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## Oxidized carbon/nano-SiC supported platinum nanoparticles as highly stable electrocatalyst for oxygen reduction reaction



Liang Dong<sup>a</sup>, Jianbing Zang<sup>a</sup>, Jing Su<sup>a</sup>, Yingdan Jia<sup>a</sup>, Yanhui Wang<sup>a,\*</sup>, Jing Lu<sup>b</sup>, Xipeng Xu<sup>b</sup>,

<sup>a</sup> State Key Laboratory of Metastable Materials Science and Technology, College of Materials Science and Engineering, Yanshan University, Qinhuangdao 066004, PR China

<sup>b</sup> MOE Engineering Research Center for Brittle Materials Machining, Huagiao University, Xiamen 361021, PR China

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#### ABSTRACT

Nano-SiC particles with derived carbon shells were prepared by an acid-etching method at room temperature. The mixture solutions of concentrated HF and HNO<sub>3</sub> were chosen to etch the nano-SiC particles, and an amorphous carbon shell absorbed by oxygen functional groups was formed on the SiC surface. The oxidized carbon/SiC (O-C/SiC) particles were used as supports for preparation of Pt electrocatalysts. The O-C/SiC supported Pt electrocatalysts showed a high catalytic activity and an excellent stability for oxygen reduction reaction. The improved stability can be ascribed to the anchoring effect of the carbon shell to Pt NPs and the high stability of nano-SiC core.

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#### Introduction

Proton exchange membrane fuel cells (PEMFCs) are considered as the most promising energy converting devices due to their environmentally friendly, high power density and low operating temperature. Platinum nanoparticles (Pt NPs) loaded on supports with high specific surface area and high conductivity are commonly used as the anode and cathode electrocatalysts for the PEMFCs [1-3]. A variety of materials have been used as electrocatalyst support, and the most common supports are carbon materials, such as carbon black, CNT and graphene. Unfortunately, the Pt electrocatalysts

supported on carbons (Pt/C) have a low stability, which is a main factor that hindered the widespread commercialization of PEMFCs [4,5]. One of reasons for the weak stability of Pt/C is corrosion and collapse of the carbon supports. The sp2 carbon supports are easily oxidized in the PEMFC working environment, particularly under the cathodic conditions including high oxygen content and high potential. The support corrosion will lead to Pt NPs shedding and agglomeration, ultimately resulting in electrocatalyst deactivation [6,7]. Thus, highly stable supports, for instance carbides, oxides and nitrides, caused wide interests from more and more investigators [8-11].

\* Corresponding authors. Tel./fax: +86 335 8387679.

E-mail addresses: diamond\_wangyanhui@163.com (Y. Wang), xpxu@hqu.edu.cn (X. Xu). http://dx.doi.org/10.1016/j.ijhydene.2014.07.161

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Silicon carbide (SiC) as a covalent crystal has excellent physical and chemical stabilities. Micron SiC was selected to be a support for fuel cell electrocatalyst by Honji et al. at early 1980s [12]. However, no further progress has been achieved for a long time due to its poor conductivity, weakly binding with Pt NPs and insufficient specific surface area. In recent years, this work has been gradually expanded and some breakthroughs have been made. Rao et al. prepared nano-SiC with huge specific surface area by plasma techniques, and used it as support for Pt NPs (Pt/nano-SiC). The Pt/nano-SiC exhibited good catalytic activity for oxygen reduction reaction (ORR) comparable with commercial Pt/C [13]. In order to improve electrode conductivity, carbon black powders were added into nano-SiC supported Pt electrocatalyst, and the composite electrode exhibited a high stability [14]. In our previous work, a graphitized SiC with a core-shell structure (SiC@C) was prepared through vacuum heat treatment. The SiC@C has an improved conductivity and affinity with Pt NPs in comparison to SiC, so the Pt electrocatalysts supported on SiC@C (Pt/ SiC@C) showed good catalytic activity and high stability [15].

In order to further increase the interaction between supports and Pt NPs, we prepared an oxidized carbon nanoshell covered nano-SiC using acid-etching at room temperature. Silicon atoms were selectively etched away by the mixture solutions of concentrated hydrofluoric acid (HF) and nitric acid (HNO<sub>3</sub>), and then an amorphous carbon nanoshell absorbed by oxygen functional groups was formed on the nano-SiC (O-C/SiC). The surface oxygen functional groups would provide more nucleation sites for Pt NPs, which form a strong interaction between supports and Pt NPs [16–18]. The catalytic activity of O-C/SiC supported Pt electrocatalysts (Pt/O-C/SiC) for oxygen reduction reaction (ORR) was studied, and the anchoring effect of the O-C/SiC for Pt NPs was investigated through accelerated durability test (ADT).

#### Material and methods

#### Preparation of O-C/SiC

Nano-SiC powder with an average size of 60 nm was bought from Anhui Kaier Nano-Power Technology Inc. Firstly, 200 mg nano-SiC powders were mixed with the acids of concentrated HF and  $HNO_3$  (a volume ratio of 1:2) in a plastic beaker. The beaker was placed inside a fume hood, and the reaction was kept for three days under ambient temperature, stiring the solution for 30 min every day. Then, the solution was washed to neutral with deionized water and the powders were filtered from the solution and dried at 80 °C, and the O-C/SiC powders were finally obtained. The morphologies of the O-C/SiC were observed by a Hitachi H2120 transmission electron microscope (TEM). Raman spectra were measured to analyze the structure of O-C/SiC using a Renishaw inVia Raman microscope. Fourier-transform infrared (FTIR) spectra of the O-C/ SiC were recorded on an EQ55 + FRA 106 FTIR instrument in the region of  $4000-400 \text{ cm}^{-1}$ .



Fig. 1 – TEM images of nano-SiC (a), O-C/SiC (b), and HRTEM images of nano-SiC (c) and O-C/SiC (d).

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