



Three-dimensional carbon- and binder-free nickel nanowire arrays as a high-performance and low-cost anode for direct hydrogen peroxide fuel cell



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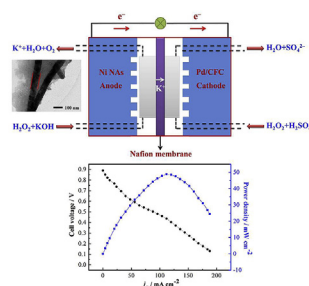
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HIGHLIGHTS

- The Ni NAs shows unique 3D open nanowire arrays structure with a large surface area.
- The Ni NAs exhibits superior catalytic activity and stability for H₂O₂ oxidation.
- The DPPFC with Ni NAs anode displays a peak power density of 48.7 mW cm⁻² at 20 °C.

GRAPHICAL ABSTRACT



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ABSTRACT

A novel three-dimensional carbon- and binder-free nickel nanowire arrays (Ni NAs) electrode is successfully fabricated by a facile galvanostatic electrodeposition method using polycarbonate membrane as the template. The Ni NAs electrode achieves a oxidation current density (divided by the electroactive surface areas of Ni) of 25.1 mA cm⁻² in 4 mol L⁻¹ KOH and 0.9 mol L⁻¹ H₂O₂ at 0.2 V (vs. Ag/AgCl) accompanied with a desirable stability, which is significantly higher than the catalytic activity of H₂O₂ electro-oxidation achieved previously with precious metals as catalysts. The impressive electrocatalytic performance is largely attributed to the superior 3D open structure and high electronic conductivity, which ensures the high utilization of Ni surfaces and makes the electrode have higher electrochemical activity. The apparent activation energy of H₂O₂ electro-oxidation on the Ni NAs catalyst is 13.59 kJ mol⁻¹. A direct peroxide–peroxide fuel cell using the Ni NAs as anode exhibits a peak power density of 48.7 mW cm⁻² at 20 °C. The electrode displays a great promise as the anode of direct peroxide–peroxide fuel cell due to its low cost, high activity and stability.

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1. Introduction

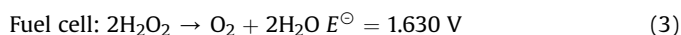
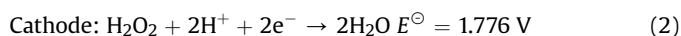
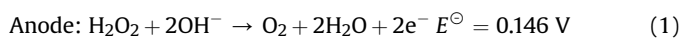
Hydrogen peroxide (H₂O₂) is a carbon-free energy carrier and a strong oxidant, and can be used as both the fuel and oxidant in a direct peroxide–peroxide fuel cell (DPPFC). In DPPFC, H₂O₂ is

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electro-oxidized at the anode in alkaline solution (Eq. (1)) and electro-reduced at the cathode in acid medium (Eq. (2)) to generate electricity and release O₂ and H₂O (Eq. (3)) [1–6]. Comparing with other types of liquid-based fuel cells (e.g. the direct methanol fuel cell, direct formic acid fuel cell and direct borohydride fuel cell) [7–12], the DPPFC has several advantages such as (a) the electrochemical oxidation and reduction of H₂O₂ involve no intermediates or by-products that can cause catalysts poisoning, (b) the products from electro-oxidation and electro-reduction of H₂O₂ are only O₂ and H₂O, which are environmentally friendly, (c) DPPFC generates power and provides O₂ at the same time, so it is a good candidate for underwater or space power sources operating without air [5,6], (d) the electro-oxidation of H₂O₂ is a simple two-electron transfer process and has lower activation energy than the electro-oxidation of methanol (six-electron transfer process) [5–8] and the electro-oxidation of BH₄⁻ (eight-electron transfer process) [10–12], (e) H₂O₂ is much cheaper and less toxic than other fuels, like methanol, formic acid, borohydride, and hydrazine [4].



S. Yamazaki et al. [1] reported a membraneless one-compartment DPPFC using H₂O₂ both as the fuel and oxidant. The fuel cell generated a maximum current density of 2.9 mA cm⁻² and achieved an open circuit voltage (OCV) of 0.13 V. Afterwards, S.A.M. Shaegh et al. [2] markedly improved the OCV of one-compartment DPPFC to 0.6 V and the power density to 1.55 mW cm⁻² using Ni anode and Prussian blue cathode in an acidic medium. S. Hasegawa et al. [3] firstly established a two-compartment DPPFC and demonstrated the OCV and output power density of two-compartment DPPFC remarkably higher than that of one-compartment configuration. Later on, A.E. Sanli et al. [4] developed a two-compartment DPPFC with the conventional fuel cell configuration. By using Ni as the anodic catalyst with basic H₂O₂ as fuel, Pt as the cathodic catalyst with acidic H₂O₂ as oxidant, they achieved a peak power density of 3.8 mW cm⁻² and an OCV of ~0.9 V. Recently, our groups [5,6,13] presented a high-performance two-compartment DPPFC using carbon fiber cloth supported metallic catalysts (Ni, Co, Pd and Au) with special structure as the anode and the Pd/CFC as the cathode. The DPPFC exhibited a stable OCV of 0.9 V and a peak power density of 21.6 mW cm⁻² at 20 °C. Thus, the DPPFC performance needs to significantly improve in order to compete with other types of fuel cells.

