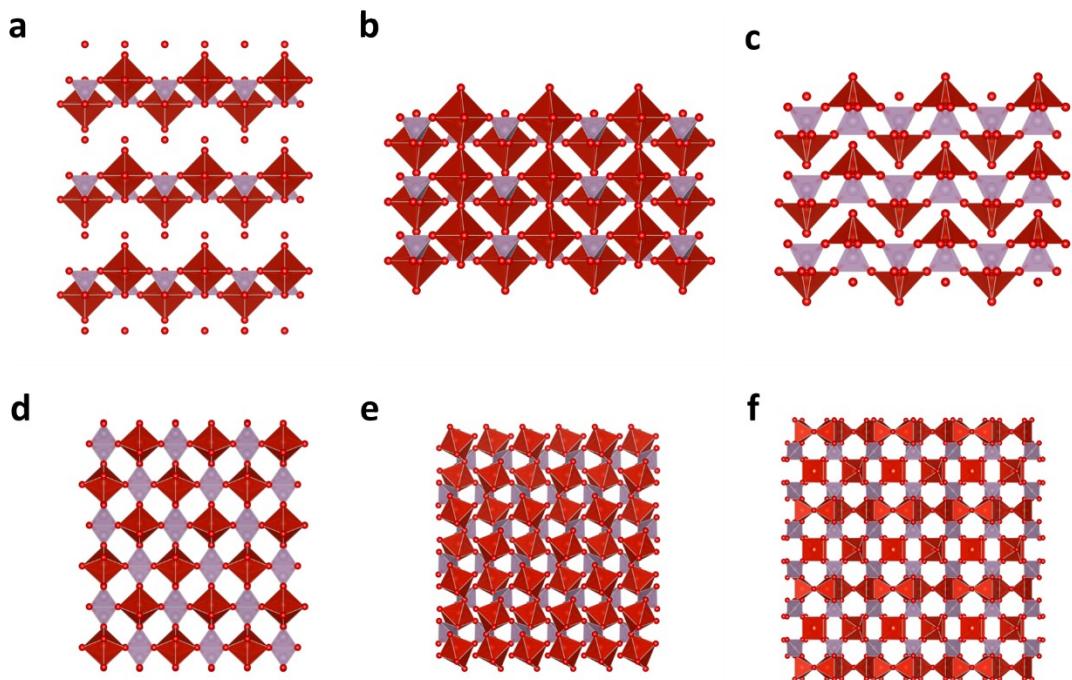
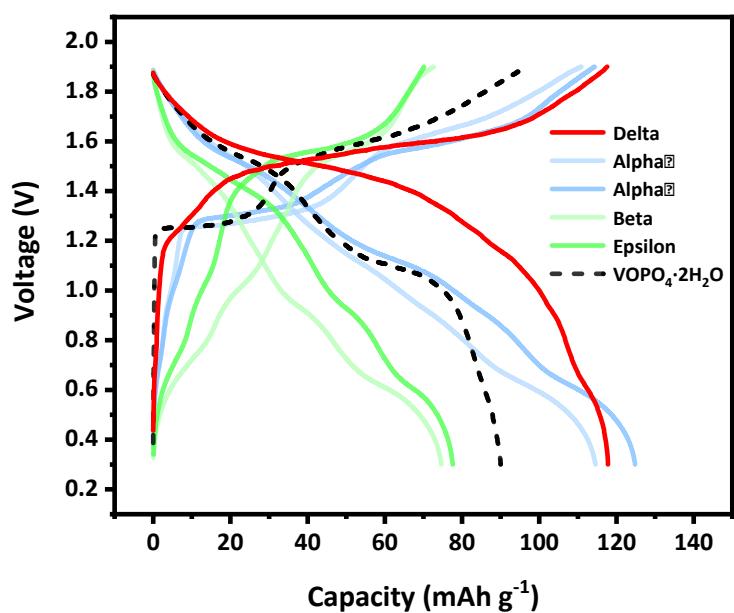


