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Facile one-step synthesis of nanocomposite based on carbon nanotubes and Nickel-Aluminum layered double hydroxides with high cycling stability for supercapacitors

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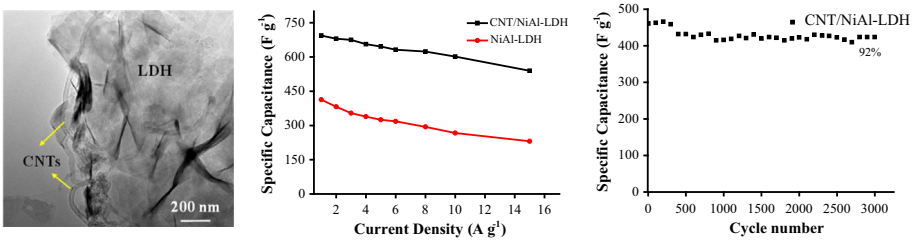
GRAPHICAL ABSTRACT

The CNTs/NiAl-LDH nanocomposite was prepared by using a facile one-step homogeneous precipitation approach, which displayed enhanced electrochemical performance and excellent cycling durability.



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ABSTRACT

Nickel-Aluminum Layered Double Hydroxide (NiAl-LDH) and nanocomposite of Carbon Nanotubes (CNTs) and NiAl-LDH (CNTs/NiAl-LDH) were prepared by using a facile one-step homogeneous precipitation approach. The morphology, structure and electrochemical properties of the as-prepared CNTs/NiAl-LDH nanocomposite were then systematically studied. According to the galvanostatic charge-discharge curves, the CNTs/NiAl-LDH nanocomposite exhibited a high specific capacitance of 694 F g^{-1} at the 1 A g^{-1}. Furthermore, the specific capacitance of the CNTs/NiAl-LDH nanocomposite still retained 87% when the current density was increased from 1 to 10 A g^{-1}. These results indicated that the CNTs/NiAl-LDH nanocomposite displayed a higher specific capacitance and rate capability than pure NiAl-LDH. And the participation of CNTs in the NiAl-LDH composite improved the electrochemical properties. Additionally, the capacitance of the CNTs/NiAl-LDH nanocomposite kept at least 92% after 3000 cycles at 20 A g^{-1}, suggesting that the nanocomposite exhibited excellent cycling durability. This strategy provided a facile and effective approach for the synthesis of nanocomposite based on CNTs and NiAl-LDH with enhanced supercapacitor behaviors, which can be potentially applied in energy storage conversion devices.

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1. Introduction

In recent years, electrochemical capacitors (ECs) have attracted widely attention owning to their high power density, fast charge-discharge and long cycle life, etc. [1,2]. In general, ECs can be divided into electrochemical double layer capacitors (EDLC) and faradaic pseudocapacitors. Layered double hydroxides (LDH), such as NiAl-LDH and CoAl-LDH, are considered to be one of the most promising electrode materials with pseudo-capacitive properties [3,4] because of their high specific capacitance, low cost, multiple oxidation states for reversible faradaic reactions and high anion exchange capacity [5–8].

However, the relative low specific surface area, poor intrinsic electrical conductivity and poor electrochemical stability of LDH restrict their applications in electrode materials. The incorporation of highly conductive carbon materials (such as active carbon, carbon nanotubes and graphene) into LDH material is thought to be one of the promising and effective strategies to solve the problem aforementioned [9]. The nanocomposite of LDH and carbon materials provide a high surface area and suitable pore-size distribution, in which more electroactive sites participate in faradic redox reactions and a fast electron transfer is guaranteed. The as-obtained nanostructures of LDH and carbon materials exhibit remarkably improved electrochemical characteristics, including high specific capacitance, high rate capability and excellent cycling stability [9]. In recent years, there are many reports on supercapacitors based on NiAl-LDH. Niu and co-workers prepared a RGO/NiAl-LDH composite with a specific capacitance of 1630 F g^{-1} at 1 A g^{-1} [10]. Wang et al. fabricated a ternary NiAl-LDH/CNT/GNS composite, exhibiting excellent specific capacitance (1562 F g^{-1} at 5 mA cm^{-2}), and high capacitance retention (96.5% at 10 mA cm^{-2} after 1000 cycle test) [11]. Wei et al. synthesized a carbon quantum dots/NiAl-LDH composite through a facile one-step solvothermal method. The composite exhibited superior specific capacitance of 1794 F g^{-1} at 2 A g^{-1} and excellent cycle performance (93% retention over 1500 cycle) [12]. Similarly, other NiAl-LDH based composites (such as RGO/NiAl-LDH [13–17], carbon nanoparticles/NiAl-LDH [18], Nicke Foam/CNTs/NiAl-LDH [19] and carbon nanofiber/NiAl-LDH [20]) have been synthesized for supercapacitor electrode materials. Carbon nanobutes (CNTs) have excellent performances of electrical conductivity, electrochemical stability, environmental friendliness and low cost. Introducing a small amount of CNTs to LDH could effectively combine the redox reactive property of LDH and good electronic conductivity of CNTs. Accordingly, CNTs/LDH nanocomposite could improve system conductivity and electron transfer ability, which will be a promising way to improve the electrochemical performance and stability [9,21]. For example, Zhao et al. fabricated a CNTs/NiMn-LDH composite with a maximum specific capacitance (2960 F g^{-1} at 1.5 A g^{-1}) and excellent rate capability (79.5% retention at 30 A g^{-1}) [22]. Yu et al. reported the synthesis of exfoliated CoAl-LDH/CNTs composite with a specific capacitance of 884 F g^{-1} at 5 mA cm^{-2} [23].

