

Supporting Information

Optimizing ammonium vanadate crystal structure by facile in-situ phase transformation of VO₂/NH₄V₄O₁₀ with special micro-nano feature for advanced aqueous zinc ion batteries

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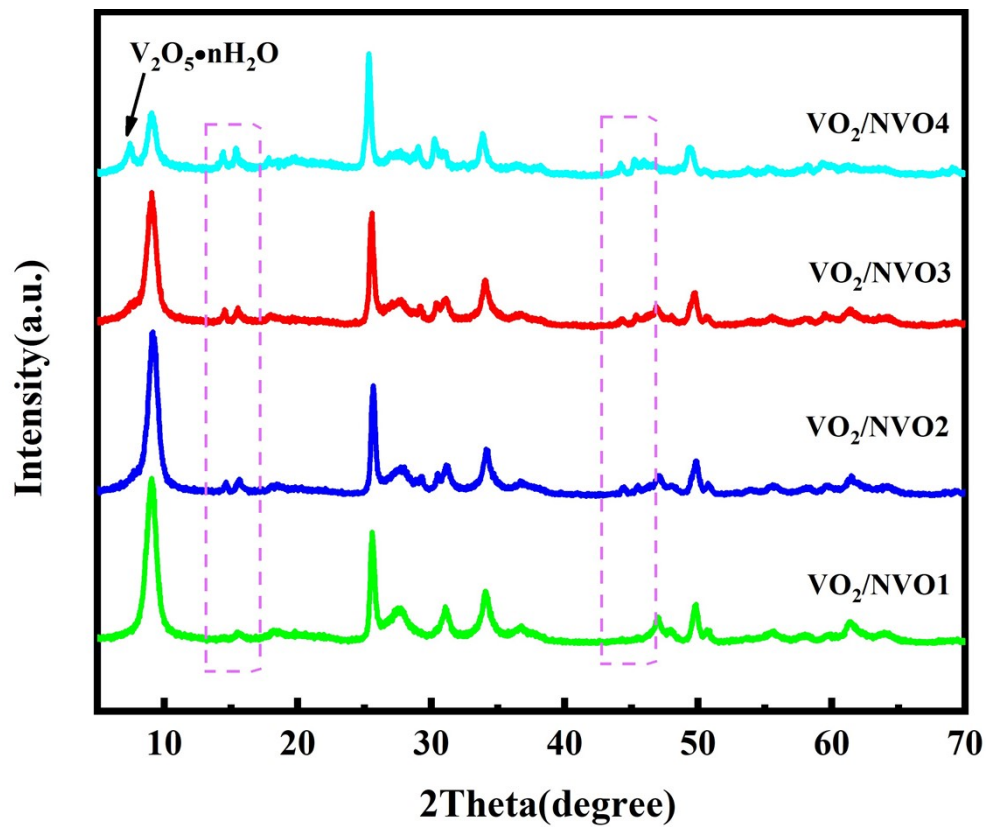


Figure S1. XRD of VO₂/NVO1, VO₂/NVO2, VO₂/NVO3 and VO₂/NVO4.

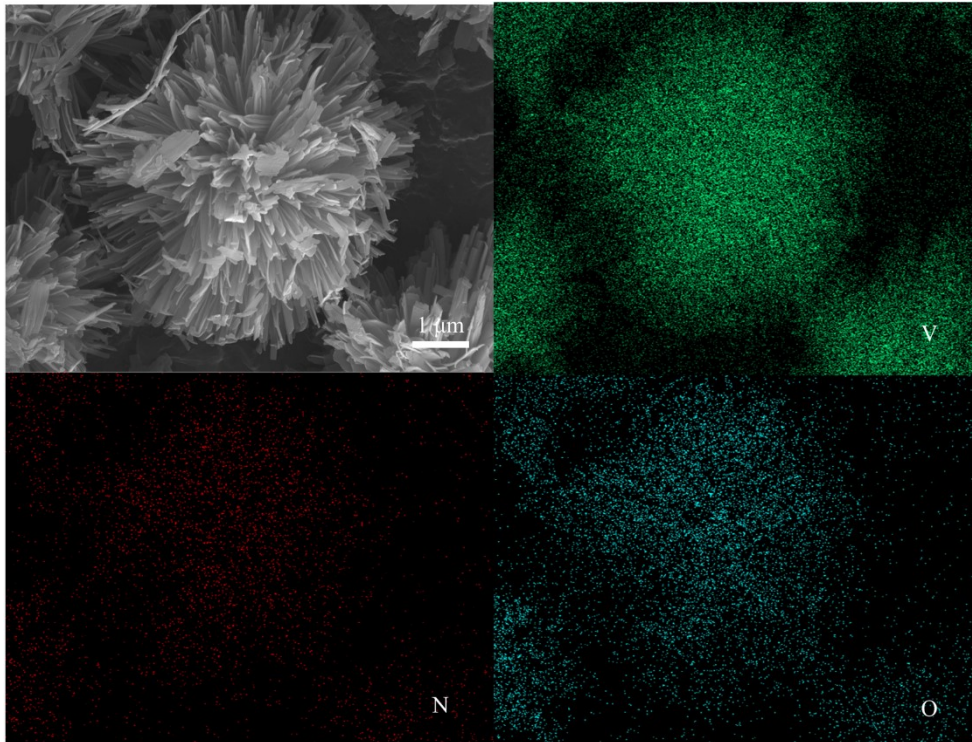


Figure S2. SEM and corresponding EDS mapping for VO_2/NVO_3 .

