

Supporting Information

Chemical Compatibility of Solid-State Electrolytes with Hydroflux Cathode-Coating Process

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Supporting Information

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Supplementary Note 1. Supplementary figures

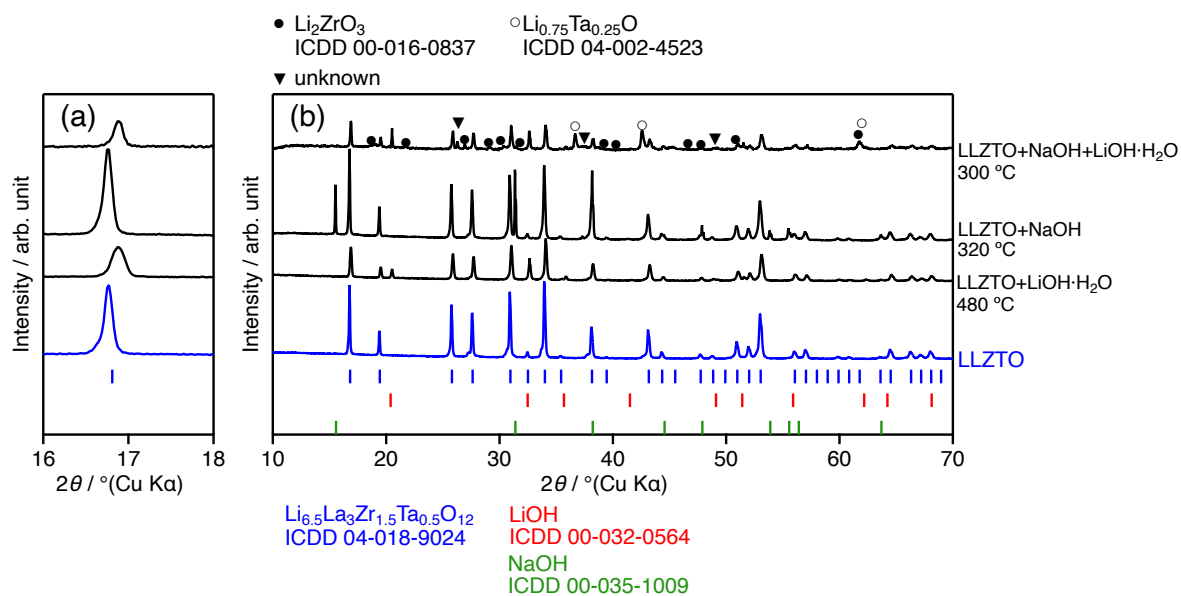


Figure S1. (a) Enlarged profiles of (b) at 16-18°. (b) XRD patterns of mixture of $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ and each alkali hydroxide calcined above their melting points for 12 hours under argon.

