

ADVANCES IN DIRECT METHANOL FUEL CELL SCIENCE & TECHNOLOGY AT LOS ALAMOS NATIONAL LABORATORY

Shimshon Gottesfeld*, Xiaoming Ren, Piotr Zelenay, Huyen Dinh, François Guyon and John Davey

Los Alamos National Laboratory
Los Alamos, New Mexico 87545, USA

This paper describes recent advances in work on direct methanol fuel cells (DMFCs) at Los Alamos National Laboratory (LANL). The effort on DMFCs at LANL includes work on potential portable power applications, supported by the Defense Advanced Research Project Agency (DARPA), and work on potential transportation applications, supported by the US DOE. We describe results obtained with DMFC stack hardware of cell width limited to 2 mm, that allows operation with low air flow and at low air pressure drops. A 30-cell stack of 45-cm² active area has been fabricated and tested. Power densities of 300 W/l and 1 kW/l (of active stack volume) seem achievable under conditions applicable to portable power and transportation application, respectively. DMFCs with significantly lower catalyst loadings have been demonstrated showing maximum power density loss of only 20-30% as the catalyst loading is lowered by an order of magnitude. A 100 mW air breathing DMFC has been fabricated and tested demonstrating 3000 hours of continuous operation. Such air breathing DMFCs are being further developed for applications in consumer electronics.

Introduction

Most, if not all recent DMFC work has strongly focused on cells with polymeric, primarily perfluorocarbon sulfonic acid (PFSA) membrane electrolytes. In work at LANL, thin film catalysts bonded to the membrane, by a decal method or direct application to the membrane, provided best results in terms of catalyst utilization and overall cell performance [1]. Recently, machined graphite hardware has been replaced by alternative, non-machined flow-field/bipolar plate hardware, which enables effective air and aqueous methanol solution distribution along the cell active area at reduced cell width of just 2 mm. We describe here development and testing of DMFC technology at LANL in the contexts of portable power sources, consumer electronic devices and potential transportation applications.

Experimental

(1) DMFC Short Stack Fabrication & Testing: In 1999, we have moved, under DARPA sponsorship, to the fabrication of a small DMFC stack to be incorporated by Ball Aerospace into a portable power system. This effort targets a 50W/160Wh DMFC power source that could potentially replace the "BA5590" primary lithium battery, used by the US Army in communication systems. At this point, we have assembled and tested a 30-cell DMFC stack of 45-cm² active area. Figure 1 presents a photograph of the 30-cell stack assembled and tested at LANL. In operation on 0.5M methanol and air, peak power near 80W was obtained at 60°C (at 14V), with ambient air stoichiometric flow of 3x. An important feature of this DMFC stack technology is a narrow width ("pitch") per cell of 2 mm, achieved while ensuring minimized pressure drop across the stack. The tight packaging has generated, under such benign operation conditions, an effective power density of close to 300 W per liter of active stack volume;

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Assuming that the weight of the auxiliaries is twice the weight of methanol fuel in a power system designed for 10 hours of operation, system energy densities in excess of 200 Wh/kg are projected [2]. At this level of energy density, DMFC power systems would compete favorably with advanced Li batteries, while providing the important advantage of ease of rechargeability.

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(2) *On Methanol Crossover* The effects of methanol crossover have been considered a severe barrier to faster development of DMFC technology. The two tools available to achieve lower methanol crossover are lower methanol feed concentration and optimized cell design [3]. We showed [3] with such optimized cell structures, that the required combination of high cell performance (close to 0.2 A/cm^2 at 0.45 V) and high fuel utilization ($> 90\%$) can be achieved, in spite of the rather "leaky" membrane employed. Water management remains, however, an important requirement in such a portable, liquid fed DMFC power system, because of the need to supply the anode with methanol of concentration not exceeding 1M in order to minimize crossover. At the DMFC system level, this challenge can be answered by returning liquid water from the cathode exhaust to anode inlet, using a neat methanol source and a methanol concentration sensor to keep methanol concentration in the anode feed at the appropriate level.

