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Interface-rich core-shell ammonium nickel cobalt phosphate for high-performance aqueous hybrid energy storage device without a depressed power density

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ABSTRACT

Introducing diffusion-controlled battery materials to supercapacitors, can significantly enhance the energy density of supercapacitors, which however encounter depressed power density due to the intrinsic sluggish charge storage kinetics of battery materials. This problem can be efficiently solved by modifying the microstructure, and enable capacitive controlled charge storage mechanism in the battery materials. In this work, a novel interface-rich core-shell structure with $(NH_4)(Ni,Co)PO_4 \cdot 0.67\ H_2O$ nanosheets @ single crystal microplatelets $(NH_4)(Ni,Co)PO_4 \cdot 0.67\ H_2O$ (NCoNiP@NCoNiP) is constructed via a facile two-step hydrothermal method, taking advantage of etching induced Kirkendall effect and Ostwald ripening. This unique structure can enable the extrinsic pseudocapacitance by providing extra charges (e.g. holes, electrons or voids) on the interfaces, and realize synergy and fast charge storage. Specifically, a maximum specific capacity of 190.3 mAh g^{-1} and ultrahigh rate performance with capacity retention of 96.1% from 1 to 10 A g^{-1} in a three-electrode test. The kinetic analysis indicates that the electrochemical response of the hybrid battery-supercapacitor storage devices shows obvious characteristic of supercapacitor especially at high scanning rates. Simultaneously, the hybrid battery-supercapacitor devices based on NCoNiP@NCoNiP exhibits a high energy density of 44.5 Wh kg^{-1} at a power density of 150 W kg^{-1}, which maintains 30 Wh kg^{-1} at high power density of 7.4 kW kg^{-1} with capacitance retention 77.5% after 7000 cycles. This work provides a novel strategy for the application of battery materials in high power devices, by enabling the capacitive charge storage mechanism of battery materials through nanostructure engineering.

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1. Introduction

With increasing concerns of environment and global climate change issues, large-scale renewable and clean energy storage becomes the key strategy to achieve a secure, sustainable energy future [1]. Supercapacitors are considered as one of the promising energy storage devices with high power density and long lifespan, which however suffer from poor energy density [2–5]. Hybrid battery-supercapacitor storage devices (HBSCs) are constructed by introducing diffusion-controlled battery-type materials such as $Na_3V_2(PO_4)_3$, niobium nitride (NbN), nickel cobalt hydroxide, Ni-Co hydroxyl carbonate, NH_4-Ni-Co phosphate etc. as electrode materials, resulting in substantial increase of the energy density [6–14]. Unfortunately, the power density of these HBSCs are inevitably depressed due to the intrinsic sluggish charge storage kinetics of the battery-type materials. Therefore, exploring efficient approach to improve the charge storage kinetics are critical for HBSCs with both high power and energy densities.

Recent studies demonstrate that, the charge storage mechanism can be to some extent transferred from the diffusion control to capacitive control by constructing a hybrid structure with different phases [15,16]. For example, Fan et al. [17] reported a core/branch arrays hybrid structure with graphene foam supported carbon nanotubes and V_2O_3 nanoflakes (GF/CNT/V_2O_3), and the resulting

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material exhibits a high capacity of 612 mAh g^{-1} at 0.1 A g^{-1}, in which the capacitive contribution is up to 72%. Park et al. [18] reported a three dimensional (3D) hybrid structure via the self-assemble of RGO and WS_2, and the capacitive contribution is up to 98.5%. Furthermore, Maier et al. [19] demonstrated a straightforward elucidation of the interface charge storage mechanism, and they found that, by designing composites containing compatible ionic conducting and electronic conducting phases, the composite materials can produce amounts of extra charges, (e.g. holes, electrons or voids) on the interfaces, and realize synergy and fast charge storage. Therefore, the charge transport kinetics can be enhanced by constructing interfaces, and the capacity can also be improved due to the existence of extra charges at the interfaces. In fact, by analyzing the energy storage behavior of various materials, battery-type materials can display capacitive characteristic under certain conditions [20]. For example, some transition metal compounds (SnS_2, V_2O_5. etc) with appropriate structure exhibit apparent capacitive behavior owing to the increased amounts of the defects and enlarged surface area [21,22]. Furthermore, parts of the capacitive behavior can be attributed to the electrical double layer capacity (EDLC) induced by the enlarged surface area, whereas, most of capacitive behavior should be ascribed to pseudocapacitance produced from the fast faradaic redox reaction on the surfaces or the reduced energy barrier of ion intercalation [23]. In comparison to the intrinsic pseudocapacitance material (RuO_2 and MnO_2. etc with aqueous electrolytes), which exhibits capacitive behavior in bulk phases, battery-type materials can only exhibit pseudocapacitive behavior with specific structure or under certain nanoscale, which is classified as extrinsic pseudocapacitance by Simon and Dunn. et al. [24–27]. These materials demonstrate much improved rate performance by breaking away the diffusion controlled charge transfer process, and the CV curves show nearly linear voltage change with current [28,29]. Therefore, the rate performance and the capacitive behavior are not only influenced by the nanoscale and electrical conductivity of the electrode material, but related to the compositing mode and compositing scale between different phases.