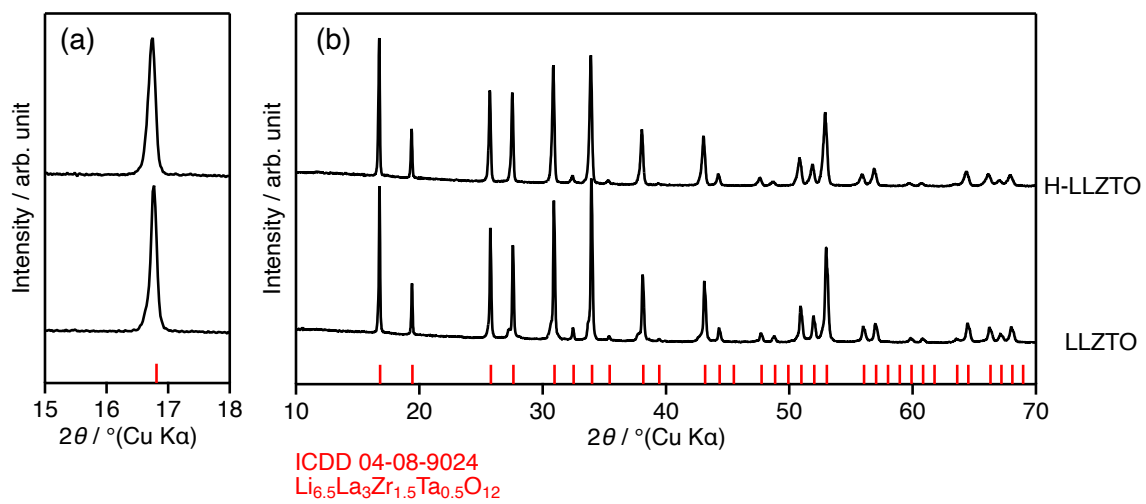


Figure S2. (a) Enlarged profiles of (b) at 16-18 °. (b) XRD patterns of synthesized $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ and $\text{H-Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$.

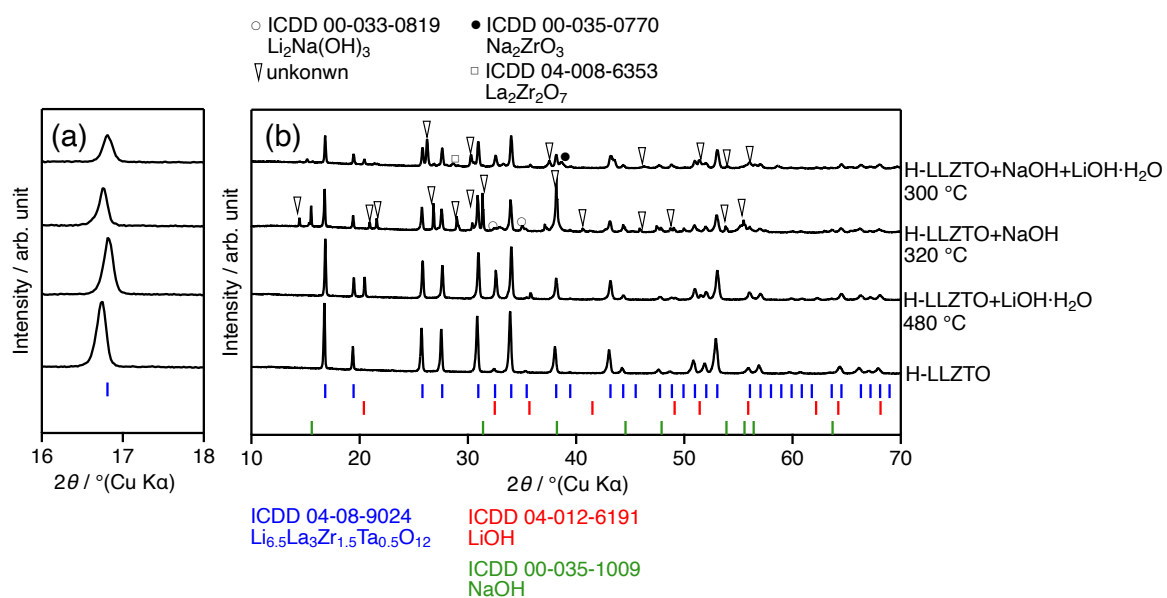


Figure S3. (a) Enlarged profiles of (b) at 16-18 °. (b) XRD patterns of mixture of $\text{Li}_{6.4-x}\text{H}_x\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ and each alkali hydroxide calcined above their melting points for 12 hours under argon.

