*Supporting Information*

# Solvothermal guided $V_2O_5$ microspherical nanoparticles constructing high performance aqueous zinc-ion batteries

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## Experimental Section

*Materials characterization*

The crystal structure of the $V_2O_5$ samples were analyzed by X-ray diffraction (XRD) and the morphology and elemental content of $V_2O_5$ was investigated by transmission electron microscopy (TEM), scanning electron microscopy (SEM) and X-ray energy spectrometry (EDS). X-ray photoelectron spectroscopy (XPS) was carried out with Thermo Scientific K-Alpha to characterize the surface chemistry of the electrode materials and the valence state of vanadium. Fourier transform infrared spectroscopy (FT-IR) was used to test the chemical bonding and molecular structure of the VOCH precursors and $V_2O_5$ samples. The composition and decomposition temperature of the ethanol or glycol complexes in the VOCH precursor were studied by thermogravimetric analysis (TGA/DSC) in an air atmosphere in the temperature range 0~500 °C at a heating rate of 10 °C $min^{-1}$. The specific surface area was determined using a fully automated specific surface area and porosity analyzer according to the Brunner-Emmett-Taylor (BET) method.

*Electrochemical measurement*

Charge-discharge cycles of the cells were tested at room temperature using a LAND-CT3002A multi-channel battery test system. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) tests were carried out on Zn||$V_2O_5$ cells using an electrochemical workstation (CHI 660E) with scanning voltage from 0.4 to 1.4 V and AC impedance test at open circuit voltage.

The diffusion coefficient of $Zn^{2+}$ ($D_{Zn}$) was calculated according to the following equation：

$$D_{Zn}=\frac{4L^2}{\pi\tau}\left(\frac{\Delta E_s}{\Delta E_t}\right)^2 \qquad \text{(S1)}$$

In Equation (S1), τ is the pulse duration of the constant current, L is the $Zn^{2+}$ diffusion path, $\Delta E_s$ is the voltage change of the termination voltage of two adjacent relaxation steps and $\Delta E_t$ is the voltage difference during the current pulse, subtracting the IR drop.

