Supporting Information

Table S1. Possible reactions taking place at the various temperatures and the respective H_2 capacities. These reactions are determined according to the Li-Al phase diagram.

Temperature	Reaction	wt.% H ₂
506	$2 \text{ LiH} + 3.23 \text{Li}_{0.126} \text{Al}_{0.874} \rightarrow 5.23 \text{Li}_{0.46} \text{Al}_{0.54} + \text{H}_2$	2.12
540	$2 \text{ LiH} + 3.28 \text{Li}_{0.131} \text{Al}_{0.869} \rightarrow 5.28 \text{Li}_{0.46} \text{Al}_{0.54} + \text{H}_2$	2.10
560	$2 \text{ LiH} + 3.40 \text{Li}_{0.142} \text{Al}_{0.858} \rightarrow 5.40 \text{Li}_{0.46} \text{Al}_{0.54} + \text{H}_2$	2.06
573	$2 \text{ LiH} + 3.42 \text{Li}_{0.144} \text{Al}_{0.856} \rightarrow 5.42 \text{Li}_{0.46} \text{Al}_{0.54} + \text{H}_2$	2.05
585	$2 \text{ LiH} + 3.48 \text{Li}_{0.150} \text{Al}_{0.850} \rightarrow 5.48 \text{Li}_{0.46} \text{Al}_{0.54} + \text{H}_2$	2.03



Figure S1. (A) Hydrogen desorption pressure – composition isotherms for reaction (2) performed at various temperatures above 600 °C. Hydrogen desorption equilibrium data below ~2.5 wt.% desorption has been displayed, and the inset of the figure provides the apparent equilibrium pressures. (B) Kinetic H₂ absorption data performed at 617.8 °C. (C) Desorption PCI followed by an absorption PCI performed at 617.8 °C. The slight hysteresis between the two curves either side of the plateau is the result of slow kinetics.



Figure S2 Powder X-ray diffraction pattern of the Li-Al-H sample after dehydrogenation at 618 $^{\circ}$ C (A) and after rehydrogenation (B).