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 arising from 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.

Keywords: Unitized regenerative fuel cell, Electrolyser, Contact resistance, Operating pressure, Clamping pressure

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Highlights

- Dependence of ICR on clamping pressure is explored.
- Maximum operating pressure as a function of clamping pressure is investigated.
- Impact of ICR on the URFC performance in electrolysis is evaluated.

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, electrolyzers 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 electrolyzers and fuels 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 electrolyzers 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]. 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.

Bogachev 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 electrolyzer 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 electrolyzer 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 electrolyzer 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 to 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 one hour 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 350⁰C 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 137⁰C for 2.5 min to fabricate the MEA [45].

2.1.4 URFC assembly

The cell assembly was prepared by holding MEA of an active area of 22.5 cm², between a PTFE gaskets and a TiN coated BPP with serpentine flow field configuration followed by a silicon rubber sheet and an end plate on both sides. The studs were welded perpendicular to one of the

end plates to reduce stacking time and act as guiding pins for the components. The different components used in the URFC assembly is shown in Fig. 2.

2.2 Modeling of electrolysis performance

Electrolysis performance has been characterized using a 3D, computational fluid dynamics (CFD) model. The effect of ICR at the interface of GDL and BPP has been included in the model. The simulated data has been validated with experimental polarization data and governing parameters have been found out.

2.2.1 Assumptions

It is assumed that the flow of gases is to be steady, laminar and single phase throughout the domain. In the case of water electrolysis, since hydrogen generation is found to be simpler than water oxidation, cathode contribution to overpotential is negligible compared to the anode. The resistivity of the electrolyte is assumed to be constant as the cell is simulated over low temperature (< 340 K) and pressure (0-8 bar) ranges. Porosity in the electrode, as well as GDL, is assumed to be uniform and the permeability to be isotropic. The transport of charge was assumed to be normal to the electrode and membrane area. External walls of the domain were kept at room temperature.

2.2.2 Governing equations:

Water can be dissociated into respective gases by supplying the electrical energy (ΔG) and the thermal energy ($T\Delta S$) i.e., the enthalpy (ΔH) of water at a given instant (eq 1 and 2).

$$\Delta H = \Delta G + T\Delta S \quad 2$$

The reactions at oxygen and hydrogen electrodes respectively could be summarized in following way:

The reversible potential [44] at a temperature T (in kelvin) and 1 atm pressure could be found out using Eq. 5.

$$E_{rev}^T = E_{rev}^0 - 1/nF \left\{ (T - 298.15) \times (\Delta C_p - \Delta S^0) - T \times \Delta C_p \times \ln \left[\frac{T}{298.15} \right] \right\} \quad 5$$

However, Nernst potential [44] gives open circuit voltage considering the effect of operating pressure (Eq. 6).

$$E^N = E^{P,T} = E_{rev}^T - (RT/nF) \times \ln \left\{ \frac{P_{H_2} \times (P_{O_2})^{1/2}}{P_{H_2O}} \right\} \quad 6$$

Volumetric current densities [46] at anode and cathode can be respectively calculated using Butler-Volmer equation as

$$J_{an} = \zeta_{an} j_{an}^{ref} \left\{ \frac{[C]}{[C]_{ref}} \right\}^{y_{an}} \left\{ e^{-\alpha F \eta_{an}/RT} - e^{+\alpha F \eta_{an}/RT} \right\} \quad 7$$

$$J_{cat} = \zeta_{cat} j_{cat}^{ref} \left\{ \frac{[C]}{[C]_{ref}} \right\}^{y_{cat}} \left\{ e^{-\alpha F \eta_{cat}/RT} - e^{+\alpha F \eta_{cat}/RT} \right\} \quad 8$$

