

Supporting Information

1. Experimental section

1.1 Chemicals and materials

All chemical reagents purchased from China National Pharmaceutical Group Corporation were analytical grade and used directly without further purification. AISI 316L stainless steel foils (Good fellow, 0.015 wt% C, 16-18 wt% Cr, 10-14 wt% Ni, 0.3 wt% Mo, and Fe balance) were cut into 10×10×0.5 mm³. All the samples were mechanically polished by abrasive papers (grade 1000 and 2000) at first. After that, they were cleaned carefully, including 10 min sonication in ethyl alcohol and 10 min sonication in distilled water.

1.2 Growth of CNT networks on stainless steel

The stainless steel samples were then placed in anhydrous ethylene glycol solution containing 5 vol% HClO₄ at a constant voltage of 20 V for 20 min in order to remove the oxide. The samples were transferred into a quartz tube furnace (diameter= 55 mm) used as the CVD reactor. Reactive and carrier gases were delivered into the reactor through mass flow controllers. The tube was primarily purged with a mixed flow of Ar and H₂ (5% H₂, 95% Ar) at a flow ratio of 120 sccm for 5 min to reduce the surface oxide when the temperature reached 900°C. During the growth step, the flow rates of Ar/H₂ and C₂H₂ were 500 and 20 sccm at 800°C for 60 min, respectively. Then the furnace cooled down to room temperature naturally. The samples were then put in a quartz tube

(diameter=20 mm) through a horizontal tubular furnace at 500 °C for 120 min in the air to obtain graphenated CNT networks and label as CNTs.

1.3 Synthesis of nitrogen-doped graphitized carbon nanotube networks (NGCNTs) on 316L stainless steel

The NGCNTs was prepared in a Teflon-lined stainless steel autoclave using the classical hydrothermal synthesis method [23]. The CNTs on the stainless steel was placed against the wall of a 50 mL Teflon-sealed stainless-steel autoclave containing 30 mL of urea precursor (molar concentration of 0.1 M, 0.3 M and 0.5 M). Afterwards, the autoclave was sealed and hydrothermally treated at 200 °C for 12 h and labeled as NGCNTs-x (x=1, 2 and 3). Finally, the as-synthesized sample was taken out, rinsed with DDW several times and dried at 60 °C for 12 h. The mass loading of NGCNTs-x was identified by a highly sensitive balance with a precision of 0.01 mg.

1.4 Structure characterization

The morphologies of the samples were investigated by SEM (Hitachi S-4800 high-resolution scanning electron microscope) and TEM (Hitachi H-7600 transmission electron microscope). The wettability was detected by OCA15PRO. Raman spectra were carried out by a Renishaw inVia Micro-Raman under a laser excitation wavelength of 532 nm. XPS measurements were conducted with a VG Microtech ESCA 2000 using monochromatic Mg K α radiation at a power of 300 W.

1.5 Electrochemical measurements

The electrochemical behavior were studied by cyclic voltammetry (CV), galvanostatic charge-discharge (GCD) and electrochemical impedance spectroscopy (EIS) using a CHI660e electrochemical workstation (Chen-hua Instrument, Shanghai, China). All electrochemical measurements were performed in 0.5 M Na₂SO₄ electrolyte. A three-electrode electrochemical system was employed, consisting of the samples as the working electrode, a platinum plate (30×30 mm²) as the counter electrode, and a saturated calomel electrode (SCE) as the reference electrode. CV of the electrode materials was performed over the potential range of 0-0.8 V. The capacitance of the electrode was measured *via* a galvanostatic charge-discharge cycle in the potential range of 0-0.8 V at constant currents of 0.5-10 A g⁻¹. The EIS frequency varies from 10 mHz to 100 kHz at open circuit voltage, and the results are analyzed by the software of ZView 2 on the basis of an electrical equivalent circuit.

The specific capacitances (C_g) were calculated according to the following equation (1):

$$C_g = 2 (I \Delta t) / (m \Delta V) \quad (1)$$

where I (A) is the constant charge/discharge current density, Δt (s) is the discharge time, and ΔV (V) is the discharge voltage, m (g) is the weight of electrode material.