Supporting Information

Understanding Thickness-Dependent Transport Kinetics in Nanosheet-Based Battery Electrodes

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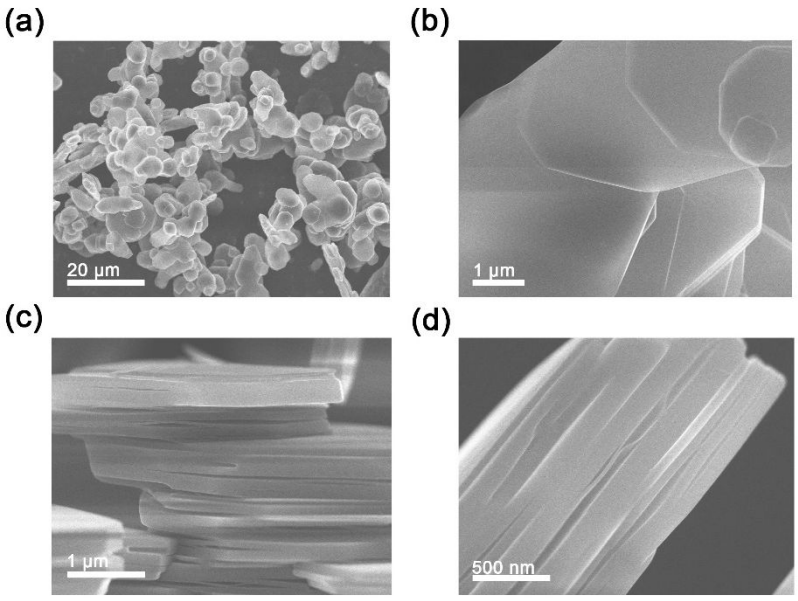

Figure S1. (a, b) Top view SEM images of bulk $VOPO_4 \cdot 2H_2O$. (c, d) Cross-section view SEM images of bulk $VOPO_4 \cdot 2H_2O$.

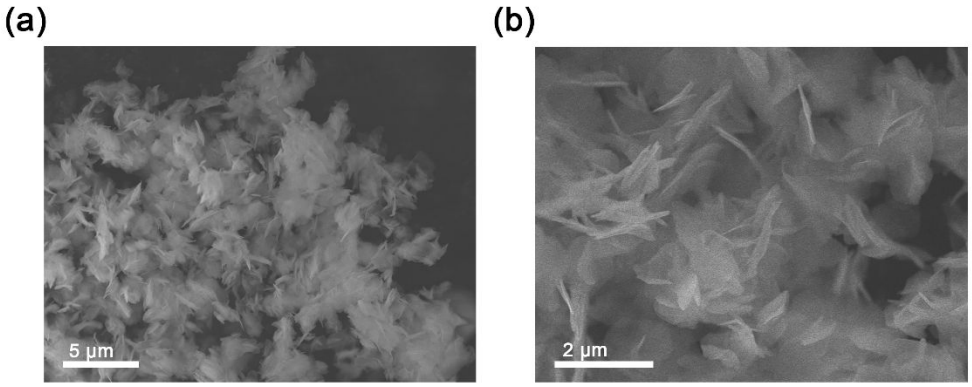

Figure S2. SEM images of exfoliated $VOPO_4$ nanosheets.

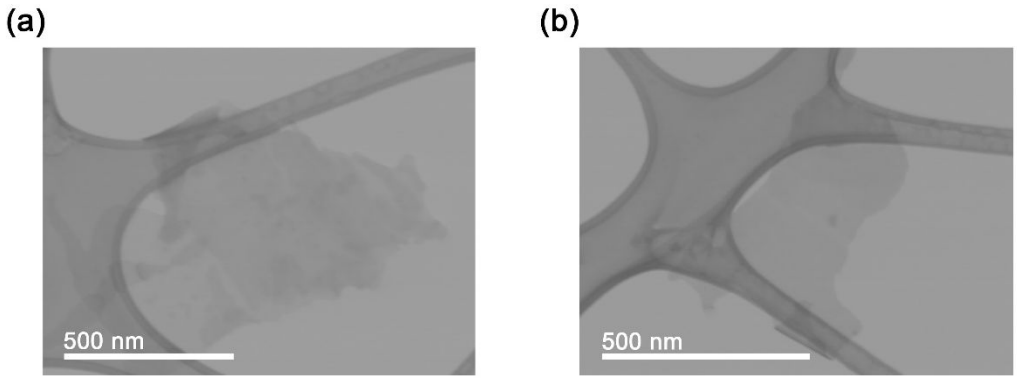

Figure S3. STEM images of exfoliated $VOPO_4$ nanosheets.