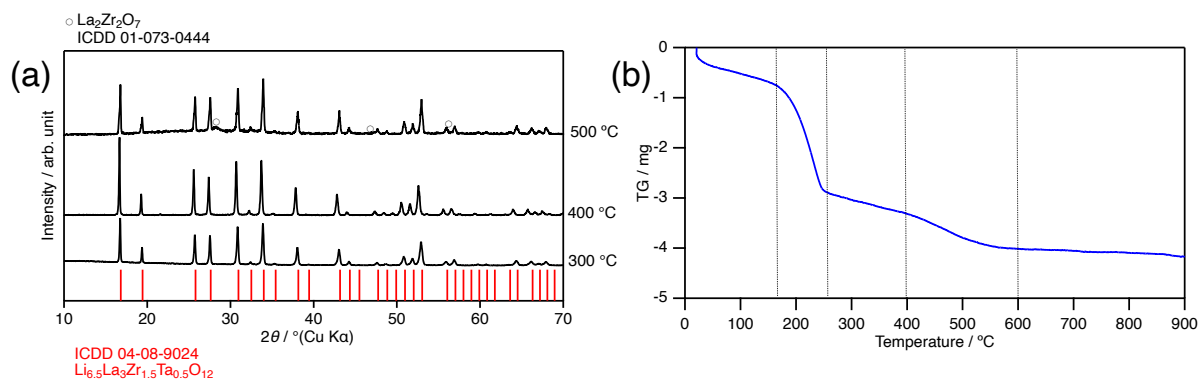


Figure S4. XRD patterns of $\text{H-Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ calcined at 300, 400 and 500 °C for 1 hour under argon. (b) TG curves of synthesized $\text{H-Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ until 900 °C under argon.

