

# Direct Methanol Fuel Cells for Automotive Power Systems

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## ABSTRACT

Direct fuel oxidization yields major fuel cell power system simplifications and potential performance advantages, particularly for an automotive power plant application. The system simplification is particularly striking when a direct fuel system is compared to an "indirect" fuel cell system in a vehicle (where the fuel on board must be "reformed" and "cleaned" to provide a hydrogen-rich gas "reformate" for use in the fuel cell system). The inherent complexity, the losses of efficiency, and the emissions associated with the fuel processor required for the indirect system combine to make the comparison to a direct fuel system extremely favorable toward the latter in all important aspects.

Although direct fuel oxidization is possible in principle for almost any hydrocarbon or alcohol fuel, at the present time the direct fuel cell system with the highest levels of system fuel efficiency and power density is the direct-hydrogen system. However, the use of hydrogen as a vehicle fuel (compressed, adsorbed, or liquefied), has one overwhelming disadvantage – the problem of effectively storing the hydrogen on-board the vehicle (added volume and weight, for example, which inhibit acceleration performance and efficiency, and intrude on passenger and payload space). These disadvantages largely negate the cell/stack advantage of a direct-hydrogen system.

In contrast, the major system simplifications and potential performance advantages of a direct fuel cell system are available, essentially without significant disadvantage, if the fuel cell system can directly use a high-energy-density liquid fuel (e.g.; an alcohol or hydrocarbon). The R&D and commercialization challenge is to develop a liquid-fueled direct fuel cell system for automotive applications which has adequate levels of fuel conversion efficiency and power density.

Within the existing technical limitations of the direct fuel cell state-of-the-art (especially the available catalysts and electrolytes), there is only one liquid fuel with sufficient reactivity to use directly in a fuel cell – that fuel is methanol (Methyl Alcohol, CH<sub>3</sub>OH, MeOH). The status and future potential for the Direct-Methanol Fuel Cell (DMFC) is an important consideration in evaluating the overall future commercial possibilities for all Fuel Cell

Vehicle (FCV) designs, and for understanding the potential long-term – and transitional -- role of methanol as an FCV Fuel. The state-of-the-art for DMFCs is reviewed here, and the issue of operation in a *load-following* vs. a *hybrid* powersystem are evaluated, together with some indications of future improvements for the DMFC. A major consideration is that the conventional wisdom that a DMFC stack *must* be operated in a hybrid power system is based on a fundamental misinterpretation of the operating characteristics of the DMFC.

## INTRODUCTION

Historically, low power density and low energy conversion efficiency (primarily caused by low fuel utilization and/or low voltage efficiency, depending on the specific DMFC type) have kept the DMFC confined to the research laboratory. However, very significant recent progress – using the polymer-electrolyte-membrane (PEM) version of the DMFC, combined with highly diluted solutions of methanol as the fuel – has revived industrial interest in the DMFC. This interest is almost exclusively focused on potential automotive applications, because of the compelling advantage for the direct use of a liquid fuel in vehicles.

The possibility of an automotive application for the DMFC depends primarily on two factors:

- system fuel conversion efficiency across a broad spectrum of power density levels,
- system power density (where the DMFC stack power density is a dominant factor for the system power density).

Certainly there are potential applications for DMFC power systems that can only be operated efficiently at relatively fixed power, or in "hybrid" power systems – even for automotive applications. However, if the potential use of the DMFC is limited solely to hybrid systems, this greatly restricts its eventual share of the vehicle powertrain market.

Although it is frequently stated that the DMFC cannot operate efficiently across the wide dynamic range of power levels demanded for automotive power systems applications, and therefore must be used in a hybridized power system – *this is incorrect*.

As will be discussed later -- it is clear that it is in fact quite realistic to design a DMFC powertrain for operation in a *load-following mode* over the wide dynamic power range needed for automotive applications. This conclusion is based on an analysis of the present experimental state-of-the art, and on a basic theoretical understanding of the DMFC cell/stack characteristics and optimum operation strategy. The conventional wisdom that a hybrid powertrain design is required for a DMFC stack is fundamentally incorrect.