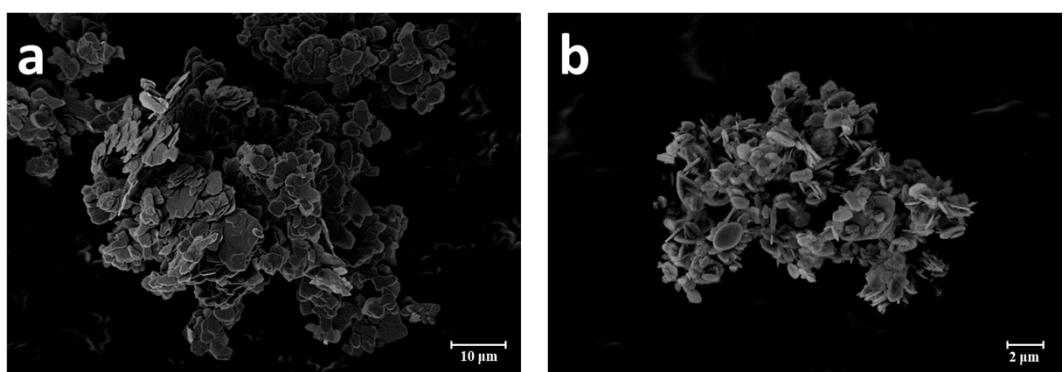
## Supporting Information



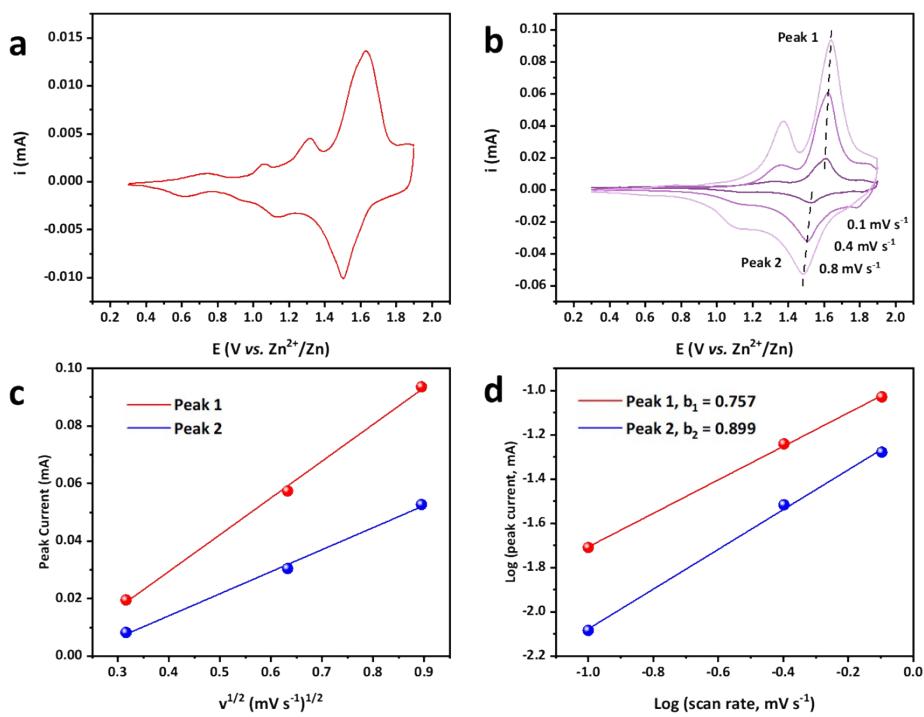
**Fig. S1** The structural models of (a) VOPO<sub>4</sub>·2H<sub>2</sub>O, (b)  $\alpha$ I, (c)  $\alpha$ II, (d)  $\beta$ , (e)  $\varepsilon$  and (f)  $\delta$ -VOPO<sub>4</sub>.



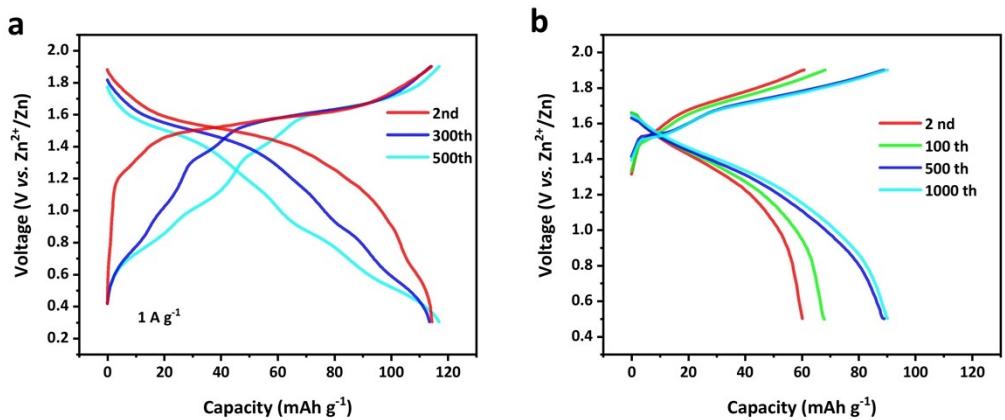
**Fig. S2** The GCD curves of VOPO<sub>4</sub>·2H<sub>2</sub>O,  $\alpha$ I,  $\alpha$ II,  $\beta$ ,  $\varepsilon$  and  $\delta$ -VOPO<sub>4</sub>.



