DYNAMICS AND CONTROL OF THE DIRECT METHANOL FUEL CELL: STATE SPACE MODELLING OF THE VOLTAGE RESPONSE

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The transient response of the direct methanol fuel cell (DMFC) is of paramount importance when it is used for transportation applications. We have applied a variety of loading cycles on a small scale single DMFC in order to evaluate the effect of the loading pattern and operating conditions on the cell response time and performance. The cell responds rapidly and reversibly to changes in magnitude and rate of change of load. Under dynamic operation the cell voltage response can be significantly better than that achieved under steady state operation. Open circuit potentials are also increased, by up to 100mV, by imposing a dynamic loading strategy. In addition the study reports the dynamic characteristics of a large-scale cell and cell stack and explains the differences in cell response. This paper also reports a model based study of the dynamics of the direct methanol fuel cell. An empirical model based on a Canonical Variate Analysis state space representation is developed to predict the voltage response of the DMFC and mutli cell stacks.

Dynamic behaviour of the direct methanol fuel cell

Various load cycles were used in this study; trying to simulate conditions of a vehicle driven in urban areas: gradual acceleration, deceleration, instantaneous loading with high power demands from zero load conditions, continuous progressive loading and unloading, and arbitrary load cycles (Figure 1a and 1b). The study has shown that the dynamic performance of the DMFC is affected by complex interactions of electrode kinetics and mass transport processes. The cell responded rapidly, and reversibly, to changes in magnitude in load and rate of change of load. Under dynamic operation the cell voltage response can be significantly better than that achieved under steady state operation. This improvement is potentially attractive in vehicle applications where higher power density and fuel efficiency can be realised.

Four different methanol solution concentrations were examined: (0.25, 0.5, 1.0 & 2.0 M) under a pulse load cycle (Figure 1c). The 2.0 M solution gave a fast and stable response for both current loads, reaching a steady state at approximately ten seconds after application of the pulsed load, but with the penalty of lower voltage. Removal of the load caused a rapid return to open circuit voltage, which was higher than the initial starting value, but which then proceeded to fall and reach a constant value after approximately fifty seconds. The 0.25 M solution gave a much slower response and can hardly sustain satisfactory operation above 100 mA cm^{-2}. The 0.5 M solution gave the best voltage output but the response time deteriorated with the increase in the current density load from 50 to 100 mA cm^{-2}. Overall in terms of electrical performance and response time, to a step change in the cell load, the 1.0 M methanol concentration showed the best compromise in overall behaviour. This was in good agreement with results from steady state operation where a concentration of approximately 1.0 M gave the best compromise between electrical performance and reduced methanol crossover.

Overall it can be said that the flow rate did not affect significantly the cell dynamic response in the case of low to mid current densities and when changes in cell loading condition were gradual. On the contrary when the cell was operated at high current densities and with sudden steep changes in the cell loading condition, the cell response time was significantly affected by the flow rate. Under all conditions the best performance was achieved at the higher cathode air pressure. The effect of cathode pressure was less significant for low to medium current densities and for gradual changes in the loading conditions. The repeated application of an increasing load served to increase the open circuit voltage after the cell was unloaded. On scale up (see Figure 1d) the response of the cell was slower and was influenced less by a change in concentration and in

methanol solution flow rate. On the other hand, a reduction in air pressure resulted in a marked deterioration in performance when the pressure approaches atmospheric.

