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Supplementary Information

Synchronous Exfoliation and Assembly of Graphene on 3D $Ni(OH)_2$ for Supercapacitor

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Experimental section

Materials

$NH_3 \cdot H_2O$, $(NH_4)_2SO_4$ (Shanghai Titan Scientific Co., Ltd., China) and Ni foam (Shen Zhen Ke Jing star technology CO., China) were purchased. All chemicals were used as received without further purification.

Methods

Synthesis of $Ni(OH)_2$/graphene composites

The $Ni(OH)_2$/graphene composites were synthesized using a unique electrochemical technique that is simple, green and low cost. In a typical synthesis, we performed in a two-electrode system using graphite flakes as anode and Ni foam as cathode (both with a working area of approximately 2.0*1.0 cm^2), respectively, and the electrolyte is a mixed aqueous solution containing 5 mL NH_3•H_2O and 100 mL 0.1 M $(NH_4)_2SO_4$. When the direct current voltage of 8 V was applied to two-electrode setup, vigorous bubbles were produced at electrodes, anodic graphite began to dissociate into graphene sheets, which was dispersed into electrolyte. Meanwhile, the cathodic Ni foam firstly transformed into $Ni(OH)_2$ through the NH_3•H_2O treatment. The reaction takes place at 25 °C for 30 min. Then the $Ni(OH)_2$/graphene composites were washed three times with H_2O and dried at 60 °C for 12h.

Synthesis of $Ni(OH)_2$ without graphene

The $Ni(OH)_2$ was synthesized using a unique electrochemical technique. In a typical synthesis, we performed in a two-electrode system using Ni foam as anode and other Ni foam as cathode (both with a working area of approximately 2.0*1.0 cm^2), respectively, and the electrolyte is a mixed aqueous electrolyte containing 5 mL NH_3•H_2O and 100 mL 0.1 M $(NH_4)_2SO_4$. When the direct current voltage of 8 V was applied to two-electrode setup, vigorous bubbles were produced at electrodes, anodic graphite began to dissociate into graphene sheets, which was dispersed into electrolyte. Meanwhile, the cathodic Ni foam firstly transformed into $Ni(OH)_2$ through the NH_3•H_2O treatment in a $(NH_4)_2SO_4$ aqueous solution, mainly due to the weak corrosion of Ni by NH_3 in aqueous solution. The mixture was allowed to react at 25 °C for 30 min. Then the $Ni(OH)_2$ was washed three times with H_2O and dried at 60 °C for 12h.

Characterization of $Ni(OH)_2$/graphene composites and $Ni(OH)_2$

The morphology of the $Ni(OH)_2$/graphene composite and $Ni(OH)_2$ were observed with SEM (JEOL JSM-7401F) with an accelerating voltage of 1.0 kV. To understand the surface information of the as synthesized samples, XPS (AXIS ULTRA DLD, Kratos, Japan) was conducted to analyse the composition of the sample surface. The Raman spectra were obtained using a laser micro-Raman spectrometer (Renishaw inVia) employing an argon-ion laser with an incident wavelength of 532 nm.

Electrochemical Measurements

The electrochemical properties were performed using a PARSTAT 4000 workstation with a standard three-electrode system. Using the prepared samples as the working electrodes with a Pt clamper, a Pt gauze as the counter electrode, and a saturated calomel as a reference electrode. Cyclic voltammetry with various scan rates between 0.0 and 0.5 V was conducted in 1 M KOH. The galvanostatic charge- discharge measurements (GCD) and cycle stability were performed on a LAND CT-2001A.

