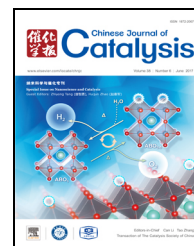


available at www.sciencedirect.comjournal homepage: www.elsevier.com/locate/chnjc

Article (Special Issue on Nanoscience and Catalysis)

Platinum stabilized by defective activated carbon with excellent oxygen reduction performance in alkaline media



Xuecheng Yan, Yi Jia *, Longzhou Zhang, Xiangdong Yao

School of Natural Sciences and Queensland Micro- and Nanotechnology Centre, Griffith University, Nathan Campus, QLD 4111, Australia

ARTICLE INFO

Article history:

Received 17 November 2016

Accepted 31 December 2016

Published 5 June 2017

Keywords:

Activated carbon

Defect

Platinum

Oxygen reduction reaction

Fuel cell

ABSTRACT

The exploration of highly active and durable cathodic oxygen reduction reaction (ORR) catalysts with economical production cost is still the bottleneck to realize the large-scale commercialization of fuel cells and metal-air batteries. Given that carbon support is crucial to the electrocatalysts, and Pt is the best-known ORR catalyst so far, in this work, we employed a simple impregnation method for synthesizing a kind of defective activated carbon (D-AC) supported low Pt content electrocatalysts for the ORR. The reduction conditions of the Pt-containing precursor were firstly optimized, and the influence of the Pt loading amount on the ORR was investigated as well. The results show that the obtained D-AC@5.0%Pt sample (contains 5 wt% Pt) has surpassed the commercial Pt/C with 20 wt% Pt for the ORR in an alkaline solution. In the meantime, it is more stable than the commercial Pt/C. The outstanding ORR performance of the D-AC@5.0%Pt confirms that both the unique defects in the D-AC and the introduced Pt particles are indispensable to the ORR. Particularly, the ORR activity of the synthesized catalysts is superior to most of the reported counterparts, but with much easier preparation methods and lower production cost, making them more advantageous in practical fuel cell applications.

© 2017, Dalian Institute of Chemical Physics, Chinese Academy of Sciences.

Published by Elsevier B.V. All rights reserved.

1. Introduction

Proton exchange membrane fuel cell is the most possible candidate to replace the current internal combustion engines in vehicles because of its high energy conversion efficiency and pollution-free characters. However, the sluggish cathodic oxygen reduction reaction (ORR) in fuel cells substantially reduces the overall cell performance, and the development of ORR catalysts with remarkable activity remains a big challenge [1]. Currently, platinum (Pt) and its alloys are still the main component of the ORR catalysts owing to their outstanding catalytic activity in both alkaline and acidic media [2–4], while the high cost of Pt and the stability issues greatly hinder its massive application [5,6]. To alleviate these shortcomings, considerable advances have been achieved on Pt alloys to reduce the utiliza-

tion of Pt, such as Pt-Fe [7–9], Pt-Ni [4,10,11], and Pt-Co [12–14]. Besides, it was found that the core-shell structures are also favorable to increase the activity and durability of Pt-based ORR catalysts [15–18]. However, the preparation of these materials normally involves the use of hazardous organic chemicals with tedious synthesis process, which is not suitable for practical large-scale production. Therefore, it is highly desirable to develop a facile method to fabricate efficient ORR catalysts with economical manufacturing cost.

Previous investigations illustrate that the support for Pt is crucial to enhance the ORR performance of the resulting catalysts [19–21]. It can be found that most of the supports are modified carbon materials, including micro/mesoporous carbons [22,23], heteroatom doped graphene [24,25], and defective carbons [21,26]. These carbon materials with tuned micro-

* Corresponding author. Tel: +61-7-37355057; Fax: +61-7-37358021; E-mail: y.jia@griffith.edu.au

structures and modified electronic environment are favorable for the electrocatalytic reactions. Recently, the defective carbons are being paid more attention owing to a new defect catalysis mechanism was proposed, demonstrating that specific types of defects in carbon materials not only could serve as the active sites for electrocatalytic reactions but also could provide more efficient anchor sites to couple metal atoms [26–29]. With respect to the pure defective carbon catalysis, albeit the activity of the defective activated carbon (D-AC) is superior to most of the reported metal-free ORR catalysts, it is still inferior to the commercial Pt/C (20 wt% Pt) [26]. In view of Pt is the best-known ORR catalyst so far, as aforementioned, if we can reduce the amount of Pt by using the D-AC as a carbon support, but without sacrificing the activity, it should be a promising approach to tackle the high cost problem of the cathodic catalyst and realize the mass production of fuel cells.

