



A novel Ni(OH)₂/graphene nanosheets electrode with high capacitance and excellent cycling stability for pseudocapacitors



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HIGHLIGHTS

- A highly conductive Ni(OH)₂/graphene nanosheets (GNs) composite is prepared.
- The Ni(OH)₂/GNs composite is used as active material for capacitor electrode.
- Good electrochemical activity and stability of the electrode is observed.
- Ion and electron transport kinetic of the composite is discussed.
- The home-made fluffy GNs are found as desirable electrode matrix.

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ABSTRACT

A novel Ni(OH)₂/graphene nanosheets (GNs) composite with Ni(OH)₂ nanoflakes dispersing within a 3D fluffy and conductive graphene network has been successfully prepared by a facile hydrothermal reaction for pseudocapacitor applications. Using the as-prepared Ni(OH)₂/GNs composite as active material, the pseudocapacitor electrode exhibits very high capacitance and encouraging rate capability, as indicated by the experimental results that its specific capacitance is 2260 F g⁻¹ at a current density of 1 A g⁻¹ and 1401 F g⁻¹ when the current density is increased to 10 A g⁻¹ (61.9% retention). In addition, it shows remarkable long-term cycling stability, with a high capacitance retention of 115.6% at 100 mV s⁻¹ after 4000 cycles and only 2.6% of decay at a current density of 16 A g⁻¹ over 1000 cycles. The excellent electrochemical performance of the Ni(OH)₂/GNs composite electrode is ascribed to its enhanced ion and electron transport kinetic arising from its unique microstructure, namely a 3D electron conductive graphene network with ion diffusive fluffy pathways. Therefore, the Ni(OH)₂/GNs composite electrode is a promising candidate for high performance pseudocapacitors; the fluffy crystalline GNs are promising matrix for advanced energy materials with ensured ion and electron conductivities.

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1. Introduction

Supercapacitor is an energy storage device featuring considerable specific capacity, high power density, long cycle life and low maintenance cost [1–4]. It has been widely used in hybrid electrical vehicles, portable electronic appliances, power source systems, and more [5,6]. Although supercapacitor has attracted the interest of a number of researchers from energy storages field, it still needs efforts to improve its overall performance by developing advanced

electrode materials. Faraday pseudocapacitors, which utilize the reversible redox reactions of electrode materials during charge/discharge processes, have much larger specific capacity than double layer supercapacitors [7]. At present, the transition metal oxides or hydroxides (e.g. RuO₂, MnO₂, NiO, and Ni(OH)₂) [8,9], conductive polymers (e.g. polyaniline, polypyrrole, and polythiophene) [10], and some oxygen/sulfur containing materials are adopted as electrode material for pseudocapacitors [5], among which Ni(OH)₂ is one of the most promising candidates due to its low cost, eco-friendly nature, and high theoretical capacity [11,12]. However, problems like poor electrical conductivity and cycling stability restrict its commercialization.

Using carbon materials as structural support for Ni(OH)₂ has been proven to be an effective strategy to improve the electrochemical performance of Ni(OH)₂ based pseudocapacitors [13–17].

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In particular, graphene, which possesses a high specific surface area ($>2500 \text{ m}^2/\text{g}$) and excellent electrical conductivity [4], has been successfully employed to increase the specific capacity and cycling stability of $\text{Ni}(\text{OH})_2$ based pseudocapacitors [18–21]. Other examples include graphite oxide (GO), reduced graphene oxide (RGO) and porous graphene [1,22,23,37]. The resulting electrochemical performance of the composite electrode materials has not reached the theoretical specific capacity of $\text{Ni}(\text{OH})_2$ due to the inherent agglomeration of graphene nanosheets or the high density of structural defect caused by the residual of oxygen-containing functional group, which significantly depress the boost in electrical conductivity and cycle life [13,20,24]. Therefore, it is critical to seek an effective strategy for suppressing the agglomeration and self-assembly of both $\text{Ni}(\text{OH})_2$ and graphene to enhance the specific capacity and cycling stability of hybrid materials. In our previous work [25], we reported a method for preparing fluffy few-layer graphene nanosheets (GNs) with crystalline structure via carbo-thermal reaction between coal-tar pitch and nano- Al_2O_3 . The as-prepared GNs provide a possibility for efficient immobilization of $\text{Ni}(\text{OH})_2$ with controlled structures. Herein, we report the successful construction of a 3-dimensional $\text{Ni}(\text{OH})_2/\text{GNs}$ nanocomposite from a facile hydrothermal reaction, which exhibits remarkable electrochemical activity for pseudocapacitors.

2. Experimental

2.1. Preparation and characterization of $\text{Ni}(\text{OH})_2/\text{GNs}$ composite

All the chemical agents were analytical grade and used without any purification. The GNs were prepared with a specific surface area of $150 \text{ m}^2 \text{ g}^{-1}$ according to our previous work [25]. The $\text{Ni}(\text{OH})_2/\text{GNs}$ composite was prepared by a hydrothermal reaction similar to literature procedure [26]. Specifically, 37 mg of fluffy GNs and 16 mg of urea were added into 30% alcohol aqueous solution and stirred for 10 min and then 4 ml of $0.4 \text{ M Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ was blended with the GNs via ultrasonic treatment. After that 20 ml of 0.2 M NaOH was added drop by drop. The mixture was then transferred into airtight Teflon-lined reactor after a vigorous shake and held at 180°C for 4 h. The product of hydrothermal reaction was took out at room temperature and washed with deionized water. The mass ratio of $\text{Ni}(\text{OH})_2$ (66.7 wt%) in the as-obtained product was calculated from the weight change between the dried composite and initial GNs.