One of the decisive factors for DPPFC to achieve the high performance is improving the anode properties. The anode catalyst material has a great impact on the electrochemical reaction rate of H₂O₂ electro-oxidation. Precious metals are active for H₂O₂ electro-oxidation [5,6,13], but the high cost of these materials largely limits their extensive utilization. Above all, noble metals are unnecessary to be employed in alkaline electrolyte because alkaline medium is less corrosive than acid. At present, an ideal low-cost transition metal nickel has been mostly investigated as the anode catalyst to reduce the price of DPPFC and displayed good stability in alkaline medium [2,4,13]. However, the planar microstructure of the nickel tremendously reduces the utilization of catalyst surfaces, resulting in the low electrochemical activity of the electrode and low performance of the DPPFC [2,4]. In general, the most used electrodes for fuel cells are carbon-supported and binder-enriched ones produced by the traditional slurry-coating technique. The electrodes are usually fabricated by binding the powder electrocatalysts onto carbon paper current collector with the usage of organic polymer

binders [14–19]. However, many researchers found that the corrosion of carbon support under fuel cell operating conditions can lead to aggregation of catalyst particles, thereby reducing the electrochemical active surface area and resulting in poor catalytic performance [20–24]. They also reported that the electrochemical kinetic of carbon corrosion is slow, so severe carbon corrosion is not easily observed under normal fuel cell operating conditions, but the phenomenon of carbon corrosion is generally existed [20–24]. As we know, H₂O₂ is a strong oxidant, so the carbon can be slowly oxidized in the harsh H₂O₂ and KOH solution, which will lead to the deactivation of electrodes. Furthermore, the binders also can be gradually degraded in the H₂O₂ and KOH solution, and the existence of binders will greatly decrease the electronic conductivity of the electrode materials and cause the low electrochemical performance [18,25]. Thus, the conventional electrodes of fuel cells are unsuitable for H₂O₂ electro-oxidation in alkaline solution. Furthermore, because O₂ was continuously generated at the anode of DPPFC, the anode should have a three-dimensional (3D) open structure to allow O₂ quickly diffusing away from the electrode surface to regenerate the catalytic active sites. Therefore, a facile and reasonable design of the open 3D structure without carbon and binder is imperative for high electrocatalytic activity towards H₂O₂ electro-oxidation in alkaline medium.

Nanowire arrays are one of the optimal candidates due to the superior 3D open structure and high specific surface area which can afford easy transport for both electrolyte ions and produced gas. Nowadays, nanowire arrays have caught much attention in the fields of high-density magnetic recording, sensors, supercapacitors and fuel cells [17,26–30]. In order to prepare nanowire arrays in a versatile and controllable way, anodic aluminum oxide (AAO) templates and track etched polycarbonate templates were introduced as the porous templates. Usually, both the templates had to be coated on one side with a conductive film (via either ion sputtering or thermal evaporation of metals) [31–34], which was costly and cumbersome. Some researchers utilized liquid mercury [35] as the conductive support or polished solid metal to a convex substrate [36] for better contact with the templates. However, these methods still had many disadvantages such as high fabrication cost, high toxicity, complicated and long technological process.

In this work, the all-metal nickel nanowire arrays (Ni NAs) electrode without carbon and binder was synthesized by one-step galvanostatic electrodeposition assisting by a facile polycarbonate template with low cost. The fully metallic electrodes supported by themselves owned unique 3D open nanostructure, which provided more catalytic active sites and enabled easy access of reactants to the catalyst surfaces. The catalytic performance of the Ni NAs electrode for H₂O₂ electro-oxidation in alkaline medium was systematically investigated. The Ni NAs materials exhibited excellent catalytic activity, desirable stability and tolerance towards H₂O₂ electro-oxidation, indicating the great potential for future applications in DPPFCs.

2. Experimental

2.1. Reagents

Nickel sulfate (Ni₂SO₄), nickel chloride (NiCl₂), boric acid (H₃BO₃), saccharin (C₇H₅O₃NS), sodium lauryl sulfate (C₁₂H₂₅SO₄Na), potassium hydroxide (KOH), hydrogen peroxide (H₂O₂), dichloromethane (CH₂Cl₂), acetone (CH₃COCH₃) and ethanol (C₂H₅OH) were obtained from Enterprise Group Chemicals Reagent Co. Ltd. China. Polycarbonate membrane (50 nm pore diameter and 6 μm thickness) coated with PVP (polyvinylpyrrolidone) as wetting agent was purchased from Whatman Ltd. Electrolytic nickel sheets were provided by HuaLiang industrial

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