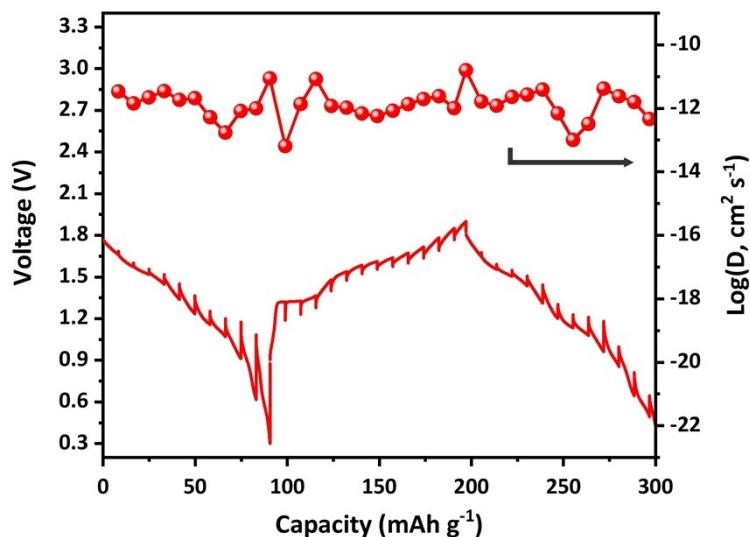
**Fig. S3** The SEM images of  $\text{VOPO}_4 \cdot 2\text{H}_2\text{O}$  and  $\text{VOHPO}_4 \cdot 0.5\text{H}_2\text{O}$ .



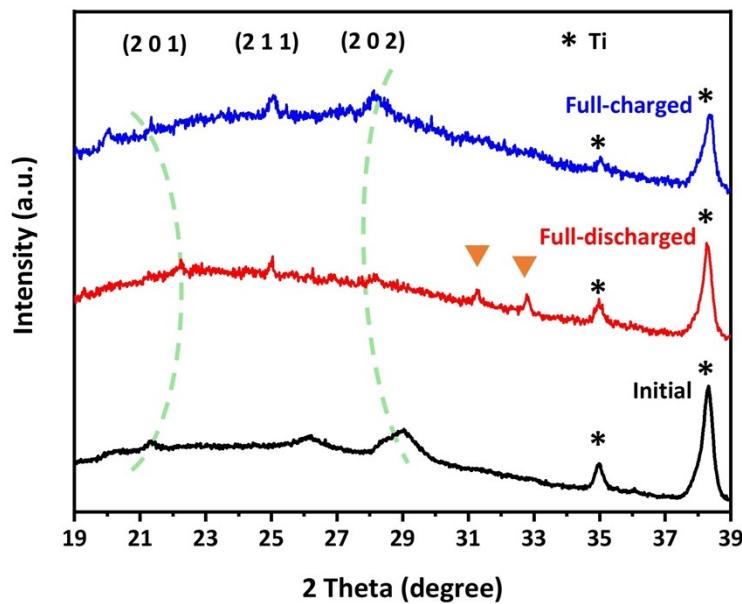
**Fig. S4** CV curves of  $\delta$ -VOPO<sub>4</sub> at the scan rate of  $0.1 \text{ mV s}^{-1}$ ; (b) CV curves of  $\delta$ -VOPO<sub>4</sub> at different scan rate; (c) Calculated slope parameters for the dependence of the current density of peaks currents and the square root of the scan rates; (d) Calculated slope parameters for the dependence of the current density of log (peak current) and the log (scan rate).