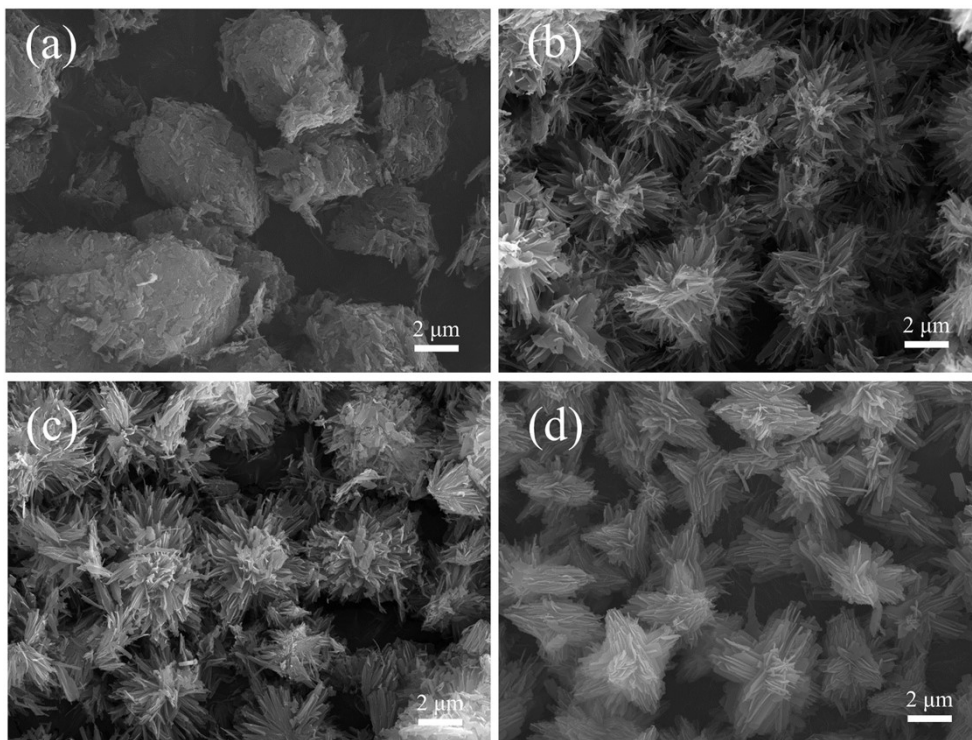


Figure S3. SEM images of (a) $\text{NH}_4\text{V}_4\text{O}_{10}$, (b) VO_2/NVO_1 , (c) VO_2/NVO_2 and (d) VO_2/NVO_4 .

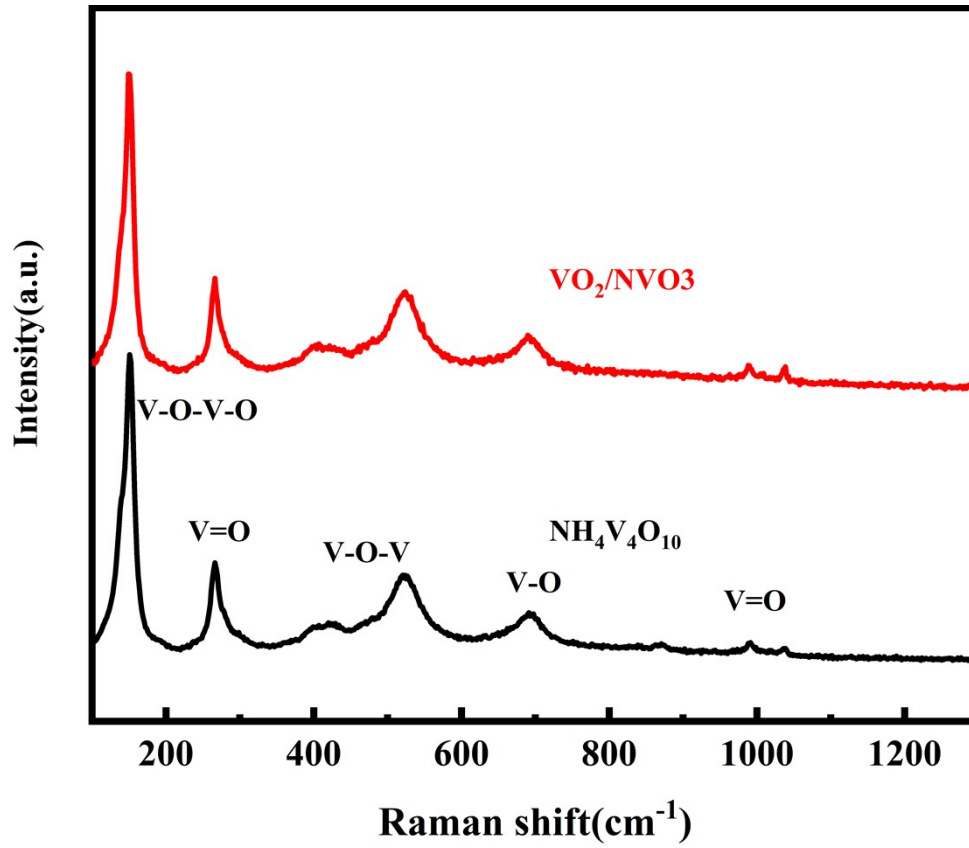


Figure S4. Raman spectrum of $\text{NH}_4\text{V}_4\text{O}_{10}$ and VO_2/NVO_3 .

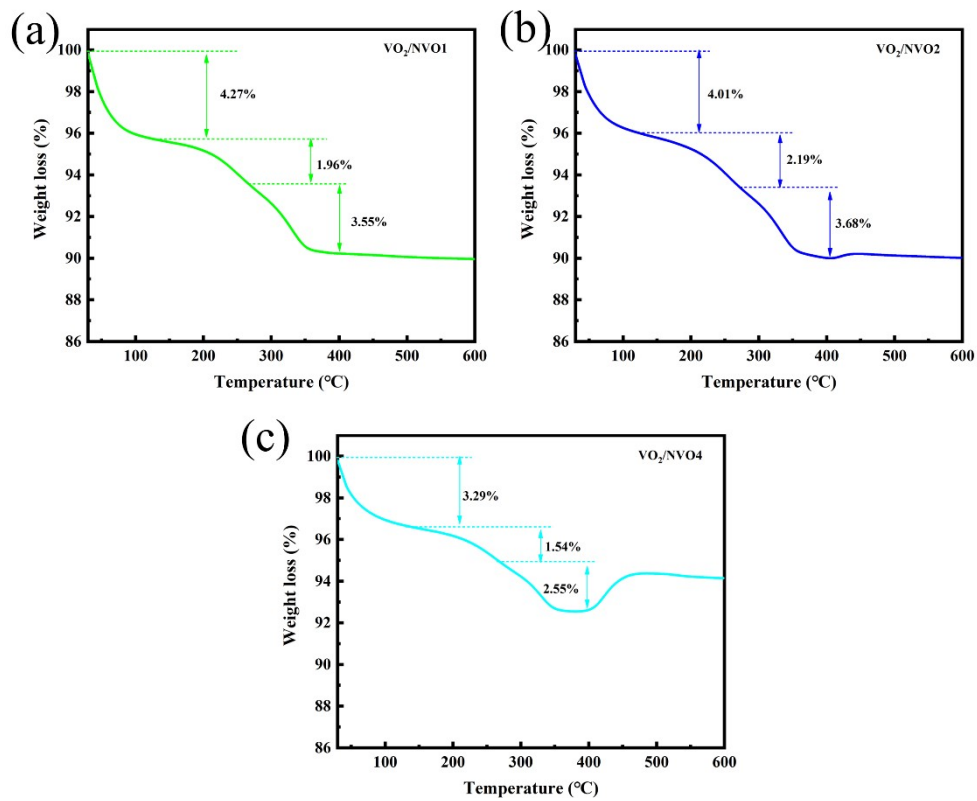


Figure S5. (a-c) TG curves of VO_2/NVO_1 , VO_2/NVO_2 and VO_2/NVO_4 materials.

