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## Development of stable electrochemical catalysts using ordered mesoporous carbon/silicon carbide nanocomposites

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### ARTICLE INFO

#### Article history:

Received 15 October 2014

Received in revised form

19 June 2015

Accepted 8 July 2015

Available online xxx

#### Keywords:

Ordered mesoporous carbon

Silicon carbide

Carbon corrosion

Nanocomposite

Electrochemical performance

### ABSTRACT

A nano-composite of ordered mesoporous carbon (OMC) and silicon carbide (SiC) was investigated as a durable support for Pt nanoparticles, in order to improve the electrochemical activity and stability in oxygen reduction reaction (ORR). The OMC and OMC–SiC were synthesized via a nano-replication method, using ordered mesoporous silica as a template and 1,10-phenanthroline as a carbon source at temperatures of 900 and 1350, respectively. Non-porous SiC material was obtained by heat treatment at 1600 °C. The OMC–SiC composite, containing 10.3 wt% of SiC, exhibited a high surface area (568 m<sup>2</sup>/g) and well-defined mesopores (2.7 nm). Highly dispersed Pt nanoparticles were supported on both the OMC and the OMC–SiC, using a polyol method. The ORR activity and the electrochemical surface area (ECSA) of Pt/OMC, Pt/OMC–SiC and commercial Pt/C catalysts were measured using a rotating disk electrode technique with the linear sweep method, and a potential-cycling test, respectively. The Pt/OMC–SiC composite showed the highest activity as well as the highest durability for ECSA and ORR, which may be attributed to the effect of the intimate hybridization of SiC with the OMC in nanoscale. These results indicate that the OMC–SiC composite is a very promising support material for electrochemical catalysts in fuel cells.

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### Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) have been regarded as alternative systems with many potential

applications in transportation, power generation and portable electronic devices, due to the high energy-conversion efficiency, low operating temperatures and environmental benefits [1,2]. Despite considerable advances over the decades [1], insufficient durability of catalysts among many major

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<http://dx.doi.org/10.1016/j.ijhydene.2015.07.044>

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technical barriers of PEMFCs still need to be overcome in the development of electrochemical catalysts for widespread commercialization of fuel cells [3]. Cathode catalysts are mainly composed of platinum nanoparticles on a carbon support, and one of the main causes of catalyst degradation is loss of the platinum surface area due to corrosion of the carbon supports and subsequent agglomeration of platinum nanoparticles. Carbon supports, such as carbon black, are thermodynamically unstable at potentials above 0.2 V, and may be prone to oxidation during the electrochemical reactions in the fuel cell [4]. This corrosion of the carbon support results in the detachment of Pt nanoparticles from the support, which may lead to a loss of catalytic activity and a decrease of the cell performance [5,6].

Recently, there have been a lot of researches in order to solve the corrosion of carbon supports. One strategy for maintaining the cell performance is to use a more stable graphitic carbon support instead of an amorphous carbon black. Wang et al. prepared a Pt catalyst using hollow carbon nanocages (CNCs) developed with a high degree of graphitization and concurrent nitrogen doping [7]. The catalyst exhibited lower loss of electrochemical surface area (ECSA) due to degradation and enhanced durability toward oxygen reduction reaction (ORR), compared to a Pt catalyst supported on less graphitized CNCs, which were attributed to the excellent comprehensive properties of CNCs required for a catalytic support, particularly their strong oxidation resistance with Pt particles due to their graphitization and concurrent nitrogen doping. In general, it is well-known that the improved corrosion resistance of the nano graphite-supported catalysts is ascribed to the graphite structure of support. Shanahan et al. reported synthesis of highly stable graphitic mesoporous carbon using the soft polymer template method at 2600 °C, demonstrating lower ECSA loss of the Pt catalyst compared to the Pt catalyst supported on carbon black, which was attributed to the reduction of corrosion due to the highly crystalline structure of the carbon support [8]. And Lin et al. reported graphite submicronparticles as an enhanced durable catalytic support for the resistance of carbon corrosion, which are attributed to the highly ordered graphite structure as well as the morphology of submicronparticles in the range of 0.2–0.3 μm [9]. However, even with the improvements of the graphitic carbon, there is still a key barrier in their practical application: carbon materials have an intrinsic instability of carbon materials in fuel cell environments, regardless of the crystallinity of the carbon support. We have also developed ordered mesoporous carbon (OMC) materials as novel supports for electrochemical catalysts, which exhibit large surface areas, highly interconnected mesopores and graphitic frameworks [10–16]. Recently, a Pt catalyst supported on a sulfur-containing OMC (Pt/S-OMC) exhibited a higher electrochemical stability as well as a catalytic activity for ORR under acidic conditions, due to the strong metal–support interaction between the Pt nanoparticles and sulfur [15]. However, the mesostructure of Pt/S-OMC appeared to collapse moderately after the 2000th voltage-cycling test. Thus, structural reinforcement of the OMC support is required to provide superior electrochemical stability [16].