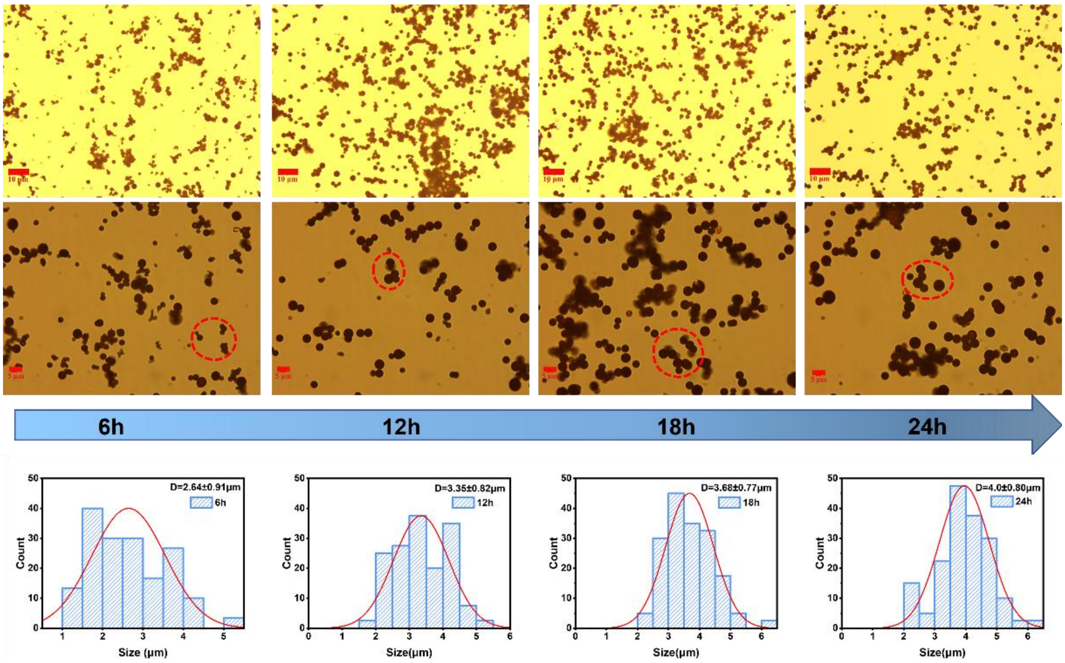


**Figure S1.** Size of the $V_2O_5$-20 microspheres at different reaction times (from 6 h to 24 h).

It can be seen for Figure S1 that the microspheres became larger and more uniform in size as the reaction time increased. The average diameter of the synthesized microspheres was 4 μm at the maximum reaction time of 24 h.