Following from that understanding, and based on the clear (but as yet unrealized) potential for major improvements in both DMFC power density and energy conversion efficiency, major R&D efforts on DMFC cells and stacks (for automotive and other applications) are now being carried forward vigorously in many locations worldwide. Participants include: US National Laboratories (JPL, LANL), industrial fuel cell developers (Ballard, Siemens), and major automotive developers of FCVs (e.g.; DaimlerChrysler and others).

The eventual commercial success of these many efforts will only come about as the result of many parallel advances in technology and process development (e.g.; catalyst, membrane, other enabling materials, fabrication processes, etc.). However, all of the advances that must be achieved are captured within the two key performance metrics cited above -- *system fuel conversion efficiency* and *system power density*.

Following a brief introduction to the unique characteristics of the PEM-based DMFC (compared to direct-hydrogen and the various indirect fuel cells), these two performance metrics for the DMFC will be examined in more detail. The following summary is a *technical assessment* of the DMFC. For opinions on *commercialization potential* -- including costs and the many other non-technical aspects of the DMFC, many widely varying views are available.

Although these issues will not be discussed in this review, for a general background on the non-technical issues surrounding the DMFC -- and contrasting views on its future potential for automotive applications -- the two most extreme positions are represented by:

- The 1998 FCTAP (Fuel Cell Technical Advisory Panel) Report to the California Air Resources Board,<sup>13</sup>
- The American Methanol Institute,<sup>14</sup>

The former is very pessimistic on the future potential for DMFCs, and the latter very sanguine -- as one might reasonably expect for the latter. Opinions on the commercialization potential, costs, and other non-technical aspects of the DMFC will not be offered in the following technical assessment.

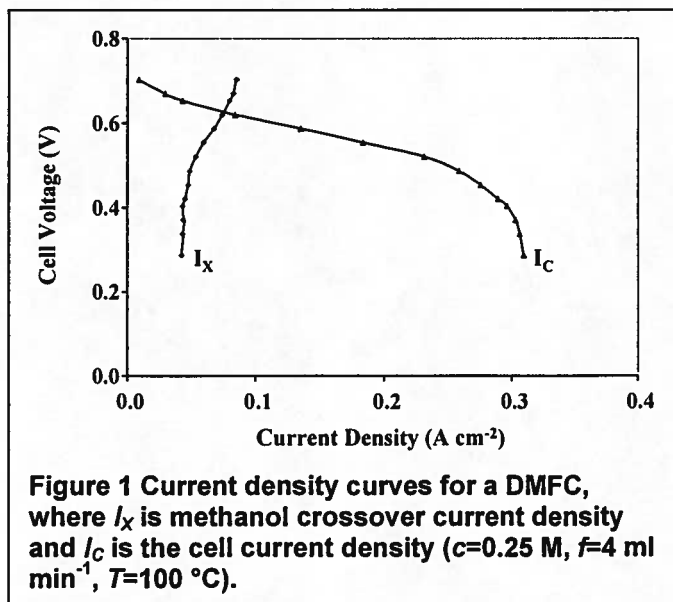
## AN OVERVIEW OF DMFC CHARACTERISTICS

A unique feature of the PEM-based direct-methanol PEM fuel cell -- when compared to a direct-hydrogen or a reformate fuel cell -- is the significant crossover of unreacted methanol from the anode through the PEM to the cathode catalyst layer.<sup>15</sup> This methanol crossover is greater (by 1-2 orders of magnitude) than the corresponding flux of unreacted hydrogen across the

PEM in the Direct-Hydrogen Fuel Cell (DHFC). The most critical result of this "crossover current" in a DMFC is a significant fuel efficiency loss due to this unproductive methanol consumption mechanism (that does not result in generation of cell current density or cell power). The methanol crossover also has a directly negative impact on the voltage efficiency of the DMFC by increasing polarization at the air electrode (fuel cell cathode).

It is conventional to treat methanol crossover analytically within the classic electrochemical framework of a "fuel utilization" parameter, while correspondingly dealing with the cell polarization curve (cell voltage vs. cell current) separately in terms of "voltage efficiency" parameter. This approach is commonly taken for reformate PEM fuel cells, and for other types of fuel cells (solid oxide, for example). However, using such an approach for the PEM-based DMFC actually disguises the strong physical interaction between the cell current and the methanol crossover current (methanol crossover expressed as equivalent current density of MeOH) in a DMFC. Such an artificial, and non-physical, separation of the crossover completely obscures the possibilities for optimizing cell current and power density vs. the methanol crossover losses -- and leads to many erroneous conclusions about the DMFC, including the conventional wisdom that the DMFC must be operated in a hybrid powertrain configuration.

