



Preparation of nickel nanowire arrays electrode for urea electro-oxidation in alkaline medium



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HIGHLIGHTS

- Fully metallic nickel NWAs electrode was templated electroformed.
- Nickel NWAs electrode showed high catalytic activity for urea electro-oxidation.
- Both EC mechanism and direct electro-oxidation of urea were proposed.

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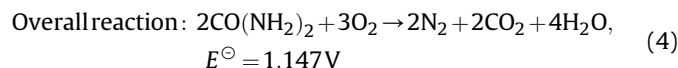
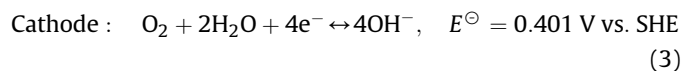
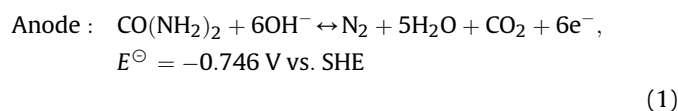
ABSTRACT

Fully metallic nickel nanowire arrays (NWAs) electrode is prepared by electrodepositing nickel within the pores and over-plating on the surface of polycarbonate template (PCT) with subsequent dissolution of the template in dichloromethane. The as-prepared electrode is characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Urea electro-oxidation reaction in KOH solution on the nickel NWAs electrode is investigated by cyclic voltammetry (CV), chronoamperometry (CA) and electrochemical impedance spectroscopy (EIS) tests. The results show that the nickel NWAs electrode achieves an onset oxidation potential of 0.25 V (vs. Ag/AgCl) and a peak current density of 160 mA cm⁻² in 5 mol L⁻¹ KOH and 0.33 mol L⁻¹ urea accompanied with considerable stability.

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1. Introduction

Urea (CO(NH₂)₂) presents in different concentrations in waste water discharged by fertilizer plants and human or animal urine. Many researches based on the electro-oxidation of urea were conducted for the purposes of urea-rich sewage disposal and hydrogen production [1,2]. Tao et al. [3] have successfully developed a working direct urea and urine fuel cell for the first time in 2010. Compared with retrieving energy from hydrogen which is produced by the electrolysis of urea, generating electricity from direct urea fuel cell (DUFC) is more efficient. DUFC is composed of urea in alkaline electrolyte as the fuel and humidified oxygen as the oxidant. The theoretical open circuit voltage of DUFC is 1.147 V, which is comparable with that of H₂–O₂ fuel cell (1.23 V). The electrode and solution reactions of DUFC are as follows:



Urea is a promising alternative anode fuel due to the reasons that: (1) urea is low-cost and derived from a wealth of sources, including industrial production and waste water; (2) urea is solid state and easy in storing and transportation; (3) the electro-

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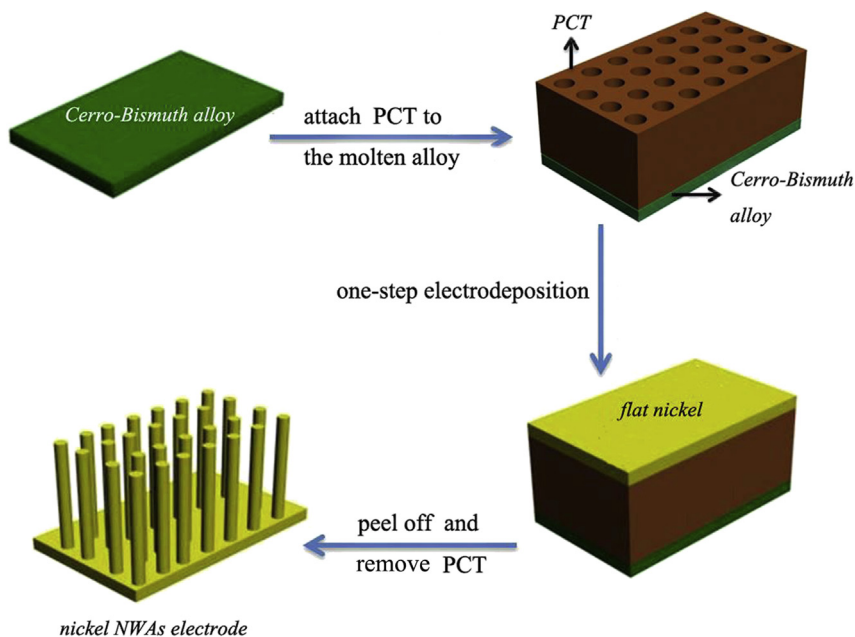


Fig. 1. Schematic of templated electrodeposition of nickel NWAs electrode.