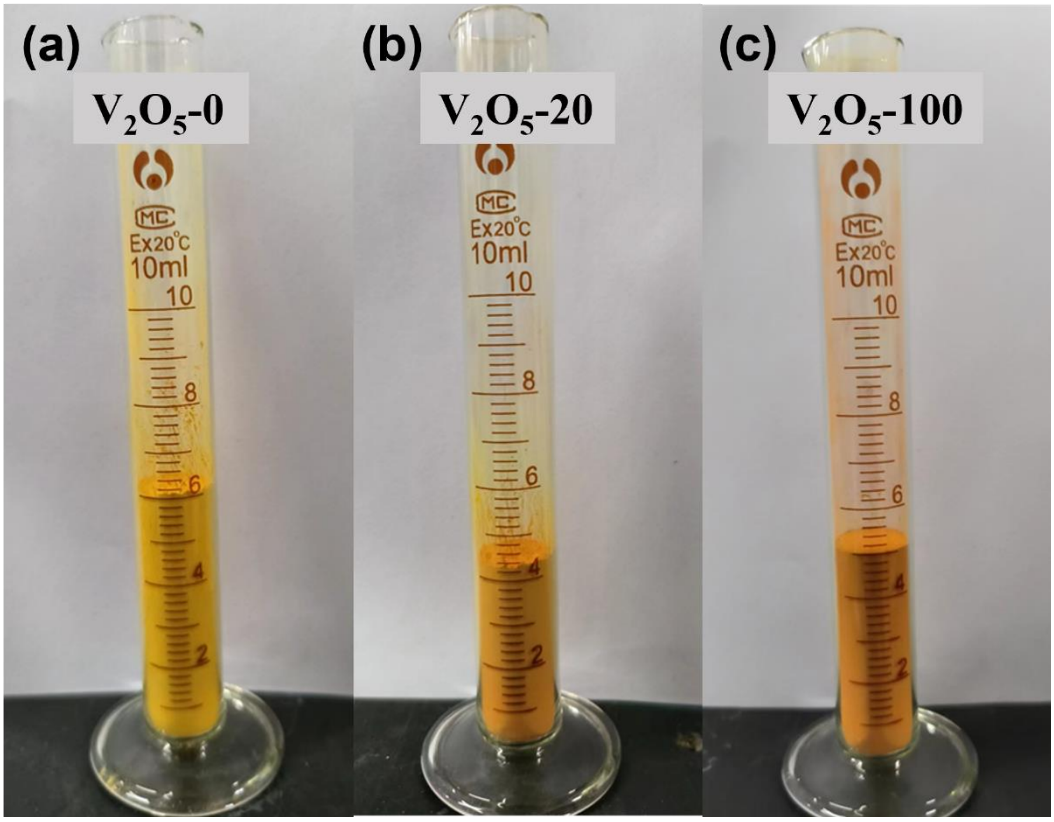


**Figure S2.** Volume comparison of (a) $V_2O_5$-0, (b) $V_2O_5$-20, and (c) $V_2O_5$-100 with the same mass (5 g).

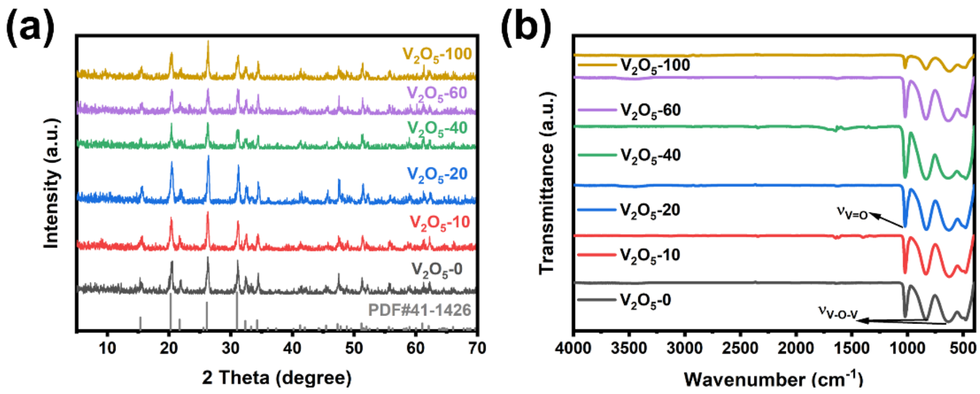


**Figure S3.** XRD patterns (a) and FT-IR spectra (b) of $V_2O_5$ samples.

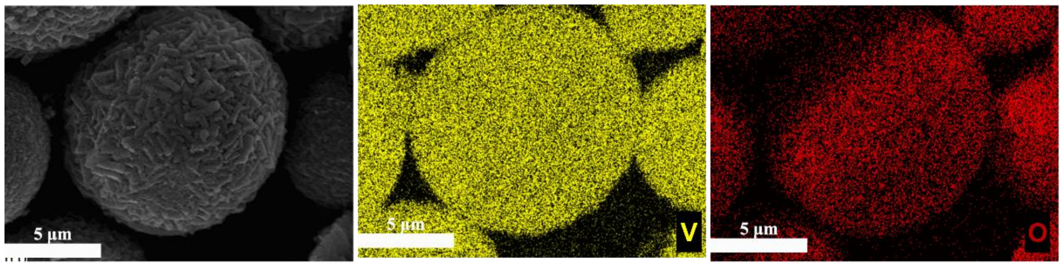


