Promising Rice-Husk-Derived Carbon/Ni(OH)$_2$ Composite Materials as a High-Performing Supercapacitor Electrode

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ABSTRACT: Improving the electrochemical performance of biomass-derived carbon electrode-active materials for supercapacitor applications has recently attracted considerable attention. Herein, we develop hybrid electrode materials from rice-husk-derived porous carbon (RH-C) materials and β-Ni(OH)$_2$ via a facile solid-state reaction strategy comprising two steps. The prepared RH-C/β-Ni(OH)$_2$ (C–Ni) was investigated using scanning electron microscopy (SEM) (energy-dispersive X-ray spectrometer (EDS)), X-ray photoelectron spectroscopy (XPS), and X-ray diffraction (XRD) to acquire the physical and chemical information, which was used to demonstrate the successful fabrication of C–Ni. Thermogravimetric analysis (TGA) measurement results confirmed that the thermal stability of C–Ni changed due to the presence of Ni(OH)$_2$. As expected, C–Ni possesses a high capacitance of ∼952 F/g at a current density of 1.0 A/g. This result is higher than that of pure biomass-based carbon materials under the three-electrode system. This facile preparation method, which was used to synthesize the electrode-active materials, can extend to the value-added utility of other waste biomass materials as high-performing supercapacitor electrodes for energy storage applications.

1. INTRODUCTION

Recently, the overuse of nonrenewable fossil fuel resources (i.e., coal and oil) has resulted in a considerable environmental pollution and energy crisis, which hinders the sustainable development of modern society. Thus, it is necessary to develop new energy technologies from renewable materials, especially from abundant and low-cost biomass-based materials.

In the last few years, supercapacitors, considered an important storage technology, have received considerable interest owing to their high power density, high charge–discharge capability, and durable cyclic lifespan. The performance of supercapacitors typically depends on electrode materials; supercapacitors can be broadly classified into three groups, i.e., (1) carbon-based, (2) metal-oxide-based, and (3) conductive-polymer-based supercapacitors. Biomass-derived carbon materials have been favored by many scholars owing to their low cost, wide distribution, and green and renewable characteristics. Various biomasses (e.g., biostraw, onion, soybean root, tannic acid, and pine nut shell) have been used as precursors to prepare carbon materials. Agricultural products, like stems and husks of grain or cereal plants, plant polyphenol, plant cellulose, and so on, are a great source of biomasses. From the long-term point of view, sustainable biomass has a considerable potential for the production of new-generation electrode materials to replace or complement traditional carbon precursors. According to the energy storage principles, carbon-based supercapacitors are considered electrochemical double-layer capacitors (EDLCs). For EDLCs, the energy storage principle is based on the physical adsorption/desorption of the electrolyte ions at the interface of the electrode, which subsequently leads to the formation of a double layer. Although EDLCs with carbon-based electrodes have the advantages of very high power density and outstanding cycling lifetime, they are restricted in practical application owing to the inferior energy density caused by their low capacity. Thus, many scientists are devoted to studying how to efficiently deal with the above-mentioned drawbacks to meet the need of practical applications. For example, He et al. determined that the capacitance of ladderlike porous carbon can be improved from 165 to 214.5 F/g at a current density of 0.5 A/g using dual N and S doping. Yang et al. have prepared a biomass-derived interconnected hierarchical micro–meso–macroporous carbon with a higher electrochemical performance, which is attributed to the comprehensive effects from the high specific surface area, multilevel porous structure, high level of graphitization, and rich heteroatoms codoping. Up to now, many studies have shown that the energy density of carbon-
based materials can be considerably improved by compounding with transition-metal oxides/hydroxides owing to their ultrahigh theoretical specific capacitance values.