In the present work, a simple impregnation method was employed to prepare effective ORR electrocatalysts by using the D-AC as a support for the Pt. The Pt particle size manipulation was achieved by controlled thermolysis process, and the influence of the Pt concentration on the ORR activity of the resulting catalysts was also discussed. The results show that the obtained D-AC@5.0%Pt sample (only contains 5 wt% Pt) has outperformed the commercial Pt/C (20 wt% Pt) in an alkaline solution, indicating that the defective carbon not only could serve as an efficient ORR catalyst alone, but also an appropriate carbon support for the Pt to promote the ORR.

2. Experimental

2.1. Catalyst preparation

The defective carbon D-AC was synthesized through a facile nitrogen-doping and removal method, as described elsewhere [26]. Specifically, the D-AC with a certain mass was dispersed into the designed $\text{H}_2\text{Cl}_6\text{Pt}\cdot 6\text{H}_2\text{O}$ /ethanol solution with 5 min sonication for better mixing. Afterwards, the mixture was placed on a stirrer to stir overnight to remove the ethanol at room temperature. The obtained precursor was reduced in the presence of a mixture gas of 5% H_2 -95% Ar at the target temperatures in a tubular furnace. The gas was purged for 30 min to expel the air inside the tube before operating the reduction program.

2.2. Characterizations

The crystalline structures of the prepared samples were characterized by X-ray powder diffraction (XRD) in a Bruker Advance X-ray diffractometer using nickel-filtered $\text{Cu K}\alpha$ X-ray source radiation ($\lambda = 0.15405$ nm). The morphologies and structures of the materials were examined by using transmission electron microscopy (TEM) and high-resolution TEM (HRTEM) (Philips Tecnai F20). Chemical compositions of the prepared samples were acquired using a Kratos Axis ULTRA X-ray photoelectron spectrometer incorporating a 165 mm hemispherical electron energy analyzer. The incident radiation is Monochromatic $\text{Al K}\alpha$ X-rays (1486.6 eV) at 150 W (15 kV, 10

mA). The energy scale was calibrated to the C 1s peak maximum at 284.5 eV. The Pt contents of the prepared samples were determined by inductively coupled plasma mass spectroscopy (ICP-MS) on the Perkin Elmer Optima 8300 ICP-OES instrument.

2.3. Electrochemical measurements

The typical three-electrode system was employed to evaluate the electrochemical properties of the prepared catalysts. Specifically, glassy carbon (GC) was the working electrode, a Pt wire was the counter electrode and the Ag/AgCl (in saturated KCl solution) was the reference electrode. All potentials were referred to the reversible hydrogen electrode by adding a value of $(0.197 + 0.059\text{pH})$ V. Cyclic voltammetry (CV), linear sweep voltammetry (LSV) and rotating ring-disk electrode (RRDE) measurements were conducted on the CHI 760E workstation (CH Instruments, Inc.) with a RRDE-3A rotator (ALS Co., Ltd).

2.3.1. Sample preparation

1 mg of the catalyst was dispersed into 1 mL mixed solution of distilled water (680 μL), ethanol (300 μL) and Nafion® 117 Solution (5%, 20 μL). Then, 10 μL of the mixture was dropped onto a polished glassy carbon electrode (4 mm in diameter, catalyst loading: 0.08 mg/cm^2) after sonicating it for at least 60 min to form a homogeneous ink. The loaded electrode was placed in a 60 °C oven for 10 min and then was taken out to cool down before all the tests.

2.3.2. CV measurement

Prior to the test, the electrolyte (KOH solution (0.1 mol/L)) was bubbled with O_2 for at least 30 min to make it saturated with O_2 , and a constant oxygen flow was maintained during the measurement. The data was recorded at the scan rate of 100 mV/s when the system became stable.

2.3.3. LSV measurement

The rotating speed of the working electrode was increased from 400 to 2500 r/min at the scan rate of 10 mV/s in an O_2 -saturated KOH solution (0.1 mol/L).

2.3.4. RRDE measurement

The rotating speed of the working electrode was fixed at 1600 r/min with the scan rate of 10 mV/s in an O_2 -saturated KOH solution (0.1 mol/L) for the RRDE test. The electron transfer number (n) and the percentage of HO_2^- were calculated via the following equations [30,31].

$$n = 4I_d / (I_d + I_r / N) \quad (1)$$

$$\% \text{HO}_2^- = 200(I_r / N) / (I_d + I_r / N) \quad (2)$$

where I_d stands for the disk current, I_r represents the ring current, and N is the current collection efficiency of the Pt ring, which is identified to be 0.43 in $\text{K}_3\text{Fe}[\text{CN}]_6$ (2 mmol/L) and KCl solution (0.1 mol/L).

3. Results and discussion

3.1. Reduction conditions optimization

Download English Version:

<https://daneshyari.com/en/article/6505896>

Download Persian Version:

<https://daneshyari.com/article/6505896>

[Daneshyari.com](https://daneshyari.com)