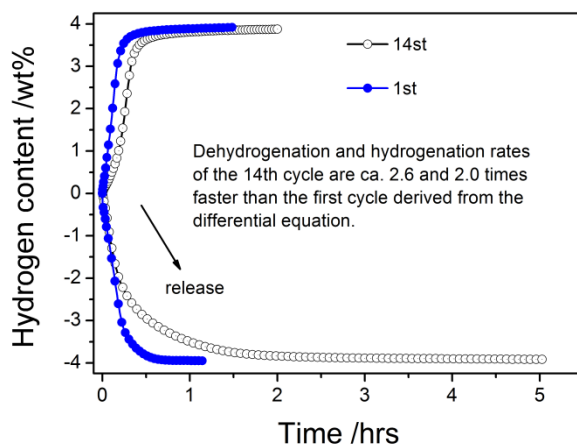
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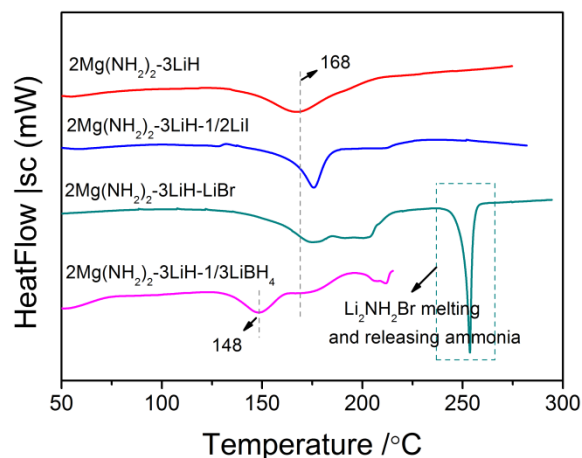
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**Fig. S1** The first and fourteenth isothermal dehydrogenation/re-hydrogenation cycle of  $S_B$  ( $2\text{Mg}(\text{NH}_2)_2\text{-}3\text{LiH}\text{-}1/3\text{LiBH}_4$ ) sample at 447 K.



**Fig. S2** The C80 spectra of  $S_P$  ( $2\text{Mg}(\text{NH}_2)_2\text{-}3\text{LiH}$ ),  $S_I$  ( $2\text{Mg}(\text{NH}_2)_2\text{-}3\text{LiH-}1/2\text{LiI}$ ),  $S_{Br}$  ( $2\text{Mg}(\text{NH}_2)_2\text{-}3\text{LiH-LiBr}$ ) and  $S_B$  ( $2\text{Mg}(\text{NH}_2)_2\text{-}3\text{LiH-}1/3\text{LiBH}_4$ ) samples. (Differential Scanning Calorimetry (DSC) measurement was carried out on a SETARAM C80 thermal analysis system equipped with a sealed cell at a ramping rate of 0.2 K/min). Due to the process of those reactions in a sealed system of C80 cannot be monitored, so the enthalpies are hardly reflected from the C80 tests accurately. However, with the C80 results, it can be known that the dehydrogenation reactions of all the doped samples are multistep reactions. It indicated that  $\text{LiNH}_2$  indeed reacted with  $\text{LiI}$ ,  $\text{LiBr}$  and  $\text{LiBH}_4$  during the dehydrogenation process. The reaction temperature of  $\text{LiNH}_2\text{-}1/2\text{LiI}$ ,  $\text{LiNH}_2\text{-LiBr}$  and  $\text{LiNH}_2\text{-}1/3\text{LiBH}_4$  mixtures are different resulted in the dehydrogenation peaks of  $S_I$ ,  $S_{Br}$  and  $S_B$  samples are different. The different of dehydrogenation peaks in C80 (close system) and TPD-MS (open system) (Fig. 4) may be due to the reaction conditions and ramping rates. Moreover, the C80 result of  $\text{LiBr}$  doped sample can be explained why the ammonia concentration of  $\text{LiBr}$  doped sample is the highest.