A conceptually better viewpoint is to view the crossover current density together with the cell current density and cell voltage -- in terms of a single "composite" polarization curve. One example of this type of composite polarization curve is shown in **Figure 1**, for a particular set of experimental conditions and a particular cell design and materials.



The cognitive value of this composite polarization curve is that it explicitly demonstrates the physical interaction between the cell and crossover current densities. The composite polarization curve can be used in either of two ways to determine the interdependence among cell voltage, cell current density, and methanol crossover current density:

- Enter the graph vertically at a value of cell current density ( $I_c$ ), read the corresponding cell voltage horizontally, and then, from the intersection of this horizontal line with the  $I_x$  curve (i.e.; at the same cell voltage), read down to the current density axis again to find the corresponding crossover current density loss (dependent on the original value of cell current density used);
- Enter the graph horizontally at a value of cell voltage, and read off the corresponding values of  $I_c$  and  $I_x$  from the appropriate intersections with the respective curves.

In this way the explicit interaction between the cell and crossover current density is clearly seen.

The cell current density ( $I_c$ ) and crossover current density ( $I_x$ ) curves in Figure 1 allow one to predict the behavior of a direct-methanol cell under variable cell current -- for example specifically illustrating the characteristic impact of increasing cell current on crossover current losses. This characteristic impact is that when the cell current increases the crossover current decreases which means that the fuel utilization increases -- in Figure 1 to almost 90% at about 0.3 V,  $0.31 \text{ A cm}^{-2}$ . In contrast (again for Figure 1), where the  $I_c$  and  $I_x$  curves cross (about 0.6 V in this figure) the fuel utilization is only 50%, and for the lowest cell current density (highest cell voltage) shown in Figure 1 the fuel utilization falls to roughly 10%.

Clearly the latter two values of fuel efficiency are unacceptable, and it is obvious that -- for a cell with the specific cell voltage-current curve shown in Figure 1 -- there is an optimum operating strategy. An intelligent designer would choose to operate at the highest possible cell current (maximum cell power density) in order to reduce the impact of the methanol crossover losses on cell conversion efficiency. This has led to the cited conventional wisdom that the DMFC *must be operated within a "hybrid" power system* -- i.e., where the fuel cell is only operated at its highest power density (as an energy source). Unfortunately this choice leads to the need for another electrical power source (e.g.; a battery) to provide the dynamically varying power levels required for a vehicle -- with accompanying problems of system control and optimization.

Fortunately, in reality the choice of a hybrid power system is actually not necessary -- since there is actually no fundamental reason to limit the operation of a DMFC stack to constant power within a hybrid power system configuration. In fact, as discussed below, high fuel utilization and system fuel efficiency can be achieved over a broad dynamic range of power densities -- using a fuel control strategy to be outlined in the next section -- without resorting to additional dynamic power devices. In general, using the DMFC in a "load-following" configuration -- without the need to control and manage the output from two separate energy and power devices in a complex hybrid power system -- is preferred because of its relative simplicity and ease of control.

