

### Supplementary Information:

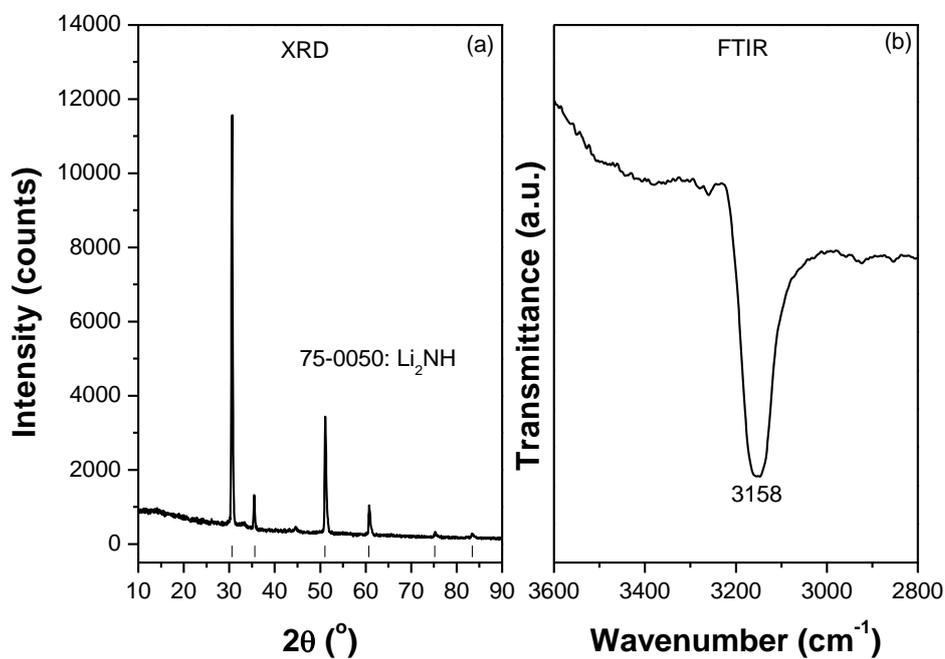


Fig. S1 XRD pattern (a) and FTIR spectrum (b) of the as-prepared Li<sub>2</sub>NH

