Investigation on the performance of reversible polymer electrolyte membrane fuel cell during the electrolysis phase

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Abstract. Due to its sustainable and zero-carbon potential, electrolysis of water is considered a suitable alternative for the production of green hydrogen. The electrolysis accounts for 2 to 5% of hydrogen production worldwide. The primary objective of this study is to simulate the performance of the RPEMFCs in producing green hydrogen. The investigation was more specifically carried out on the reversible polymer electrolyte membrane fuel cell (RPEMFC) unit with Nafion-115 membrane with different anode and cathode catalyst loadings at the current density of 1 A/cm2. This study acknowledges the significance of the ramp-up phase in the investigation of the steady state electrolysis process at fixed current density. Also, it is observed that cell voltage tends to increase with an increase in current density from 0.1A/cm2 to 1A/cm2. The operating voltage was reported as 1.89 V at the current density of 1A/cm2. Furthermore, the voltage efficiency of the reversible polymer electrolyte membrane fuel cell (RPEMFC) electrolysis was 65% under the given conditions.

1 Introduction

An engineering system that is capable of producing electrical energy from chemical energy is known as a fuel cell. Similarly, combustion-based systems also involve the transformation of chemical energy into mechanical or electrical energy [1]. Therefore, both systems may look similar; however, in reality transformation process of both systems is quite different. In contrast to combustion based systems; in case of simple fuel cells, electrical energy is produced directly from the chemical reaction using an electrochemical reaction by harnessing electron's energy due to their conversion from higher energy bonds to low-energy bonds [2]. Other than simple energy generation, fuel cells can also be used for the storage of electrical energy. In comparison to conventional batteries, fuel cells have advantages in terms of better operating life without recharging, low toxicity [3,4].

The reversible polymer electrolyte membrane fuel cell (RPEMFC) or hydrogen fuel cell is an improved variant of the polymer electrolyte membrane fuel cell (PEMFC), combining characteristics of a fuel cell and electrolyser [5]. The electrolysis phase involves the splitting

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of water (H_2O) into hydrogen (H) and oxygen (O) with the passage of electricity through the system. On the other hand, the fuel cell phase works on reverse electrolysis, i.e. H and O react to form H_2O, heat and electricity. Most of the RPEMFC designs operate on the same principle; however, the characteristics of electrolytes may vary as per design considerations [6]. The role of RPEMFCs on environment pollution is directly related to the method of hydrogen production [5][12]. The use of fossil fuels to produce hydrogen accounts for more than 90 % of the world, resulting in higher carbon emissions. On the other hand, water electrolysis accounts for only 2 to 5% of hydrogen production worldwide [5]. Due to its sustainable and zero-carbon potential, electrolysis of water is the best option for producing green hydrogen, leading to a higher importance of reversible polymer electrolyte membrane fuel cell (RPEMFC). Therefore, to make PEMFCs more eco-friendly and economically suitable, reversible polymer electrolyte membrane fuel cells (RPEMFC) are utilised.

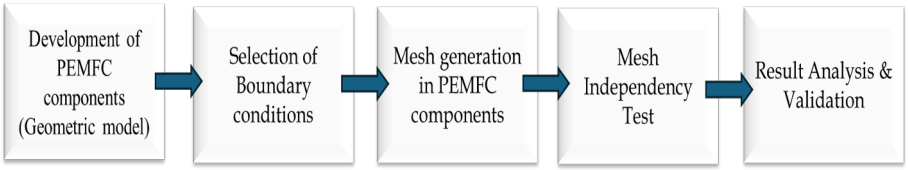
Since a RPEMFC unit involves direct conversion of energy using electrochemical reactions, the efficiency of these systems is reported to be higher than conventional combustion-based systems. However, its efficiency in terms of energy storage is less than other conventional electrochemical energy storage systems [7]. The efficiency of RPEMFC units is reported to be around 50-60% [8]. Due to the limited efficiency of the RPEMFCs between 50 to 60%, the remaining amount of energy is dissipated in the form of waste heat, which results in unfavourable thermal conditions [2]. The reversible PEMFC units are specially designed for temperatures between 60°C to 100°C [2] [9]. The higher waste heat will result in higher operating temperatures, which will affect the overall efficiency of RPEMFC systems. The increase in temperature within reversible PEMFCs results in improved electrochemical activity, resulting in higher efficiency [10,11]. However, higher operating temperature will also produce more dominant negative effects like electrode dehydration, non-uniform temperature distribution, large ionic resistance, Ohmic losses and material degradation [9]. The electrolyte dehydration is generally caused due to higher protonic resistance leading to interrupted supply of water to PEM. Also, this effect is further intensified by electro-osmotic force which pushes H_2O from anode to cathode section. Other than this, non-uniform distribution of temperature at membrane electrode assembly (MEA) also leads to reduced durability and life of membrane unit. Therefore, suitable thermal conditions are quite significant in obtaining higher fuel cell efficiencies. However, in order to design a suitable thermal management system for reversible PEMFCs, it is important to have a better understanding of various heat sources and their effects on overall fuel cell performance.