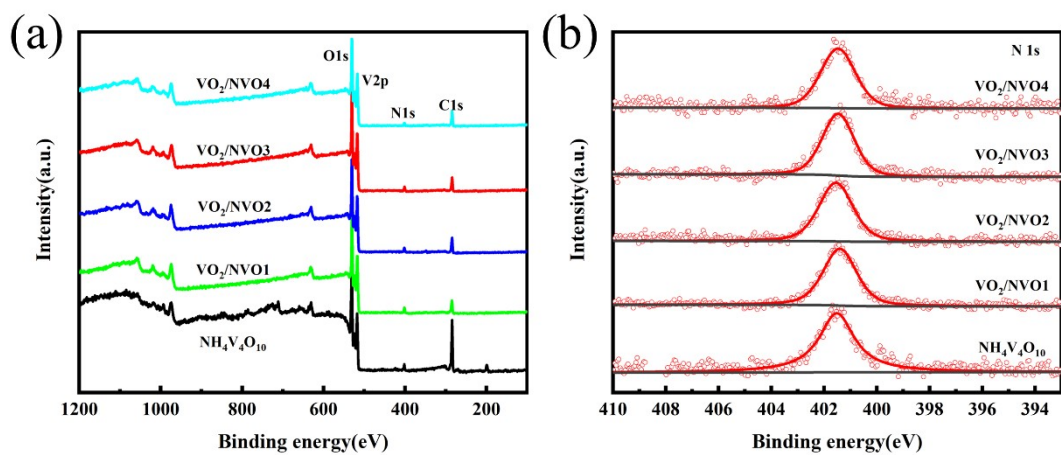


Figure S6. (a) XPS spectrum and (b) N 1s high-resolution of $\text{NH}_4\text{V}_4\text{O}_{10}$, $\text{VO}_2/\text{NVO1}$, $\text{VO}_2/\text{NVO2}$, $\text{VO}_2/\text{NVO3}$ and $\text{VO}_2/\text{NVO4}$ materials.

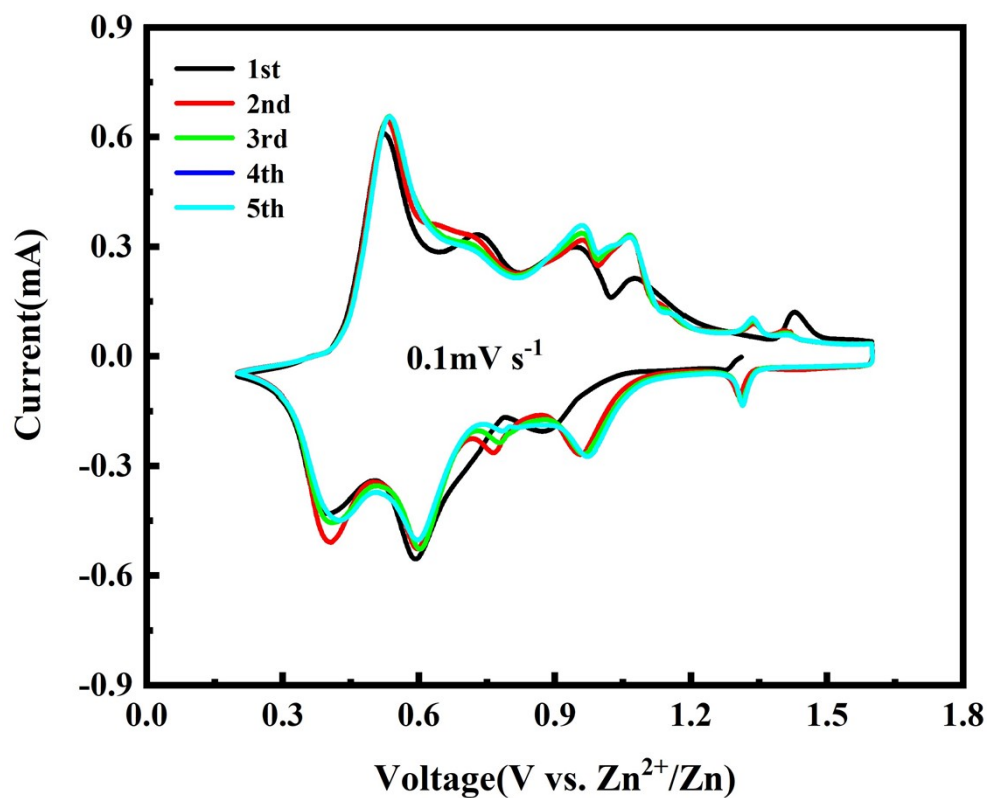


Figure S7. The first five CV curves of $\text{NH}_4\text{V}_4\text{O}_{10}$ electrodes at 0.1 mV s^{-1} .

