Supporting Information

$NH_4V_4O_{10}$/rGO Composite as High Performance Electrode Material for Hybrid Capacitive Deionization

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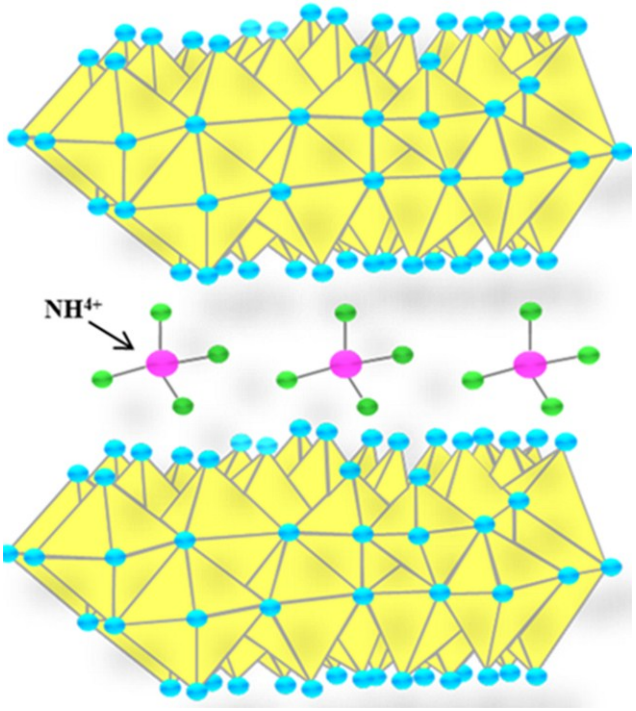

Fig. S1 Crystal structure diagram of $NH_4V_4O_{10}$.

