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 ºC min^{-1} and then kept for 24 h.

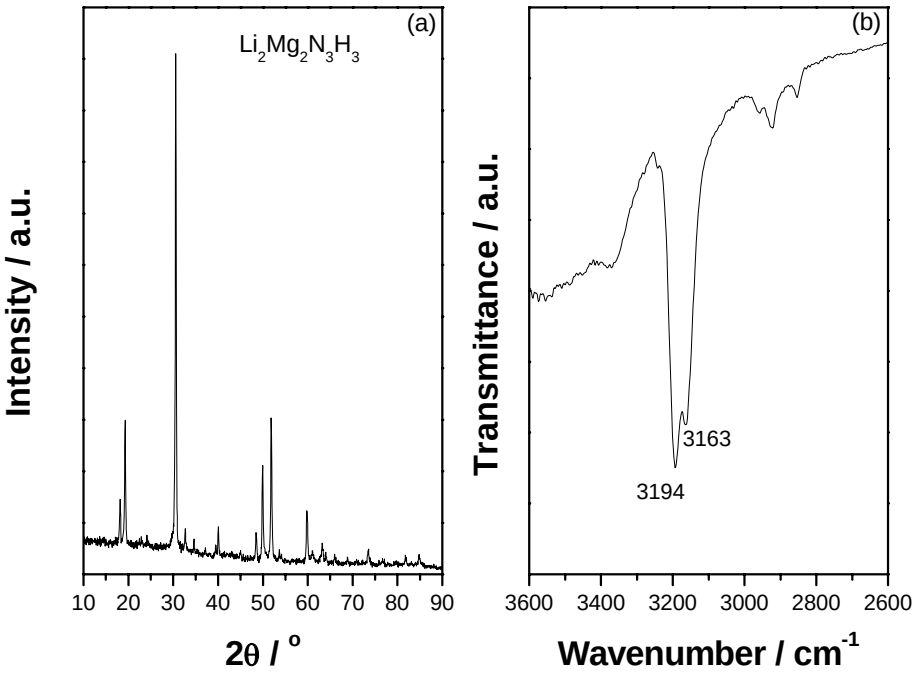

Fig. S1 XRD pattern (a) and FTIR spectrum (b) of the as-prepared $Li_2Mg_2N_3H_3$ sample. Three absorbances at 2800-2990 cm^{-1} may originate from the C-H stretchs of the ethanol contamination which was employed to wash the press mold.

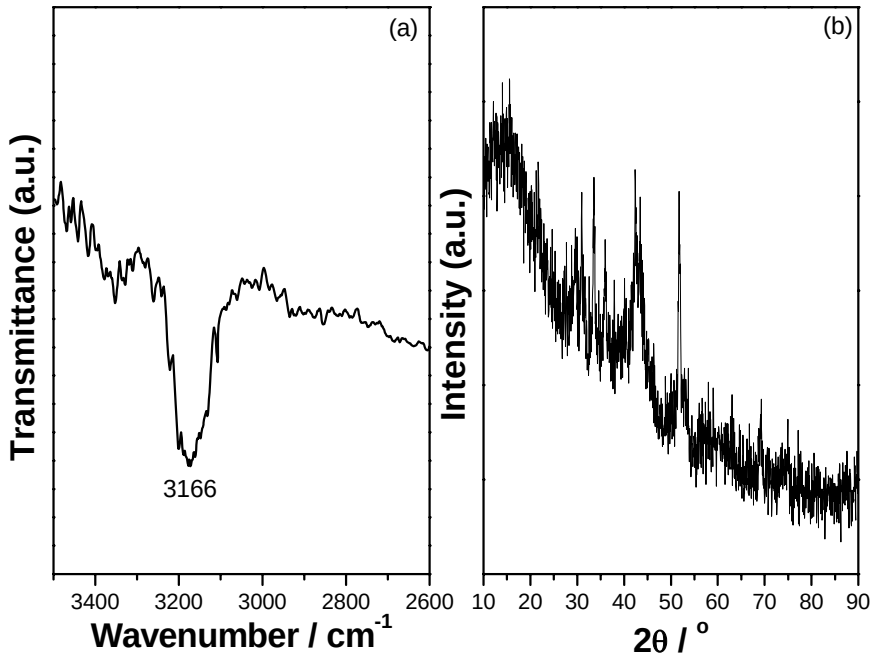

Fig. S2 FTIR spectrum (a) and XRD pattern (b) of the post-milled MgNH.

