Journal of Power Sources 242 (2013) 70–77

Contents lists available at SciVerse ScienceDirect

Journal of Power Sources

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

A study of the effect of water management and electrode flooding on the dimensional change of polymer electrolyte fuel cells

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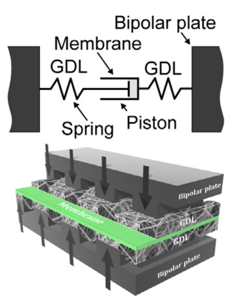
HIGHLIGHTS

- Direct correlation between membrane thickness and conductivity.
- Initial hydration of membrane leads to significant swelling of MEA.
- Dynamic electro-mechanical analysis on operational MEAs during flooding demonstrated.
- Flooding leads to a significant change in membrane conductivity and thickness change.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 28 August 2012
Received in revised form
24 February 2013
Accepted 9 May 2013
Available online 21 May 2013

Keywords:
Water management
Diagnostics
Flooding
Electrochemical impedance spectroscopy
Swelling
Compression

ABSTRACT

Water management and flooding play an important role in the performance and durability of polymer electrolyte fuel cells (PEFCs). In this study, a dynamic electro-mechanical analysis is performed to examine the performance of a working PEFC during hydration transients and flooding events. Cell resistance is measured using electrochemical impedance spectroscopy (EIS), and the stress/strain characteristics – cell compression and membrane electrode assembly (MEA) dimensional change – are studied using a controlled compression unit (CCU).

Ex-situ measurements of membrane thickness as a function of hydration level provide a direct correlation between ionic conductivity and thickness. During initial hydration of Nafion membranes there is a direct relationship between membrane conductivity and dimensional change (swelling) of MEAs. Electrode flooding is found to result in membrane hydration and an increase in stress or strain, depending on the compression mode of the fuel cell. Results suggest that hydration cycles and flooding events can lead to cell degradation due to the stresses imposed.

1. Introduction

Polymer electrolyte fuel cells (PEFCs) have a major role to play in the transition from a carbon intensive economy to a sustainable low carbon future. Intensive research and development in this area has focused on key issues such as catalyst development, low cost materials, performance and durability. A large section of research in the area of performance degradation has focused on water management, an area that has been extensively reviewed [1–3].

Water management inside a PEFC is a function of generation (reaction), various transport processes and the effect on the proton conductivity of the membrane. It has a significant impact on PEFC performance and is one of the major challenges facing the development of this technology [4]. Mechanisms of particular importance include electro-osmotic drag, associated with the migration of protons, which draws water from the anode through

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http://dx.doi.org/10.1016/j.jpowsour.2013.05.045

the electrolyte to the cathode [5]; back diffusion of water from the cathode to the anode due to hydraulic pressure difference; hydration of the membrane which effects the conductivity of the electrolyte [6] and the fact that water is produced at the cathode catalyst layer (CCL) by the oxygen reduction reaction. Accumulation of water limits the performance of the PEFC due to mass transport limitation of flooded electrodes [7]. Effective water management requires careful consideration of fuel cell component design, the materials used and the operating conditions imposed.

When water builds up at the cathode CCL, the mechanisms for removing it include the hydrophobic nature of the elements of the gas diffusion layer (GDL), the operating temperature of the fuel cell (removal as water vapour), and back diffusion through the electrolyte. When the rate of water accumulation at the CCL exceeds the removal rate, the pores in the GDL, and ultimately the flow channel, get blocked (flooding). When this occurs the catalyst is effectively starved of reactant and hence the performance of the cell decreases.

Extensive research has taken place into understanding water management and developing new materials and cell designs to mitigate flooding; this includes examining the effects of GDL material [8,9], PTFE content [10,11], micro-porous layers (MPL) [12,13], porosity of the GDL/MPL structure [14,15], flow field design [16,17], CCL materials and microstructure [18,19] and fuel cell operating conditions [20–22]. The GDL is a particularly important component for water management; an understanding on the various chemical and physical properties of which is particularly important for effective MEA design [23].

