

RAPID COMMUNICATION

Biomimetic design of monolithic fuel cell electrodes with hierarchical structures



Rongyue Wang^a, Drew C. Higgins^a, Dong Un Lee^a,
Sagar Prabhudev^b, Fathy M. Hassan^a, Victor Chabot^a,
Gregory Lui^a, Gaopeng Jiang^a, Ja-Yeon Choi^a,
Lathankan Rasenthiram^a, Jing Fu^a, Gianluigi Botton^b,
Zhongwei Chen^{a,*}

^aDepartment of Chemical Engineering, Waterloo Institute for Nanotechnology, Waterloo Institute for Sustainable Energy, University of Waterloo, 200 University Avenue West, Waterloo, Ontario, Canada, N2L 3G1

^bDepartment of Materials Science and Engineering, McMaster University, Hamilton, Ontario, Canada, L8S 4L8

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Abstract

Despite the significant improvement of polymer electrolyte membrane fuel cell catalyst activities, a cost-effective and stable membrane electrode assembly is still lacking, which greatly inhibits the commercialization of this efficient and environmental friendly technology in stationary and transportation applications. The main reason is that the engineering of different components of an electrode, such as catalytically active metals, electron transport and reactant diffusion paths in a compatible way is very challenging. Here we show the design and preparation of a monolithic fuel cell electrode with a compatible wire on wire structure that mimics the configuration of a pine tree. We developed a procedure to make a flexible carbon thin film composed of porous nanofibers with a thickness of ~ 100 nm and centimeter scale lengths. Platinum nanowires (ca. 3 nm diameter) were deposited on these microscale carbon nanofiber films, resulting in a hierarchical structure. The platinum nanowires were then decorated with a porous bismuth coating to modulate the atomic structure and induce catalytic activity toward formic acid electrooxidation. The end result is a monolithic structure used as a fuel cell electrode that combines microscale diffusive pathways and nanoscale catalyst structures. Prepared by a process that is readily scalable, this design strategy offers a new way to tailor catalytic functions at a system level.

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*Corresponding author. Tel.: +1 519 888 4567x38664.

E-mail address: zhwchen@uwaterloo.ca (Z. Chen).

Introduction

When powered by renewable fuels, polymer electrolyte membrane (PEM) fuel cells offer efficient and environmentally benign solutions for our future power source demands in sectors ranging from transportation to small electronic devices [1-3]. Despite two decades of intensive investigations and advancements achieved in fundamental electrochemistry and nanocatalyst synthesis, the implementation of PEM fuel cells at large scale is still hindered by the lack of cost-effective and operationally stable membrane electrode assemblies (MEAs) [4-10]. This situation is caused by the fact that most of fuel cell catalyst development efforts have focused on the specific aspect of fuel cell electrodes such as activity or durability, while less attention was paid on the engineering requirements of an MEA. This situation urges the investigation of component interfaces and their interaction within MEAs from a system point of view, while simultaneously considering the feasibility of mass-production [7,11-13].

Poor catalyst utilization and non-ideal mass transport properties commonly result from the traditional electrode fabrication strategy used for membrane electrode assembly (MEA) preparation. The strategy involves mixing of platinum nanoparticles (supported on carbon black or unsupported) in an alcohol solution with proton conducting ionomer, and then coating on a gas diffusion layer (GDL) using a brush or spray gun [14]. Introducing large size pores in the electrode was found beneficial for effective mass transport [15]; however, the optimization of Pt utilization in these traditional electrodes is very challenging. Several alternative strategies have been investigated to improve the mass transport properties, water management and catalyst utilization in fuel cell electrodes [16-26]. A straightforward strategy is the direct electro-deposition of electrocatalysts on carbon paper or graphene foam [16-18] that possesses an inherently open and interconnected porous structure. This structure leads to easy reactant transport and electron mobility, and has been demonstrated to provide direct formic acid fuel cell performance improvements in comparison to traditional MEA configurations [16,17]. A significant issue however is that the direct growth of highly dispersed catalysts on carbon fibers with microscale

diameters of $\sim 10\ \mu\text{m}$ (or $\sim 100\ \mu\text{m}$ for graphene foam) is very difficult. Furthermore, the true catalyst surface area that can be achieved on carbon papers or graphene foams is limited by the small surface area of these substrates. Although employing self-assembled carbon nanotube meshes offers higher substrate surface areas [19,20], the uniform and stable integration of Pt nanocatalysts on these supporting materials remains challenging. The nanostructured thin film catalysts developed by physical vapor deposition of Pt or Pt alloy onto organic whiskers represent the state of the art $\text{H}_2\text{-O}_2$ fuel cell catalysts from the aspect of low catalyst loading performance and durability [21]. However, the uniformity of deposited catalytic metals through the whole thickness of the organic whisker substrate is still far from ideal. Furthermore, the use of these catalysts in liquid fuel cells, such as direct methanol or formic acid fuel cells is very challenging because the production of thicker catalyst layers using the physical vapor deposition method is not possible. On the other hand, nanoporous metals have been investigated as catalyst supports in both $\text{H}_2\text{-O}_2$ and liquid fuel cells [22-25]. With ultra-high Pt utilization and fast electron transportation within the inter-connected metal ligaments, these catalysts show the highest Pt mass specific performance in real fuel cells [25]. However, the mass transport within these 20-30 nm pores, especially for liquid fuel cells, effectively limit the areal power densities of these nanoporous metal based electrodes [25]. Ordered macroporous Pt electrodes with pore sizes of $\sim 500\ \text{nm}$ can overcome this by readily facilitating mass transfer, although the optimization of Pt utilization in this structure is very challenging and the electrode preparation process is very complex, which may limit its real application [26]. In order to achieve high performance in fuel cell with optimized Pt catalyst utilization, the design of catalytic electrode should be started from a system point of view and the compatibility of preparation process with existing technology should also be considered [7,11].

In nature, many processes rely on complex structures that are compatibly organized with different levels of sub-structures, leading to functional, hierarchical design. A characteristic example is a pine tree, where the needle-like leaves are grown uniformly on the surface of twigs, which in turn are attached to the branches (see Figure 1a

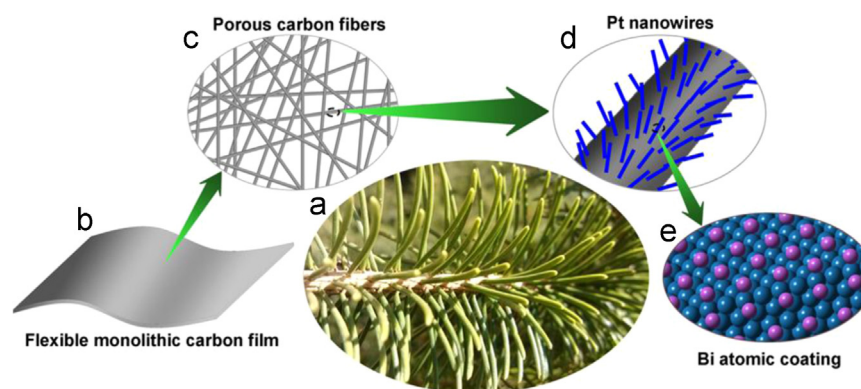


Figure 1 Hierarchical structures in nature and by design. (a) Optical photo of a branch showing the hierarchy wire on wire structure of a pine tree. (b-e) Monolithic thin film catalyst composed of porous carbon nanofibers on top of which Bi decorated Pt nanowires were grown.

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