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Carbon nanotubes as durable catalyst supports for oxygen reduction electrode of proton exchange membrane fuel cells

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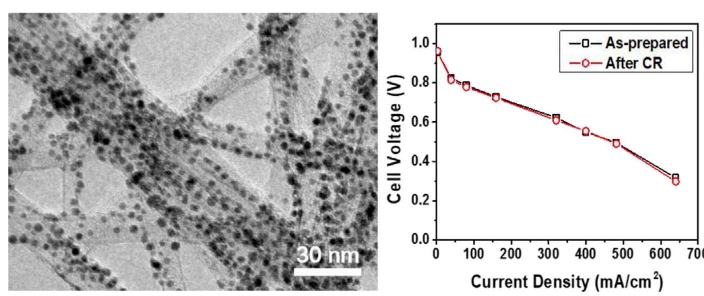
HIGHLIGHTS

- Pt/CNTs exhibits electrochemically durable oxygen reduction reaction property.
- Pt/CNT catalyst maintained the initial performance after 100 rounds of cell reversal.
- High crystallinity and low defect density of CNTs suppress the carbon oxidation reaction.
- Pt/CNTs demonstrates consistent cell potential, ESCA, and electrical resistance.

GRAPHICAL ABSTRACT



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ABSTRACT

Herein, we report electrochemically durable oxygen reduction reaction catalysts for proton exchange membrane fuel cells under conditions of hydrogen starvation. Insufficient hydrogen supply during transient conditions leads to a cell reversal state in a proton exchange membrane fuel cell, and oxidation of the carbon support severely decreases the cell performance. The use of carbon nanotubes as a support for the oxygen reduction reaction catalyst is found to suppress carbon oxidation due to the high crystallinity, low defect density, and high aspect ratio, which results in a consistent cell voltage, electrochemical surface area, and electrical resistance, even after 100 rounds of cell reversal. This study demonstrates the excellent durability of proton exchange membrane fuel cells with a Pt/Carbon nanotube electrode, as confirmed by X-ray diffraction, transmission electron microscopy, Raman spectroscopy and cyclic voltammetry analysis.

1. Introduction

Proton exchange membrane fuel cells (PEMFCs) have attracted considerable attention for power sources of zero emission vehicles as they are compact, lightweight, clean, and have a high power density. The oxygen reduction reaction (ORR) at the cathode of a PEMFC plays a key role in determining the performance of a fuel cell [1,2], and durable ORR catalysts able to endure over 5000 h of operation are essential for the commercial adoption of fuel cells [3,4]. Platinum (Pt) is regarded as a standard ORR catalyst in PEMFCs because of its high catalytic activity, and the catalyst is generally prepared by loading Pt on a carbon black (CB) support [5,6]. However, Pt/CB catalysts suffer from low

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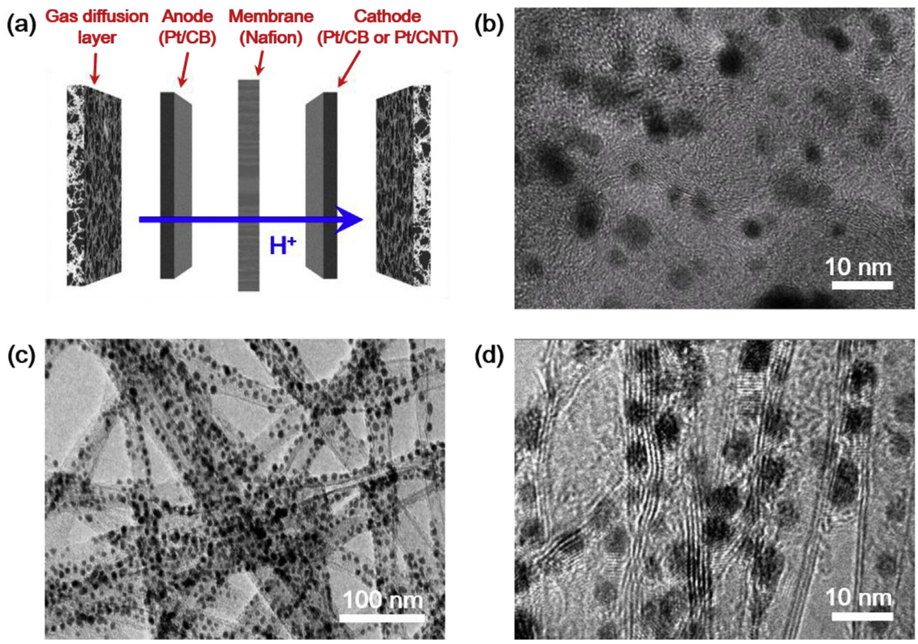

