Journal of Power Sources 196 (2011) 4264–4269

Contents lists available at ScienceDirect

Journal of Power Sources

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

Non-dimensional analysis of PEM fuel cell phenomena by means of AC impedance measurements

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ARTICLE INFO

Article history:
Received 3 July 2010
Received in revised form 18 October 2010
Accepted 2 November 2010
Available online 9 November 2010

Keywords:
PEMFC
AC impedance
Charge transfer resistance
Bode plot
Characteristic time scale
Damkhöler

ABSTRACT

AC impedance or electrochemical impedance spectroscopy (EIS) is becoming a fundamental technique used by researchers and scientists in proton exchange membrane (PEM) fuel cell analysis and development. In this work, in situ impedance measurements are presented for a series of operating conditions in a 50 cm^2 fuel cell. The electrode charge transfer resistance was determined from the corresponding arcs of the Nyquist diagrams. The analyses were performed for H_2/O_2 and H_2/air operation at different stoichiometric factors and reactant gases humidification. Characteristic time scales of charge transfer processes at the different operating conditions were estimated from the corresponding Bode plots. These values were used for a non-dimensional analysis of the different fuel cell electrochemical and transport processes, namely electrochemical reaction versus GDL reactant transport. Fuel cell adapted Damkhöler numbers are thus presented, where the results indicate that the GDL diffusion transport is the limiting process for the cases under analysis, especially when air is used as oxidant. Additional analysis of channel convective mass transport versus GDL diffusive mass transport is also presented.

1. Introduction

AC impedance (electrochemical impedance spectroscopy) is an experimental technique widely used in the analysis of electrochemical systems, and has been established as a fundamental diagnostic and research tool for PEM fuel cells during the last years [1,2]. One of the main advantages of AC impedance techniques is that it allows for the determination of the different resistances occurring in a PEM fuel cell, corresponding to the different electrochemical and transport processes: activation or charge transfer resistance, ohmic resistance, and transport or concentration resistance. On contrast, the polarization curve itself does not provide detailed information about the different processes as they are tightly coupled and the polarization curve describes only the integral output of all processes. AC impedance is widely applied to multiple MEA analysis such as influences of the catalyst loading, PTFE content, Nafion content, GDL structure, manufacturing methods, membrane thickness, and others [1]. In an important number of studies EIS analysis has been applied to characterize fuel cell processes. Parthasarathy et al. [3], Antoine et al. [4] and Neyerlin et al. [5] analysed the oxygen reduction reaction (ORR) kinetics. Xu et al. [6] and Neyerlin et al. [7] also analysed the effect of relative humidity conditions on the ORR kinetics of PEM fuel cells. HOR (hydrogen oxidation reaction) kinetics and the effect of CO poisoning were studied by Wagner and Gülzow [8]. Transport losses were analysed by Springer et al. [9], Eikerling and Kornyshev [10], Lefebvre et al. [11], and Saab et al. [12,13], who also analysed the influence of the processing conditions of the electrode. Liu et al. [14] also studied the resistance to the protonic conduction at the cathode ionomer layer using AC impedance. The cell ohmic resistance is also commonly determined by means of AC impedance, as in the work of Cooper and Smith [15]. Romero-Castañón et al. [16], Song et al. [17] or Wagner [18] have applied AC impedance for the evaluation and optimization of the membrane electrode assembly (MEA). Additional applications are the analysis of electrode and GDL flooding or membrane dry-out, what is commonly referred to as MEA State-of-Health (SOH), as in the work reported by Fouquet et al. [19] or Mérida et al. [20]. The results are presented for the stack under analysis, but the analysis of single cells in a stack is also feasible as reported by Hakenjos et al. [21], who presented a measurement set-up featuring a multichannel frequency response analyser (FRA) for the simultaneous measurement of impedance spectra of single cells in a fuel cell stack.

Non-dimensional analysis has proven to be a powerful tool for engineering design and analysis, especially in the chemical engineering field, although its application to complex systems such as fuel cells is not yet fully established [22]. However, there is a need for comprehensive and numerically less expensive description of cell performance in order to implement it into full stack models

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doi:10.1016/j.jpowsour.2010.11.004

