Supplementary Information (SI) for Journal of Materials Chemistry A.

Supplementary Material

Section S1 Calculation methodology

1.1 Calculation of capacitance and diffusion control contributions

The peak response current (i, mA) versus scan rate (v, mV s^{-1}) can be fitted to different scan rates by the following equations[1, 2]

$$i=av^b \qquad \text{(Eq. S1)}$$

a and b are constants. The contribution of capacitance can be analyzed qualitatively through the power law relationship between v and i. The contribution of capacitance can be analyzed qualitatively through the power law relationship between b and v. When the value of b is about 0.5, diffusion dominates the charge transfer process. When the value of b is about 1.0, capacitance dominates the charge transfer process.

The current (i, mA) at a certain potential can be categorized into capacitance-controlled effect (k_1v) and diffusion-controlled effect ($k_2v^{1/2}$) as shown in the following equation

$$i = k_1v + k_2v^{1/2} \qquad \text{(Eq. S2)}$$

1.2 Calculation of the diffusion coefficient of Zn^{2+}

The diffusion coefficient (D_{Zn}) of zinc ions in the cathode was measured by constant current intermittent titration technology (GITT), and the calculation formula is as follows[3, 4]

$$D_{Zn} = \frac{4}{\pi\tau}\left(\frac{mV}{MS}\right)^2\left(\frac{\Delta E_s}{\Delta E_\tau}\right)^2 \qquad \text{(Eq. S3)}$$

Among them, τ represents the pulse duration (s), M represents the mass of the active substance (g), and M and V represent the molar mass (g mol^{-1}) and molar volume (cm^3 mol^{-1}), respectively. S represents the contact area between the electrode and the electrolyte (cm^2), and ΔE_s and ΔE_τ, respectively, represent the voltage changes (V) under the action of steady-state and constant current pulses.

