

## Electronic Supplementary Information

### **Oxygenated copper vanadium selenide composite nanostructures as a cathode material for zinc-ion batteries with high stability up to 10 000 cycles**

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## **1. Experimental procedure**

### ***1.1. Materials***

Following chemicals, such as copper (II) nitrate trihydrate ( $\text{CuH}_6\text{N}_2\text{O}_9$ , Sigma Aldrich, 99%), selenious acid ( $\text{Se}_2\text{H}_2\text{O}_3$ , Sigma Aldrich, 99%), 2-methylimidazole ( $\text{C}_4\text{H}_6\text{N}_2$ , Sigma Aldrich, 99%), ammonia meta vanadate ( $\text{NH}_4\text{VO}_3$ , Sigma Aldrich, 99%), metallic Zn (Sigma Aldrich, 0.1 cm thickness), zinc trifluoromethanesulfonate ( $\text{Zn}(\text{CF}_3\text{SO}_3)_2$ , Sigma Aldrich, 98%), and ZnCF (Sigma Aldrich, 98%), were utilized without further purification. The carbon fiber cloth (CFC) textile was received from the Nara Cell-Tech Corp., South Korea. Distilled water was produced in our laboratory by using a Milli-Q water equipment.

### ***1.2. Material characterizations***

The phase confirmation of the prepared samples was analyzed by X-ray diffraction (XRD; Rigaku MiniFlex 600 X-ray diffractometer with  $\text{Cu K}_\alpha$  radiation). The morphology and fine structure of the powder sample were characterized by using a field-emission scanning electron microscope (FE-SEM; JEOL JSM-7500F) and a transmission electron microscope (TEM, JEOL- JEM). The valance states of the sample were examined by X-ray photoelectron spectroscopy (XPS; PHI 5000 VersaProbe ESCALAB 250xi).

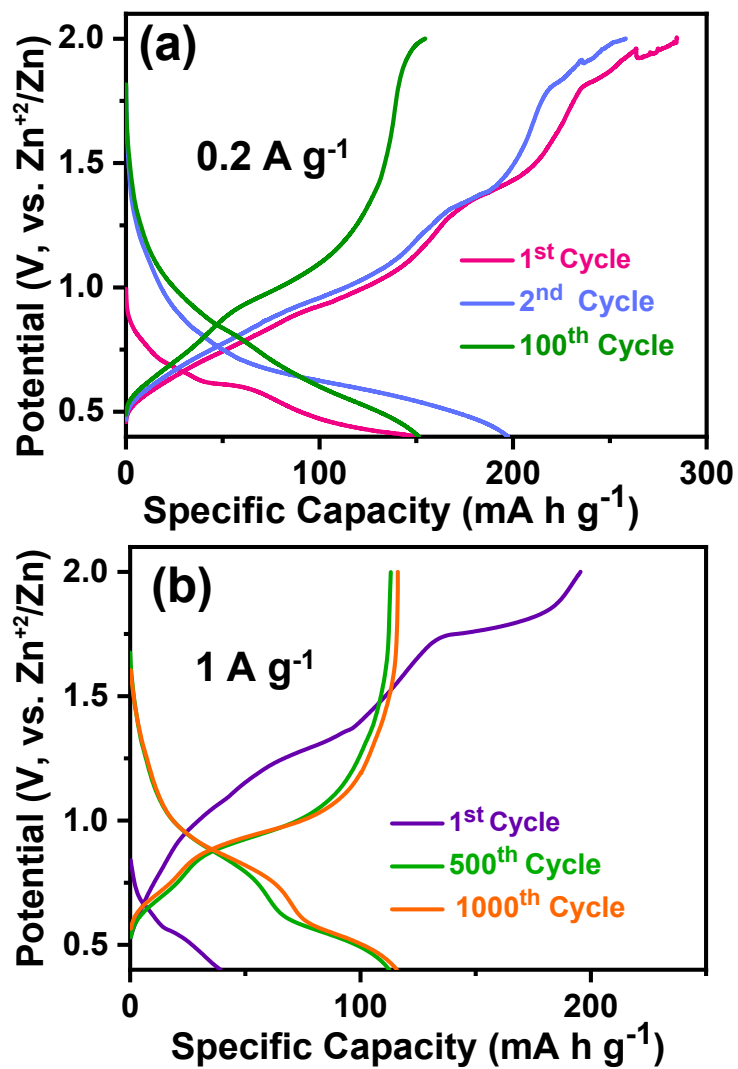
### ***1.3. Electrochemical measurements***

