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# Facile solvothermal fabrication of polypyrrole sheets supported dendritic platinum-cobalt nanoclusters for highly efficient oxygen reduction and ethylene glycol oxidation



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#### G R A P H I C A L A B S T R A C T



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#### ABSTRACT

Herein, uniform dendritic PtCo nanoclusters supported on sheet-like polypyrrole (PtCo NCs/PPy) were prepared by a facile one-pot solvothermal method. Cetyltrimethylammonium chloride (CTAC) and pyrrole worked as the capping agent and reductant, respectively, and pyrrole was *in-situ* polymerized to form PPy sheets under solvothermal conditions. The dendritic PtCo NCs/PPy had the enlarged electrochemically active surface area (EASA,  $30.95 \text{ m}^2\text{g}^{-1}$ ), and showed the superior catalytic performance and durability towards oxygen reduction reaction (ORR) and ethylene glycol oxidation reaction (EGOR) in comparison with Pt<sub>1</sub>Co<sub>3</sub> nanoparticles (NPs), Pt<sub>3</sub>Co<sub>1</sub> NPs and commercial Pt/C catalysts. This work displays a new strategy for rational design and synthesis of advanced functional nanocomposites as electrocatalysts in fuel cells.

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#### 1. Introduction

Recently, bimetallic nanoalloys have drawn great interest for their promising catalytic properties in fuel cells, sensing, and catalysis [1]. Among them, Pt-based nanostructures have attracted increasing attention because of their excellent catalytic characters

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https://doi.org/10.1016/j.jcis.2018.06.095 0021-9797/© 2018 Published by Elsevier Inc. [2]. However, their commercial applications are seriously hindered by the high cost and limited resource of Pt, Ostwald ripening and deactivation from poisonous CO-like intermediates on the catalysts [3,4]. Thus, it is of vital importance to develop Pt-derived nanocatalysts to replace pure Pt counterparts.

The incorporation of non-precious 3d transition metal (M = Co, Ni, Cu or Fe) into Pt attracts increasing attention [5], since the resultant PtM alloy can remarkedly reduce the cost and surface adsorption of poisonous intermediates and/or reaction products,

exposing enriched active sites for catalytic reactions [6–8]. Among PtM catalysts, PtCo catalysts are of great significance owing to their prominent catalytic performance towards ethylene glycol oxidation reaction (EGOR) and oxygen reduction reaction (ORR) [9,10]. For example, Yu and her coworkers fabricated platinum<sub>69</sub>-cobalt<sub>31</sub> nanosheet nanoassemblies (Pt<sub>69</sub>Co<sub>31</sub> NSNSs) with improved catalytic activity and durability towards EGOR and ORR [11]. In another example, Kadirgan's group prepared PtCo nanoparticles (NPs)/C, showing the great enhancement in ORR activity and stability compared with commercial Pt/C [12].

Besides, supporting materials are usually introduced for the immobilization of the nanocatalysts, which can greatly enhance the dispersion, stability, and durability of the deposited catalysts [13]. In recent years, conducting polymers such as polyaniline, polypyrrole (PPy) and polythiophene have attracted significant attention due to their good chemical stability, high electric conductivity, scalable and low cost [14]. Particularly, the encapsulating PPy layer is regarded as ideal coating material in commercial applications, which strengthens the protecting function [15]. On the one hand, the PPy sheet can promote the formation of an electric field on the substrate, which limits the migration of electron transfer occurring on the substrate surface. On the other hand, the PPy layer can improve the stability [16].

Herein, a one-pot hydrothermal synthesis was developed to synthesize PtCo NCs supported on sheet-like PPy (PtCo NCs/PPy) with superior catalytic performance in the presence of pyrrole and CTAC. The combination of dendritic PtCo NCs and PPy promoted the electrocatalytic properties owing to the synergistic effects of the encapsulated PtCo NCs with PPy, as examined through ORR and EGOR as the two benchmark systems.

2. Experimental

#### 2.1. Synthesis of dendritic PtCo NCs/PPy

For the typical construction of dendritic PtCo NCs/PPy, the metal precursors  $(0.009 \text{ g of } Co(acac)_3 \text{ and } 0.010 \text{ g of } Pt(acac)_2)$ ,

and 0.032 g of CTAC were placed into 5 mL of oleylamine under stirring, followed by slowly injecting 20.8  $\mu$ L of pyrrole monomer into the above mixed solution. The mixture was ultrasonicated for 60 min, which was then heated to 160°C in oil bath and remained for 8 h, followed by cooling down to room temperature naturally. The sample was collected by centrifugation and repeatedly washed with hexamethylene and ethanol. Finally, the product was dried at 60°C for further characterization.

For contrast materials, Pt<sub>1</sub>Co<sub>3</sub> NPs/PPy and Pt<sub>3</sub>Co<sub>1</sub> NPs/PPy were prepared by adjusting the initial ratios of Pt(acac)<sub>2</sub> to Co(acac)<sub>3</sub> to 1:3 and 3:1, respectively, while keeping the other experimental conditions unchanged. More detailed information of the *Characterization* and *Chemicals* were provided in Supporting Information (SI).

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.jcis.2018.06.095.

#### 2.2. Preparation of dendritic PtCo NCs/PPy modified electrode

To prepare dendritic PtCo NCs/PPy modified electrode, 1 mg of the product was dissolved by 1 mL of ethanol, which was then ultrasonicated for 0.5 h to get a well-homogeneous suspension. Then, 3  $\mu$ L of the suspension was successively casted onto a glassy carbon electrode (GCE, 3 mm in diameter) or glassy carbon rotating disk electrode (RDE, 3 mm in diameter), accompanied by dropping 4  $\mu$ L of Nafion (0.05 wt%) to tightly unite the deposit and drying at room temperature. Similarly, Pt<sub>1</sub>Co<sub>3</sub> NPs/PPy, Pt<sub>3</sub>Co<sub>1</sub> NPs/PPy and commercial Pt/C (50 wt%) catalysts modified electrodes were prepared for comparison.

More detailed information of the *Electrochemical measurements* were provided in SI.

#### 3. Results and discussion

#### 3.1. Characterization

Transmission electron microscopy (TEM) is a powerful technique for structure characterization [17]. Fig. 1 shows the



Fig. 1. (A) Low-, (B) medium-, (C-E) high-resolution TEM images of dendritic PtCo NCs/PPy. (F) The SAED pattern. Inset in B shows the particle-size distribution.

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