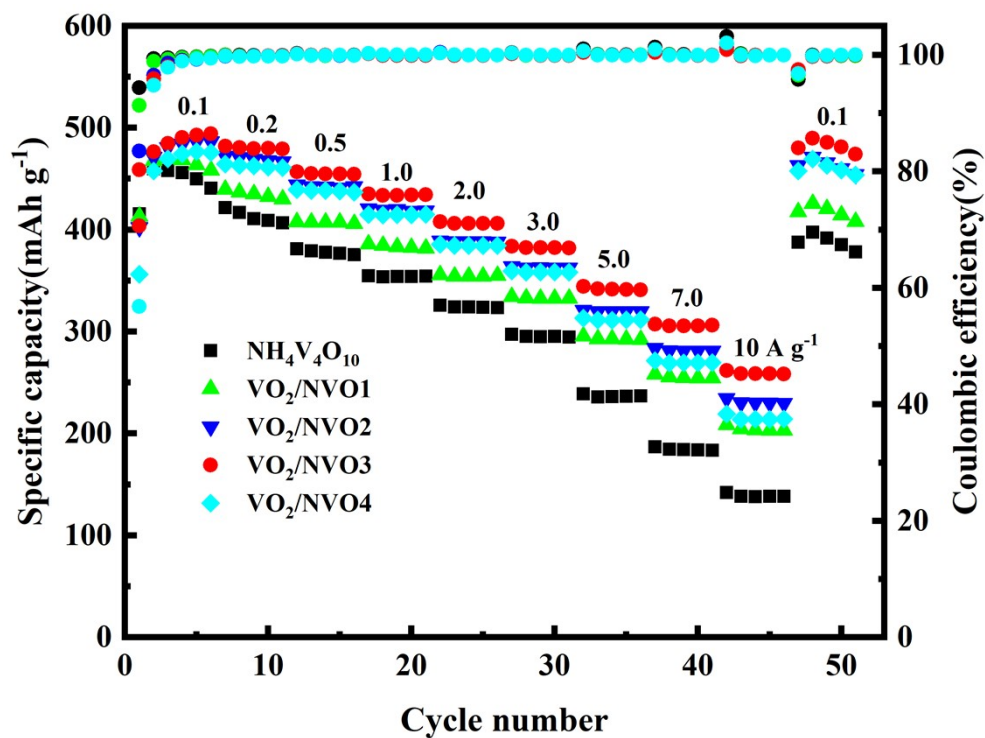


Figure S8. Rate performance of $\text{NH}_4\text{V}_4\text{O}_{10}$, $\text{VO}_2/\text{NVO1}$, $\text{VO}_2/\text{NVO2}$, $\text{VO}_2/\text{NVO3}$ and $\text{VO}_2/\text{NVO4}$ electrodes.

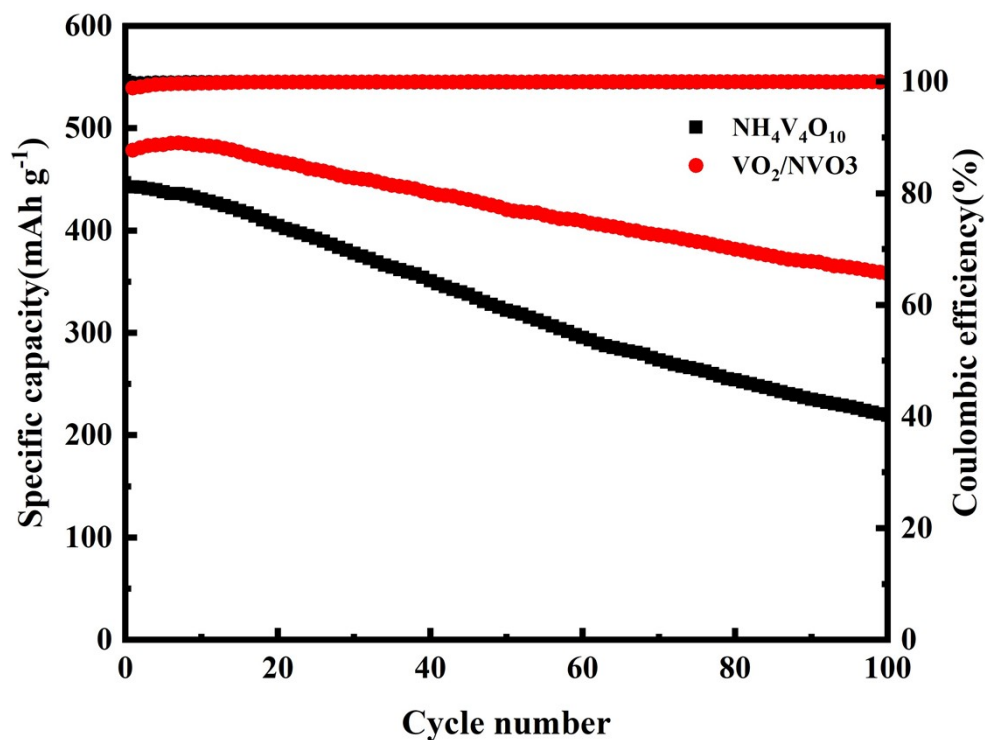


Figure S9. Cycle performance of $\text{NH}_4\text{V}_4\text{O}_{10}$ and $\text{VO}_2/\text{NVO3}$ electrodes at 0.2 A g^{-1} .

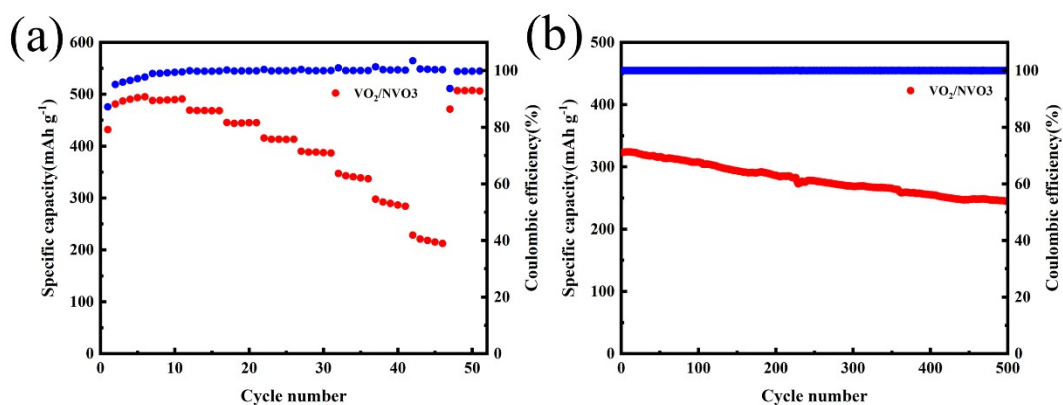


Figure S10. (a) Rate performance at various current density and (b) cycle performance at 5 Ag⁻¹ of the VO₂/NVO₃ electrode with high loading mass of 6 mg cm⁻².

