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Enhanced performance of direct peroxide/peroxide fuel cell by using ultrafine Nickel Ferric Ferrocyanide nanoparticles as the cathode catalyst

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ABSTRACT

Direct peroxide-peroxide fuel cell (DPPFC) employing with H_2O_2 both as the fuel and oxidant is an attractive fuel cell due to its no intermediates, easy handling, low toxicity and expense. However, the major gap of DPPFC is the cathode performance as a result of the slow reaction kinetics of H_2O_2 electro-reduction and thus the target issue is to design cathode catalysts with high performance and low cost. Herein, different with using noble metal of state-of-the-art, we have successfully synthesized ultra-fine Ni–Fe ferrocyanide (NiFeHCF) nanoparticles (the mean particles size is 2.5 nm) through a co-precipitation method, which is used as the cathode catalyst towards H_2O_2 reduction in acidic medium. The current density of H_2O_2 reduction on the resultant NiFeHCF electrode after the 1800 s test period at -0.1 , 0 and 0.1 V are 121, 93 and 76 $mA\ cm^{-2}$, respectively. Meanwhile, a single two-compartment DPPFC cell with NiFeHCF nanoparticles as the cathode and Ni/Ni foam as the anode is assembled and displayed a stable OCP of 1.09 V and a peak power density of 36 $mW\ cm^{-2}$ at 20 °C, which is much higher than that of a DPPFC employed with Pd nano-catalyst as cathode.

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Introduction

Fuel cells, in which direct converting chemical fuel into electric energy with high efficiency, have been regarded as preferable energy converter system in the application of handy power supply. In that of the abundant amount in air, oxygen (O_2) is the most used oxidant in fuel cells. However, take a

consideration of some air-free conditions (such as underwater or outer space), hydrogen peroxide (H_2O_2) has been investigated as the oxidizer to replaced oxygen depending on its fast reduction kinetics, convenient storage and manufacture [1]. Recently, a new type of fuel cell named as direct peroxide-peroxide fuel cell (shorten as DPPFC) is assembled by employing H_2O_2 both as the fuel and oxidant based on the

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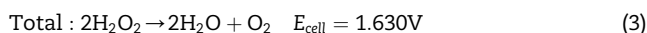
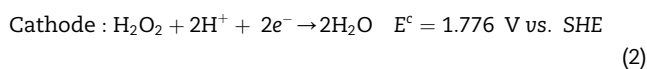
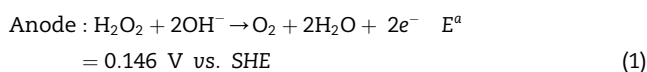
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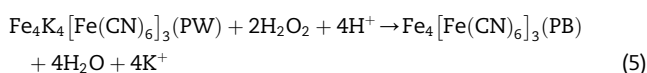
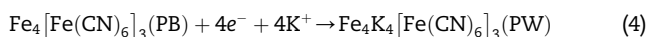
different potentials between the oxidation (Eq. (1)) and reduction of H_2O_2 in different pH medium (Eq. (2)) [2].



Comparing with other types fuel cells, for example, DMFC (M denote as methanol) [3–5], DFAFC (FA: formic acid) [6–8] and DBFC (B is referring to borohydride) [9–11], DPPFC has the advantages of no intermediates or by-products, therefore there would not have the issue of catalysts poisoning, faster oxidation kinetics, low toxic and cost [12–16]. Besides, the theoretical cell voltage of 1.630 V for DPPFC is higher than that of 1.23 V for a PEMFC and that of 1.21 V for DMFC. Furthermore, O_2 will produce during the DPPFC working process, making it to be an excellent candidate power sources for underwater and/or space application.