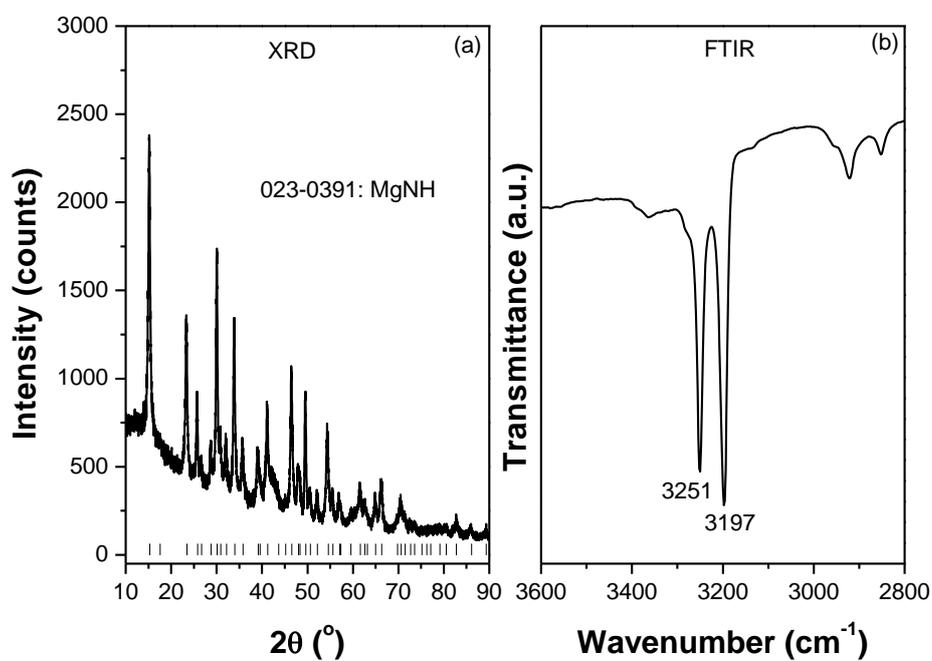


Fig. S2 XRD pattern (a) and FTIR spectrum (b) of the as-prepared MgNH

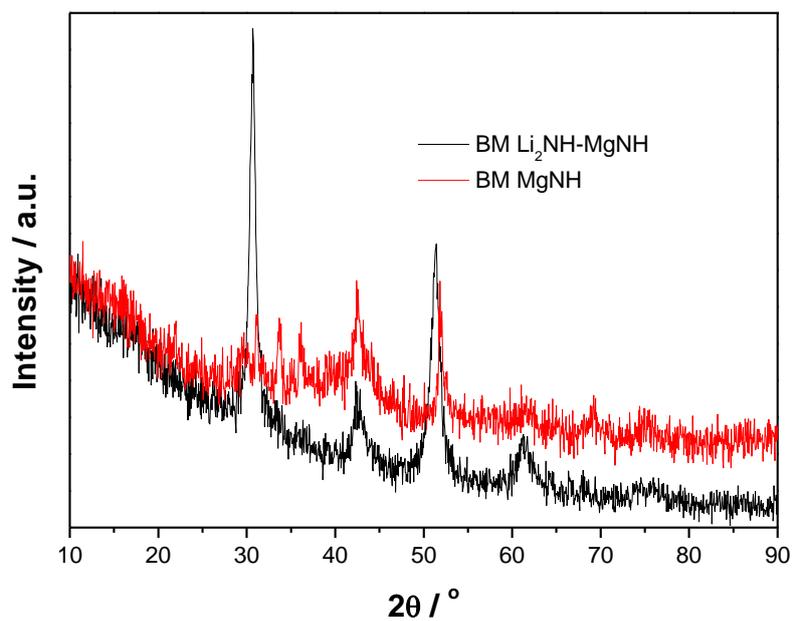


Fig. S3 Comparison on XRD patterns of the as-milled MgNH and Li<sub>2</sub>NH-MgNH

