## **Supporting Information**

## Effective participation of $Li_4(NH_2)_3BH_4$ in the dehydrogenation

## pathway of Mg(NH<sub>2</sub>)<sub>2</sub>-2LiH composite

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thermal treating (TT).



**Fig. S2:** Sixth cycle of dehydrogenation (A) and rehydrogenation (B) at 200 °C of the LM and LMB samples. Absorption hydrogen pressure: 6.0 MPa; Desorption hydrogen pressure: 0.05 MPa.





Fig. S3: XRPD patterns of the LMB sample at points 1, 2, 3 and 4 of the PCI curve (Fig. 6B).



Fig. S4: FTIR spectra of sample LMB at points 1, 2, 3 and 4 of the PCI curve (Fig. 6B).

Table S1: Phase abundance (mol % and wt %) of sample LMB at points 1, 2, 3 and 4 of the PCI (Fig.

Stage in PCI	LiNH <sub>2</sub>		Mg(NH <sub>2</sub> ) <sub>2</sub>		LiH		Li₄BN₃H <sub>10</sub>		Li <sub>2</sub> Mg <sub>2</sub> (NH) <sub>3</sub>		a-Li <sub>2</sub> Mg(NH) <sub>2</sub>		b-Li <sub>2</sub> Mg(NH) <sub>2</sub>		Rwp	Experimental H <sub>2</sub> release <sup>1</sup>	Calculated H <sub>2</sub> released <sup>2</sup>
	% Iom	wt %	% lom	wt %	% lom	wt %	% Iom	wt %	% Iom	wt %	% lom	wt %	% lom	wt %		wt %	wt %
1	0	0	32	63	62	17	6	20	0	0	0	0	0	0	8.23	0.0	0.0
2	0	0	21	26	49	9	10	20	19	45	0	0	0	0	7.08	2.3	2.6
3	12	5	0	0	37	5	12	19	32	62	7	9	0	0	6.14	3.0	3.9
4	0	0	0	0	0	0	7	9	0	0	50	49	13	42	9.28	5.6	5.4

6B). All data have been obtained by Rietveld analysis using Fullprof software showed in Fig. S3.

<sup>1</sup> Hydrogen content determined experimentally. PCI measurement at 200 °C was repeated using different samples. Each run was stopped at selected values of hydrogen released, indicated in Fig. 6B with points 1, 2, 3 and 4. The quenched and withdrawn samples were subsequently analyzed by XRPD. The patterns were refined using the Rietveld method and the Rwp value obtained from each refinement is indicated in Table S1.

2 Hydrogen release was calculated from the measured wt% values by assuming constant the total mass of all elements excepting hydrogen (Li, Β, Ν and Mg).