And the source terms and sink terms [47] in case of electrolysis could be written as following;

$$S_{H_2} = \left\{ M_{w,H_2} / 2F \right\} J_{cat} > 0 \quad 9$$

$$S_{O_2} = \left\{ M_{w,O_2} / 4F \right\} J_{an} > 0 \quad 10$$

$$S_{H_2O} = \left\{ M_{w,H_2O} / 2F \right\} J_{an} < 0$$

Total resistance [28] that arises from ionically and electronically conducting mediums are given by

$$R_{tot} = R_{act} + R_{ohmic} + R_{e\_tot} \quad 12$$

$$R_{e\_tot} = R_{GDL} + R_{BPP} + \left(R_{ICR} / A_{electrode} \right)$$

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The local current density can now be deduced from operating potential with the help of the following equation:

$$J_{avg} = \frac{E}{R_{tot} \times A_{electrode}} \quad 14$$

2.2.3 Computational procedure

The geometry was created in Ansys 14.5 and hexahedral type meshing was adopted in all domains. Mesh optimization was carried out after importing the geometry in solver (Ansys Fluent 16). The governing equations were discretized using SIMPLE algorithm and boundary conditions were set on respective surfaces followed by solution initialization. The solution was under-relaxed against the stability of the species and energy conservation equations. This was achieved by setting the value of 0.8 for the species and 0.8 for the energy term in the beginning. In due course of the solution, these values were increased depending upon the nature of the solution. The solution was deemed to be converged when the residuals of all the conserved quantitative dropped below 10^{-6} .

2.2.4 Boundary conditions:

The average molar flow rate for hydrogen produced during water electrolysis could be calculated using following equation.

$$\dot{m}_{H_2} = \frac{J_{avg} \cdot A_{electrode}}{2F} \quad 15$$

Where, J_{avg} is the average current density applied to the cell at a particular voltage; $A_{electrode}$ is the active area of the electrode.

The temperature of water at the inlet was set to be 333 K for all simulations and pressure outlet type boundary condition (atmospheric pressure) was enforced at the outlets. Area specific contact resistance and operating pressure i.e. gas pressure were varied with respect to operating conditions. Electric potential was specified at the anode side whereas the same was assumed to be zero at the cathode side.

3. Results and discussion:

3.1 Interfacial contact resistance

Fig. 3 shows the variation of contact resistance with contact pressure (CP) between carbon paper and TiN based bipolar plate. It could be seen that the area-specific ICR decreased exponentially with the increase in the contact pressure as reported by L. Wang et al. [48] thereby emphasizing the fact that the contact pressure plays a dominant role in the internal resistance of the URFCs. The value of the area specific ICR decreased with the increase in the contact pressure and dropped to a value of $0.059 \Omega \text{ cm}^2$ at 0.46 MPa. This decrease is attributed to more area of carbon paper coming in contact with BPP as and when the pressure was increased. The experimental results have been fitted using following empirical equation which was further incorporated in CFD model.

$$R_{ICR} = -1.02012 + \left(2.92053/CP\right) + (0.09624 \times CP) \quad 16$$

Although there has been slight deviation of the fit from experimental curve predominantly in the initial range of contact pressure (≤ 2.6 MPa), it was acceptable as higher contact pressures have been considered in most of the simulations. The slight decrease in bulk resistance of carbon paper was observed as it gradually got compressed with an increase in contact pressure; mainly because of bringing more fibers in contact with each other.

3.2 Distribution and validation of contact pressure

Clamping pressure of 1 MPa has been taken as a case for comparative study and detailed analysis of spatial stress variation over electrode has been carried out. The simulated distribution of the contact pressure on the electrode surface is shown in Fig. 4a and the pressure distributions of similar assembly obtained from the pressure film measurement are presented in Fig. 4b. Higher stresses on the edges could be observed in both the studies due to excessive pressing on the electrodes by the BPP mainly due to the positioning of bolts along the edges of the cell. Fig. 4c shows the statistical distribution of the contact pressure distribution obtained from the simulated results. From the figure, it is clear that majority of the areas lies between 0.8 MPa to 1 MPa. The average contact pressure obtained from the simulation were experimentally validated for different clamping pressures. Approximately, a linear relationship was observed between the two in the case of both experimental as well as simulation results (Fig. 4d) and both seem to be in good agreement. The properties of the materials those were used in the structural simulation are summarized in Table 1. The properties for the gaskets were obtained experimentally using nano-indentation equipment.