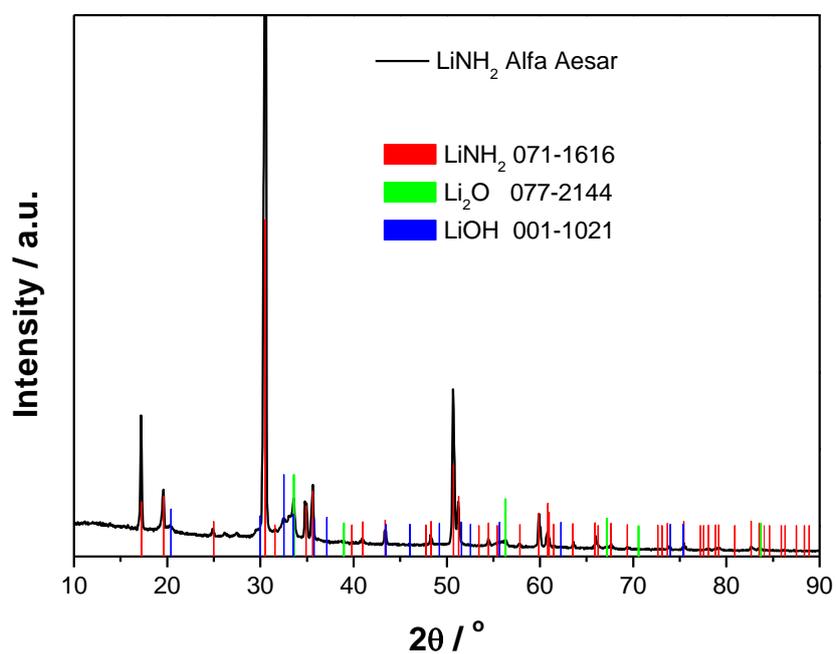


Fig. S4 XRD pattern of the as-received LiNH<sub>2</sub>

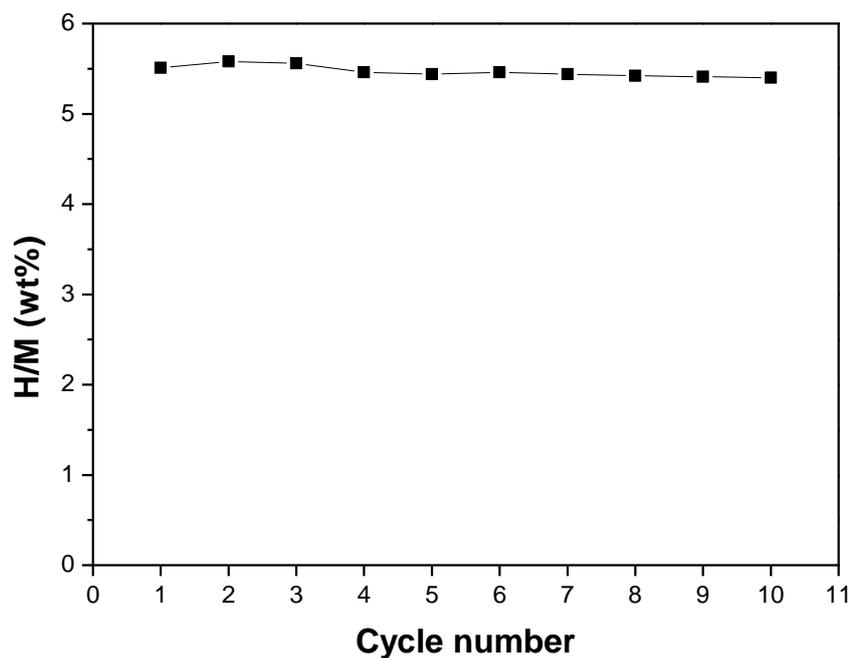


Fig. S5 Hydrogen desorption capacities of the hydrogenated  $\text{Li}_2\text{NH}$ -0.5MgNH sample within first ten cycles

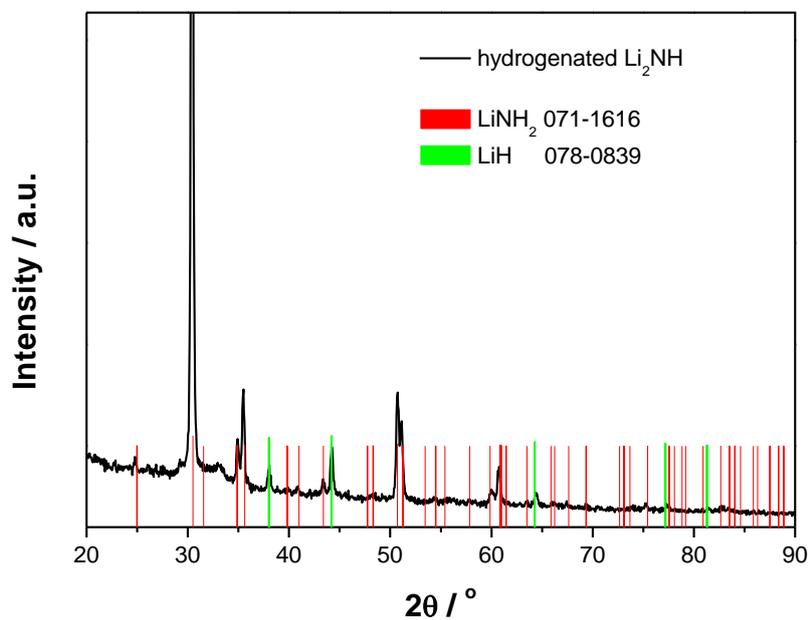


Fig. S6 A full profile analysis on XRD pattern of the hydrogenated  $\text{Li}_2\text{NH}$  sample