**Figure S4.** Elemental distribution of $V_2O_5$-20.

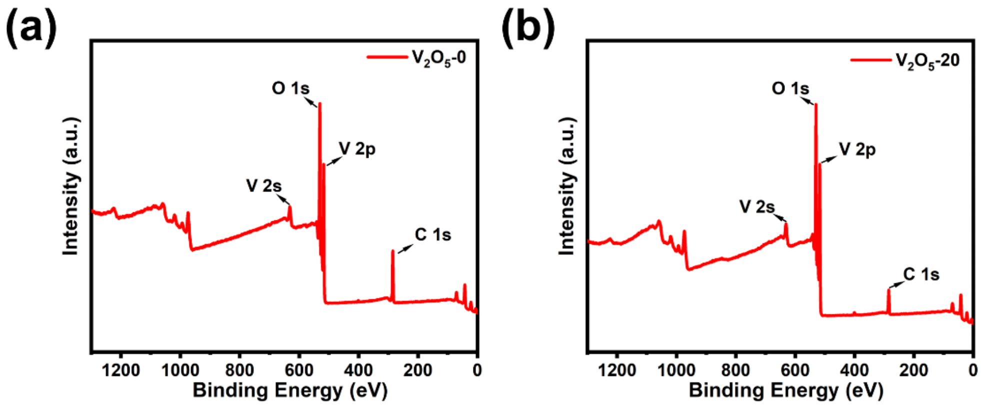


**Figure S5.** XPS survey spectra of (a) $V_2O_5$-0, (b) $V_2O_5$-20.

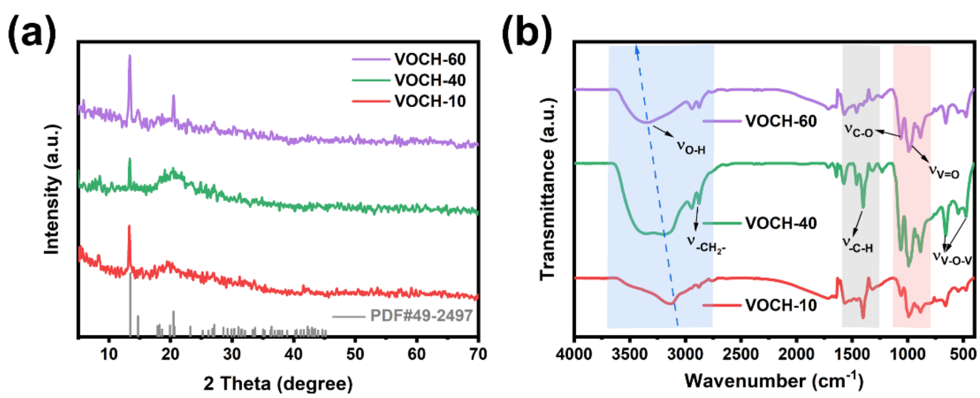


**Figure S6.** XRD patterns (a) and FT-IR spectra (b) of VOCH precursors.

