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Optimisation of air cooled, open-cathode fuel cells: Current of lowest resistance and electro-thermal performance mapping

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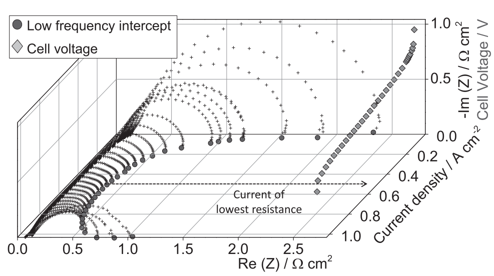
HIGHLIGHTS

- Current of lowest resistance used as a metric for performance comparison.
- Electro-thermal performance maps identify optimal performance point.
- Higher cooling flow rate increases the current of lowest resistance, and reduces the actual resistance.

GRAPHICAL ABSTRACT



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ABSTRACT

Selecting the ideal operating point for a fuel cell depends on the application and consequent trade-off between efficiency, power density and various operating considerations. A systematic methodology for determining the optimal operating point for fuel cells is lacking; there is also the need for a single-value metric to describe and compare fuel cell performance. This work shows how the 'current of lowest resistance' can be accurately measured using electrochemical impedance spectroscopy and used as a useful metric of fuel cell performance. This, along with other measures, is then used to generate an 'electro-thermal performance map' of fuel cell operation. A commercial air-cooled open-cathode fuel cell is used to demonstrate how the approach can be used; in this case leading to the identification of the optimum operating temperature of ~45 °C.

1. Introduction

Polymer electrolyte fuel cells (PEFCs) fuelled with hydrogen are among the most promising energy conversion technologies for a broad range of applications, including portable, stationary and automotive power delivery. In recent years PEFCs have shown tremendous advances in terms of performance and durability, with wide-scale commercialisation imminent. However, new techniques are sought to optimise performance by understanding the internal workings of these devices. Understanding aspects of thermal and water management is of particular interest as they have a profound effect on the performance and durability.

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In developing PEFC technology, it is important that appropriate comparisons are made between different operational strategies (liquid/air cooled, humidified/dry gases), designs (closed/open cathode, through-flow/dead-ended) and materials (graphitic/metallic bipolar plates, etc.). Current and voltage are the key metrics of performance; however, there is no standard metric that can report the performance of a fuel cell as a single number; rather, whole polarisation plots are typically compared. It is common to quote the current and voltage at the maximum power point in order to compare the performance between designs and operating conditions [1–7].

Whilst the maximum power density is considered to indicate the highest performance, it is obtained under extreme conditions (typ. voltage lower than 0.45 V), at a current close to the limiting current density. If operated in this region, the system is likely to undergo local dehydration and/or membrane flooding and reactant starvation. Operation at the highest current density could reduce the cell's lifetime and increase the risks of catastrophic failures [8–12]. Also, it has been shown that the fuel efficiency is less than 50% at maximum power, because the internal impedance must match the load impedance at maximum power. Operating below maximum power improves fuel efficiency [13]. The actual optimal fuel efficiency depends on the combination of the load/internal impedance ratio and the fuel recovery (recycle system) or waste (dead ended system). From a system perspective, parasitic power consumption will be higher at the maximum power density due to the heat and water management, therefore lowering the net system power [14,15]. Therefore, the maximum power is not a suitable indicator of how a fuel cell will operate under most practical conditions, and does not take into account operating parameters.

In addition, the current/voltage characteristics alone neglect a further critical parameter of performance: temperature. The temperature of a fuel cell has a profound effect on electrochemical performance, influencing the thermodynamics, electro-kinetics, transport processes and water distribution, which collectively dictate balance-of-plant requirements, system efficiency and long-term durability. It is desirable to maintain control over the stack temperature, and to minimise any inevitable heterogeneities in the temperature distribution within the stack, in order to maximise performance and durability. Therefore, the critical parameter of temperature, and indeed its spatial variation, should be considered alongside current and voltage when characterising fuel cell performance and searching for optimum operating conditions. This is important for all fuel cell types and operating modes, but is particularly relevant to air-breathing/cooled fuel cells, for which the air supply acts to provide both reactant and cooling for the system.

