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Key issues in the microchemical systems-based methanol fuel processor: Energy density, thermal integration, and heat loss mechanisms

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Abstract

Microreactor technology is a promising approach in harnessing the high energy density of hydrocarbons and is being used to produce hydrogenrich gases by reforming of methanol and other liquid hydrocarbons. However, on-demand H_2 generation for miniature proton exchange membrane fuel cell (PEMFC) systems has been a bottleneck problem, which has limited the development and demonstration of the PEMFC for highperformance portable power. A number of crucial challenges exist for the realization of practical portable fuel processors. Among these, the management of heat in a compact format is perhaps the most crucial challenge for portable fuel processors. In this study, a silicon microreactorbased catalytic methanol steam reforming reactor was designed, fabricated, and demonstrated in the context of complete thermal integration to understand this critical issue and develop a knowledge base required to rationally design and integrate the microchemical components of a fuel processor. Detailed thermal and reaction experiments were carried out to demonstrate the potential of microreactorbased on thermal characterization experiments, the heat loss mechanisms and effective convective heat coefficients from the planar microreactor structure were determined and suggestions were made for scale up and implementation of packaging schemes to reduce different modes of heat losses.

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1. Introduction

As fuel cell (FC) gain attention as potential energy solution of the future, one of the most promising fields of application is portable power, as a prominent alternative to batteries [1]. Batteries are a multi-billion dollar market and the technology has made significant strides in the last couple of decades to meet growing power demand of advanced portable computing, handheld personal devices, and other microelectronics systems. The rechargeable battery technology has progressed from nickel cadmium (NiCD) to nickel meta hydride (NiMH) to lithium ion (Li-Ion). Until recently, the gradual improvement of Libased batteries was enough to power mobile computing devices. Unfortunately, the energy density available from current battery technology is inadequate to match the power demands

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of emerging portable devices [2]. This need has encouraged researchers to continue to vigorously explore FCs as miniature power sources.

High energy density is required for advanced portable applications for consumer use (e.g., laptop computers, PDAs, cellular phones, medical and telecommunication devices), and for military and intelligence use (e.g., remote sensors (<20 W), systems for the soldier (<500 W), silent power generation and battery recharging (200 W–2 kW), and mobile power generation and auxiliary power units (>2 kW)) [3], with the goal of allowing these devices to operate for longer times with less recharging. Miniatures FCs offer promise for the conversion of chemical energy into electrical energy in a highly efficient way achieving the goals of power dense, light weight, and compact power sources.

The proton exchange membrane fuel cell (PEMFC) is particularly attractive and promising for portable applications because of its ability to provide high power density and nearly instant power. Moreover, its simplicity in design and operation allow-

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Nomenclature	
Α	surface area (m ²)
$A_{\rm w}$	cross-sectional area of the wire (m^2)
с _{MeoH}	methanol concentration in feed (mol cm $^{-3}$)
d_{h}	hydraulic diameter (m)
D_{AB}	diffusivity of methanol in gas mixture $(m^2 s^{-1})$
Ε	Emissive power (W m^{-2})
$E_{\mathbf{b}\lambda}$	spectral blackbody emissive power
	$(W m^{-2} \mu m^{-1})$
h	heat transfer coefficient (W m ^{-2} K ^{-1})
Ι	current (A)
I_{λ}	radiative intensity (W m ^{-2} s ^{-1})
kg	mass transfer coefficient (m s ^{-1})
$k_{\lambda}^{\mathrm{Py}}$	spectral absorption coefficient of Pyrex TM (m ⁻¹)
$k_{\lambda}^{\hat{S}i}$	spectral absorption coefficient of silicon (m^{-1})
l	length of the wire (m)
N _{Nu}	Nusselt number
$N_{\rm Ra}$	Rayleigh number
P'	perimeter of the wire (m)
$Q_{ m conv.}$	heat loss by convection
$Q_{ m in}$	heat input (W)
$Q_{ m loss}$	total heat loss (W)
$Q_{\mathrm{rad.}}$	heat loss by radiation
$Q_{ m wires}$	heat loss from the attached wires
<i>r</i> _{MeOH}	methanol reaction rate (mol cm ^{-3} s ^{-1})
Sh	Sherwood number
$T_{\rm s}$	surface temperature (K)
T_{∞}	ambient temperature (K)
V	voltage (V)
Greek letter	
α	constant used in equation (5)
~	

ing fabrication of a compact and lightweight cell, mild operating conditions (temperature less then $100 \,^{\circ}$ C, pressure in the range of a few atmospheres), and relatively low loading of precious metal catalysts are additional strong advantages [1,4].