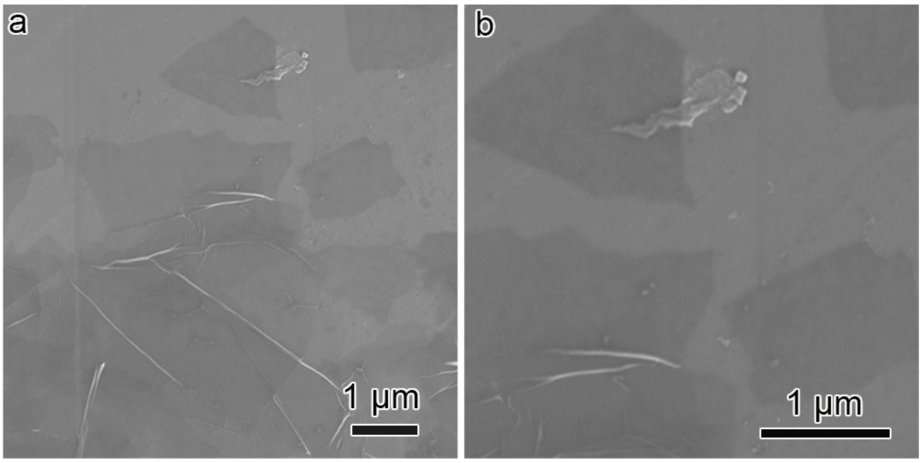

Fig.S1 SEM image of the exfoliated graphene with different magnifications.

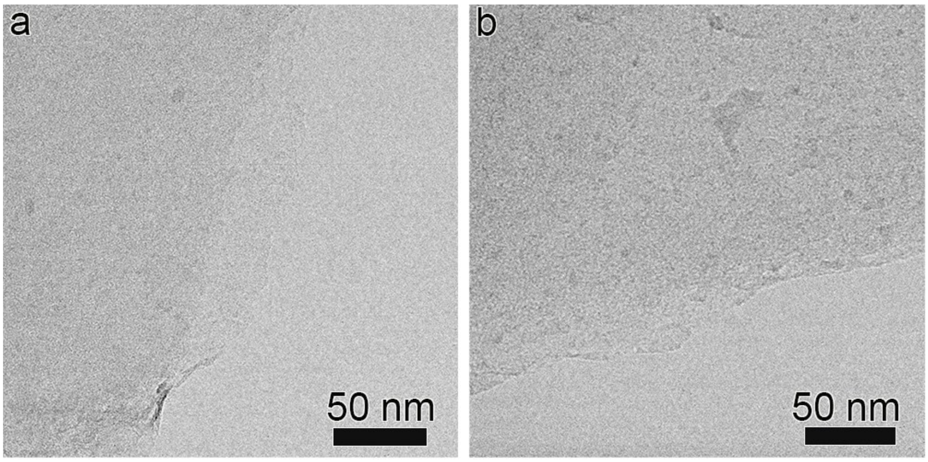

Fig. S2 TEM image of the exfoliated graphene with different magnifications.

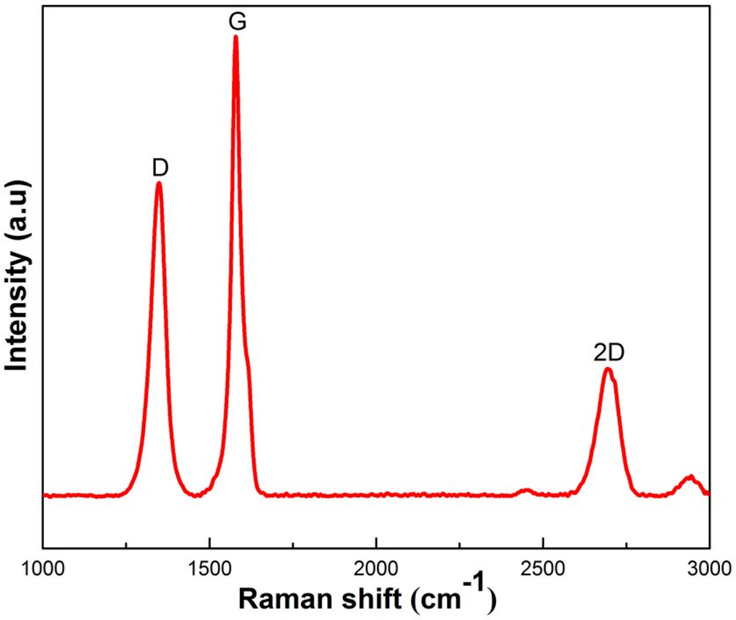

Fig. S3 Raman pattern of the exfoliated graphene.

