Supporting Information

Layer-by-layer Stacked $(NH_4)_2V_4O_9 \cdot 0.5H_2O$ Nanosheet Assemblies with Intercalation Pseudocapacitance for High Rate Aqueous Zinc-Ion Storage

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Extended data

Supplementary Note 1: Electrochemical kinetics analyses of NVO cathode.

Sweep voltammetry at varied scan rates from 0.1 to 1 mV s^{-1} was conducted to investigate the electrochemical kinetics origin. Generally, the peak current (i) has a relationship with the scan rate (v) that can be described as below :

$$i = av^b$$

$$\log(i) = b \times \log(v) + \log(a)$$

Where the slope *b* could be calculated according to the log(i) versus log(v) plots, always in a range between 0.5 to 1. When *b* = 0.5, the capacity is considered to be totally provided by the diffusion-controlled process, corresponding to a faradaic insertion/extraction reaction. While *b* = 1 means a surface-controlled capacitive process.

Additionally, the capacitive contributions at varied scan rates can be calculated by the following equation:

$$i = k_1 v + k_2 v^{1/2}$$

$$i / v^{1/2} = k_1 v^{1/2} + k_2$$

Where $k_1 v$ and $k_2 v^{1/2}$ represent the capacitive and diffusion-controlled contribution,

respectively.

S**upplementary Note 2**: Discussion of the Galvanostatic Intermittent Titration Technique (GITT) measurement.

The GITT was employed to in-depth analyze the solid-state diffusion kinetics of the electrode, where the solid-state diffusion coefficient of Zn^{2+} was obtained based on the following equation:

$$D_{Zn^{2+}}^{GITT} = \frac{4}{\pi\tau}\left(\frac{n_M V_M}{S}\right)^2\left(\frac{\Delta E_S}{\Delta E_t}\right)^2$$

Where τ is the constant current pulse time (s); n_M and V_M are the moles (mol) and molar volume ($cm^3\ mol^{-1}$) of active material, respectively; S is the electrode/electrolyte contact area (cm^2); ΔE_s is the steady-state potential change (V) due to the current pulse; ΔE_t is the potential change (V) during the constant current pulse after eliminating the iR drop. In our GITT study, a cell was charged or discharged at the current density of 50 mAh g^{-1} for 20 min, followed by a 1 h open circuit step to allow relaxation back to equilibrium, as depicted in Figure N2.

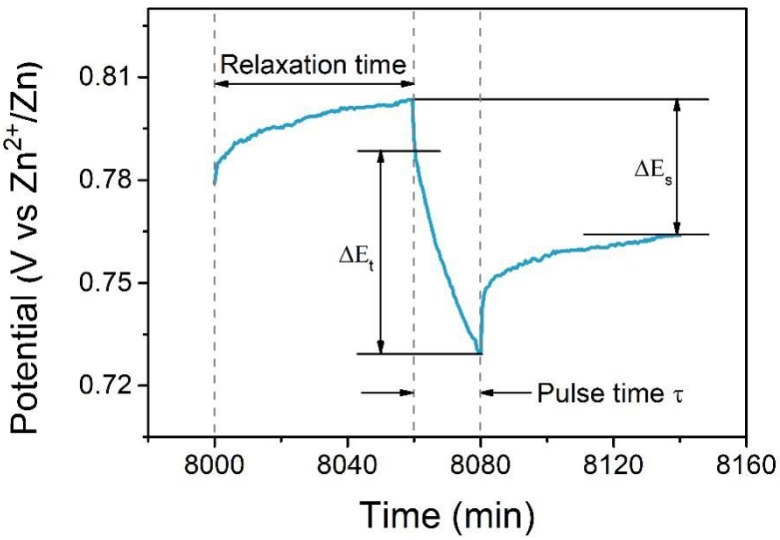

Figure N2. Schematic illustration of a single step of the GITT during discharge process.

