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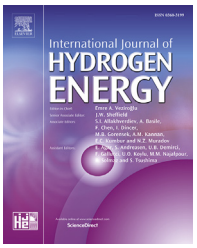

Peony petal-like 3D graphene-nickel oxide nanocomposite decorated nickel foam as high-performance electrocatalyst for direct glucose alkaline fuel cell

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ABSTRACT

Nickel foam modified by various electron mediators is an ideal non-noble metal anode for direct glucose alkaline fuel cell. However, high cost, low stability, and toxicity of electron mediators largely hamper their practical application. Herein, we demonstrated a one-step electrodeposition method to produce high-performance nickel foam electrode decorated with reduced graphene oxides and nickel oxides. The structure and morphology of the resulting 3D graphene-nickel oxide nanocomposites were characterized by UV–Vis, Raman spectroscopy, X-ray photoelectron spectroscopy, and scanning electron microscopy. Furthermore, a composite anode was prepared by rolling an activated carbon layer on the fabricated nickel foam electrode. At ambient temperature, the fuel cell equipped with the composite anode exhibited excellent performance with a high peak power density of 13.48 W m^{-2} under the condition of 1 M glucose, 3 M KOH, which was 39.30% higher than that of the bare activated carbon anode cell. In particular, electrochemical measurements demonstrated the high performance of the nanocomposite modified activated carbon anode was likely attributed to the synergistic effect of high conductivity of graphene and the catalytic activity of trivalent nickel towards glucose oxidation.

Introduction

Fuel cells have been widely concerned in recent years because they represent a highly efficient and environmentally friendly alternative technology for energy production [1,2]. Glucose, as the most abundant and important simple sugar in nature, has great potential for being used as high-density hydrogen carrier and sustainable energy source. It is cheap, easily available, non-explosive, nontoxic, and non-volatile [3,4]. Furthermore, glucose can yield 2870 kJ mol^{-1} energy under the condition of complete oxidation to CO_2 via 24-electron transfer [5,6]. The

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theoretical energy density of glucose is 4430 Wh kg^{-1}. This value is comparable to that of the most common substrate for direct fuel cell like methanol (6100 Wh kg^{-1}) [7,8]. Considering its advantages, glucose is more suitable for various portable applications, such as cell phones, power computers, and other portable low-power devices [1]. However, in these fields, glucose fuel cell is still at the prior stage compared with methanol and ethanol fed fuel cells [1,9–15].

Based on the catalyst material form, glucose fuel cells can be primarily classified into three kinds: enzymatic fuel cell, microbial fuel cell, and abiotic fuel cell [1,16]. The design of enzymatic glucose fuel cell is initially simple because the catalysts are selective, though enzymes are easy to be progressively deactivated for long-term application [17–19]. Microorganisms are more resistant to poisoning and loss of activity [1], but the most critical problems of microbial glucose fuel cell are their low performance and potential health risk [8,20]. The abiotic glucose fuel cells, by contrast, have long-term stability, affording them considerable durability. However, their prominent barriers are high cost, catalyst poisoning, and slow reaction kinetics of glucose oxidation [1,8]. Researchers have studied a variety of alloy catalysts with long lifetime and operational stability, Basu et al. [21] used $NaBH_4$ reduction technique to add Pd and Au with Pt precursor to form Pt–Pd–Au/C (metal ratio 1:1:1) catalyst that can efficiently electro-oxidize glucose in direct glucose fuel cell. In addition, there are many other alloy catalysts, such as: Pt–Au/C [22], Pt/GNS [23], Pd–Au/C [24], Pd/cMWCNT [25]. In general, Ni and other transition metals (e.g. Co_3O_4 [26], ZnO–Al_2O_3 [27], $NiCoO_2$@CNT [28], CuO [29] and Mn_3O_4 [30]) are capable of catalyzing glucose oxidation in alkaline solutions even better than Pt [1]. Iwu et al. [31] fabricated a novel Ni nanofoam electrode applied for non-enzymatic glucose sensing by chemical bath deposition and thermal annealing. Kung et al. [32] covered a single layer of nickel hydroxide nanoparticles on the surface of a nickel foam, and found that the redox couple of $Ni(OH)_2$/NiOOH formed on the electrode surface greatly enhanced the oxidation of glucose. Eshghi et al. [33] synthesized Nickel–Iron Double Hydroxide nanocomposites on graphene/glassy carbon electrodes by electrochemical method, and illustrated that graphene/NiFe LDH exhibits a high diffusion coefficient (1.80×10^{-4} cm^2 s^{-1}) as an electro catalyst for glucose electro oxidation. Although all these work have promoted the oxidation of glucose remarkably, the slow reaction kinetics still impedes the widespread use of glucose fuel cells [34].