The presence of heat in electrolysis operation in RPEMFCs has a positive effect on its overall efficiency. The reduced need for electricity is reported for the electrolysis process in comparison to the amount of chemical energy produced in the same process [1]. The lower external electricity requirement during electrolysis results in higher efficiency of operation. From previous studies, it can be observed that parameters like membrane type, catalyst layer, fluid channel configurations, and current density, temperatures have a significant impacton efficiency of RPEMFC during electrolysis operation. This study aims to investigate and simulate the performance of the RPEMFCs in producing hydrogen during electrolysing operation under given conditions. This investigation has been carried out on the RPEMFC unit with Nafion-115 type membrane at the current density of 1 A/cm^2.

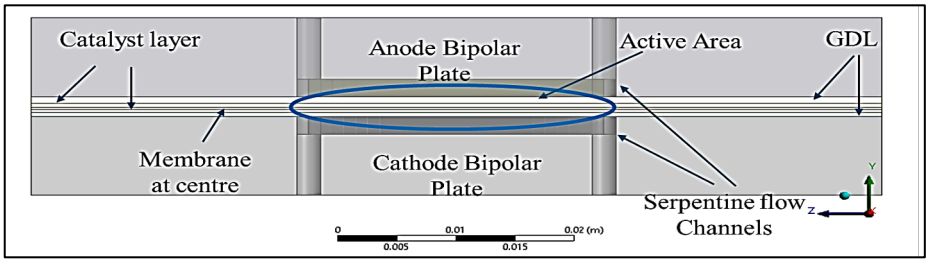
2 Problem description and modelling

The primary objective of the study is to investigate the performance of the RPEMFC unit during the electrolysis process, i.e. charging phase, in the context of the PEM battery unit. The methodology followed throughout this investigation is given in Figure 1(a). This process involves development of RPEMFC unit with given components like Bipolar plates, gas