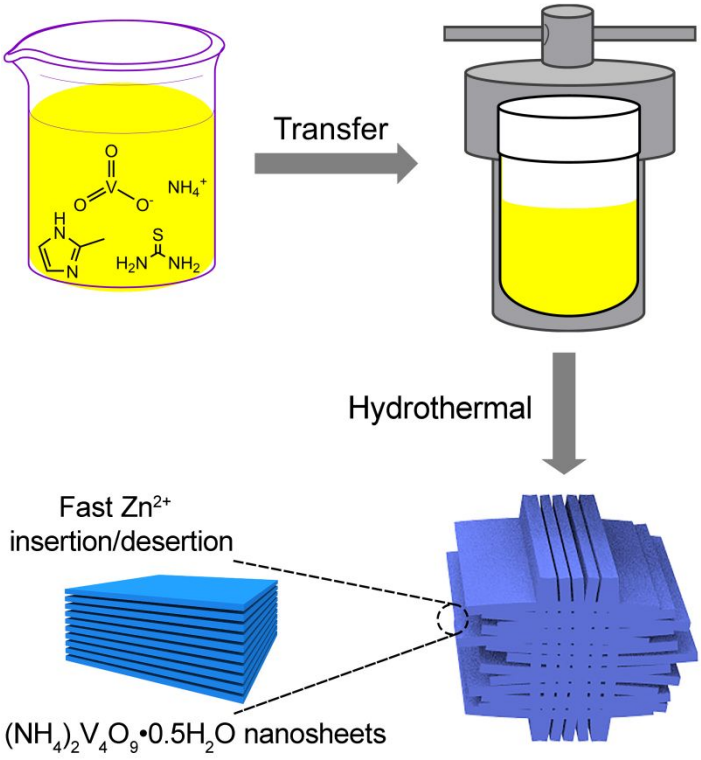

Figure S1. Illustration of the synthetic process of layer-by-layer stacked $(NH_4)_2V_4O_9 \cdot 0.5H_2O$ nanosheet assemblies.

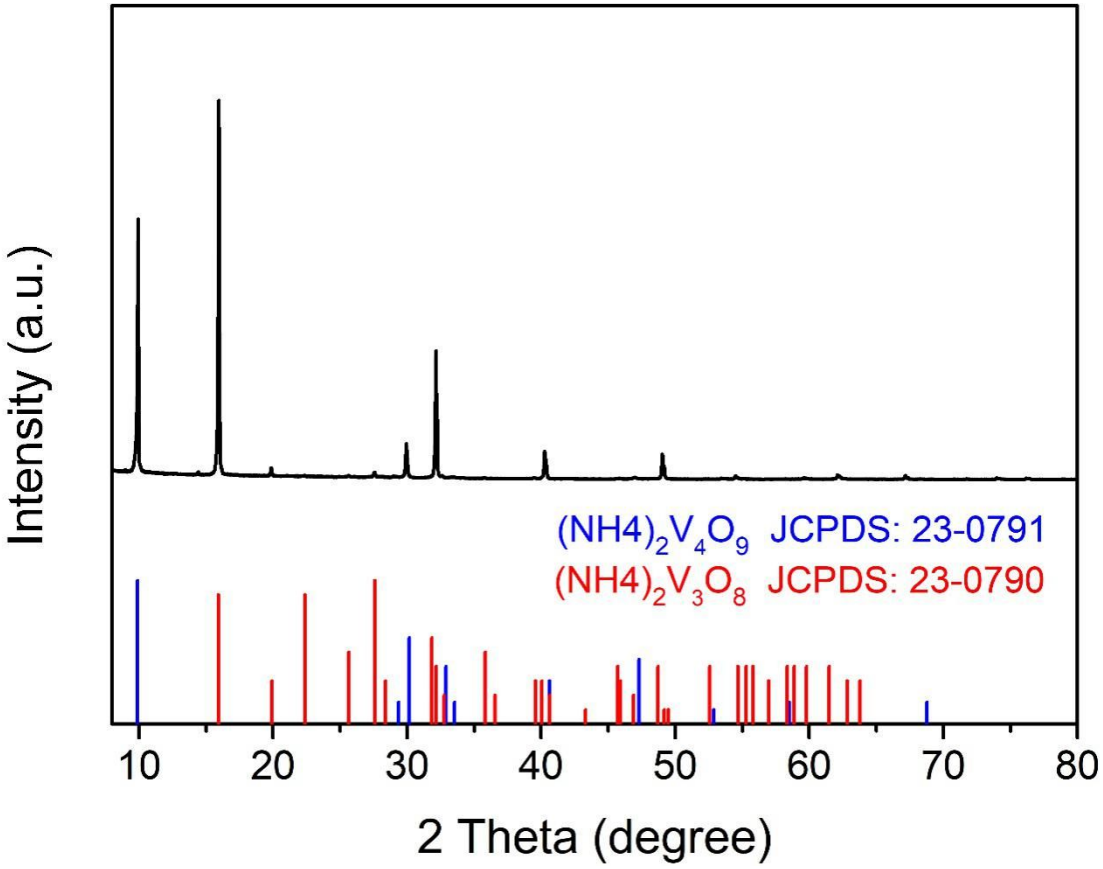

Figure S2. The XRD pattern of the final product synthesized with the absence of 2-Methylimidazole.