The O-CuVSe cathode material slurry was prepared by mixing 70 wt% of active material, 15 wt% polyvinylidene fluoride, and 15 wt% of super P carbon in N-methyl 2 pyrrolidinone solvent. The well-mixed slurry was coated on a CFC substrate using a brush. The electrode was dried at 80 °C for 12 h in a vacuum over. After proper drying, O-CuVSe coated CFC was cut into circular discs. The mass loading of active material over the CFC substrate is around 1–2  $\text{mg cm}^{-2}$ . The CR2032 coin-type cells were assembled in an air atmosphere using 2M ZnCF as an electrolyte, Zn metal

was employed as an anode, and GF/D glass fiber membrane (Whatman) was used as a separator. The galvanostatic charge-discharge (GCD) curves at various current densities and cyclic voltammetry (CV) curves at various scan rates were measured using the Wonna Tech battery cycler between the potential window of 0.4-2.0 V (vs. Zn/Zn<sup>2+</sup>).

**Table S1.** Comparison of the electrochemical performance of the O-CuVSe cathode material along with the previously reported cathode materials.

Cathode material	Electrolyte	Voltage (V)	Reversible capacity (mA h g <sup>-1</sup> )	Current density (A g <sup>-1</sup> )	Cycles	Ref.
Cu <sub>2-x</sub> Se	2M ZnSO <sub>4</sub>	0.4–1.6	70	5	3000	1
ZnMn <sub>2</sub> O <sub>4</sub> /Mn <sub>2</sub> O <sub>3</sub>	2M ZnSO <sub>4</sub>	0.8–1.9	80	1	1000	2
ZnMn <sub>2</sub> O <sub>4</sub>	2M ZnSO <sub>4</sub> with 0.05M MnSO <sub>4</sub>	0.8–1.9	106.5	0.1	300	3
Ni <sub>x</sub> Mn <sub>3-x</sub> O <sub>4</sub>	(2M ZnSO <sub>4</sub> with 0.15M MnSO <sub>4</sub>	1.0-1.8	128.8	0.4	850	4
ZnNi <sub>0.5</sub> Mn <sub>0.5</sub> CoO <sub>4</sub> @C	2M ZnSO <sub>4</sub> with 0.1M MnSO <sub>4</sub>	1.0-1.8	110	0.2	500	5
MoS <sub>2</sub>	3M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	0.2-1.3	125	2	500	6
MoS <sub>2</sub> @N	3M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	0.2-1.3	100	1	600	7
MoO <sub>3</sub>	1M ZnSO <sub>4</sub>	0.2-1.9	35	1	1000	8
VS <sub>2</sub> @N-doped carbon	3 M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	0.2-1.8	144	1	600	9
α-MnO <sub>2</sub> @C	1M ZnSO <sub>4</sub>	1.0-1.8	189	0.066	50	10
MnO@NGS	2M ZnSO <sub>4</sub>	0.9 -1.8	112.3	0.5	300	11
Cu <sub>3</sub> V <sub>2</sub> O <sub>7</sub> (OH) <sub>2</sub> ·2H <sub>2</sub> O	2.5M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	0.2-1.6	100.1	0.2	130	12
CuHCF	1M ZnSO <sub>4</sub>	1.4-2.0	44	0.1	50	13
CuHCF	1M ZnSO <sub>4</sub>	0.8-1.9	40	0.02	20	14
CuHCF	20mM ZnSO <sub>4</sub>	1.4-2.1	53	1	50	15
ZnHCF	1M ZnSO <sub>4</sub>	0.8-2.0	49.4	0.06	100	16
ZnHCF@MnO <sub>2</sub>	0.5M ZnSO <sub>4</sub>	1.4-1.9	70	0.5	1000	17
ZnNi <sub>1/2</sub> Mn <sub>1/2</sub> CoO <sub>4</sub>	0.3M Zn (OTf) <sub>2</sub> in MeCN	0.9-2.15	174	1	200	18
Mo <sub>6</sub> S <sub>8</sub>	1 M ZnSO <sub>4</sub>	0.25-1.0	87.4	0.18	150	19
CC@MnO <sub>2</sub> @MXene	0.1M Mn(CH <sub>3</sub> COO) <sub>2</sub> with 0.01M Na <sub>2</sub> SO <sub>4</sub>	0.8-1.9	80.6	1	800	20
<b>O-CuVSe</b>	<b>2M Zn(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub></b>	<b>0.4-2.0</b>	<b>166.6 117</b>	<b>1 2 5</b>	<b>1000 2000 10000</b>	<b>This work</b>