(3) *DMFCs as Potential Power Sources for Potential Transportation Applications* Introduction of a DMFC as primary power source for transportation, has the potential to achieve the combined attractive properties of a liquid fuel of good potential availability, high system simplicity ("liquid fuel + air *in*, DC power *out*"), good potential for packaging as required to achieve 350 miles range in a passenger vehicle, and a good potential for ZEV characteristics. Our approach in working on potential transportation applications of DMFCs, has been based on raising the DMFC temperature to around 100°C [1, 3] to achieve competitive power densities. To date, we have demonstrated operation near 100°C of a DMFC, 5-cell short stack based on LANL stack hardware, operating on aqueous methanol and pressurized (30 psig) air. Maximum power density generated by the stack under these conditions was 50 W, corresponding to 1.1 W/cm^3 of active volume, or 1 kW/liter of active volume. For operation of this stack at 100°C , we measured, by detailed mass balance, methanol utilization rate of 95% with an anode feed of 0.75 M . This high fuel utilization was achieved by optimizing cell structure, rather than by significant membrane modification. An overall conversion efficiency, methanol chemical energy to DC power of 37% could consequently be achieved near 80% of the DMFC short stack peak power.

To address the important issue of cost, we have further pursued recently the target of lowering catalyst loadings in DMFCs while maintaining performance. Figure 2 illustrates DMFC results obtained near 100°C with relatively lower loadings of Pt. To achieve better catalyst utilization, we reexamined carbon supported PtRu anode catalysts. The state-of-the-art catalyst technology for direct and indirect methanol fuel cell systems can be roughly described as a ratio of 2.5:1 between DMFC stack and (overall system) reformate/air precious metal loadings demonstrated to date per kW. While certainly significant and a future target for further effort, this gap is smaller than usually perceived. We also refer the reader to a recent work performed at LANL [4], showing that maximized surface area together with maximized population of *metal* alloy surface sites of composition close to $\text{Pt:Ru} = 1:1$, is apparently the key for higher DMFC anode catalyst activity.

(4) *Potential Applications for Consumer Electronics:* To allow consideration for some consumer electronics applications, we recently demonstrated successfully operation of our DMFCs in "air breathing" mode. In this mode of operation, oxygen is supplied to the cathode only by diffusion from surrounding air, i.e., no active flow of air is applied. Figure 3 shows such an air-breathing MFC, comprising of two cells connected in series, which provided around 10 mW per cm^2 of MEA area when operating on 0.5 M methanol and breathed air. In a test of this cell at Motorola, the voltage was up-converted electronically to provide the required voltage level for charging the battery in a cellular phone. Continuous testing of this cell for 3000 hours at Motorola (1999) demonstrated the viability of DMFC operation in passive mode.

References

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- [4] H. Dinh, X. Ren, F. H. Garzon, P. Zelenay and S. Gottesfeld, *J. Electroanal. Chem.*, in press.

Figures

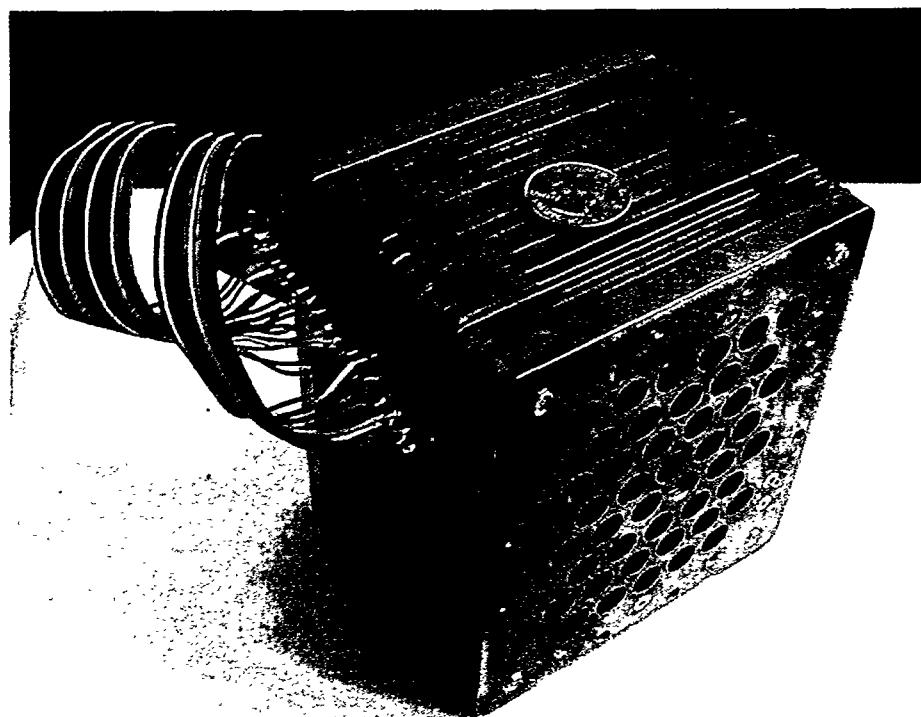


Figure 1: Photograph of 30-cell DMFC stack of 45 cm^2 active area, fabricated and tested recently at LANL.

