

MICROFLUIDIC FUEL CELLS THAT LACK A PEM

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ABSTRACT

Microscale systems research over the last decades has focused on miniaturization of their functional components, leaving miniaturization of power sources as one of the major challenges. This paper discusses research in the area of micro fuel cell power sources for which there is an increasing societal demand. The specific fuel cell type utilizes a concept unknown to the fuel cell community: multi-stream laminar flow. Such a membrane-free fuel cell system will avoid several of the technical issues related to the use of proton exchange membranes as used in conventional fuel cells. The physicochemical phenomena that govern chemical conversion and all accompanied energy and mass transport phenomena in our laminar flow-based microfuel cell will be discussed. Based on our current fundamental understanding of these characteristics we will also discuss the promise of a membrane free fuel cell.

INTRODUCTION

The study and development of microscale systems is an emerging area in the physical, chemical, and engineering sciences. Most research over the last decades has focused on miniaturization of the functional components of these microsystems, leaving miniaturization of power sources as one of the major challenges. This situation is rapidly changing: both researchers and funding agencies have recognized the need for high-energy microscale power sources as replacements for batteries in portable electronics, clinical and diagnostic devices, micro-analytical devices, and in a wide variety of Micro-Electro-Mechanical Systems

(MEMS). In this respect, (micro) fuel cells, power sources that allow for the direct conversion of the chemical energy stored in liquid fuels into electrical energy, have received a lot of attention, in particular those that are based on liquid fuels. Liquid fuels such as methanol and formic acid have a much higher energy density than solid fuels such as those used in Li-ion batteries.

In conventional fuel cells, a Proton-Exchange Membrane (PEM) keeps the streams of fuels separated while they react in the cathodic and the anodic compartments. In addition, this membrane allows for the diffusion of protons from the anode to the cathode to complete the electrochemical process. The fuel cell type that we present here utilizes the unique characteristics of fluid flow at the microscale. Two differently dyed aqueous streams that are brought together in the same channel will proceed to flow

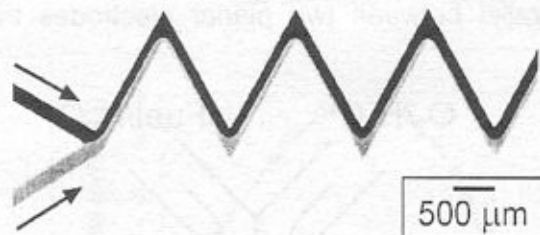


Figure 1. Multistream Laminar Flow in a microchannel: aqueous streams with contrasting dyes.

laminarily in parallel due to the lack of turbulence at low Reynolds number flow [1] (Figure 1). The only remaining mechanism of mixing is molecular diffusion across the liquid-liquid interface between the two streams [2]. This concept of multistream laminar flow forms the basis for a fuel cell that lacks a

PEM membrane. Such a membrane-less fuel cell system avoids several of the technical issues related to the use of PEM membranes in microfuel cells [3-7]. Fuel crossover can be avoided by controlling convective time scales with respect to diffusive time scales. By adjusting channel dimensions and flow rates, the residence time can be manipulated so that the fuel will not diffuse to the cathode and hinder cell performance. Operation at elevated temperatures is possible without fear of membrane dry-out. There will be no problems with water removal since this is a flowing system. There will be no issues with maintenance of the liquid membrane as it is created instantaneously upon start of fluid flow, and will therefore be cheaper in fabrication.

Fuel Cell Design and Fabrication

The dimensions of our micro fuel cell system are such that the fluid flow is characterized by a low Reynolds number and is thus laminar [1]. Typical channel widths and heights range from 100 to 1000 μm . The system consists of a Y-shaped microfluidic channel design (Figure 2), which allows two liquid streams containing fuel and oxidant to merge and continue to flow laminarily in parallel between two planar electrodes that

line the opposing channel walls. The electrodes are covered with catalyst (Figure 2). The two liquid streams are in diffusive contact with each other, allowing for H^+ ions to diffuse across the channel as is necessary for the electrochemical process.

