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Nanocatalysts for Low Temperature Fuel Cells

A.M. Kannan

The Polytechnic School, Ira A. Fulton Schools of Engineering, Arizona State University, Mesa 85212 USA

Abstract

Zeolitic Imidazolate Frameworks (ZIFs) are one of the potential candidates as highly conducting networks with surface area with a possibility to be used as catalyst support. In the present study, highly active state-of-the-art Pt-NCNTFs catalyst was synthesized by pyrolyzing ZIF-67 along with Pt precursor under flowing Ar-H_2 (90-10 %) gas at 700 °C. XRD analysis indicated the formation of Pt-Co alloy on the surface of the nanostructured catalyst support. The high resolution TEM examination showed the particle size range of 7 to 10 nm. Proton exchange membrane fuel cell performance was evaluated by fabricating membrane electrode assemblies using Nafion-212 electrolyte using H_2/O_2 gases (100 % RH) at various temperatures. The peak power density of 630 mW.cm^2 was obtained with Pt-NCNTFs cathode catalyst and commercial Pt/C anode catalyst at 70 ºC at ambient pressure.

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Keywords: ZIF-67; ORR catalysts; Membrane-Electrodes Assembly; PEM Fuel Cell

* Corresponding author.
E-mail address: *amk@asu.edu*

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1. Introduction

Proton exchange membrane fuel cell (PEMFC) is the most promising energy conversion technology for stationary as well as automotive applications due to its advantages such as lower operating temperature and higher power density as compared to other types of fuel cells. However, for wide-scale commercialization of PEMFCs, it should overcome several challenges including reducing the cost, maximizing the utilization of platinum catalyst, improving the performance and durability of the membrane electrode assembly. Oxygen reduction reaction (ORR) is the most important one due to the potentially lower exchange current density value in the PEMFC [1,2,3]. In order to have reasonable reaction kinetics, noble metals based nanocomposites are the most efficient electrocatalysts towards ORR. However the noble metal ORR catalysts are highly expensive and are the leading barriers for the PEMFC commercialization [4,5]. In this context, it is important to develop highly efficient and durable electrocatalysts for ORR [6]. Recently, metal-organic frameworks (MOFs) have emerged as a novel class of porous crystalline materials both as template and precursor to produce nanoporous carbons for gas storage [7], catalyst support [8], and electrode materials for lithium batteries [9], sensors [10], and supercapacitors [11]. Zeolitic imidazolate frameworks (ZIFs) as a subclass of MOFs are excellent materials for the synthesis of nanocarbon electrocatalysts with abundant carbon and nitrogen [12,13]. MOF-derived nanocomposites reported as poor ion and electron transport materials due to poor graphitic degree and microporous structures [14-16]. The use of ZIFs as a precursor for the synthesis of N-doped CNTs (NCNTs) structures has rarely been reported as ORR electrocatalyst [17-19]. In the preliminary study, cathode with ultra-low loading of Pt on N-doped carbon nanotube frameworks (Pt-NCNTFs) was developed and evaluated towards ORR catalyst in the PEMFC. The Pt-NCNTFs nanocatalyst was characterized using x-ray diffraction, scanning electron microscope and transmission electron microscope for composition, and nature of particle morphology and distribution. Membrane electrode assemblies fabricated with Pt-NCNTFs cathode nanoelectrocatalyst catalyst (loading of 0.12 mg Pt per cm^2) showed a peak power density of ~630 $mW.cm^{-2}$ at 70 ◦C with H_2 and O_2 gases at 100 % RH under ambient operating pressure, with excellent performance stability.

2. Experimental

2.1 Synthesis of ZIF-67 particles

ZIF-67 samples were synthesized as described in the published literature [3]. In a typical synthesis, 2-methylimidazole (1.97 g) was dissolved in a mixed solution of 20 ml of methanol and 20 ml of ethanol. $Co(NO_3)_2.6H_2O$ (1.746 g) was dissolved in another mixed solution of 20 ml of methanol and 20 ml of ethanol. The above two solutions were then mixed under continuous stirring for few minutes and held for 20 h at room temperature. The purple precipitate was collected by centrifuging the solution, washed in ethanol several times and dried at 80 °C overnight. The flowchart in Figure 1 provides the process steps sequentially.

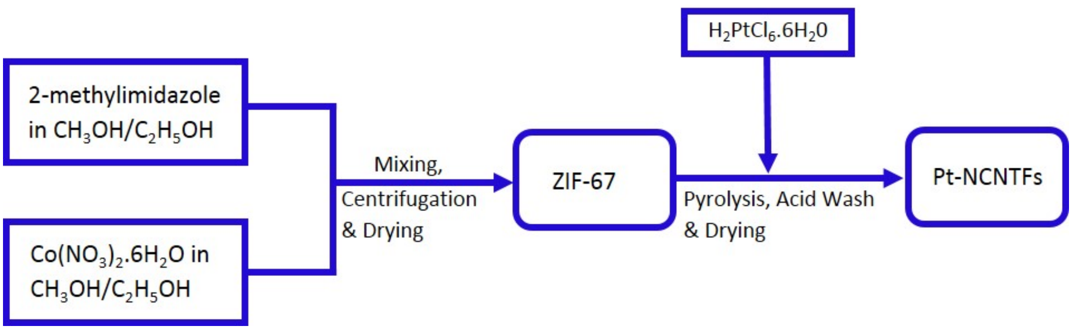

Figure 1. Process flowchart for Pt-NCNTFs Synthesis.

2.2 Synthesis of Pt-NCNTFs

The ZIF-67 particles were soaked with $H_2PtCl_6.6H_2O$ solution (5 wt % solution in DI water) and the resulting material was dried at 80 °C for 1h. The dried powder was heated at 350 °C for 1.5 h then raised to 700 °C at a ramp rate of 2 °C per minute and pyrolyzed for 3.5 h under flowing Ar/H_2 (90%/10% in volume ratio) atmosphere (see