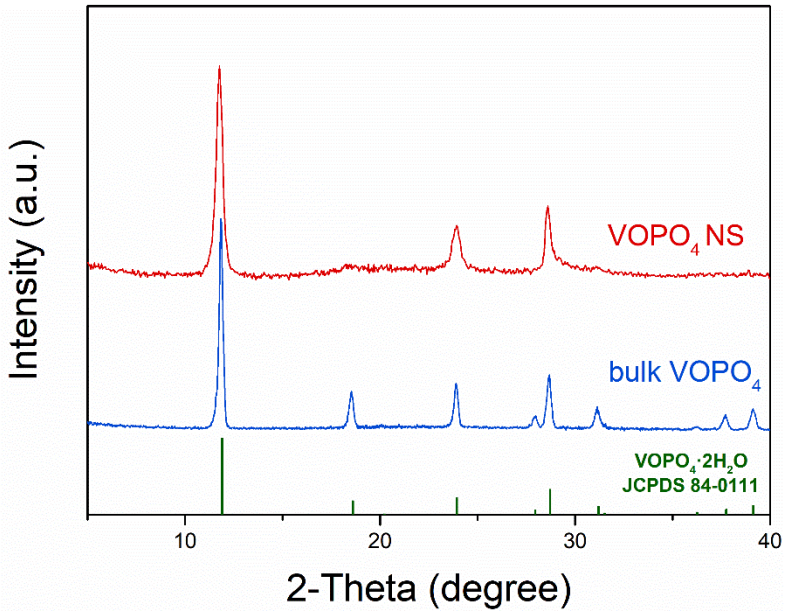

Figure S4. XRD patterns of bulk $VOPO_4$ (blue line) and exfoliated $VOPO_4$ nanosheets (red line).

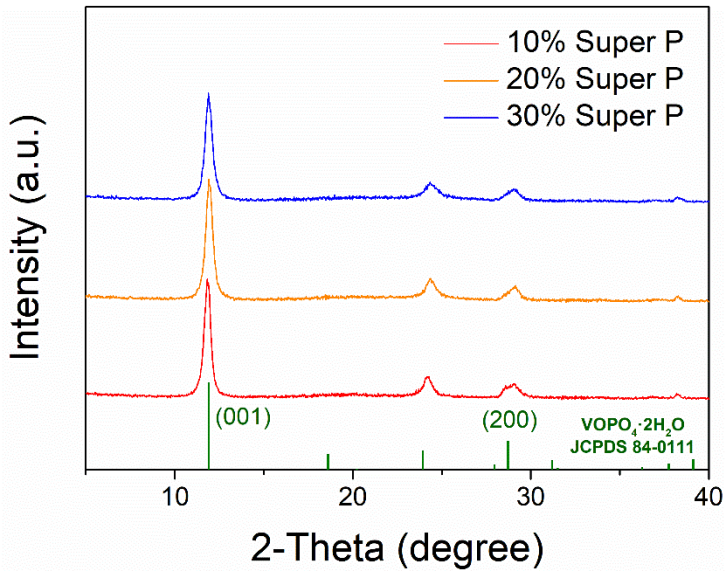

Figure S5. XRD patterns of drop-casted $VOPO_4$ electrodes with different contents of Super P as conductive additive. (Red line: 10 wt.%, orange line: 20 wt.% and blue line: 30 wt.%)

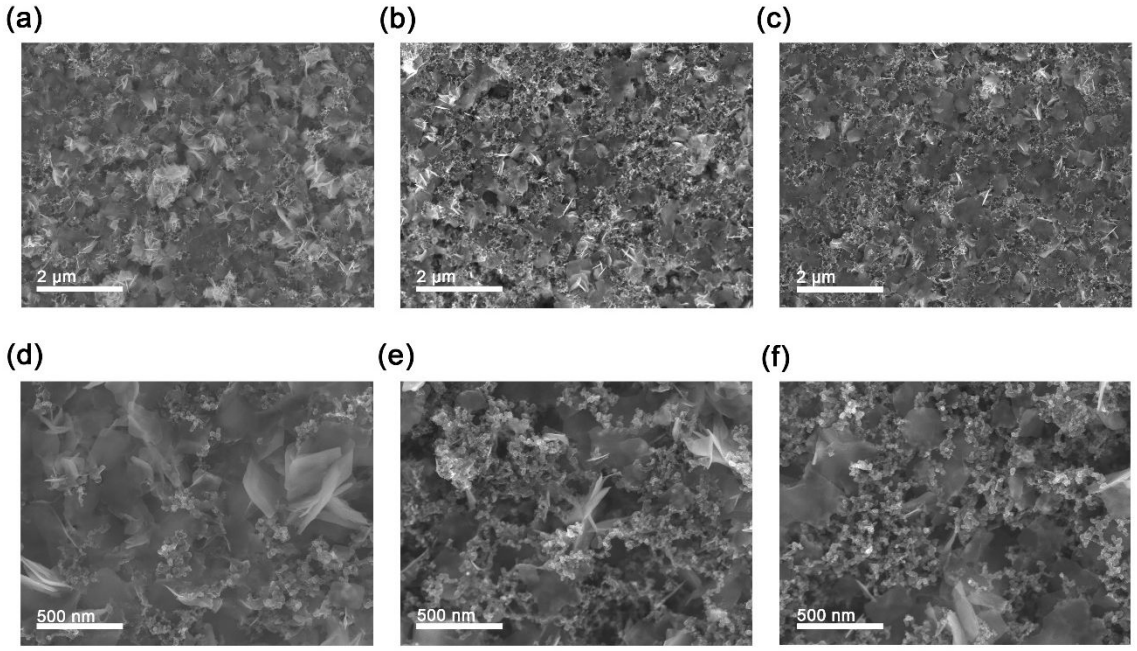

Figure S6. SEM images of drop-casted $VOPO_4$/Super P electrodes. (a, d) SEM images of electrodes with 10 wt.% Super P. (b, e) SEM images of electrodes with 20 wt.% Super P. (c, f) SEM images of electrodes with 30 wt.% Super P.