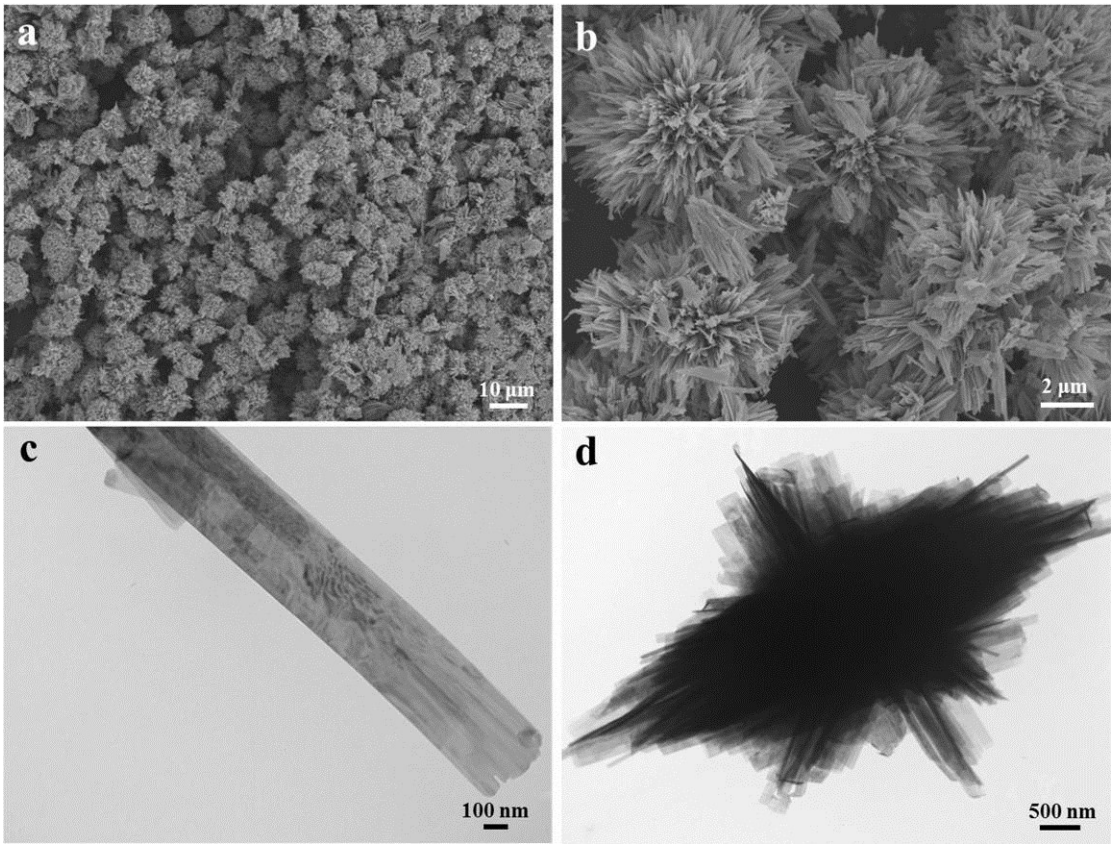

Fig. S2 (a, b) SEM images and (c, d) TEM images of NHVO.

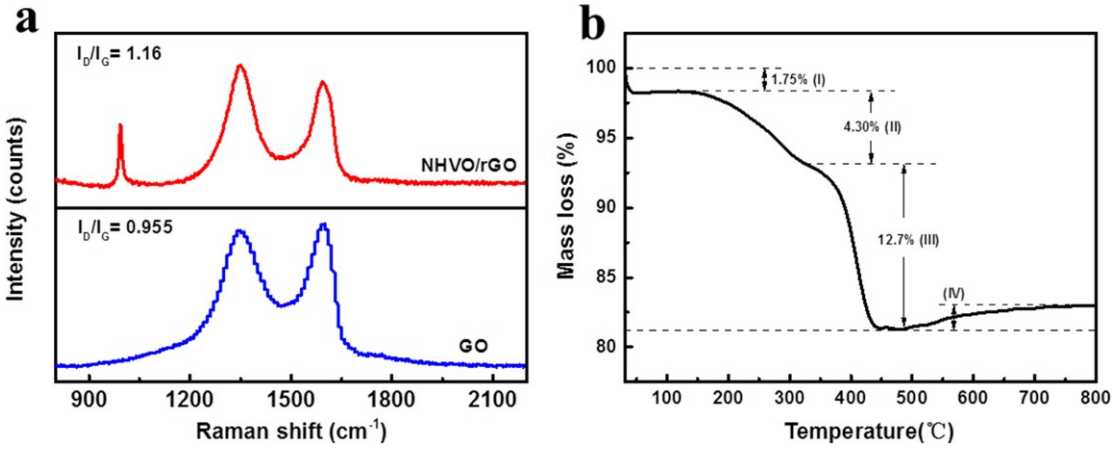

Fig. S3 (a) Raman spectra of NHVO/rGO composite and pure GO; (b) TGA curve of NHVO/rGO composite.

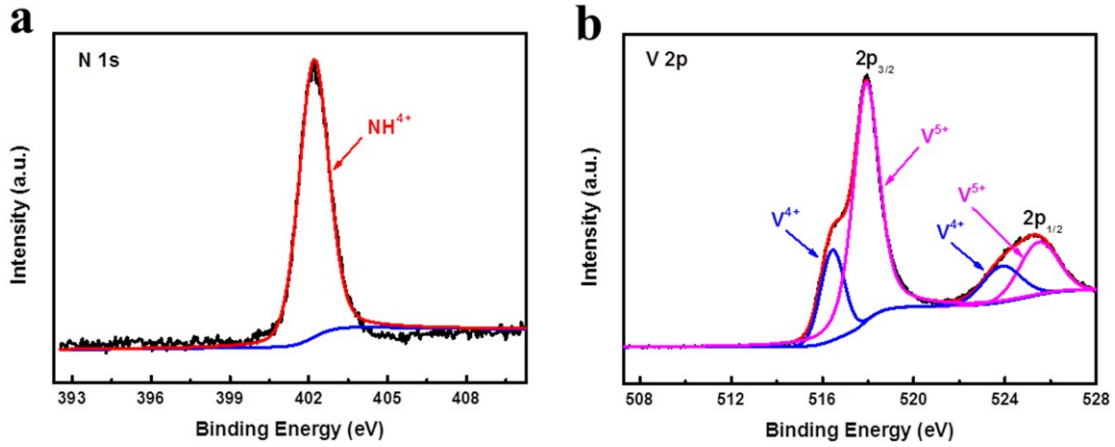

Fig. S4 (a) N 1s and (b) V 2p XPS spectrum of NHVO.