The structure and morphology of sample were characterized by field emission electron microscope (FESEM, Carl Zeiss ULTRA 55 FESEM) and transmission electron microscope (TEM, JEM-2010, Jeol). X-ray diffraction pattern of sample was taken out by Ultima III X-ray model diffractometer (Rigaku, Japan) with $\text{Cu K}\alpha$ radiation at scanning rate of $10^\circ \text{ min}^{-1}$ in a reflection mode. Fourier transform infrared (FT-IR) spectrum of sample was carried out by utilizing a Nicolet-5700 FT-IR spectrometer (Thermo, USA). Raman spectroscopy was performed with a Raman spectrometer (Renishaw Invia Reflex, Britain) with a 532 nm Ar-ion laser. X-ray photoelectron spectroscopy (XPS) was performed on a Thermo ESCALAB 250 high performance electron spectrometer using monochromatic $\text{Al K}\alpha$ as the excitation source.

2.2. Preparation of electrode and electrochemical measurements

All of the electrochemical measurements were carried out via a three electrode system using CHI 660E electrochemical workstation. Nickel foam was used as current collector and 6 M KOH aqueous solution as electrolyte. The electrode was consisted of $\text{Ni}(\text{OH})_2/\text{GNs}$ composite (90 wt%) and polyvinylidene fluoride (PVDF) (10 wt%). The mass of the active material on the electrode

was 2 mg cm^{-2} . Platinum foil ($2 \text{ cm} \times 2 \text{ cm}$) and Ag/AgCl electrode were used as counter and reference electrode, respectively. The average specific capacitance was calculated by following equation:

$C = \frac{\int IdV}{m \cdot \Delta V}$, where I is the voltammetric current, v is the potential scan rate, m is the mass of active material, and V is the potential in one sweep segment. The specific capacitance was derived from galvanostatic discharge curve according to the equation: $C = \frac{It}{m \cdot \Delta V}$, where I is the constant current, t is the time of discharge, m is the mass of active material, V is the change of potential after a full discharge. Electrochemical impedance spectroscopy tests were performed by applying an alternating voltage with 5 mV amplitude in a frequency range from 10^{-2} Hz – 10^5 Hz at the open circuit potential. Cycle life was evaluated by cyclic voltammetry (CV) and galvanostatic charge/discharge process.

3. Result and discussion

Fig. 1(a) shows the XRD patterns of the crystalline GNs, $\text{Ni}(\text{OH})_2$ and as-prepared product, respectively. The XRD pattern of the as-prepared product has the combined features of the crystalline GNs and $\text{Ni}(\text{OH})_2$, indicating that the $\text{Ni}(\text{OH})_2/\text{GNs}$ composite had been well prepared. The characteristic peaks at 19.7° , 33.1° , 38.7° , 51.7° , 59.1° and 62.8° correspond to (001), (100), (101), (102), (110), and (111) diffraction plane respectively. It is in keeping with the

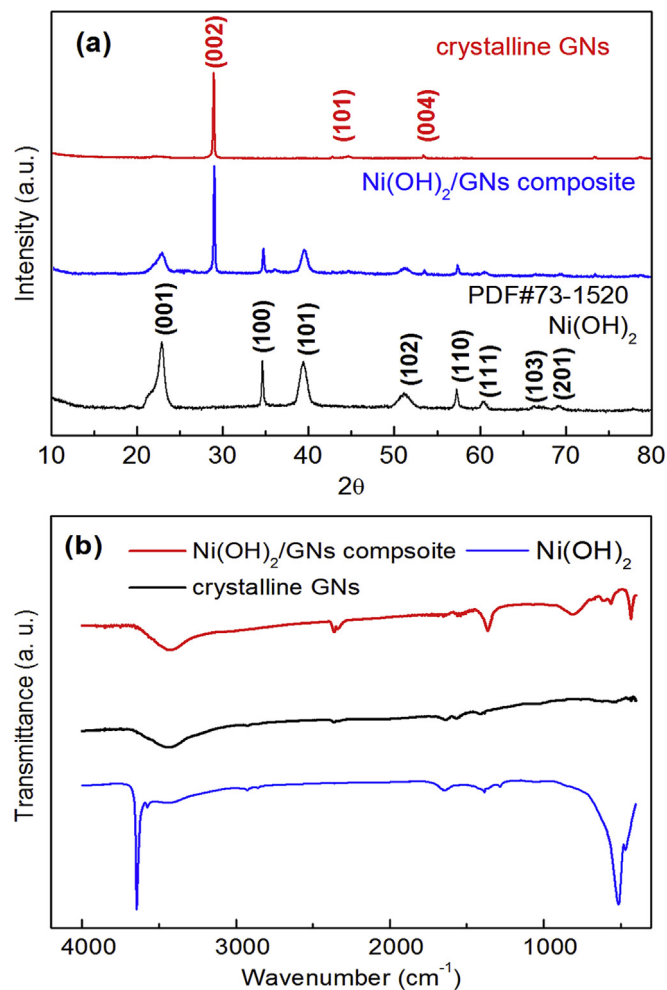


Fig. 1. (a) XRD patterns of the crystalline GNs, $\text{Ni}(\text{OH})_2$ and as-prepared $\text{Ni}(\text{OH})_2/\text{GNs}$ composite. (b) FT-IR spectra of the crystalline GNs, $\text{Ni}(\text{OH})_2$ and as-prepared $\text{Ni}(\text{OH})_2/\text{GNs}$ composite.

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