**Three-dimensional ultrathin Ni(OH)₂ nanosheets grown on nickel foam for
high-performance supercapacitors**

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Abstract

In this paper, we report the growth of ultrathin Ni(OH)₂ nanosheets on nickel foam at room temperature via a cost-effective and simple process, oxidizing fresh nickel foam in a wet environment followed by a morphology transformation in a mixed alkaline and oxidative solution without the need for any additional nickel sources, templates, or surfactant. When tested as electrode for a supercapacitor, the Ni(OH)₂ nanosheets grown on nickel foam displayed excellent performance, demonstrating specific capacitance of 2384.3 F g⁻¹ at a charge and discharge current density of 1 A g⁻¹ and 1288.1 F g⁻¹ at 5 A g⁻¹ with a good cycling ability (~75% of the initial specific capacitance remained after 3000 cycles). The excellent electrochemical performance is attributed to its unique nanostructures, which may facilitate rapid ion transport near electrode surfaces, while allowing facile redox reactions associated with charge storage by the nanosheets. The demonstrated high specific capacity and the remarkable rate performance of the Ni(OH)₂ nanosheets, together with the flexibility of the nickel foam substrate, make the three-dimensional nanostructured electrodes ideally suited for low-cost, high-performance supercapacitor applications.

Keywords: nickel hydroxide nanosheets, nickel foam, high-performance, supercapacitor

Introduction

Supercapacitors have attracted increasing attentions as novel energy storage devices because of their higher power density and longer cycling life than conventional

rechargeable batteries [1,2]. One type of supercapacitor is termed a “pseudocapacitor”, which utilizes near-surface redox reactions of transition metal oxides and hydroxides materials such as RuO_2 [3], MnO_2 [4,5], NiO [6], Co_3O_4 [7-9], and Ni(OH)_2 [10-12] providing very high energy storage capacity (or energy density) while maintaining capacitor-like performance (high power density and long cycling life). Among various pseudoactive materials, nickel hydroxide (Ni(OH)_2) is an especially attractive candidate for pseudocapacitors due to its layered structure of large interlayer spacing, offering high theoretical specific capacitance. Nickel based compounds like Ni(OH)_2 are also readily available, environmentally friendly, and highly stable in alkaline electrolytes [10,13]. Despite these attractive features, the actual specific capacitance values reported for a wide variety of unique nanostructures is still far below the theoretical value [14,15], likely because the pseudocapacitive effect is limited only to a region near the surface (or poor utilization of active materials). To ensure high capacitances and facile ionic transportation, it is vital to create highly porous Ni(OH)_2 -based materials of high specific surface area on a suitable current collector. Accordingly, Ni(OH)_2 nanostructures [16], and Ni(OH)_2 -carbon composites involving conductive materials of high surface area (such as carbon nanotubes and graphene) have been extensively studied [12,15,17]. Such nanostructured materials are typically mixed with ancillary materials such as carbon black or binder and then coated on current collector, which greatly decreases the overall specific capacitance. Since electroactive nanostructures grown on three-dimensional (3D) conductive substrates provide more efficient electrical contact, they can be directly used as binder-free

electrodes, potentially offering simpler electrode structure with higher energy density [18,19]. Nickel foam has been used extensively as supporting substrate for active materials in supercapacitors and batteries because it has large surface area, high porosity, good electric conductivity, and excellent chemical stability in a wide variety of liquid electrolytes. Several methods to grow electroactive nanostructures on nickel foam have been demonstrated including hydrothermal treatment [20], chemical bath deposition [21,22] and electrodeposition [19]. However, all the above methods require elevated temperature or additional nickel sources. Another disadvantage is that the grown nanostructures may not be in good contact with the nickel foam and thus, effective electron transfer between them cannot be ensured.