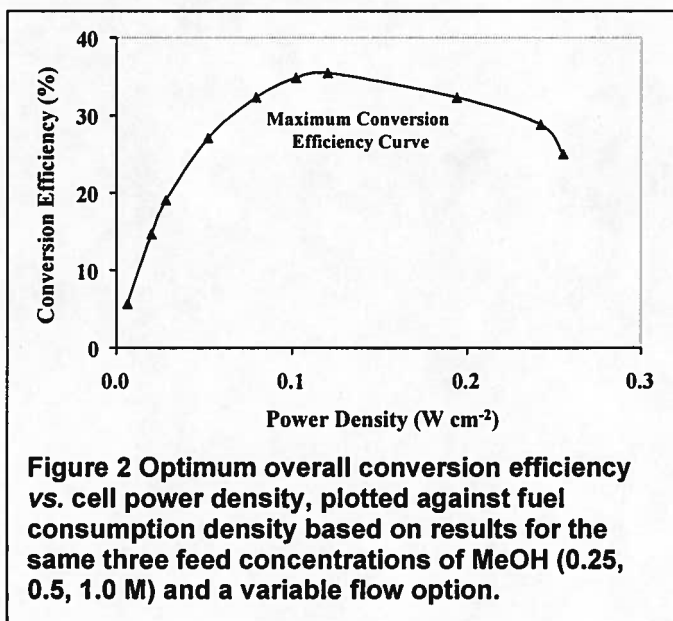
## ENERGY CONVERSION EFFICIENCY

In optimizing the operation of a DMFC for maximum efficiency across a broad dynamic power range, the key cognitive step is to realize that the example shown in Figure 1 is for a *particular combination* of anode (and

cathode) conditions. That is, it does not represent a limiting behavior of the DMFC, nor a complete summary of its fuel conversion and power delivery capabilities, but is simply the DMFC composite characteristic for a particular combination of fuel rate (and at a fixed air rate and pressure). That is, for a different set of anode conditions, (i.e., a different methanol solution concentration and solution flow rate) the crossover current, cell current, voltage and power density will all change, in a given cell, in a predictable and controllable way.

So, for each different choice of methanol fuel solution concentration and solution flow rate to the anode (analogously to the fuel rate and oxygen stoichiometry in an internal combustion engine), there will be a different pair of  $I_x$  and  $I_c$  curves vs. cell voltage that characterize a particular cell design and materials combination. It is therefore possible to minimize the crossover losses over a broad range of power densities by manipulating the anode feed conditions to an optimum pair of values corresponding to each power density level required -- in exactly the same way as is done for a conventional internal combustion engine (where fuel rate varies roughly linearly with the required power level, in most cases, and the oxygen stoichiometry is maintained for efficient combustion).

Specifically, it is possible to select an optimum fuel solution flow rate for each methanol concentration value. The result of this optimization procedure is shown in Figure 2 in a somewhat different format -- namely, conversion efficiency vs. power density. This format is particularly useful for dynamic power system analysis in an automotive application. The exact details of this optimization procedure are discussed in the original paper that pioneered this optimization process.



As a result of this optimization procedure, the major conclusions regarding the cell power density and cell fuel conversion efficiency for a DMFC are:

1. At any required cell power density level (up to the peak available), the DMFC power density can be maximized for a given methanol fuel consumption rate by simultaneously controlling the methanol fuel concentration and solution flow rate to the anode

(i.e.; the fuel rate). This optimum concentration/flow combination is specific to the methanol consumption rate, the required power level, and the characteristics of the particular DMFC cell design.

2. The envelope of all such points of maximum power density vs. fuel rate provides the maximum conversion efficiency for the complete range of fuel consumption and provides the fuel concentration/flow conditions for the specific DMFC design.
3. The optimized conversion efficiency curve provides a conceptual basis for a DMFC system control strategy that will maximize the DMFC energy conversion and power output over a broad dynamic range.

A key advantage of the resulting optimized curve is its relative flatness versus power density (Figure 2). This means that the fuel efficiency of the DMFC power system in a vehicle will be largely independent of the driving cycle (in sharp contrast to the results for conventional vehicles, and even for other types of fuel cell power systems).

This uniform efficiency behavior vs. power level is illustrated in Figure 2 – using data for state-of-the-art DMFC technology – where it is possible to achieve greater than 30% efficiency over a power density range from about 70 to 230 mW cm<sup>2</sup>. This is a highly desirable shape of efficiency vs. power for applications where power demand has a broad dynamic range – such as an automotive application.

Overall, the major operational conclusion regarding DMFC fuel conversion efficiency is that it is possible (certainly in principle) to optimize the conversion efficiency of a DMFC power system by manipulating the anode feed stream fuel rate dynamically as a function of the system power demand. Successful application of such a fuel rate control strategy in a real power system and vehicle would, of course, require implementation of a variable-concentration and variable-flow control system for the methanol solution supply to the anode of the DMFC. In addition, development of appropriate sensors and uniform solution delivery technology is a non-trivial technical prerequisite for this dynamic optimization process.

