Supplementary information:

Preparation procedure of Mg(NH$_2$)$_2$: Mg(NH$_2$)$_2$ was synthesized by reacting the pre-milled Mg powder with ammonia. The typical procedure is as follows: ~ 2 g of Mg powder (99%, Sinopharm) were first ball milled in a gaseous ammonia atmosphere on a planetary ball mill (QM-3SP4) at 500 rpm for 36 h at room temperature. The as-milled products were then transferred to a stainless-steel tube reactor in a glovebox. The tube reactor was connected to a homemade gas-solid reaction apparatus. After outgassing at room temperature, ~ 7 bar ammonia was loaded into the reactor, and the sample temperature was gradually raised to 300 °C at a heating rate of 2 °C min$^{-1}$ and then kept for 24 h.

Fig. S1 XRD pattern (a) and FTIR spectrum (b) of the as-prepared Li$_2$Mg$_2$N$_3$H$_3$ sample. Three absorbances at 2800-2990 cm$^{-1}$ may originate from the C-H stretches of the ethanol contamination which was employed to wash the press mold.
Fig. S2 FTIR spectrum (a) and XRD pattern (b) of the post-milled MgNH.

Fig. S3 XRD pattern (a) and FTIR spectrum (b) of the re-dehydrogenated sample.
Fig. S4 MS profiles of the hydrogenated Li₂Mg₂N₃H₃ sample with temperatures