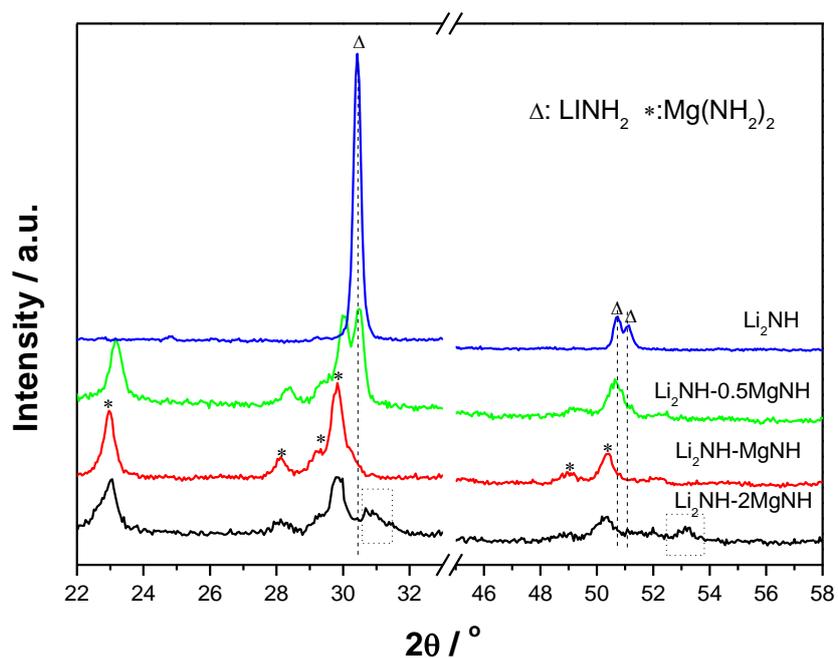


Fig. S7 XRD patterns with enlarged scale for the hydrogenated  $\text{Li}_2\text{NH}-x\text{MgNH}$  samples

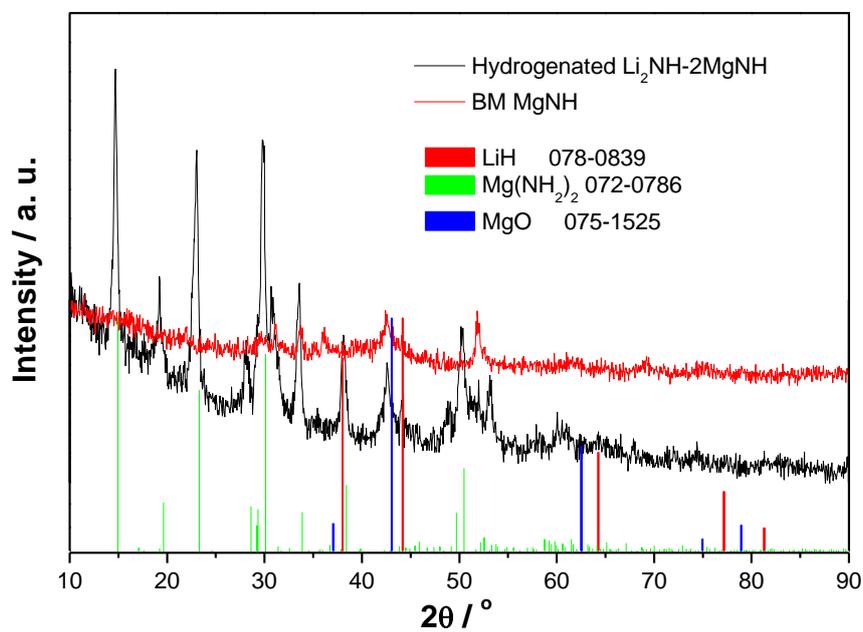


Fig. S8 A full profile analysis on XRD pattern of the hydrogenated  $\text{Li}_2\text{NH}-2\text{MgNH}$  samples