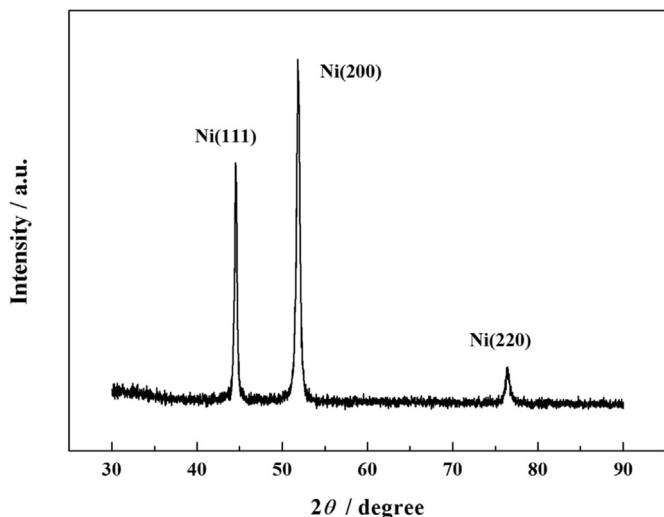


Fig. 2. XRD pattern of nickel NWAs electrode.

oxidation of urea not only generates electricity but also purifies wastewater; (4) the products of urea electro-oxidation are benign nitrogen and carbonate (Eqs. (1) and (2)).

In order to search for highly efficient electrocatalysts for urea electro-oxidation, several kinds of materials including noble metals, non-noble metals, metal oxides and metal hydroxides have been studied. Although noble metals (Pt, Ir, Pd, Rh) exhibited excellent electrocatalytic performance for the electro-oxidation of alcohol and hydrogen peroxide [4–8], they showed poor activity towards urea [1]. More importantly, noble metals are not necessary to be utilized in alkaline electrolyte because alkaline medium is less corrosive than acid. At present, much attention has been paid to non-noble transition metals. Among various candidates, Botte et al. [1,2,9–11] found nickel was the most favorable electrocatalyst for the electro-oxidation of urea. However, nickel as the catalyst has a defect that the onset oxidation potential of urea reached up to

0.45 V vs. SHE, which would cause inevitable polarization loss of DUF. They also observed that the onset potential of urea electro-oxidation was nearly the same as the potential where NiOOH was formed (0.49 V vs. SHE). This phenomenon demonstrated that urea was possibly electro-oxidized on NiOOH. In an attempt to reduce the large overpotential (1.196 V), Yan et al. [12] synthesized Nickel–Cobalt bimetallic hydroxide electrocatalysts with the method of electrodeposition. The cobalt content greatly affected the morphology of the electrode as well as the onset oxidation potential of urea electro-oxidation. At the optimum amount of cobalt, the onset oxidation potential moved negatively from 0.4 V (vs. Hg/HgO) to 0.25 V (vs. Hg/HgO). Lan et al. [13] prepared nano-sized nickel (~5 nm) as the catalyst for urea electro-oxidation. DUF with nano-sized nickel as anode showed higher open circuit voltage of 0.65 V and a maximum current density of 1.6 mA cm^{-2} , while that with commercial nickel (4–10 μm) only achieved 0.22 V and 0.34 mA cm^{-2} when 1 mol L^{-1} urea was used as anode fuel at 20 °C. The results displayed that the structure and composition of catalysts were rather influential on the performance of urea electro-oxidation.

Nanowire arrays (NWAs) nowadays have triggered much attention because of potentials in high-density magnetic recording, sensors and fuel cells [14–16]. To fabricate nanowire arrays in a controllable and versatile way, anodic aluminum oxide (AAO) template and track etched polycarbonate template (PCT) are introduced as porous templates. Both of the templates usually have to be coated on one side with a conductive film (via either ion sputtering or thermal evaporation of metals) [17], which is costly and cumbersome. Others have utilized liquid mercury [18] as the conductive support or polished solid metal to a convex substrate [19] for better contact with the templates. However, those methods still have many disadvantages in mass production.

In this work, we prepared nickel NWAs electrode with the aid of PCT by one-step galvanostatic electrodeposition. The fully metallic electrode has a unique three-dimensional nanostructure, which provides more catalytic active sites and enables easy access of reactants to the catalyst surfaces. The electrocatalytic performance of the nickel NWAs electrode for urea electro-oxidation in alkaline

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