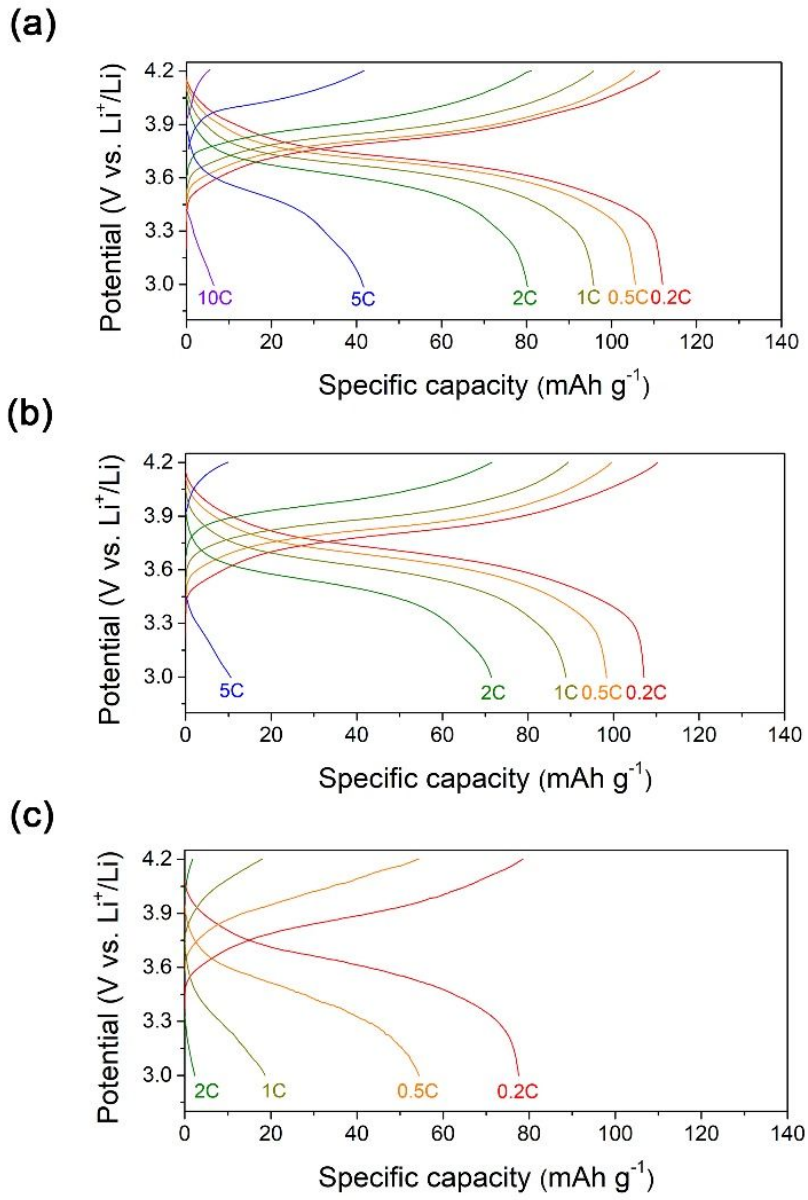

Figure S7. Galvanostatic charge-discharge curves of drop-casted $VOPO_4$ electrodes with 10 wt.% Super P. Areal mass loading: (a) 1 mg cm^{-2}, (b) 5 mg cm^{-2} and (c) 10 mg cm^{-2}.