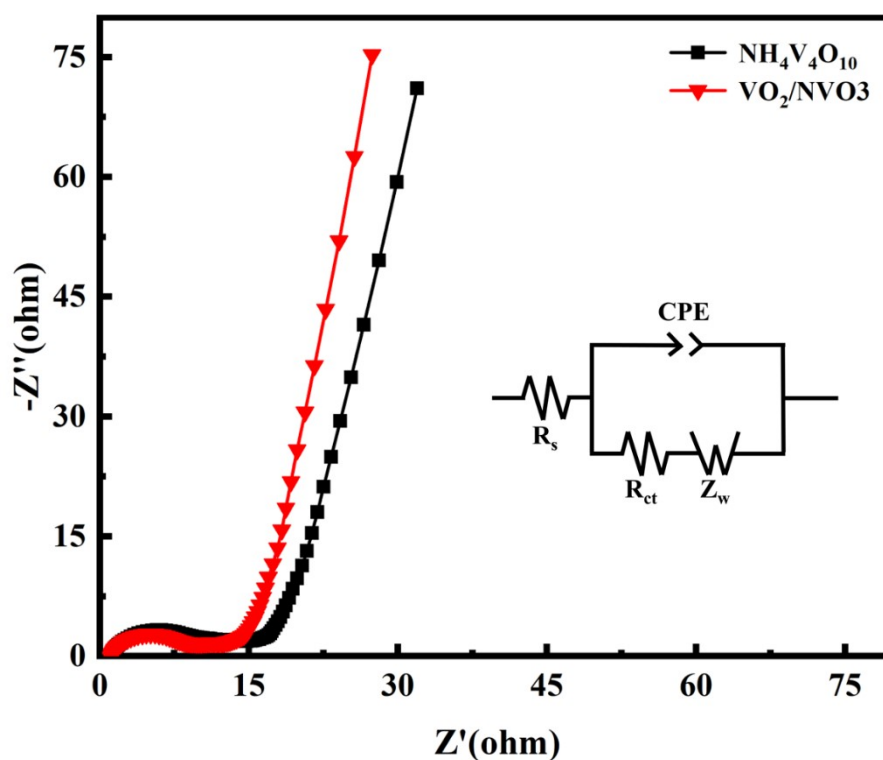


Figure S11. EIS curves of NH₄V₄O₁₀, VO₂/NVO₁, VO₂/NVO₂, VO₂/NVO₃ and VO₂/NVO₄ electrodes.

GITT: The battery was discharged or charged for 10 min at the current density of 0.1A g⁻¹, followed by relaxation for 60 min to back to equilibrium. And the Zn²⁺

diffusion coefficient ($D_{Zn^{2+}}$) was calculated by the galvanostatic and intermittent titration technique (GITT), which was based on the following equation:

$$D_{Zn^{2+}} = \frac{4}{\pi\tau} \left(\frac{m_B V_M}{M_B S} \right)^2 \left(\frac{\Delta E_s}{\Delta E_\tau} \right)^2 \quad (S1)$$

Where τ represents current pulse time, m_B is the mass of the active material. M_B is the molecular weight (g mol^{-1}), V_M is the molar volume ($\text{cm}^3 \text{mol}^{-1}$) and S delegates the surface area of electrode. The ΔE_τ and ΔE_s correspond to the voltage change of constant current pulse and the steady-state voltage change of the current pulse, respectively.

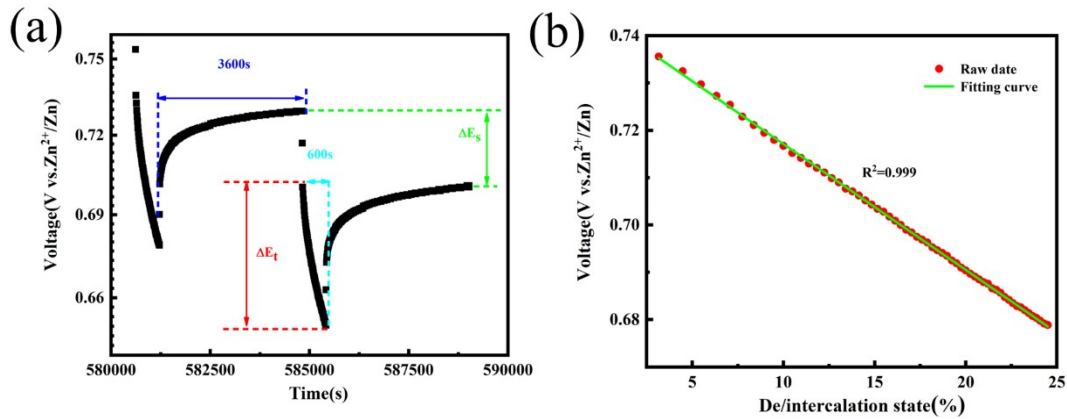


Figure S12. (a) Schematic illustration of partial enlarged GITT curve and (b) the linear relationship between E and $\tau^{1/2}$ for VO_2/NVO_3 electrode.

