

Supplementary Information:

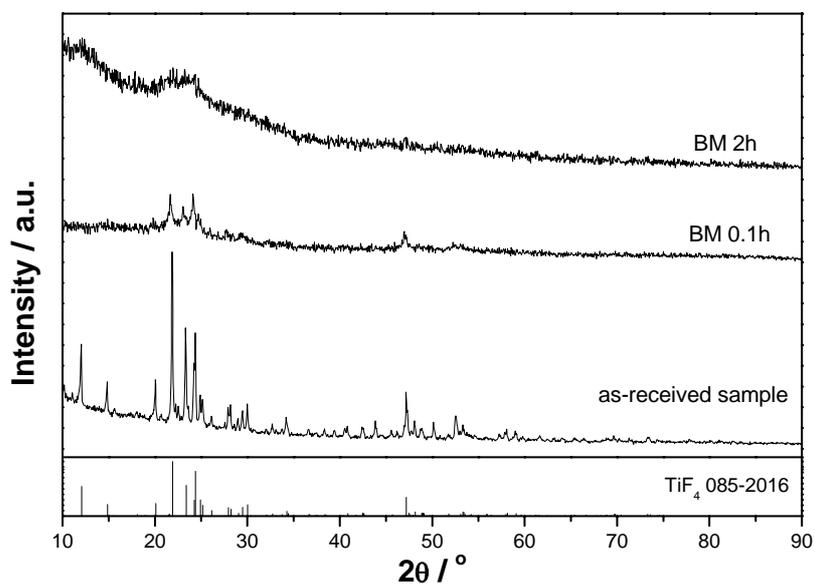


Figure-S1 XRD patterns of TiF₄ with different ball milling durations (After ball milling for 2h, the TiF₄ sample exhibited obviously the amorphous form.)

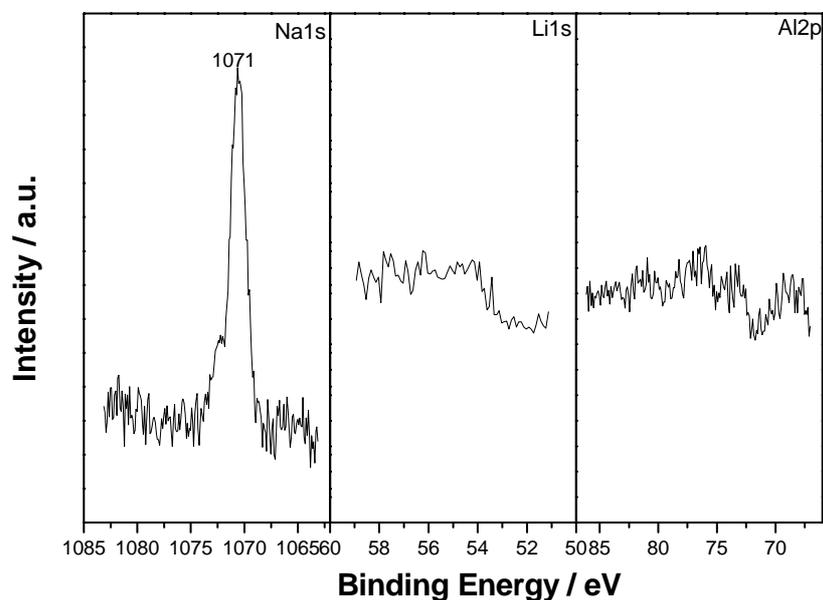


Figure-S2 High-resolution Na 1s, Li 1s and Al 2p XPS spectra of the as-prepared Na₂LiAlH₆ (Only Na signal was detected at the surface of the as-prepared Na₂LiAlH₆.)

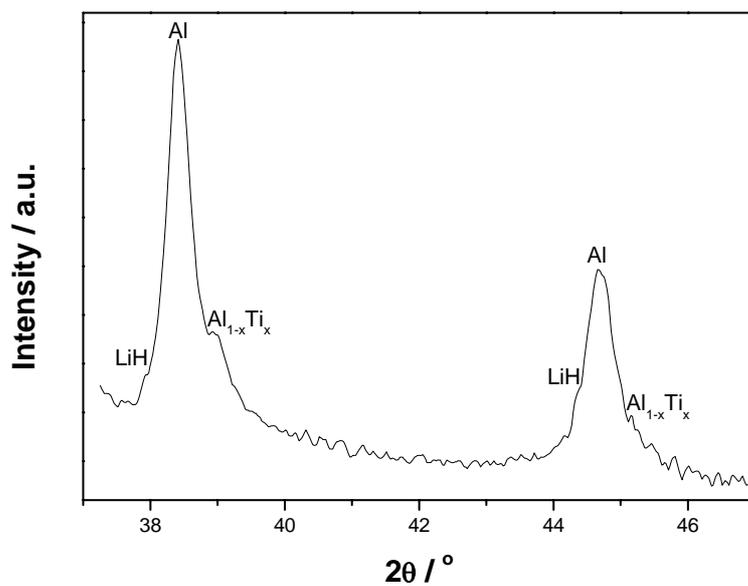


Figure-S3 XRD pattern of the dehydrogenated sample of the 20wt% TiF_4 -added $\text{Na}_2\text{LiAlH}_6$ at $260\text{ }^\circ\text{C}$ ($2\theta = 37 - 47^\circ$)

(It can be clearly seen that the diffraction peaks of Ti-Al solid solution with considerable intensities emerge at the high-angle side of the Al reflections.)