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Model-supported characterization of a PEM water electrolysis cell for the effect of compression



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ABSTRACT

This paper investigates the influence of the cell compression of a PEM water electrolysis cell. A small single cell is therefore electrochemically analyzed by means of polarization behavior and impedance spectroscopy throughout a range of currents $(0.01 \, \text{A cm}^{-2} \text{ to } 2.0 \, \text{A cm}^{-2})$ at two temperatures $(60^{\circ} \text{C} \text{ and } 80^{\circ} \text{C})$ and eight compressions $(0.77 \, \text{MPa}-3.45 \, \text{MPa})$. Additionally, a computational model is utilized to support the analysis. The main findings are that cell compression has a positive effect on overall cell performance due to decreased contact resistances, but is subject to optimization. In this case, no signs of severe mass transport problems due to crushed transport layers are visible in either polarization curves or impedance plots, even at high currents. However, a Tafel plot analysis revealed more than one slope throughout the current range. The change in the Tafel slope is therefore discussed and connected to the electrochemical reaction or an ohmic contribution from a non-electrode component.

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1. Introduction

Energy storage is considered crucial for a successful transition to carbon neutral energy production. A hydrogen based energy production can be a key technology as hydrogen can be stored and distributed as a liquid or gas, and utilized in fuel cells (FC) to produce electrical energy and heat [1]. Nowadays, the main source of hydrogen is from steam reformed hydrocarbons or alcohols [2] with inevitable release of greenhouse gases. However, hydrogen can also be produced through water electrolysis (WE), where water is electrochemically split into its elements, hydrogen and oxygen. If the required electrical energy comes from renewable energy sources, this process can be considered CO₂ neutral. Additionally, electrolyzers can potentially provide grid-services for frequency stability [3].

Polymer electrolyte membrane (PEM) electrolysis can be considered an evolving technology that is entering the market and has attracted more research interest in the past few years due to certain advantages [2,4]. Among others, PEM electrolyzers can be manufactured in compact systems due to their solid electrolyte

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In the context of grid stability, they are especially interesting due to good dynamic response times and start/stop behavior, and a wide operating range [5]. Comparably fast starts are possible due to the operation at low temperatures of around 60° C to 100° C. Furthermore, the hydrogen gas purity can be maintained fairly high at 99.999% which makes further purification redundant [6]. PEM electrolyzers offer the possibility to operate under high pressures which allows hydrogen gas production at more than 100 bar [7]. Depending on the application, internal pressurization may make external gas compressors redundant and therefore reduce system complexity and cost [8]. Pure oxygen as a byproduct might be utilized and monetized in PEM FC or other applications, or released to the atmosphere [9]. The major drawback is the high cost due to the need of precious materials. Additionally, acceptable lifetime under dynamic operation has yet to be proven [5].

membrane with small footprints and offer relatively high efficiency.

An improvement in performance and durability requires proper characterization methods. Besides classic polarization curves (IV), other tools that researchers employed on WE include current interrupt, cyclic voltammetry, more visual approaches, such as electron microscopy, and electrochemical impedance spectroscopy (EIS) [10,11]. EIS is a versatile characterization technique that is already widely used in fuel cell applications [12,13] and many researchers have also implemented EIS as a tool in their investigations on PEM electrolyzers [14,15]. With the above





Electrochimica Act

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mentioned characterization techniques, performance investigations for different membrane-electrode-assemblies (MEAs) in terms of materials and loading were compared [16,17], as well as studies on the impact of the feed water flow type [18] and stoichiometry [19].

However, not many systematic studies are available in the literature that reveal more insight about how various operation conditions affect cell performance. Moreover, with the design consisting of multiple solid layers clamped between two end plates, the compression pressure is an interesting parameter and possibly subject to optimization. While the compression pressure is reasonably well researched for FC, WE lacks studies [5,20]. EIS can provide useful insights for analyzing compression [21,22].

This paper investigates the performance of a single cell under different current and temperature operation points at various compressions. The purpose is to better understand what the most relevant operation parameters are. The experimental work is supported by computational modelling in order to extract characteristic parameters such as the exchange current density. The combination of experimental data and a model approach also helps to divide the different resistance contributions between the components.