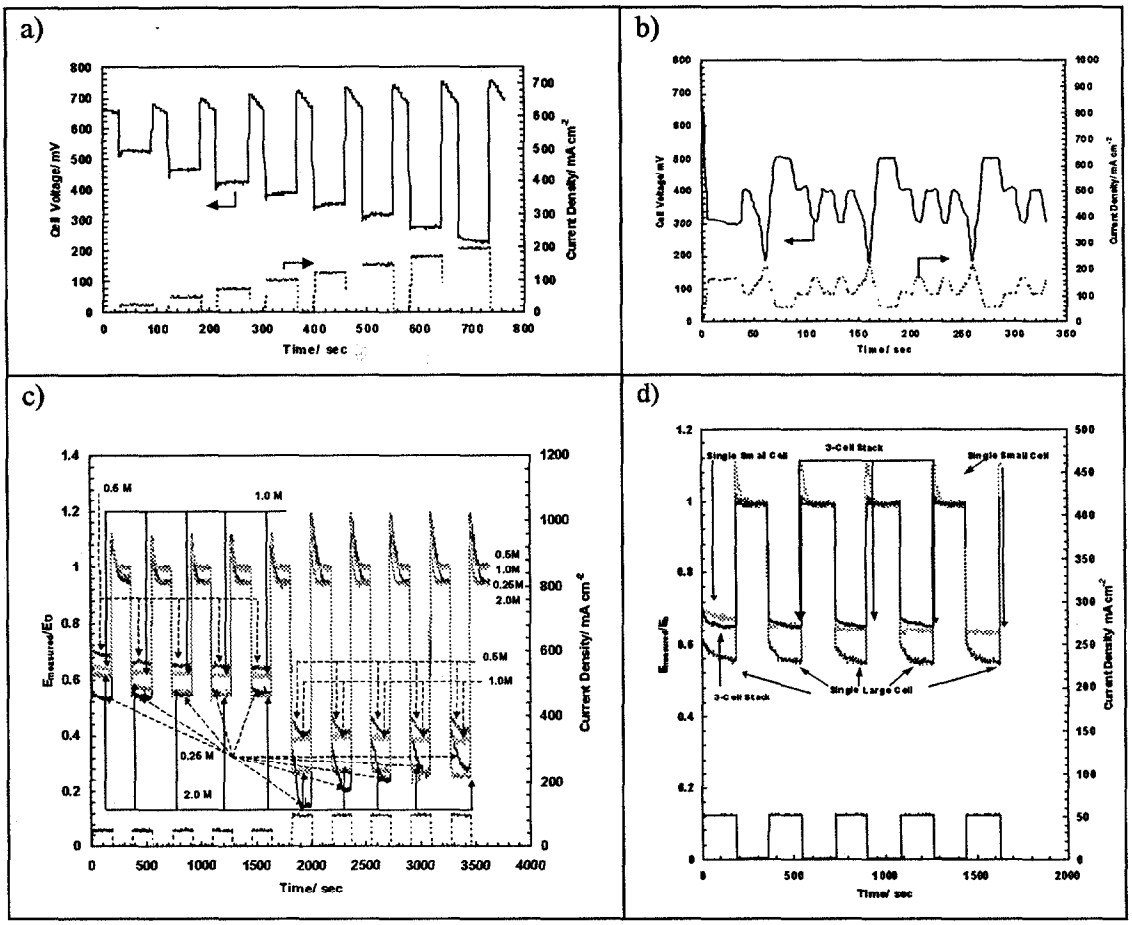

Figure 1: *The voltage response of the DMFC under variable load and operating conditions: a) Pulse type load b) Arbitrarily varying load c) The effect of methanol solution concentration d)The effect of scale up and cell stacking.*

Control of the Direct Methanol Fuel Cell: State space modelling of the voltage response

In order to achieve high performance control of a commercial system it is essential to have a methodology that will accurately predict the stack voltage from a minimum number of sensors and with the smallest time delay after vehicle's start up. In particular, predictive control will enable system components to be adjusted to meet future demands and offer the performance needed to meet driveability requirements.

