Supporting information for

Porous V₂O₅ Yolk-Shell Microspheres for Zinc Ion Battery Cathode: Activation Responsible for Enhanced Capacity and Rate Performance

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Figure S1. SEM images of V_2O_5 -YS at a) low magnification and b) high

magnification.



Figure S2. SEM image of broken microsphere.



Figure S3. N_2 adsorption/desorption isotherms of commercial V_2O_5 and as-prepared V_2O_5 -YS at 77K.



Figure S4. SEM Images of V_2O_5 -YS cathode cycling for 100 cycles at the current density of 1.0 A g⁻¹ at a) low magnification and b) high magnification, where nanofibers are fragments of glass fiber membrane.



Figure S5. Cycling performance of the V₂O₅-YS cathode at current density of 0.2 A

g⁻¹.



Figure S6. Cycling performance of the pristine V_2O_5 at current density of 5 A g⁻¹.



Figure S7. The equivalent circuit used for fitting the EIS curves of Figure 4c, where $R_s = bulk$ resistance, $R_{ct} = charge$ transfer resistance, CPE = constant phase element, W=Warburg impedance.



Figure S8. Galvanostatic discharge curve of the V_2O_5 -YS cathode in a three-electrode cell in 5 mM H_2SO_4 electrolyte.



Figure S9. a) SEM image of V₂O₅-YS electrode soaked in aqueous electrolyte (nanoflakes: $Zn_3V_2O_7(OH)_2 \cdot 2H_2O$) b) Element quantification of nanoflakes and the ratio of Zn, V, O is accorded with $Zn_3V_2O_7(OH)_2 \cdot 2H_2O$.



Figure S10. The magnified profile of a) (004) plane in the first cycle. b) (007) plane in the 11st cycle

| Materials | Capacity | Cycling | Reference |
|---|---|-------------------------------|-------------------------|
| | | performance | |
| V_2O_5 -YS | 410mA h g ⁻¹ at | 80% after 1000 | This work |
| | 0.1A g ⁻¹ | cycles at 5A g ⁻¹ | |
| | 182mA h g^{-1} at | | |
| | 20A g ⁻¹ | | |
| V_2O_5 | 224mA h g ⁻¹ at | 37% after 400 | Chem. Commun., 2018, |
| | 0.1A g ⁻¹ | cycles at 1A g ⁻¹ | 54, 4457 |
| V_2O_5 | 132mA h g^{-1} at | 82% after 6000 | Electrochimica Acta 306 |
| | 10A g ⁻¹ | cycles at 10A g- | (2019) 307e316 |
| | | 1 | |
| V_2O_5 | $336 \text{mA} \text{ h g}^{-1} \text{ at}$ | 85% after 5000 | Nano Energy 60 (2019) |
| | 50mA g ⁻¹ | cycles at 10A g ⁻¹ | 171–178 |
| V ₂ O ₅ -CNT | 219mA h g ⁻¹ at | 80% after 500 | Nano Energy 60 (2019) |
| | 10A g ⁻¹ | cycles at 10A g ⁻ | 752–759 |
| | C | 1 | |
| $Mg_{0.34}V_2O_5 \cdot nH_2O$ | 81mA h g ⁻¹ at | 97% after 2000 | ACS Energy Lett. 2018, |
| | 5A g ⁻¹ | cycles at 5 A g ⁻¹ | 3, 2602–2609 |
| $Zn_{0.25}V_2O_5 \cdot nH_2O$ | 223mA h g ⁻¹ at | 82% after 1000 | Nature Energy 2016, 1, |
| | 4.5A g ⁻¹ | cycles at 4.5A g ⁻ | 16119 |
| | - | 1 | |
| Expanded | 222mA h g ⁻¹ at | 72% after 3000 | Nano Energy 62 (2019) |
| $V_2O_5 \cdot 2.2H_2O$ | 10A g ⁻¹ | cycles at 5A g ⁻¹ | 94-102 |
| V ₂ O ₅ ·nH ₂ O-Graphene | 248 mA h g ⁻¹ | 71% after 900 | Adv. Mater. 2018, 30, |
| | at 30A g ⁻¹ | cycles at 6A g ⁻¹ | 1703725 |
| Li _x V ₂ O ₅ ·nH ₂ O | 170 mA h g ⁻¹ | 67% after 50 | Energy Environ. Sci., |
| | at 10A g ⁻¹ | cycles at 1A g ⁻¹ | 2018,11, 3157-3162 |
| Ag _{0.4} V ₂ O ₅ ·nH ₂ O | 180 mA h g ⁻¹ | 74% after | Energy Storage |
| • | at 2A g ⁻¹ | 1000cycles at | Materials 18 (2019) 10- |
| | C | 5Å g ⁻¹ | 14 |
| $K_{2}V_{8}O_{21}$ | 247mA h g ⁻¹ at | 90% after 300 | Nano Energy 51 (2018) |
| | 0.3A g ⁻¹ | cycles at 6A g ⁻¹ | 579–587 |
| $NH_4V_4O_{10}$ | 150 mA h g ⁻¹ | 76% after 100 | J. Mater. Chem. A,2019, |
| | at 10A g^{-1} | cycles at 1A g ⁻¹ | 7, 940–945 |
| $Ca_{0.67}V_8O_{20}$ ·3.5H ₂ O | 291mA h g ⁻¹ at | 74% after 2000 | ACS Nano 2019, 13, 12, |
| | 5A g ⁻¹ | cycles at 5A g ⁻¹ | 14447-14458 |
| Ca _{0.25} V ₂ O ₅ ·nH ₂ O | 72mA h g^{-1} at | 96% after 3000 | Angew. Chem. |
| | 5.76A g ⁻¹ | cycles at 5.76A | 10.1002/ange.201713291 |
| | - | g ⁻¹ | |

Table S1. Comparison of battery performance of V_2O_5 -YS with other V_2O_5 -based cathode materials

| Cell | $R_{\rm s}\left(\Omega ight)$ | CPE (µF) | $R_{\mathrm{ct}}\left(\Omega ight)$ | |
|------------------|-------------------------------|----------|-------------------------------------|--|
| Pristine | 0.6 | 18.3 | 380.0 | |
| After activation | 4.6 | 696.2 | 39.8 | |

Table S2. Simulated parameters from EIS curves in Figure 4c using equivalent circuitin Figure S7.

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Supplementary Note1: Discussion of the Galvanostatic Intermittent Titration Technique (GITT)

GITT tests were employed to determine the kinetic behavior of materials by calculating the Zn2+ diffusion coefficient based on the following equation:

$$D = \frac{4L^2}{\pi\tau} (\frac{\Delta E_s}{\Delta E_t})^2$$

Where L is diffusion length (cm) of Zn^{2+} , which is approximate to thickness of electrode here, τ is the relaxation time (s), and ΔE_s is the steady-state voltage change (V) by the current pulse. ΔE_t is the voltage change (V) during the constant current pulse after eliminating the iR drop. All of the parameters are illustrated as follows.