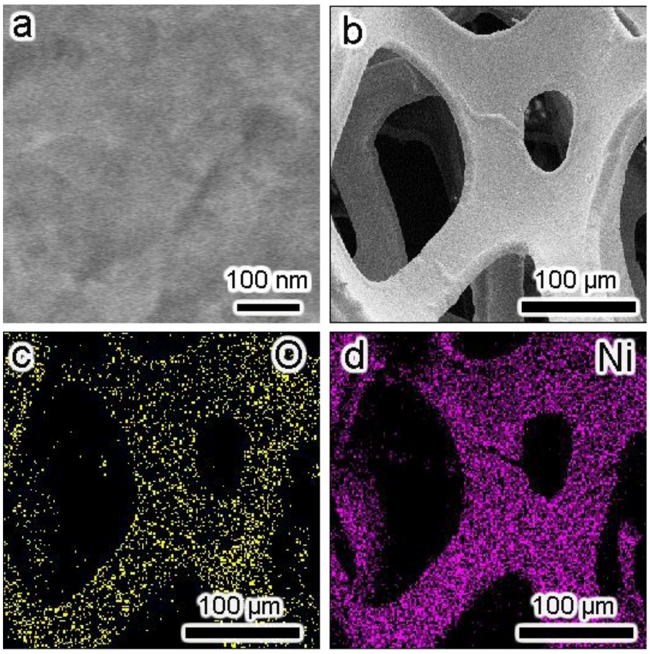

Fig. S4 SEM images of the $Ni(OH)_2$ without graphene. (a) SEM images of the $Ni(OH)_2$; (b) SEM image and the corresponding EDS elemental mapping images of (c) oxygen, and (d) nickel.

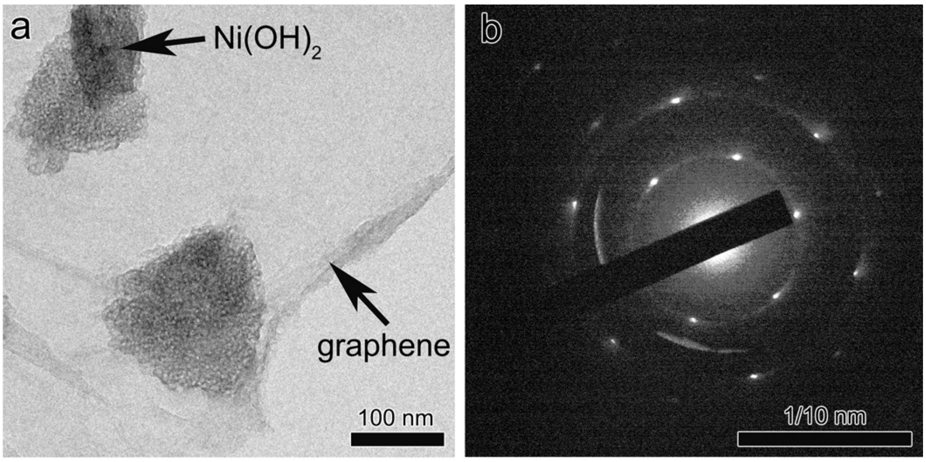

Fig. S5 TEM images of the $Ni(OH)_2$/graphene composites (a) and corresponding selected-area electron-diffraction pattern (b).

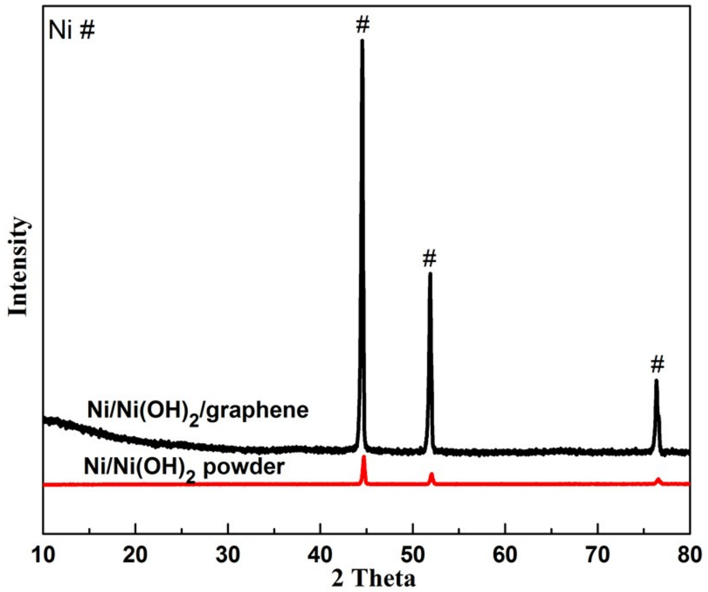

Fig. S6 X-ray diffraction (XRD) measurement of the $Ni(OH)_2$/graphene composites and $Ni(OH)_2$ power (the same synthesis condition without graphene).