Section S2 Supplementary figures

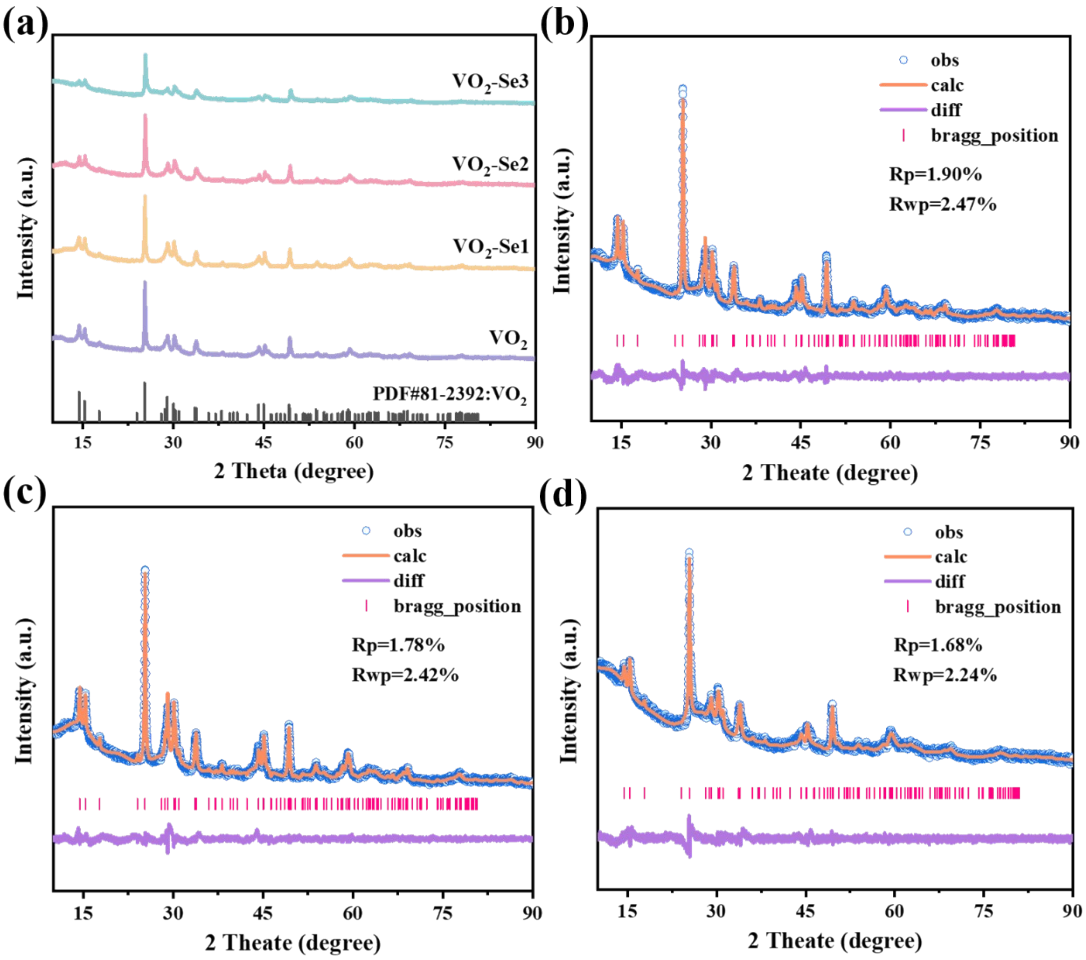

Figure S1. (a) XRD pattern; Rietveld refined XRD patterns of samples with different Se doping amounts, (b) VO_2, (c) VO_2-Se1, (d) VO_2-Se3.

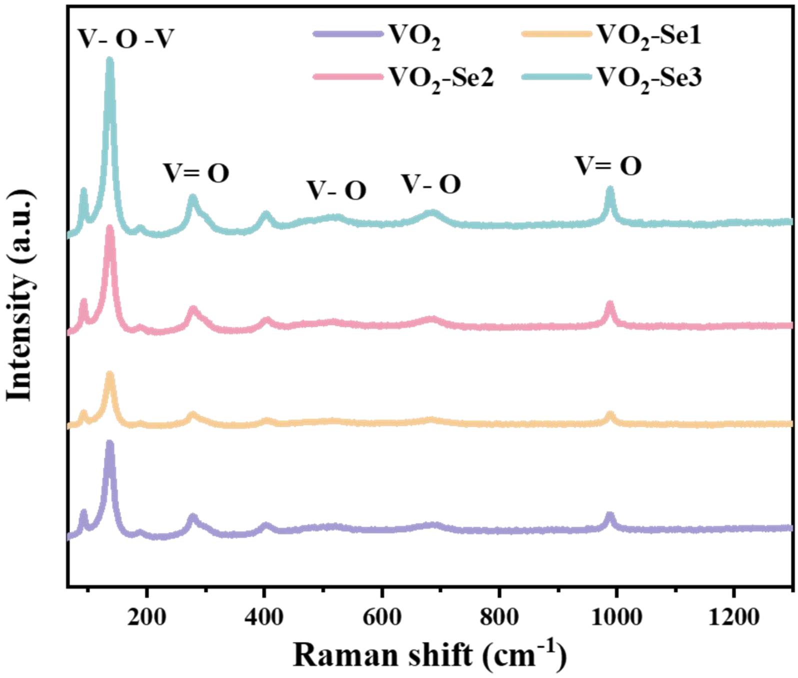

Figure S2. Raman spectra of the prepared VO$_2$-based samples.

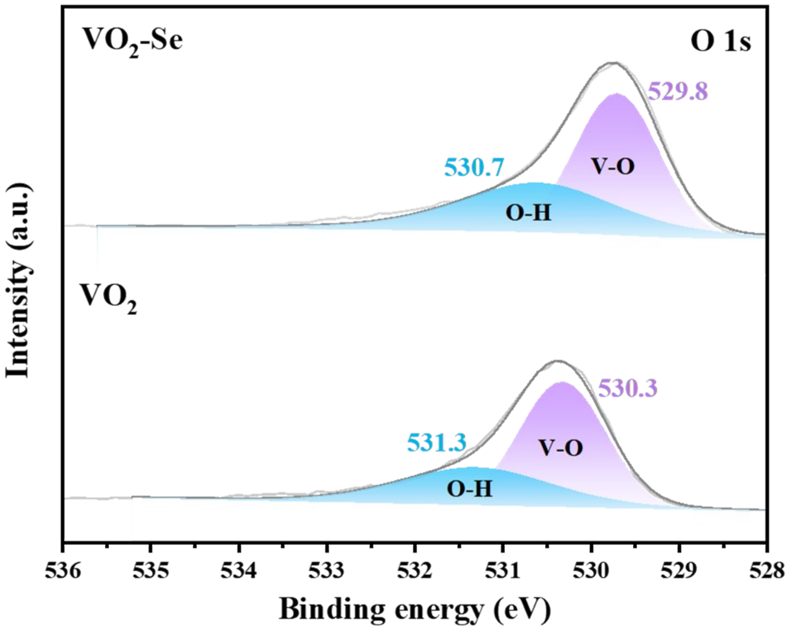

Figure S3. VO$_2$ and Se-doped VO$_2$ in O 1s XPS fine spectra.