2. Methodology

All experiments are carried out on a single cell set-up. Custom made MEAs by EWII Fuel Cells A/S (Denmark) with an active area of 2.89 cm^{-2} (1.7 cm^{*}1.7 cm) are tested. The assembly consists of a commercial Nafion 117 membrane coated with Iridium oxide (IrO₂, 0.3 mg cm^{-2}) on the anode and carbon-supported Platinum (Pt/C, 0.5 mg cm^{-2}) on the cathode. The porous transport layers (PTL) are carbon cloth (Sigracet 35DC) and titanium felt on the cathode and the anode, respectively. The Ti felt is 350 µm thick, with a porosity of 81% and a fiber diameter of 20 μ m. Additionally, an Iridium contact layer is employed at the anode between the catalyst layer and the Ti-felt. The layer consists only of pure Iridium metal with a loading of 2.48 mg cm^{-2} . The purpose of this layer is to provide better contact between the components and may be considered when calculating the total noble metal loading of the cell, especially when taking financial considerations into account. Other studies of this cell layout have been performed [23]. The coarse metallic Ir may oxidize fully or partially to IrO₂ during operation, which has to be considered when performing degradation studies [24].

The test bench is designed so that the cell is heated by the feed water, which is held at a constant temperature. The temperature is monitored close to the water inlet channel at the anode and close to the hydrogen outlet channel at the cathode. The water flow is constant around 270 mL min⁻¹, which represents a high overstoichiometry. That way, steady temperature is established and mass transport contributions are minimized.

A Gamry Reference 3000 potentiostat/galvanostat is used for all IV and EIS measurements in a two-electrode set-up, meaning that the anode is connected to the working electrode and the cathode acts as the reference electrode. Fig. 1 illustrates the experimental set-up.

Polarization curves are measured in variable steps of $0.01 \,\mathrm{A} \,\mathrm{cm}^{-2}$ to $0.2 \,\mathrm{A} \,\mathrm{cm}^{-2}$ in ascending direction. Each current step is held for 60 s before measuring the potential. The feed water flow rate is kept constant, which implies that the stoichiometry is in fact slightly changed throughout the experiment due to changes in current according to Faraday's law of electrolysis. However, the effect should not have a significant impact on the measurements since the over-stoichiometry is very high at any operation point with around 17000 at $1.0 \,\mathrm{A} \,\mathrm{cm}^{-2}$.

Impedance spectra are, if not otherwise annotated, measured



Fig. 1. Test set-up including the cell, feed water tank, data acquisition, and power source.

galvanostatic from 60000 Hz to 0.1 Hz with 10 points per decade. The potentiostat supplies a variable AC disturbance current of 5% of the DC current operation point. The data is fit to an equivalent circuit as depicted in Fig. 2. The circuit and its interpretation is discussed more in Sections 3.2 and 3.3.

The MEA compression is controlled through the clamping pressure applied to the cell in eight steps from 0.77 MPa to 3.45 MPa (C0 to C7) according to Table 1.

Estimating the actual clamping pressure applied to the MEA precisely is not as straight forward as it may seem. Additionally, the pressure distribution is generally a topic of interest which is investigated and optimized experimentally, and through simulations [25,26]. Different approaches in fuel cell research can be found in the literature, which can be transferred to electrolysis research. However, the mechanisms may not be strictly derived from fuel cells since the geometry and fluid compositions differ. Furthermore, it is arguable which area should be considered to calculate the compression. In this work, four die springs are used to establish a pressure distribution as homogeneous as possible. Actual compression pressure as well as homogeneity are tested exsitu with a pressure sensitive film in Sec. 3.1. The film exhibits a red color at points of pressure that is dependent on the pressure value. The color intensity was analyzed with image processing software and can be related to the applied pressure through reference curves supplied by the manufacturer.

Moreover, contact resistances are measured between several components that are accessible. Each contact is investigated separately through a sample with a high precision Ohmmeter while being clamped between two copper plates. The three experimentally determined contact resistances are between the anode bipolar plate (BPP) and the anode current distributor (CD), between the anode BPP and the Ti felt, and between the cathode BPP and cathode CD.



Fig. 2. Equivalent circuit for impedance data fit.

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