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Miniature fuel-cell system complete with on-demand fuel and oxidant supply



Janet I. Hur*, Chang-Jin Kim

Mechanical and Aerospace Engineering Department, University of California, Los Angeles (UCLA), 420 Westwood Plaza, Engineering IV Bldg., Los Angeles, CA 90095-1597, USA

HIGHLIGHTS

- We realized a complete miniature fuel-cell system free of ancillary component.
- Pure oxygen gas is supplied to cathode on-demand in dramatically small volume.
- The fuel-cell system can be stacked in multiple to satisfy higher power demands.

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ABSTRACT

The size of a functioning “system” rather than the individual components determines the success of many miniaturization efforts. While most of the existing micro fuel-cell research has been focusing on the fuel-cell stack, our approach has been to systematically eliminate all the ancillary components with the goal of miniaturizing the full system. In this paper, we present a miniature fuel-cell system that combines the self-pumping of fuel and self-generation of oxidant altogether in a box-shape device of a few centimeters. Since the fuel is pumped on demand inside the system without requiring any external assistance, the device is self-sufficient and portable. Furthermore, the oxygen is generated on demand inside the system without requiring the ambient air, so that the device can be stacked in multiple. Constructed simply as liquids in a solid container, this active fuel-cell system resembles a battery to the user.

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1. Introduction

Long-lasting portable power sources are in great need to supply power to portable consumer electronics integrated with emerging technologies [1–3]. Today's military also needs light and compact power sources with longer life to support the soldiers in the field [4]. Furthermore, as microelectromechanical systems (MEMS) technology advanced, numerous micro sensors and devices have become available, such as remote sensors and diagnostic systems, demanding microscale power sources on chip [5–7]. The need for the miniature power sources is stronger than ever.

Being environmentally friendly, fuel cells are attractive in both macro and micro scales. Especially in microscale, the simple construction compared to internal combustion engines and high theoretical energy density compared to batteries make fuel cells a

strong candidate. Miniature fuel cells, based on direct methanol fuel cell (DMFC) technology [8–10] and direct formic acid fuel cell (DFAFC) [11–13], have been actively reported using a membrane electrode assembly (MEA) [14,15] or a membraneless technique [16–19]. Moreover, the U.S. Department of Transportation has approved the use of some fuel cells in airplanes, removing what companies have called a major barrier keeping micro fuel cells out of laptops and cell phones [20].

One of the main remaining barriers against commercialization is the lack of ability to miniaturize the fuel cell as a system because of the so-called “packaging penalty”. Current state-of-the-art proton exchange membrane (PEM) fuel-cell systems still consist of several ancillary components [20,21]. Although many fuel cells reported in the literature are well shrunk in size and named micro fuel cells, almost all of them are micro fuel-cell *stack*, not a micro fuel cell complete as a standalone system. To operate as a complete system, the micro fuel-cell stacks are attached to ancillary components that significantly increase the volume and lower the energy density of a system. More specifically, they require: a pump to flow the liquid

* Corresponding author. Tel.: +1 (310) 825 3977.
E-mail address: janethur@ucla.edu (J.I. Hur).

fuel and to force the flow against carbon dioxide bubble clogging; a gas separator to remove the bubbles; and often a pressurized oxygen tank to deliver oxygen to the system. The schematic drawing of the components needed in a usual microfluidic fuel cell is shown in Fig. 1. Unfortunately, miniaturization and packaging of the entire system is prohibitively challenging since these supporting components take up significant portions of the total volume, posing limitation in miniaturization. When the supporting structures are miniaturized as much as possible and integrated into the system, the inactive (i.e., non-fuel) materials take up a considerable volume of the fuel-cell system, diminishing the fuel and oxidant storage capacity. The packaging penalty becomes unacceptably high if a fuel-cell system (not a fuel-cell stack) should be below ~1 cm in size – the main reason why the flurry of activities in micro fuel cells subsided after early 2000s and failed to provide a chance to compete with batteries. With only a small amount of fuel available in the system, the advantage of high energy density is simply lost in a complete (i.e., full, standalone) fuel-cell system when miniaturized. Furthermore, the complexity of integrating the moving ancillary components in a small space would make the miniature system expensive and unreliable.

We have previously reported a self-pumping fuel cell that has an embedded ability to pump the liquid fuel within microfluidic channels, allowing one to miniaturize fuel cells (at least the anodic side) without the packaging penalty [22,23]. Unlike other passive fuel cells [9,13,24], the self-pumping fuel cell is an active fuel cell; it actively pumps to deliver fresh fuel from a reservoir, where fuel concentration is kept relatively constant. Utilizing the carbon dioxide byproduct on anode was the key idea of the self-pumping technology. By making the growing carbon dioxide bubbles to always expand from the inlet side to the outlet before being vented out, the fuel was pumped in one direction. Doing so enabled us to eliminate the mechanical pump and gas separator, as shown in Fig. 2. This method is in principle applicable and flexible to any kind of liquid fuel, as long as the fuel produces carbon dioxide as byproduct on anode. While the fuel was supplied through self-pumping system, oxygen was provided through air-breathing cathode directly from ambient air. Although the oxygen tank was eliminated, relying on oxygen supply from air limited the application of the miniature fuel-cell system, because the system could not be stacked for higher power output.