## POWER DENSITY: CELL AND STACK

A very recent paper has made a system level comparison of the attributes for a DMFC powered vehicle vs. a DHFC powered FCV.<sup>8</sup> Overall, this study concludes that a DMFC-powered FCV could meet the requirements for a general-purpose ZEV, and could thereby be an effective competitor to a DHFC-based FCV – in any locality or market niche where zero exhaust emissions capability is a condition of doing business.

The conclusion of the cited study is that the DMFCV (Direct-Methanol Fuel Cell Vehicle) would have an equivalent range of 350 miles, and would provide the same class of performance (acceleration) as an advanced design hydrogen-fueled FCV,<sup>10,11,12</sup> if:

1. the DMFC stack power density were at 0.35 kW l<sup>-1</sup>,

2. the DMFC stack conversion efficiency (over the required test drive cycle) is at 50% of the equivalent DHFC stack drive cycle efficiency.

These relative performance criteria (DMFC vs. DHFC) are already marginally met at the cell level for the experimental data representing the current state-of-the-art DMFC (i.e.; Figure 2) – which corresponds to the data used in the cited DMFC vs. DHFC comparison.<sup>8</sup>

Another factor that facilitates the positive outlook for the DMFC system's competitive position is the prospect of very effective cell packaging into a compact and high-power-density stack configuration. This higher cell pitch (or packing density) occurs because of large potential simplifications in the cell design that result from very simple temperature control and membrane hydration in the DMFC. Recent experimental mini-stack fabrications suggest the possibility of the DMFC stack approaching or even exceeding the published state-of-the-art stack power density for the DHFC (Direct-Hydrogen Fuel Cell).

Specifically, the above requirement of DMFC stack power density at a level of 35% of a DHFC stack appears rather modest compared to recent DMFC "mini-stack" fabrication at LANL (Los Alamos National Laboratory) – a demonstrated DMFC stack cell pitch of 2 mm.<sup>9</sup> This cell pitch is smaller than the publicly announced state-of-the-art for a DHFC stack (i.e.; the linear density of cells is higher for the DMFC mini-stack), and, if naively projected to a full scale DMFC stack, leads to an estimated DMFC stack power density of roughly 1.25 kW/Liter – using only the experimental state-of-the-art DMFC cell power density shown in Figure 2 as the basis for the projection (i.e.; without any projected improvements in the DMFC cell characteristics).

This DMFC mini-stack cell pitch has yet to be demonstrated in a full size stack (several hundred cells in series, nominally 50 kW) or with full size cells (>500 cm<sup>2</sup> area per cell). The engineering design and materials requirements for a full size stack with an output of 50-100 kW should not be trivialized. Major and creative engineering design is needed, for example, to efficiently provide the required reactant supply (methanol, water, air) and product removal (CO<sub>2</sub>, water) for large area cells in a thermally and electrically coupled stack configuration. However, even so, the initial mini-stack achievements cited above are quite impressive – and indicate some of the great promise of a DMFC power system for automotive applications.

## A NOTE ON INDIRECT FUEL CELL SYSTEMS

Finally, a few words should be said about the efficiency and power density for fuel cell stacks based on indirect methanol and indirect hydrocarbon ("gasoline") fuels, and some rough comparisons should be offered to provide context for the conclusions re DMFC systems.

The most favorable estimate for a methanol steam-reformer stack can be calculated as follows. Using the upper values of 90% relative voltage efficiency (direct hydrogen dilution loss in the stack), 80% reformer efficiency, and 85% fuel utilization (excess fuel stoichiometry) yields a relative overall fuel efficiency of about 60% for a steam methanol reformat stack relative



to the fuel efficiency of a DHFC stack.<sup>8</sup> The primary impact on the peak power density of the indirect-methanol stack is the increased anode polarization (hence reduced cell voltage and current) due to the effect of the diluted hydrogen in the methanol reformat – and an additional, potentially catastrophic, impact of CO-poisoning that is not included in these estimates. For the range of dilution expected, *i.e.*, nominally 70% hydrogen mole fraction, the stack power density would be reduced to about 90% of the DHFC stack.<sup>8</sup>