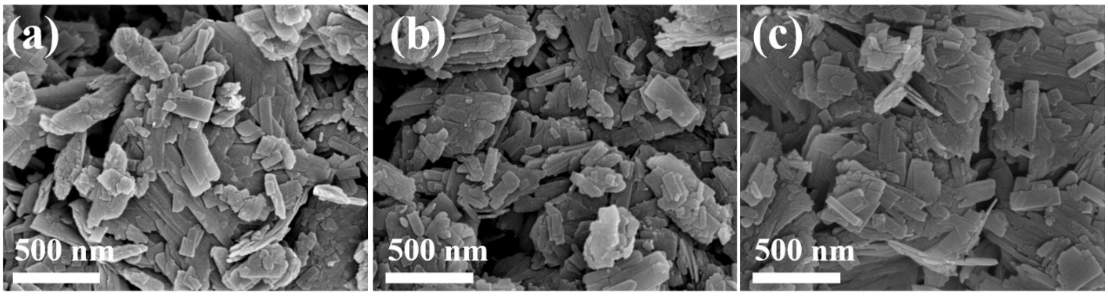

Figure S4. SEM images of VO_2 with different Se doping concentrations: (a) VO_2, (b) VO_2-Se1, (c) VO_2-Se3.

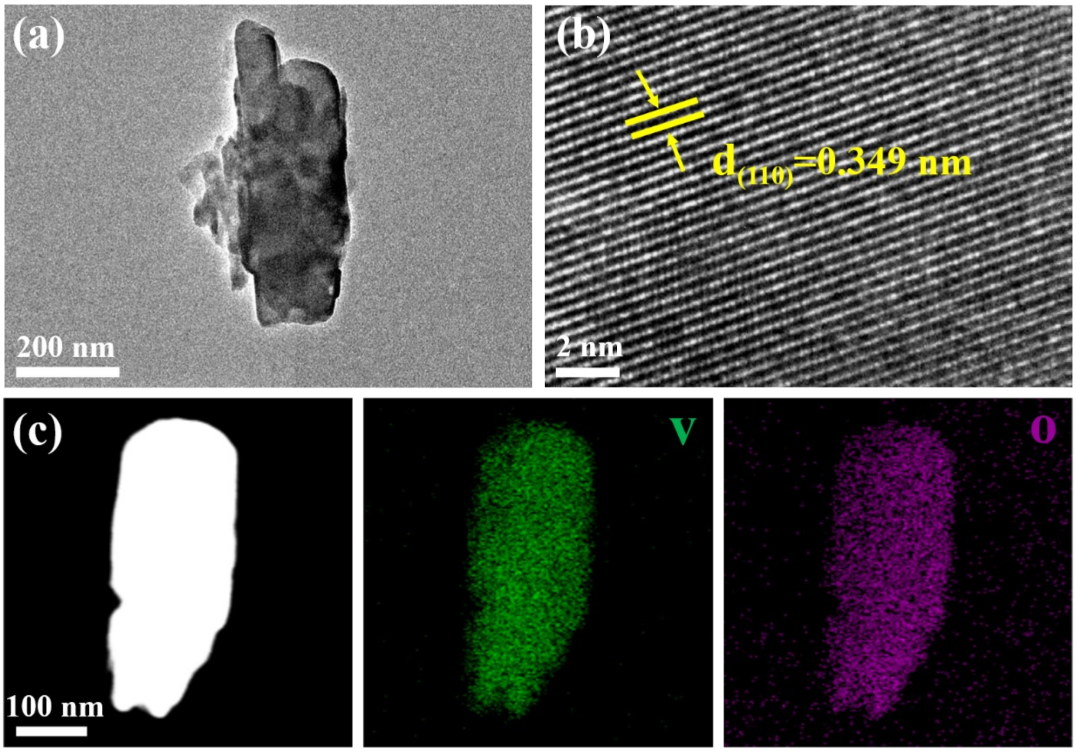

Figure S5. (a) TEM, (b) HRTEM, (c) TEEM-EDS of VO_2.