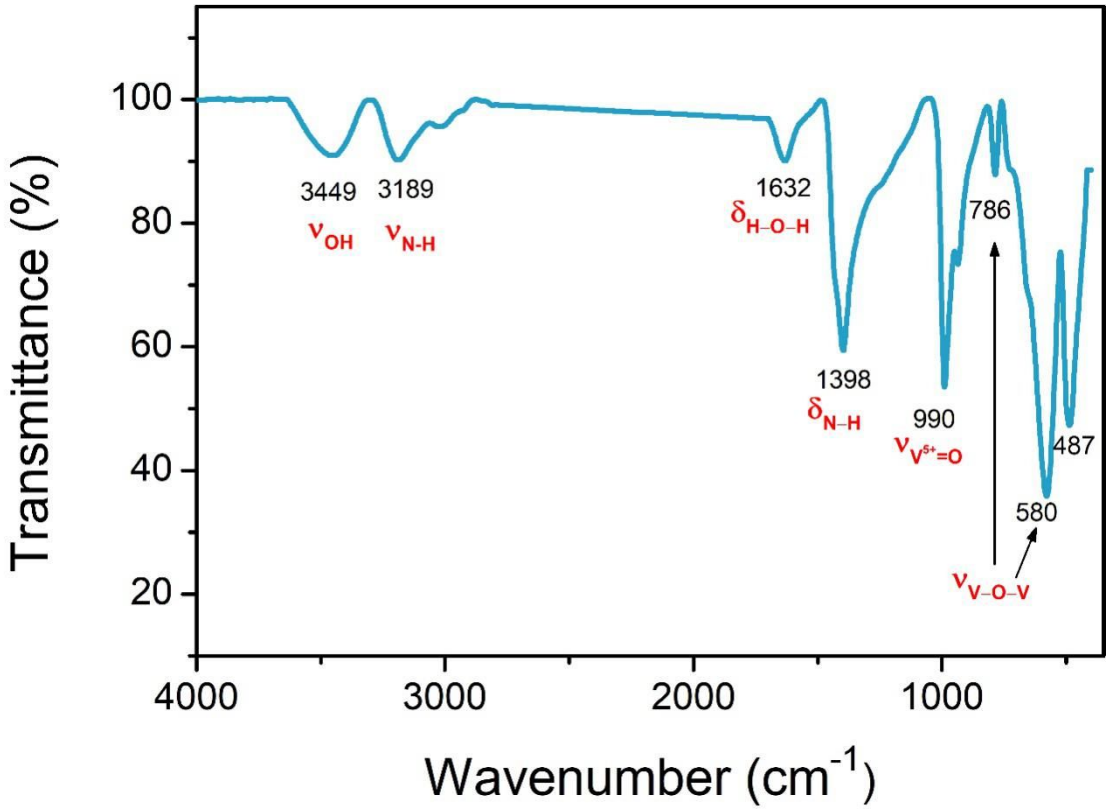

Figure S3. The FTIR spectra of NVO nanosheet assemblies.

