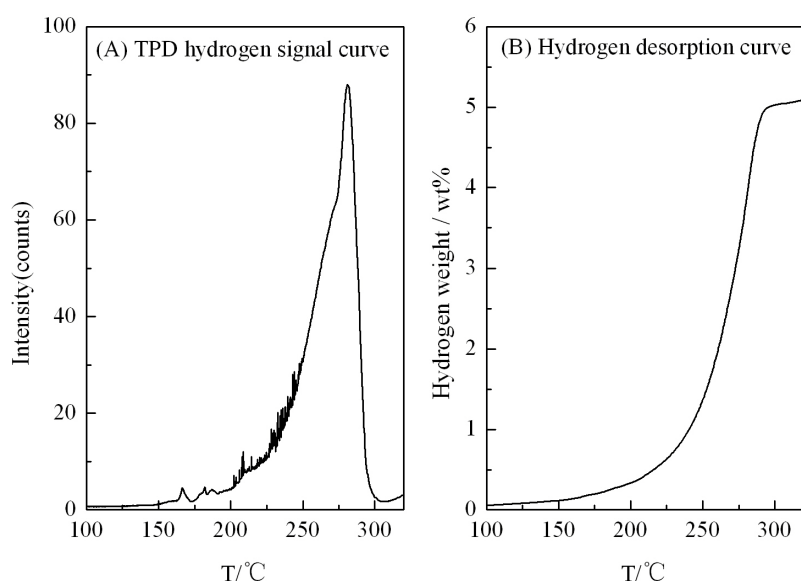
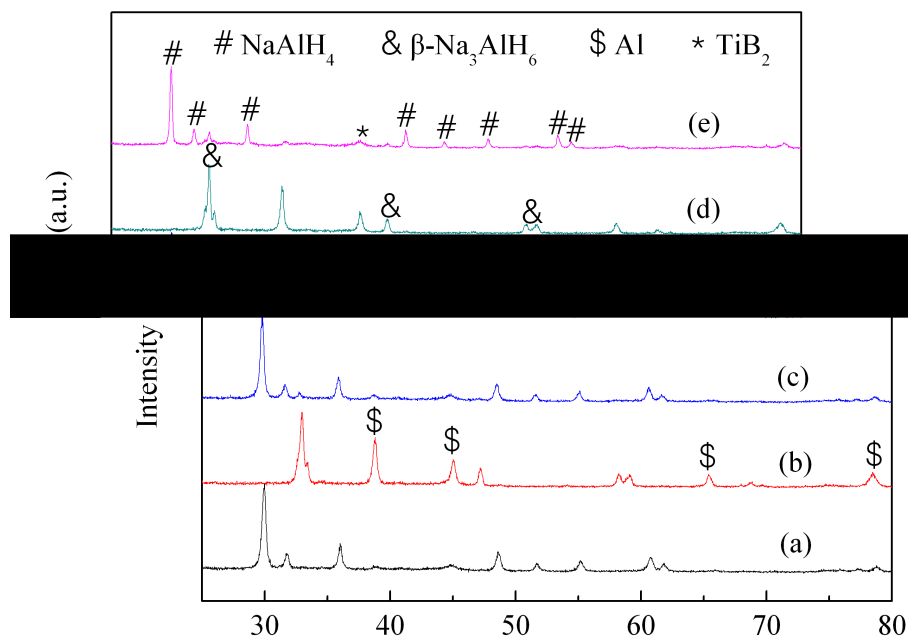


## Supporting Information



**Fig. S1** (A)TPD hydrogen signal of the commercial  $\text{NaAlH}_4$  milled for 70 h, (B) Hydrogen desorption curves of the commercial  $\text{NaAlH}_4$  milled for 70 h. (the heating rate is  $2^\circ\text{C}/\text{min}$ ).



**Fig. S2** XRD patterns of the synthesized  $\text{TiB}_2\text{-NaAlH}_4$  mixture in different experiment stages: (a) as-synthesized  $\text{TiB}_2\text{-NaAlH}_4$  mixture; (b) dehydrogenation of the first cycle; (c) rehydrogenation of the first cycle; (d) dehydrogenation of the second cycle; (e) rehydrogenation of the second cycle. Dehydrogenation:  $90^\circ\text{C}/\text{vacuum}$ ; hydrogenation:  $100^\circ\text{C}/4\text{ MPa}$ .