In this report, we first report the development of a O₂-generating plate that, when in contact with a hydrogen peroxide solution on one side, grows a thin layer of oxygen gas on the other side in a self-regulating manner. In pursuit of realizing a complete fuel-cell system, we then integrate the O₂-generating mechanism with our previously proven air-breathing fuel-cell system [23]. This unique oxygen supply takes up a much smaller volume than the existing designs, which would require ancillary components, such as a

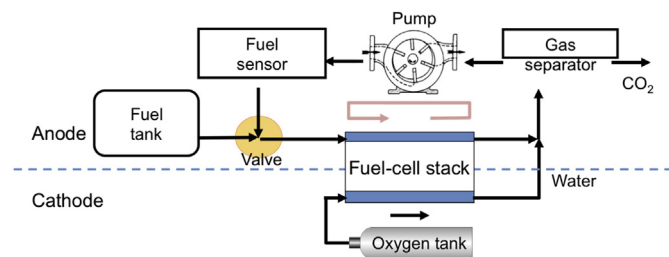


Fig. 1. A fuel-cell system needs ancillary components to provide and regulate the fuel and oxidant in addition to the fuel-cell stack. In a miniaturized configuration, these supporting components take up significant portions of the total volume, posing a serious limitation against miniaturizing the system.

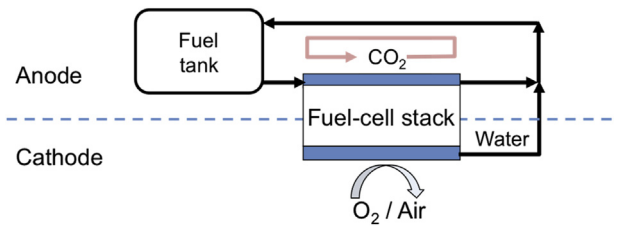


Fig. 2. Simplified fuel-cell system previously developed from our group [23]. Oxygen is provided from the ambient air.

pressurized oxygen tank with a regulator. Elimination of the oxygen tank or the need to access the ambient air allows the fuel-cell system to be designed as a standard unit that can be easily stacked.

2. Mechanism

The fuel-cell system reported in this paper, schematically illustrated in Fig. 3, was designed to integrate two main technologies—self-regulated fuel pumping and self-regulated oxygen supply—into one device. The top portion marked A, consisting of a fuel cartridge and a fuel channel, is essentially an air-breathing fuel-cell system developed in Ref. [23] and shown Fig. 2, which self-pumps the fuel in a self-regulating manner. The bottom portion marked B is an oxygen supply cartridge, which supplies oxygen directly to the gas diffusion electrode (GDE), i.e., the cathode, of the fuel channel in a self-regulating manner. The O₂-supply cartridge was developed to solve the problem of mixed potential in the O₂-generating surface in our preliminary study [25], as detailed in the Supplementary.

2.1. Working mechanism of the fuel-cell system

In the top portion A of Fig. 3, the fuel in the fuel channel is pumped by directional growth during the fuel-cell reaction, following [22]. When the fuel-cell circuit is closed to draw the

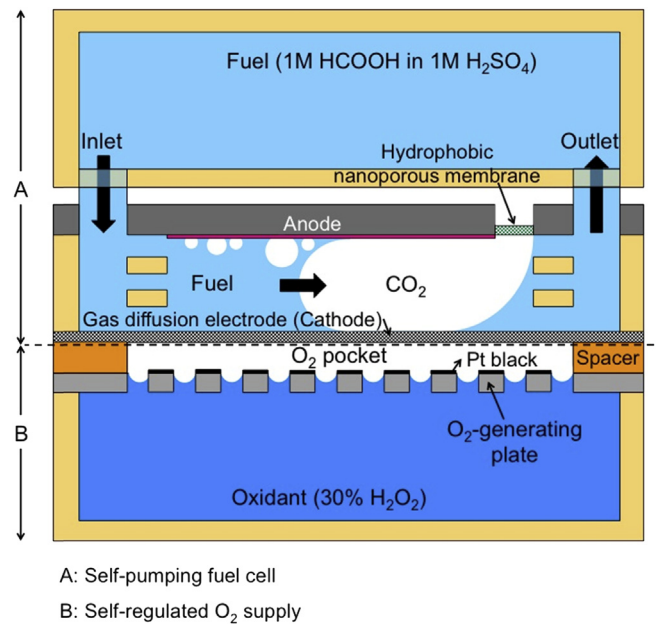


Fig. 3. Schematic description of the proposed fuel-cell system, which integrates two technologies into one device: top portion marked A is the membraneless, self-pumping, air-breathing fuel-cell system [23] and the bottom portion marked B is the self-regulated, oxygen-generation mechanism inspired by [25].

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