The Y-shaped microfluidic channels are fabricated following a rapid prototyping methodology based on replica molding as developed by Whitesides and coworkers [8,9]. First, a master of the Y-shaped channel system is made in photoresist (SU-8, 100-1000 μm high 100-1000 μm wide) by photolithography, using a high-resolution transparency film as the mask. Second, this negative-relief master is replicated by molding in an elastomeric rubber, such as

polydimethylsiloxane (PDMS). The resulting PDMS mold is replicated, again via molding, into a chemical resistant material membrane. The resulting membrane forms the centerpiece of the microfluidic system as it defines the dimensions of the Y-shaped channel (Figure 3). A metallic seed layer is applied to the sidewalls of the channel system carved out in the chemically resistant membrane by evaporative deposition. Then the catalytic layer is applied on the metallic seed layers by whichever method is appropriate for the catalyst at hand. Finally

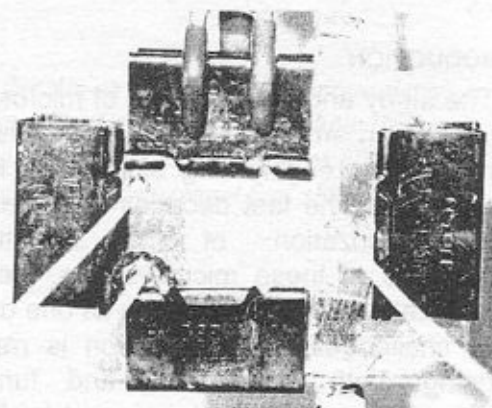
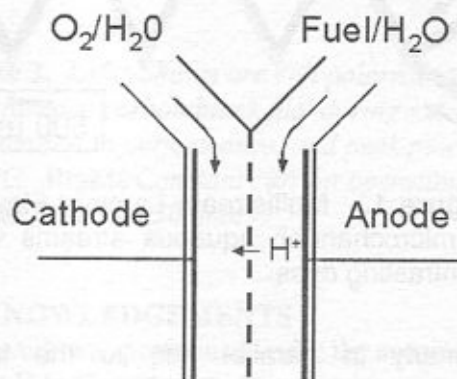


Figure 2. *Left:* Schematic of laminar flow-based fuel cell: Two streams, one containing 5–10% fuel and one saturated with oxygen or other oxidant, convene in a microfluidic channel and flow over the cathode and anode. *Right:* An optical micrograph (top view) of the actual prototype laminar flow-based fuel cell. The electrodes as well as the tubes going into the microfluidic system can clearly be seen.

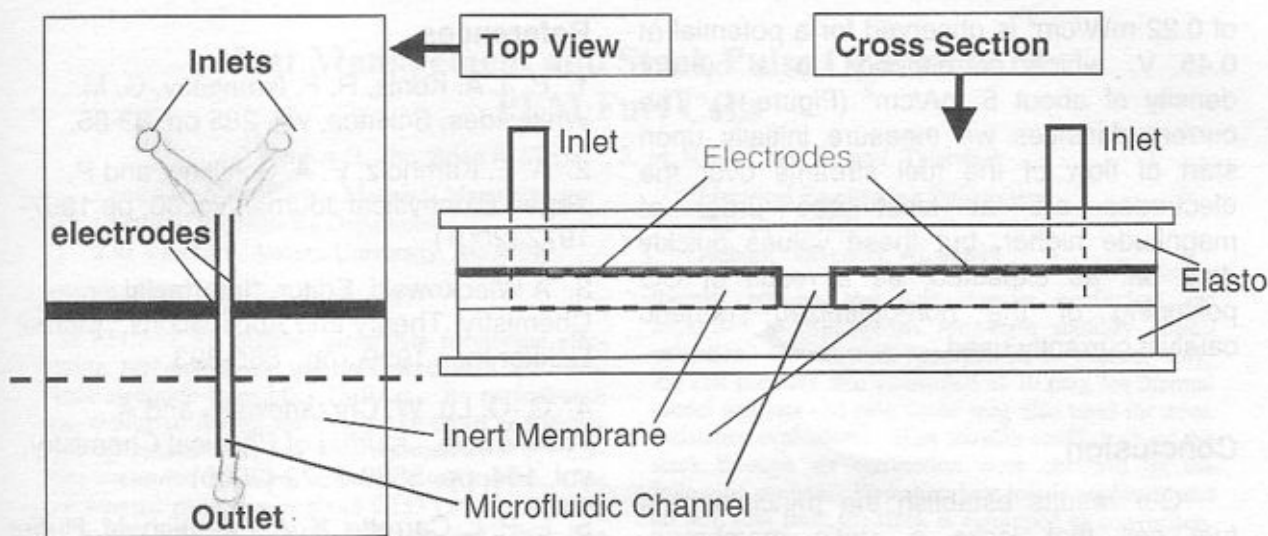


Figure 3. Fabrication and stacked assembly of Y-shaped microfluidic fuel cell system. Centerpiece is a polyurethane layer that forms the sidewalls of the Y-shaped channel. These sidewalls carry both the anode and the cathode (in red).

the membrane, now carrying the two electrodes, is clamped between two slabs of rubber to form the top and bottom walls of the microfluidic channel system. Figure 3 shows a cross-section of the microfluidic fuel cell as assembled by stacking of the different layers. These microfluidic systems can be easily connected to microsyringes via polyethylene tubing. In this prototype, precise control over fluid flow is achieved by placing this microsyringes in a syringe pump. Fluid flow can eventually also be driven by other methods in order to reduce the amount of energy needed to run the fuel cell system.