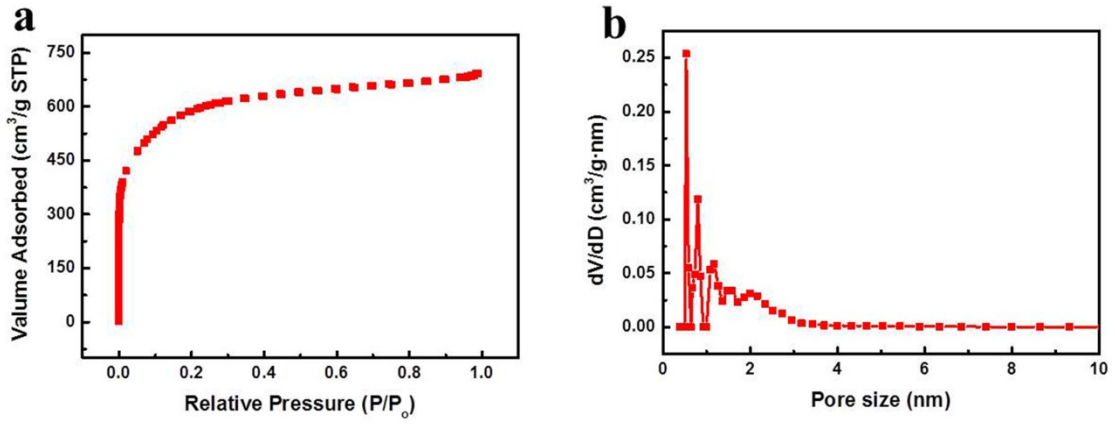

Fig. S5 (a) Nitrogen adsorption-desorption isotherm and (b) pore-size distribution of

AC.

The BET surface area of AC used in our CDI system is 2153.7 m^2/g. The pore size distribution is mainly microporous.

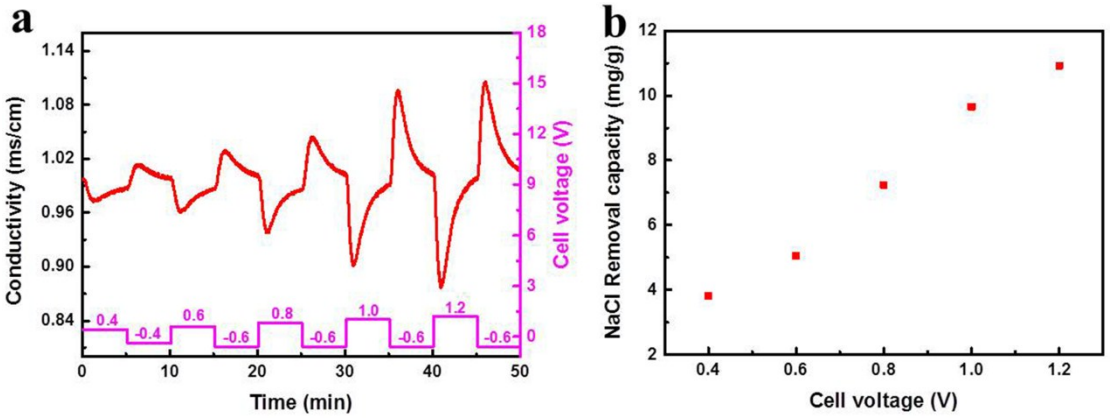

Fig. S6 (a) conductivity change curves of effluent and (b) NaCl removal capacity of rGO//AC cell (rGO as the cathode and AC as the anode) in 500 mg/L NaCl solution at different cell voltages. (The mass of active material (containing AC and rGO) is 45.0 mg. The thickness of AC electrode, and rGO electrode is about 255.2 μm and 326.1 μm, respectively.)