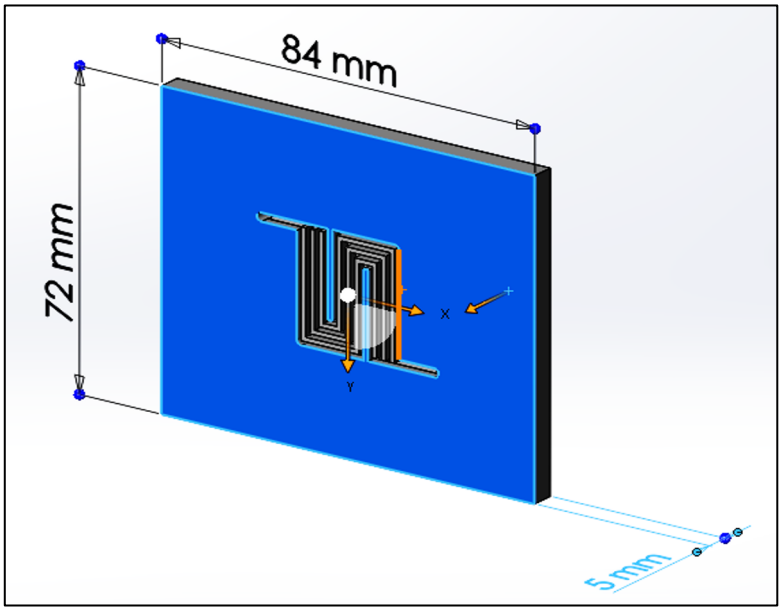
diffusion layers (GDL), catalyst layers (CL) and Membrane. The schematic for the geometric model of the RPEMFC unit is presented in Figure 1 (b). The anode and cathode Bipolar plates (82mm X 72mm X 5mm) with serpentine flow channel (1mm-wide and 2 mm-high) have been used for reactant and product distribution. Geometric models of both serpentine channel and bipolar plate configuration are clearly illustrated in Figures 1(c) and (d). The Nafion-115 (operating temperature = 80°C) with a thickness of 0.127 mm are used as an electrolyte membrane with suitable catalyst loading or layer (CL), i.e. Iridium Ruthenium Oxide +Platinum layer on the anode side and only platinum layer on the cathode side. The overall active area for this study has been calculated as 366.43 mm^2. The thickness of the CL and GDL is considered as 0.02 mm and 0.22 mm, respectively. The optimum averaged density, specific heat and porosity for Iridium Ruthenium Oxide + Platinum Black as catalysts layer were taken as 13.31 $g\text{-}cm^{-3}$, 165 $J{\cdot}kg^{-1}{\cdot}K^{-1}$ and 0.5 respectively on the basis of previous literatures and investigations. All the components of the RPEMFC have been modelled in the modelling software along with their specific material properties. Various input parameters and boundary conditions used in this study have been presented in Table 1. Under suitable input parameters and boundary conditions, as shown in Table 1, the RPEMFC unit is numerically investigated and simulated with the help of ANSYS-Fluent 2023.



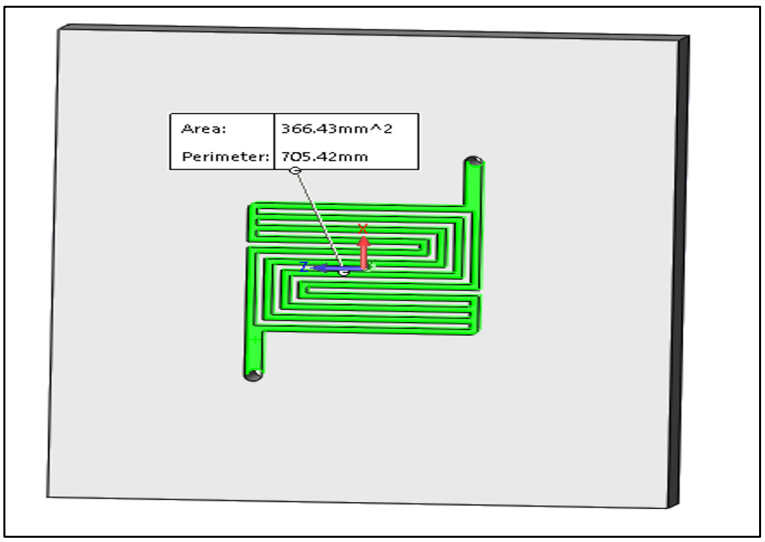
(a) Step wise methodology followed for the current investigation.



(b) Assembly of PEMFC components in geometric model.



(c) Geometric dimensions of bipolar plates.



(d) Active area (serpentine flow channel).

Fig. 1. Geometric parameters and assembly of PEMFC unit.

Table 1. Various Input Parameters and Boundary conditions for numerical analysis of the RPEMFC unit.