The relative efficiency of a partial-oxidation "gasoline" reformer stack is lower than that computed above for the methanol steam reformer case. A rough figure for the "gasoline" case can be calculated from the following figures – 80% efficiency due to the anode polarization dilution losses, 70% reformer efficiency; and 75% fuel utilization of hydrogen supplied to the stack.<sup>8</sup> Since the partial oxidation reformation process for "gasoline" is exothermic, there is very limited opportunity to recover the excess (fuel cell exhaust) hydrogen energy within the overall partial oxidation fuel cell system. The overall efficiency of such a system is therefore calculated to be about 40% of that of the DHFC system – *i.e.*, roughly equivalent to the "system" efficiency for the present DMFC state-of-the-art.<sup>8</sup> For the range of hydrogen dilution expected from a partial oxidation "gasoline" fuel processor – *e.g.*, about 20% hydrogen mole fraction – the indirect-gasoline stack power density would be reduced to 80%, or somewhat less, relative to that of the DHFC stack.<sup>8</sup>

## CONCLUSION

In principle, a DMFC powertrain can be designed for operation in a **load-following mode** over the wide dynamic power range needed for automotive applications. This conclusion is based on an analysis of the present experimental DMFC state-of-the-art, and on a basic theoretical understanding of the DMFC cell/stack characteristics and optimum operation strategy. The conventional wisdom that a hybrid powertrain design is required for a DMFC stack is fundamentally incorrect. A broad region of high fuel conversion efficiency vs. power level is possible for DMFC operation. This is illustrated in Figure 2 for state-of-the-art DMFC technology. Figure 2 demonstrates that the DMFC state-of-the-art can achieve greater than 30% efficiency over a DMFC power density range from about 70 to 230 mW cm<sup>2</sup>. This is a highly desirable shape of efficiency vs. power for applications where power demand has a broad dynamic range – such as an automotive application.

The possibility of a load-following power system design, plus the clear (but as yet unrealized) potential for major improvements in both DMFC power density and energy conversion efficiency, has generated worldwide R&D efforts on DMFC cells and stacks for automotive applications. The eventual commercial success of these R&D efforts will only come about as the result of many parallel advances in technology and process development (*e.g.*; catalyst, membrane, other enabling materials, fabrication processes, etc.).

Overall, the major operational conclusion regarding DMFC fuel conversion efficiency is that it is possible (certainly in principle) to optimize the conversion efficiency of a DMFC power system by manipulating the anode feed stream fuel rate dynamically as a function of

the system power demand. Successful application of such a fuel rate control strategy in a real power system and vehicle requires the implementation of a variable-concentration and variable-flow control system for the methanol solution supply to the anode of the DMFC. In addition, development of appropriate sensors and uniform solution delivery technology is a non-trivial technical prerequisite for this dynamic optimization process.

Recent DMFC preliminary "mini-stack" fabrication at LANL (Los Alamos National Laboratory) has demonstrated DMFC stack cell pitch of 2 mm.<sup>9</sup> This cell pitch is smaller than the publicly announced state-of-the-art for a DHFC stack (*i.e.*; the linear density of cells is higher for the DMFC mini-stack), and, if naively projected to a full scale DMFC stack, this DMFC cell pitch leads to an estimated DMFC stack power density of roughly 1.25 kW/Liter – using the experimental state-of-the-art DMFC cell power density shown in Figure 2 as the basis for the projection (*i.e.*; without any projected improvements in the DMFC cell characteristics).

This DMFC mini-stack cell pitch has yet to be demonstrated in a full size stack (several hundred cells in series, nominally 50 kW) or with full size cells (>500 cm<sup>2</sup> area per cell). The engineering design and materials advances required to demonstrate and commercialize a full size DMFC stack with an output of 50-100 kW should not be trivialized. Major and creative engineering design is needed, for example, to efficiently provide the required reactant supply (methanol, water, air) and product removal (CO<sub>2</sub>, water) for large area cells in a thermally and electrically coupled stack configuration. However, even with this caution, the initial mini-stack achievements cited above are quite impressive – and indicate some of the great promise of a DMFC power system for automotive applications. Much remains to be done to convert this promise to reality, but the potential impact on FCV design – and on the long-term potential pathway of methanol as an FCV fuel – is equally impressive.

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