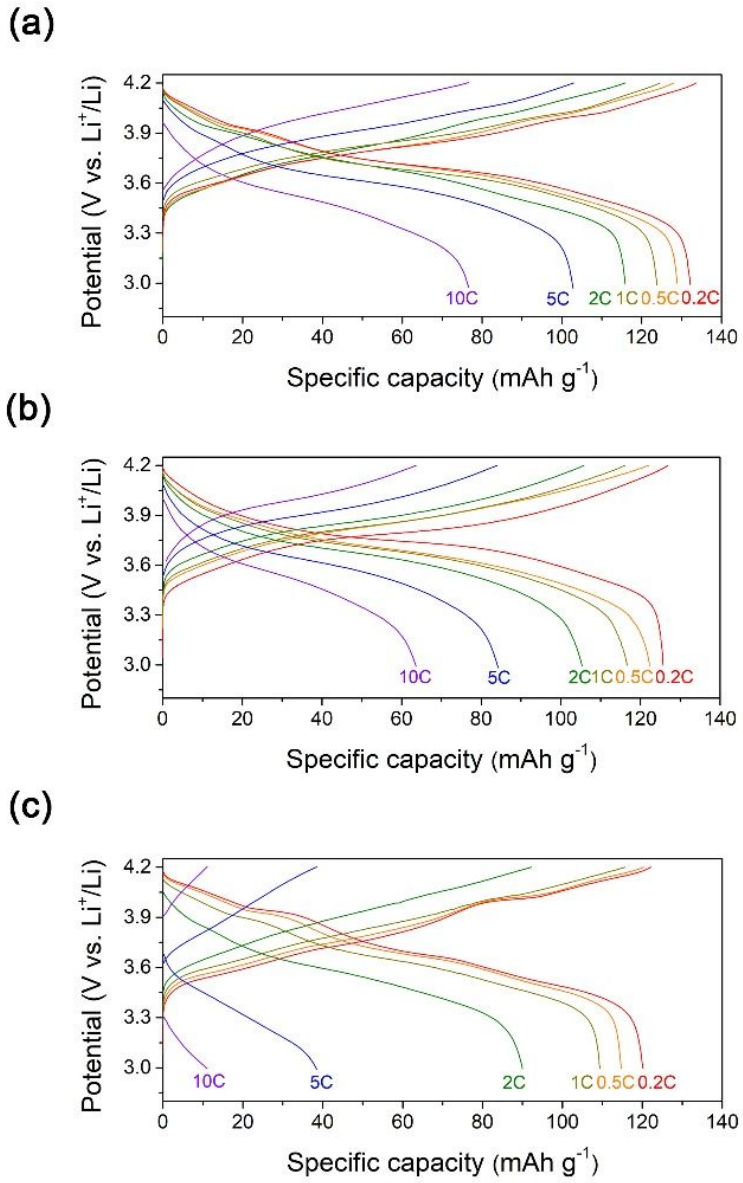

Figure S8. Galvanostatic charge-discharge curves of drop-casted $VOPO_4$ electrodes with 20 wt.% Super P. Areal mass loading: (a) 1 mg cm^{-2}, (b) 5 mg cm^{-2} and (c) 10 mg cm^{-2}.

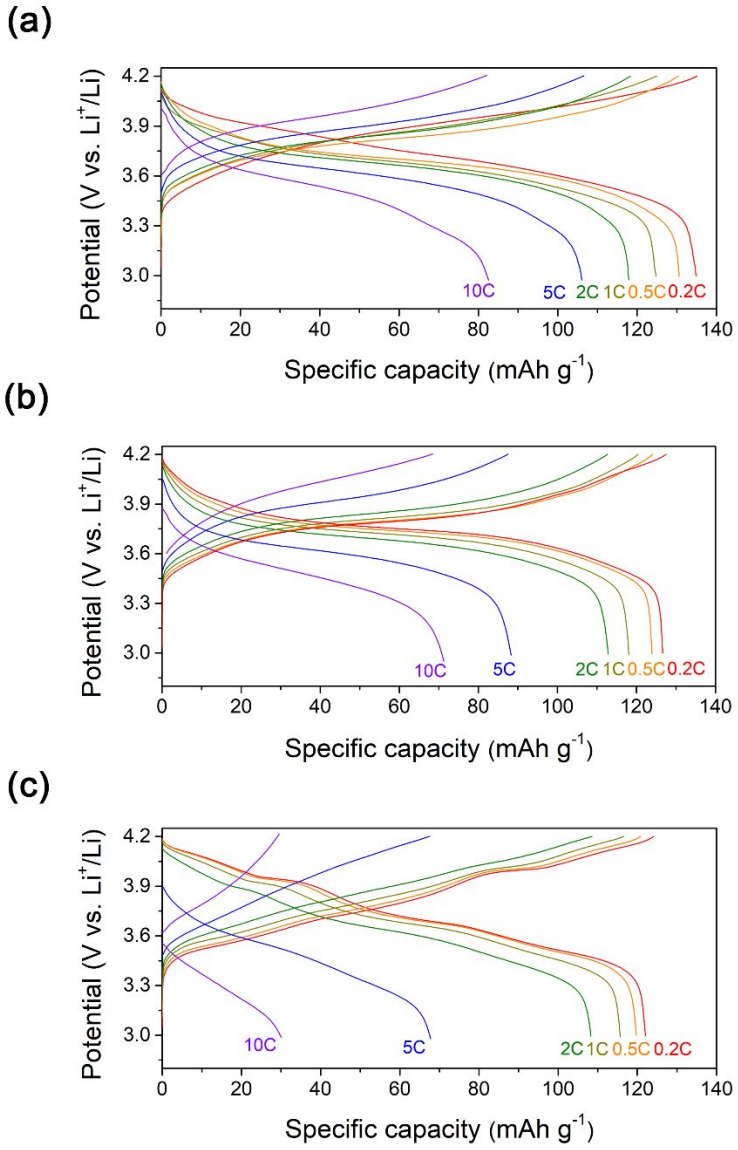

Figure S9. Galvanostatic charge-discharge curves of drop-casted $VOPO_4$ electrodes with 30 wt.% Super P. Areal mass loading: (a) 1 mg cm^{-2}, (b) 5 mg cm^{-2} and (c) 10 mg cm^{-2}.

