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Research paper

Cabbage-like α-$Ni(OH)_2$ with a good long-term cycling stability and high electrochemical performances for supercapacitor applications

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ABSTRACT

Nanostructured $Ni(OH)_2$ is a promising candidate to be used as an extrinsic pseudocapacitors material. Here, nickel hydroxide ($Ni(OH)_2$) was grown on the surface of a Ti plate, galvanostatically via electrodeposition method. In the electrodeposited films a stable α-$Ni(OH)_2$ phase with particles in cabbage form and average crystallite size of 20 nm is obtained. The α-$Ni(OH)_2$ sample exhibits excellent electrochemical performance at different current densities and good cycling stability. A maximum specific capacitance of 1903 $F\,g^{-1}$ is achieved at a current density of 1 $mA\,cm^{-2}$. The energy and power density values found are 42.31 $W\,h\,kg^{-1}$ and 430 $W\,kg^{-1}$.

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1. Introduction

Supercapacitors (SCs), also called electrochemical capacitors or ultracapacitors have been recognized for over 25 years for having superior performance, becoming for many applications, such as, the high efficiency energy storage for electric and hybrid vehicles and portable wireless communication, competitive to batteries [1]. They possess advantages such as high power, high cycle life, require no maintenance, safe and non-aggressive environment [2].

The SCs, based on their charge-storage mechanism, can be classified in three types. The electrical double-layer capacitors (EDLC), dominated by electrostatic charge diffusion and accumulation at the electrode/electrolyte interface, exhibiting high power density and cyclic stability, but they have the disadvantage to offer low capacitance values [3]. In these devices the electrode material is carbon based [4–6]. Meanwhile the pseudocapacitors (PC), governed by Faradaic reactions at the electrode materials develop high capacitance values, but present low conductivity and cyclic stability problems [3]. The electrode material in this type of supercapacitors is based primarily on transition metal oxides and conductive polymers [7]. Advanced approaches to increase the energy density and the stability are to hybridize the electrode materials by adding electrochemically active materials to a carbon-particle-based electrode layer [8–11].

Nickel hydroxide has aroused great interest as an electrode material for supercapacitors. This material has a high theoretical capacitance, excellent electrochemical properties, stability and low production cost. Nickel hydroxide can be found in two polymorphic forms: β-$Ni(OH)_2$, very stable thermodynamically and α-$Ni(OH)_2$, more electrochemically active but unstable in basic medium. The α-$Ni(OH)_2$ phase is transformed to β-$Ni(OH)_2$ phase when the material is subjected to charging and discharging cycles in basic electrolytes [12]. Several synthesis methods have been used to produce nickel hydroxide, including: hydrothermal [13], sol-gel [8], electroplating [9], deposition with templates [14] and electrodeposition [15]. The electrodeposition method is very attractive because of its flexibility and relative simplicity.

In the present work, the electrodeposition method was used for the synthesis of nickel hydroxide directly on titanium substrate. Nickel sulfate was used as electrolyte for the deposition, without using templates or additional material. The followed route presented a quick and simple method to obtain materials for electrodes used in supercapacitor applications.

2. Experimental

2.1. Synthesis of $Ni(OH)_2$

All chemicals were analytical grade without any previous purification. A Ti substrate (with a surface area of 1 cm^2 and 99.7% purity

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(TiBrazil)) was previously cleaned by sandblasting, followed by thermochemical treatment at 10% (w/v) in oxalic acid solution during 20 min. These electrodes were washed with ultrapure water (Milli-Q) and dried at 105 °C for 4 h. The electrodeposition was performed in a standard three-electrode glass cell consisting of the clean Ti substrate working electrode, a platinum plate as the counter electrode and Ag/AgCl as a reference electrode at room temperature. Nickel hydroxide was electrodeposited upon Ti substrate from 0.1 mol L^{-1} $Ni(SO_4)_2 \cdot 6H_2O$ aqueous electrolyte through a galvanostatic deposition for 30 min and a current of −2.0 mA. Then, this nickel hydroxide electrode was carefully rinsed several times with ultrapure water and finally dried in air.

2.2. Characterization

The surface morphology of the nickel hydroxide thin film was examined by SEM (Quanta FEG-20, FEI Company) and TEM (Morgagni FEI 80 kV, USA). XRD data were collected using a Philips X'Pert diffractometer. The electrochemical characterization was carried out using the thin-film prepared on the titanium substrate as the working electrode, a platinum wire as the counter electrode, and Ag/AgCl/saturated KCl as the reference electrode. A solution of 1.0 mol L^{-1} KOH was used as the electrolyte solution. Cyclic voltammograms were acquired in a potential range between 0.0 and 0.9 V at different scan rates, and galvanostatic charge–discharge (GCD) cycles were recorded within a potential range of 0.2 –0.6 V at the different current densities. The electrochemical impedance spectra were collected at the frequency range from 0.1 to 10^4 Hz with an Open Circuit Potential (OCP). All measurements were conducted using an Autolab Potentiostat (PGSTAT 302N).

