



# Antimony doped tin oxide applied in the gas diffusion layer for proton exchange membrane fuel cells



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## ABSTRACT

In this paper, antimony doped tin oxide (ATO) was firstly used as a conductive material in the microporous layer (MPL). ATO had higher electrochemical stability than XC-72 carbon powder. With the same thickness of MPL, the electrochemical corrosion behavior of gas diffusion layers (GDLs) prepared with ATO and XC-72 carbon powder in the simulated PEMFC operation environment were examined and the results showed that the GDL prepared with ATO had higher durability after oxidation for 55 h. Besides, the fuel cell performance with the GDLs prepared with ATO and XC-72 carbon powder as the cathode GDLs were evaluated and a very small difference between those two cells was observed at low current densities, which demonstrated that GDL prepared with ATO had wide application prospects.

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## 1. Introduction

Proton exchange membrane fuel cell (PEMFC) has been drawing much interest in recent years as a promising alternative power source for stationary and mobile applications [1,2]. Situated between the catalyst layer and bipolar plate, a gas diffusion layer (GDL) within a PEMFC facilitates the transport of reactant gases and produced water, electrical and thermal conduction, etc. [3,4]. Usually, the GDL consists of a carbon paper or carbon cloth substrate and a microporous layer (MPL) is composed of a mixture of carbon powder and polytetrafluoroethylene (PTFE) on one side of the substrate facing the catalyst layer. The addition of MPL could improve water removal and prevent mass transport losses at higher current densities and facilitate the transport of liquid water and current collection, which is beneficial to achieve higher current density and thus better fuel cell performance [5]. Besides, MPL serves key functions in decreasing the contact resistance and providing a better mechanical compatibility with the catalyst layer. As a result, much attention has been paid to optimize the fabrication of MPL including the examination of the optimal loadings of carbon powder [6] and PTFE [7,8], the types of carbon powder [9,10] and the structure of MPL [11–13].

Durability is a major challenge and pursued currently to promote the development and commercialization of PEMFC [14]. In terms of operating environment, the PEMFC is prone to change the GDL from hydrophobic to hydrophilic due to high temperature, high water content, low pH, high oxygen concentration, high potential, catalyst, various operating conditions, etc. [15]. The hydrophobicity degradation could induce the accumulation of excess water blocking the flow channels and the pores of the GDL and lead to reactant starvation instantly [16]. Many studies showed the carbon corrosion mechanism under electrochemical conditions and indicated that fuel starvation and frequent start-up/shut down could accelerate the carbon corrosion [17,18]. Besides, taking into account the MPL near catalyst layer within the PEMFC, the corrosion of carbon powder in the MPL could be accelerated by the presence of Pt. The improvement of the electrochemical durability of MPL is vital to improve the durability of PEMFC [19,20].

In our earlier work [21,22], the causes for the hydrophobicity degradation of GDL were investigated and it was found that the formation of oxygen functional groups due to the oxidation of carbon powder and the loss of PTFE were the main reasons for the decrease of hydrophobicity. However, few literature pay attention to improve the durability of MPL, though more and more evidence indicating research in this area is required. In our present work, to improve the electrochemical durability of GDL a new kind of material, antimony tin oxide (ATO), was adopted to replace carbon powder. ATO is a typical n-type semiconducting material with excellent stability because of its specially physical and

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chemical properties [23]. It was found that the GDL prepared with ATO had better electrochemical durability than the GDL prepared with XC-72 carbon powder.

## 2. Experimental

### 2.1. Corrosion of materials

The measurement was conducted at CHI730 electrochemical working station with a three-electrode system using Pt foil and saturated calomel electrode (SCE) as the counter electrode and reference electrode, respectively. Five milligrams of tested sample, 20  $\mu\text{L}$  Nafion solution (5 wt%) and 2.5 mL isopropyl alcohol were dispersed ultrasonically and 10  $\mu\text{L}$  ink was transferred onto the glassy carbon electrode, which was used as the working electrode and exposed to a constant potential (1.2 V vs. SCE) in 0.5 M  $\text{H}_2\text{SO}_4$  saturated with  $\text{N}_2$  for 2 h to examine the electrochemical stability of samples. The cyclic voltammetry (CV) curve before and after potentiostatic oxidation was measured from  $-0.241$  to  $0.959$  V (vs. SCE) at the rate of  $50$  mV  $\text{s}^{-1}$ .

### 2.2. Preparation of GDLs

The GDL was fabricated with Toray carbon paper as the substrate. The MPL slurry was prepared as described in our earlier work [24]. 400 mg ATO (NERCN Co., China) or 80 mg XC-72 carbon powder was added into 20 ml PVP (58,000 MW, Aladdin Co., USA) aqueous solution using ultrasonication and mechanical stirring and then 640 mg PTFE aqueous suspension (5 wt.%) was added. The slurry was coated onto one side of the carbon paper substrate, followed by drying at  $80$   $^\circ\text{C}$  and then sprayed with some PTFE aqueous suspension on the other side of the substrate. After coating MPL on one side and spraying PTFE on the other side, the GDL was sintered at  $240$  and  $340$   $^\circ\text{C}$  for 30 min, respectively. To make sure the thickness of the two kinds of GDLs are the same, the equal-volume fraction method was adopted by a pressed disc method.

