Facile Fabrication of Nickel Aluminum Layered Double Hydroxide/Carbon Nanotube Electrodes Toward High-Performance Supercapacitors

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ABSTRACT: The electrode, as one of the key components in supercapacitors, has a pivotal effect on the overall performances. In this work, a series of composite electrode materials are proposed via the combination of nickel aluminum layered double hydroxides (NiAl-LDHs) and carbon nanotubes (CNTs). To begin with, materials with different ratios of the two compositions are fabricated with a coprecipitation method. After that, various characterization methods indicate that the NiAl-LDH/CNT composites exhibit an irregular thin platelet structure with a well-constructed conductive network inside. Furthermore, the effect of the CNT ratio on the electrochemical property is subsequently investigated, which proves that the conductive network of CNTs is beneficial for the transport of the electrons and strengthens the platelet structure. The results show that when the amount of CNTs reaches 1.5 wt %, it can yield a high specific capacitance of 2447 F g⁻¹ at 2 A g⁻¹, with a good cycling stability of 90.1% after 2500 cycles, indicating high application potential in positive electrodes of pseudocapacitors. The synergistic effects of NiAl-LDHs and CNTs are thought to be the main reasons for the good properties of NiAl-LDHs/CNTs composites.

1. INTRODUCTION

On account of global warming and fossil energy depletion, it is highly essential to develop renewable and clean energy resources and technologies, including new energy storage systems with better performance.¹⁻³ Supercapacitors (SCs) with long cycle life and high power density are deemed as one of the most promising substitutes to fulfill energy requirements in the future.⁴⁻⁶ Owing to charge-storage mechanisms, SCs include electrochemical double-layer capacitors (EDLCs) and pseudocapacitors.⁷⁻⁹ EDLCs entail ion adsorption and desorption for electrical energy storage, which can be reversed. Pseudocapacitors, whose materials include metal oxides/hydroxides and their binary systems, store electrical energy by reversible redox reactions upon the surfaces of the electrodes. It can be concluded that electrode materials have a pivotal effect on the property of SCs since both EDLCs and pseudocapacitors enhance the surface phenomena on their electrode exterior. To obtain an electrode with excellent electrochemical performance, electrode materials with larger porosity are taken into consideration, which may promote electron transmission by means of shortening the transportation distance.¹⁰⁻¹² By virtue of the remarkably enhanced surface area of electrodes using nanostructured materials, many related studies on electrode materials with various microstructures have been reported.¹³⁻¹⁶

Recently, layered double hydroxides (LDHs) have aroused great interest, resulting from their inherent properties, including large surface area, high redox activity, tunable metal composition, intrinsic high stability, and intercalating capability.¹⁷⁻²⁵ In previous studies, supercapacitor electrodes including Ni-based LDHs were investigated in detail.²⁶ For example, the nickel foam electrodes with NiAl-LDH nanostructures revealed a specific capacitance (SC) of 1017 F g⁻¹ at 200 mA g⁻¹.²⁷ Wang et al. synthesized NiAl-LDH nanosheets with hollow microspheres, which showed a high SC of 1578 F g⁻¹, and 93.75% remained after 10 000 cycles at a current density (CD) of 20 A g⁻¹.²⁸ Luo et al. fabricated a kind of NiCo-LDH with a significant SC of 1765 F g⁻¹ at 1 A g⁻¹, but when the CD reached 10 A g⁻¹, the SC decreased rapidly to 940 F g⁻¹.²⁹ Zheng et al. reported a pathway to prepare NiCoAl-LDH/rGO composites using a hydrothermal method. The composites exhibited a good SC of 1544 F g⁻¹ at 1 A g⁻¹, whereas the cycling stability (CS) was relatively poor (only 87% was retained after 2000 cycles).³⁰ Although the Ni-based LDH always displays a high SC, the CS requires to be improved.³¹ To solve this problem, considerable effort has been made toward the correct choice of additives.³²⁻³⁷
Among different kinds of additives, incorporation of LDHs with carbon-based materials has aroused great interest. Carbon-based materials have become the focus of investigations in the domain of supercapacitors because of their distinctive stability. Nevertheless, it is difficult to maximize the intrinsic capacitance property for most carbon-based materials. Especially, carbon nanotubes (CNTs) are often used in composite conductive materials. Wu et al. constructed a CNT-isolated conductive network in a polymer matrix, which exhibited excellent electrical conductivity. As a conductive network, CNTs with internal and external interfaces can facilitate the collection/transport of the electrons as well as buffer the change of volume during long-term cycling, leading to higher SC and improved CS. Moreover, the inner resistance of a composite electrode declines owing to the interfacial contact. For instance, a NiAl-LDH/CNT nanocomposite shows a high SC of 1849 F g\(^{-1}\) at 1 A g\(^{-1}\) and after 1000 cycles, only 12% of the initial capacitance deteriorated. Herein, we propose a strategy to design a NiAl-LDH/CNT composite, via the combination of NiAl-LDHs and CNTs. When the amount of CNTs is 1.5 wt %, a maximum SC of 2447 F g\(^{-1}\) at 2 A g\(^{-1}\) can be obtained with a good CS of 90.1% after 2500 cycles. It is thought that the well-connected conductive network in NiAl-LDHs that improves the transport of the electrons could be the main reason for the good properties of NiAl-LDH/CNT composites. This method is not only feasible for large-scale production but also environmentally friendly since it saves energy without producing a large amount of heat during preparation. Based on the results, more studies can be carried out by adding new ingredients into the system to achieve better CS and higher SC.

