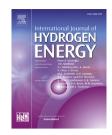
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international journal of hydrogen energy XXX (2017) 1–8 $\,$



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Short Communication

An aluminum/cobalt/iron/nickel alloy as a precatalyst for water oxidation

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ARTICLE INFO

Article history: Received 22 October 2017 Received in revised form 23 November 2017 Accepted 4 December 2017 Available online xxx

ABSTRACT

Among different strategies, water splitting toward hydrogen production is a promising process to store energy from intermittent sources. However, the anodic water oxidation is a bottleneck for water splitting. In this paper, we report an aluminum/cobalt/iron/nickel alloy as a precatalyst for the electrochemical water oxidation. The alloy electrode contains different metal ions including cobalt, iron, and nickel which all are efficient for water oxidation is tested. We characterized this electrode using scanning electron microscopy, transmission electron microscopy, diffuse reflectance infrared Fourier transform

Please cite this article in press as: Najafpour MM, et al., An aluminum/cobalt/iron/nickel alloy as a precatalyst for water oxidation, International Journal of Hydrogen Energy (2017), https://doi.org/10.1016/j.ijhydene.2017.12.025

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Keywords: Alloy Artificial photosynthesis Hydrogen production Water Water-oxidizing catalyst Water oxidation spectroscopy, Fourier transform infrared spectroscopy, X-ray diffraction, X-ray photoelectron spectroscopy and electrochemical methods. After stabilization, the electrode shows an onset overpotential of 200.0 mV and affords a current density of 3.5 mA cm⁻² at an overpotential of 600.0 mV in KOH solution at pH 13.

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Introduction

Water splitting conducted by different methods to form hydrogen is a promising approach to store sustainable energies [1-16]. Among various strategies, electrochemical water splitting has attracted more attention. In electrochemical water splitting, although the cathodic reaction is of major interest, the anodic water oxidation which proceeds through a complicated four-electrons mechanism at large overpotentials is considered as a bottleneck and has been largely investigated. The stability of the electrode is also an important factor that should be addressed in the electrochemical water oxidation [17-19]. Under the electrochemical condition, the current density (j) of more than 1 mA/cm² at a low overpotential is necessary for the water oxidation reaction [20-24]. To lower the overpotential needed for the electrochemical water oxidation, different compounds were used as a water-oxidizing catalyst in water-splitting systems. Manganese, iron, cobalt, nickel, chromium, and vanadium are among promising compounds for electrocatalytic water oxidation that have been extensively studied. As an interesting strategy, we used a mixed oxidebased catalyst containing cobalt, iron, and nickel ions for water oxidation.

Experimental section

Materials

All reagents and solvents were purchased from commercial sources. The alloy aluminum/cobalt/iron/nickel alloy (composition: aluminum: 7.5–8.7 wt%; cobalt: 23.5–25 wt%; copper: 2–3.5 wt%; iron: 45.3–52.85 wt%; nickel, 12.5–15.5 wt %; silicon: 1.5 wt%; titanium, 0.15–0.5 wt%) were purchased from Sigma–Aldrich and were used without further purification. The alloy was first treated by KOH (1.0 M, 24 h) (1), then treated by KOH (0.1 M, 24 h) under a potential of 2.0 V (2).

Methods

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were carried out with LEO 1430VP and FEI Tecnai G² F20 transmission electron microscope, TF20 (200 kV), respectively. The X-ray powder patterns were recorded with a Bruker D8 ADVANCE diffractometer (CuK_{α} radiation). Electrochemical experiments were performed using an EmStat³⁺ device from the PalmSens Company (Netherlands). The alloy was directly used as an electrode. A three-electrode system was applied for the investigation of electrochemical properties. The chronoamperometric data was used to calculate the Tafel plots, for which the applied potential was increased stepwise and held for 600 s at each step. The Hg|HgO, Pt foil and the alloy were used as the reference, counter, and working electrodes, respectively. In the paper, potentials were reported based on the Hg|HgO reference electrode. Oxygen evolution from aqueous solutions under amperometry at 25 °C was investigated using an HQ40d portable dissolved oxygen-meter connected to an oxygen monitor with a digital readout.

Results and discussion

The aluminum/cobalt/iron/nickel alloy is commercially available. First, the alloy was placed in a KOH solution (1.0 M, 24 h) (1), but a stabilization was not observed. In the next step, it was placed in KOH (0.1 M, 24 h) under potential (2.0 V) for a day to be stabilized (2). In this condition, dealloying occurred and the surface of alloy was converted to a nano-structure, although a solution of KOH (1.0 M, 24 h) in the absence of the potential also caused a few destructure of the surface.

Such dealloying has received great attention as some alloys generate porosity during dealloying, which is important in increasing catalytic activity or other properties [25,26].

SEM images indicated that the surface of the alloy is relatively smooth before the reaction (Fig. 1a, Figs. S1–S4, Electronic Supplementary Information) and no nanostructure was observed on the surface of it. However, the layers and nanoporosity were observed on the surface of 2 (Fig. 1b and c). Such waves and valley like structures could significantly increase catalytic sites on the surface of 2. The thickness of the layers is ≤ 100 nm, but the depth and width are ≥ 100 nm. Because all metals in the alloy are easily oxidizable under water oxidation, such destructuring are expected.

TEM and HRTEM images of mechanically separated layers of 2 in a few areas showed inter-planar distances of 2.9, 2, and 1.4 Å attributed to 30.6° , 44.7° , and 64.4° . However, in

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