In order to study fuel cells and obtain a better understanding of their internal workings, a range of diagnostics have been developed [24]; of these, several key techniques have been used to analyse *in-situ* effects of water management and flooding. An electrochemical impedance spectroscopy (EIS) technique was used by Canut*et al.* to study membrane conductivity during drying and flooding [25]. Barbir *et al.* showed, again using EIS, a relationship between cell resistance and humidification level with pressure drop analysis used concurrently to show the onset of water flooding [26]. Membrane conductivity spatial mapping has been demonstrated by Brett *et al.* [27] and applied by Hakenjos *et al.* in conjunction with temperature distribution analysis to examine water flooding [28].

Experimental diagnostics of water management and in particular water flooding have been extensively researched with particular focus on imaging techniques such as optical visualisation [22,29–31], X-ray imaging [32,33], neutron imaging [34–36] and magnetic resonance imaging [37,38].

The diagnostic techniques applied to the study of water management and flooding have tended to concentrate on the effect of membrane conductivity and the mass transport limiting effect of water in the GDL and electrode. Extensive work has been dedicated to the understanding of Nafion in order to characterise the structure and distribution of water [39]; however, little has been done to examine the effect of dimensional change associated with changes in the hydration of the membrane and its impact on cell and stack performance. This paper applies a new technique to investigate the effects of water flooding on operating PEFCs by looking at the resistance of the membrane and its thickness change in real time.

2. Experimental

Fuel cell operation was carried out using a commercially available cell compression unit (CCU) (Pragma Industries SAS, France), which allows controlled compression (resolution of 0.01 MPa) or displacement of the fuel cell with simultaneous relative real time displacement measurement (resolution of 1 μm). The CCU features a 'floating piston' style fuel cell (Fig. 1) that allows compression to be applied evenly onto the active area of the fuel cell to allow

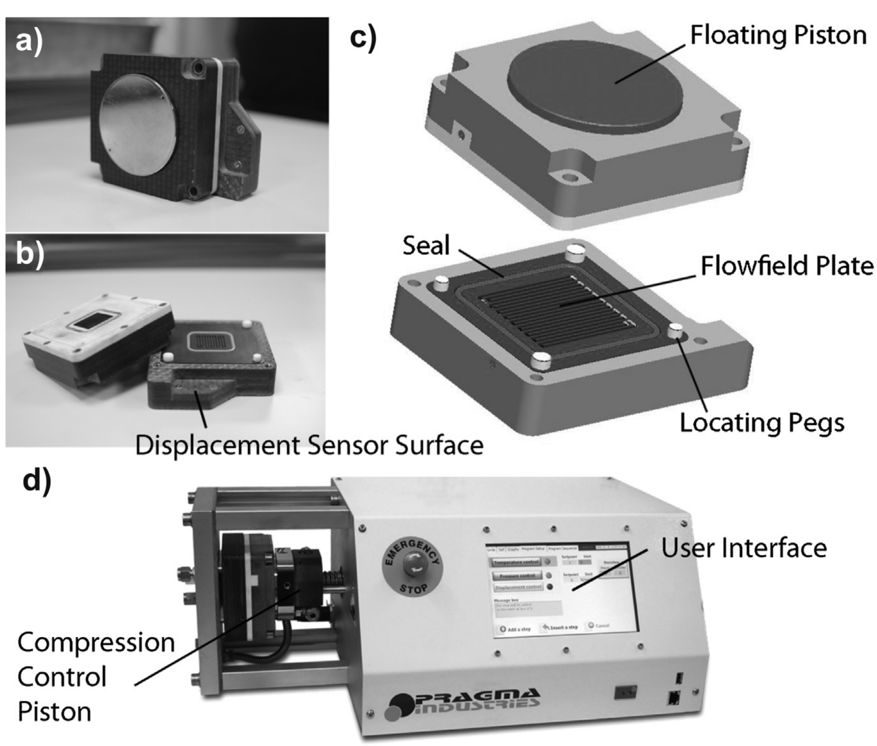

Fig. 1. Cell compression unit (CCU) with floating piston fuel cell. (a–c) show the floating piston cell with the single serpentine flow field design and gas seals, with the displacement sensor surface located on the static anode side of the cell. (d) shows the CCU with the cell loaded with the compression control piston acting on the 'floating' cathode flow field. (Images (c) and (d) courtesy of Pragma Industries SAS, France).