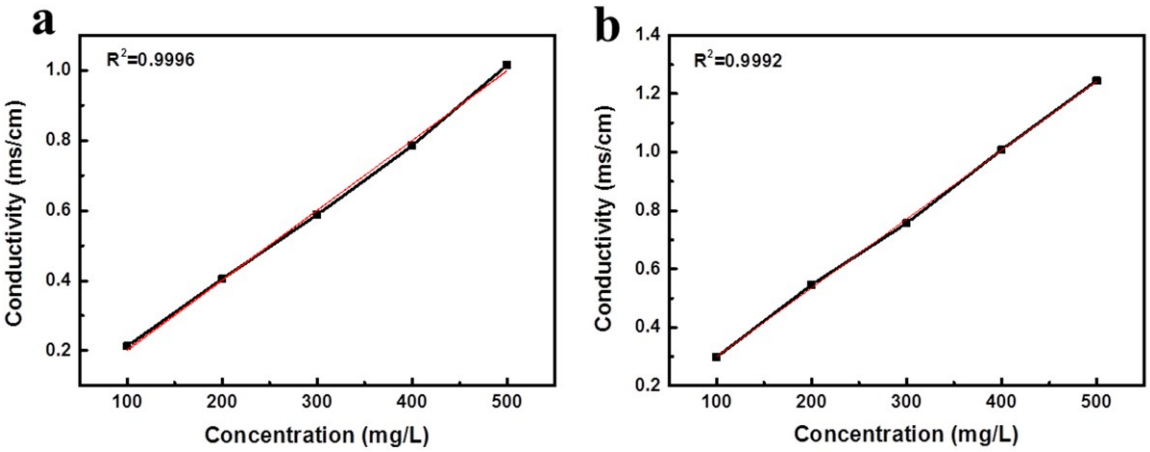

Fig. S7 (a) relationship between conductivity and concentration of $CaCl_2$; (b) relationship between conductivity and concentration of $MgCl_2$

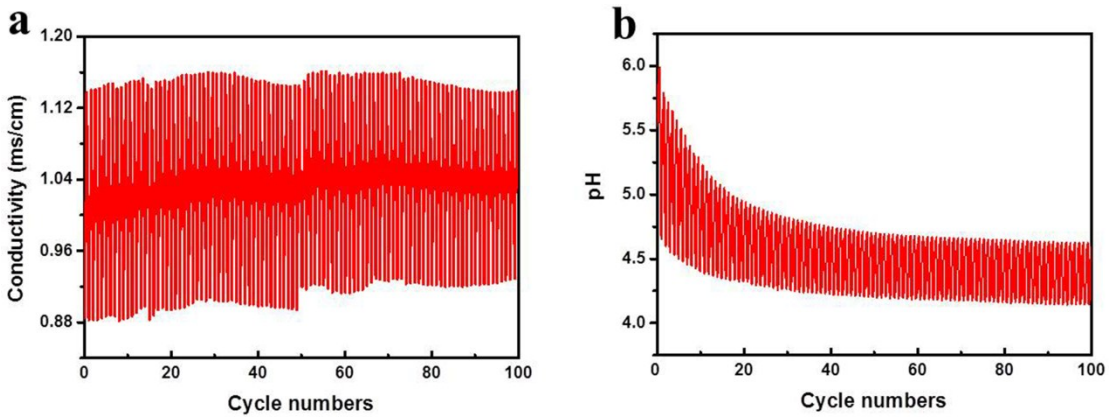

Fig. S8 (a) the effluent conductivity variation and (b) effluent pH changes of NHVO/rGO cell during 100 cycles of charging and discharging process in 500 mg/L NaCl solution at the cell voltage of 0.8 V.

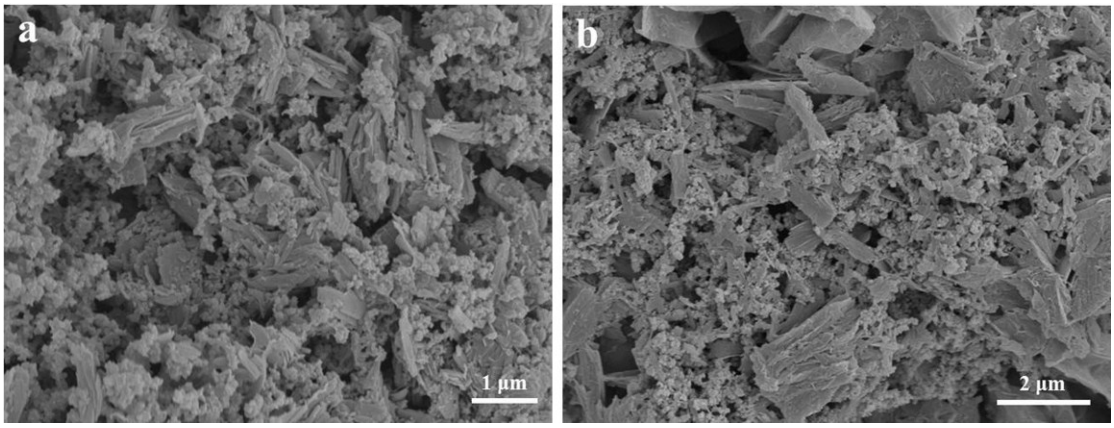

Fig. S9 (a, b) SEM images of NHVO/rGO composite after 100 cycles in 500 mg/L NaCl solution at the cell voltage of 0.8 V.