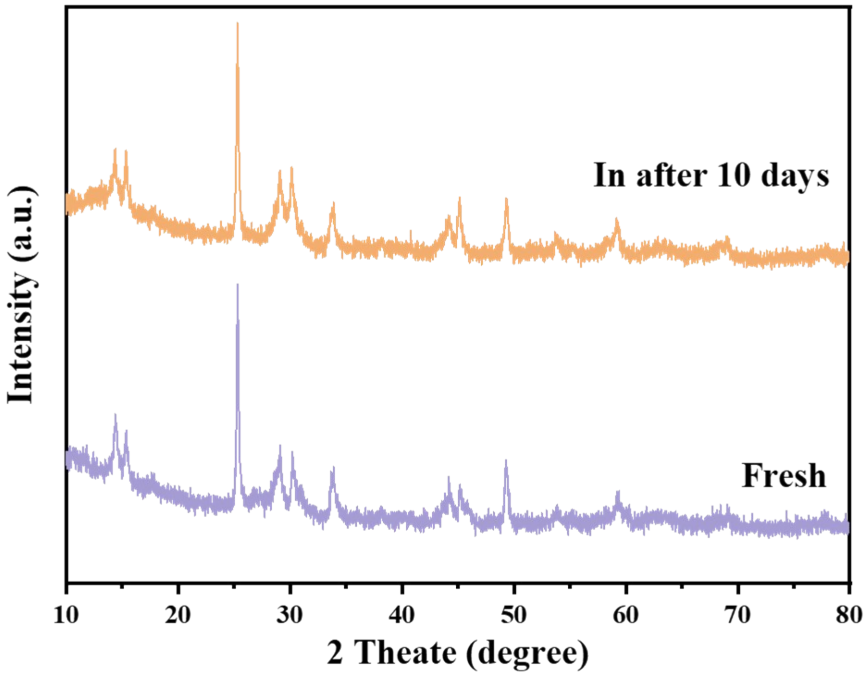

Figure S6. XRD of VO_2-Se2 under natural environmental conditions.

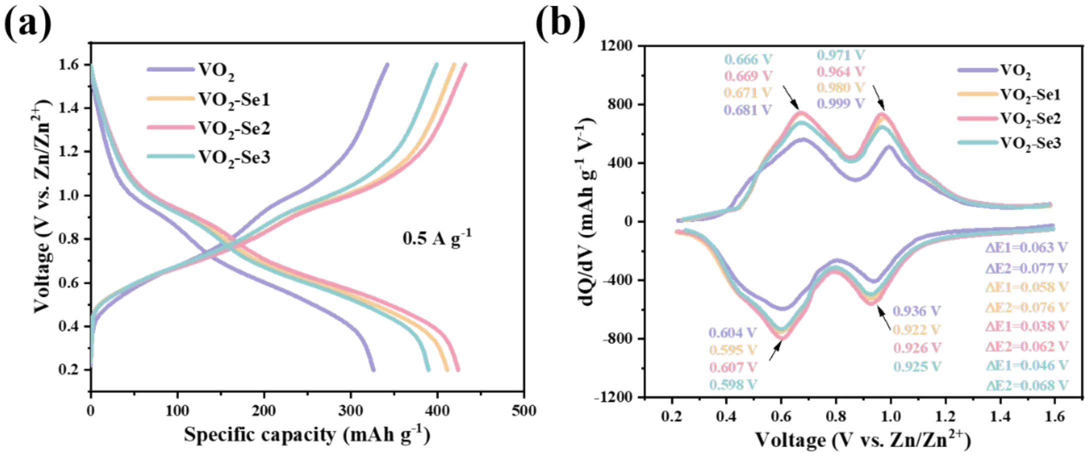

Figure S7. (a) GCD curves and (b) corresponding differential capacity-voltage curves of VO_2 electrodes with different Se doping concentrations at 0.5 a g^{-1}.

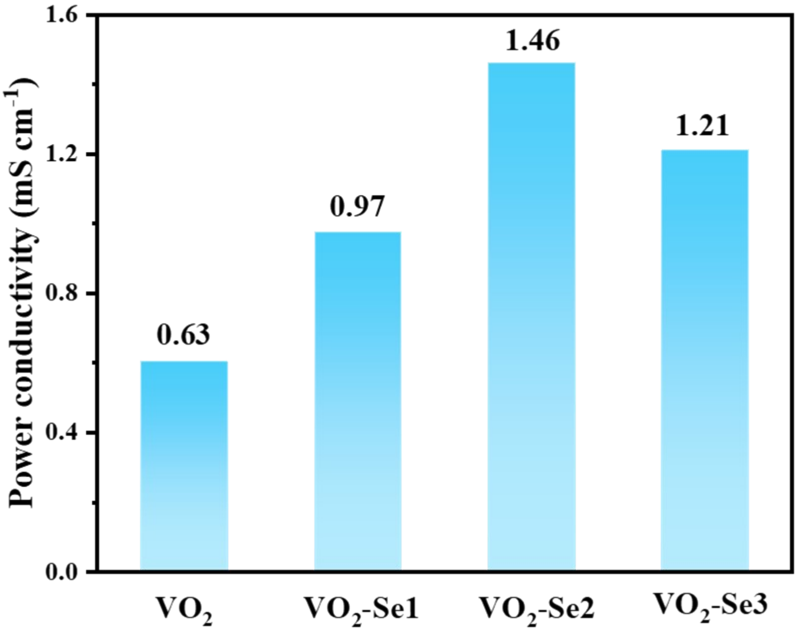

Figure S8. Powder conductivity of VO$_2$ samples with different Se doping concentrations.