**Fig. S1** Cycling performance of the O-CuVSe cathode at (a) 0.2 A g<sup>-1</sup> and (b) 1 A g<sup>-1</sup>.

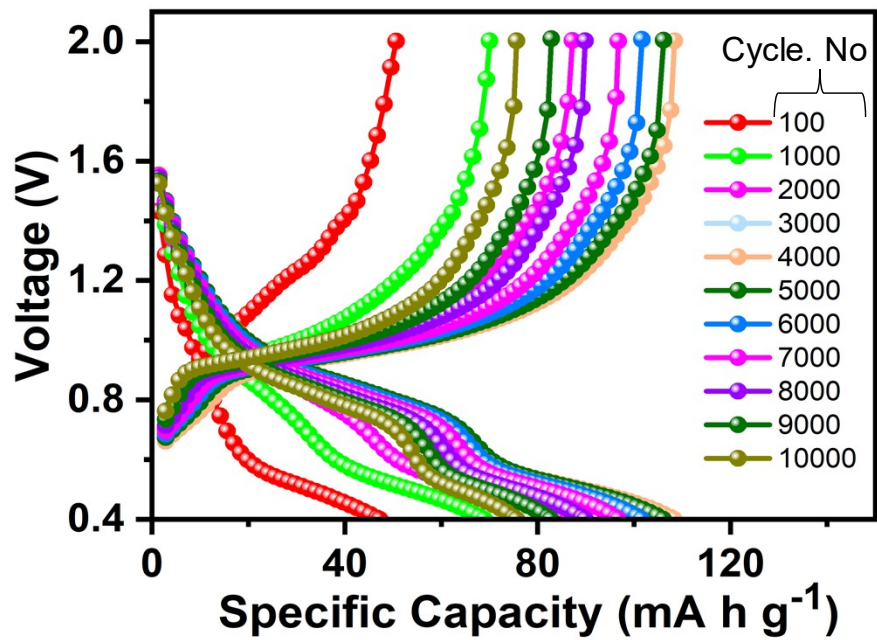
