### **Electronic Supplementary Information**

# Layer-by-Layer Stacked Vanadium Nitride Nanocrystals/N-doped

## Carbon Hybrid Nanosheets toward High-performance Aqueous Zinc-

#### ion Batteries

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Fig. S1. <sup>1</sup>H NMR spectrum of PV molecule.



**Fig. S2.** (a) The high-resolution SEM image of VN/NC. The morphology of "thin-sheet" and "rich-pores" can be seen clearly. (b) SEM image of VN-control.



**Fig. S3**. HRTEM image of VN/NC and VN-control. (a) the pores in the VN/NC are circled by the yellow dotted line. (b) the multilayered structure of VN/NC. It can be seen 3 layers at least in different focal planes from the top right to the bottom left. (c, d) morphology of VN-control.



Fig. S4. The AFM image of NC. The pores in the NC may be due to the etching of VN.



**Fig. S5.** The nitrogen adsorption-desorption isotherm curve and pore size distribution of VN/NC. The BET surface area is 56.4 m<sup>2</sup> g<sup>-1</sup> and pore size distribution is 2-50 nm.



Fig. S6. Charge–discharge curves of NC and VN/NC at a current density of 0.1 A  $g^{-1}$ .



**Fig. S7.** The electrochemical performance of VN-control. (a) Charge–discharge curves at a current density of 0.5 A  $g^{-1}$ , (b) Galvanostatic charge–discharge plots at different current densities, and (c) Rate performance.

The discharge capacities of VN-control are 476, 449, 408, 313, 243, 152 and 88 mA h g<sup>-1</sup> at current densities of 0.2, 0.5, 1.0, 2.0, 5.0, 10.0 and 20.0 A g<sup>-1</sup> respectively.



Fig. S8. EIS plot of VN-control and VN/NC based cells.



**Fig. S9.** The SEM images of (a) VN-control and (b) VN/NC after 500 cycles at 10 A g<sup>-1</sup>. The VNcontrol blocks have been broken into small pieces, while the VN/NC sheets still maintain their morphologies.



**Fig. S10.** The TEM images of VN/NC after 500 cycles at 10 A g<sup>-1</sup>. The right image is the high magnification of the white dotted-line area in the left image. The individual small VN nanocrystals on the layered NC has been preserved. The new appearing lattice spacing 0.281 nm, corresponding to the peak of 31.82° (20,  $\theta$  = 15.91°, calculated by the Bragg equation: 2*dsin* $\theta$  =  $n\lambda$ , d = 0.281 nm, n = 1 and  $\lambda = 0.15406$  nm) in the XRD test, should come from  $Zn_x(CF_3SO_3)_y(OH)_{2x-y}\cdot nH_2O$ , which is inevitably formed in the cathodes using the Zn(CF<sub>3</sub>SO<sub>3</sub>)<sub>2</sub> electrolyte<sup>1</sup>.

#### Notes and references

1 P. Oberholzer, E. Tervoort, A. Bouzid, A. Pasquarello and D. Kundu, *ACS Appl. Mater. Interfaces*, 2019, **11**, 674-682.