Herein, we reported a cost-effective and simple process for growing ultrathin Ni(OH)_2 nanosheets directly from nickel foam at room temperature without adding any additional nickel sources, templates, or surfactant. Due to the high specific surface area and good contact with substrate, the Ni(OH)_2 nanosheets have a specific capacitance of 1288.1 F g^{-1} when tested as electrodes for a supercapacitor (without adding any binder and conductive agent) and maintained 75% of the initial capacity after 3000 cycles at a current density of 5 A g^{-1} . These results show that Ni(OH)_2 nanosheets grown on nickel foams could be a promising electrode for high-performance electrochemical capacitors.

Experimental

Materials Synthesis

The ultrathin Ni(OH)_2 nanosheets were grown on nickel foams as follows. All of the chemicals were of analytical grade and used without further purification. In a

typical procedure, nickel foams with a thickness of 1.6 mm (Novamet, USA) were cut into size of $3 \times 3 \text{ cm}^2$, carefully cleaned using a 3 M HCl solution in an ultrasonic bath for 30 minutes in order to remove the surface oxide layer, and then cleaned in absolute ethanol and deionized water for 15 min in sequence by ultrasonication. The wet foam was sealed in a glass vials and stored for 24 hours. After that, the nickel foam was taken out and immersed in mixed solution of NaOH (20 mL, 3M) and $(\text{NH}_4)_2\text{S}_2\text{O}_8$ (10 mL, 1M) in a 60 mL beaker for 6 hours at room temperature. The thin film on the metal substrate was rinsed several times with distilled water and ethanol and dried at 150°C for 6 hours.

In order to determine the mass of $\text{Ni}(\text{OH})_2$, the synthesized $\text{Ni}(\text{OH})_2$ films (1 cm^2) were reduced in a tube furnace at 600°C for 5 h with a heating rate of $5^\circ\text{C} \cdot \text{min}^{-1}$ and a constant H_2 flow rate of 50 sccm. According to the reaction of $\text{Ni}(\text{OH})_2 = \text{NiO} + \text{H}_2\text{O}$ and $\text{NiO} + \text{H}_2 = \text{Ni} + \text{H}_2\text{O}$, the active mass of $\text{Ni}(\text{OH})_2$ was obtained from the change of weight before and after reduction (Tab.S1, see Supporting Information). The mass was weighed by a microbalance (Citizen, CX265) with a minimum measurement increment of 0.01 mg.

Characterization

The phases of the samples were examined by X-ray diffraction (XRD) analysis (PAN'alytical X'Pert Alpha 1, using Cu K- α 1 x-rays). The microstructure and morphology were examined using a scanning electron microscope (LEO 1530 field emission SEM) and a high resolution transmission electron microscope (JEOL 4000 EX).

Electrochemical Measurement

Cyclic voltammetry (CV) and galvanostatic charge-discharge tests were carried out using a classical three-electrode configuration. The thin Ni(OH)_2 film on the nickel foam (1 cm^2) was used as the working electrode. A platinum foil (1.0 cm^2) and a saturated calomel electrode were used as the counter and the reference electrode, respectively. The cyclic voltammograms were acquired in a potential range between -0.1 and 0.65 V at different scan rates, and the charge/discharge processes were performed by cycling the potential from 0 to 0.5 V at different current densities in a 2 M KOH aqueous electrolyte. The cyclic stability was evaluated by galvanostatic charge-discharge measurements at a current density of 5 A g^{-1} for over 3000 cycles. All the electrochemical measurements were conducted using a Solartron SI 1286 electrochemical workstation.

Results and discussion

The synthetic strategy for the ultrathin Ni(OH)_2 nanosheets from the nickel foam is illustrated in Fig.1. First, a dense Ni(OH)_2 layer is grown from the nickel foam after being washed with 3 M HCl and water and stored in sealed glass vials, which provides nickel sources for later Ni(OH)_2 nanosheets formation. The amount of Ni(OH)_2 grown from nickel foam can be controlled by changing the storage time (Fig.S1,S2 and Tab.S1). Subsequently the obtained nickel foam is immersed into a mixed alkaline and oxidative solution. Herein the dense and random Ni(OH)_2 film adhered on the Ni surface is transferred into ultrathin nanosheets. The growth mechanism responsible for the formation of the Ni(OH)_2 nanosheets is similar to the facile, yet effective chemical

bath deposition (CBD) method, through which $\text{Ni}(\text{OH})_2$ nanosheets are grown on 3D conductive substrates via adding soluble nickel salts into alkaline and oxidative mixing solution in previous reports [21,22].

