Supplementary Informations

Hydrogen reversibility of $LiBH_4$-MgH_2-Al Composites

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Rietveld refinement

Rietveld refinement is performed on selected PXD patterns from *in situ* SR-PXD measurements and presented below in Figure S1 and S2. The extracted data is presented in Table 2.

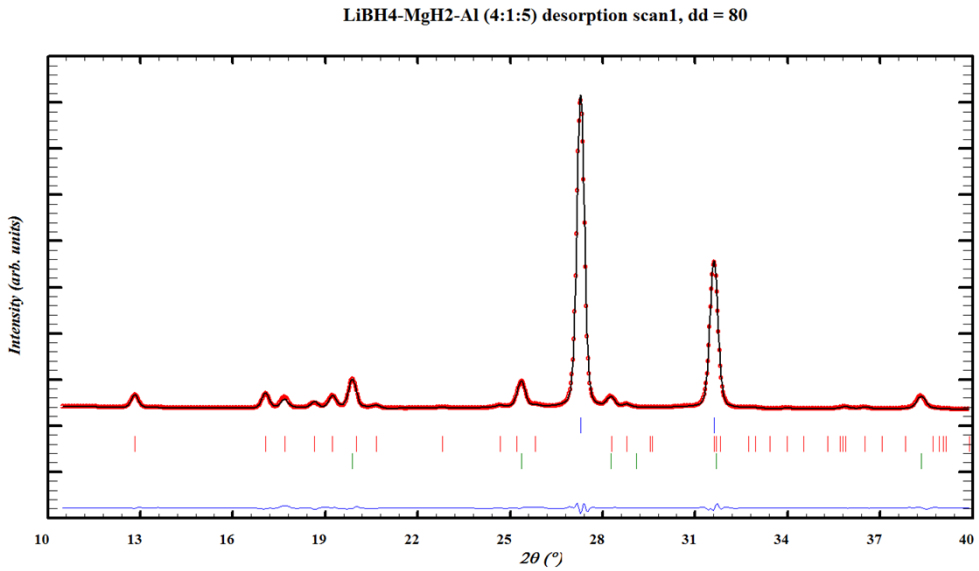

Figure S1. Rietveld refinement profile (Y_{calc}, upper black line) of SR-PXD data (observed data Y_{obs}, red) for $LiBH_4$-MgH_2-Al (4:1:5, S2) measured at *RT before desorption* (λ = 1.10205 Å). This data is shown in Figure 2. Ticks: Al (upper, blue), *o*-$LiBH_4$ (middle, red) and MgH_2 (lower, green). The difference profile ($Y_{obs} - Y_{calc}$) is shown as the lower (blue) curve. Reliability factors, R_{Bragg}, are R_{Bragg}(*o*-$LiBH_4$) = 6.37, R_{Bragg}(Al) = 0.167 and R_{Bragg}(MgH_2) = 1.44.

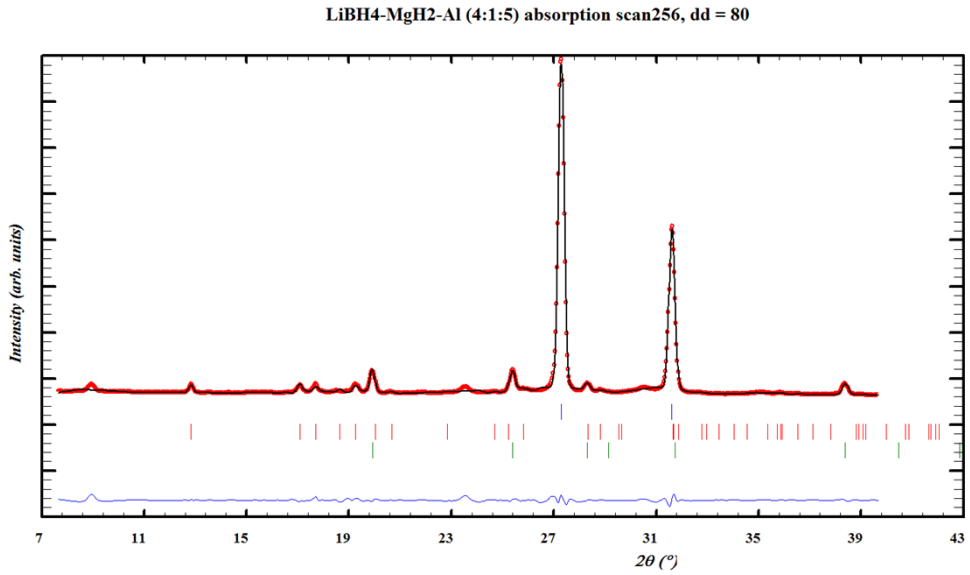

Figure S2. Rietveld refinement profile (Y_{calc}, upper black line) of SR-PXD data (observed data Y_{obs}, red) for $LiBH_4$-MgH_2-Al (4:1:5, **S2**) measured at *RT after absorption* (λ = 1.10205 Å). This data is shown in Figure 5. Ticks: Al (upper, blue), *o*-$LiBH_4$ (middle, red) and MgH_2 (lower, green). The difference profile ($Y_{obs} - Y_{calc}$) is shown as the lower (blue) curve. Reliability factors, R_{Bragg}, are R_{Bragg}(*o*-$LiBH_4$) = 26.1, R_{Bragg}(Al) = 0.514 and R_{Bragg}(MgH_2) = 4.54.

Table S1. Crystallographic data for compounds observed in the $LiBH_4$–MgH_2–Al system.

Compound	Space group	*a* (Å)	*b* (Å)	*c* (Å)	Reference
o–$LiBH_4$	*Pnma*	7.179	4.437	6.803	[1]
h–$LiBH_4$	*P*63*mc*	4.276	---	6.948	[1]
LiH	*Fm*-3*m*	4.083	---	---	[2]
Al	*Fm*-3*m*	4.049	---	---	[3]
$Li_{0.3}Mg_{0.7}$	*Im*-3*m*	3.517	---	---	[4]
$Mg_{17}Al_{12}$	*I*4-3*m*	10.5438	---	---	[5]
Mg	*P*63/*mmc*	3.2094	---	5.2103	[6]
$Mg_{0.9}Al_{0.1}$	*P*63/*mmc*	3.1692	---	5.1581	[7]
$Mg_{0.5}Al_{0.5}B_2$	*P*6/*mmm*	3.047	---	3.366	[8]
MgO	*Fm*-3*m*	4.217	---	---	[9]
LiAl	*Fd*-3*m*	6.376	---	---	[10]
WC	*P*6-*m*2	2.907	---	2.837	[11]

Table S2. Reflections observed for unknown compounds denoted 1, 2 and 3

Unknown 1*			Unknown 2**			Unknown 3***		
2θ (°)	**d-spacing (Å)**	**Intensity (%)**	**2θ (°)**	**d-spacing (Å)**	**Intensity (%)**	**2θ (°)**	**d-spacing (Å)**	**Intensity (%)**
21.22	4.18	100	21.12	4.20	100	8.91	9.91	88
34.97	2.56	70	35.84	2.50	80	23.51	3.78	100

*Observed in $LiBH_4$-MgH_2-Al (411, **S1**) absorption, scan 32-38 (T = 275-340 °C)
Observed in $LiBH_4$-MgH_2-Al (411, **S1) absorption, scan 125-182 (T = 400-245 °C)
*** Observed in *in situ* SR-PXD desorption and absorption of **S2** and in samples of **S1** and **S2** after three hydrogen release and uptake cycles. Data obtained *after* absorption measurement of **S2** at *RT*.

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