Herein, pure NiAl-LDH and nanocomposite of CNTs and NiAl-LDH were prepared by using a facile one-step homogeneous precipitation approach. The morphology and structure of the CNTs/NiAl-LDH nanocomposite were systematically studied by using X-ray diffractometer (XRD), Transmission electron microscopy (TEM) and IR spectra. Electrochemical properties of the obtained materials were characterized by cyclic voltammetry, galvanostatic charge-discharge and electrochemical impedance spectroscopy. As expected, the as-prepared CNTs/NiAl-LDH nanocomposite displayed an excellent specific capacitance of 694 F g^{-1} at 1 A g^{-1} and long cycling life with specific capacitance retention of 92% at 20 A g^{-1} after 3000 cycles.

2. Experimental

2.1. Materials

All chemicals were of analytical grade and used without further purification. CNTs with a diameter of 10–20 nm were purchased from Shenzhen Nanotech Port Co. Ltd.

2.2. Modification of CNTs

In order to form negatively charged CNTs, the pristine CNTs were introduced of carboxyl and hydroxyl groups onto the surface. CNTs were dispersed in 9 M nitric acid with the assistance of sonication for 30 min. Then the suspension was refluxed at 100 °C for 12 h. The resulting acid-treated CNTs were filtered and washed with deionized water until pH = 7. The product dried at 80 °C for 12 h in vacuum oven.

2.3. Preparation of the CNTs/NiAl-LDH nanocomposite

The CNTs/NiAl-LDH nanocomposite was prepared via a facile one-step homogeneous precipitation approach. The acid-treated CNTs were dispersed in 80 mL deionized water with the assistance of stirring for 30 min to form a homogeneous CNTs dispersion (0.5 mg/mL). Subsequently, $Ni(NO_3)_2{\cdot}6H_2O$ (0.2320 g), $Al(NO_3)_3{\cdot}9H_2O$ (0.1500 g) and urea (0.1680 g) were added into the above CNTs suspension, respectively. After being vigorously shaken for another 20 min, the suspension was then hydrothermally treated at 100 °C for 24 h. The resulting product was filtered, washed with deionized water and alcohol until pH = 7, and dried at 80 °C for 12 h in a vacuum oven.

For comparison, pure NiAl-LDH was also prepared in the same procedure using $Ni(NO_3)_2{\cdot}6H_2O$, $Al(NO_3)_3{\cdot}9H_2O$ and urea but without CNTs.

2.4. Electrode fabrication

The working electrodes were prepared by mixing the CNTs/NiAl-LDH nanocomposite (80 wt%) with acetylene black (15 wt%) and polyvinylidene fluoride (PVDF, 5 wt%) [24]. The three former constituents were mixed together with small amounts of ethanol to obtain a homogeneous slurry. Then the slurry was coated onto Ni foam and dried at 80 °C for 12 h in a vacuum. After drying, the coated foam was pressed at pressure of 10 MPa to make the electrode material adhere to the collector.

2.5. Characterization

X-ray diffraction (XRD) analysis was carried out with a D/Max2550VB+/PC X-ray diffractometer with Cu Kα (λ = 0.15406 nm), using an operation voltage and current of 40 kV and 30 mA, respectively. Transmission electron microscopy (TEM) images were collected using a JEM-2100 microscope working at 200 kV. Specimens for observation were prepared by dispersing the samples into alcohol by ultrasonic treatment and dropped on carbon-copper grids. Fourier transform infrared (FT-IR) spectra were obtained on a Brucher EQUINX55 FT-IR spectrophotometer by a standard KBr disk method in the range 400–4000 cm^{-1}. The cyclic voltammogram (CV) and electrochemical impedance spectroscopy (EIS) were measured by a CHI 660E electrochemical workstation. The galvanostatic charge/discharge curves and cycle life were conducted on a LAND CT2001A test system. The electrochemical performance of the samples was studied in a three-electrode test system. A platinum-foil (4 cm^2), Hg/HgO electrode and 6 M KOH aqueous solution were used as the counter electrode, reference electrode

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