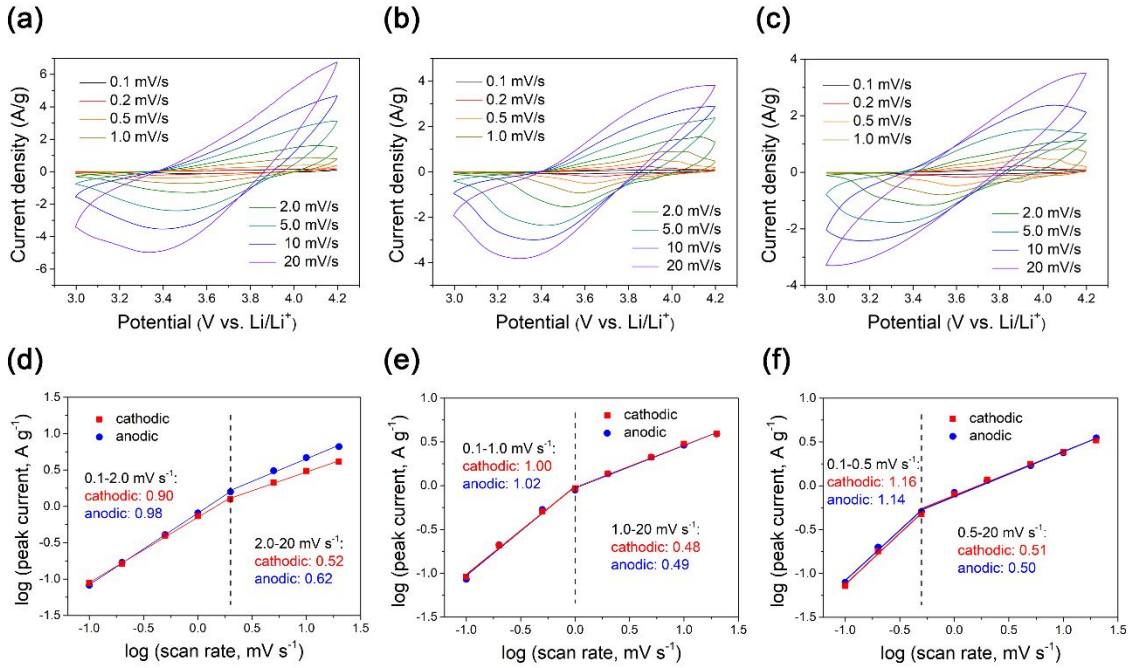

Figure S10. Cyclic voltammetric curves at various scan rates from 0.1 to 20 mV s^{-1} and corresponding b value determination. Areal mass loading: (a, d) 1 mg cm^{-2}, (b, e) 5 mg cm^{-2} and (c, f) 10 mg cm^{-2}.