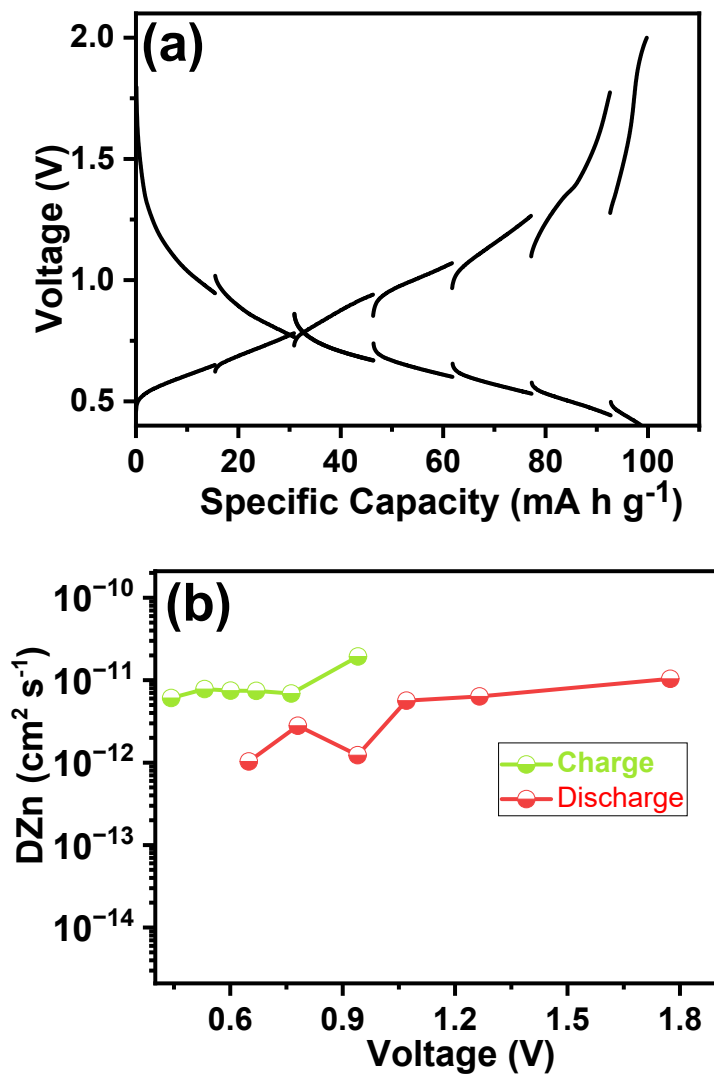
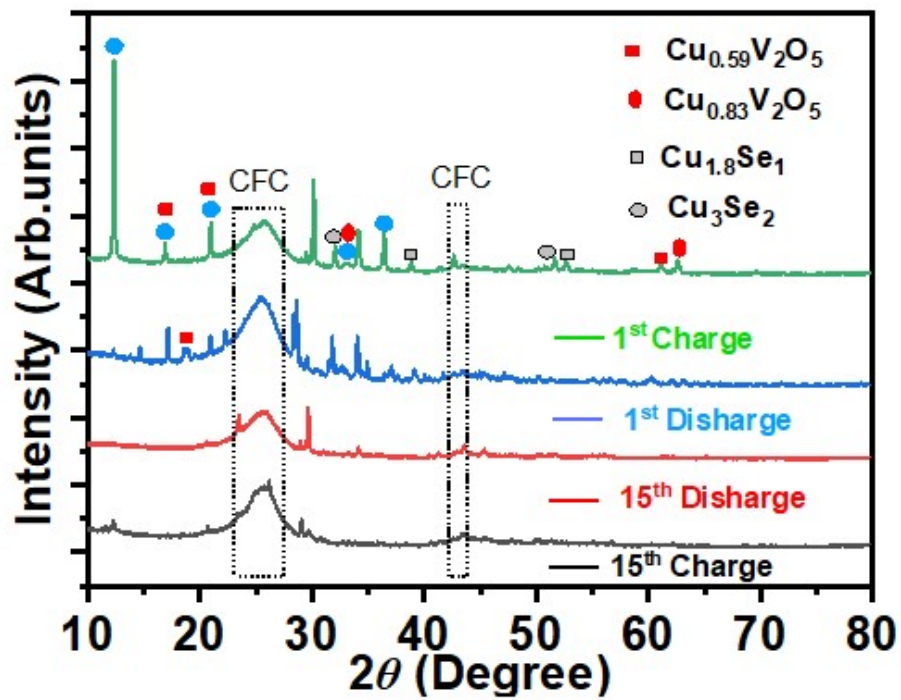


Fig. S2 Voltage profile curves of the O-CuVSe cathode at 5 A g<sup>-1</sup>.

