



Growth of nickel (111) plane: The key role in nickel for further improving the electrochemical property of hexagonal nickel hydroxide-nickel & reduced graphene oxide composite



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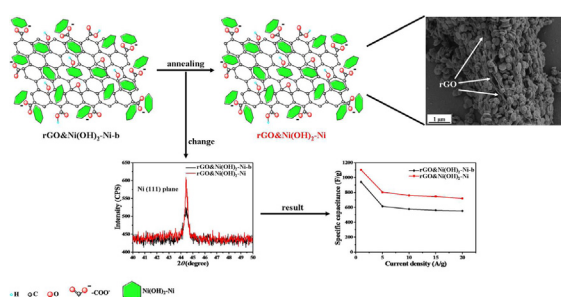
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HIGHLIGHTS

- A reduced graphene oxide and Ni(OH)₂/Ni (rGO&Ni(OH)₂-Ni) composite is got easily.
- The existence of Ni can enhance the thermal stability of Ni(OH)₂.
- The coexistence of rGO and Ni can greatly improve the electrical conductivity.
- The (111) plane plays a key role in Ni for improving the electrochemical activity.
- The rGO&Ni(OH)₂-Ni composite exhibits superior electrochemical property.

GRAPHICAL ABSTRACT



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ABSTRACT

Hexagonal nickel hydroxide/nickel decorated on reduced graphene oxide (rGO&Ni(OH)₂-Ni) has been prepared via easy one-step chemical precipitation method and subsequent annealing treatment. The results show that the thermal stability of Ni(OH)₂ is enhanced by the incorporation of Ni. Besides, hexagonal Ni(OH)₂-Ni nanoplate which can supply a short diffusion and migration pathway for electron and electrolyte ion contacts closely with the surface of rGO, resulting that the agglomeration of rGO is effectively prevented. Due to the good electrical conductivity of Ni, the incorporation of Ni can also improve the electrical conductivity of Ni(OH)₂. More importantly, the Ni (111) plane is grown after annealing treatment, which plays a key role in Ni for further improving the electrochemical activity of composite. Consequently, the rGO&Ni(OH)₂-Ni electrode exhibits high capacitance, high energy density, excellent rate capability, good cycle stability, etc. The advantages of easy preparation and excellent electrochemical performance imply the great potential application of rGO&Ni(OH)₂-Ni in supercapacitors. Moreover, it is worthy noting that this work will offer a new approach for using metal nanoparticle in improving the electrochemical property of supercapacitor electrode material.

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1. Introduction

Supercapacitors, one of the new and promising energy storage and conversion devices, have attracted tremendous attention owing to higher energy density than that of conventional dielectric capacitors, larger power density and longer cycle life than those of batteries, low self-discharging, low maintenance cost and environmental friendliness [1–4]. Nowadays, supercapacitors have found extensive applications, including industrial power, mobile electrical system, portable consumer electronics, power and memory back-up system, etc [5,6]. Generally speaking, supercapacitors store energy based on two types of mechanisms: fast surface redox reaction (pseudocapacitors) and ion adsorption (electrochemical double layer capacitors, EDLCs) [7]. Unfortunately, the specific capacitance of EDLCs is usually low, which can not meet the ever-increasing need for some fields where high energy density is a key requirement. Therefore, using pseudocapacitive materials for supercapacitors has caused much interest since the energy density associated with Faradaic reaction is substantially higher by at least one order of magnitude than that of EDLCs [1,8–14].

Pseudocapacitive materials mainly contain metal oxides/hydroxides and conductive polymers, which are capable of rich redox reaction because of multiple oxidation states/structures. Among them, nickel hydroxide ($\text{Ni}(\text{OH})_2$) is a promising candidate for supercapacitors due to its high theoretical specific capacitance (ca. 3750 F g^{-1}), well-defined electrochemical redox activity, low cost and easy synthesis [15]. It is noteworthy that the specific capacitance of $\text{Ni}(\text{OH})_2$, which may depend on its structure and morphology, is usually lower than its theoretical value. Thus, many established methods are used to synthesize nano-sized $\text{Ni}(\text{OH})_2$ with various architectures (e.g., flower, flake and plate) to obtain improved electrochemical performance [16–18]. Nevertheless, the major issue of $\text{Ni}(\text{OH})_2$ used as electrode for supercapacitors is its poor electrical conductivity, which can not make $\text{Ni}(\text{OH})_2$ meet fast electron transport under high-rate charge and discharge, resulting in the compromises of rate capability and reversibility. For solving this predicament, considerable research efforts have been carried out, where $\text{Ni}(\text{OH})_2$ is usually mixed with highly conductive carbon materials, such as carbon nanotube [19], graphene [20,21] and mesoporous carbon [22].