Numerous transition-metal oxides/hydroxides, e.g., CuO,25 Ni(OH)2,26,27 MnO2,28,29 Co(OH)2,30 and Co3O4,31 are excellent electrode materials and have been used to construct high energy and power supercapacitor devices because these materials possess high theoretical capacitances. Among the above-mentioned pseudocapacitive materials, Ni(OH)2 is the most promising owing to the advantages of a distinct redox reaction, controllable morphology, and low production cost.32,33 Our group has reported that Ni(OH)2 could significantly increase the electrochemical performance of carbon nanofibers after they were synthesized on the surface of the matrix materials through a facile solid-state reaction to produce supercapacitor electrodes.34

In this study, we successfully prepared C–Ni composite materials through a simple two-step solid-state reaction using biomass-based carbon materials (Figure 1). The samples were characterized by several common techniques, i.e., SEM, XRD, and XPS, to acquire the chemical and physical information about them. TGA measurement was used to determine the thermal stability of composite materials. A three-electrode setup was used to study the electrochemical performance of C–Ni composite. Supercapacitor electrodes, which were obtained by compounding biomass-based carbon materials and Ni(OH)2, delivered a high specific capacitance of ∼952 F/g at a current density of 1.0 A/g. The developed facile electrode material preparation process presented in this work can extend to the value-added utility of other waste biomass materials, which can be used as electrode-active materials in high-performing supercapacitors and other energy storage equipment.

2. RESULTS AND DISCUSSION

2.1. Characterization of C–Ni Composite Active Materials. Figure 2 presents the microstructure of C–Ni and RH-C. After grinding, the obtained RH-C materials (Figure 2a) appeared as a microsized fragmentary block and possessed a smooth surface; the surface of C–Ni composite materials, as shown in Figure 2b, became relatively tough because of the accumulation of Ni(OH)2 particles on the surface, which suggested the successful preparation of the C–Ni composite. The elemental distribution on the surface of carbonized C–Ni composite materials was investigated using an EDS. According to Figure 2c–f, a uniform distribution of Ni and O could be observed throughout the carbon material surface, which is consistent with the SEM result. XPS was also used to confirm the presence of Ni on the carbon material surface, and the typical peaks in the full survey spectrum are shown in Figure 3a. For pure carbon materials, the peaks at ∼284 and ∼533 eV are ascribed to C 1s and O 1s, respectively. However, a typical wide scan of C–Ni composite materials showed C 1s (∼284 eV), Ni 2p (∼856 eV), and O 1s (∼531 eV) species, which suggested the presence of Ni in the carbon material substrate. The high-resolution spectra of the Ni 2p peak (Figure 3b) confirm the presence of two major binding energies at ∼874 and ∼856 eV, which are assigned to Ni 2p3/2 and Ni 2p1/2, respectively.55 XRD was used to determine the crystal structure of the samples and used for further qualitative analysis. The XRD patterns of RH-C and C–Ni material are shown in Figure 3c. The XRD profile of RH-C shows two main broad diffraction peaks at 21.6 and 43.8°, which correspond to the (002) and (100) crystalline planes of graphite, respectively. After the solid-state reaction, several clear diffraction peaks appear at 18.5, 33.2, 39.4, 51.4, 59.3, and 62.8°, which correspond to the (001), (100), (101), (102), (110), and (111) planes of the β-phase nickel hydroxide, respectively. This result confirms the formation of C–Ni composite. The thermal stability of composites was evaluated by TGA. The weight loss of the samples was recorded in the TG curves in detail (Figure 3d). For carbon materials, the weight remained nearly unchanged and was 96.74% at 800 °C. The weight of C–Ni composites decreased considerably. Weight loss can be divided mainly into three stages. (1) A small weight loss occurred in the first stage from 30 to 200 °C owing to the evaporation of adsorbed and bound water. (2) The second stage occurring from 200 to 600 °C corresponds to the thermal decomposition of Ni(OH)2 into NiO. (3) During the third stage, the composite had almost no significant weight loss. The results of TGA characterization showed that C–Ni had lower stability than that of pure carbon materials due to the introduction of Ni(OH)2.
2.2. Electrochemical Performance of C–Ni Composite Materials. The electrochemical performance of C–Ni was examined using cyclic voltammetry (CV), galvanostatic charge–discharge (GCD), and electrochemical impedance spectroscopy (EIS) tests, and the results are presented in Figure 4. Figure 4a reveals that the area under the CV curve of carbon materials without Ni(OH)$_2$ is relatively small, which indicates a lower capacitance, compared with that of C–Ni composite. Two peaks appeared in the CV profile of C–Ni at 10 mV/s scan rates (i.e., a cathodic peak at ~0.20 V and an anodic peak at ~0.49 V), which were ascribed to the Faradic reaction of Ni(OH)$_2$. Specifically, the mechanism corresponds to the redox reaction in the reversible conversion between Ni$^{2+}$ and Ni$^{3+}$ as follows:

\[
\text{Ni}^{2+} + \text{OH}^- + e^- \rightarrow \text{Ni}^{3+} + \text{H}_2\text{O}
\]

\[
\text{Ni}^{3+} + \text{H}_2\text{O} \rightarrow \text{Ni}^{2+} + \text{OH}^- + \text{H}^+
\]