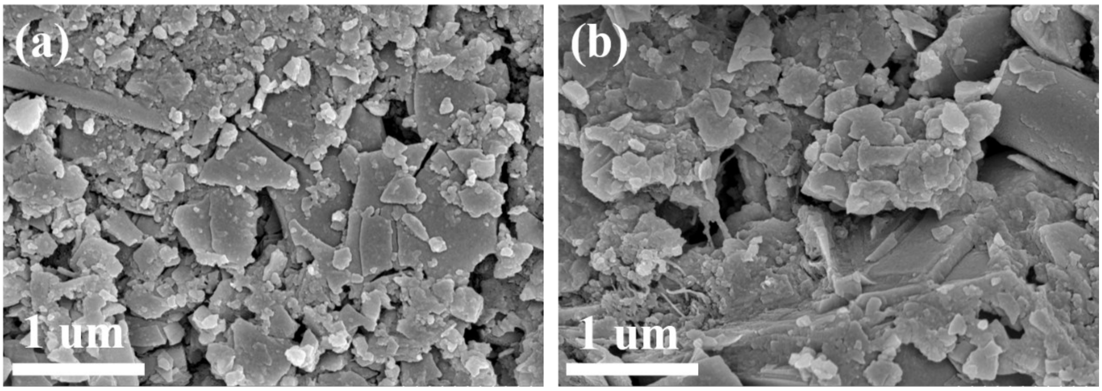

Figure S9. SEM images of the VO_2-Se electrode: (a) the original state, (b) 5000 cycles at 5 A g^{-1}.

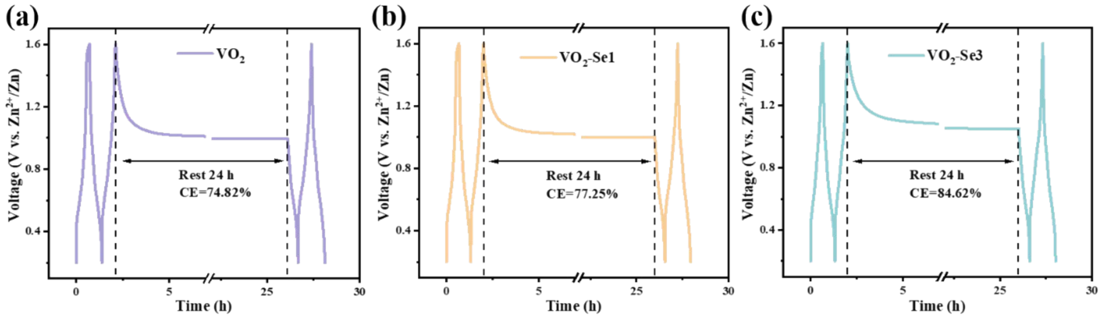

Figure S10. Voltage-time curves of the prepared VO$_2$-based samples during self-discharge of 0.5 A g^{-1} in button cells: (a) VO$_2$, (b) VO$_2$-se1, (c) VO$_2$-se3.