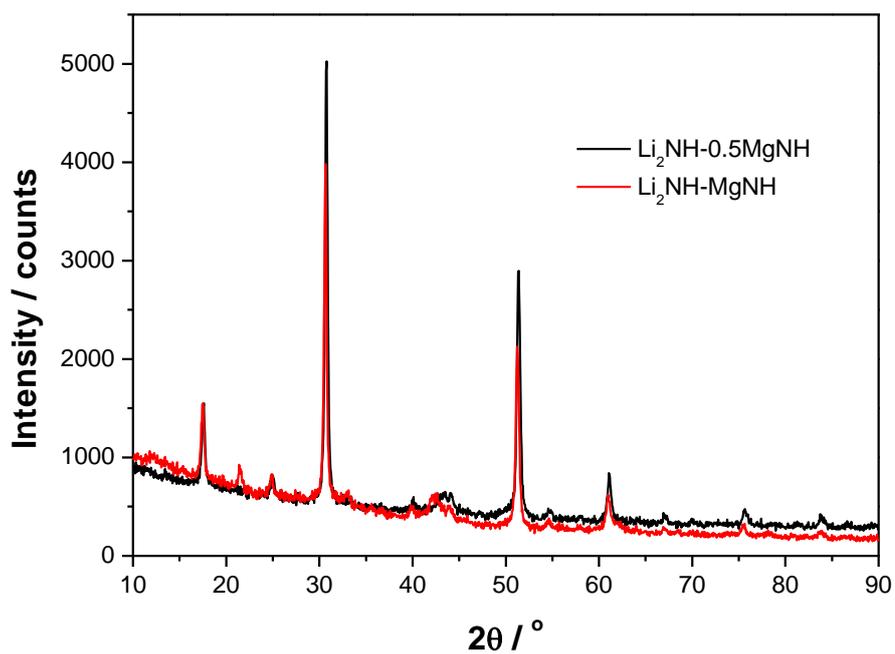


Fig. S9 Comparison of XRD patterns of the  $\text{Li}_2\text{NH}-0.5\text{MgNH}$  and  $\text{Li}_2\text{NH}-\text{MgNH}$  samples after one hydrogenation/dehydrogenation cycle

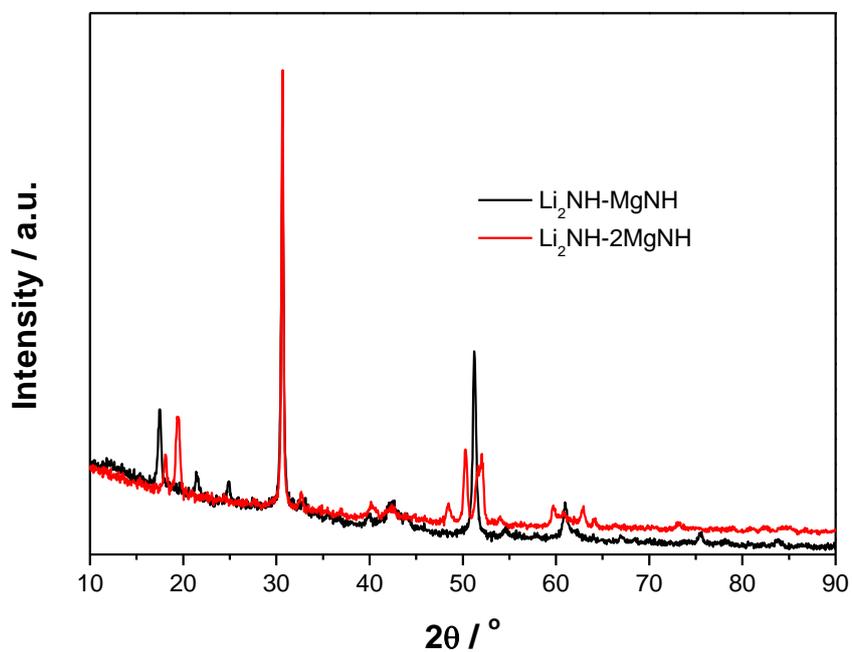


Fig. S10 Comparison of XRD patterns of the  $\text{Li}_2\text{NH}-\text{MgNH}$  and  $\text{Li}_2\text{NH}-2\text{MgNH}$  samples after one hydrogenation/dehydrogenation cycle