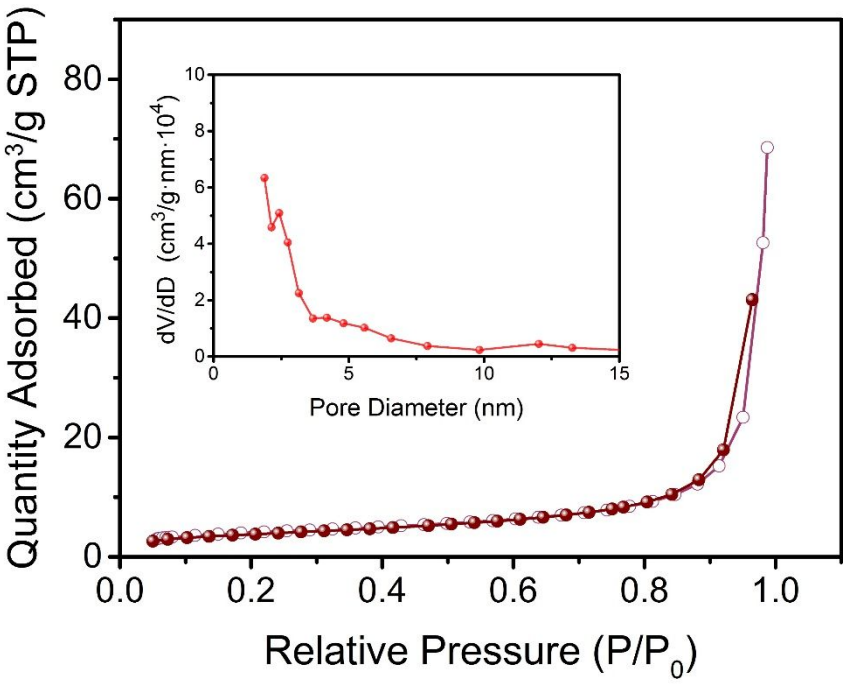

Figure S4. N_2 adsorption-desorption isotherms of NVO. The inset shows the pore distribution.

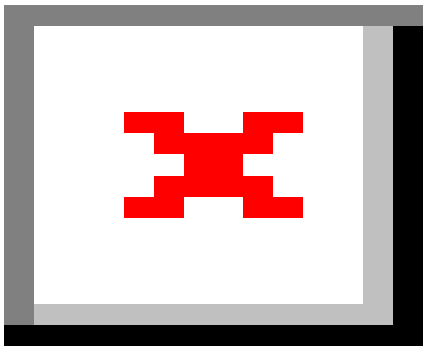

Figure S5. A high-magnification FESEM image of NVO nanosheet assemblies.

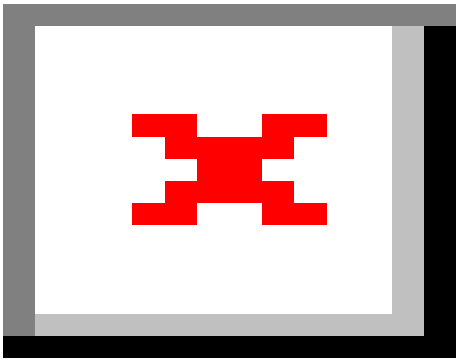

Figure S6. The FESEM image of the NVO electrode after 400 cycles at 1 A g^{-1}.

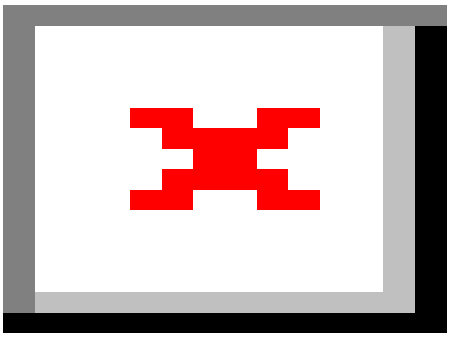

Figure S7. The FESEM image of the NVO electrode after 1000 cycles at 5 A g^{-1}.