Input Parameters	Range	Boundary Zone	Type
Membrane	Nafion-115 (0.127 mm thickness)	Anode water inlet	Mass flow inlet
Fluid channel profile	Serpentine	Anode oxygen outlet	Pressure outlet
Active Area	366.43 mm^2	Cathode hydrogen outlet	Pressure outlet
Water mass flow rate	$5.1307*10^{-7}$kg/s for the entire active area.	Anode current collector	Current density =0 A/m^2
Surface Current Density	10000A/m^2 or 1 A/cm^2	Cathode current collector	Current density =10000 A/m^2

3 Discretisation of PEMFC unit

The discretisation is the process involving the division of the given geometry into various small grids or finite volumes. The RPEMFC model used in our study has been discretised into structured hexahedron elements, as shown in Figure 2, due to its potential in providing better convergence, less memory consumption and computation time. In order to ensure higher accuracy and precision in results, a higher density of mesh elements was maintained at the centre of anode bipolar plates and cathode bipolar plates, i.e. active membrane area. The decision was made due to direct exposure of the active area to the electrochemical reactions under the electrolysis process, resulting in higher complexity levels. The density and type of meshing in the fluid domain can be seen in Figure 2(a). The discretisation quality is analysed in the context of skewness, aspect ratio and orthogonal quality as shown in Table 2. A suitable mesh density is mandatory for analysis to perfectly balance time consumption, load and accuracy of results. Therefore, suitable mesh density was selected by carrying out mesh independency test. The voltage at current density 0.1 A/cm^2 with different number of mesh elements was evaluated for different mesh element numbers by changing element sizes. After careful observation, structured hexahedron mesh with 951897 elements was selected for this study. Furthermore, it was also observed that 10e-8 is suitable for the converging of solutions to desired accuracy levels.

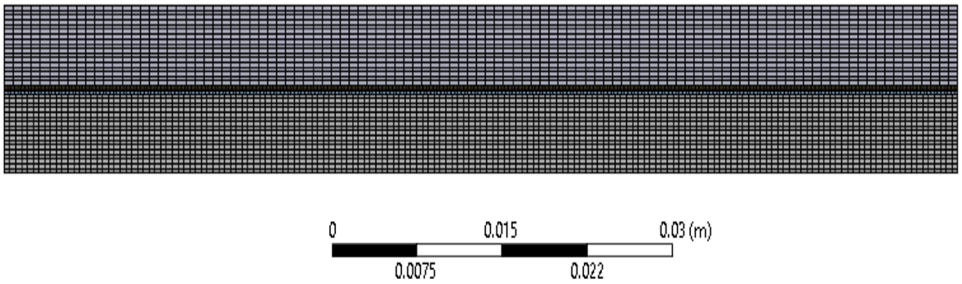


Fig. 2. Discretisation of the RPEMFC unit into hexahedral elements.

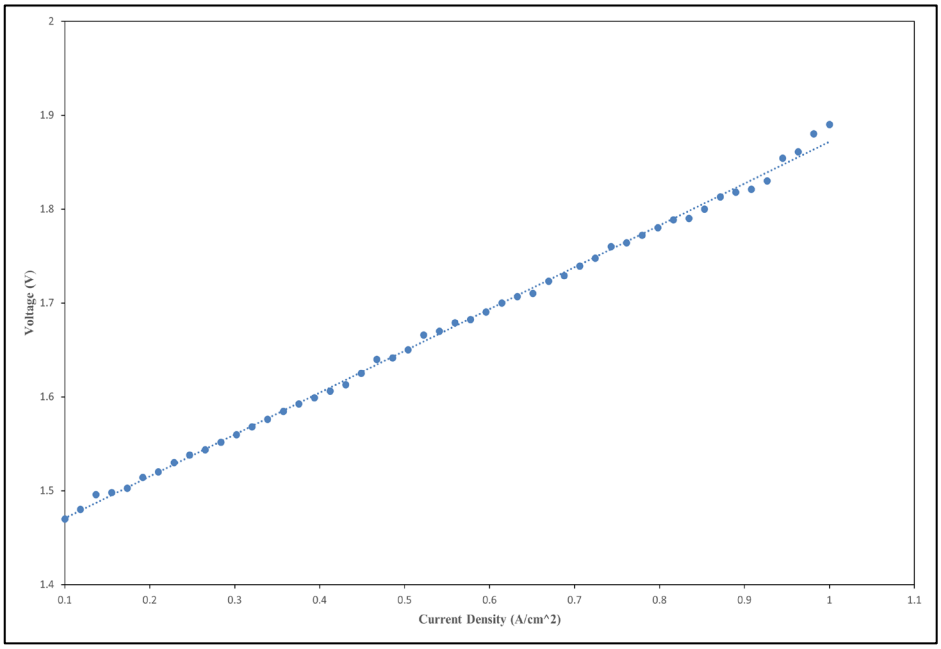
Table 2. Discretisation parameters maintained for the current study.