An empirical model based on a Canonical Variate Analysis state space representation was developed to predict the voltage response of the DMFC cell and mutli cell stacks to a wide range of varying dynamic loads and operating conditions. The methodology was centred on the experimental dynamic performance of two cell systems: a small-scale single cell and a three-cell stack assembly. The advantage of CVA state space modelling is that no *a priori* knowledge of the system parameters, dynamics, or time-delays is required. CVA was first applied to the data and pseudo-states of the system were calculated. These pseudo-states were fair approximations of the true states of the real system since they were "optimal" predictors of the future outputs. Here

"optimal" implies that the prediction error was minimised. The pseudo-states were then used to develop a state space model considered as the most general representation of a linear time-invariant system. It was thus considered reasonable to use such a CVA based model to describe both the steady-state and dynamic operation of the DMFC. The Canonical Variate Analysis approach was able to describe with high accuracy (typically above 90% without model optimisation) the system dynamics using only two measurement sources and provideed acceptable inferential and one step ahead predictions even when the systems were operated without having reached a steady state (See Table 1 for details).

Operating Conditions \ Data Set	**Data Set (A)**	**Data Set (B)**	**Data Set (C)**	**Data Set (D)**	**Data Set (E)**	**Data Set (F)**
Solution Concentration (M)	2	1	0.25	2	1	1
Cathode Pressure (bar)	2	0.5-2	2	2	1.4	1.7
Cell Temperature ($^{\circ}$C)	64-71	79-81	85	80	63-71	43-74
Methanol Inlet Temperature ($^{\circ}$C)	80-90	51-54	50-55	51-57	66-77	43-76
Methanol Outlet Temp. ($^{\circ}$C)	69-79	-	-	-	68-78	40-72
Air Inlet Temperature ($^{\circ}$C)	-	-	-	-	22-23	21-29
Air Outlet Temperature ($^{\circ}$C)	-	-	-	-	36-50	29-54
Anode Flow Rate (cm^3min^{-1})	150	5-15	10	15	3500	5500
Total Active Area (cm^2)	9	9	9	9	816	816
Number of Cells	1	1	1	1	3	3
Applied Load	5-70	0-201	0-100	0-200	18-76	18-76
Number of Data Points	13912	9502	3630	766	11288	6908
Inferential Estimates (%)	**96.60**	**97.53**	**98.25**	**96.53**	**99.39**	**99.15**
One-step-ahead predictions(%)	**95.41**	**96.65**	**98.02**	**94.76**	**98.99**	**98.64**

Table 1: *Operating conditions for the DMFC experiments and accuracy of inferential estimates and one-step-ahead predictions of the six data sets using CVA modelling*

CVA state space dynamic models have been used to infer and predict in a one-step-ahead sense, with reasonable accuracy, the scale up voltage responses of a three-cell DMFC stack that was significantly different, both in terms of geometry and physiology, than the small scale system. In contrast, using the CVA state space dynamic model, built from the three-cell stack data, to predict the voltage response of the small single-cell system, i.e. scale-down, although feasible, gave predictions that were slightly inferior to the case of scale-up. The issue of scale-down is rather unusual as most development work starts from small-scale cells in order to minimise development time and costs, and where engineering issues are of less importance for correct operation. In this context the proposed CVA state space representation is a promising route for investigating the scale up of DMFC systems. There are, however, situations where during process development, a designer needs to develop a simulation of a large scale system and in this instance the ability to scale-down would be a major advantage. For example in the production of power supplies for miniaturised transmitters. Although for the scale-down experiments, accuracy's of only 90-95% are achieved, such overall accuracy's were still far better than those that can be produced from highly complicated deterministic models. In addition, the time and effort to develop a CVA state space dynamic model is much less than that for mechanistic models and more important is non-system specific. This is a major benefit of this new approach. A further advantage of the present dynamic modelling approach is that it can be used for on-line real-time cell performance monitoring as well as for on-line fault detection

The accuracy achieved utilising only two sensor measurements (see Figure 2) provides impetus for the continuation of the study into the use of CVA state space methodologies for the dynamic

modelling of the DMFC. One of the main problems is the issue of the time delay between system start-up and the time required using traditional control techniques to collect sufficient appropriate data to control the system effectively. The state space CVA technique, in part, addresses this issue. Only five values were required before the model was computed and able to provide predictions resulting in minimum time delay. This is potentially a major advantage in automotive applications.