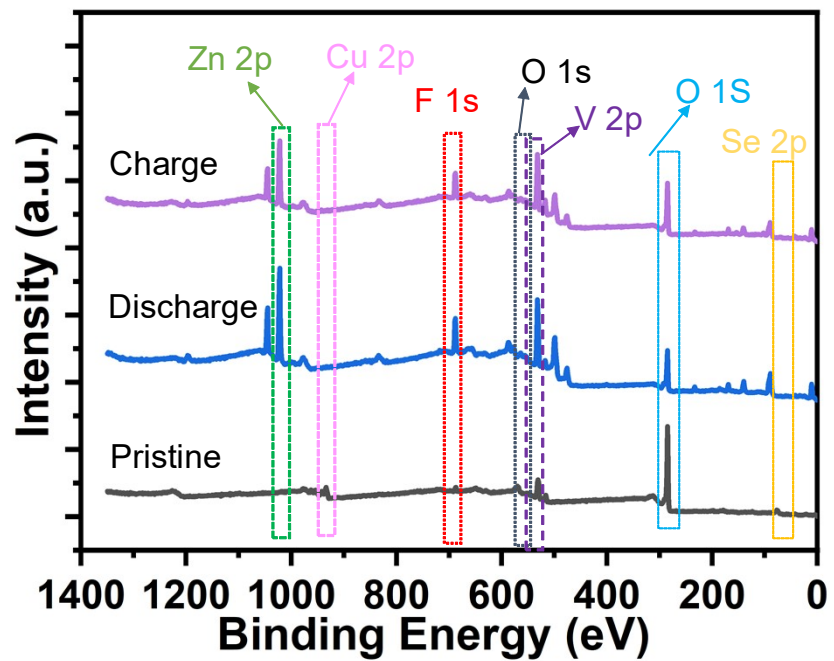


**Fig. S3** (a) GITT curves and (b) corresponding diffusivity coefficient for Zn<sup>2+</sup> in Zn/O-CuVSe battery vs. voltage.