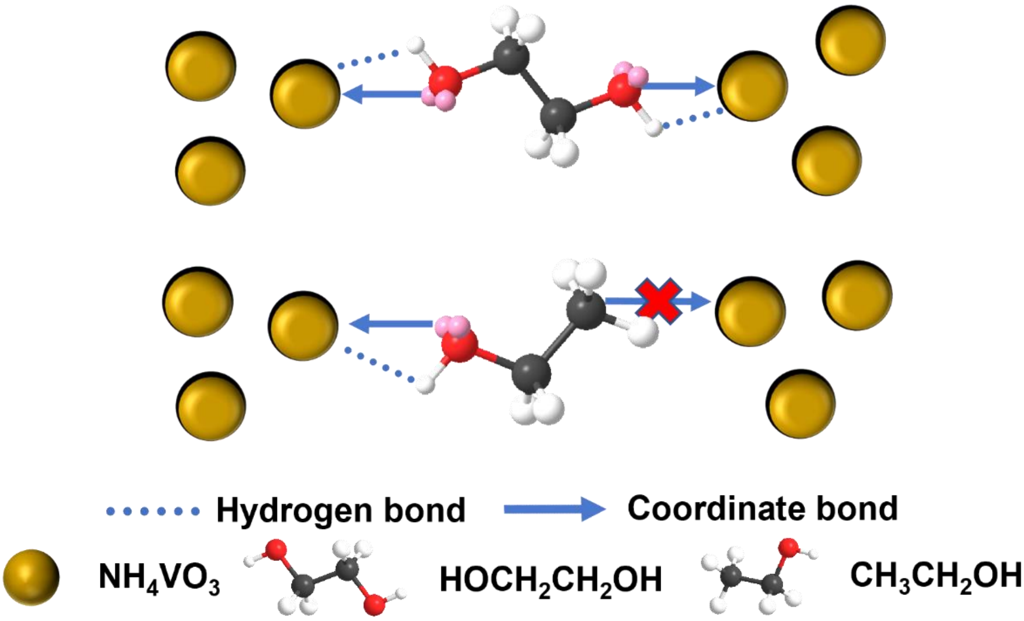


**Figure S7.** Schematic illustration of the chaining of ethylene glycol or ethanol with nanoparticles.

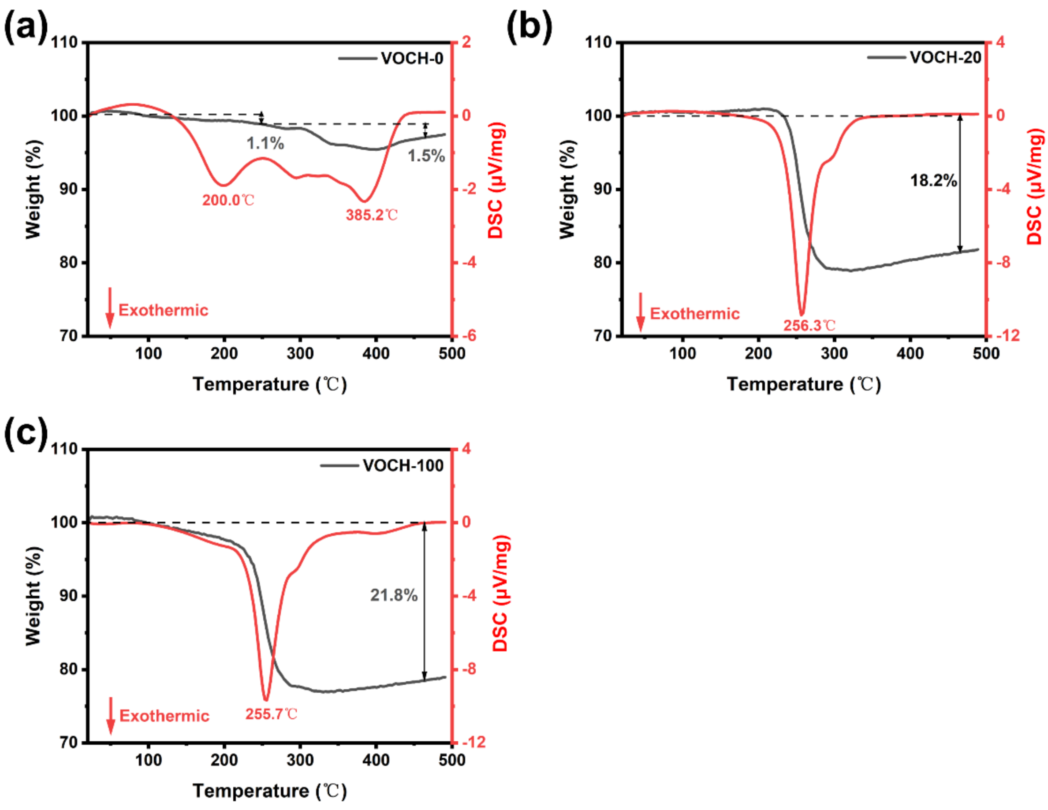


**Figure S8.** TGA/DSC profiles of (a) VOCH-0, (b) VOCH-20 and (c) VOCH-100 before calcination.

