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Regular Article

Nickel cobaltite nanosheets coated on metal-organic framework-derived mesoporous carbon nanofibers for high-performance pseudocapacitors

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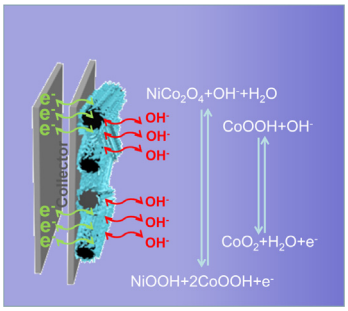
GRAPHICAL ABSTRACT

$NiCo_2O_4$ nanosheets coated on zinic-trimesic acid fiber-derived mesoporous carbon nanofibers within surface O-functionalities as high-performance electrode for supercapacitors.



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ABSTRACT

Core-shell structured carbon nanofiber@metal oxide is one of the most promising hybrid electrodes as supercapacitors, in which the pseudocapacitive metal oxides can be fully exerted and stabilized on the carbonaceous scaffolds. However, facile fabrication of mesoporous carbon nanofibers and integration of them with metal oxides are challenging. Herein, we report a new type of mesoporous carbon nanofibers (MCNs), derived from zinc-trimesic acid fibers, acting as the scaffolds to anchor nickel cobaltite ($NiCo_2O_4$) nanosheets after surface O-functionalization. The resultant core-shell OMCN@$NiCo_2O_4$ nanostructure is demonstrated by scanning electron microscope (SEM), elemental mapping, bright-field/high-resolution transmission electron microscope (TEM), selected area electron diffraction (SAED) studies. The anchored $NiCo_2O_4$ nanosheets are dense (97.4%), and have a strong interaction with OMCN, as revealed by X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), thermogravimetric analysis (TGA) and H_2-temperature programmed reduction (H_2-TPR) techniques. As expected, the OMCN@$NiCo_2O_4$ is highly efficient, showing a high specific capacitance of 1631 F g^{-1} at the current density of 1 A g^{-1}, excellent rate capability and superior cycling stability up to 5000 cycles within a high capacitance retention ratio of 94.5%. This research opens the avenue to fabricate high-efficiency carbon-metal oxide electrodes using metal-organic framework fiber-derived mesoporous carbon nanofibers and integration of them with $NiCo_2O_4$ nanosheets by increasing the interfacial interaction.

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1. Introduction

With the great demand for energy consumption and the urgent concern about the environmental problem, there has been intensive research focusing on energy conversion and storage [1]. Supercapacitors are ideal candidates for energy storage because of their high power density, fast charge-discharge process and long life span. It is known that an electrode material is the key component that governs device performance, and an ideal electrode should integrate the merits of large capacitance, superior stability and low cost. Currently, Faradaic reaction-based pseudocapacitors are much favored because they exhibit substantially larger specific capacitance and thus higher energy density than electrical double-layer capacitors (EDLCs) [2]. Among various pseudocapacitor materials studied so far, transition metal oxides show great potentials owing to their miscellaneous structures, multiple oxidation states that enable rich redox reactions, natural abundance, and higher stability than conductive polymers [3].

Spinel nickel cobaltite ($NiCo_2O_4$) is one of the most promising candidates that has high theoretical capacity (>3000 F g^{-1}), low diffusion resistance to protons and cations and good electrolyte penetration, and it is superior to the single component oxides of NiO and Co_3O_4 [4]. However, the capacitance of bulk $NiCo_2O_4$ is far from satisfactory since the elctroactive surface accessible is very limited. Engineering $NiCo_2O_4$ at the nanoscale offers unique properties by increasing the active surface area and shortening the ion transport pathway. Several types of $NiCo_2O_4$ nanostructures, including nanoparticles [5], nanoplatelets [6], nanotubes [7] and nanosheets [8], show enhanced capacitive performances as compared to bulk $NiCo_2O_4$. There are still many challenges that need to be addressed, such as poor rate capability and cycling stability that are related to low conductivity, large volume change and rapid aggregation/pulverization of $NiCo_2O_4$ during the repeated charge/discharge process. Recent studies have indicated that superior electrode capability and stability can be achieved by anchoring $NiCo_2O_4$ nanostructures on conductive substrates. Growing $NiCo_2O_4$ nanoneedles on conductive Ni foam [9] and carbon textile [10] renders binder-free electrodes, showing improved rate capability and cycling stability. However, high mass density of Ni foam and carbon textile leads to the decrease of the capacity and energy density finally. Direct growing $NiCo_2O_4$ nanostructures on light carbons, including carbon nanofiber (CNF), carbon nanotube (CNT), graphene and mesoporous carbon, may compromise between capacity, capability and stability. The $NiCo_2O_4$-carbon nanostructures will show unprecedented advantages of excellent flexibility, light weight and chemical inertness to acidic and basic electrolytes.

