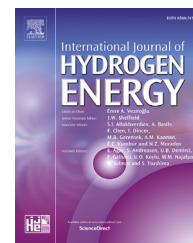


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# Freestanding palladium nanonetworks electrocatalyst for oxygen reduction reaction in fuel cells

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## ABSTRACT

Still it's a main challenge to design of highly efficient electrocatalysts to reduce the high overpotential of the oxygen reduction reaction (ORR). The 1 dimensional (1D) palladium nanonetworks (Pd-Net) can be a promising alternative to platinum (Pt)-based electrocatalyst for ORR. In this study, the Pd-Net electrocatalysts have been synthesized via a simple wet-chemical method with the assistance of cetyltrimethylammonium bromide (CTAB) and zinc precursor. Further investigation indicates that the thickness of Pd-Net can be regulated by simply changing the molar ratio of CTAB and the  $5 \pm 0.1$  nm is proven as an efficient ORR electrocatalyst without any support material. The freestanding 1D Pd-Net has shown 2.2 and 3.6-fold higher electrochemical surface area than that of commercially available Pt/C and homemade Pd nanoparticles (PdNPs) catalysts, respectively. As a result, it provides a higher density of ORR active sites and facilitated the electron transport. The Pd-Net catalyst shows 2.1 and 4.1 times higher mass activity and 1.3 and 3.1 higher specific activity at 0.85 V (vs. RHE) with better ORR kinetics than that of Pt/C and PdNPs, respectively. Additionally, the Pd-Net catalyst has been demonstrated a significant tolerance to the anodic fuels (i.e. methanol) and enhanced durability than the Pt/C and PdNPs catalysts for ORR.

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## Introduction

The four-electrons ( $4e^-$ ) involving direct oxygen reduction reaction (ORR) [1,2] catalysis is an interesting research area because of its important role in the application of energy storage and conversion devices, such as fuel cells (FCs) in alkaline media [1–7]. Thus, the development of efficient ORR

electrocatalysts is crucially important for clean energy production technology. Typically, the platinum (Pt) and its alloy materials have been regarded as the most efficient electrocatalyst for ORR [8–11] because of the boosting sluggish kinetic rate of ORR. Unfortunately, Pt-based materials are susceptible to the crossover effect caused by the diffusion of fuel molecules from the anode, CO poisoning, and poor durability [8,12–14]. Moreover, the limited availability and

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expense of Pt are also great obstacle to reduce the cost of FCs [15].

Recent intensive research efforts have led to the development of less expensive and more active palladium (Pd)-based nanomaterials for ORR catalysis to overcome the aforementioned challenges [16–25]. However, Pd-based ORR catalysts have gained remarkable attraction because it serves as an attractive alternative in the development of Pt-free catalysts with even better catalytic activity than Pt [4,17,19,26]. For example, the Pd surface electrode exhibited over 3 times higher kinetic current density ( $j_k$ ) in comparison with the Pt surface electrode for ORR [27]. Many researchers have reported that the Pd electrode exhibits higher stability towards ORR than the Pt electrode [10,19,23]. Furthermore, Pd is abundantly available in nature and cheaper than Pt (Pt:927\$/Oz; Pd 872\$/Oz), this fact is also designates it as an ideal alternative to Pt [24,28]. Therefore, effort should be exerted to identify alternative catalysts that are readily available and cost effective with comparable or even better electrocatalytic activity than Pt for cathodic ORR in FCs.

Current catalyst materials are still far from meeting the requirements of combined high catalytic activity, better durability and low cost [29]. Thus, many efforts such as, nanoparticles (NPs), nanonetwork (Net), nanoflowers, nanotubes, and nanodendrites have been explored to develop more electroactive and efficient Pd-based nanocatalysts for FCs application [20,30–35], while the size and shape of catalysts are strongly related with their electrocatalytic performances [36]. Among them, metallic (i.e. Pd)-Net can maximize the Pd utilization and enhance the catalytic activity because of the structural benefits such as, self-support, high surface area, low density, and special electrical properties, resulting in an increased electrochemically active surface area (ECSA) associated with the higher catalytic performance [37,38]. Although, many efforts have been focused on the preparation of bimetallic nanonetwork-like catalysts with precious metals (i.e. Au, Ag) [30,31,38], which actually led to the higher metal loading, high cost and low ECSA. For example, bimetallic PtPdTe/CF network catalyst [38] showed a quite low ECSA