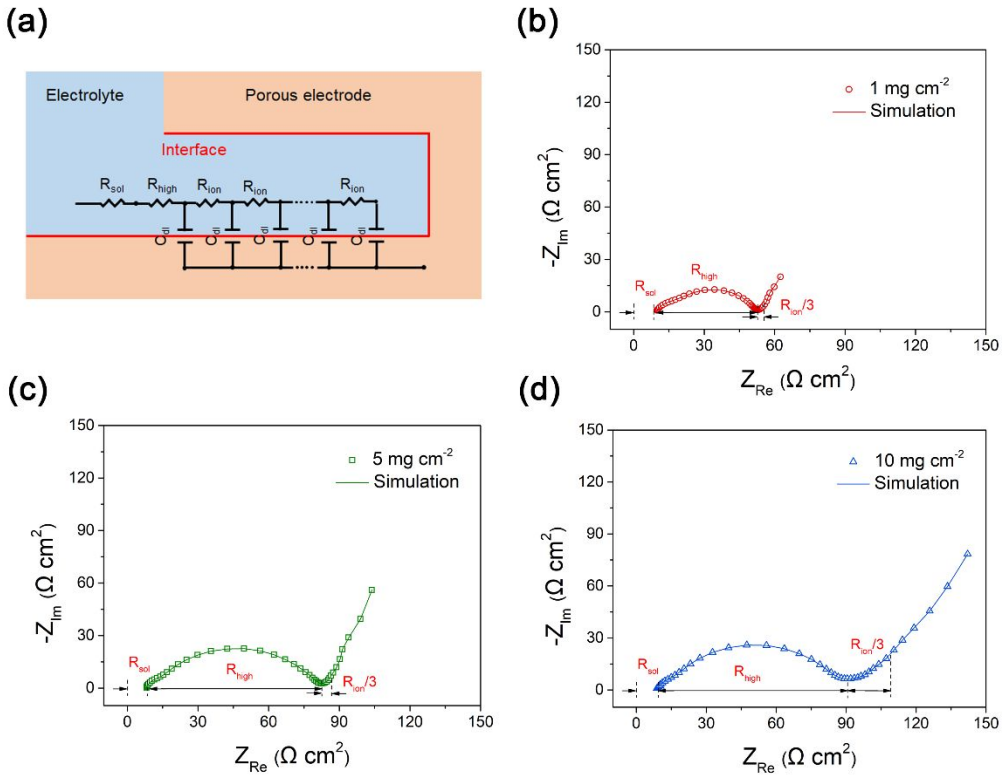

Figure S11. (a) Transmission line equivalent circuit of porous electrodes at 0% state of charge (SOC). Nyquist plots characterized by using a symmetric cell with two identical electrodes at 0% SOC. Areal mass loading: (b) 1 mg cm^{-2}, (c) 5 mg cm^{-2} and (d) 10 mg cm^{-2}.

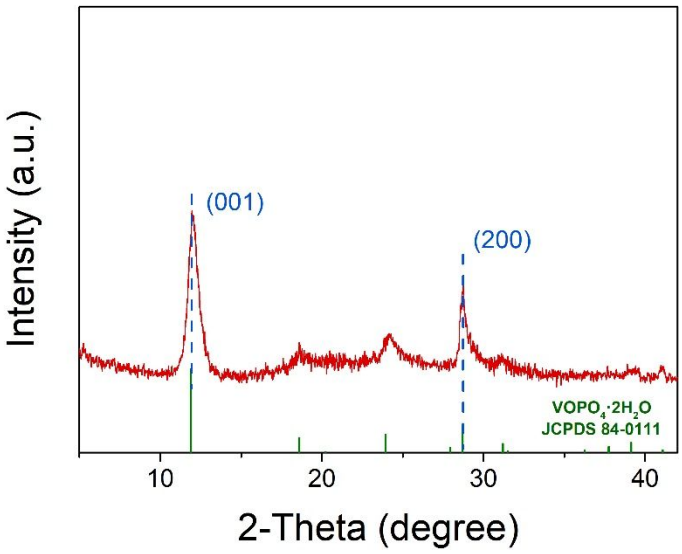

Figure S12. XRD pattern of an ice-templated $VOPO_4$ electrode.

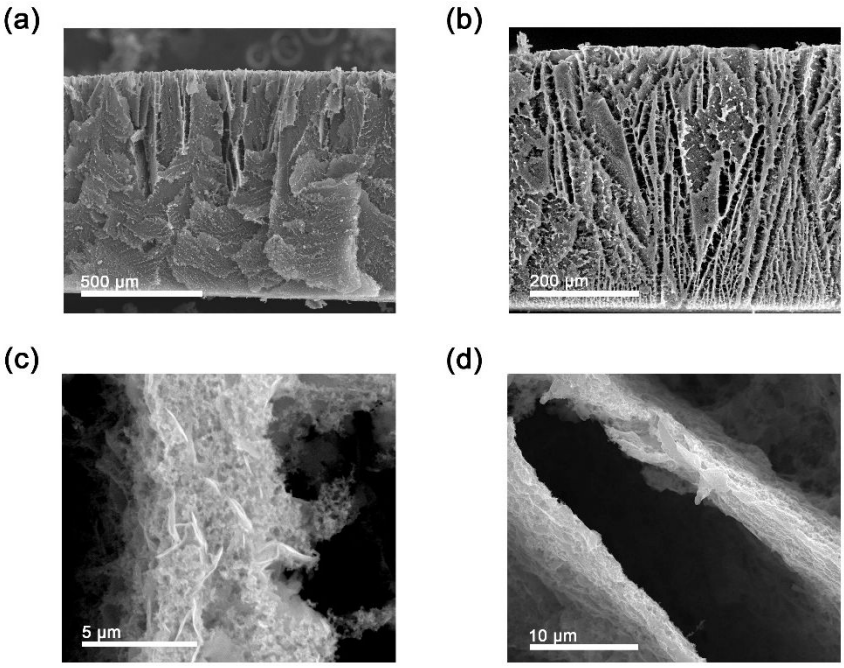

Figure S13. Cross-section SEM images of ice-templated $VOPO_4$ electrodes with areal mass loading: (a) 10 mg cm^{-2} and (b) 5 mg cm^{-2} and corresponding electrode thickness is approximate 800 µm and 400 µm, respectively. Zoom-in cross-section SEM image (c) and top-view image (d).