Fig.1 Growth mechanism and schematic illustration of the formation of $\text{Ni}(\text{OH})_2$ nanosheets by a facile process.

SEM images in Fig.2(a) show that after storage in sealed vials, the nickel foam with a smooth surface and clear texture converts into a random and homogenous layer on top, below which was a dense layer of nanoparticles (Fig.2(b)). After immersion in the mixed solution of NaOH and $(\text{NH}_4)_2\text{S}_2\text{O}_8$, the film exhibits a highly porous cross-linked structure with pore diameter ranging from 200 to 600 nm (Fig.2(c) and (d)). The interconnecting network was made up of nanosheets with thickness of about 10 nm and no coalescence between neighboring nanosheets or obstruction of the pores was observed, indicating the strong mechanical strength of the $\text{Ni}(\text{OH})_2$ nanosheets. With such a unique hierarchical structure, the space between the $\text{Ni}(\text{OH})_2$ nanosheets can be efficiently utilized, allowing the electrolyte ions easier access to the surface of the

active material. TEM was used to analyze the detailed crystal structure of Ni(OH)_2 nanosheets as shown in Fig.2. As is seen in Fig. 2(e), the nanosheets exhibited foldaway silk-like morphology with a high degree of transparency, indicating their ultrathin nature. The corresponding selected area electron diffraction (SAED) pattern (Fig. 2(f)) showed the disperse rings were well indexed to the crystal planes of (100), (101), (102), (110), (111), and (200), confirming well crystalline feature of $\beta\text{-Ni(OH)}_2$.

Fig.2 Low- and high-magnification SEM images of (a and b) dense Ni(OH)_2 on nickel foam (insert is SEM image of fresh nickel foam), (c and d) Ni(OH)_2 nanosheets grown from nickel foam, TEM images (e) and SAED pattern (f) of Ni(OH)_2 nanosheets.

The phase and structures of dense Ni(OH)_2 film and porous Ni(OH)_2 nanosheets on Ni foam were verified by powder XRD and Raman spectrum. As shown in Fig.3(a), the three most intense peaks marked by “#” belonged to the Ni substrate and all other

diffraction peaks can be indexed as hexagonal β -Ni(OH)₂ (JCPDS card no. 14-0117). The difference of the relative intensity indicated the different degree of crystallinity. The abnormal broadening of the (101) diffraction line for dense Ni(OH)₂ film demonstrated the presence of a large amount of stacking faults [23,24], which was consistent with the SEM shown in Fig.2. The structural features of the Ni(OH)₂ were further confirmed using Raman measurements. As illustrated in Fig.3, dense Ni(OH)₂ film and porous Ni(OH)₂ nanosheets almost demonstrate the same Raman spectrum, displaying three peaks associated with well-crystallized β -Ni(OH)₂ at 3570, 445 and 310 cm⁻¹ [25,26]. The 3570 cm⁻¹ peak was attributed to the symmetric stretching of the hydroxyl groups (ν OH), the band at 445 cm⁻¹ was assigned to the Ni-O stretching (ν NiO), and the signal at 310 cm⁻¹ has been attributed to an E-type vibration of the Ni-OH lattice. Except for the three peaks of well-crystallized β -Ni(OH)₂, the Raman spectrum showed two additional peaks, in agreement with the spectrum obtained by for disordered β -Ni(OH)₂ at respectively 520 and 3640 cm⁻¹. The peak at 3640 cm⁻¹ was not attributed to particular vibration modes yet, but it is probably associated with OH stretching of free hydroxyl groups and the 520 cm⁻¹ peak can be associated with the existence of defects. A unknown peak at 380 cm⁻¹ has been also detected for the dense Ni(OH)₂ film, but its intensity was very weak. In conclusion, both the XRD and Raman spectrum demonstrate that only β -Ni(OH)₂ remains on the surface of nickel foam after storage in humid environment or immersed in oxidative solution.