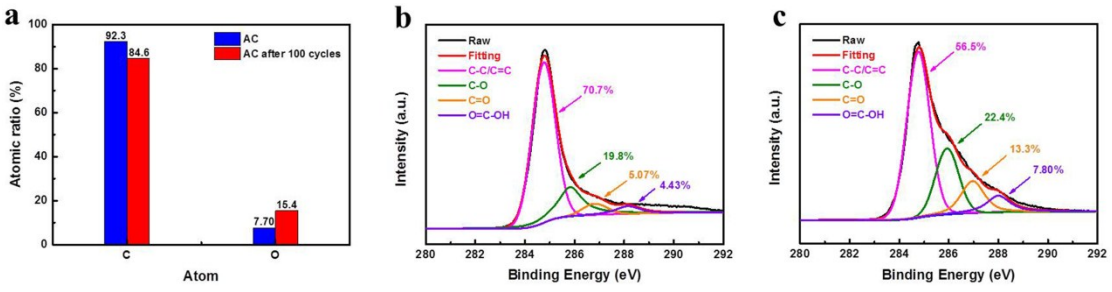

Fig. S10 (a) atomic ratio of C and O on the surface of AC after 100 cycles; (b) C 1S XPS spectrum of AC before cycling; (c) C 1s XPS spectrum of AC after 100 cycles.

Table S1 Comparison of desalting performance among different capacitive deionization systems

Material	Voltage or Current density	NaCl concentration	SAC	Electrode Mass	Reference
$Na_3V_2(PO_4)_3$@C	1.0 V	100 mM	137.2 mg/g	10-20 mg	[1]
hV_2O_5-MWCNT	166 mA/g	600 mM	23.6±2.2 mg/g	15 mg	[2]
$Na_3V_2(PO_4)_3$@C wire	100 mA/g	1000 mg/L	98.0 mg/g	10 mg	[3]
A mixture of $VOHPO_4 \cdot 0.5(H_2O)$ and $Na_{0.5}VOPO_4 \cdot 2(H2O)$	50 mA/g	100 mM	24.3 mg/g	64 mg	[4]
$NH_4V_4O_{10}$/rGO	1.2 V	500 mg/L	20.1 mg/g	75 mg	This work

By contrast, the SAC of $NH_4V_4O_{10}$/rGO CDI system in this work can reach the level that had been already reported in literatures.

Reference

1. J. Cao, Y. Wang, L. Wang, F. Yu and J. Ma, $Na_3V_2(PO_4)_3$@C as Faradaic Electrodes in Capacitive Deionization for High-Performance Desalination, *Nano Lett.*, 2019, **19**, 823-828.
2. J. Lee, P. Srimuk, K. Aristizabal, C. Kim, S. Choudhury, Y. C. Nah, F. Mucklich and V. Presser, Pseudocapacitive Desalination of Brackish Water and Seawater with Vanadium-Pentoxide-Decorated Multiwalled Carbon Nanotubes, *ChemSusChem*, 2017, **10**, 3611-3623.
3. W. Zhao, L. Guo, M. Ding, Y. Huang and H. Y. Yang, Ultrahigh-Desalination-Capacity Dual-Ion Electrochemical Deionization Device Based on $Na_3V_2(PO_4)_3$@C-AgCl Electrodes, *ACS Appl. Mater. Inter.*, 2018, **10**, 40540-40548.
4. J. Lee, P. Srimuk, R. Zwingelstein, R. L. Zornitta, J. Choi, C. Kim and V. Presser, Sodium ion removal by hydrated vanadyl phosphate for electrochemical water desalination, *J. Mater. Chem. A*, 2019, **7**, 4175-4184.