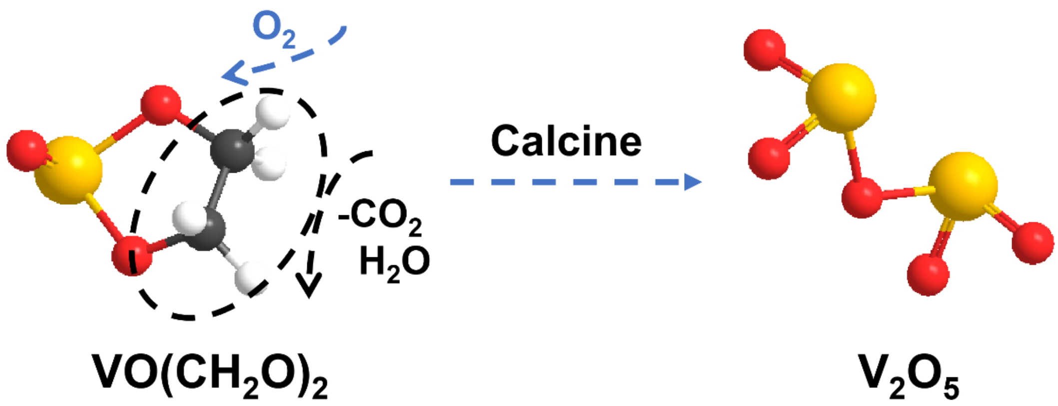


**Figure S9.** Schematic of $V_2O_5$ formation by calcination of VOCH precursors.

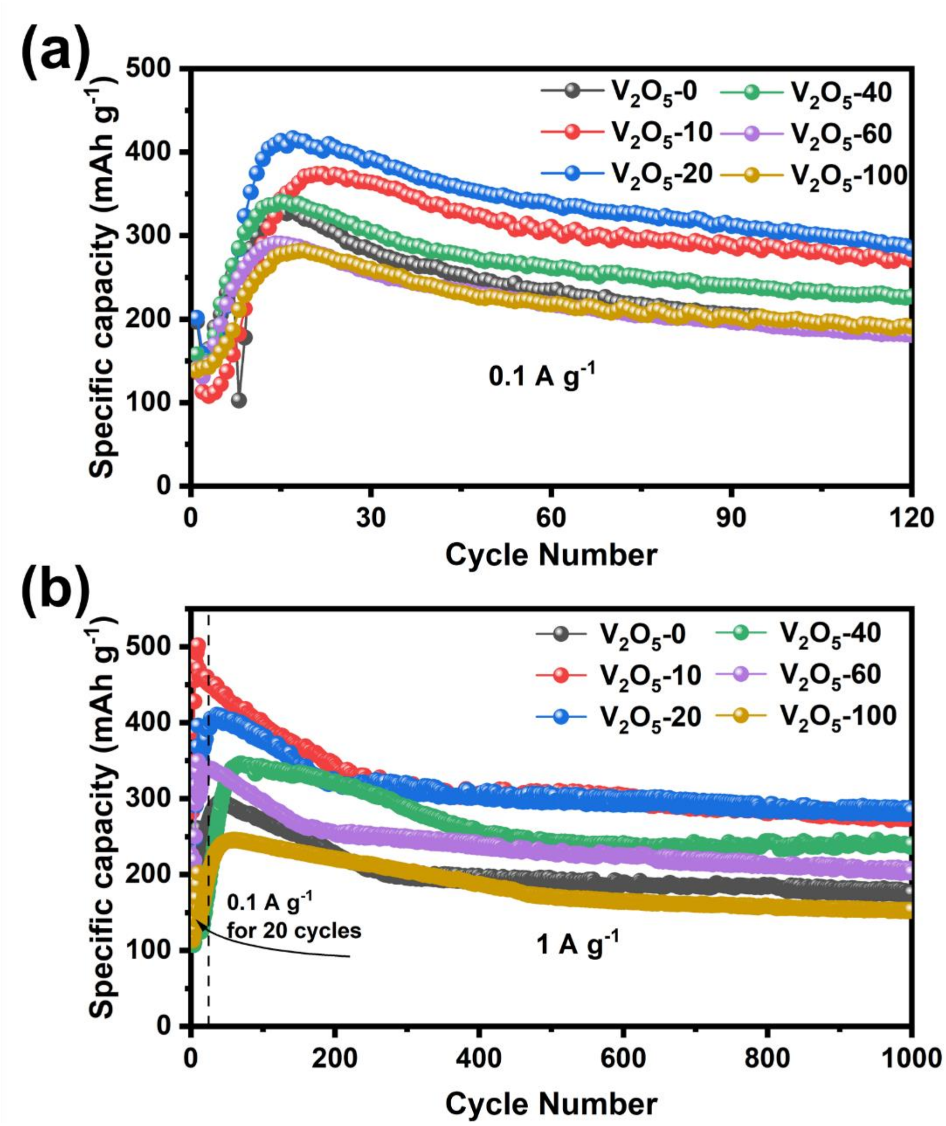


**Figure S10.** Cycle performance of different $V_2O_5$ electrodes at (a) 0.1 A $g^{-1}$, (b) 1 A $g^{-1}$.

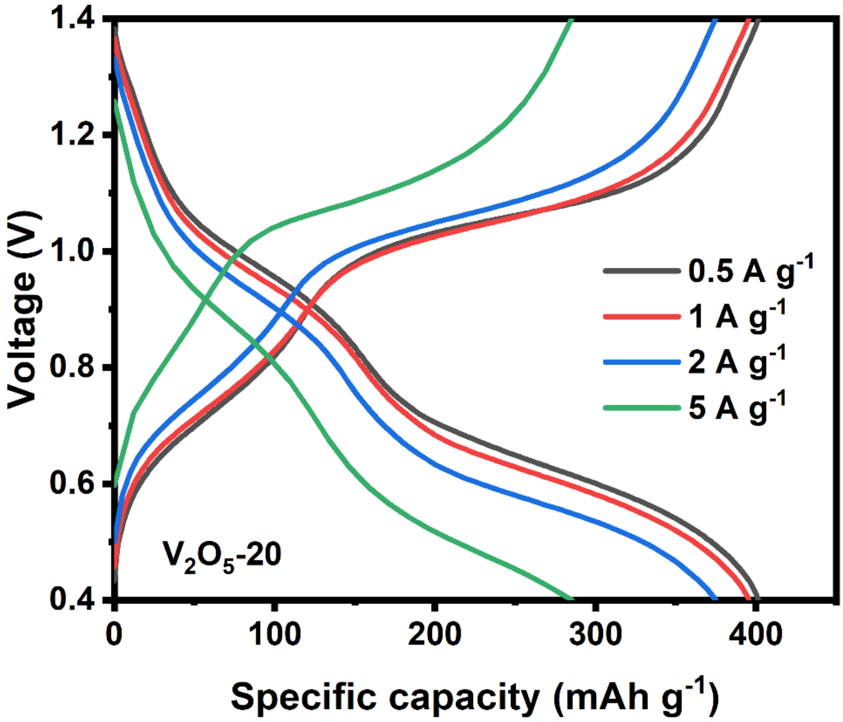


