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Supplementary Information

High-performance supercapacitor based on free-standing $V_4C_3T_X$@NiO-reduced graphene oxide core-shell hierarchical heterostructured hydrogel electrodes

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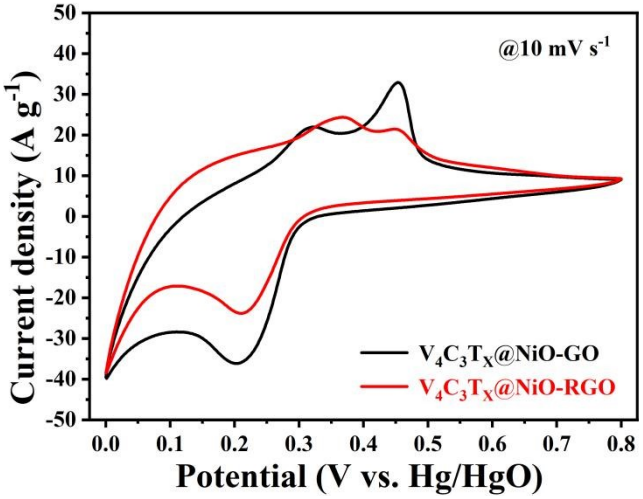

Fig. S1 CV curves of $V_4C_3T_X$@NiO-GO and $V_4C_3T_X$@NiO-RGO hydrogel at the potential scan rate of 10 mV s^{-1}.

"As shown in Fig. S1. there is a significant enhancement of the peak near 0.25 V compared to that of CV curve with RGO, indicating that the GO have been fully reduced into RGO."

