Electrochimica Acta 180 (2015) 307–315

Contents lists available at ScienceDirect

Electrochimica Acta

journal homepage: www.elsevier.com/locate/electacta

The Hydro-electro-thermal Performance of Air-cooled, Open-cathode Polymer Electrolyte Fuel Cells: Combined Localised Current Density, Temperature and Water Mapping

Quentin Meyer[a], Sean Ashton[a], Rhodri Jervis[a], Donal P. Finegan[a], Pierre Boillat[c,d], Magali Cochet[c], Oliver Curnick[b], Tobias Reisch[b], Paul Adcock[b], Paul R. Shearing[a,e,**], Dan J.L. Brett[a,*]

[a] Electrochemical Innovation Lab, Department of Chemical Engineering, UCL, London, WC1E 7JE, United Kingdom
[b] Intelligent Energy, Charnwood Building Holywell Park, Ashby Road, Loughborough Leicestershire, LE11 3GB, United Kingdom
[c] Electrochemistry Laboratory (LEC), Paul Scherrer Institute (PSI), 5232 Villigen, Switzerland
[d] Neutron Imaging and Activation Group (NIAG), Paul Scherrer Institute (PSI), 5232 Villigen, Switzerland
[e] UCL/Zeiss Centre for Correlative X-ray Microscopy, UCL, London, WC1E 7JE, United Kingdom

ARTICLE INFO

Article history:
Received 10 June 2015
Received in revised form 18 August 2015
Accepted 19 August 2015
Available online 24 August 2015

Keywords:
Air-cooled open-cathode polymer electrolyte fuel cell
water mapping
neutron imaging
temperature mapping
current mapping

ABSTRACT

In situ diagnostic techniques provide a means of understanding the internal workings of fuel cells so that improved designs and operating regimes can be identified. Here, a novel metrology approach is reported that combines current and temperature mapping with water visualisation using neutron radiography.

The approach enables a hydro-electro-thermal performance map to be generated that is applied to an air-cooled, open-cathode polymer electrolyte fuel cell. This type of fuel cell exhibits a particularly interesting coupled relationship between water, current and heat, as the air supply has the due role of cooling the stack as well as providing the cathode reactant feed via a single source. It is found that water predominantly accumulates under the cooling channels (thickness of 70-100 μm under the cooling channels and 5-25 μm in the active channels at 0.5 A cm^{-2}), in a similar fashion to the lands in a closed-cathode design, but contrary to passive open-cathode systems. The relationship between current, temperature and water accumulation is complex and highly dependent on location within the cell. However, there is a general trend that higher currents and cooling limitations, especially above 0.7 A cm^{-2} and below 3.9×10^{-3} m^3 s^{-1}, leads to temperatures above 60 °C, which dehydrate the membrane (water thickness of 10-25 um) and the cell operates below 0.5 V.

1. Introduction

Polymer electrolyte fuel cells (PEFC) fuelled with hydrogen are among the most promising energy conversion technologies for a broad range of applications, including portable, stationary and automotive power delivery. A range of diagnosis techniques have been developed to understand and improve the heat and water management in these devices with a view to improving performance, extending durability and informing advanced design.

1.1. Current and temperature mapping in fuel cells

Current mapping studies have proven to be insightful and revealed large current density gradients attributed to factors such as: uneven fuel consumptions [1–4], operating conditions [5–7], stoichiometric ratios [8–11], the reactant flow orientation [3,7], and water management issues [12].

Temperature distribution has also been extensively studied, identifying areas of higher electrochemical activity, hot-spot formation and fuel depletion. Thermocouples can provide a crude measure of temperature inside fuel cells [13–16] but cannot provide high spatial resolution. Moreover, thermocouples need to be inserted inside the fuel cell, which often requires design modifications. In contrast, infrared thermal imaging can provide very high spatial and temperature resolution [17–22], yet typically requires use of modified fuel cells with an infrared transparent

---

[*] Corresponding author. Tel.: +44 20 7679 3310.
[**] Corresponding author at: Electrochemical Innovation Lab, Department of Chemical Engineering, UCL, London, WC1E 7JE, United Kingdom.
E-mail addresses: p.shearing@ucl.ac.uk (P.R. Shearing), d.brett@ucl.ac.uk, http://www.ucl.ac.uk/electrochemical-innovation-lab (D.J.L. Brett).

http://dx.doi.org/10.1016/j.electacta.2015.08.106

window, or is otherwise confined to open-cathode fuel cells [23–25] or the outer surface of a cell or stack [26,27].

Combined temperature and current mapping studies offer an extra dimension of information and allow the impact of each parameter on the other to be assessed [15,26,19,27,28]; however, the other important component in this equation, water, needs to be considered in unison to see the whole picture.