Nomenclature

a	specific catalyst surface area ($m^2\ m^{-3}$)
d	channel depth (m)
D_0	diffusion coefficient ($m^2\ s^{-1}$)
Da_{PEMFC}	Damkhöler number
F	Faraday constant (96,485 As mol^{-1})
f_{peak}	frequency (Hz)
h	mass transfer coefficient ($m\ s^{-1}$)
HFR	high frequency resistance (Ω)
I	current intensity (A)
J	diffusive flux ($kg\ m^{-2}\ s^{-1}$)
L	channel total length (m)
M_w	molecular weight ($g\ mol^{-1}$)
P	pressure (Pa)
Q_v	volume flow ($ml_N\ min^{-1}$)
R	bulk resistance (Ω)
R_c	contact resistance (Ω)
R_{cat}	cathode exchange current density ($A\ m^{-3}$)
S_{O_2}	oxygen consumption ($kg\ m^{-3}\ s^{-1}$)
Sh	Sherwood number
T	temperature (K)
V	flow velocity ($m\ s^{-1}$)
y	mass fraction
Greek symbols	
δ_{GDL}	GDL thickness (m)
ε	porosity
ρ	density ($kg\ m^{-3}$)
τ	characteristic time scale (s)
λ	stoichiometric factor
Subscripts	
0	reference state
BP	bipolar plate
ch	channel
chMT	channel convective mass transfer
chrt	flow residence time
diff	diffusion
EC	electrochemical
eff	effective
GDL	gas diffusion layer
MEA	membrane electrode assembly
MT	mass transfer
S	catalyst surface

that cannot be analysed by means of Computational Fluid Dynamics (CFD) models except for small-scale units [23]. Therefore, a detailed description based on dimensional analysis of the cell phenomena would be of high relevance for further stack research and development, and important contributions are already in progress as established in the work of Gyenge [22].

This work presents AC impedance results for a 50 cm^2 PEM fuel cell. The electrode activity and membrane protonic conductivity were determined elsewhere [24] based on the measurements performed for different operating conditions, aimed at the fuel cell parameter estimation needed for numerical CFD simulations [25]. In this work, the application of AC impedance measurements to the non-dimensional analysis of PEM fuel cell phenomena is presented, and results for a 50 cm^2 PEM fuel cell are reported. In particular, Nyquist and Bode plots are presented for a fuel cell with serpentine flow field, operating with oxygen and air at different humidification conditions. Measurements at several current densities are presented in order to better assess the influence of the current density on the limiting processes affecting the performance of the cell.

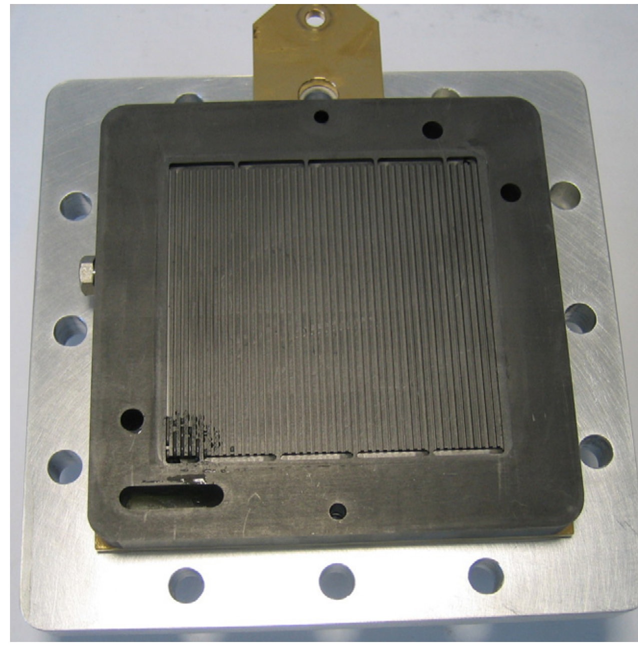

Fig. 1. Serpentine bipolar plate used in the analysis.

2. Cell description, operating conditions and experimental analysis

2.1. PEM single cell description

The single fuel cell analysed consists of commercial graphite bipolar plates with a five-channel serpentine flow field design (Fig. 1) from ElectroChem Inc. (USA). The GDL is a Sigracet 10 CC from SGL Group (Germany), with 420 μm thickness, porosity 0.82 and 10% PTFE content, featuring a micro porous layer (MPL). The 50 cm^2 membrane used is a CCM type (catalyst coated membrane) from Baltic fuel cells (Germany), with Nafion-117 and 0.3 mg Pt/cm^2 and 0.6 mg Pt/cm^2 catalyst load in anode and cathode respectively. The ratio of ionomer/carbon (I/C ratio) is 1.2/2, with 70% Pt over carbon, and a ratio ionomer/catalyst = 1/4.

2.2. Operating conditions

The operating conditions used in the analysis are listed in Table 1. Cell temperature and pressure were maintained constant during all the analysis at 60 °C and 4 bar (absolute), which are the operating conditions recommended by the cell supplier.

2.3. Experimental analysis

The experimental work was conducted in a FuelCon CT-1000 (Germany) test station, located at INTA facilities. The experimental methods and procedures were carried out following the details described in the FCTESTNET documents for single cells [26], where

Table 1
Operating conditions used for the experimental analysis.

λ_a	RH_a (%)	λ_c	RH_c (%)	Oxidant
1.5	60	5.0	60	Air
1.5	100	5.0	100	Air
2.0	100	10.0	100	Oxygen