In-Situ Fabrication Three-Dimensional Ultrathin Graphite/Carbon Nanotubes/NiO Composite as Binder-Free Electrode for High-performance Energy Storage

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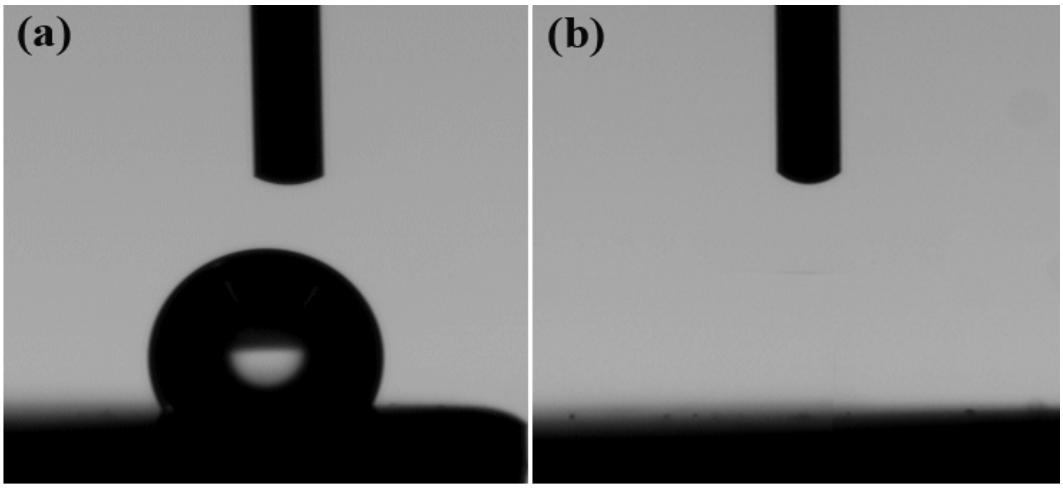

Figure S1 Contact angle of the bare nickel foam/UGF/CNTs (a) before and (b) after treated by O_2 plasma.

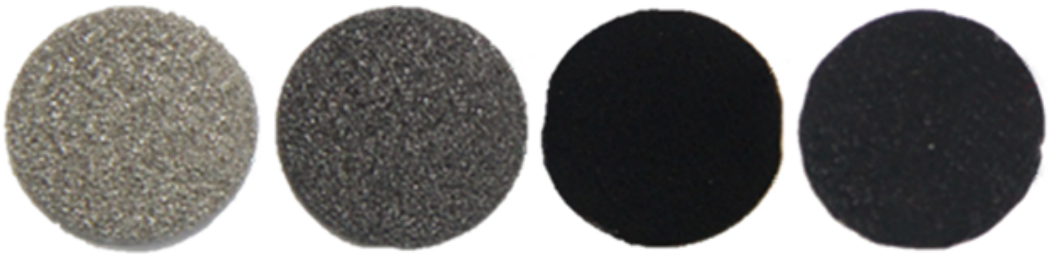

Figure S2 Photo images of nickel foam, UGF grown on nickel foam, nickel foam/UGF /CNTs and nickel foam/UGF /CNTs/NiO, respectively (From left to right).