Graphene, a single-atom-thick two-dimensional graphitic carbon material, has attracted multidisciplinary attention thanks to its high electrical conductivity, large surface area and good chemical stability [23,24]. In recent years, extensive studies have developed the graphene-based materials for supercapacitors [25,26]. The results of these studies show improved electrochemical performance, because the advantages of each component can be combined and the special properties may be offered by the reinforcement or modification of each other. However, due to the strong Van der Waals interaction, graphene sheets usually suffer from serious agglomeration or re-stacking during fabrication process, which will lead to great loss of its excellent property. Consequently, the capacitive behavior of graphene-based material is much lower than the expected value in practical application. On the other hand, some researchers have reported that nickel ion in the $\text{Ni}(\text{OH})_2$ lattice is partially substituted by other metal ions (e.g., Co [27], Zn [28] and Al [29]) or $\text{Ni}(\text{OH})_2$ is mixed with non-carbon materials like metal oxide (e.g., Co_3O_4 [30]) and metal nanoparticle (e.g., Ag [31]). These research results also show the enhanced electrochemical property, which may be due to the synergistic effect among individual components that can improve electron and ion conduction, chemical stability and mechanical stability as well as pseudocapacitance.

In this paper, we propose an advanced electrochemical pseudocapacitive material based on reduced graphene oxide and nickel hydroxide/nickel ($\text{rGO}\&\text{Ni}(\text{OH})_2\text{-Ni}$) via easy one-step chemical precipitation method and subsequent annealing treatment. First, the carboxyl of graphene oxide (GO) is mostly ionized through absorbing thermal energy when the GO solution is heated, which causes added negative charge of the GO surface, resulting that a great deal of Ni^{2+} can be adsorbed and enriched on the surface of GO. As a result, the reduction of GO and the generations of $\text{Ni}(\text{OH})_2$ and Ni are achieved simultaneously with the assistance of hydrazine hydrate ($\text{N}_2\text{H}_4\cdot\text{H}_2\text{O}$). The results show that the existence of Ni can enhance the thermal stability of $\text{Ni}(\text{OH})_2$. Besides, $\text{Ni}(\text{OH})_2$ is mixed with Ni to form hexagonal nanoplate that can provide a short diffusion and migration pathway for electron and electrolyte ion. Such hexagonal $\text{Ni}(\text{OH})_2\text{-Ni}$ nanoplate closely contacts on the surface of rGO, resulting that the agglomeration of rGO is effectively prevented. In addition, the coexistence of rGO and Ni can much better improve the electrical conductivity of $\text{Ni}(\text{OH})_2$ than the single existence of rGO, thanks to the good electrical conductivity of Ni. More importantly, the (111) plane of Ni is grown after annealing treatment, which plays a key role in Ni for further improving the electrochemical activity of composite. Consequently, the $\text{rGO}\&\text{Ni}(\text{OH})_2\text{-Ni}$ composite exhibits excellent electrochemical properties, including high capacitance, high energy density, excellent rate capability, good cycle stability, etc. The advantages combining easy preparation and remarkable electrochemical performance suggest the great potential application of $\text{rGO}\&\text{Ni}(\text{OH})_2\text{-Ni}$ in supercapacitors. Moreover, the synthesis developed in this work will offer a new approach for using metal nanoparticle in improving the electrochemical property of supercapacitor electrode material.

2. Experimental section

2.1. Materials

Graphite powder (spectral pure) was purchased from Sino-pharm Chemical Reagent Co., Ltd. Nickel nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2\cdot 6\text{H}_2\text{O}$) was purchased from Jinhuada Chemical Reagent Co., Ltd (Guangzhou, China). Hydrazine hydrate (80%) was supplied by Fuyu Chemical Reagent Co., Ltd (Tianjin, China). All other reagents and solvents were obtained from commercial suppliers. All aqueous solutions were prepared with ultrapure water ($>18 \text{ M}\Omega$) from a Milli-Q Plus system (Millipore).

2.2. Synthesis of sample

GO was prepared by oxidizing natural graphite powder based on a modified Hummers method as originally presented by Kovtyukhova and colleagues [32,33]. The as-prepared GO was dispersed in deionized water (1 mg mL^{-1}) under ultrasonication and the homogeneous solution was placed in a water bath at 90°C for 14 h. The resulting product was washed with deionized water until $\text{pH} = 7$ and was dispersed again in deionized water. Afterwards, 5 g of nickel (II) nitrate hexahydrate was added into the above solution and the mixture was ultrasonicated for 0.5 h. Then, 20 mL 80% hydrazine hydrate was added in and the mixture was reacted at 95°C for 2 h. The obtained product was washed with deionized water until $\text{pH} = 7$ and was freeze-dried. Such acquired powder was designated as $\text{rGO}\&\text{Ni}(\text{OH})_2\text{-Ni-b}$. Finally, the powder was further annealed in nitrogen atmosphere at 300°C for 2 h in a tube furnace and the product was designated as $\text{rGO}\&\text{Ni}(\text{OH})_2\text{-Ni}$. Similarly, rGO was prepared according to the same method without the addition of nickel (II) nitrate hexahydrate and the annealing treatment.

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