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Binary cobalt ferrite nanomesh arrays as the advanced binder-free electrode for applications in oxygen evolution reaction and supercapacitors



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HIGHLIGHTS

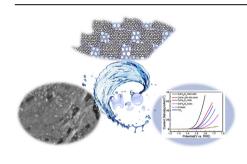
- CoFe₂O₄ nanomesh arrays are successfully synthesized on the nickel foam.
- Porous CoFe₂O₄ nanomesh arrays own the large surface area and active sites.
- Synergistic effect of Co and Fe provides eminent electrochemical performance.
- CoFe₂O₄ NM-As/Ni owns a potential catalytic/capacitive performance.

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G R A P H I C A L A B S T R A C T



ABSTRACT

The porous CoFe₂O₄nanomesh arrays are successfully synthesized on nickel foam substrate through a high temperature and pressure hydrothermal method, following by the thermal post-treatment in air. The CoFe₂O₄ nanomesh arrays own numerous pores and large specific surface area, which is in favor of exposing more active sites. In consideration of the structural preponderances and versatility of the materials, the CoFe₂O₄ nanomesh arrays have been researched as the binder-free electrode materials for electrocatalysis and supercapacitors. When the CoFe₂O₄nanomesh arrays on nickel foam (CoFe₂O₄ NM-As/Ni) directly act as the free-binder catalyst toward catalyzing the oxygen evolution reaction (OER) of electrochemical water splitting, CoFe₂O₄ NM-As/Ni exhibits an admirable OER property with a low onset potential of 1.47 V(corresponding to the onset overpotential of 240 mV), a minimal overpotential ($\eta_{10} = 253$ mV), a small Tafel slope (44 mV dec⁻¹), large anodic currents and long-term durability for 35 h in alkaline media. In addition, as an electrode of supercapacitors, CoFe₂O₄ NM-As/Ni obtains a desired specific capacitance (1426 F/g at the current density of 1 A/g), remarkable rate capability (1024 F/g at the current density of 20 A/g) and eminent capacitance retention (92.6% after 3000 cycles). The above results demonstrate the CoFe₂O₄ NM-As/Ni possesses great potential application in electrocatalysis and supercapacitors.

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1. Introduction

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http://dx.doi.org/10.1016/j.jpowsour.2016.07.104 0378-7753/© 2016 Elsevier B.V. All rights reserved. Facing ever-increasing demands in energy consumption and concerns about environmental pollution, there is an urgent pursuit



of renewable energy and emerging technique for energy storage and transformation [1,2]. Among various energy storage and conversion equipments, electrocatalysis and supercapacitors (SCs) are considered to be the most effective technology that can help to settle the above problems. Electrochemical water splitting for oxygen and hydrogen $(2H_2O \rightarrow 2H_2+O_2)$ provides an attractive strategy to generate and store the hydrogen from renewable energy sources, thereby fulfilling the energy requirements [3]. In this process of water splitting, the hydrogen evolution reaction (HER) is a relatively easy course with require of two electron transfer steps that can readily happen at low overpotential [4]. However, for oxvgen evolution reaction (OER) $(40H^- \rightarrow 02 + 2H20 + 4e^-)$, it is more hardship to realize effective and stable gas evolution due to the kinetically slow anodic process with sluggish four electron transfer steps, which accompany with the breakage of O–H bond and the combination of attendant O–O bond [5]. Therefore, the process of OER is related to several procedures with high kinetic impedes resulting in the large overpotential, which severely hinders the efficiency of massive production of hydrogen from water splitting on account of the undesirable waste of energy [6]. Thus, compared to the thermodynamic potential, the electrolysis generally needs a higher potential. To date, the state-of-the-art electrocatalysts for OER are on the basis of precious metals like Ir or Ru, such as IrO₂ or RuO₂, but their inherent imperfection, like scarcity and high price, significantly imposes barrier to their practical applications [7]. Besides, it has been well recorded that the IrO₂ and RuO₂ catalysts present poor stability, especially in alkaline solution [8]. Bear this in mind, it is crucial to explore highly active and costeffective OER catalysts based on the strength of earth-abundant metals. Lately, great researches have been committed to the hydroxides/oxides of first-line transition metals as potential electrocatalysts [9–11]. In particular, the low-price CoFe-derived layered hydroxides and spinel oxides have been demonstrated the promising properties of OER in alkaline solutions, as reported in the literatures [12–14]. Supercapacitors (SCs) are another highefficiency energy storage device with big power density, excellent kinetics reversibility and long-term cycling stability. SCs could be divided into two types, such as pseudocapacitors and electrical double-layer capacitors (EDLCs), based on the different chargestorage mechanisms. Thereinto, pseudocapacitors have drawn more concerns, recently, compared to EDLCs, because pseudocapacitors own the higher energy and power density. Until now, diverse traditional materials, for example, carbonaceous materials [15] and polymers [16], have been found to suffer from the dissatisfied specific capacitance, mechanical decomposition and limited working-life, which seriously impede their practical application in the field of SCs. Transition metal oxides (TMOs) with multiple oxidation states for oxidation and reduction reaction are expected to be a potential candidate for SCs, because they endow the high specific capacitance, abundant reserve, low-price and environmental benignity. Especially, binary TMOs display many characteristics for SCs applications due to the co-existence of two metal ions in a compound, which is benefit to enhancing the capacitive activity and stabilization. Especially, CoFe₂O₄ with good chemical stability and high theoretical specific capacitance is deemed to be a promising electrode material in SCs. Thus, it can be concluded that the binary CoFe₂O₄ has great potential applications in both electro-catalysis and SCs. However, few researches reported the CoFe₂O₄ with uniform morphology as the electrode materials for electro-catalysis and SCs.