3. Results and discussion

Fig. 1a presents the X-ray diffraction patterns for $Ni(OH)_2$ films prepared by electrodeposition method. Fig. 1a shows the diffraction peaks that can be indexed as α-$Ni(OH)_2$ phase (JCPDS File No. 22-0444), with lattice parameters of $a = b = 3.08$ Å and $c = 8.0$ Å, and space group *P*3*lm*. The diffraction peaks at 11.1°, 22.2°, 35.5°, 39.0°, 45.3° and 52.4° can be ascribed to the (001), (002), (111), (200), (004) and (210) crystal planes to the α-Ni$(OH)_2$ phase. The crystallite size of the particles was estimated from X-ray peak broadening techniques, applying the Scherer's equation [16]. The average crystallite size estimated was smaller than 20 nm.

Fig. 1b shows FESEM images of an electrochemically deposited nickel hydroxide thin film onto a Ti foil. It can be seen in Fig. 1b that the films exhibit a nanoscaled 3D structure with cabbage-like morphology. A closer examination of the FESEM images suggests the presence of smaller nanoparticles, which are agglomerated forming nanorods which together give the appearance of cabbage. In addition, the nanorods exhibit a hierarchical array feature with empty space among adjacent nanorods. It is advantageous for the electrolyte ions to access the active materials, which may contribute to enhancement of capacitive performance. TEM images were carried out to further investigate the surface morphology of α-$Ni(OH)_2$ films. Fig. 1c shows α-$Ni(OH)_2$ nanoparticles with an estimated size less than 20 nm. These particles are

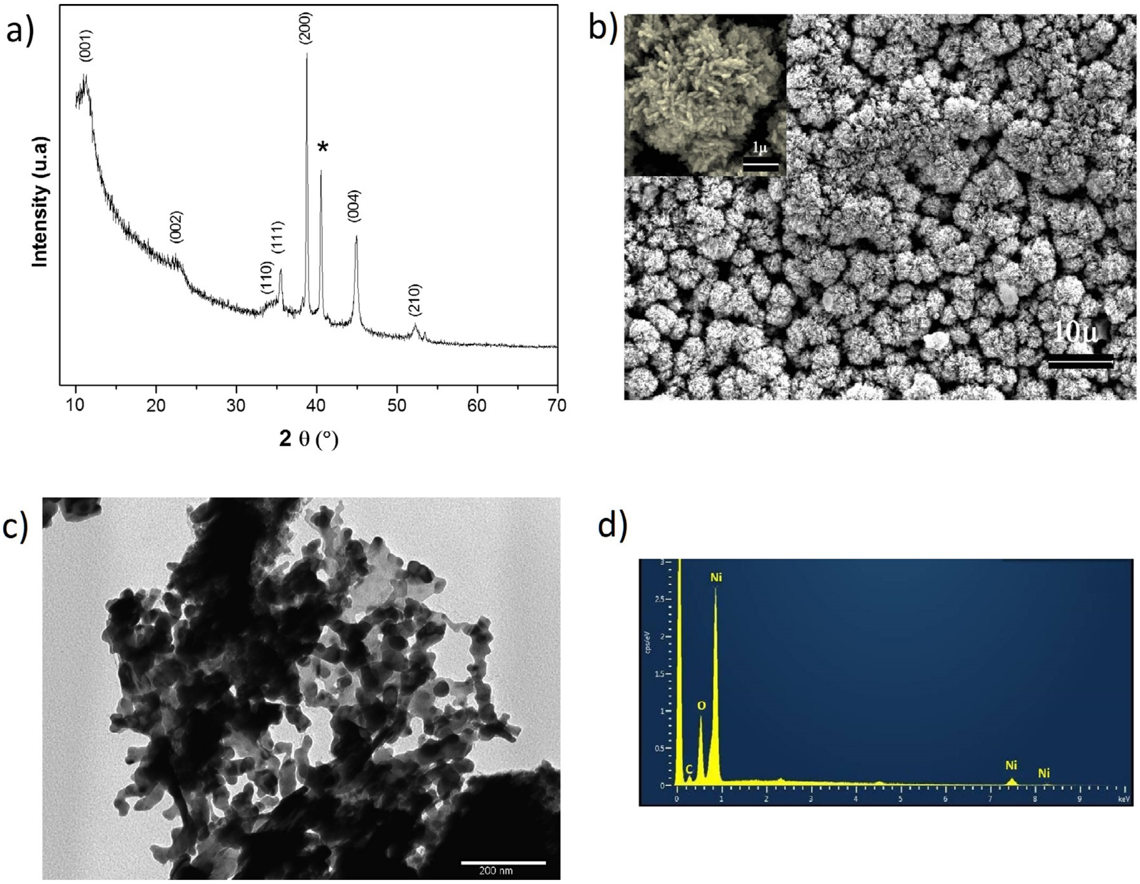

Fig. 1. (a) X-ray diffraction patterns of α-$Ni(OH)_2$ film. (b) SEM image of the α-$Ni(OH)_2$ film. (c) TEM images of α-$Ni(OH)_2$ film. (d) EDS spectrum of α-$Ni(OH)_2$ film;

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