Fig.3 (a) XRD patterns of dense Ni(OH)₂ film and Ni(OH)₂ nanosheet on nickel foam, (b and c)

Raman spectrum of dense Ni(OH)₂ film and Ni(OH)₂ nanosheet on nickel foam (Black line: dense Ni(OH)₂ film, Red line: Ni(OH)₂ nanosheet)

As illustrated in Fig.2, many more exposed interfaces of Ni(OH)₂ were generated after immersion in a mixed solution of NaOH and (NH₄)₂S₂O₈ that will greatly increase the ion diffusion and thus lead to a better electrochemical performance. Stimulated by the 3D self-growth ultrathin Ni(OH)₂ nanosheets architectures, the Ni(OH)₂ nanosheets were investigated as binder- and conductive- agent-free electrodes for pseudocapacitors. The CVs of fresh nickel foam and nickel foam with dense Ni(OH)₂ at a scan rate 5 mV s⁻¹ were shown for comparison (Fig.4(a)). A pair of well-defined redox peaks were clearly visible for dense Ni(OH)₂ film and Ni(OH)₂ nanosheets, which corresponds to

the reversible reactions of $\text{Ni}^{2+} \leftrightarrow \text{Ni}^{3+}$. Meanwhile, the fresh nickel foam shows a negligible current response in the CV test. The well-accepted surface Faradaic reaction of nickel hydroxide [12] can be expressed as:

The larger separation between the anodic peak potential and cathodic peak potential for the Ni(OH)_2 nanosheets compared to the dense Ni(OH)_2 film (See Fig.S5) may be ascribed to the longer electronic pathway (larger Ohmic resistance). More importantly, the higher peak current values for the Ni(OH)_2 nanosheets indicated its significantly higher specific capacitance. This can be attributed to the remarkably increased accessible surface area between electrolyte and active materials through transforming the dense Ni(OH)_2 to ultrathin nanosheets. It confirms that this facile approach allows us to dramatically enhance the electrochemical performance of Ni(OH)_2 . This phenomenon was further confirmed by chronopotentiometry measurements. As shown in Fig.4(b), the discharge curves of the samples at the current density of 2 mA cm^{-2} show a significant deviation from a straight and flat line, indicating that the capacitance mainly comes from the Faradaic redox reactions, which is consistent with the CV curves. Compared to the dense Ni(OH)_2 film, the discharge time of Ni(OH)_2 nanosheets was subsequently increased from 108 s to 1080 s at a current density of 2 mA cm^{-2} (within a voltage window of 0-0.45 V), resulting in an ultrahigh areal capacitance of 4.8 F cm^{-2} . Fig.4(c) shows the CV response of Ni(OH)_2 nanosheets at different scan rates ranging from 5 to 40 mV s^{-1} . With the increase of scan rate, the current response increased accordingly, indicating the good rate property of the

Ni(OH)₂ nanosheets composite electrode. Fig.4(d) shows the galvanostatic discharge curves of Ni(OH)₂ nanosheets at different current density (voltage window: 0-0.5 V). The specific galvanostatic capacitances (F·g⁻¹) were calculated from the formula below [27,28] using the discharge curves presented in Fig.4(b) and (d):

$$C = \frac{2i_m \int V dt}{V^2 \big|_{V_i}^{V_f}} \quad (1)$$

where $i_m = I/m$ (A·g⁻¹) is the current density, I is the current, m is the active mass of the electrode, V is the potential with an initial and final value of V_i and V_f , respectively. The calculated specific capacitance as a function of the discharge current density was plotted in Fig.4(e). The specific capacitance was as high as 2384.3 F g⁻¹ when measured at the discharge current density of 1 A g⁻¹ (the current density and specific capacitance were normalized based on the mass of the Ni(OH)₂ nanosheets and voltage of 0.5 V). The specific capacitance gradually decreased with the increment of current density due to an incremental voltage drop and insufficient active material involved in redox reaction. It is worthwhile to note that when the current density reaches 10 mA cm⁻², the specific capacitance remained approximately constant, implying good ion diffusion and electron transportability at high current density. This rate performance highlights advantages of 3D self-growth ultrathin Ni(OH)₂ nanosheets, which provides fast and efficient diffusion of the electrolyte ions to the active material surface and excellent electron transport within Ni(OH)₂.