Mesh Parameters	Range
No. of mesh elements	951897
Skewness factor	0 to 0.50
Aspect ratio	1 to 8
Orthogonal quality	0.5 to 1

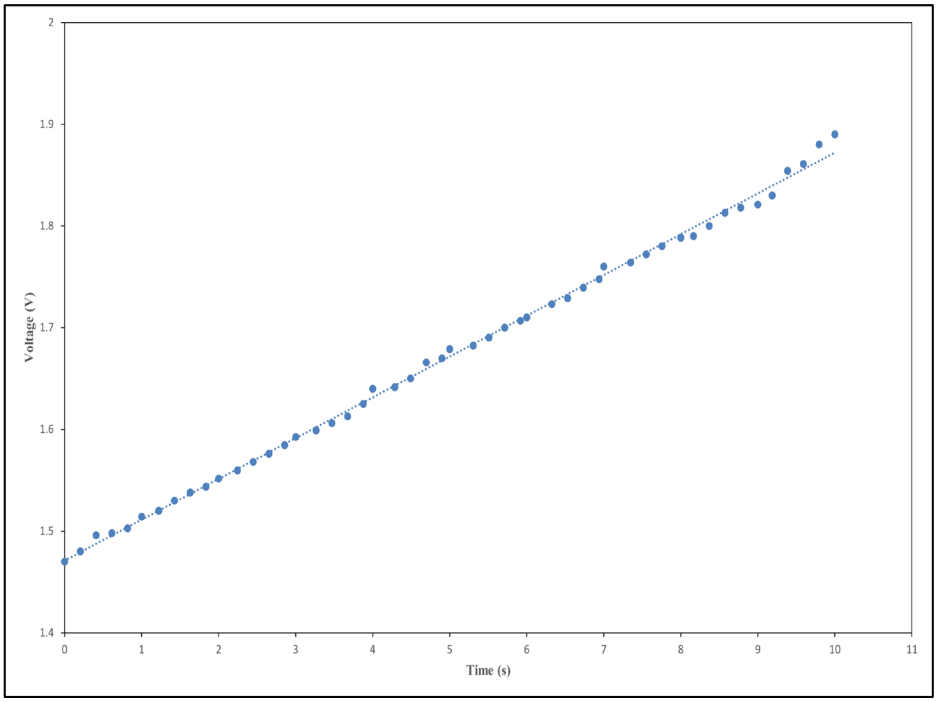
4 Results and discussion

The performance of RPEMFC unit during steady state electrolysing operation was analysed at the current density of 1A/cm^2 (i.e. 3.66A of total current) for a ramp-up phase from 0s to 10s. The ramp-up phase is essential to prevent electrochemical or mechanical shocks due to an instant increase in target load. The ramp-up phase in this study represents the time interval in which the system moves smoothly from 0.1 A/cm^2 to its target load, i.e. 1A/cm^2. The voltage was reported to increase with an increase in time from 0 to 10s, as shown in Figure 3 (b). Additionally, the variation of voltage with an increase in current density from 0.1A/cm^2 to 1A/cm^2 is depicted in Figure 3(a). This increase in voltage at lower current densities is essentially dominated by activation overpotential due to electrode microstructure and catalyst activity. However, at higher current densities, ohmic losses are more dominant as ohmic losses increase linearly with an increase in current density. At 1A/cm^2 current density, the operating voltage was reported at 1.89 V.

The results of this study were validated with Kumar and Himabindu [13], who reported similar trends with variation of voltage between 1.55-2.0 V for different catalyst loadings in the case of Nafion-115 at a current density of 1 A/cm^2. Therefore, the results reported in the simulation of the RPEMFC electrolysis operation are consistent with the experimental findings of Kumar and Himabindu [13]. Additionally, the electrical power (P=Voltage*current) required in RPEMFC was calculated as 6.92 W. The voltage efficiency at 1A/cm^2 current density was reported at 65 %. The close agreement is primarily due to use of validated electrochemical parameters, with suitable gas diffusion layer, catalyst layer and membrane. This consistency validates reliability of the given model and their capability to understand dominant electrochemical behaviors reported in experimental investigations.



