



Modeling and experimental validation of a unitized regenerative fuel cell in electrolysis mode of operation



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ABSTRACT

Unitized regenerative fuel cell (URFC) is considered to be the compact solution to generate and utilize hydrogen. It possesses combined capabilities of operating in fuel cell and electrolyser modes. In the present study, the performance of a URFC in electrolyser mode is modelled and also experimentally validated. The performances are being modelled using a combination of structural and CFD analysis tool. The effect of the operating gas pressure on the variation in the contact pressure between GDL and BPP on the performances are studied. The clamping pressure, as well as the operating pressure of the electrolyser, are seen to have a high impact on the contact resistance and thereby the performance as well. It is observed that the simulated polarization behavior is in good agreement with the experimental results. To restrict the area specific resistance below $150 \text{ m}\Omega \text{ cm}^2$ the operating pressure should be maintained below 5.9 bar at clamping pressure of 1.5 MPa.

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1. Introduction

In the light of global warming and negative consequences of the dependence on the conventional fossil fuel resources, hydrogen fuel based economy is believed to be a promising alternative solution [1]. Although hydrogen fuel offers several advantages compared to the fossil fuel and also abandoned in nature, it must be derived from other resources and requires energy input. Waste heat from nuclear power plant [2–4] could be considered as an intermediate solution for the energy sources and renewable energy viz., solar energy [5–7] and wind energy [8,9] can be considered as a long-term sustainable solution for the input for deriving hydrogen fuel. Different roles of hydrogen in the renewable system have been demonstrated in industrial scale [10–12] as well as small scale [13]. In renewable system, hydrogen is derived from water using electrolyser and it is utilized in fuel cells to meet electrical demand.

Fuel cells are electrochemical energy conversion devices that possess several operational advantages include quick start-up, high efficiency, low operating temperature, noise free operation and lower emissions than fossil fuel-based devices [14–17]. Alike fuel cells, electrolysers are also electrochemical devices capable of splitting water into hydrogen and oxygen electrochemically. In

most of the renewable system where hydrogen is explored as a medium for energy storage to cater the mismatches in the demand and supply of power, combination of electrolysers and fuel cells are explored. Unitized regenerative fuel cells (URFCs) are able to work in both fuel cell mode and an electrolyser mode [18,19]. This makes them be an alternative candidate to club with renewable energy sources for hydrogen generation and utilization, particularly for the standalone system [20–22]. However, URFCs are still undergoing intense research and development to address issues related to the complexity associated with the scale ups and durability. The durability is affected due to the redox reaction of oxygen at oxygen electrode resulting in faster decay in performance compared to that of fuel cells and electrolysers individually [23,24]. A URFC is composed of the membrane electrode assembly (MEA) with bipolar plate (BPP) on each side which is further enclosed within a pair of end plates. The entire assembly is brought into an electrical contact using clamping pressure applied using bolts. If clamping pressure is non-uniform and non-optimized, the performance of the cell will be affected. The decrease is primarily due to interfacial ohmic loss, which may be significant if non-optimal clamping pressure is applied. In particular, the loss is very sensitive to the electrical contact at the interface of the BPP and gas diffusion layer (GDL) [25,26].

As the issue of the contact resistance is also of relevance to the fuel cells, extensive work has been carried out to study its behavior [27–29] in connection with the clamping pressure [30,31].

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Researchers have suggested changing the surface topology of BPP viz., surface treatment [32] and conductive coatings [33–35] as the possible solutions to reduce Interfacial Contact Resistance (ICR). Oyarceet et al. [36] observed a decrease in contact resistance with respect to the temperature and relative humidity (RH) of gasses. However, higher current density resulted in the production of excessive water on the cathode side which increased the contact resistance, especially for the uncoated SS316 sample. Bates et al. [37] analytically observed the contact pressure distribution between GDL and BPP for the PEMFC stack. It was observed that the pressure at the interface was negligible regardless of the clamping pressure. Dey et al. [38] experimentally validated their theoretical model which dealt with the ICR between the electrode and interconnect for the solid oxide fuel cell.

Lee et al. [39] studied the influences of the GDL and clamping torque on the performances of the fuel cells at a fixed stoichiometric flow rate of gases. It was observed that incompressible GDL showed the highest performance at low clamping torque. However, increase in clamping torque enhanced the performance of cell with softer GDL which was attributed to respective changes in porosity and contact resistance.

Bograchev et al. [40] proposed a 2D elastoplastic model (elastic for metallic components and plastic for membrane) for fuel cell against mechanical stresses. The model proved plastic deformation of membrane below gasket and bipolar plate at 15.8 N-m which was confirmed experimentally.