The success of PEMFC technology for portable power depends heavily on the development of efficient ways of delivering the appropriate fuel to the cell. The PEMFC operating on hydrogen as the fuel holds the most promise, offering the best possible efficiency and power density [5,6]. However, while hydrogen offers high energy density on a per weight basis, it has a poor energy density per volume. Gaseous hydrogen is therefore difficult and inefficient to store and several high energy density storage methods have been investigated such as highly pressurized cylinders, metal hydrides, chemical hydrides, and even carbon nanotubes [7,8]. Since on-board storage of hydrogen is unsafe and energy inefficient, two main alternatives are commonly envisioned and employed for portability.

One approach, the direct FC employs fuels like methanol, ethanol, formic acid, etc., directly in the cell to generate electricity. The direct FC has the advantage of a smaller total system and weight, as it does not need an in-situ fuel processing (FP) unit to extract hydrogen. Among all direct FCs investigated at the portable scale, more research and development efforts are focused on the development of direct methanol fuel cell (DMFC). Several groups are pursuing low power DMFCs for portable power applications [9]. Although the DMFC offers promising possibilities as a miniature power source, the technology has to overcome a number of challenges to be competitive with battery technology in terms of efficiency, power density, and cost. Methanol crossover and the relatively slow reaction rate of methanol oxidation to hydrogen ions and carbon dioxide at the anode are persisting challenges that currently limit the DMFC to less stringent applications. In addition to DMFC, the direct ethanol fuel cell (DEFC) and direct formic acid fuel cell (DFAFC) are also being developed. Ethanol is considered an attractive fuel mainly because of its higher theoretical energy density than methanol, non-toxicity, availability, and renewability [10,11]. However, the available power density from DEFC is currently about half that of DMFC resulting from relatively slower electrooxidation reaction kinetics at the anode. Recently, the DFAFC has been demonstrated as an attractive alternative [12]. The theoretical energy density of formic acid is less than one-third of methanol. However, formic acid has other advantages like faster anode kinetics and smaller crossover through Nafion membranes. Unlike DMFC where methanol crossover limits the use of higher methanol concentration solutions, it is possible to run the DFAFC with high concentrations of formic acid. However, it was recently observed that the performance of the cell dropped as the formic acid concentration was increased beyond 10 M because of decreases in anode catalytic activity and an increase of the fuel crossover [13].

The other, higher performance alternative is to generate pure hydrogen on site and on demand by processing easily stored, high energy density liquid fuels [14]. FP adds complexity, weight, volume, and cost compared to the DMFC or a system running directly on hydrogen, however the effective energy storage density is significantly higher.

A wide variety of energy dense hydrocarbons such as natural gas, methanol, gasoline, and diesel can be reformed into hydrogen-rich streams for FCs. Different processes are available for hydrogen extraction and subsequent purification, therefore, the type of hydrocarbon and method used to produce the hydrogen play a major role in the design of a FP system. Processing of liquid hydrocarbons to hydrogen involves a series of steps including fuel vaporization, reforming (three main processes available are partial oxidation, steam reforming (SR), and autothermal reforming), water gas shift (WGS), and CO clean up (preferential oxidation (PrOx), hydrogen separation by permeation through a hydrogen-selective membrane, or pressure swing adsorption) before feeding hydrogen rich gases to the FC.

Methanol is the fuel of choice for mobile portable power application as it is sulfur free and can be reformed at relatively low temperatures (250–300 $^{\circ}$ C), simplifying the micro FP design. Moreover, it offers other advantages like high hydrogen–carbon ratio, high energy density, and ready availability.

The following chemical reactions take place during the SR process.

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