Another approach to improve the durability of the fuel cell catalysts is the utilization of electrochemically stable

alternatives, instead of the carbon supports [17,18]. Basic requirements for these alternative supports in the fuel cell catalysts are high surface area, favored dispersion of catalyst metal particles, high stability in an acidic media, excellent conductivity and high electrochemical steadiness under fuel cell operating conditions. In addition, the alternative supports should also have a strong physicochemical and electronic interaction with the catalytic metals to improve their catalytic activity and durability [19,20]. Considerable researches have been carried out in the past decades with respect to the exploration of diverse supports for fuel cell catalysts, which include the use of metal, nitride, carbide, mesoporous silica, conducting polymers and metal oxides [21–26]. Among these alternative supports, silicon carbide (SiC) materials have been proposed to be of interest due to their hardness, chemical inertness and high thermal stability up to 1200 °C in an oxidative environment [27–32]. Dhiman et al. attempted to prepare a Pt catalyst supported on SiC nanocrystals that were obtained from a reaction of SiO<sub>2</sub> vapors with carbon black at 1450 °C [29]. The electrochemical measurements indicated that the ECSA of the Pt/SiC catalyst was larger than that of a commercial Pt/C catalyst (20 wt% Pt, E-TEK). These results opened up a possibility for the use of a new class of SiC materials as the fuel cell catalyst support, because of their good chemical and mechanical stability. However, even though the SiC nanocrystal support exhibited surface areas of 55–75 m<sup>2</sup>/g, they were not sufficiently large to accommodate the high loading of Pt, above 40 wt%. In addition, the low conductivity of SiC itself (around 10<sup>-6</sup> S/cm) should be improved. Carbon black, with high electrical conductivity, was added to the Pt/SiC catalyst system to supplement for the insufficient conductivity of the SiC support, but fast degradation of fuel cell performance was observed, due to the oxidation of additional carbon black [30,31].

For the first time, we report a unique nanocomposite OMC support with the SiC on the surface of carbon frameworks, to utilize both the ordered mesopore of the OMC and the high electrochemical stability of the SiC materials. This was accomplished by a controlled carbothermal reduction process, as shown in Fig. 1. The carbothermal reduction conditions were carefully controlled during the nano-replication using an ordered mesoporous silica as a hard template, in order to obtain OMC–SiC composites with high surface areas and well-defined mesopores. A Pt catalyst supported on the OMC–SiC composite exhibited excellent electrochemical activity as well as stability, compared to the Pt catalysts on OMC and the commercial Pt/C.

## Experimental

### Synthesis

An ordered mesoporous silica, SBA-15, with a rod-type morphology [33] was utilized as the template for the preparation of OMC, OMC–SiC and SiC via a nano-replication method. 1,10-phenanthroline and *para*-toluene sulfonic acid was simultaneously infiltrated within the mesopores of the silica template by an incipient wetness impregnation method [15], and the carbon precursor-silica template composites

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