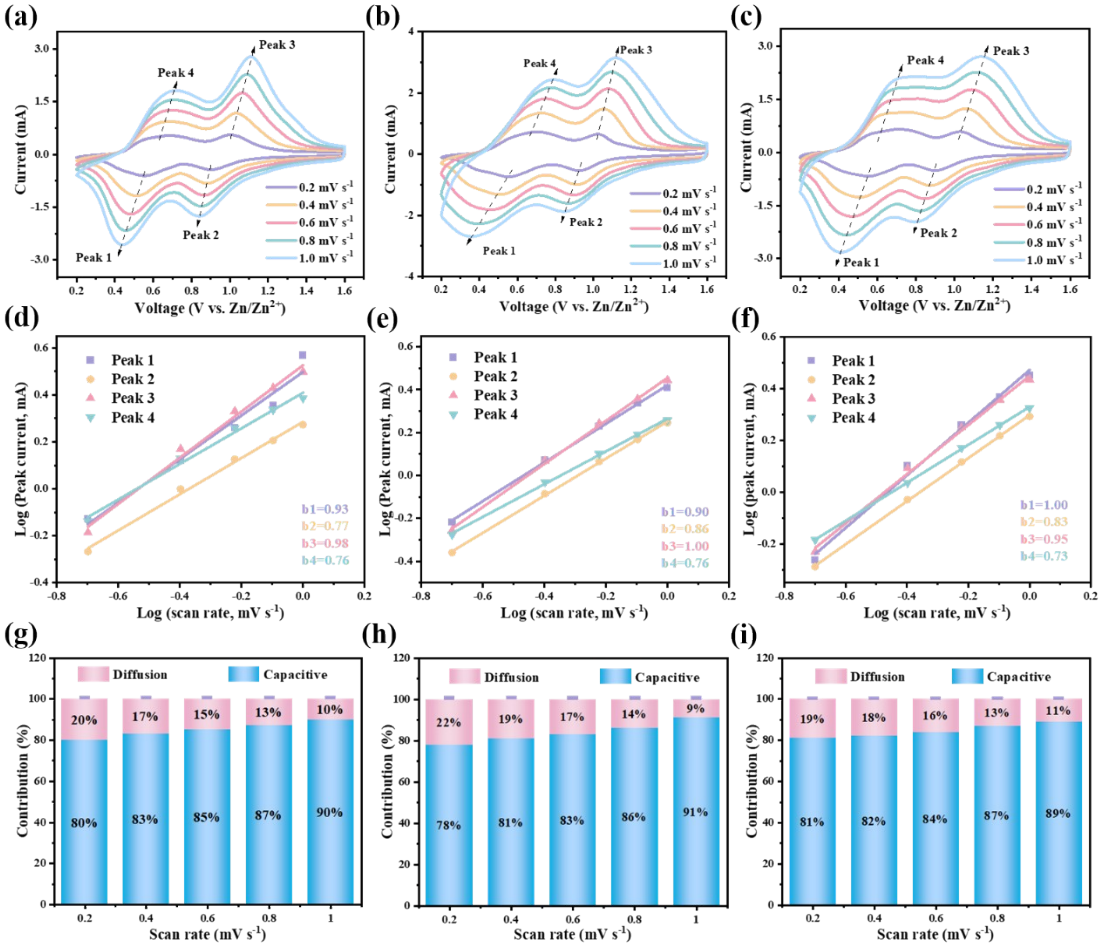

Figure S11. (a, b, c) CV curves at different scan rates, (d, e, f) relationship curves between logi and logv, and (g, h, i) contribution rates of capacitance control process and diffusion control process at different scan rates of VO_2, VO_2-Se1, VO_2-Se2, and VO_2-Se3 electrodes.

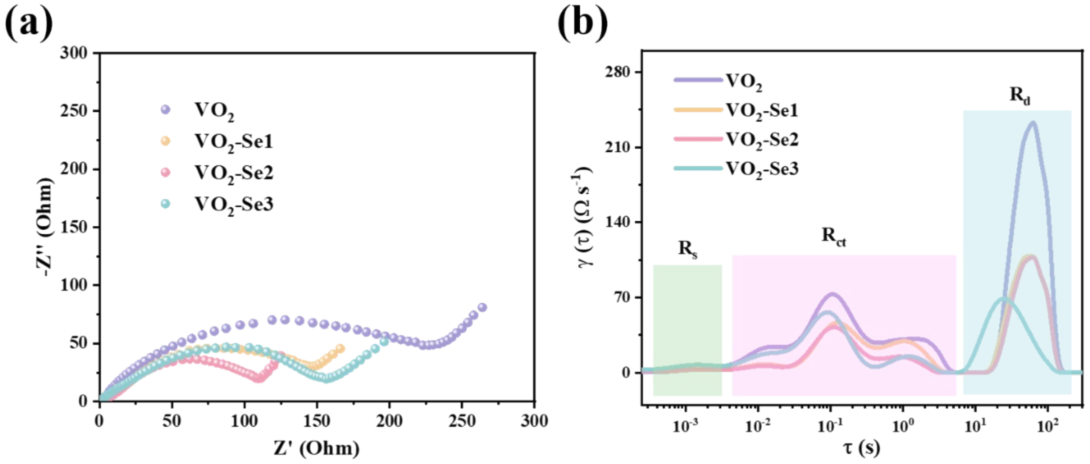

Figure S12. (a) EIS Nyquist plot of VO_2, VO_2-Se1, VO_2-Se2, and VO_2-Se3 electrodes and (b) associated DRT analysis.