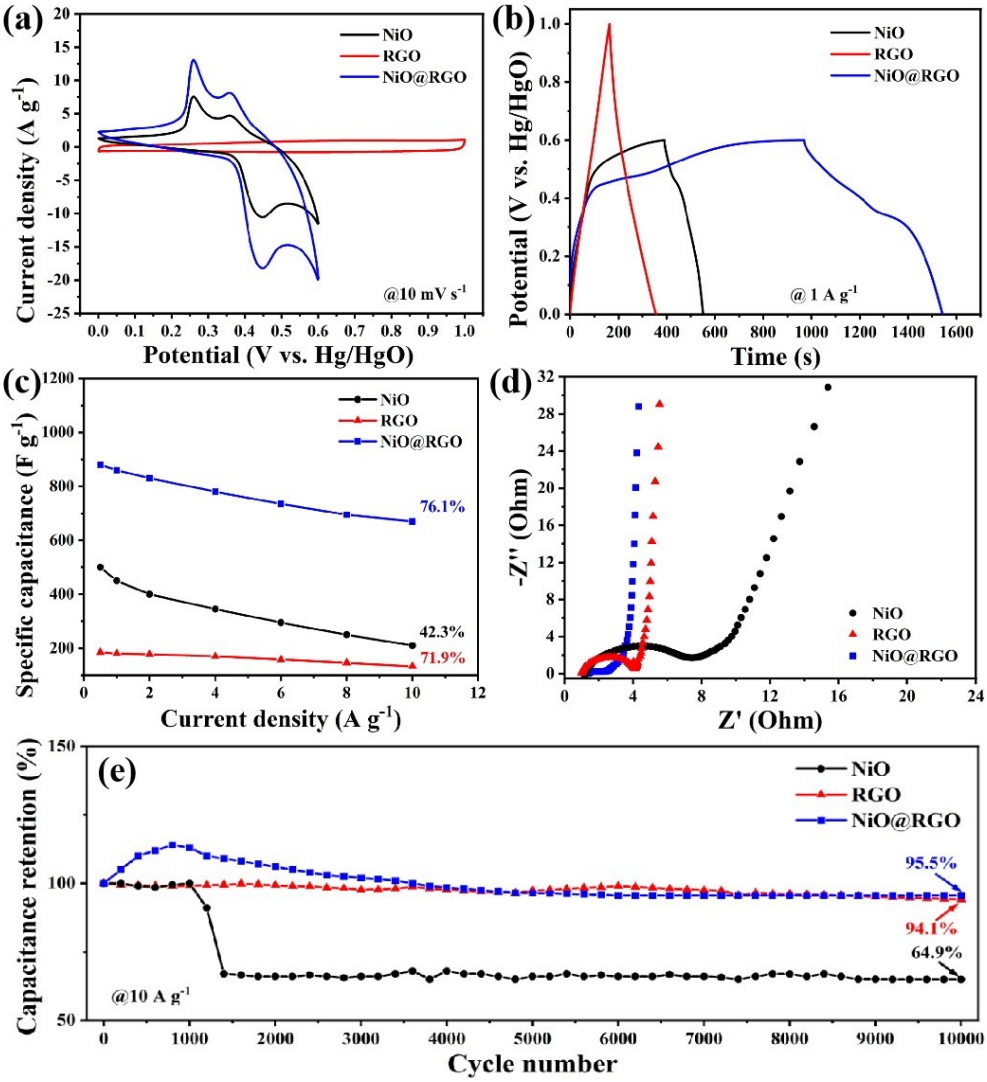

Fig. S2 Electrochemical properties of these samples in three-electrode system: (a) CV curves of NiO, RGO, and NiO@RGO electrodes at 10 mV s^{-1}. (b) GCD curves of NiO, RGO, and NiO@RGO electrodes at 1 A g^{-1}. (c) Specific capacitance vs. current density plot of NiO, RGO, and NiO@RGO electrodes. (d) Nyquist impedance plots of NiO, RGO, and NiO@RGO electrodes in a frequency range from 0.01 Hz to 100 k Hz. (e) Cycling stability of NiO, RGO, and NiO@RGO electrodes measured at 10 A g^{-1} for 10,000 cycles.

"The electrochemical properties of these samples were examined at the potential

sweep rate of 10 mV s^{-1} in a 1 M KOH electrolyte. In cyclic voltametric (CV) assessments (Fig. S2a), the CV curve of RGO is quasi-rectangular in shape, indicative of good capacitive behavior. In comparison with RGO, NiO and NiO@RGO electrodes all displayed a pair of redox peaks in 1 M KOH, arising from the variation of the Ni valence state in alkaline media. More importantly, the NiO@RGO electrode exhibited a far higher current density and greater integrated charge area, and hence a higher specific capacity, than RGO and NiO counterparts. This demonstrates that the assembly of the hierarchical core-shell heterostructure indeed improves the pseudocapacitive performance of NiO@RGO.

To further explore the capacitive characteristics of these samples, GCD studies were conducted at a unified current density of 1 A g^{-1}. From Fig. S2b, the GCD curve of the RGO electrode remained largely symmetrical, and there is no obvious IR drop, indicative of good capacitive behavior, consistent with the CV results. While, the nonlinear GCD curves of NiO and NiO@RGO electrodes featured a clear plateau, consistent with the typical pseudocapacitive behavior and reversible and fast charge/discharge kinetics. Additionally, the GCD profiles show an almost symmetric triangular-shaped shape, further confirming the reversible nature of the faradic reactions. Furthermore, NiO@RGO electrode shows only a moderate decrease of the specific capacitance with the current densities, from 880 F g^{-1} at 0.5 A g^{-1} to 670 F g^{-1} at 10 A g^{-1} (Fig. S2c). The 76.1% retention of the original specific capacitance suggests sufficient surface utilization and outstanding rate capability. As a comparison, the specific capacitance of RGO and NiO decreases from 185 to 133 F g^{-1} (71.9% retained) and 500 to 210 F g^{-1} (42.3% retained), respectively.

To investigate the conductivity behavior of these samples, electrochemical impedance measurements were conducted within the 0.01 to 10^5 Hz frequency range. From the Nyquist plots (Fig. S2d), NiO@RGO electrode exhibits a lower charge transport resistance (Rct, 3.8 Ω) than NiO (9.6 Ω), indicating faster electrochemical kinetics. Furthermore, the device series resistance (Rs) was ca. 4.8 Ω for NiO and 2.3 Ω for NiO@RGO, suggesting that synthetic of the NiO@RGO heterostructure indeed increased the electrical conductivity of the composite. Evidently, the slope of the linear segment for NiO@RGO is greater compared with that of NiO, indicating increasingly capacitive behavior of the former. Long-cycle lifetime is vital to practical applications, which was evaluated via GCD measurements at 10 A g^{-1} for 10,000 cycles, as shown in Fig. S2e. It can be observed that about 95.5% retention of its initial capacitance (94.1% of RGO and 64.9% of NiO) and the GCD profiles did not vary obviously in shape, indicating a small leakage current and good cycling performance of the NiO@RGO."