(a) Variation of voltage in RPEMFC with current density variation (0.1A/cm^2 to 1A/cm^2).



(d) Variation of voltage in RPEMFC with time from 0 to 10 s.

Fig. 3. Variation of voltage for a reversible polymer electrolyte fuel cell.

The linear relationship is reported between cumulative hydrogen production and operating time, as shown in Figure 4. Furthermore, hydrogen production is also directly proportional to

the current, which leads to a smooth ramp-up in hydrogen output. At the beginning of the electrolysis reaction, it can be observed that the hydrogen production rate is quite low, essentially due to the low concentration of reactive ions in the active region. The cumulative hydrogen production output at current density 1A/cm^2 was reported as 0.00021 mol. More accurate illustration of the hydrogen production at 1A/cm^2 is provided in the Figure 5(a). The Figure 5(a) presents mass fractions of Water, Oxygen and Hydrogen in RPEMFC unit at both anode and cathode sides at the current density of 1A/cm^2. At the anode side as shown in Figure 5(b), it has been observed that electrochemical reactions are more dominant at the central section of the channel, leading to higher mass fraction of oxygen. This is primarily due to flow of reactants through the serpentine channels having bends and corners leading to reduced velocity. This enhances the reaction time at the catalyst layer leading to intense chemical interactions and ultimately resulting in higher oxygen and hydrogen ion values. Even though higher reaction improves overall performance of fuel cell unit, non-uniform behaviour shown for the electrochemical reaction will also lead to formation of hotspots on the membrane. The hotspot will cause higher temperature in the region and reduce the diffusion efficiency of the membrane leading to degradation of membrane.