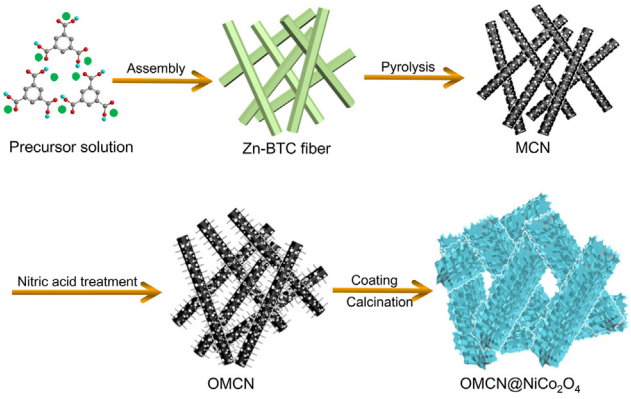
The merging concept of core-shell carbon@metal oxide nanostructures has proved to be effective to fabricate high-efficiency electrodes with large capacity, high rate capability and excellent cycling stability. Hierarchical CNT@$NiCo_2O_4$ nanostructure has been reported and it can shorten the electron/ion diffusion path during the electrochemical process, and thus exhibits a superior capacitive performance [11]. CNF is superior to CNT in the aspect of mechanical robustness (high tensile strengths of up to 7 GPa and high moduli up to 900 GPa) that is required to construct well-defined hierarchical nanostructures [12]. However, facile fabrication of high-efficiency core-shell structured CNF@$NiCo_2O_4$ is challenging. Direct pyrolysis of electrospun fibers that are composed of metal salts and carbon sources, can yield $NiCo_2O_4$-containing carbon nanofibers facilely. Nevertheless, the electroactive surface is very low since most $NiCo_2O_4$ nanoparticles are embedded into the carbonaceous network [13]. Alternatively, anchoring $NiCo_2O_4$ nanosheets or nanoneedles on pre-formed carbon nanofibers renders more $NiCo_2O_4$ exposed, whereas, the loosely attached $NiCo_2O_4$ nanostructures are inclined to leach during the repeated charge-discharge process [14,15]. Currently, Te nanowire [16] and SBA-15 [17] are utilized as the hard templates for the controlled growth of $NiCo_2O_4$ nanorods and/or nanosheets on glucose-derived porous carbon nanofibers, as highly active and stable supercapacitor electrode and lithium storage. However, such a hard-templating approach is less favored owing to the tedious synthesis and high cost. Therefore, facile fabrication of hierarchical CNF@$NiCo_2O_4$ nanostructures with sufficient active surface, intimate contact and low cost, is highly desirable.

Herein, we report a new type of mesoporous carbon nanofibers (MCNs), derived from zinc-trimesic acid fibers, acting as the scaffolds to anchor $NiCo_2O_4$ nanosheets after surface O-functionalization (Scheme 1). The resultant nanostructure has dense but well-dispersed $NiCo_2O_4$ nanosheets firmly coated on O-functionalized MCN (OMCN). As expected, the elaborately fabricated OMCN@$NiCo_2O_4$ exhibits remarkable capacitive performance with large specific capacity, high rate capability and excellent cycling stability, all of which make it a promising electrode for high-performance pseudocapacitors.

2. Experimental

2.1. Synthesis

Controlled synthesis of zinc-trimesic acid fibers was first performed by assembling zinc salts and ligands in N,N-Dimethylformamide (DMF). Typically, 1.10 g of $Zn(Ac)_2{\cdot}2H_2O$ (6 mmol) combined with 1.26 g of trimesic acid (H_3BTC, 5 mmol) was dissolved in 50 mL of DMF under constant agitation at the ambient temperature for 40 min. The resultant mixture was solvothermally treated at 140 °C for 12 h to give zinc-trimesic acid (Zn-BTC) fibers. These fibers were collected by filtration, dried under vacuum, and then pyrolyzed at 910 °C for 2 h to yield mesoporous carbon nanofibers, MCNs. The resultant MCNs were acidified and then used as the scaffolds to anchor Ni-Co hydroxides, affording OMCN@$NiCo_2O_4$ after calcination. Typically, 1.0 g of MCN combined with 50 mL of nitric acid (65–68 wt%) was treated at 60 °C for 12 h, yielding OMCN. Then 120 mg of OMCN was dispersed in 3.3 mL of EtOH, and 0.048 g of $Ni(NO_3)_2{\cdot}6H_2O$ (0.17 mmol), 0.097 g of $Co(NO_3)_2{\cdot}6H_2O$ (0.33 mmol) and 0.105 g of hexamethylenetetramine (HMTM, 0.75 mmol) dissolved in 6.7 mL of distilled water were introduced. The resultant mixture was heated at 90 °C for 4 h, and then centrifugated and washed to render OMCN@Ni-Co hydroxide. The following calcination was conducted at 350 °C for 2 h in air, yielding OMCN@$NiCo_2O_4$. In this study, $NiCo_2O_4$ nanosheet and MCN@$NiCo_2O_4$ were also prepared for comparison (Supporting Information).



Scheme 1. Schematic illustration for the OMCN@$NiCo_2O_4$ synthesis.

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