Although structurally fuel cells and electrolyzers are almost similar, different materials are used for the components in the electrolyser as their operating environment is different. Based on such concept, Selamet et al. [41] examined the effect of the bolt torque on the contact resistance in the PEM electrolyser using different gaskets. They observed the pressure distribution with pressure sensitive films between the gaskets and found that contact resistance and thereby performance did not decrease drastically for PTFE gasket under different clamping pressure. Siracusano et al. [42] experimented with the effect of thickness variation of GDL/current collector on ICR for PEM electrolyser stack and found that thicker titanium grid had lower contact resistance possibly due to better contact with BPP. Moreover, no further enhancement in the performance was observed between 15 and 25 kg cm⁻².

However, the effect of the operating gas pressure on contact pressure and thereby ICR has not been studied in the case of electrolyzers. As gas pressure rises, it is suspected that it would try to counter the contact pressure that maintains electrical contact between BPP and GDL. This will definitely cause the contact pressure between them to drop during operation which will end up in increasing the ICR. Authors, therefore, investigate such effect at different clamping pressure for URFC in the electrolysis mode. It is important to observe the contact pressure developed at the interface of GDL and BPP which would relate to corresponding contact resistance between them. Experimental validation has been done to support the modeling. The analysis done herewith shall be useful in predicting the approximate pressure of gas that the cell could withstand against acceptable contact resistance for a given clamping pressure.

2. Methodology

Interfacial contact resistance mainly, between BPP and GDL inside a unitized regenerative fuel cell, contributes significantly, under a non-optimal clamping condition. It is also affected by the operating pressure. Hence, it is important to investigate the contact resistance behavior of the interfacial plates under different clamping pressure and gas operating pressure. The experimental details for measuring the interfacial contact resistance under

different clamping pressure is discussed in the following sections.

2.1. Experimental setup

2.1.1. Measurement of contact pressure distribution

Spatial distribution of interfacial pressure between different components in the URFC plays an important role in the overall performances. In general, the spatial distribution of the contact pressure between the electrode and bipolar plate is observed in a large area cell which dictates the local internal resistance of a cell. In the present study, pressure sensitive films were explored to study the contact pressure distribution at the interface. Pressure sensitive films suitable for use in the range from 0.2 till 0.5 MPa were used in present study.

2.1.2. Measurement of ICR

In order to convert the interfacial resistance distribution from the pressure distribution, obtained from the pressure sensitive films, the correlation between the contact pressure and contact resistance is important. In the present study, the contact resistance between a sample of SS 316 coated with titanium nitride (TiN) and carbon paper, were cut into appropriate size (1 cm × 1 cm). The experimental in-house device used for the measurement of contact resistances is shown in Fig. 1. The threaded screw was used to convert rotary motion into axial motion. The contact pressure with respect to spindle turns was calibrated using pressure sensitive films. Carbon paper was fixed on the bottom of the top plate whereas coated SS sample was fixed on top of bottom plate ensuring that the samples could face each other all the time. The connections were drawn from the samples and connected to the DAQ that was monitored through a computer.

2.1.3. Membrane electrode assembly

In order to characterize and validate the simulation results, MEA was prepared by initially treating the membrane (NR-212) with HNO₃, H₂O₂ and H₂SO₄ [43] respectively for 1 h each to remove any organic and inorganic impurities and sulfonate at the end. GDL was prepared by coating an ink of Vulcan XC-72 (Loading 1.5 mg cm⁻²) mixed with PTFE (15 wt%) in iso propyl alcohol (IPA) [44]. The coated paper was later sintered at 350 °C for 30 min to help PTFE distribute uniformly over the paper. The catalyst ink with 0.1 mg cm⁻² loading of 40% Pt/C with 7 wt% of nafion solution was then coated on GDL with a soft brush to make hydrogen electrode. However, 40% Pt/C and IrO₂ together were mixed in IPA along with 30 wt% of N.S. (total loading 0.5 mg cm⁻²) to coat the ink for oxygen electrode. The treated membrane was then sandwiched between the electrodes in a hot press with 65 kg cm⁻² at 137 °C for 2.5 min to fabricate the MEA [45].

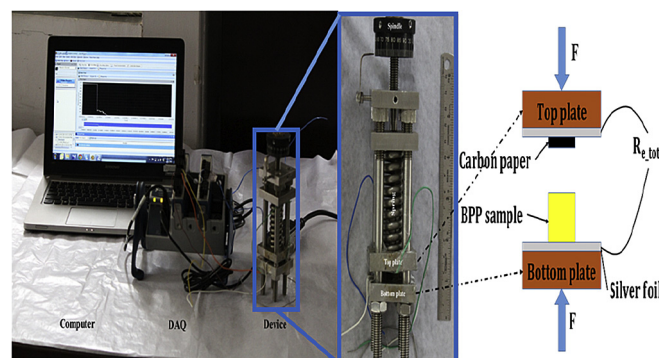


Fig. 1. Experimental setup (left) with device schematically illustrated (right).

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