# **Supporting Information for:**

## Construction of waffle-like NS-ZIF@V<sub>2</sub>CT<sub>x</sub> heterostructures for

## high-performance potassium ion batteries

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### **1. Experimental Method**

#### 1.1 Materials

The materials used here include:V<sub>2</sub>AlC (98 wt%, 400 mesh, Ichiyi Technology Co. Ltd.), TMAOH (tetramethylammonium hydroxide, 25 wt%, Macklin Co. Ltd.), methanol (99.5%), polyvinylpyrrolidone (PVP, GR), and ascorbic acid were purchased from Sinopharm Chemical Reagent Co. HCl (36-38 wt%), Zn(NO<sub>3</sub>)<sub>2</sub>-6H<sub>2</sub>O (99.99%), Co(NO<sub>3</sub>)<sub>2</sub>-6H<sub>2</sub>O (99.99%), dimethylimidazole (98%), potassium metal from potassium ion batteries (K,99.9%),1,2dimethoxyethane (DME, AR, 99.5%) and potassium bis(fluorosulfonyl)imide (KFSI, AR, > 95%) were purchased from Aladdin Industries.

### 1.2 Materials characterization

The morphology of the resulting products was observed and photographed using a scanning electron microscope (SEM) (Hitachi SU-8010, Japan) at a deceleration voltage of 1.5 kV. Transmission electron microscopy images were taken using a JEOL JEM-2010 LaB<sub>6</sub> high-resolution transmission electron microscope (200 kV). High resolution transmission electron microscope (200 kV). High resolution transmission electron microscope at 200 kV. X-ray diffraction (XRD) patterns were recorded using a Bruker AXS D8 Advance X-ray diffractometer with a cu-k- $\alpha$  radiation source ( $\lambda = 1.5406$  Å). Raman spectra were collected using a DXR Raman confocal microscope (Thermo Fisher Scientific, USA), 532 nm @10 mW laser. X-ray photoelectron spectroscopy (XPS) measurements were performed using Al K $\alpha$  radiation as the excitation source (VG Multilab 2000 Photoelectron Spectrometer, 2 × 10-6 Pa vacuum).

#### 1.3 Electrochemical characterization

A CR2032 button cell was selected to test various electrochemical properties. The mixtures consisted of prepared material (70 wt%), polyvinylidene fluoride (PVDF, 10 wt%) and cochineal black (20 wt%). The mixtures were then applied to a copper foil (collector) to prepare V<sub>2</sub>CT<sub>x</sub>, ZIF@V<sub>2</sub>CT<sub>x</sub> and NS-ZIF@V<sub>2</sub>CT<sub>x</sub> electrodes (mass loadings of  $0.2 \text{ mg cm}^{-2}$  and diameters of 8 mm). Potassium metal foil was used as the reference electrode for the assembly of the semi-PIBs, and polypropylene membranes (Celgard 2400) were used for the separator. The electrolyte was selected as 3 M KFSI (DME) and 200 µL of each CR2032 button cell was used. Charge-discharge tests and GITT tests were performed using a NEWARE battery tester (0.01-3 V, room temperature, 0.1 mV s<sup>-1</sup> ~ 1.0 mV s<sup>-1</sup>) and EIS spectra (5 mV, 0.01 Hz ~ 100 kHz) tests were performed on the IviumStathtype electrochemical workstation.



Figure S1. SEM images of (a) V<sub>2</sub>CT<sub>x</sub>, (b) NS-ZIF@V<sub>2</sub>CT<sub>x</sub>, TEM images of (c) NS-ZIF@V<sub>2</sub>CT<sub>x</sub>, and (d) NS-ZIF



**Figure S2.** TEM images (a) ZIF@V<sub>2</sub>CT<sub>x</sub>, (b) NS-ZIF@V<sub>2</sub>CT<sub>x</sub>, (c) ANS-ZIF@V<sub>2</sub>CT<sub>x</sub>, SAED diffraction pattern of (d) ZIF@V<sub>2</sub>CT<sub>x</sub>, (e) NS-ZIF@V<sub>2</sub>CT<sub>x</sub>, (f) ANS-ZIF@V<sub>2</sub>CT<sub>x</sub>.



Figure S3. EDS elemental analysis map of NS-ZIF@V<sub>2</sub>CT<sub>x</sub>.



Figure S4. (a) TEM image, (b) EDS image of ZIF@V<sub>2</sub>CT<sub>x</sub>.



Figure S5. N<sub>2</sub> adsorption-desorption isotherms and pore size distribution of (a)  $V_2CT_x$ , and (b)  $ZIF@V_2CT_x$ .



Figure S6. Thermogravimetric curve of NS-ZIF@V<sub>2</sub>CT<sub>x</sub>.



**Figure S7**. (a) Cycling performance of  $V_2CT_x$ , ZIF@ $V_2CT_x$ , NS-ZIF@ $V_2CT_x$ , and NS-ZIF@ $V_2CT_x$  electrodes at current density of 100 mA g<sup>-1</sup>, Cycling performance of ANS-ZIF@ $V_2CT_x$  electrode at (b) 1000 mA g<sup>-1</sup> and (c) 2000 mA g<sup>-1</sup>.



Figure S8. GITT plots of V<sub>2</sub>CT<sub>x</sub>, NS-ZIF@V<sub>2</sub>CT<sub>x</sub> and ZIF@V<sub>2</sub>CT<sub>x</sub>.



Figure S9. The calculated diffusion coefficient (D\_{K}^{+}) of ZIF@V\_2CT\_x electrode.



 $\label{eq:Figure S10.} \mbox{(a) SEM image of NS-ZIF} @V_2CT_x \mbox{ electrode and (b) TEM image of SEI layer after} \\ 200^{th} \mbox{ charge/discharge cycles.}$ 



Figure S11. (a) SEM, (b) TEM, (c) EDS of ANS-ZIF@V<sub>2</sub>CT<sub>x</sub> samples.



Figure S12. (a) XRD patterns, (b) Raman spectra, (c) FT-IR spectra of  $V_2CT_x$ , ZIF@ $V_2CT_x$ , NS-ZIF@ $V_2CT_x$ , and NS-ZIF@ $V_2CT_x$ .



Figure S13. XPS data for ANS-ZIF@V<sub>2</sub>CT<sub>x</sub> sample of (a) XPS survey, (b) C 1s, (c) O1s, (d) V 2p, (e) Zn 2p, and (f) Co 2p spectrum.



**Figure S14**. (a) CV curves, (b) GCD profiles, (c) GDV profiles, (d) CV curves at different sweep speeds, (e) anode peaks corresponding to the b-value, and (f) capacitive contributions of ANS- $ZIF@V_2CT_x$  electrode.



Figure S15. (a) GITT test, (b) the calculated diffusion coefficient ( $D_K^+$ ) of ZIF@V<sub>2</sub>CT<sub>x</sub> electrode.



Figure S16. Plots of EIS data for (a) 0 cycles of EIS, (b) after 20 cycles of, and (c) the simulated  $R_{ct}$ , and  $R_{SEI}$  values.