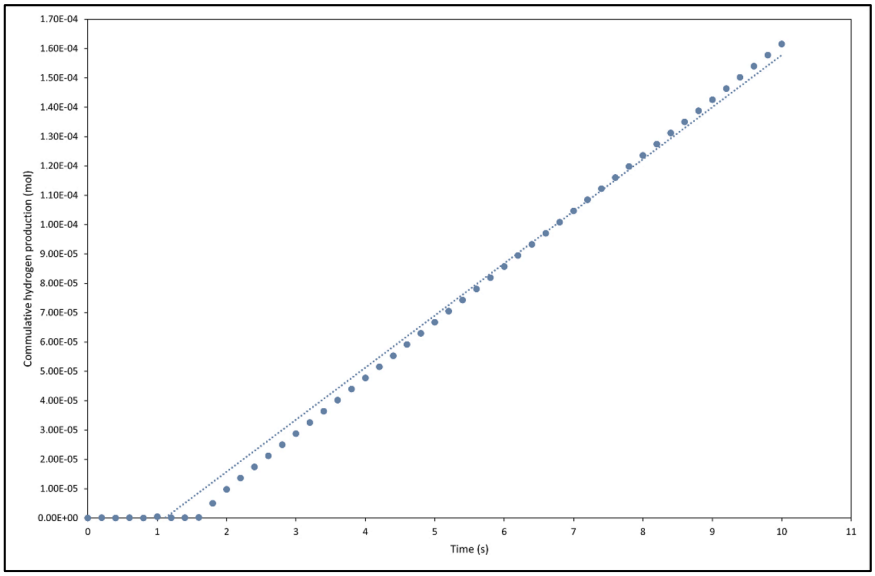
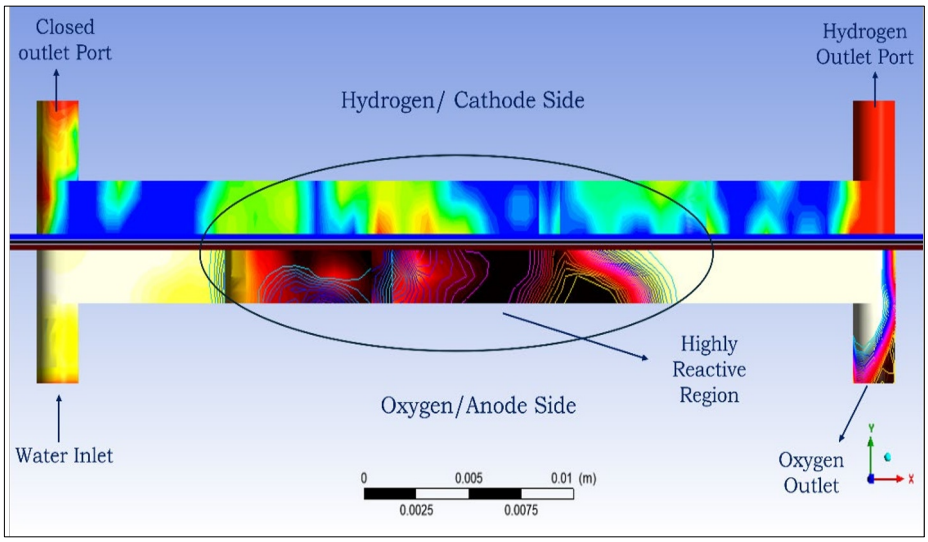
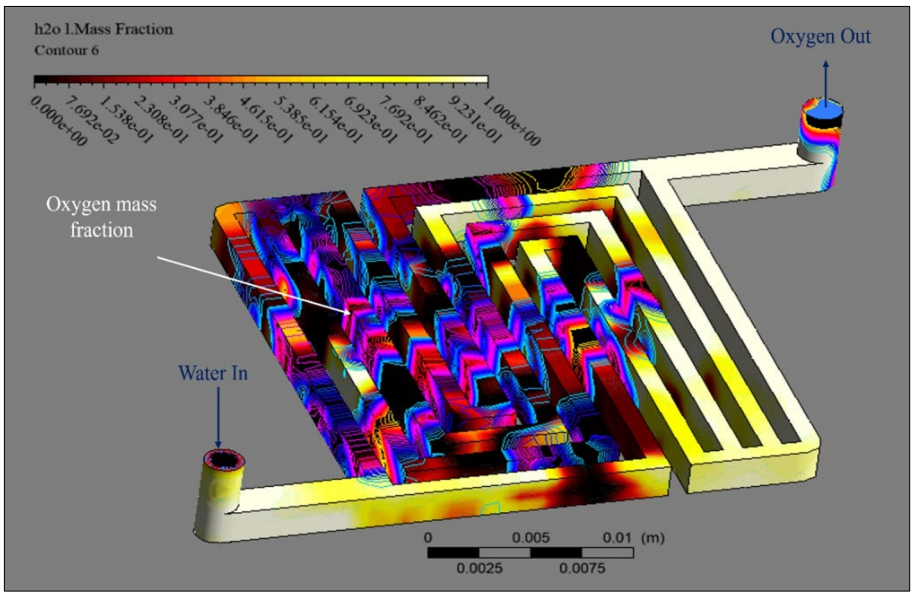


Fig. 4. Variation of cumulative hydrogen production at different time intervals for a RPEMFC.



(a)



(b)

Fig. 5. (a) Contours of water, oxygen, and hydrogen in RPEMFC unit at both anode and cathode sides (At a current density of 1A/cm^2), and (b) A mixture of oxygen and water (liquid) on the anode side of RPEMFC during electrolysis in serpentine channels.

5 Conclusion

The primary objective of this study was to simulate the performance of the RPEMFCs in producing hydrogen during electrolysing operation under given conditions. This investigation was carried out on a geometric model of an RPEMFC unit with Nafion-115 type membrane at a current density of 1 A/cm^2. Iridium Ruthenium Oxide and Platinum

Black on the anode side and platinum on the cathode side were used as catalysts in the electrode assembly.