The upper peak corresponds to the oxidation of Ni$^{2+}$ to Ni$^{3+}$, and the lower peak corresponds to the reduction of Ni$^{3+}$ to Ni$^{2+}$. This redox reaction is responsible for the Faradic capacitance of the C–Ni composite.
Ni(OH)$_2$ + OH$^-\rightleftharpoons$ NiOOH + H$_2$O + e$^-$

As seen in Figure 4b, the current density increased with an increase in the scan rate (between 1 and 20 mV/s), and all CV curves had analogous profiles, which indicated the excellent rate capability of the electrode.$^{32}$ However, anodic and cathodic peaks were clearly shifted toward positive and negative potentials, respectively, with increasing scan rate due to the controlled internal diffusion and limited charge transfer in the electrode surface.$^{41,42}$ GCD curves were recorded to further characterize the electrochemical behavior of the as-synthesized C–Ni composite at different current densities (Figure 4c). The nonlinear charge-discharge plateaus of the GCD curves further demonstrated that the typical Faradic redox capacitance of C–Ni materials was mainly due to the reversible conversion between Ni(OH)$_2$ and NiOOH, which was consistent with the CV profiles. Moreover, nearly symmetric nonlinearities in GCD curves confirmed electrochemical reversibility and the rapid I–V response of these materials.$^{33}$ The specific capacitance values under various current densities are presented in Figure 4d. C gradually increased with an increase in the current density owing to ion diffusion resistance. Under faster-charging conditions, electrolytic ions did not have sufficient time to enter the interior of C–Ni composite materials through the pores, which caused the internal active materials to become inaccessible for the redox reaction and provided the corresponding capacity. At a low current density, the electrolytic ions filled the internal structure of these active materials after a sufficient amount of time. C–Ni delivered high gravimetric capacitance of ~952 F/g at 1 A/g. Even at a high current density of 30 A/g, ~126 F/g can be achieved for the sample. It should be pointed out that this capacitance value is comparable to those of some of the biomass-derived carbon composite electrodes (see Table 1).

3. CONCLUSIONS

In summary, we successfully fabricated biomass-based carbon and β-Ni(OH)$_2$ hybrid electrode materials via a facile solid-state reaction method involving two steps. The physical and chemical properties of C–Ni composite materials were studied with the use of several characterization techniques, including SEM, XPS, and XRD. The results of TGA showed that the thermal stability of composite materials changed owing to the incorporation of Ni(OH)$_2$. The gravimetric capacitance of C–Ni electrode materials reached ~952 F/g at 1.0 A/g based on a three-electrode system configuration. The developed material can be applied for the development of high-performing supercapacitor electrodes for energy storage applications. This facile preparation approach of electrode materials can extend to the value-added utility of other waste biomass materials.

4. MATERIALS AND METHODS

4.1. Materials. Rice husk was purchased from taobao.com (Alibaba, China). A hydrochloric acid solution with >31.6% HCl and potassium hydroxide (KOH) was purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Nickel(II) acetate tetrahydrate [Ni(CH$_3$COO)$_2$·4H$_2$O] and oxalic acid (C$_2$H$_2$O$_4$·2H$_2$O) were provided by Shanghai Shangpu Chemical Co., Ltd. (Shanghai, China) and Tianjing Kaitong Chemical Co., Ltd. (Tianjing, China), respectively. All reagents were of analytical grade, and they were used as received without further purification; all aqueous solutions were used with deionized water.

