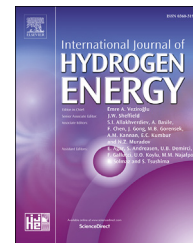




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Co(OH)₂ nanoparticles deposited on reduced graphene oxide nanoflake as a suitable electrode material for supercapacitor and oxygen evolution reaction in alkaline media

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ARTICLE INFO

Article history:

Received 20 March 2017

Received in revised form

18 May 2017

Accepted 26 May 2017

Available online xxx

Keywords:

Cobalt hydroxide nanoparticles

Reduced graphene oxide nanoflake

Hydrothermal process

Pseudocapacitors

Oxygen evolution reaction

ABSTRACT

In this work, cobalt hydroxide nanoparticles are simply synthesized (size is about 50 nm) and deposited on the reduced graphene oxide nanoflake by the hydrothermal method. Then, the ability of glassy carbon electrode modified with this low-cost nanocomposite is examined as a supercapacitor and oxygen evolution electrocatalysts in 2.0 mol L⁻¹ KOH by a three-electrode system. The modified electrode as a pseudocapacitor with potential windows of 0.35 V, exhibits a powerful specific capacitance (235.20 F g⁻¹ at 0.1 A g⁻¹ current density), energy density, stability (about 90% of the initial capacitance value maintain after 2000 cycles at 1.0 A g⁻¹) and fast charge/discharge ability. Furthermore, the modified electrode displays a good electrocatalytic activity for oxygen evolution reaction with a current density of 10.0 mA cm⁻² at 1.647 V, small Tafel slope of 56.5 mV dec⁻¹, good onset potential of 1.521 V vs. RHE and suitable durability.

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Introduction

In the 21st century, requests for renewable energy and energy storage/conversion instrument have increased due to the decline of fossil fuels and increasing human passion for using different portable electronic devices [1–3]. Hydrogen fuel is one of the renewable and environmental friendly energies generated from water splitting in cathodic reaction known as a hydrogen evolution reaction (HER) [4]. The water splitting in the anodic electrode consists of oxygen evolution reaction (OER) which needs high overpotential and is the main key in water splitting, fuel cells and metal-air batteries [5–7].

Although high capacitance power, in brief time, is very important in the various electrical systems, batteries and fuel

cells don't provide a high power on a short time scale. So, supercapacitors with intermittent high-power conditions have more usage as an assist to improve reliability, power density, reversibility and durability [2,8–10]. Supercapacitors are generally divided into two kinds: a) electric double-layer capacitors (EDLCs) which can keep the ions in the electrolyte at the surface of nanostructure electrodes such as carbon that cause to storage electrical charge; and b) pseudocapacitors (PCs) with certain transition metal oxides/hydroxide or a complex of them with heteroatoms which is known as electrochemical capacitors. In PCs, the materials with high and reversible charge/discharge capability is applied at the surface or nearby the surface reaction [2,10–13]. According to the two different storage mechanisms, the EDLCs display higher rate

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<http://dx.doi.org/10.1016/j.ijhydene.2017.05.193>

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capability and cycle stability, but poorer capacitance and energy density than the PCs [2].

Among transition metal compounds, ruthenium compounds as a noble metal have the excellent specific capacity and high electrocatalytic activity for PCs and OER, respectively. But the price of ruthenium compounds is an important limitation for using them in wide application [5,10,14].

In the energy storage field and electrocatalyst for OER, many researchers have focused on oxides, hydroxides and heteroatoms compound of low-cost transition metals like V, Mn, Fe, Co, Ni and Ni are which combined with conductive polymers or/and carbonaceous in various architecture designs to improve properties of PCs electrodes and OER electrocatalytic activity [5,6,8,14–31]. However, the construction of PC electrode materials with a great electrochemical operation like high capacitance, reversibility and speed capability are an important challenge. While, the important factors for electrocatalyst materials in OER are low onset potential, small Tafel slope and high current density [32]. Furthermore, high stability and low-cost of materials are important for both of them. In particular, Co and Ni compounds as a low-cost and abundant materials have been demonstrated as promising alternatives to IrO_2 and RuO_2 for OER and are widely used as supercapacitors because of their effective catalytic activity and good stability in alkaline media [29,33].