3.3 Gas pressure vs contact pressure at different clamping pressure

Structural simulation for the cell was carried out for different gas pressures at various clamping pressures. The variation in the average contact pressure at the interface of the BPP and GDL is shown in Fig. 5. It was observed that the average contact pressure decreases linearly with increase in the gas pressure. The effect was observed as the gas pressure tried to separate out the contact between BPP and MEA and thereby GDL.

3.4 ICR vs gas pressure at different clamping pressure

The variation of operating gas pressure was further extended to see its impact on ICR at different clamping pressure as given in Fig. 6. In the case of lower clamping pressure (0.5 and 0.75 MPa), little increase in gas pressure further separated GDL and BPP locally. As there was already low contact pressure at the interface, the local separation led to an increase in the area specific ICR exponentially as investigated by P. Zhou et al. [49]. However, in the case of higher clamping pressure (1.25 and 1.5 MPa), relatively more gas pressure was required to separate out GDL and BPP. Therefore, slow increase in ICR was initially seen with the increase in gas pressure. The exponential increase in ICR after a threshold value (e.g. 6 bar in case of 1.5 MPa) was observed because of low average contact pressure at the interface. The maximum limit of the operating pressure for the URFC in the electrolyser mode of operation was estimated from the plot assuming the maximum allowed contact pressure to be $150 \text{ m}\Omega \text{ cm}^2$ and observed that the maximum operating pressure varies linearly with the clamping pressure as given in the inset of Fig. 6.

3.5 Polarization behavior

The polarization behavior of the URFC in the electrolyser mode was studied at 65°C with a flow rate of water maintained approximately at 10 mL/min at constant voltage mode (CV mode). The

settling time for current against each voltage set was 90 s. The simulated polarization curve obtained from CFD modelling was validated with experimental results as shown in Fig. 7. It is observed that simulation results were in the good agreement with the experimental results. The effect of ICR was further studied on performances of the URFC. The variations in the polarization behavior for different clamping pressure is shown in Fig. 8a, keeping the operating pressure constant at 4 bar. Similarly, the variation in the polarization behavior of the URFC for different operating pressures keeping the clamping pressure constant at 1 MPa are presented in Fig. 8b. The effect of an increase in gas pressure was seen in terms of increasing the cell resistance thereby reducing the performance.

4 Conclusions

In the present study, the performance of a URFC in electrolyser mode is modelled and also experimentally validated. The effect of the operating gas pressure arising from the variation in the contact pressure between GDL and BPP is studied. It is observed that the contact pressure decreases almost linearly with the increase in the operating gas pressure. Such trend is observed for all clamping pressures. Distribution of area specific ICR is observed to be relatively higher at the central portion of the electrode and lower at the peripheral portion of GDL. However, the exponential relation between ICR and contact pressure makes relatively larger clamping pressure to hold more gas pressure (as observed by C. Turan et al. [50]). 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. It should be noted that excessive clamping could bring ICR little lower, however, it will severely affect the cell components like GDL and membrane. It will also increase the mass transfer losses. The further incorporation of ICR in CFD for simulating the performance of URFC in electrolyser leads to an important conclusion of its impact on how

drastically the performance could vary. Operating gas pressure and clamping pressure have been correlated with each other in the study with the help of modelling and it is observed that the maximum operating pressure of the URFC varies linearly with the clamping pressure.

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Nomenclature:

$A_{electrode}$	Active area of the cell, m^2
α	Transfer coefficient
$[C]$	Local species concentration, $kmol\ m^{-3}$
$[C]_{ref}$	Reference concentration, $kmol\ m^{-3}$
CP	Average contact pressure, bar
ΔC_p	Specific heat of the reaction, $J\ mol^{-1}\ K^{-1}$
E	Operating potential of electrolyser, V
E_{rev}^0	Reversible potential of electrolyser at STP conditions, V
E_{rev}^T	Reversible potential of electrolyser at temperature T, V
$E^N = E^{P,T}$	Nernst or reversible potential at different operating pressure and temperature, V
F	Faraday constant ($96485\ C\ mol^{-1}$)
ΔG	Gibbs free energy (electrical input for water splitting), $kJ\ kg^{-1}$
ΔH	Change in enthalpy, $kJ\ kg^{-1}$
J_{an}	Volumetric current density for anode, $A\ m^{-3}$
J_{cat}	Volumetric current density for cathode, $A\ m^{-3}$
j_{an}^{ref}	Reference exchange current density per active surface area for anode, $A\ m^{-2}$
j_{cat}^{ref}	Reference exchange current density per active surface area for cathode, $A\ m^{-2}$
J_{avg}	Average current density supplied for water electrolysis, $A\ cm^{-2}$
ζ_{an}	Specific active surface area of anode catalyst, m^{-1}
ζ_{cat}	Specific active surface area of cathode catalyst, m^{-1}
$\dot{m}_{H_2}$	Molar flow rate of hydrogen produced, $mol\ s^{-1}$
M_{w,H_2}	Molecular weight of hydrogen, $kg\ mol^{-1}$

M_{w,O_2}	Molecular weight of oxygen, kg mol ⁻¹
M_{w,H_2O}	Molecular weight of water, kg mol ⁻¹
MEA	Membrane electrode assembly
n	Number of electrons per mole, mol ⁻¹
η_{an}	Activation overpotential of oxygen electrode (anode), V
η_{cat}	Activation overpotential of hydrogen electrode (cathode), V
OCV	Open circuit voltage, V
P_{H_2}	Partial pressure of hydrogen = operating pressure of hydrogen, atm
P_{O_2}	Partial pressure of oxygen = operating pressure of oxygen, atm
P_{H_2O}	Activity of water
R	Universal gas constant (8.314 J mol ⁻¹ K ⁻¹)
R_{BPP}	Bulk resistance of BPP, Ω
R_{GDL}	Bulk resistance of GDL, Ω
$R_{connections}$	Bulk resistance of electrical connections, Ω
R_{e_tot}	Total electronic resistance, Ω
R_{tot}	Total resistance in the cell, Ω
R_{act}	Resistance due to activation, Ω
R_{ohmic}	Ionic resistance, Ω
R_{ICR}	Resistance due to ICR, $\Omega\text{ m}^2$
ΔS	Change in entropy (thermal input for water splitting), kJ kg ⁻¹ K ⁻¹
ΔS^0	Entropy of the reaction at STP condition, J mol ⁻¹ K ⁻¹
S_{H_2}	Source term for hydrogen, kg m ⁻³ s ⁻¹
S_{O_2}	Source term for oxygen, kg m ⁻³ s ⁻¹

S_{H_2}	Source term for water, $\text{kg m}^{-3} \text{s}^{-1}$
T	Absolute temperature of electrolysis, K
y	Concentration dependence

Abbreviations

BPP	Bipolar plate
CFD	Computational fluid dynamics
CP	Average contact pressure
DAQ	Data acquisition system
FC	Fuel cell mode
GDL	Gas diffusion layer
ICR	Interfacial contact resistance
IPA	Iso-propyl alcohol
MEA	Membrane electrode assembly
PEM	Polymer electrolyte membrane/ proton exchange membrane
PTFE	Polytetrafluoroethylene
RH	Relative humidity
Ti	Titanium
TiN	Titanium nitride
URFC	Unitized regenerative fuel cell

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Table 1. Material properties of different components of URFC

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

Fig. 5.

Fig. 6.

Fig. 7.

Fig. 8.

Table 1

Component	Material	Young's modulus (GPa)	Density (kgm ⁻³)	Poisson's ratio
End plate [51]	SS 316	193	8000	0.3
Silicon Gasket	Silicon	0.017	1500	0.47
BPP [51]	SS 316 coated with TiN	193	8000	0.3
PTFE gasket	PTFE	79	2200	0.46
GDL [52]	Toray paper (TGP-H-120)	10	450	0.3
Membrane [53]	NR-212	0.25	2000	0.25