4.2. Preparation of RH-C. Rice husk was subjected to carbonization for 2 h at 1000 °C in an N$_2$ atmosphere at the heating and cooling rates of 4 °C/min. The entire process of carbonization was conducted in a closed corundum tube. The as-prepared rice-husk-based carbon was ground into powder and then purified by immersing in a 10% (v/v) HCl solution for 3 h, followed by soaking in a NaOH solution (2 M) at 80 °C for 2 h to remove inorganic salts and silica. Finally, porous carbon materials derived from biomass were obtained for further use in supercapacitor electrodes in the next step.

4.3. Fabrication of C–Ni. C–Ni was synthesized via a facile solid-state reaction, which involved two reaction steps, according to our group’s previous study with minor modification.$^{34}$ First, a mixture of oxalic acid and nickel acetate (1:1, mole ratio) was ground for 0.5 h and then rinsed numerous times with distilled water and ethanol. Then, the dried precursor and RH-C were loaded in an agate mortar and ground for 0.5 h. Further, 0.123 g of KOH was then added to the compounds and RH-C were washed with distilled water and eventually dried at 80 °C to achieve the C–Ni composite. The schematic of the preparation process for C–Ni is presented in Figure 1.

Table 1. Specific Capacitance ($C_s$) of Biomass-Derived Carbon Composite Electrode Materials Reported the Literature

| biomass-derived carbon composite materials | electrolyte  | $C_s$ (F/g) | refs |
|------------------------------------------|-------------|-------------|------|
| fungal conidium-derived carbon/MnO$_2$    | 1.0 M Na$_2$SO$_4$ | 263.5 (1 A/g) | 44 |
| pine cone flowers-derived carbon/Ni(OH)$_2$ | 1.0 M KOH | 916.4 (1 A/g) | 45 |
| willow catkin-derived carbon MnO$_2$      | 1.0 M Na$_2$SO$_4$ | 189 (1 A/g) | 46 |
| Faidherbia albida fruit shell-derived carbon MnO$_2$ | 3.0 M KOH | 426.66 (1 A/g) | 47 |
| celery-derived carbon/polyaniline        | 1.0 M H$_2$SO$_4$ | 402 (1 A/g) | 48 |
| banana peel-derived carbon MnO$_2$       | 1.0 M Na$_2$SO$_4$ | 139.6 (0.3 A/g) | 49 |
| bamboo leaves-derived carbon/CuOx        | 3.0 M KOH | 147 (1 A/g) | 50 |
| husk-derived carbon/Ni(OH)$_2$           | 6.0 M KOH | 952 (1 A/g) | this work |
4.4. Characterization. The surface morphology of the samples was investigated using SEM (S-3000N; Hitachi, Japan). XPS (Axis Ultra DLD) was performed to determine the surface elemental composition. XRD measurement was used to obtain the crystallographic information and performed on a PANalytical Empyrean instrument (Netherlands) with a Cu Kα radiation in the 2θ range of 10°–70°. Thermal stability was analyzed using a METTLER TOLEDO TGA/DSC/110SF instrument in a N2 atmosphere from 30 to 800 °C with a ramping rate of 10 °C/min.

4.5. Electrochemical Testing. A three-electrode setup with 6 M KOH electrolyte was used for all electrochemical measurements (which consisted of a platinum foil as the counter electrode, Hg/HgO electrode as the reference electrode, and a working electrode), and the electrochemical performance was evaluated using an electrochemical workstation (CHI660E; Shanghai Chenhua, China) at room temperature. The preparation of the working electrode was according to a previously reported procedure. Briefly, the electrode was prepared by pressing a slurry into a 1 cm × 1 cm nickel foam; the slurry contained active materials, polyvinylidene fluoride), and carbon black with the mass ratio of 80:5:15 in a small amount of N-methylpyrrolidone solvent; then, the mixture was dried at 80 °C. The prepared C–Ni composite coated onto a Ni-foam substrate served as a working electrode for the electrochemical measurements. Galvanostatic discharge (GCD) curves were used to determine the specific capacitance ($C_s$) of the electrode (F/g) based on the following equation:

$$C_s = \frac{I \times \Delta t}{m \times \Delta V}$$

where $\Delta V$ is the potential window (V), $m$ is the mass of the as-prepared active material (g), $I$ is the discharge current (A), and $\Delta t$ is the discharge time (s).

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