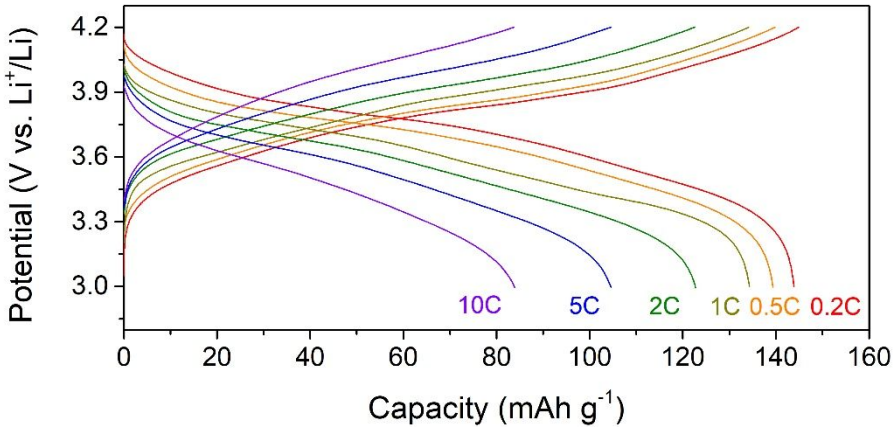

Figure S14. Galvanostatic charge-discharge curves of ice-templated $VOPO_4$ electrodes with areal mass loading: 10 mg cm^{-2}.

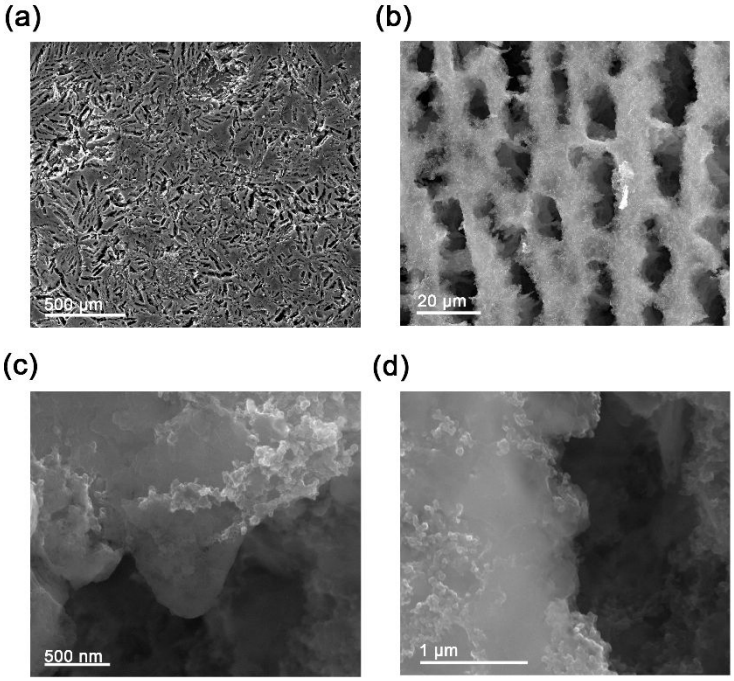

Figure S15. Morphologies of ice-templated $VOPO_4$ electrodes after cycling. (a, c) Top view SEM images and (b, d) cross-section view SEM images of the electrodes.