### 2.3. Electrochemical oxidation tests

The electrochemical durability of GDL was tested in a homemade three-electrode cell setup. The prepared GDL ( $3.0 \times 3.0$  cm) was prepared as a working electrode. A graphite board and a saturated calomel electrode (SCE) were employed as the counter-electrode and reference electrode, respectively. The electrodes were held vertically in a chamber filled with 0.5 M  $\text{H}_2\text{SO}_4$ , which was kept in a water bath at  $65$   $^\circ\text{C}$ . During

the whole tests, the  $\text{H}_2\text{SO}_4$  solution was saturated with  $\text{N}_2$  and the GDL was exposed to a constant potential (1.2 V vs SCE) for 55 h.

### 2.4. Characterization

The surface morphologies of GDLs were examined with a field emission scanning electron microscopy (FESEM; Hitachi, S-4800) and the surface contact angles of MPLs were measured by a contact angle system (Drop Shape Analyzer 100, Kruss, Germany). In addition, the gas permeability of different GDLs was characterized by pore size distribution analyzer (PSDA-20) and calculated by Eq. (1).

$$\mu = \eta \times \frac{L}{A} \times \frac{Q}{\Delta P} \quad (1)$$

Where  $\eta$  was the gas viscosity,  $L$  was the thickness of the sample and  $A$  was the cross-sectional area through which the gas passes,  $Q$  was the volumetric flow rate and  $\Delta P$  was the pressure difference across the sample.

Besides, the through-plane contact resistance of GDL was measured using material testing machine. The tested sample was placed between two gold bullions supplied with electrical source. The electrical current was set at 5A and the area of the tested sample was  $9$   $\text{cm}^2$ . The tested pressure was applied from  $1$  to  $16$   $\text{kg}\cdot\text{cm}^{-2}$  with  $1$  kg increment with the rate of  $3$  N  $\text{s}^{-1}$  between measurements. When the tested pressure

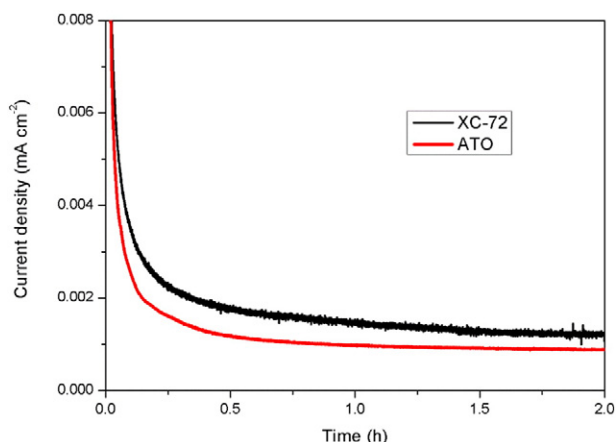


Fig. 1. Curves of oxidation current of ATO and XC-72 carbon powder versus time.

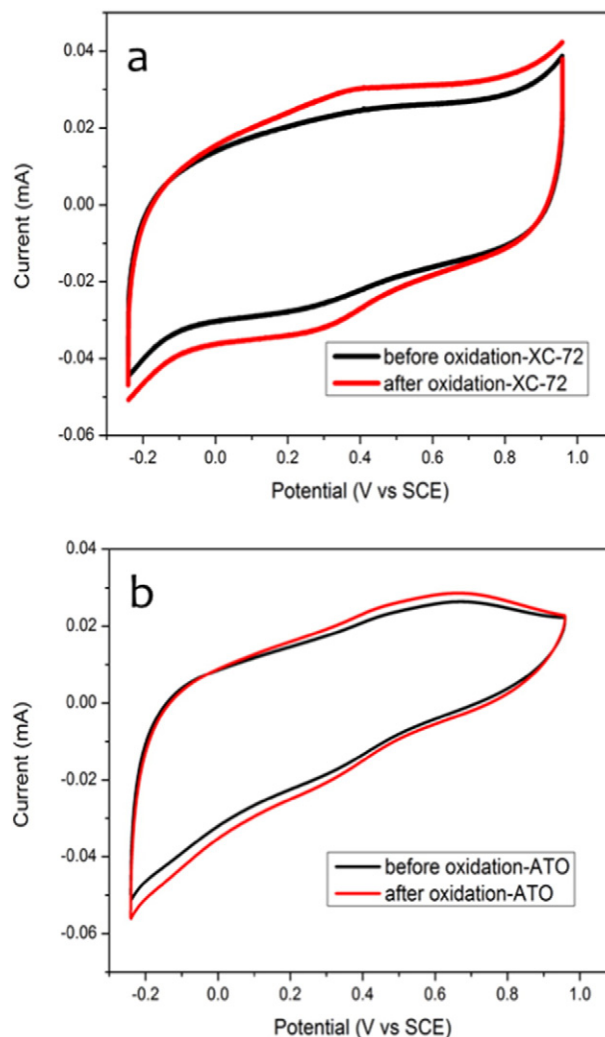


Fig. 2. CV curves of (a) XC-72 carbon powder and (b) ATO before and after oxidation for 2 h.

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