The stability of the Ni(OH)₂ nanosheets was evaluated using a long-term galvanostatic charge/discharge process (Fig.4(f)). The capacitance increased during the first 200 cycles, which may be attributed to the activation of Ni(OH)₂ that allows

trapped ions (ions can be trapped between the Ni(OH)_2 crystalline layers during nanosheets) to diffuse out [29-31]. The capacitance stabilized in the last 500 cycles and a capacitance retention of about 75% was achieved after 3000 cycles at a current density of 5 A g^{-1} . The Ni(OH)_2 nanosheets exhibited decent long-term electrochemical stability, as further evident from the stable charge-discharge curves for the last 10 cycles (the inset in Fig.4(f)). The morphology of the electrode after the cycling test was recorded to analyze the reason of degradation, see SEM images (Fig.S6). The deformation of the nanosheets may be ascribed to the degradation, however, no peeling off from the nickel foam was observed indicating a good contact between the porous Ni(OH)_2 nanosheets with nickel foam. The good electrochemical performance, in terms of cycle performance and rate capability, of 3D self-growth Ni(OH)_2 nanosheets should originate from the following merits: First, the direct growth from nickel foam enable good mechanical adhesion and fast charge transfer pathways. It eliminates the need for polymer binders or conducting additives and substantially reduces the “dead volume” in electrode materials. Second, the nanosheets largely increase the amount of contact areas between electrolyte and active materials, which will greatly facilitate ion and electron diffusion. Finally, the interspaces between the nanosheets provides sufficient space to buffer the volume change because the accessibility of electrolyte and OH^- ions to a bulk Ni(OH)_2 phase may result in the volume expansion [12].

Fig.4 Cyclic voltammograms at a scan rate of 5 mV s^{-1} (a) and constant current discharge curves at 2 mA cm^{-2} (b) of a nickel foam electrode in a 3-electrode cell with 2 M KOH aqueous solution: (1) Fresh nickel foam, (2) dense Ni(OH)_2 on nickel foam, (3) Ni(OH)_2 nanosheets on nickel foam. (c) CV curves of the Ni(OH)_2 nanosheets electrode at different scan rates from 5 to 40 mV s^{-1} within a voltage window of -0.1 - 0.65 V . (d) Constant current discharge curves of Ni(OH)_2 nanosheets electrode at different current densities. (e) Specific capacitances of the Ni(OH)_2 nanosheets at different current densities. (f) Charge/discharge cycling test at a current density of 5 A g^{-1} for the Ni(OH)_2 and hybrid electrodes; the inset shows the galvanostatic charge/discharge cyclic curves of the last 10 cycles for Ni(OH)_2 nanosheets.

Conclusion

In summary, Ni(OH)_2 nanosheets on nickel foam have been synthesized by a facile

and cost-effective process as binder-free and conductive-agent-free supercapacitor electrode. The as-fabricated $\text{Ni}(\text{OH})_2$ nanosheets electrode exhibits excellent supercapacitor performance with a specific capacitance of 2384.3 F g^{-1} at 1 A g^{-1} and excellent rate capability and good cycle life. The ultrathin thickness and sufficient spaces between the nanosheets in direct contact with the substrate, as well as high surface area account for the excellent performance of the obtained pseudocapacitors. Our work shows that cost-effective synthesis of $\text{Ni}(\text{OH})_2$ nanosheets and excellent electrochemical performance provide great potential as an active electrode for electrochemical pseudocapacitors.

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Figure captions

Fig.1 Growth mechanism and schematic illustration of the formation of $\text{Ni}(\text{OH})_2$ nanosheets by a facile process.

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