Compared with the one-compartment DPPFC without separating membrane between the anode and cathode [14], a two-compartment DPPFC reported by Sanli [13] of basic H_2O_2 as fuel and acidic H_2O_2 as oxidant, has a higher open-circuit voltage (OCV, 0.9 V) and power density (3.8 mW cm^{-2}). However, the obtained power density is far from the practical application that needs to be greatly improved. Based on our recent research [17], we found that H_2O_2 electrooxidation has faster reaction kinetics than that of H_2O_2 electroreduction. Therefore, the shortage of DPPFC is the cathode performance and an effective way to overcome this drawback is designing cathode catalysts with high performance towards H_2O_2 electroreduction in acid medium. Variety of materials have been investigated as H_2O_2 electro-catalyst, including metals [18–22], transition metal oxides [23–25], macrocycle complexes of transition metals [26,27], and enzymes [28,29]. Among them, noble metal catalyst showed a better integral property with high catalytic performance and superior stability, but the high prices restricted their extensive utilization. Hence, reducing the usage of precious metals and improving the comprehensive properties have been attracted much attention at present.

Prussian Blue (PB), also named ferric ferrocyanide ($\text{Fe}_4\text{III}[\text{FeII}(\text{CN})_6]_3$) and its analogs are a type of broadly discussed octahedrally coordinated compounds, which have been researched roomy for their noticeable magnetic, electrochromic and electrochemical properties, which have been widely used in H_2 adsorptions [30,31], charge storage devices [32], ion selective electrodes and electrochemical sensors [33]. In particular, they also represent as electro-catalyst for H_2O_2 reduction in acid medium [34–37] due to their redox transformations between with the reduced (Prussian White, PW) forms [38], which is described as following:



Compared with traditional precious metal nanoparticles, the positive potential of working electrode based on PB obviously reduced (approximate 0 V), preventing the conflict with minimizing species (e.g. ascorbic acid), and this feature is also widely used as biosensor for H_2O_2 detection [39–41]. According to previous reports, the critical influence on electrocatalytic properties for H_2O_2 reduction is the size and shape of PB materials [35,42]. However, the solubility constant of PB is as low as 3.3×10^{-41} , which makes the reaction is too fast and hard to control the morphology and the particle size of product.

In this work, we proved a plain and effective method to manufacture Nickel Ferric Ferrocyanide (NiFeHCF) nanoparticles with ultra-fine diameter through a co-precipitation way under mild conditions. The catalytic activity of NiFeHCF nanoparticles towards H_2O_2 electro-reduction has been systematically investigated in H_2SO_4 solution and an integral property in terms of high catalytic performance and superior stability are obtained. The performance of a single DPPFC assembled with the as-prepared NiFeHCF nanoparticles as the cathode is inspected and an expressively outstanding performance is achieved, higher than the preceding literature (eg. Pd/CFC).

Experimental

Preparation of the NiFeHCF ultrafine nanoparticles

The ultrafine NiFeHCF nanoparticles were synthesized by a co-precipitation method. Briefly, 30 mL NiCl_2 (100 mmol L^{-1}) and 30 mL FeCl_3 (100 mmol L^{-1}) were simultaneously pumped into 120 mL $\text{K}_3[\text{Fe}(\text{CN})_6]$ (25 mmol L^{-1}) with a flow rate of 2 mL min^{-1} . During this process, the solution was kept at 80°C with constant stirring. After a further 1.5 h stirring, the obtained solid precipitate was centrifuged, washed, and dried at 80°C . In order to investigate the effect of Ni and Fe atom ratio on the electrochemical performance, we have synthesized NiFeHCF with different Ni and Fe ratio by adjusting the volume ratio of NiCl_2 and FeCl_3 . The samples were designated as NiFeHCF ($n_{\text{Fe}}/n_{\text{Ni}}$, the mole ratio of FeCl_3 and NiCl_2 adding) for distinguish.

Characterization

The morphology of NiFeHCF nanoparticles was characterized by a SEM (JEOL JSM-6480) and TEM (FEI 2010). XRD (Rigaku TTR III) with a Cu $K\alpha$ radiation ($\lambda = 0.1514 \text{ nm}$) was employed to verify the crystalline phases of the resultant materials. In order to check the Ni and Fe mole ratio, the as-prepared NiFeHCF sample was checked with ICP test (Xseries II, Thermo Scientific). N_2 adsorption was performed on an ASAP 2460 (Micromeritics, USA) at 77 K to measure the specific surface area of the as-prepared NiFeHCF sample.

Electrochemical measurements

To prepare an electrode, the as-prepared NiFeHCF nanoparticles were first mixing with acetylene black and polyvinylidene difluoride (PVDF) according to a mass ratio of

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