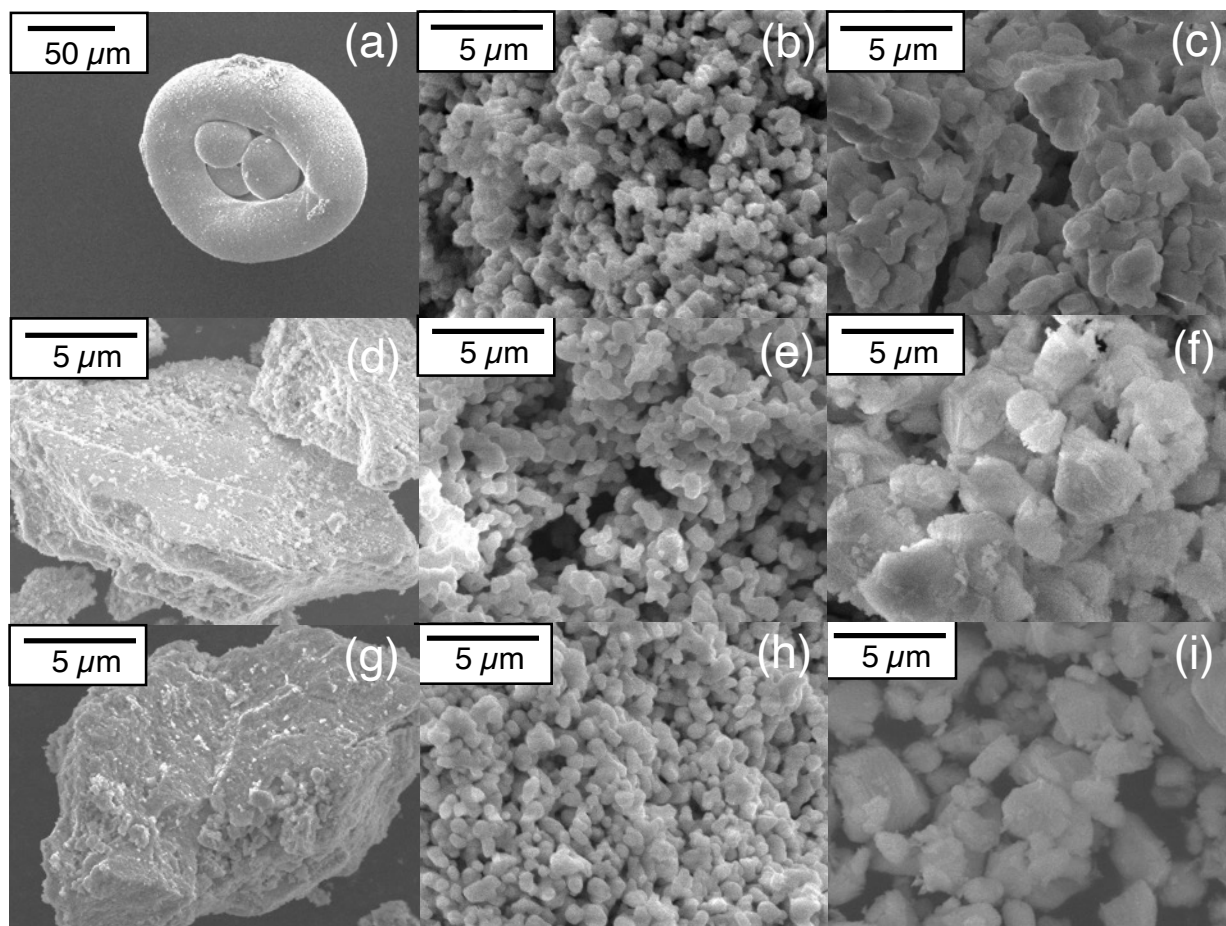


Figure S5. SEM images of commercial materials (a) ZrO_2 , (b) Ta_2O_5 and (c) La_2O_3 . (d),(e) and (f) are SEM images of the mixtures of each transition metal oxide (a,b, and c) with $\text{LiOH}\cdot\text{H}_2\text{O}$ after calcination at $480\ ^\circ\text{C}$ and washing respectively. (g), (h) and (i) are also SEM images of the mixtures of each transition metal oxide (a,b, and c) with NaOH after calcination at $320\ ^\circ\text{C}$ and washing respectively.