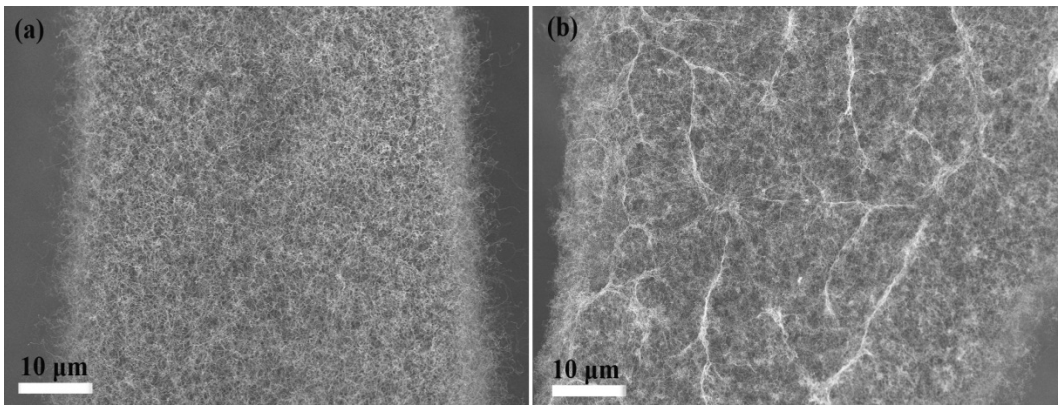

Figure S3 SEM images of bare nickel foam/UGF/CNTs (a) before and (b) after ultra-sounded for 10 min.

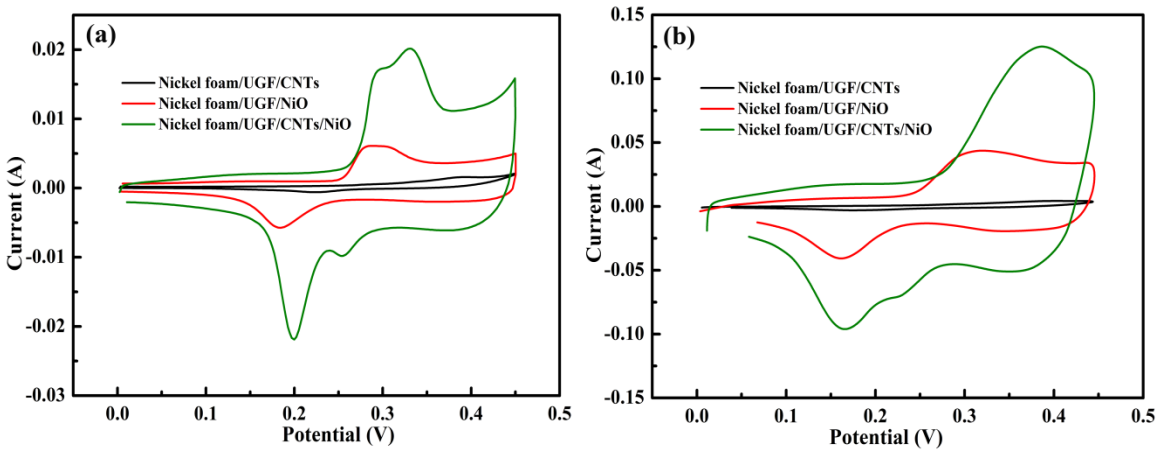

Figure S4 Comparision of the CV curves of bare nickel foam/UGF/CNTs, nickel foam/UGF/NiO, nickel foam/UGF/CNTs/NiO at the scan rate of (a) 10 $mV \cdot s^{-1}$ and (b) 100 $mV \cdot s^{-1}$.

Herein, the weights of ultrathin-graphite film (UGF), CNTs, and NiO in nickel foam/UGF/CNTs/NiO composite are 0.73 mg, 0.64 mg, and 0.59 mg, respectively, which are measured several times by Mettler Toledo XP26 Delta Range balance with

readability of 0.002 mg. It should be mentioned that the specific capacitance, the power and energy density are based on the weight of NiO. For a comparison, nickel foam/UGF/CNTs and NiO nanosheets on nickel foam/UGF (nickel foam/UGF/NiO) are also prepared under the same annealing temperature in air. Capacitance (*C*) of a electrode evalulates its chare storage capacity. It can be derived from the CV curve according to the following equation:

$$C = \frac{\int_{E_1}^{E_2} i(E)dE}{(E_2 - E_1) \cdot v}$$

where E_2, E_1, $i(E)$, v are the upper, lower cut-off potentials, instantaneous current with repect to potential E and potential scan rate, respectively. Obviously, under the same scan rate and potential window, the capacitance is propotional to $\int_{E_1}^{E_2} i(E)dE$, which is integral area of CV curve. As shown in **Figure S4**, the integral area of CV curve of the bare nickel foam/UGF/CNTs is much smaller than that of nickel foam/UGF/CNTs/NiO, indicating that the capacitance contribution from bare nickel foam/UGF/CNTs is neglilible. Furthermore, the integral area of CV curve of the nickel foam/UGF/CNTs/NiO is much larger than that of nickel foam/UGF/NiO, suggesting that the 3D nickel foam/UGF/CNTs/NiO significantly enhances the electrochemical performances.