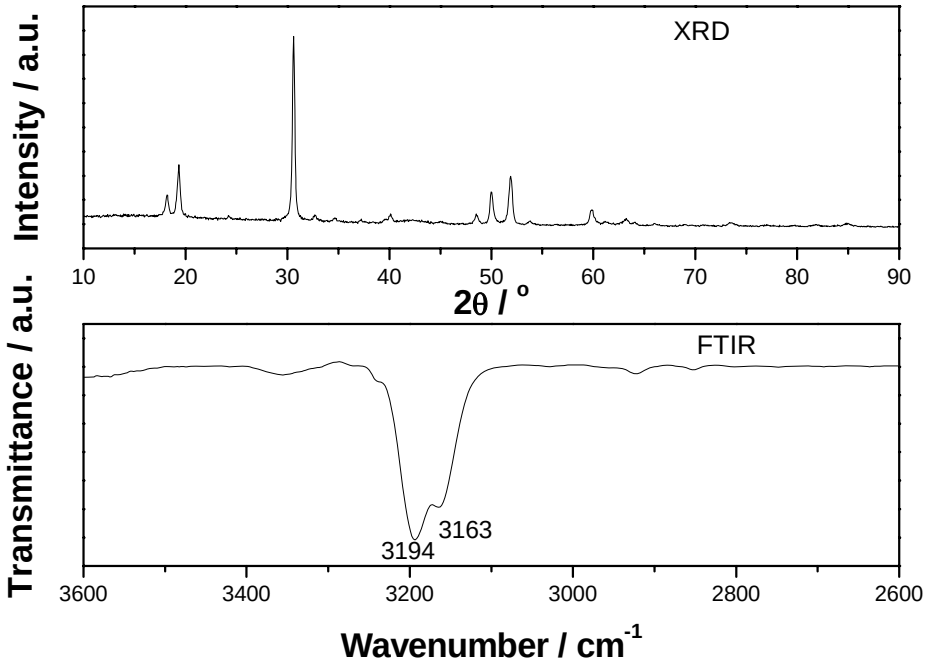

Fig. S3 XRD pattern (a) and FTIR spectrum (b) of the re-dehydrogenated sample.

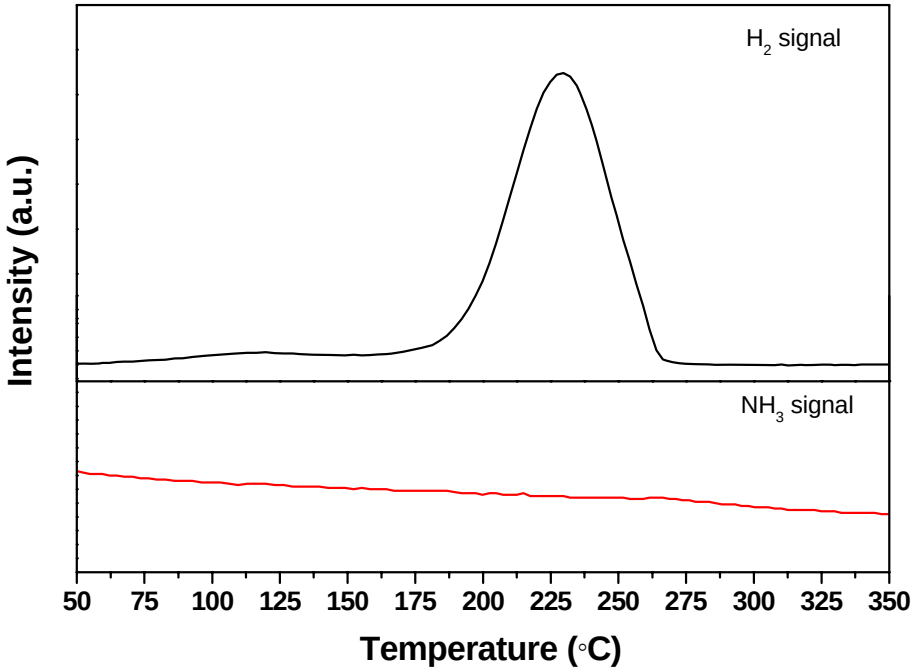

Fig. S4 MS profiles of the hydrogenated $Li_2Mg_2N_3H_3$ sample with temperatures