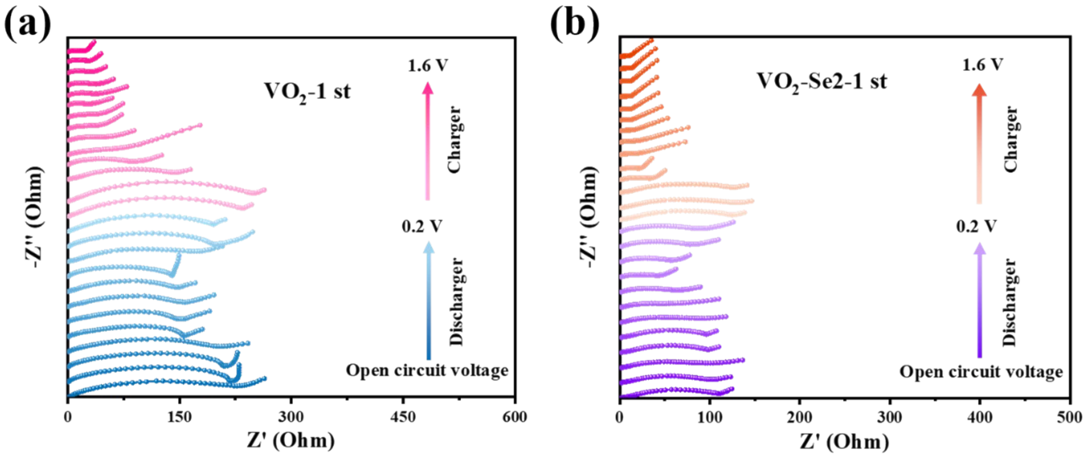

Figure S13. In-situ EIS: (a) VO_2, (b) VO_2-se2.

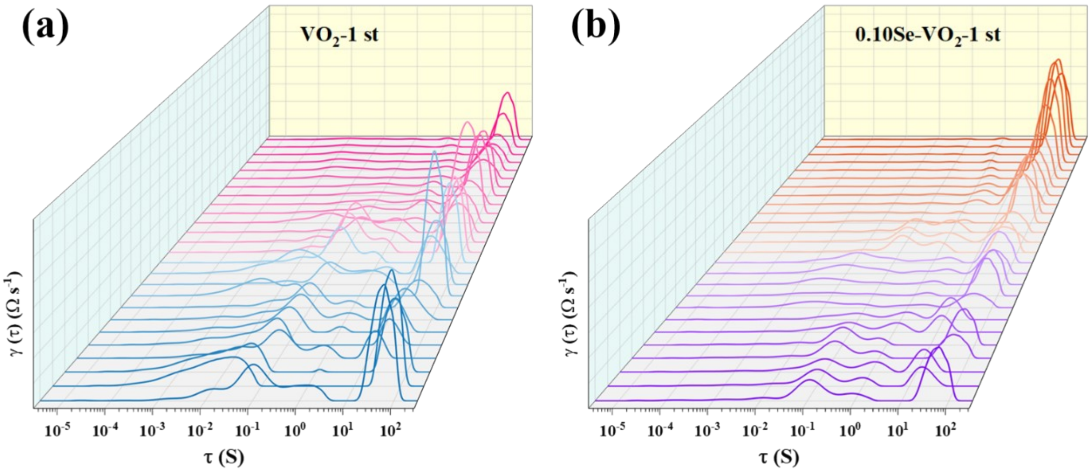

Figure S14. In-situ DRT curves: (a) VO_2, (b) VO_2-Se2.

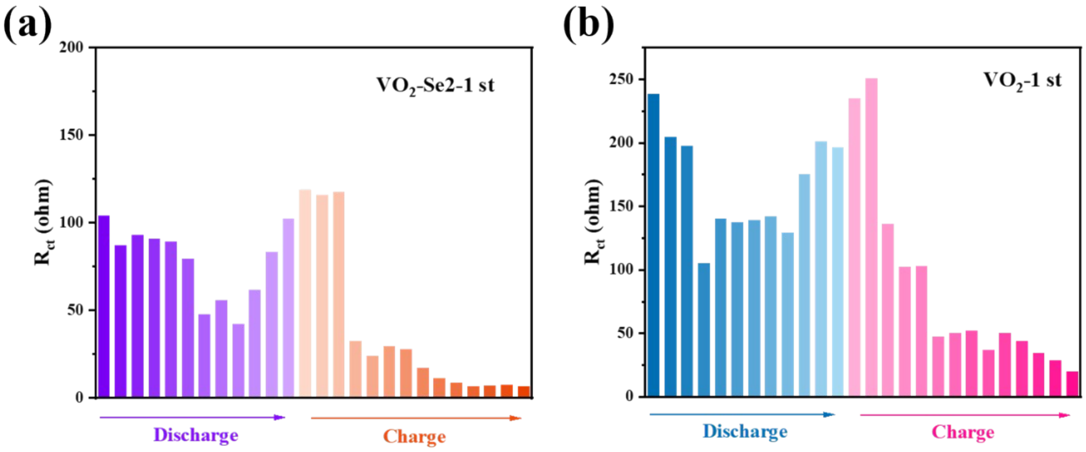

Figure S15. R_{ct} values for (a) VO_2 and (b) VO_2-Se2 during the first cycle.