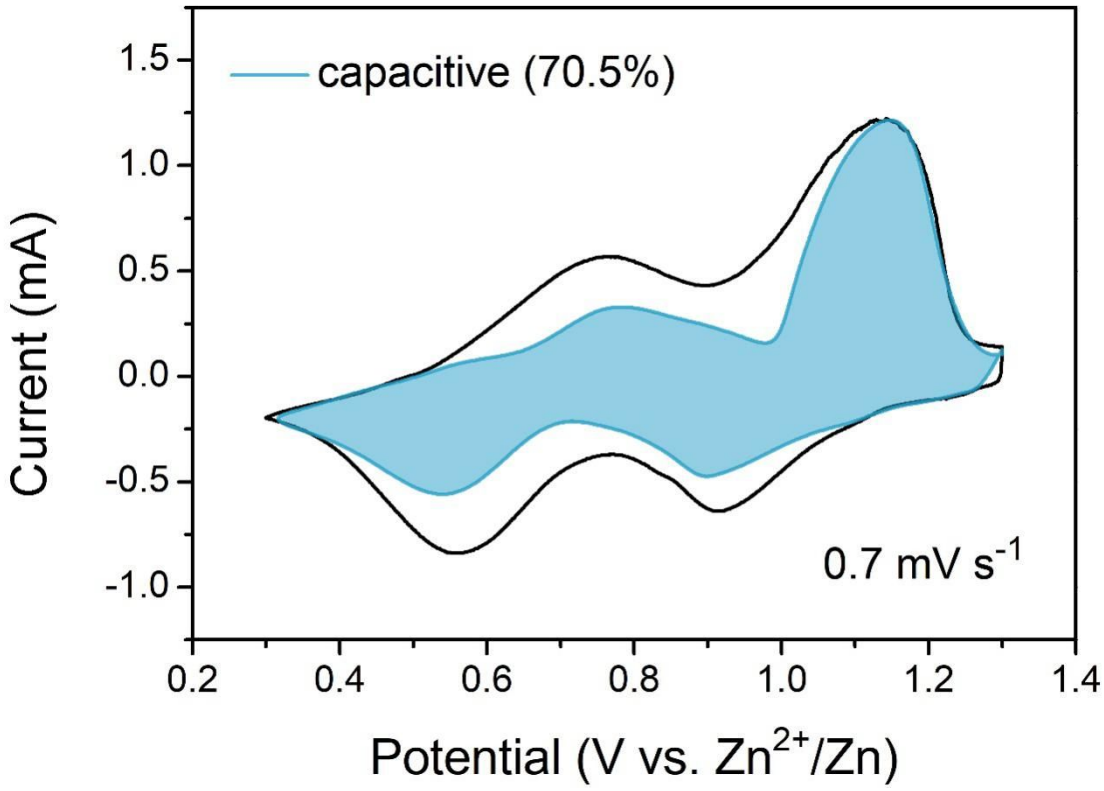

Figure S8. The contribution of capacitive charge storage as a function of the potential at the scan rate of 0.7 mV s^{-1} calculated based on the k_1 analysis.

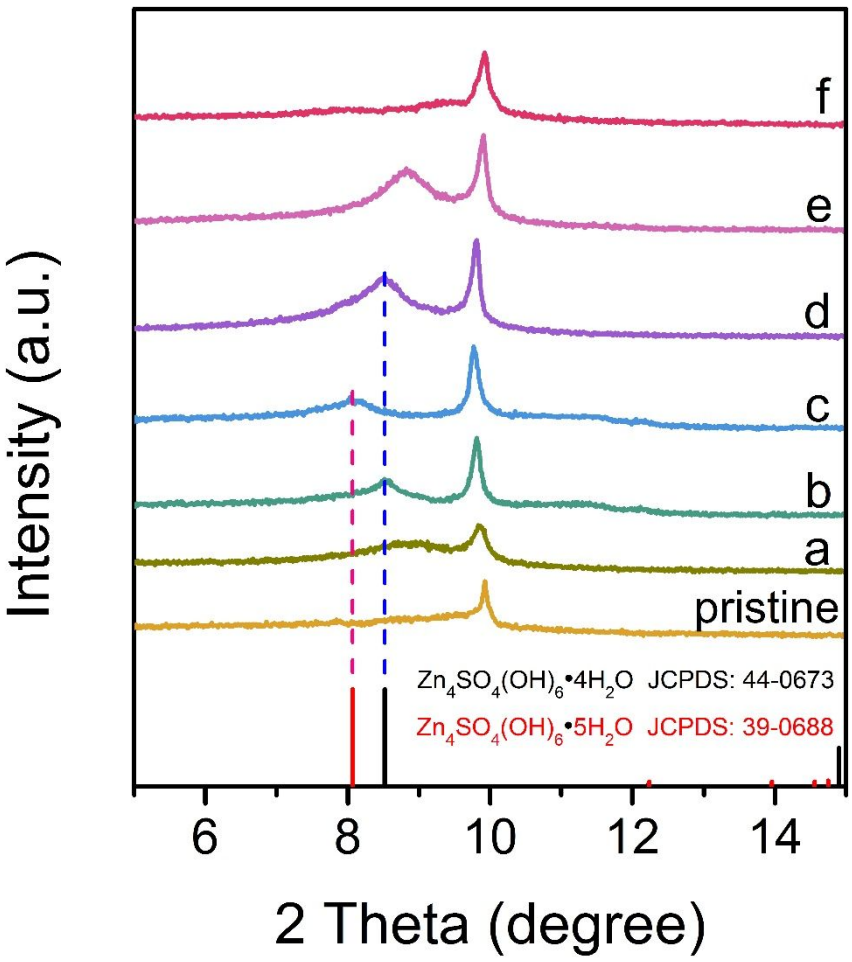

Figure S9. The amplified *ex situ* XRD patterns.