**Figure S11.** Specific capacity of $V_2O_5$-20 cathode at various current densities.

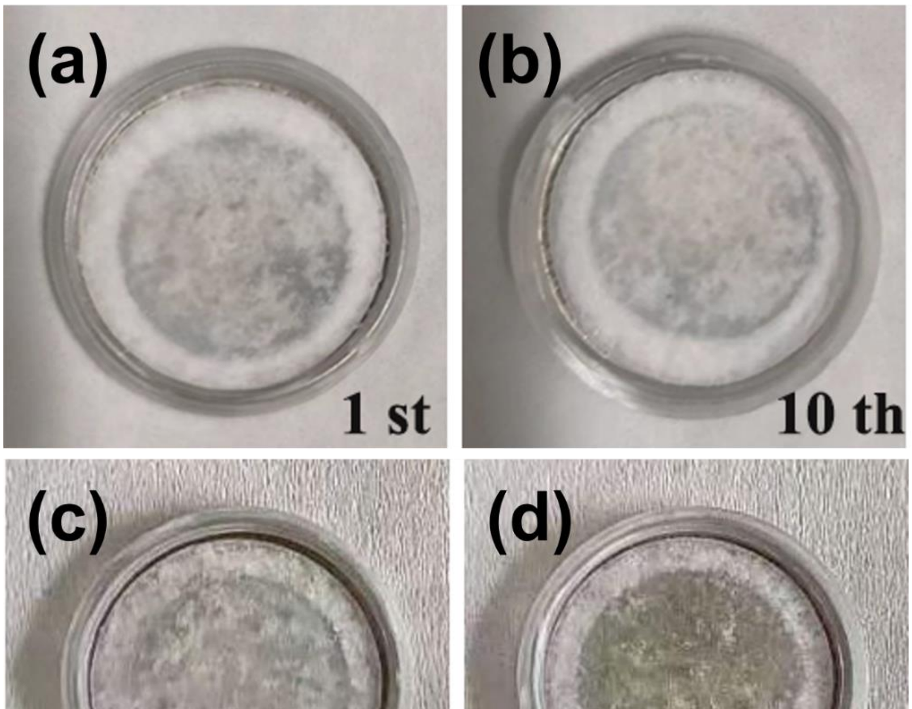


**Figure S12.** The pictures of battery separators after different recycle times: (a) 1st, (b) 10th, (c) 100th and (d) 500th.