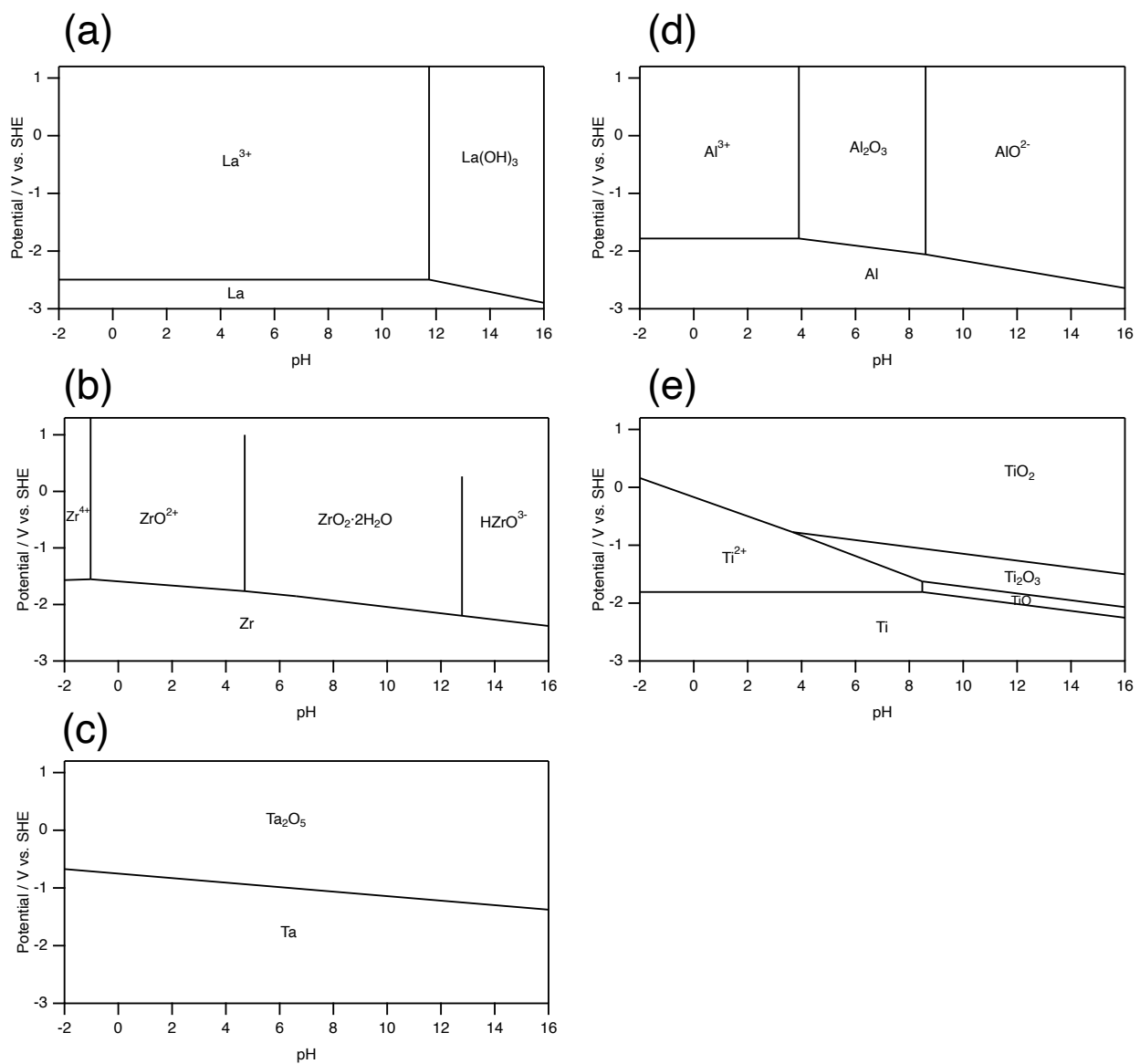


Figure S6. Pourbaix diagram of each element of the (a) La, (b) Zr, (c) Ta, (d) Al and (e) Ta respectively.¹

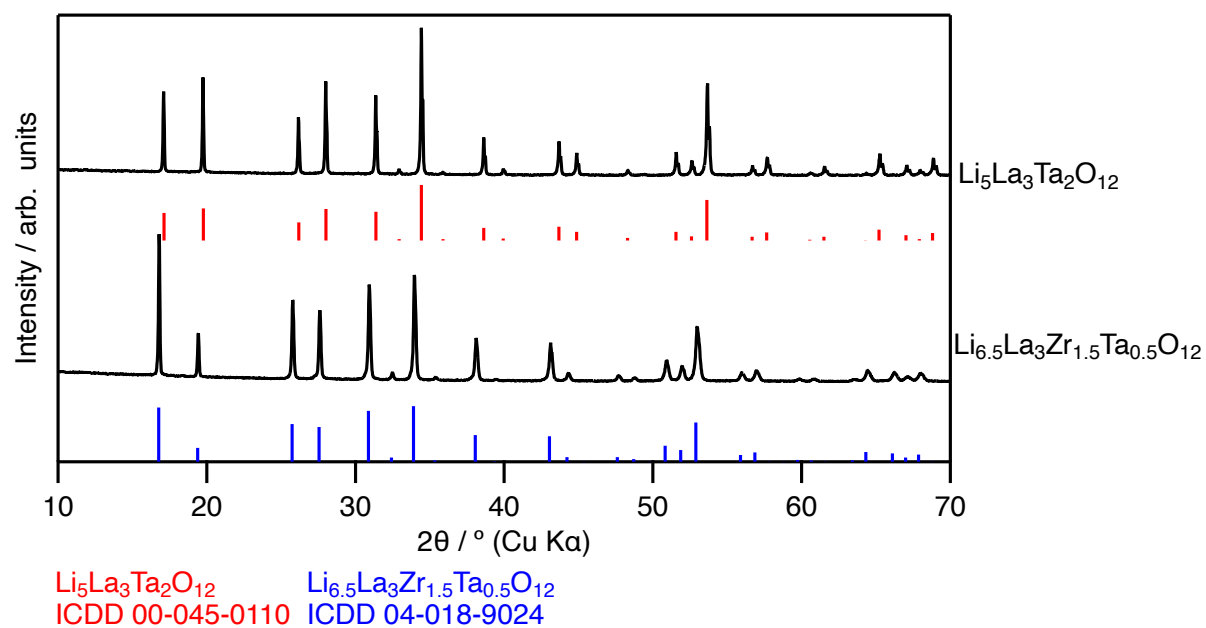


Figure S7. XRD patterns of $\text{Li}_5\text{La}_3\text{Ta}_2\text{O}_{12}$ obtained by substituting all Zr site for Ta.