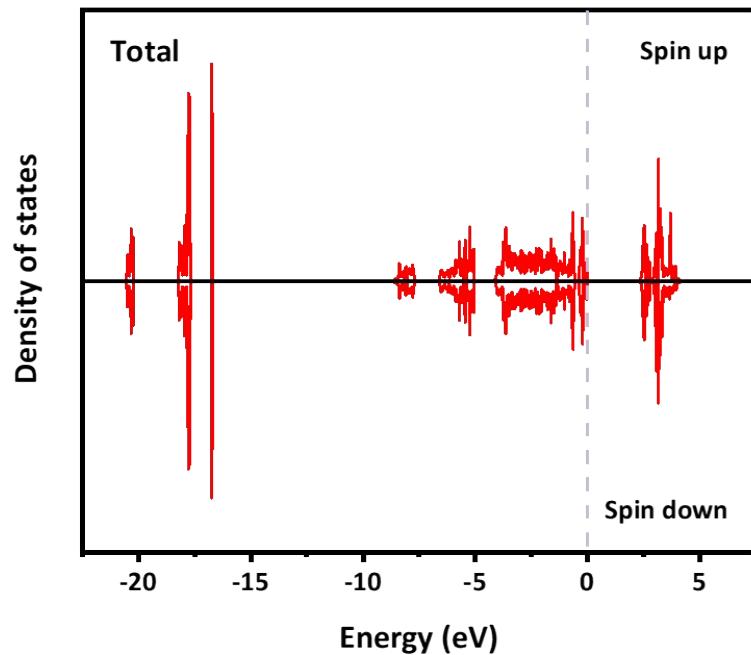
**Fig. S5** The typical charge-discharge curves of  $\delta$ -VOPO<sub>4</sub> at (a)  $1 \text{ A g}^{-1}$  and (b)  $10 \text{ A g}^{-1}$ .



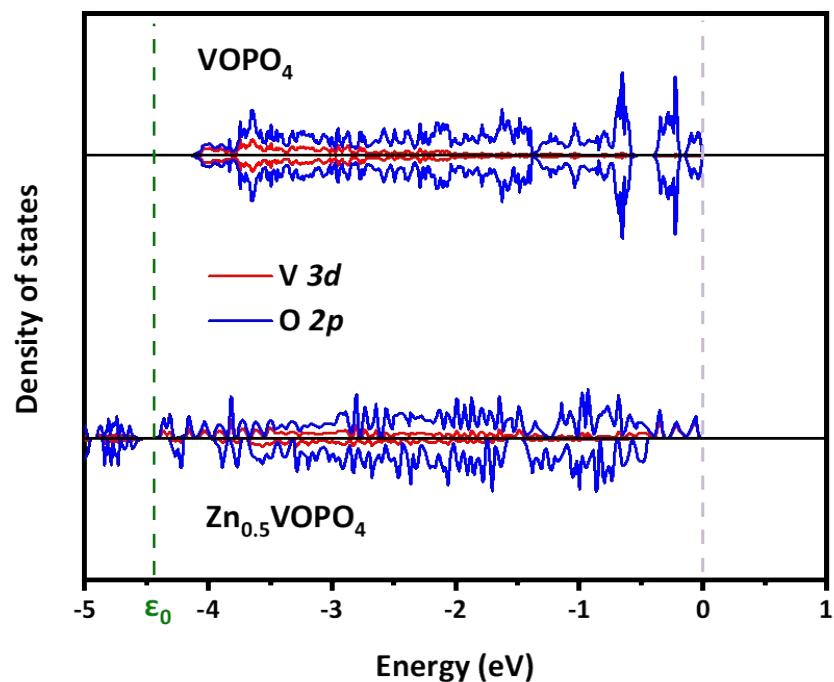
**Fig. S6** The GITT profiles of  $\delta$ -VOPO<sub>4</sub> and the calculated Zn<sup>2+</sup> ion diffusion coefficients.



**Fig. S7** *Ex-situ* XRD patterns of  $\delta$ -VOPO<sub>4</sub> after the first discharge and charge.



**Fig. S8** The total density of states of  $\delta$ -VOPO<sub>4</sub>.



**Fig. S9** The density of states of  $\delta\text{-VOPO}_4$  from -4.45 eV ( $\epsilon_0$ ) to 0 eV (the Fermi energy level).