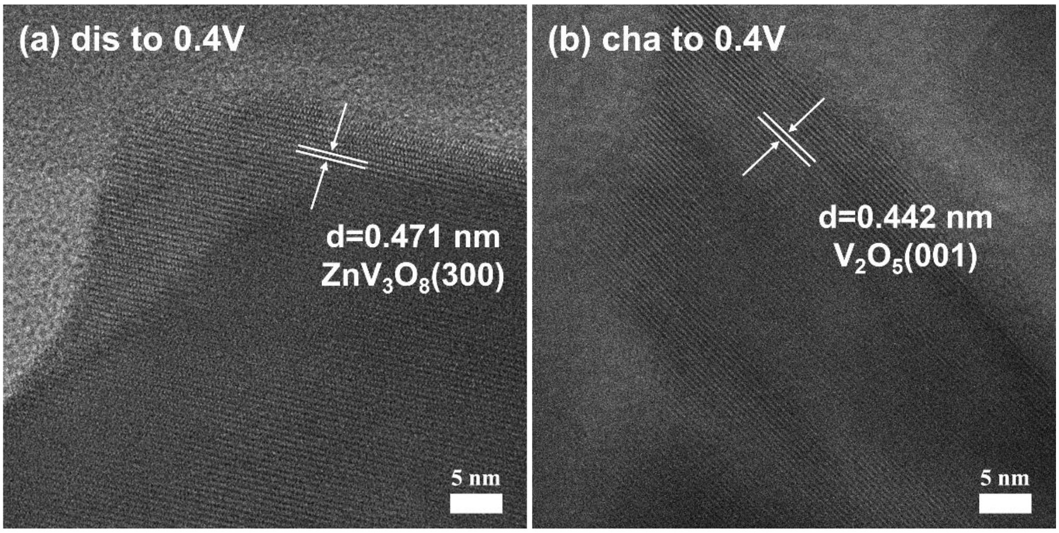


**Figure S13.** HRTEM images of $V_2O_5$-20 electrode (a) discharged to 0.4 V, (b) charged to 1.4 V.

**Table S1.** The BET surface area, pore volume and average pore size of $V_2O_5$-0 nanoparticles and $V_2O_5$-0 microspheres samples.

| Sample | BET surface area ($m^2g^{-1}$) | Pore volume ($cm^3\ g^{-1}$) | Average pore diameter(nm) |
|---|---|---|---|
| $V_2O_5$-0 | 6.8468 | 0.03272 | 10.1882 |
| $V_2O_5$-20 | 9.4024 | 0.04763 | 37.0358 |

**Table S2.** A survey of $V_2O_5$-based electrode materials with three-dimensional structures for AZIBs.

| Cathode material [Structure characteristic] | Electrolyte | Specific capacity | Cycling performance | Reference. |
|---|---|---|---|---|
| $V_2O_5$ (porous microspheres) | 3M $Zn(CF_3SO_3)_2$ | 401mAh/g (0.1 A/g) | 73% (1000) (2A/g) | [15] |
| $V_2O_5$ (hollow spheres) | Saturated $ZnSO_4$ | 280mAh/g (0.2 A/g) | 82% (6200) (10A/g) | [22] |
| $V_2O_5$ (nanospheres) | 3M $ZnSO_4$ | 327mAh/g (0.1 A/g) | 69% (6000) (10A/g) | [41] |
| $V_2O_5$@CNTs (irregular spherical) | 1 M $ZnSO_4$/1 $Na_2SO_4$ | 293mAh/g (0.3 A/g) | 72% (6000) (5A/g) | [42] |
| $VO_2$ (hollow nanospheres) | 3M $Zn(CF_3SO_3)_2$ | 440mAh/g (0.1 A/g) | 47% (860) (1A/g) | [43] |