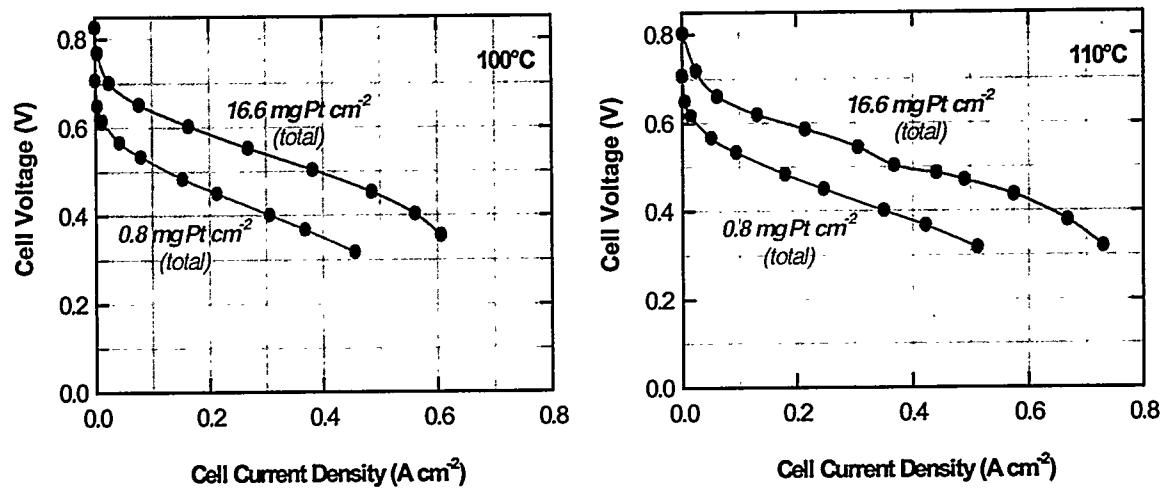


Figure 2: Favorable trade-off between (total) DMFC catalyst loading and cell performance. Unsupported PtRu is used in the high loading cases shown and carbon-supported PtRu in the cases of low catalyst loading shown.

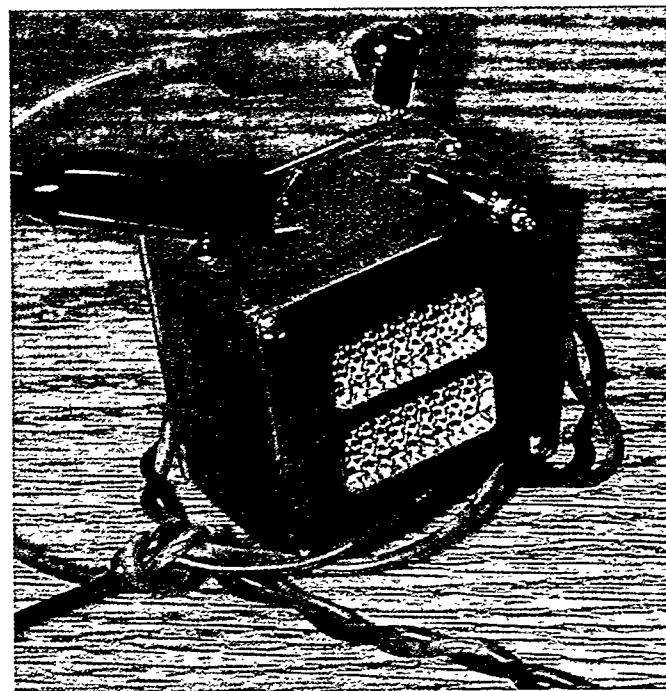


Figure 3: Air Breathing DMFC that demonstrated 3000 hours of continuous operation.