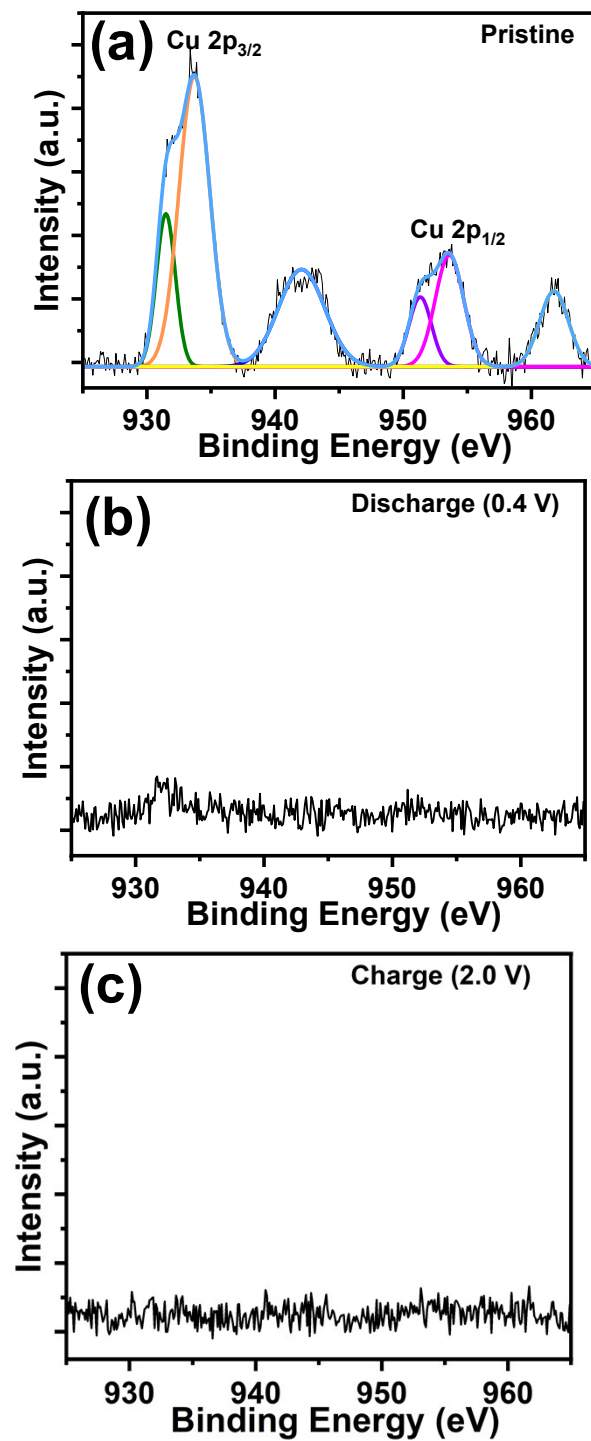


**Fig. S4** Ex-situ XRD patterns of the O-CuVSe composite cathode measured after the 1<sup>st</sup> charge/discharge and 15<sup>th</sup> charge/discharge cycles.