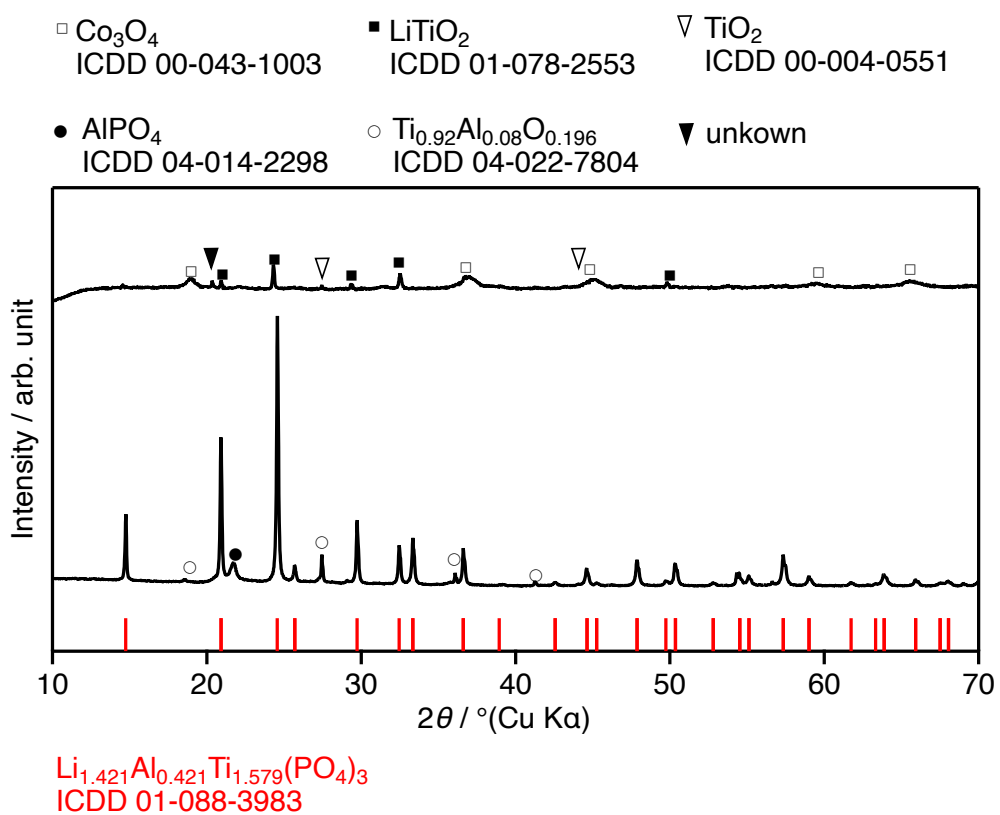


Figure S8. XRD patterns of LATP and LiCoO_2 precursors calcined at 300°C for 12 hours under O_2 flow.

The thermal stability of $\text{Li}_{6.4-x}\text{H}_x\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ by itself is investigated to evaluate the effect of water on $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$. (Figure S4.) All peaks shifted towards a lower angle and the FWHM increased due to the reaction with water. (Figure S4.) This increase in the lattice parameter is attributed to the replacement of stronger Li-O bonds by weaker O-H bonds²⁻⁵. The decrease in crystallinity is induced in elemental rearrangements due to the H^+/Li^+ exchange⁶. The H^+ desorption temperature from $\text{Li}_{6.4-x}\text{H}_x\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ is identified by Thermogravimetry-Differential Thermal Analysis (TG-DTA). (Figure S4 (b).) In the TG curve, two steps of weight loss are observed at 200 °C and 500 °C. XRD patterns of $\text{Li}_{6.4-x}\text{H}_x\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ are shown calcined at 300, 400 and 500 °C for 1 hour in Figure S6. The single phase of $\text{Li}_{6.4-x}\text{H}_x\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ is observed until 400 °C. At 500 °C, an increase in background associated with the amorphisation and the decomposition product $\text{La}_2\text{Zr}_2\text{O}_7$ phase are observed. Heat treatment induces decomposition of the metastable phase $\text{Li}_{6.4-x}\text{H}_x\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$, due to the release of H_2O from the structure and the charge imbalance.

Supplementary Note 3. Supporting references

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