The hybrid structure can be built by taking advantage of the Kirkendall effect, which is based on different ion-exchange rate mechanism, and further produce amounts of active sites with improved the specific capacity [30–33]. In this work, on the basis of our previous work [11,12], a novel core-shell structure with $(NH_4)(Ni,Co)PO_4 \cdot 0.67H_2O$ nanosheets @ single crystal microplatelets $(NH_4)(Ni,Co)PO_4 \cdot 0.67H_2O$ (NCoNiP@NCoNiP) is synthesized via a facile method, taking advantage of the synergistic effect of Kirkendall effect and Ostwald ripening [34–36]. The composites display a maximum specific capacity of 190.3 mAh g^{-1} with ultrahigh capacity retention of 96.1% from 1 to 10 A g^{-1}. Meanwhile, the storage mechanism of NCoNiP@NCoNiP with core-shell structure can be to some extent transferred from diffusion control to capacitive control with capacitive contribution up to 54%. Meanwhile, the HBSC based on NCoNiP@NCoNiP and a 3D hierarchical porous carbon (HPC) exhibits a high energy density of 44.5 Wh kg^{-1} at a power density of 150 W kg^{-1}, which maintains 30 Wh kg^{-1} at 7.4 kW kg^{-1}.

2. Experimental

2.1. Synthesis of NCoNiP@NCoNiP

The synthesis of NCoNiP@NCoNiP with core-shell structure was realized through a facile hydrothermal method based on our previous works [11,12]. The targeting material was prepared by the following three steps. Firstly, nickel cobalt hydroxides ($(Ni,Co)(OH)_2$) were synthesized as an intermediate for the subsequent preparation of nickel cobalt ammonium phosphate (NCoNiP). The detailed synthetic process of $(Ni,Co)(OH)_2$ and NCoNiP is provided in supporting information. After that, 0.5 g NCoNiP was dispersed in 50 ml of deionized water and sonicated for 15 min until a light pink solution was formed. Then 0.5 g $Ni(NO)_3 \cdot 6H_2O$ and $Co(NO)_3 \cdot 6H_2O$ were added into the light pink solution and magnetic stirred for 1 h. Subsequently, appropriate amount of urea solution was slowly and dropwise added into the above solution under vigorous stirring for 1 h. After complete dissolution, the as-formed solution was pour into a 100 ml Teflon lined autoclave and maintained at 120 °C for 4 h. The precipitate was washed with water several times, and dried in oven at 80 °C. Thus, the final product was obtained and denoted as NCoNiP@NCoNiP.

2.2. Characterizations

The crystal phase of the as prepared materials was confirmed using powder X-ray diffraction (XRD) Rigaku, $\lambda = 1.5418$ Å), and the morphology was investigated by transmission electron microscopy (TEM) equipped with the selected area electron diffraction (SAED) (Hitachi-7650, 100 kV 10 μA). Nitrogen adsorption/desorption isotherms were measured on a Micromeritics ASAP2020 analyzer.

2.3. Electrochemical measurements

The working electrodes were prepared in the way that was adapted from our previous work [9–12]. The electrochemical performance of the as-prepared NCoNiP@NCoNiP was firstly performed in a three electrode system. In this system, as-prepared NCoNiP@NCoNiP coated on nickel foam, platinum foil, and Hg/HgO (1 mol/L KOH) were used as the working electrode, counter electrode and reference electrode, respectively. The electrochemical test was performed within the voltage window of 0–0.5 V. A hybrid battery-supercapacitor device was assembled using NCoNiP@NCoNiP, hierarchical porous carbon (HPC) [37–39], and glass fiber membrane as positive electrode, negative electrode and separator respectively, and the test was carried out within the voltage range of 0–1.55 V. Cycle voltammetry (CV) experiments were conducted on a CHI660C, and galvanostatic charge-discharge (GCD) tests were implemented on a LAND battery program-control test system. The whole experiment above mentioned was performed in a 6 mol/L KOH aqueous electrolyte. See detailed information in the supporting Information.

3. Results and discussion

The NCoNiP@NCoNiP with core-shell structure was prepared through the synergistic effect between etching induced Kirkendall effect and Ostwald ripening growth process as illustrated in Fig. 1. NCoNiP microplatelets are partly dissolved in a mild alkaline circumstances caused by the hydrolyzing of urea to produce OH^- ions [40,41]. Meanwhile, $(Ni,Co)(OH)_2$ can be obtained through the reaction of liberated Ni^{2+}, Co^{2+} and OH^-. The $(Ni,Co)(OH)_2$ is less thermodynamically stable than NCoNiP due to its lower Ksp values so that released PO_4^{3-} can enable the dissolution $(Ni,Co)(OH)_2$ through the ion exchange between PO_4^{3-} and OH^- based on the Kirkendall effect mechanism [11]. Subsequently, the liberated Ni^{2+}, Co^{2+}, PO_4^{3-} and NH_4^+ can nucleate and further grow into the nanosheets at the surface of NCoNiP microplatelets through the Ostwald ripening growth and finally form the core-shell structure with $(NH_4)(Ni,Co)PO_4 \cdot 0.67\ H_2O$ nanosheets @ single crystal microplatelets $(NH_4)(Ni,Co)PO_4 \cdot 0.67\ H_2O$ (NCoNiP@NCoNiP).

To determine the morphology and the crystalline phase of NCoNiP@NCoNiP, TEM and XRD analysis were performed. The TEM images of NCoNiP@NCoNiP (Fig. 2a and b) indicate the core-shell

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