Air-breathing PEFCs have attracted increasing interest over the last decade. Unlike closed-cathode systems, self-breathing designs offer the advantages of simpler design and integration into systems, taking air directly from the atmosphere. Passive air-breathing systems are limited to a maximum current density of ~600 mA cm^{-2} [1,6,16–18] due to heat and water management issues, since there is no water removal from the membrane, apart from evaporation [6,19]. Forced convection of air using fans improves performance in the so-called open-cathode configuration, and enables higher current densities to be attained [3,5,20–22].

In air-cooled, open-cathode systems the temperature depends on the voltage and current density [16,23], air cooling flow rate [3,21], and heat transfer characteristics of the stack. Temperature monitoring is therefore crucial to avoid thermal runaway or hot spot formation at high currents densities. In practice, this is normally performed using a single-point thermocouple inserted in the centre of the cell [20,24,25] or using multiple micro thermocouple measurements at various locations in the fuel cell [26–28].

Electrochemical impedance spectroscopy (EIS) is an established and powerful tool for fuel cell characterisation, providing insightful information on the various resistive losses and capacitive effects that determine fuel cell operation [29–31]. EIS has been used to characterise PEFC response to CO poisoning [32], decouple anode and cathode operation [33], and to isolate and explore the effect of specific components (e.g. platinum loading, membrane thickness, GDL structure) [29]. EIS has also found applications in localized measurements [34,35], fault detection and flooding/drying events [10,36,37], and more recently, dynamic processes such as dead-ended anode operations using a reconstructive impedance measurement technique [25]. Focusing exclusively on the high frequency intercept with the real axis of the Nyquist plot (zero phase shift point) provides a measure of the purely Ohmic impedance of a fuel cell, which can be used to measure changes in the conductivity, and therefore hydration level, of the polymer electrolyte membrane [24,37–43]. However, the low frequency intercept has been neglected as a diagnostic feature, despite representing the total impedance of the system and therefore potentially being a way to identify the minimum impedance point on the polarisation curve.

This work presents a novel approach to characterising fuel cell performance that: (i) uses the low-frequency intercept with the Nyquist plot as a means of identifying the minimum impedance point in the polarisation curve, (ii) highlights how this can be used as an optimisation strategy in order to determine the most suitable conditions, and (iii) proposes an application of this new method, in order to determine the optimum air flow rate and current density, considering the influence of the temperature as an integral part of the performance 'map', alongside current and voltage. The minimum impedance point can be used as a key performance metric. In the absence of other considerations, the minimum impedance point represents a sensible trade-off between efficiency and power, and is likely to be preferable to the maximum power point when seeking to maximise stack lifetime. 'Electro-thermal mapping' allows optimum performance regions to be identified and decouple the effect of temperature on factors such as membrane hydration (conductivity) and cooling.

2. Experimental

Fuel cell testing – A 5-cell (60 cm^2 active area) air-cooled/air-breathing fuel cell stack was used for testing (Intelligent Energy Ltd., UK). The membrane electrode assembly was composed of commercially available gas diffusion layers (GDLs) and commercially available membranes with Pt loading of 0.1 and 0.4 mg cm^{-2} on the anode and cathode, respectively.

The test station supplied dry hydrogen at ambient temperature (with a purity of 99.995%) to the anodes and air was blown by three fans (SanAce 36, Sanyo Denki) to the open cathode channels [25,44]. The exhaust hydrogen flow rate in through-flow mode was measured using a thermal mass flow meter (MassVIEW, Bronkhorst) to be 4.7 SLPM. The fans, which provide cooling and air supply to the cathode, were controlled by a programmable power supply (3649A Agilent). The current drawn from the PEFC was controlled by an electronic load (PLZ664WA, Kikusui) in galvanostatic mode. An in-house computer controlled system controls the air, hydrogen, cooling and electrical valves (LabVIEW, National Instruments) as well as recording and presenting data using a data acquisition card (PCI 6221, National Instruments). It was used in the $\pm$1 V range with a resolution down to 30 μV (i.e. 2 V/2^{16}). Ambient temperature, pressure (absolute) and relative humidity (RH) were measured, being of around 25 °C, 1.02 bar and 40% RH respectively, during all tests. The software enables a maximum sampling rate of 5 Hz. The operation of this fuel cell in terms of open-cathode design, cooling and active channels and materials [25,44],