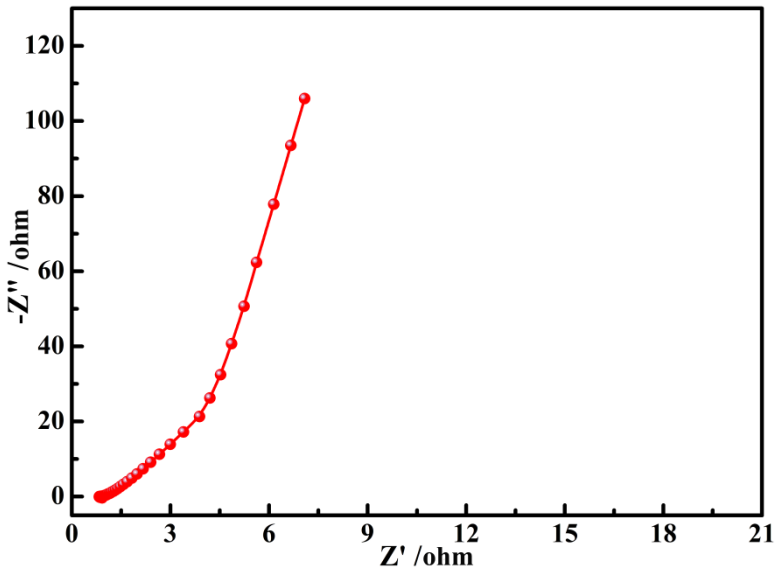

Figure S5 EIS spectrum of the 3D nickel foam/UGF/CNTs/NiO electrode.

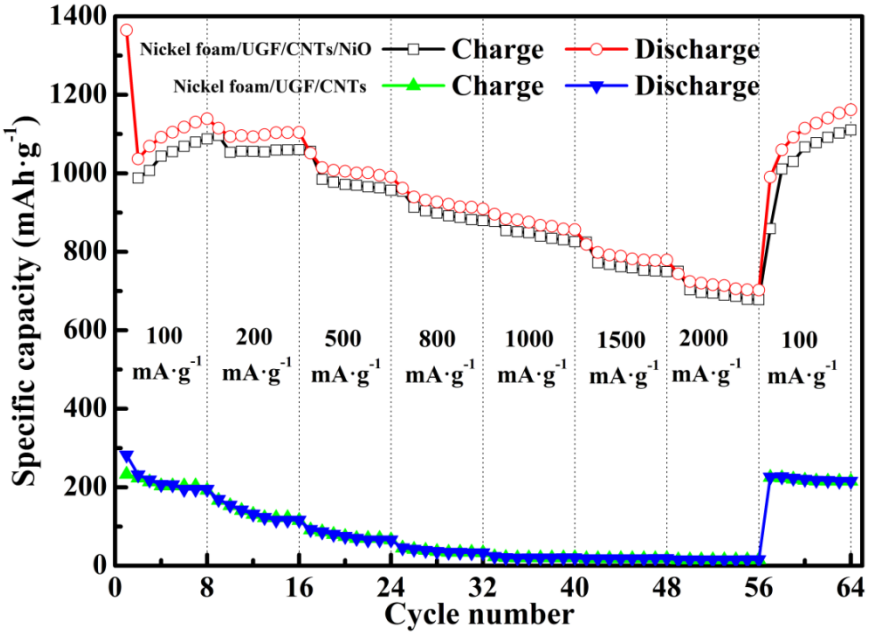

Figure S6 Charge/discharge capacities of bare nickel foam/UGF/CNTs and nickel

foam/UGF/CNTs/NiO composite at different current rates.