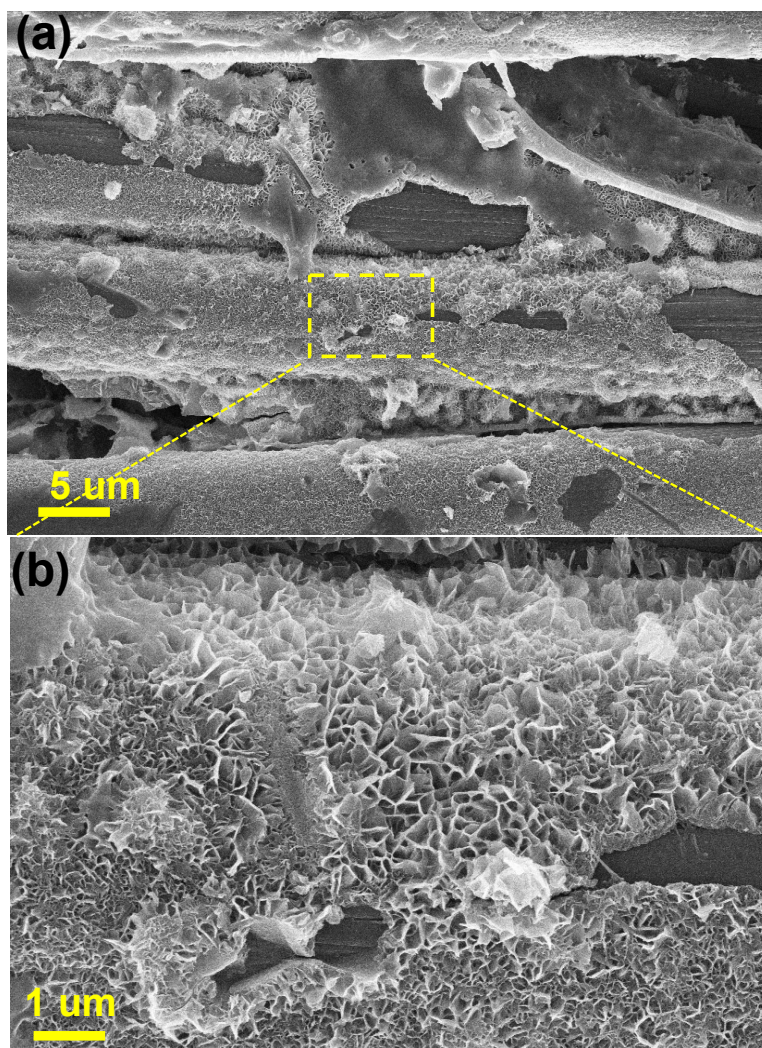




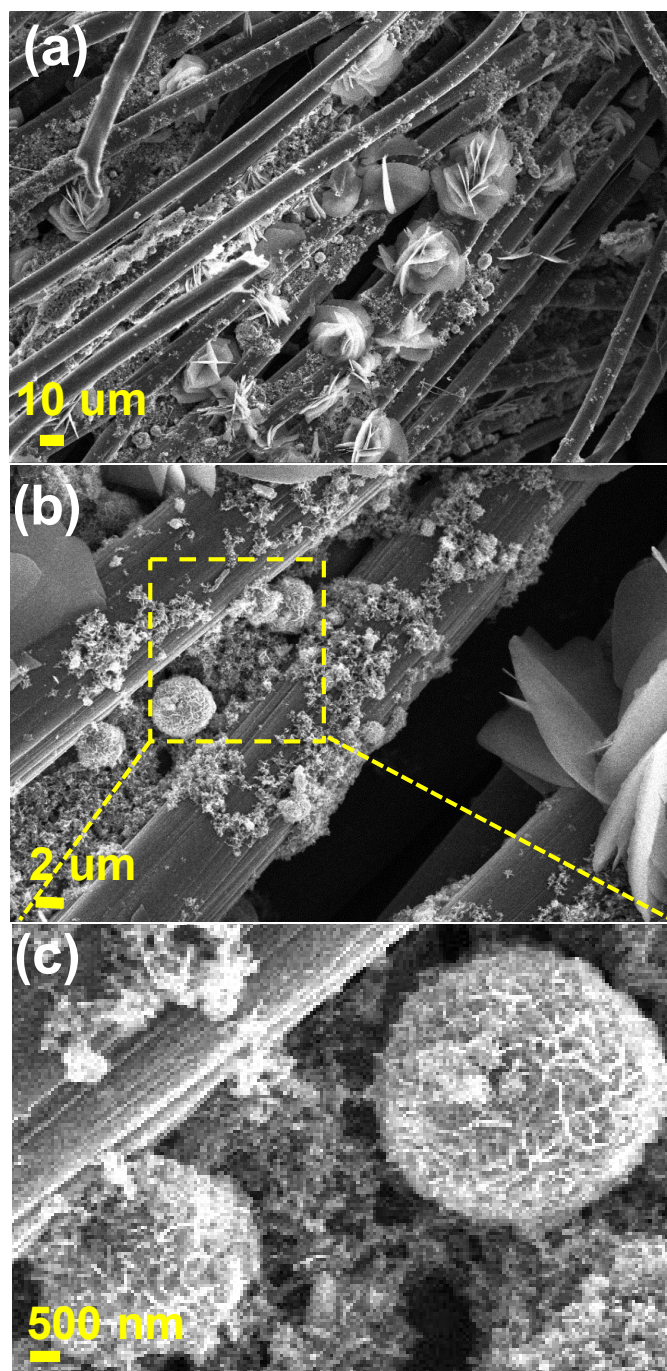
**Fig. S5** XPS survey scan spectra of the O-CuVSe cathode in pristine, charge, and discharge states.



**Fig. S6** High-resolution Cu 2p XPS spectra: (a) pristine state, (b) discharge state, and (c) charge state.



**Fig. S7** (a) Low- and (b) high-magnification FE-SEM images for the O-CuVSe cathode after 2000 cycles at 2 A g<sup>-1</sup>.



**Fig. S8** (a-c) FE-SEM images under different magnifications for the O-CuVSe cathode after 10000 cycles at 5 A g<sup>-1</sup>.

## References

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