2. RESULTS AND DISCUSSION

2.1. Characterization of Morphology and Structure. The NiAl-LDH/CNT products are named LDH-X, where X represents the mass of CNTs of 10, 20, 30, and 40 mg, respectively. The crystal structures of as-synthesized samples were determined by the XRD test. The XRD patterns of CNTs, LDHs, LDH-10, LDH-20, LDH-30, and LDH-40 are displayed in Figure 1a. The characteristic diffraction peak of CNTs at 26.0° belongs to the (002) plane. The diffraction peaks of the NiAl-LDHs at 11.2, 22.8, 34.5, 60.0, and 61.3° are attributed to the (003), (006), (009), (015), and (110) planes of NiAl-LDHs, respectively (JCPDS No. 15-0087). However, the diffraction peak of CNTs in the XRD patterns is not so clear in the composites, which is mainly attributed to the small addition compared with NiAl-LDHs, leading to the low intensity of the peak. It can be concluded that the samples have all of the characteristic peaks of LDHs, indicating the successful preparation of target samples.

Figure 1. (a) XRD patterns for all samples and XPS spectra of (b) NiAl-LDHs/CNTs, (c) Ni 2p, (d) Al 2p, (e) C 1s, and (f) O 1s.
from the incomplete grinding process. Figure 2b shows the structure of CNTs, which exhibits tubes twisting together, and the ratio of length to diameter is extremely high. In contrast, as illustrated in Figure 2c, CNTs form an interconnected network with the irregular thin platelets in LDHs, facilitating the electrons’ transport and promoting the electrochemical performance. In addition, the presence of CNTs also makes the structure more stable to promote cycling stability. Moreover, the energy-dispersive X-ray spectroscopy (EDS) image of NiAl-LDHs/CNTs displayed in Figure 2d clearly indicates that the molar ratio of Al to Ni is about 1:5, corresponding to the amount added.

More information about the compositions of NiAl-LDHs/CNTs is obtained from TEM and HRTEM. Figure 3a shows the structure of pure CNTs, consistent with the images of SEM. It can be clearly deduced from Figure 3b that a platelet structure forms. As depicted in Figure 3c, the platelet structure can be clearly found as predicted with the interplanar spacing value of 0.26 nm approximately, matching with the (009) plane of the NiAl-LDH phase well. Figure 3d shows that the nanotube structure is buried between thin platelets. It can be found from Figure 3e that another lattice fringe of 0.35 nm appears, indicating the existence of CNTs. Besides, the selected area electron diffraction (SAED) shown in Figure 3f further confirms the above conclusion. Based on the above analysis, the SC values of five samples at different discharged CD values, which range from 2 to 20 A g⁻¹, are displayed in Figure 4c. It can be noticed that the capacitance of LDH-30 is 2111 F g⁻¹ at a high CD of 20 A g⁻¹, which equates to retention of 86% compared with the SC at 2 A g⁻¹. The capacitance retention (CR) of 90.1% is illustrated in Figure 4d after 2500 cycles, demonstrating that the electrochemical stability of the LDH-30 electrode is good. Therefore, LDH-30 exhibits excellent rate capability (RC), from which it can be deduced that when it is applied to actual capacitors, the capacitors will show very good power densities. Simultaneously, the RC and SC of NiAl-LDHs are lower than those of the other four samples containing CNTs, further confirming that the addition of CNTs can indeed play a positive role in improving the electrochemical property.