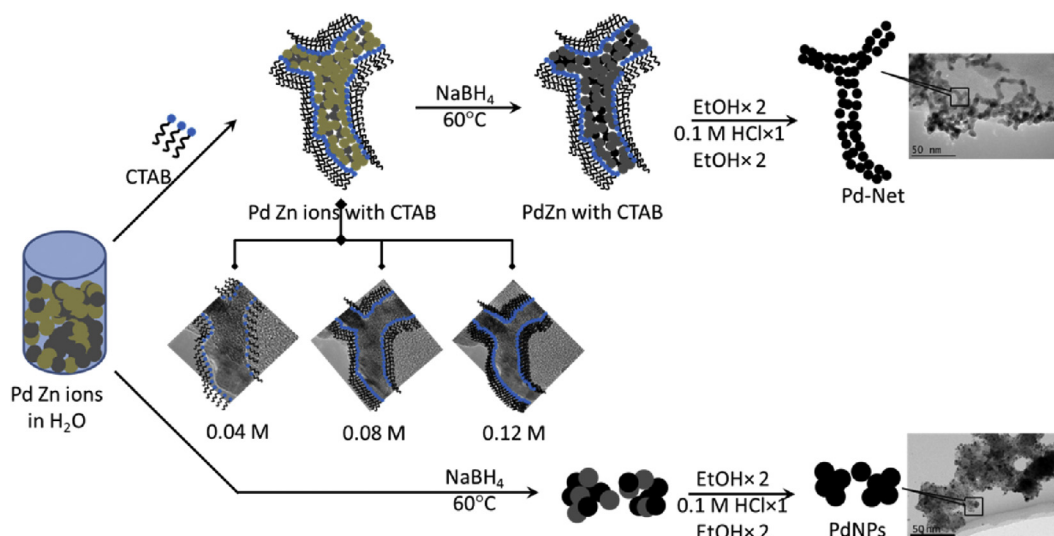
( $14.9 \text{ m}^2 \text{ g}_{\text{Pt,Pd}}^{-1}$ ) when normalize against total metal mass. Also, the durability and catalytic activity under real operational conditions often hampered by the corrosion of the support materials (i.e. carbon or its allotropes), and the weak interaction between the metallic site (i.e. NPs) and support [32,39]. Therefore, the development of monometallic and free-standing Pd nanonetwork is still an urgent topic, particularly, for better ORR catalysis in FCs application.

Considering above discussions, we have developed a simple strategy to synthesize freestanding Pd-Net with 1D porous nanoarchitecture. For the first time, the Pd-Net has been employed successfully as monometallic freestanding electrocatalyst for ORR catalysis. The as-prepared Pd-Net exhibited superior electrocatalytic activity for ORR with excellent stability in alkaline media which comparable to the Pt/C and homemade PdNPs. The porous Pd-Net ensured a high density of active sites as well as excellent mass and charge transport during ORR. As a result, the as-prepared Pd-Net is demonstrated to be a promising low-cost, highly efficient electrocatalyst as an efficient alternative for Pt-based catalysts.

## Experimental

### Material synthesis

The synthesis of all catalysts has demonstrated by the schematic diagram in Scheme 1. For the preparation of Pd-Net, 1 mL of 0.01 M  $\text{K}_2\text{PdCl}_4$  and 1 mL of 0.01 M  $\text{ZnCl}_2$  were taken together into a 20 mL vial, followed by the addition of 1 mL of 0.08 M cetyltrimethylammonium bromide (CTAB) under vigorous stirring to ensure adequate mixing. Under continuous stirring, 1 mL of 0.1 M  $\text{NaBH}_4$  was added to the solution drop wise. Afterwards, the above solution kept in an oven for steady-state heating at  $60^\circ\text{C}$  for 12 h. The solution was then centrifuged thoroughly with excess amount of ethanol for twice. To remove Zn, 0.1 M HCl was added for etching treatment and finally followed by the cleaning with ethanol for couple of time by centrifugation. The as-obtained Pd-Net was



Scheme 1 – The preparation of Pd-Net and homemade PdNPs.

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