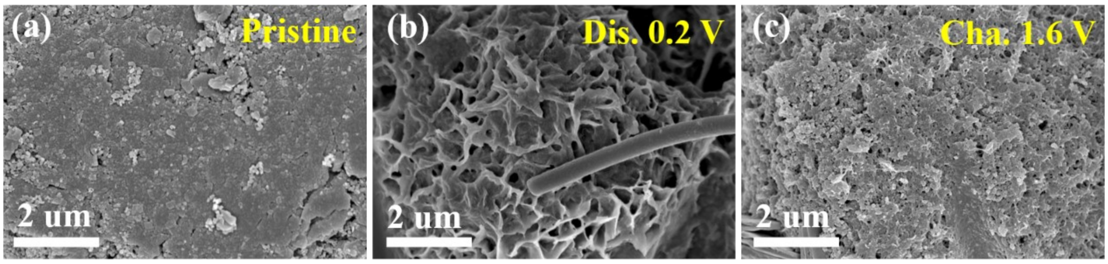

Figure S16. SEM images of VO_2-Se2 under different voltage conditions: (a) Pristine, (b) 0.2 V, (c) 1.6 V.

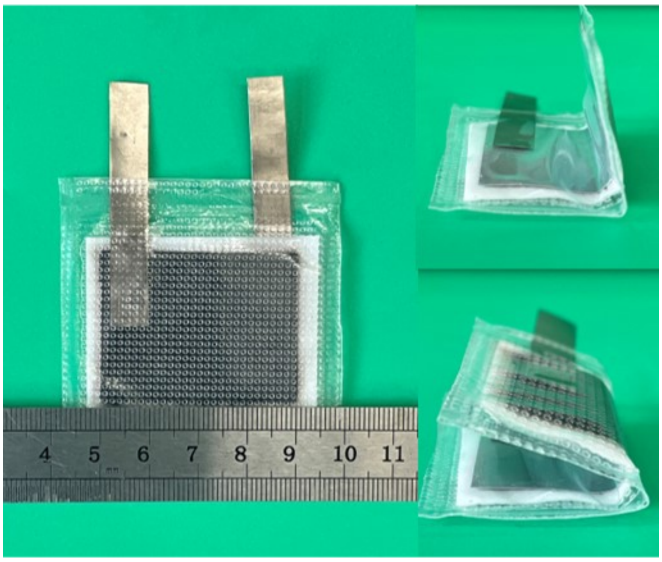

Figure S17. Optical image of the assembled VO_2−Se2||Zn pouch−type battery.

Figure S18. Open−circuit voltage of a fresh soft−pack battery.

Table S1. The Rietveld refined structural parameters of the prepared VO_2-based samples

		VO_2	**VO_2-Se1**	**VO_2-Se2**	**VO_2-Se3**
Crystal system		Monoclinic			
Space group		C2/m			
Cell parameters / Å	a	12.0039	12.0044	12.0048	12.0053
	b	3.6896	3.6905	3.6912	3.6927
	c	6.4213	6.4302	6.4358	6.4377
Cell volume / Å^3		272.12	272.58	272.88	273.08

Table S2. Analysis of VO_2 and Se-doped VO_2 by inductively coupled plasma-mass spectrometry (ICP-MS).

Sample	**Raw material Ratio (VO_2/Se wt%)**	**Vanadium (mg/L)**	**Selenium (mg/L)**	**Se/V (wt. %)**	**Se/V (at. %)**
VO_2	/	60.64	/	/	/
VO_2-Se1	4:1	57.93	0.88	1.52	1.0
VO_2-Se2	2:1	56.17	0.95	1.69	1.1
VO_2-Se3	4:3	55.41	1.09	1.97	1.3

References

1. M. Li, M. Liu, Y. Lu, G. Zhang, Y. Zhang, Z. Li, Q. Xu, H. Liu and Y. Wang, Adv. Funct. Mater., 2024, **34**, 2312789.
2. X. Wang, Y. Wang, A. Naveed, G. Li, H. Zhang, Y. Zhou, A. Dou, M. Su, Y. Liu, R. Guo and C. C. Li, Adv. Funct. Mater., 2023, **33**, 2306205.
3. K. Wang, S. Li, X. Chen, J. Shen, H. Zhao and Y. Bai, ACS Nano, 2024, **18**, 7311-7323.
4. S. Kong, Y. Li, X. Zhang, Z. Xu, X. Wang, Y. Feng, W. Gong, C. Liu, K. Tian and Q. Li, Small, 2023, **19**, 2304462.