Results

Figure 4 shows preliminary performance data, a load curve and a power density curve, of the prototype laminar flow-based microfuel cell system shown in Figure 2. The shape of both curves are identical to the shape typically observed for conventional PEM-based fuel cells indicating similar behavior. Using aqueous streams of fuel and oxidant dissolved in water measured potentials range from 0.2 to 1.0 V depending on the load applied (Figure 4). A maximum power density

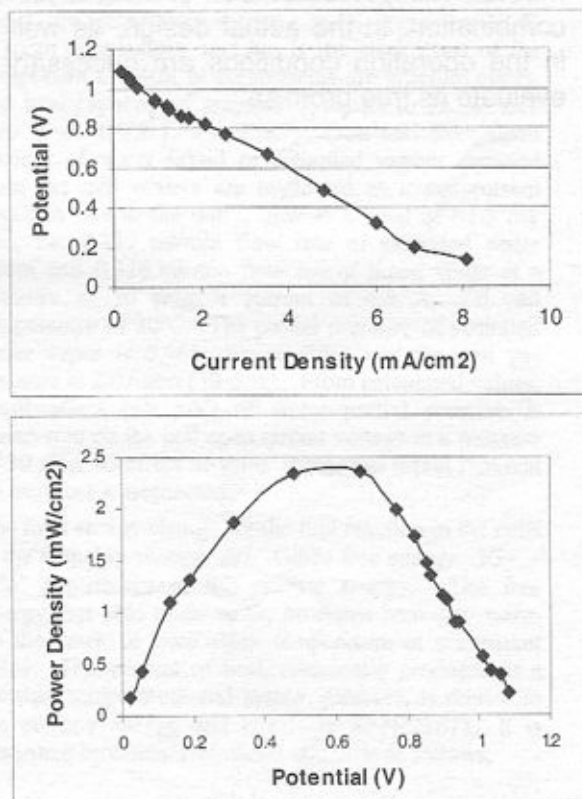


Figure 4. Preliminary data: load curve (top) and power density curve of a laminar flow-based microfuel cell using aqueous streams of fuel and oxidant as the respective streams.

of 0.22 mW/cm^2 is observed for a potential at 0.45 V , which corresponds to a current density of about 5 mA/cm^2 (Figure 4). The current densities we measure initially upon start of flow of the fuel streams over the electrodes are at least one order of magnitude higher, but these values quickly drop off, as expected, as a result of CO poisoning of the non-optimized, generic catalyst currently used.

Conclusion

Our results establish the principle of a fuel cell that lacks a static membrane. Whether laminar flow based fuel cells are going to be competitive with or superior over existing microfuel cell technology such as PEM-based fuel cells is unclear at this point. Further improvements in catalyst/fuel combination, in the actual design, as well as in the operation conditions are necessary to evaluate its true promise.

References

1. P. J. A. Kenis, R. F. Ismagilov, G. M. Whitesides, *Science*, vol. 285 pp. 83-85.
2. A. E. Kamholz, E. A. Schilling, and P. Yager, *Biophysical Journal*, vol 80, pp 1967-1972 (2001)
3. A. Wieckowski, Editor, "Interfacial Chemistry: Theory and Applications," Marcel Dekker Inc., 1999. pp. 885-893.
4. G.-Q. Lu, W. Chrzanowski, and A. Wieckowski, *Journal of Physical Chemistry*, vol. 104, pp. 5566-5572 (2000)
5. L. P. L. Carrette, K. A. Friedrich, M. Huber and U. Stimming, *Phys. Chem. Chem. Phys.*, vol 3, pp. 320-324, (2001)
6. L. Carrette, K. A. Friedrich, U. Stimming, *Chem. Phys. Chem.*, vol. 1, pp. 162-193 (2000)
7. S. Wasmus and A. Kuver, *Journal of Electroanalytical Chemistry*, vol 461, pp. 14-31 (1999)
8. Y. Xia and G. M. Whitesides, *Angew. Chem. Int. Ed.*, vol 37, pp. 550-575 (1998)
9. D. C. Duffy, J. C. McDonald, O. J. A. Schueller, and G. M. Whitesides, *Analytical Chemistry*, vol. 70, pp. 4974-4984 (1998)
10. G.-Q. Lu, A. Crown, and A. Wieckowski, *Journal of Physical Chemistry*, vol 103, pp. 9700-9711 (1999)