To further confirm the electrochemical properties of the NiAl-LDHs/CNTs, a symmetric supercapacitor was prepared using LDH-30 both as the negative and positive electrodes. Figure 5a displays the CV curves of the capacitor. The symmetric supercapacitor exhibits a typical leaflike shape, which is attributed to the pseudocapacitance characteristics ranging from 0.1 to 1.4 V. It can also be concluded that when the scan rate increases, the CD increases as well. GCD tests are further conducted to verify the SC of the LDH-30 capacitor. The curves illustrated in Figure 5b appear to have a triangle-like shape, which is a combination of the faradic pseudocapacitance and the EDLCs. All of these results imply that the NiAl-LDH/CNT symmetric supercapacitor has good electrochemical properties, optimistically guaranteeing its future usage as a supercapacitor for daily use.

Table 1 presents the SC and CR of the LDH-based nanomaterials. We can see that the as-synthesized NiAl-LDHs/CNTs in this work reveal the advantages of high SC and good SR. It is worth noting that the N-CNTs/CoNi-LDHs possess the most outstanding SC of 2170 F g⁻¹ when the CD is 1 A g⁻¹. However, its CR is only 49.7% after 5000 cycles, lower than most of the others. Evidently, NiAl-LDHs/CNTs fabricated in this work can reach an SC of 2447 F g⁻¹ at a higher CD of 2 A g⁻¹, whose CR is 90.1%, reaching a good
balance between SC and CR, superior to the previous results of CNT-based LDH composites.

The supercapacitor mechanism of NiAl-LDHs/CNTs is further discussed. Ni-based materials have the characteristics of high SC. The radii of Al³⁺ and Ni²⁺ are not very different. When Al³⁺ replaces Ni²⁺ to form a structure of LDHs, anions and water molecules are introduced between the layers, which enhances the stability of the layer structure. At the same time, the valence state of aluminum ions is stable, and it is not easy to change the structure during the long-term charge and discharge process, therefore improving the CS of the material.²⁸ As shown in Figure 6, the electrons tend to move on the network of CNTs owing to the high conductivity, which suggests that the transport of the electrons is promoted, resulting in a higher SC. Moreover, the conductive network enhances the strength of the structure during long-term

Table 1. LDH-Based Nanomaterials and Their Electrochemical Performances

| sample                     | SC (F g⁻¹) | CR (cycling number) | references |
|----------------------------|------------|---------------------|------------|
| Co₃O₄@glucose-LDH          | 1644 (1)   | 94.2% (5000)        | 11         |
| CoAlNi-LDH                 | 1399 (1)   | 82.2% (3000)        | 14         |
| NiCo-LDH@NiMoO₄            | 2100 (1)   | 91% (5000)          | 15         |
| CoAl-LDH@rGO               | 1492 (1)   | 94.3% (5000)        | 20         |
| CNTs@NiCo-LDH              |            | 90.2% (5200)        | 38         |
| Ni-Al LDH/CNTs             | 1849 (1)   | 88% (1000)          | 43         |
| N-CNTs/NiCo-LDHs           | 2170 (1)   | 49.7% (5000)        | 45         |
| NiAl-LDHs/CNTs             | 2447 (2)   | 90.1% (2500)        | this work  |

Figure 4. (a) CV curves of various electrodes measured at 5 mV s⁻¹. (b) GCD curves of different electrodes measured at a CD of 2 A g⁻¹. (c) Rate capability of various electrodes measured at a CD of 2–20 A g⁻¹. (d) CR curve of LDH-30 after 2500 cycles.

Figure 5. (a) CV curves of a NiAl-LDH/CNT symmetric capacitor at scan rates of 2–20 mV s⁻¹. (b) GCD curves of a NiAl-LDH/CNT symmetric capacitor at a CD of 2–20 A g⁻¹.