1.2. Liquid water mapping in fuel cells

Effective water management is of paramount importance for fuel cell operation (dehydration / flooding can lead to performance decay and stack failure). Water generation and removal, and transport processes in the gas diffusion layer, membrane and flow-field have been extensively modelled [30–36]. However, the reliability of these models depends on the level of validation, which requires appropriate experimental inputs. Therefore, a number of experimental methods have been investigated, in order to evaluate, quantify, measure and / or visualise the water dynamics and distribution under different modes of operation. Such techniques should ideally satisfy three requirements as defined by Stumper et al. [37]: (i) *in situ* applicability, (ii) minimal invasiveness and (iii) ability to provide information on the distribution of liquid water over the active area.

High frequency impedance enables the ohmic resistance of a fuel cell to be measured, which can be used to monitor changes in the membrane conductivity, and therefore hydration content [1,7,16,29,38–44]. Localised electrochemical impedance spectroscopy (EIS) has been achieved as well, and provides more insight on the hydration / dehydration processes distributed across electrodes [1,5,29,45].

To investigate water content, it is possible to weigh the fuel cell before and after operation [46], or to visualise liquid water via optical imaging open channels [24,47]. These methods are attractive because of their simplicity, but the most powerful method for water visualisation, (satisfying all three criteria from Stumper et al. [37]) is neutron imaging. This technique is based on attenuation of a neutron by hydrogen-containing compounds such as water, and transparency to neutrons of most fuel cell construction materials (aluminium, stainless steel). Neutron imaging can identify water in the in-plane orientation (with the membrane place parallel to the beam) and through-plane orientation (with the membrane plane perpendicular to the beam), enabling in the first case to differentiate the water content from the cathode and the anode [48–50] and in the second case the effect of different designs, components, and operating conditions [45,51–65]. Neutron imaging has been combined with other modelling and experimental techniques, such as current mapping [66], CFD models validation [32,51,65], optical imaging [47], neutron scattering [61] and localised EIS [45].

1.3. Air-cooled, Open-Cathode Fuel Cells

Unlike conventional closed-cathode fuel cells, self-breathing fuel cells offer the advantages of simpler design and integration into systems, using diffusion from the atmosphere without compressors. Passive air-breathing systems are typically limited to a maximum current density of ~0.6 A cm^{-2} [67–71] due to heat and water management issues, since water cannot be removed from the membrane, except through evaporation [69,72]. In the so-called 'air-cooled, open-cathode' configuration, air is forced through the cathode channels using fans, which improves performance and enables higher current densities to be attained [73–77]. In air-cooled, open-cathode systems the temperature depends on the voltage and current density [46,67], air cooling flow rate [73,76], and heat transfer characteristics of the stack. Temperature monitoring is therefore crucial to ensure effective and durable operation. In practice, this is normally performed using a single-point thermocouple inserted in the centre of the cell [16,26,75], or for development work using multiple micro thermocouple measurements at various locations in the fuel cell [13,78,79].

Here, we present the results obtained by applying a novel metrology approach to an air-cooled, open-cathode two-cell stack, operated without external humidification: the technique combines water visualisation using neutron imaging, with current and temperature mapping using a printed circuit board (PCB) sensor plate [80]. The effect and relationship between the key hydro-electro-thermal properties allows important new insight into this type of fuel cell to be achieved.

2. Experimental

Fuel cell testing - A 2-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 [26] supplied dry hydrogen at ambient temperature (with a purity of 99.995%) to the anodes and air was forced through the stack by a single fan (SanAce 36, Sanyo Denki) to the open-cathode channels (Fig. 1). The exhaust hydrogen flow rate in through-flow mode was measured using a

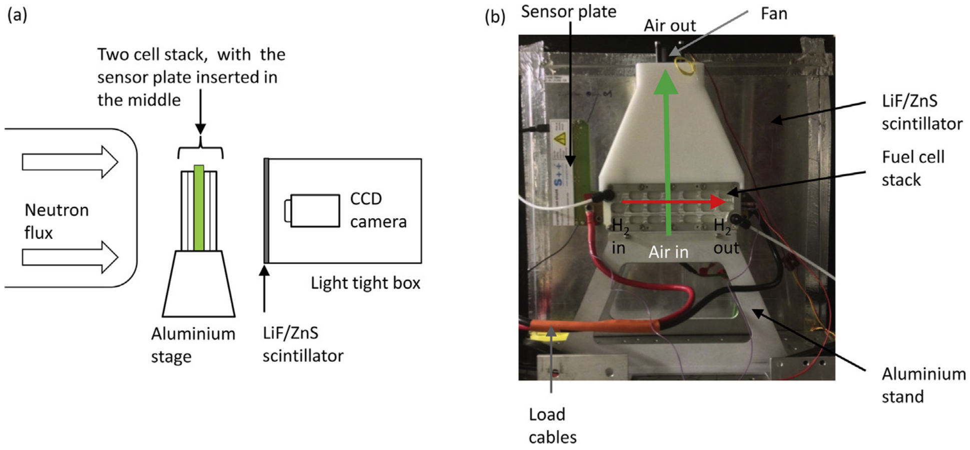

Fig. 1. (a) Simplified Schematic and picture (b) of the fuel cell set-up for through-plane measurement in NEUTRA [81], facing the LiF/ZnS scintillator.