Unfortunately, one of the primary issues existed in binary spinel TMOs is the low electrical conductivity, which significantly limits the electron transport and consequently degrades the electrochemical performance [17]. Developing active materials on conductive substrates with high-surface-area is considered as an advisable strategy to improve the electro-conductivity. The materials attach on conductive substrates are not only convenient for electrochemical measurements, but also advantageous to avoid the extra contact resistances from the binder and reaction site obstruction from the additive, which have negative impact on the stability and electrochemical activity [18,19]. Various nano-carbon materials, for instance carbon nanotubes [20], carbon cloth [21] and graphene [22,23], have been introduced into binary spinel TMOs to improve the electronic conductivity. However, as for the 1D carbon nanotube and 2D graphene, they are confronted with the severely aggregation or stack each other caused by the intense van der Waals forces, which impede the electrolyte-access and gasrelease processes, bringing about the poor structural hierarchy. With this in mind, nickel foam may be an ideal substrate to deposit active materials, because it not only endows large surface area and excellent electro-conductivity, but also provides the firm framework to the loaded binary spinel TMOs without aggregation. In general, co-precipitation method is used to prepare the precursors of binary TMOs, in which all the raw materials are directly mixed together at the outset of the reaction. In consequence, the tremendous loss of concentration (supersaturation) in precursor solution during the reaction is unavoidable, which contributes to an inexplicable and undesirable controlled growth of the product. So the precursors synthesized by this method usually suffer from serious aggregation to form irregular particles with a broad size distribution. Therefore, most binary spinel oxides stemmed from such precursors are existed in the form of particle agglomerates and require additional binder-assisted film casting or coating procedures when evaluating their catalytic and capacitive properties [17,24,25], which decreases the specific surface area and leads to the poor conductivity. However, at high temperature and high pressure, the more energetic reaction conditions can improve the phase miscibility and solubility of precursor to form the compounds with complex structure and morphology that can't be realized in low-temperature and atmospheric pressure aqueous solution reactions [26]. Moreover, in hydrothermal environment, water closely reaches to the critical point, resulting in the decrease of the solution viscosity. Therefore, the diffusion of precursors becomes more readily, which is benefit to the growth of high quality crystals [27]. So, it can be seen that the morphology-engineering in nanoscale is an advisable measure to ameliorate the electrochemical performance of the materials [28].

In this work, a high temperature and pressure hydrothermal strategy is used to synthesize the CoFe-precursor nanosheet arrays on Ni foam. Then, with the calcinations process in air, CoFe-precursor nanosheet arrays convert to porous $CoFe_2O_4$ nanomesh arrays. Most importantly, the $CoFe_2O_4$ nanomesh arrays, as the multifunctional materials, endow not only the superior OER properties, but also the prominent capacitive performances, because the numerous pores in $CoFe_2O_4$ nanomesh arrays bring about larger surface area and more active sites. Moreover, $CoFe_2O_4$ nanomeshes vertically grow on conductive substrate-nickel foam to form the array structure, which could prevent the aggregation. Therefore, when directly used as free-binder electrode for OER and SCs, $CoFe_2O_4$ nanomesh arrays on nickel foam could avoid the contact resistance stemmed from additive, enhancing the electron conductivity.

2. Experimental sections

2.1. Materials synthesis

Firstly, the preparatory work is adopted for subsequent synthesis. A slice of nickel foam with the size of $1 \times 2 \text{ cm}^2$ is immersed in 3 M HCl aqueous solution with an ultrasound bath for 10–15 min,

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