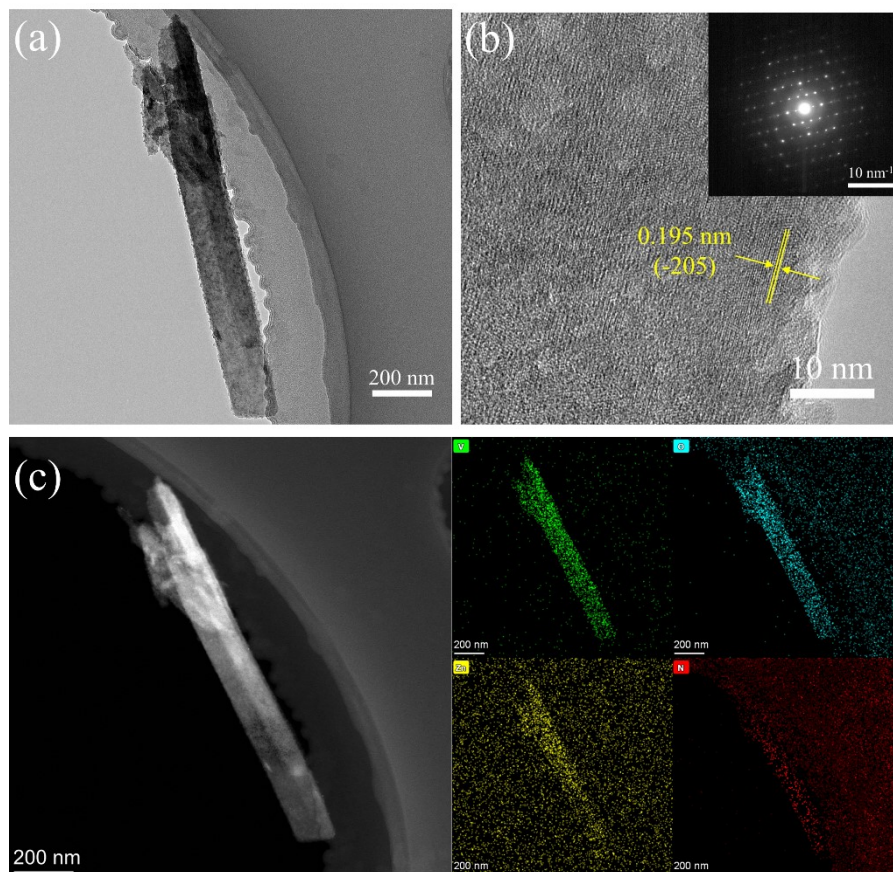


Figure S13. The TEM image of VO₂/NVO₃ after 3 cycles

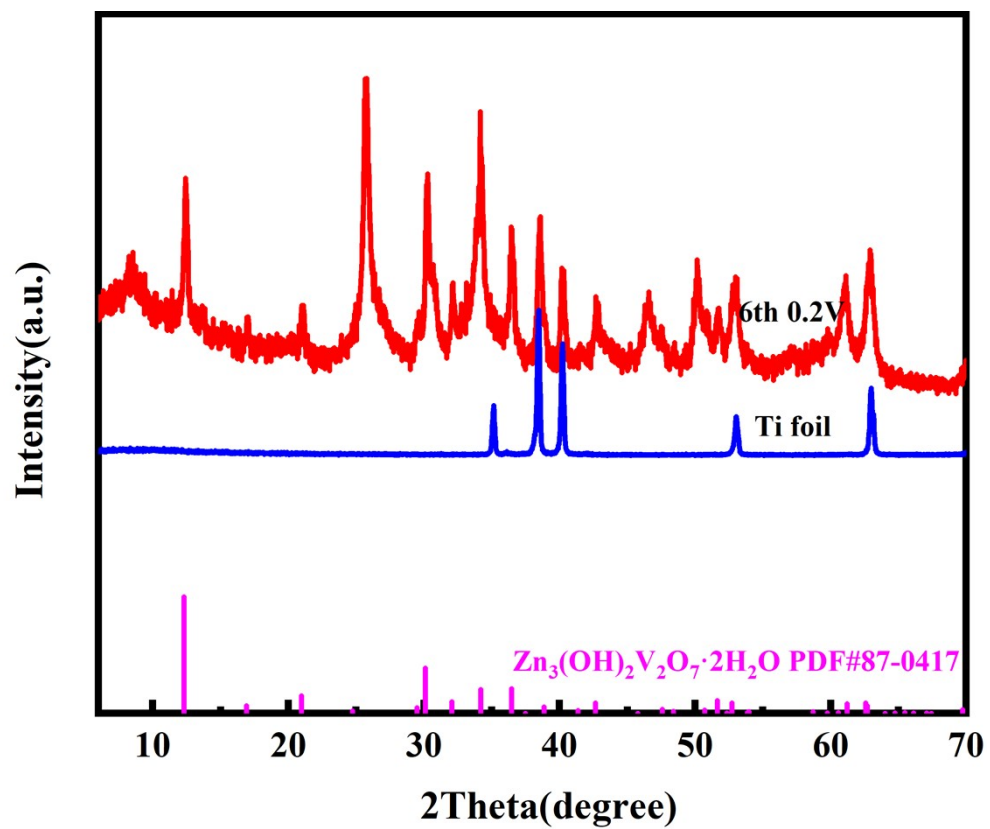


Figure S14. XRD pattern of VO_2/NVO_3 electrode at 0.2 V after suffering 6 cycles.

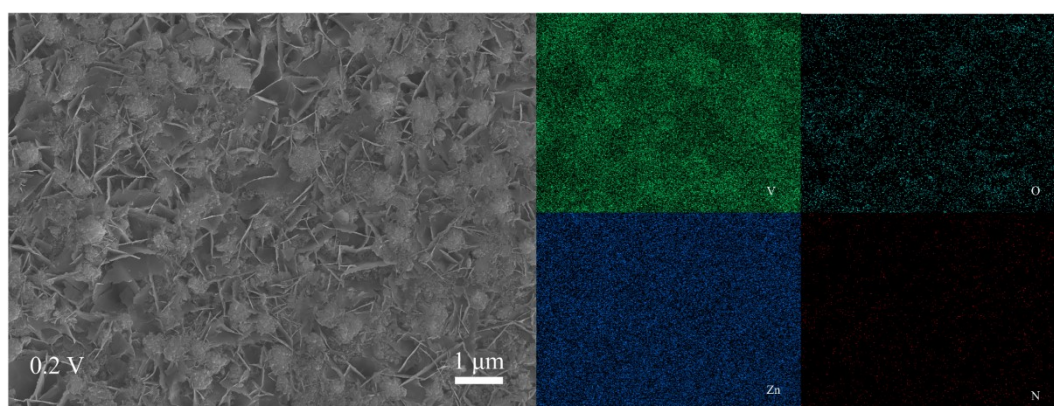


Figure S15. SEM image of VO_2/NVO_3 electrode at 0.2 V after suffering 6 cycles.