Graphene is a one-atom-thick layer of carbon atoms bonded by sp^2 bonds. This configuration provides this material with unusual properties, such as: large accessible surface area, excellent thermal and electrical conductivity [35]. Owing to these extraordinarily properties, graphene is an attractive carbon material to carry various catalysts [36]. Jafri et al. [37] prepared Pt loaded nitrogen doped graphene as electrocatalyst for proton exchange membrane fuel cell that showed excellent methanol oxidation activity. Chi et al. [38] fabricated a Pd–Pt/rGO/NFP composite through one-step mild reduction process. By changing the proportion of catalyst components, the authors found that the anode with the $Pd_1Pt_{0.98}$/rGO/NFP catalyst exhibited a strong activity for glucose oxidation. Yang et al. [34] reported the fabrication of the porous Co_3O_4@graphene microspheres for enzyme-free biosensor applications. Wang et al. prepared a three-dimensional (3D) reduced graphene oxide-nickel foam as an anode for microbial fuel cell (MFC) [30]. Yang et al. [39] described a simple, cheap, and green method to produce porous graphene/nickel foam electrodes and its application in supercapacitors. Zhao et al. [40] developed a 3D graphene aerogel decorated with Pt nanoparticles as an efficient anode for MFC. K. Hoshi et al. [41] used a graphene-coated carbon fiber cloth electrode to absorb more enzymes on its surface. Although graphene is extensively explored as an electrode material for supercapacitors, sensors [42] and MFCs, there are few reports on its application in direct glucose alkaline fuel cells (DGAFCs).

Electron mediators are normally used to facilitate the electron transfer and enhance the fuel cell performance [13]. In prior work, we reported the preparation of various high-performance activated carbon anodes modified by with different electron mediators, such as methyl viologen (MV) and 2-hydroxy-1, 4-naphthoquinone (NQ) [5,13,16]. However, most electron mediators are high-cost and have adverse impact to the environment [5]. The development of high-performance anode electrocatalyst for glucose oxidation is still in urgent need to avoid the use of additional electron mediators.

In this work, a highly active 3D graphene-nickel oxide nanocomposite modified nickel foam electrode was prepared by a simple potentiostatic method. To the best of our knowledge, this is the first report of one-step electrodeposition of 3D graphene-nickel oxide nanocomposite on nickel foam electrode and application in DGAFCs. UV–Vis, Raman, XPS and SEM were used to study the electrochemical reduction of graphite oxide and the morphology of the electrode. Furthermore, various electrochemical techniques, such as linear sweep voltammetry (LSV), electrochemical impedance spectroscopy (EIS), and Tafel tests, were employed to get further insight into the possible action mechanism. Finally, the performance of DGAFC equipped with the prepared anode was examined.

Methods

Materials

Graphite powder (purity: 99.95%, mesh: 325) was purchased from Jinrilai Graphite Co. Ltd (Qingdao, China). 2-hydroxy-1,4-naphthoquinone (NQ) was purchased from J&K Scientific Co. Ltd (Beijing, China). 60 wt% PTFE solution was provided by Heshen Inc. (Shanghai, China). Activated Carbon (AC) powder (YEC-8A) was obtained from Yihuan Carbon Co. Ltd (Fuzhou, China). Nickel foam was purchased from Liyuan New Material Co. Ltd (Changde, China) (purity: 99.9%, number of pores per inch: 110, density: 380 g m^{-2} ± 20 g m^{-2}, average pore size: 590 mm, thickness: 1.7 mm). Glucose, $KMnO_4$, $NaNO_3$, Na_2SO_4, NiSO4, HCl, H_2SO4, H_2O_2 (30%), and KOH were all of analytical grade. Deionized water (Millipore, Milli Q, 18.3 MU) was used to prepare all the solutions.

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