The electrocatalytic properties of cobalt and nickel compounds are referred to d orbitals and improve with deposited on carbon substrate. Carbon nanostructure materials like reduced graphene oxide (RGO) with high surface area, mechanical strength, structural flexibility and electrical conductivity are being used as a substrate for nanocatalyst in fuel cells, biosensors, batteries and supercapacitors [34–41]. Newly, a simple procedure for preparation of reduced graphene oxide nanoflake (RGONF) from graphite at low temperature (160 °C) was introduced by our group. Unlike previous synthetic methods of RGONF, it doesn't need any chemical reduction sources for producing RGONF from graphite [42].

Herein, a nanocomposite material of Co(OH)_2 nanoparticles deposited on the reduced graphene oxide nanoflake (Co(OH)_2 @RGONF), is examined as an electro material for supercapacitors and oxygen evolution reaction (OER) in the alkaline solution. The linear sweep voltammetry (LSV), cyclic voltammetry (CV) and galvanostatic charging/discharging measurements were investigated to assess the properties of modified glassy carbon electrode with Co(OH)_2 @RGONF nanocomposite (Co(OH)_2 @RGONF/GCE) in supercapacitors reaction and OER. Furthermore, the electrochemical impedance spectroscopy (EIS) technic was used to study the kinetic and calculate the charging/discharging ability, imaginary and real capacitance of the Co(OH)_2 @RGONF/GCE.

Experimental

All the chemical materials used in this manuscript are purchased from Merck or Sigma in analytical grade and double distilled (DD) water was used for the preparation of aqueous solutions.

Synthesis of reduced graphene oxide nanoflake (RGONF)

In a typical synthesis, RGONF was prepared by a previous method in our laboratory [42]. Into a 100-ml flask containing an acidic solution H_2SO_4 (Merck) (70.0 g), HNO_3 (Merck) (10.0 g) and 5.0 g DD water, 4.0 g $\text{K}_2\text{Cr}_2\text{O}_7$ (Merck) were added and all of the $\text{K}_2\text{Cr}_2\text{O}_7$ (Sigma) was dissolved under ultrasonication. After that, 1.0 g graphite powder was mixed with above solution and refluxed about 1.0 h in 80 °C and 160 °C, respectively (heating speed was 10 °C min^{-1}). After cooling the resulting suspension, 10.0 ml of hydrogen peroxide (Sigma) increased to the flask and the homogenous black suspension was rinsed with DD water to achieve neutral pH (~7). Finally, the resulting black homogenous suspension was collected via centrifuging and dried at 50 °C.

Synthesis of Co(OH)_2 nanoparticle

Co(OH)_2 nanoparticle was synthesized as below, in 100-ml capacity Teflon-lined autoclave, 0.1170 g $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ (Merck), and 0.0375 g trimethyl tetradecyl ammonium bromide (TTAB) from Merck were added. Then 2.50 ml hexylamine and 50.0 ml hexane (Merck) were appended respectively and sonicated for 5 min. After that, the sealed autoclave was heated at 150 °C for 10 h. Ultimately, the pink precipitate was collected via centrifuging, rinsed with DD water and dried at room temperature.

Synthesis of Co(OH)_2 @RGONF

For the preparation of cobalt hydroxide @RGONF (Co(OH)_2 @RGONF) nanocomposite, 15.0 mg of RGONF was dispersed in 49.0 mL of DD water and 1.0 ml ethanol. After a 30 min sonication, the black homogeneous suspension of RGONF appeared. Then, 7.5 mg of Co(OH)_2 was added to the previous suspension and refluxed at boiling point for 2 h. Finally, the suspension was centrifuged and the resulting nanocomposite was dried at 60 °C.

Characterization and electrochemical measurements

The X-ray diffraction (XRD) pattern (Bruker D8/Advance X-ray diffractometer) with Cu $K\alpha$ radiation (1.5406 Å) was used for investigation of crystallite structure and data were measured in the 2θ range between 10 and 90 with the step size of 0.05°. The surface appearance of the nanocomposite was checked by field emission scanning electron microscopic (FE-SEM) using NOVA NANOSEM 230 equipped with EDX, Cold Field Emission. Transmission electron microscopy, (TEM) Philips XLC, was used to investigate and show the size of Co(OH)_2 nanoparticles. Atomic force microscopy (AFM: Bruker Nanos instrument made in Germany) was applied to investigate the surface porosity of the modified electrode.

The electrochemical behavior of the nanohybrid was tested in 2.0 mol L^{-1} potassium hydroxide solution in a three-electrode glass cell.

The modified GCE, Pt wire and Ag/AgCl (saturated KCl) were used as working, counter and reference electrode, respectively. The CV, EIS tests and galvanostatic charge/discharge cycling studies were performed via Autolab

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