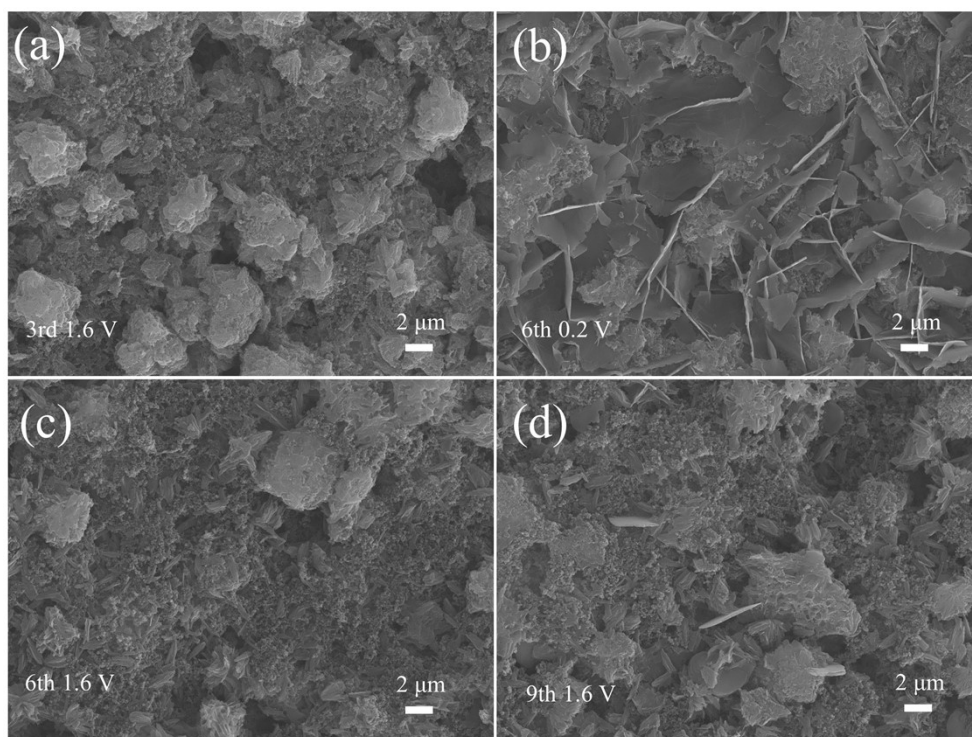


Figure S16. (a-d) SEM image of VO₂/NVO₃ electrode at various charge/discharge state after different cycles.

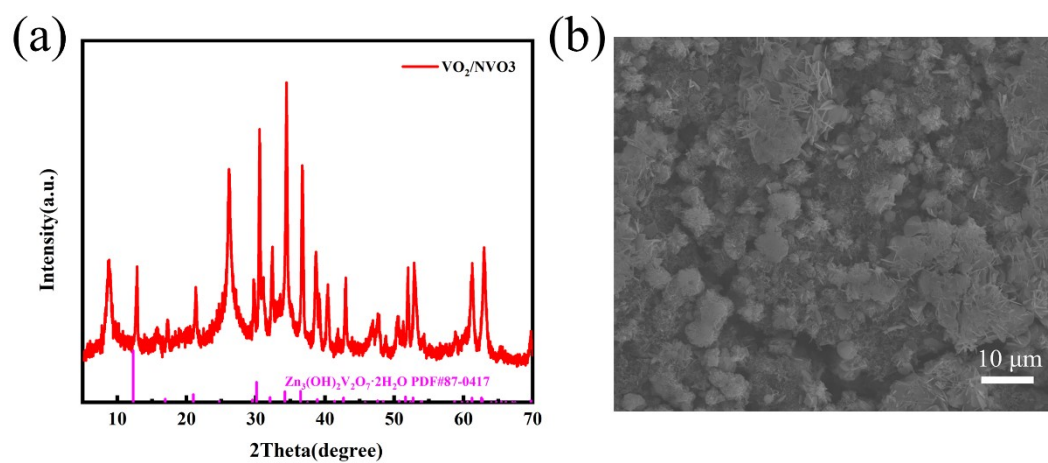


Figure S17. (a) XRD pattern and (b) SEM images of VO₂/NVO₃ electrode after 1000 cycles at the current density of 5 A g⁻¹.

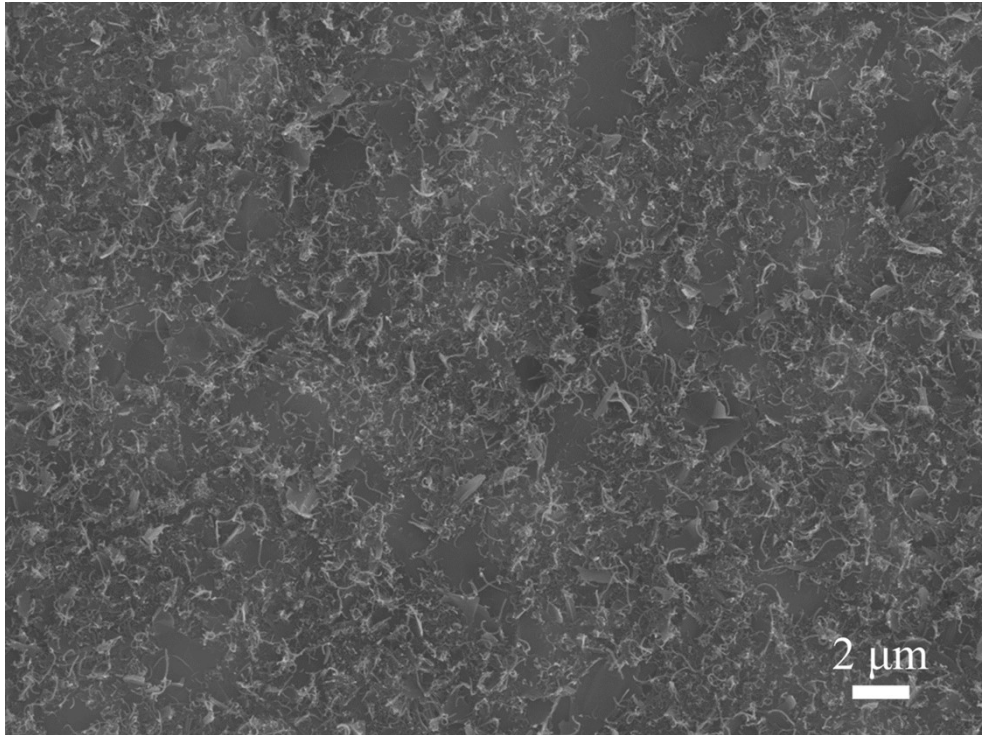


Figure S18. SEM image of VO₂/NVO₃/CG electrode.

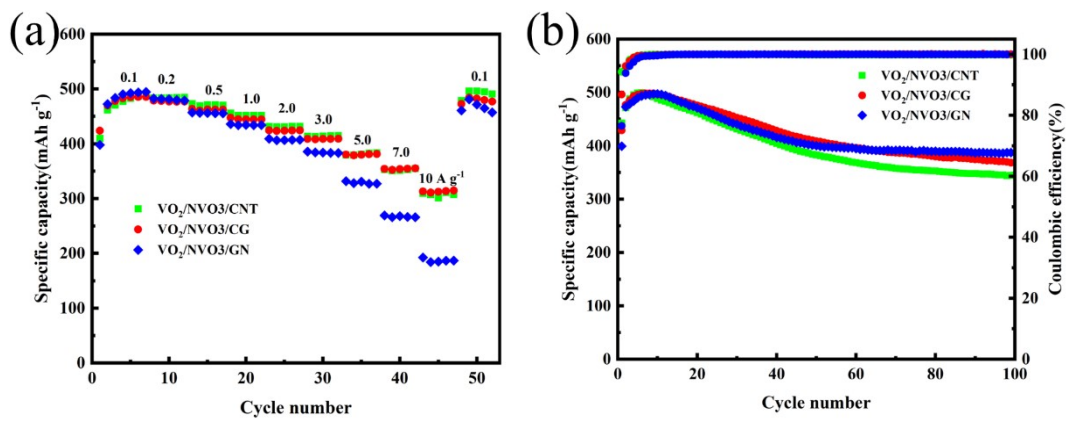


Figure S19. (a) Rate performance and (b) cycle performance at 0.1 A g⁻¹ of VO₂/NVO₃/CNT, VO₂/NVO₃/CG and VO₂/NVO₃/GN membrane electrodes.