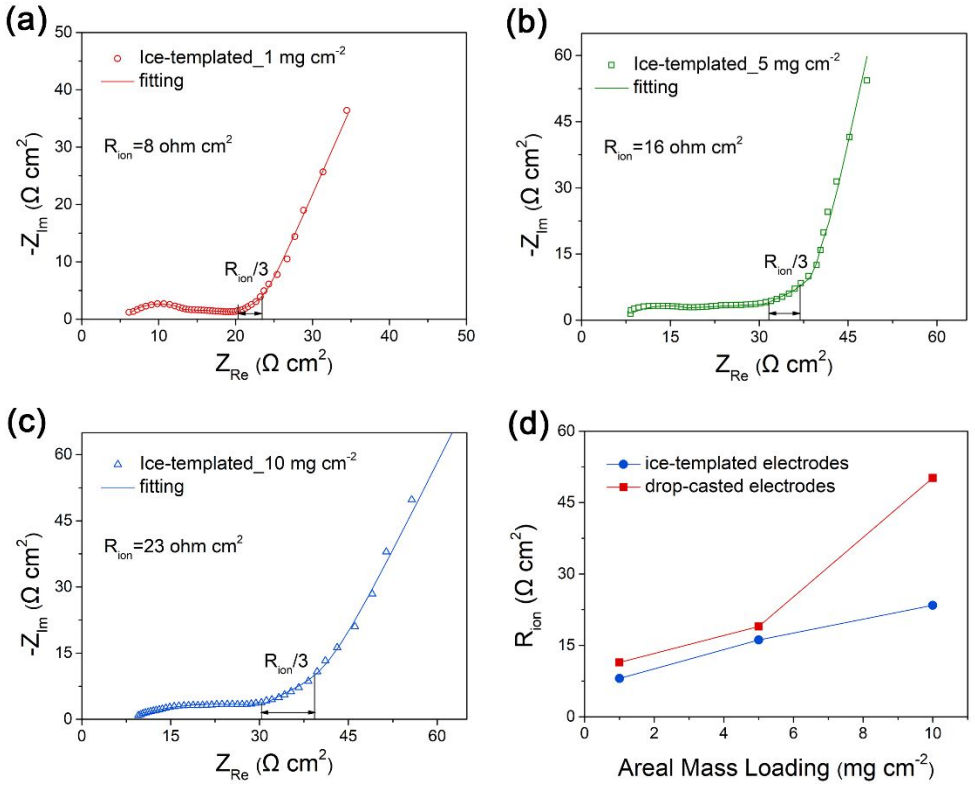

Figure S16. Simulated Nyquist plots of ice-templated electrodes characterized by using a symmetric cell with two identical electrodes at 0% SOC. Areal mass loading: (a) 1 mg cm^{-2}, (b) 5 mg cm^{-2} and (c) 10 mg cm^{-2}. (d) R_{ion} comparison between ice-templated and drop-casted electrodes with various areal mass loadings.

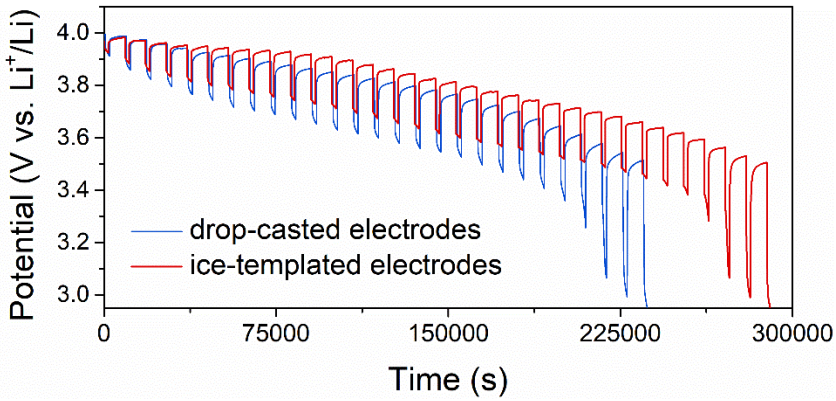

Figure S17. Voltage curves (1st discharge) for GITT cycles of drop-casted and ice-templated electrodes. Areal mass loading of drop-casted and ice-templated electrodes: 10 mg cm^{-2}.

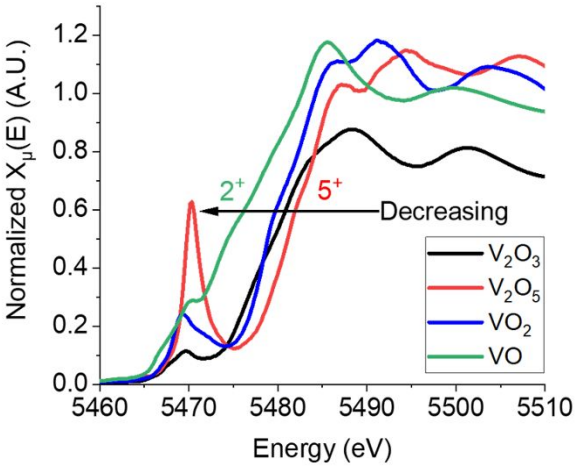

Figure S18. XANES spectra for the vanadium 5^+, 4^+, 3^+, and 2^+ reference materials.

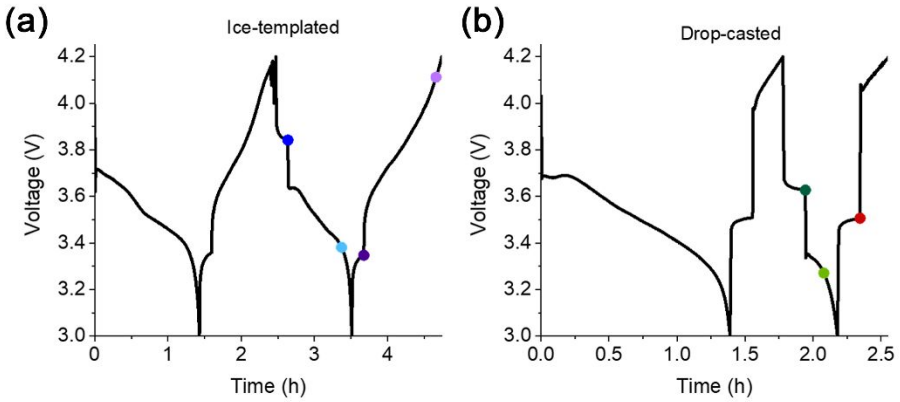

Figure S19. First and second discharge and charge voltage profiles as a function of time for in situ cells prepared using the (a) ice-templated and (b) drop-casted electrodes. Areal mass loading of drop-casted and ice-templated electrodes: 10 mg cm^{-2}.

Table S1. Charge-transfer resistance determined for half-cells with various mass loadings.

Areal mass loading (mg cm^{-2})	R_{ct} (Ω)	$R_{ct,A}$ (Ω cm^2)
1	1589	8.98
5	1246	35.2
10	1067	60.3