Finally, the use of a dynamic state space modelling approach to represent the dynamics of other fuel cells, e.g. hydrogen, should also be possible. In principle this is a relatively straightforward task since the two polymer electrolyte membrane systems are quite similar in terms of geometry, design and materials. If for the DMFC, the assumption that the accuracy of the scaled-down models was limited, due to the changes in the cell physiology through vapour liquid equilibrium and gas management, both of which are related to the liquid nature of the feed, then in the case of hydrogen cells, the resulting model predictions may well be improved. These issues are currently under consideration.

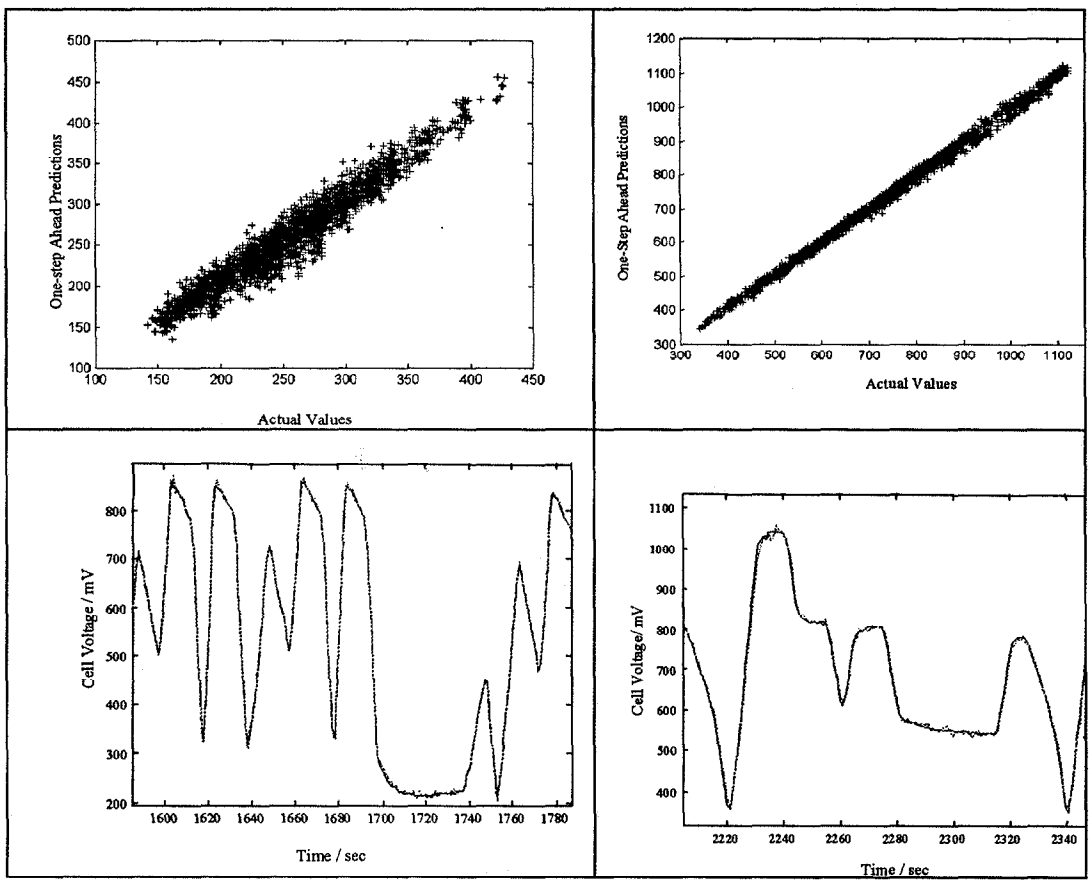

Figure 2: *Sample prediction of DMFC voltage response with the aid of CVA modelling*

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