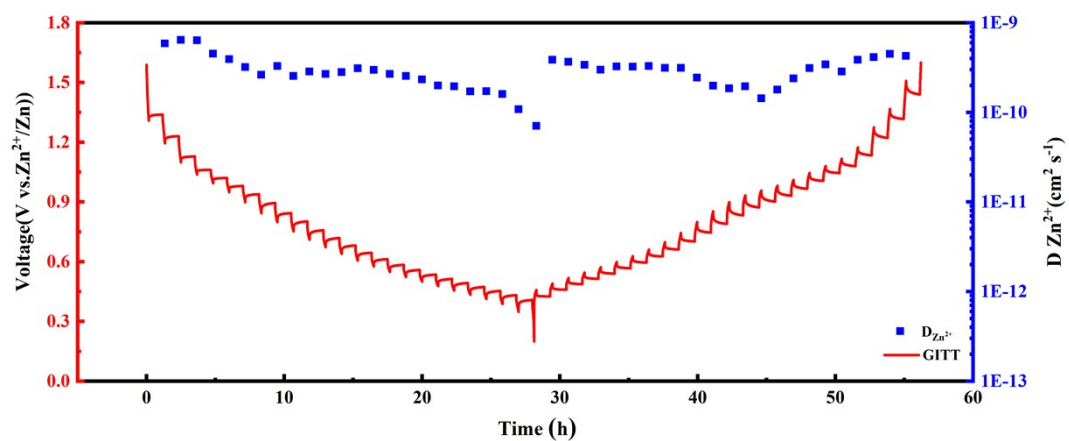


Figure S20. GITT curves and evaluated Zn^{2+} diffusion coefficient of $VO_2/NVO_3/CG$ electrode.

Table S1. The electrochemical property comparison of VO₂/NVO₃ and reported vanadium based materials.

Cathode materials	Specific capacity	Rate performance	Power density	Energy density	Ref.
NH ₄ V ₄ O _{10-x} /rGO	391 mAh g ⁻¹ at 1.0 A g ⁻¹	187 mAh g ⁻¹ at 20 A g ⁻¹	657.2 W kg ⁻¹	260 Wh kg ⁻¹	1
V ₂ O ₅ ·nH ₂ O/PPy	383 mAh g ⁻¹ at 0.1 A g ⁻¹	281 mAh g ⁻¹ at 2 A g ⁻¹	95 W kg ⁻¹	358 Wh kg ⁻¹	2
V ₂ O ₅ ·nH ₂ O/rGO	465 mAh g ⁻¹ at 0.1 A g ⁻¹	230 mAh g ⁻¹ at 15 A g ⁻¹	67 W kg ⁻¹	312 Wh kg ⁻¹	3
H ₁₁ Al ₂ V ₆ O _{23.2}	416.3 mAh g ⁻¹ at 0.3 A g ⁻¹	138.9 mAh g ⁻¹ at 5 A g ⁻¹	220.1 W kg ⁻¹	307.4 Wh kg ⁻¹	4
K _{0.43} (NH ₄) _{0.12} V ₂ O _{5-δ}	373.7 mAh g ⁻¹ at 0.5 A g ⁻¹	216.8 mAh g ⁻¹ at 10 A g ⁻¹	71.1 W kg ⁻¹	269 Wh kg ⁻¹	5
NH ₄ V ₄ O ₁₀ -300	334 mAh g ⁻¹ at 0.5 A g ⁻¹	210 mAh g ⁻¹ at 10 A g ⁻¹	209 W kg ⁻¹	245 Wh kg ⁻¹	6
Cs _{0.24} V ₂ O ₅ ·0.19H ₂ O	400 mAh g ⁻¹ at 0.2 A g ⁻¹	224 mAh g ⁻¹ at 20 A g ⁻¹	147 W kg ⁻¹	294 Wh kg ⁻¹	7
NH ₄ V ₄ O ₁₀ /C ₃ N ₄	391.6 mAh g ⁻¹ at 1.0 A g ⁻¹	194.7 mAh g ⁻¹ at 20 A g ⁻¹	348.6 W kg ⁻¹	289.3 Wh kg ⁻¹	8
Od-VO ₂ ·xH ₂ O/PPy	346.5 mAh g ⁻¹ at 0.1 A g ⁻¹	206 mAh g ⁻¹ at 10 A g ⁻¹	67.6 W kg ⁻¹	223 Wh kg ⁻¹	9
VO ₂ /NVO ₃	493.98 mAh g ⁻¹ at 0.1 A g ⁻¹	258.60 mAh g ⁻¹ at 10 A g ⁻¹	72.10, 5938.85 W kg ⁻¹	356.34, 155.40 Wh kg ⁻¹	This work

Table S2 The comparison of Zn^{2+} diffusion coefficient of VO_2/NVO_3 and previous reported V-based cathode materials.

V-based cathode materials	Zn^{2+} diffusion coefficient ($\text{cm}^2 \text{s}^{-1}$)	References
NVO-300/CC	$10^{-12} \sim 10^{-10}$	10
Ti-NVO	$10^{-11} \sim 10^{-10}$	11
KNVO/CC	$10^{-13} \sim 10^{-11}$	12
Na-NVO	$10^{-9.2} \sim 10^{-10}$	13
$\text{V}_2\text{O}_5 \cdot n\text{H}_2\text{O}$	$10^{-8} \sim 10^{-9}$	14
$\text{PANI}_{0.22} \cdot \text{V}_2\text{O}_5 \cdot 0.88\text{H}_2\text{O}$	$10^{-8} \sim 10^{-10}$	15
$\text{Ca}_{0.24}\text{V}_2\text{O}_5$	$10^{-8} \sim 10^{-10}$	16
VO_2/NVO_3	$10^{-10.1} \sim 10^{-9.6}$	This work
$\text{VO}_2/\text{NVO}_3/\text{CG}$	$10^{-9.8} \sim 10^{-9.4}$	This work

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