Table S1 Element composition of the Ni$(OH)_2$/graphene composites.

Element	Wt.%	Atom %
C	2.05	8.02
O	6.45	18.90
Ni	91.50	73.08
Total	100.00	100.00

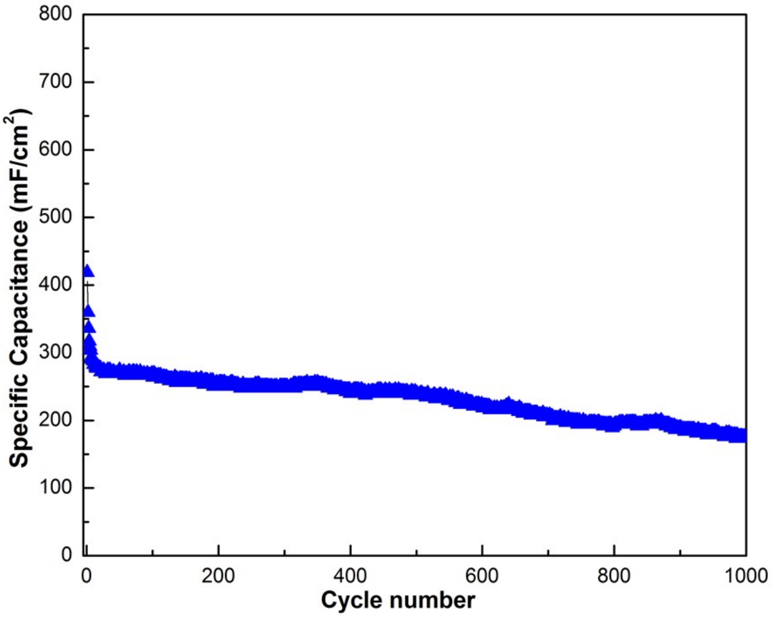

Fig. S7 Cycling performance of $Ni(OH)_2$ without graphene at 2 mA/cm^2.

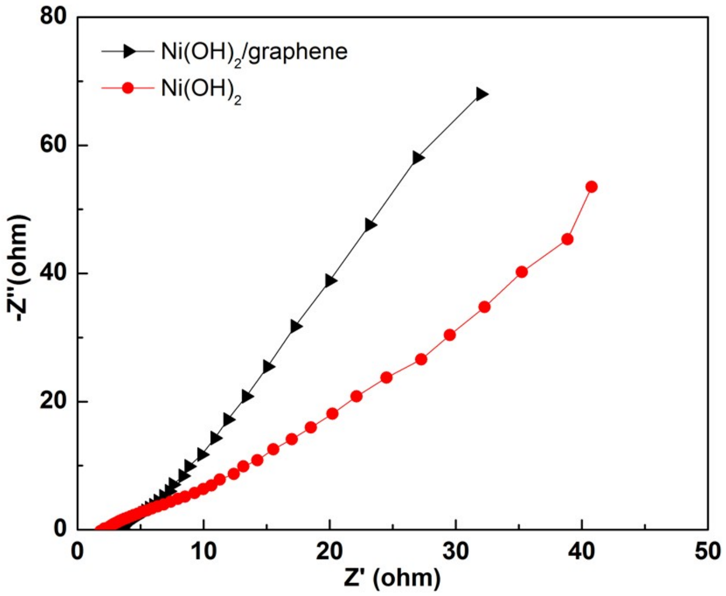

Fig. S8 EIS of Ni(OH) $_2$ and Ni(OH)$_2$/graphene composites electrodes in the frequency range from 100 kHz to 0.01 Hz.