Fig. 1. (a) Schematic illustration of the membrane electrode assembly. (b) High-magnification TEM image of as-prepared Pt/CB, and low- (c) and high- (d) magnification TEM images of Pt/CNT catalyst.

Table 1
Compositions of anodes and cathodes of MEAs.

MEA	Catalyst		Pt loading (mg/cm^2)		Catalyst: Nafion (weight ratio)	
	Anode	Cathode	Anode	Cathode	Anode	Cathode
MEA-a	Pt/CB	Pt/CB	0.4	0.4	3: 1	3: 1
MEA-b	Pt/CB	Pt/CNT				

long-term durability and large overpotential loss [1,6,7], and catalyst degradation is accelerated during transient conditions, such as during startup/shutdown and cell reversal (CR) processes [3,4].

Fuel starvation during fuel cell operation can be caused by several factors such as water flooding within the fuel cell, ice formation in winter, and abnormal operation of the reactant gas supply [3]. Insufficient hydrogen (H_2) supply in the anode results in CR of the PEMFC, and the CB support of the Pt/CB catalyst quickly reacts with water to sustain the stack current [8,9]. The carbon oxidation reaction (COR) greatly decreases the amount of CB support within the cathode, resulting in unfavorable PEMFC operation conditions such as

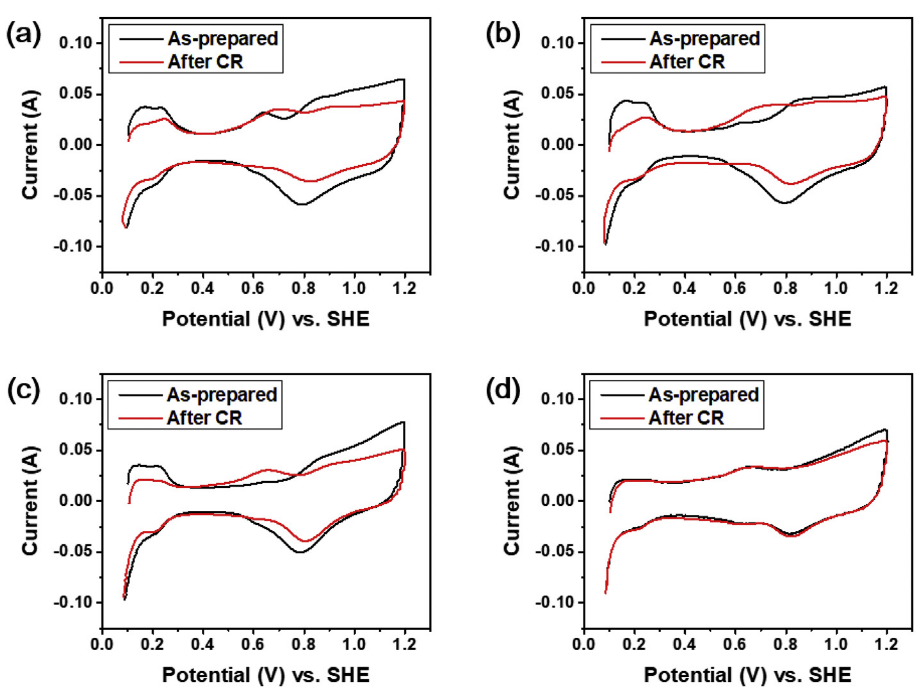

Fig. 2. The first CVs of as-prepared (black) and after cell reversal (red) for (a) an anode of MEA-a, (b) cathode of MEA-a, (c) anode of MEA-b, and (d) cathode of MEA-b. The scan rate was 30 mV/s. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)