Figure 6. Schematic illustration of the proposed supercapacitor mechanism of NiAl-LDHs/CNTs.
cycling, leading to a better CS.42 Further, the addition of CNTs decreases the distance of electron transmission and reduces the resistance of the charge transfer, leading to a remarkably enhanced rate capability for the LDHs. In short, the good CS and high SC of NiAl-LDHs/CNTs are mainly owing to the synergistic effects of NiAl-LDHs and CNTs.

3. CONCLUSIONS
A unique material composed of NiAl-LDH irregular thin platelets and CNTs with excellent electrochemical performance has been developed with a coprecipitation method. The CNTs can promote the transport of the electrons as well as reduce the volume change of the active species during long-term cycling as a conductive network. Benefiting from CNTs, the NiAl-LDH/CNT electrode can achieve a high SC of 2447 F g⁻¹ at 2 A g⁻¹, with high RC and good CR (90.1% capacitance after 2500 cycles), owing to the high conductivity of the materials and their synergistic effects leading to structural stability. The electrode developed in this work with superior electrochemical properties is a prospective candidate for supercapacitors.

4. EXPERIMENTAL SECTION

4.1. Synthesis of NiAl-LDH/CNT Composites. Figure 7 illustrates the synthesis process of NiAl-LDH/CNTs. First, a certain amount of carboxyl-CNTs was dissolved in 100 mL of deionized water and sonicated until the CNTs were completely dispersed. Then, 1590 mg of Ni(NO₃)₂·6H₂O and 410 mg of Al(NO₃)₃·9H₂O were added into 100 mL of deionized water and the mixture was kept stirring for 30 min to obtain a uniformly mixed salt solution. Na₂CO₃ (530 mg) and 700 mg of NaOH were added into 62.5 mL of deionized water and sonicated until the CNTs were completely dispersed. Then, 1590 mg of Ni(NO₃)₂·6H₂O and 410 mg of Al(NO₃)₃·9H₂O were added into 100 mL of deionized water and the mixture was kept stirring for 30 min to obtain a uniformly mixed salt solution. Na₂CO₃ (530 mg) and 700 mg of NaOH were added into 62.5 mL of deionized water and sonicated until the CNTs were completely dispersed. Then, 1590 mg of Ni(NO₃)₂·6H₂O and 410 mg of Al(NO₃)₃·9H₂O were added into 100 mL of deionized water and the mixture was kept stirring for 30 min to obtain a uniformly mixed salt solution. Na₂CO₃ (530 mg) and 700 mg of NaOH were added into 62.5 mL of deionized water and sonicated until the CNTs were completely dispersed.

4.2. Preparation of the Working Electrode. The electrode paste consists of NiAl-LDH/CNT composites, acetylene black, and PVDF (3%) at a mass ratio of 8:1:1. First, the NiAl-LDH/CNT composites and acetylene black were mixed well and thoroughly ground. Then, the binder of PVDF (3%) was added. After stirring, the electrode paste was uniformly coated onto a carbon fiber cloth with an area of 1 cm². Finally, the coated carbon fiber cloth was dried under vacuum at 60 °C for 12 h. The working electrode was successfully prepared for further use.

4.3. Characterization. The structural investigation of the samples was observed via field emission scanning electron microscopy (QUANTA200) and field emission transmission electron microscopy (JEM-1400plus). Further, the crystalline information of the samples was collected by X-ray diffraction (XRD, X’Pert3 Powder) employing a Cu Kα (1.5406 nm) source. X-ray photoelectron spectroscopy (XPS) was performed on ESCALAB 250Xi.

4.4. Electrochemical Measurements. The electrochemical performance of all samples was evaluated with an electrochemical workstation in a 6 M KOH electrolyte. Employing the samples as the working electrode, Hg/HgO as the reference electrode, and the platinum electrode as the counter electrode, cyclic voltammetry (CV) and galvanostatic charge–discharge (GCD) measurements were recorded in a three-electrode system. CV curves were obtained with a potential scope from 0.1 to 0.4 V at scan rates of 5 mV s⁻¹. GCD curves were tested under the voltage range of 0–0.37 V at various current densities.

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Notes
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