The performance of reversible polymer electrolyte fuel cell (RPEMFC) during steady state electrolysing operation was analysed at the current density of 1A/cm^2 (i.e. 3.66A of total current) for a ramp-up phase from 0s to 10s. This study acknowledges the significance of the ramp-up phase in the investigation of the steady state electrolysis process at fixed current density. In this study, it is observed that the cell voltage tends to increase with varying current density from 0.1A/cm^2 to 1A/cm^2. At 1A/cm^2 current density, operating voltage was reported at 1.89 V. At lower current densities voltage increase is significantly dominated by activation overpotential. However, at higher current density values, ohmic losses become more dominant than activation overpotential. The voltage efficiency of RPEMFC electrolysing operation was reported as 65% at 1A/cm^2 current density. This study also observes cumulative hydrogen production of 0.00021 mol at a current density of 1A/cm^2.

Furthermore, higher capital cost also presents significant challenges in the commercialisation of RPEMFCs. More sustainable and innovative approaches are required to bring its cost down. In addition, this study indicates that more investigations can be carried out with higher loads, different catalyst loadings, and different membrane materials to further enhance the applicability of the reversible polymer electrolyte fuel cell (RPEMFC). Future investigations may also be carried out in the field of hydrogen storage and thermal management segment of RPEMFC units. Due to challenges processed by storage of hydrogen in liquid or gaseous form, more suitable alternatives such as activated carbon, graphene oxides etc can be considered for storage of hydrogen. On the other hand, use of RPEMFCs in applications with higher power requirement, leads to higher power generation, which leads to higher heat losses in the unit. Therefore, various passive thermal management techniques such as phase change materials, heat spreaders, and metal foam can be considered to ensure the smooth and efficient functioning of RPEMFC unit.

References

1. R. O'HAYRE, S.-W. CHA, W. G. COLELLA, and F. B. PRINZ, *Fuel cell fundamentals*, 3rd ed. Hoboken, New Jersey: John Wiley & Sons, 2016. doi: 10.1007/978-0-387-73532-0_1.
2. A. Faghri and Z. Guo, "Challenges and opportunities of thermal management issues related to fuel cell technology and modelling," *Int. J. Heat Mass Transf.*, vol. 48, no. 19–20, pp. 3891–3920, 2005, doi: 10.1016/j.ijheatmasstransfer.2005.04.014.
3. Q. Chen, G. Zhang, X. Zhang, C. Sun, K. Jiao, and Y. Wang, "Thermal management of polymer electrolyte membrane fuel cells: A review of cooling methods, material properties, and durability," *Appl. Energy*, vol. 286, no. December 2020, p. 116496, 2021, doi: 10.1016/j.apenergy.2021.116496.
4. M. M. Tellez-cruz, J. Escorihuela, and O. Solorza-feria, "Proton Exchange Membrane Fuel Cells (PEMFCs): Advances and Challenges," pp. 1–54, 2021.
5. S. Z. Zhiznin, V. M. Timokhov, and A. L. Gusev, "Economic aspects of nuclear and hydrogen energy in the world and Russia," *Int. J. Hydrogen Energy*, vol. 45, no. 56, pp. 31353–31366, 2020, doi: 10.1016/j.ijhydene.2020.08.260.
6. B. Cook, "Introduction to fuel," no. December, pp. 205–216, 2002, [Online]. Available: https://doi.org/10.1049/esej:20020601
7. Y. Wang, P. P. Mukherjee, J. Mishler, R. Mukundan, and R. L. Borup, "Cold start of polymer electrolyte fuel cells: Three-stage startup characterization," *Electrochim. Acta*, vol. 55, no. 8, pp. 2636–2644, 2010, doi: 10.1016/j.electacta.2009.12.029.

8. O. Z. Sharaf and M. F. Orhan, “An overview of fuel cell technology: Fundamentals and applications,” *Renew. Sustain. Energy Rev.*, vol. 32, pp. 810–853, 2014, doi: 10.1016/j.rser.2014.01.012.
9. T. J. P. Freire and E. R. Gonzalez, “Effect of membrane characteristics and humidification conditions on the impedance response of polymer electrolyte fuel cells,” *J. Electroanal. Chem.*, vol. 503, no. 1–2, pp. 57–68, 2001, doi: 10.1016/S0022-0728(01)00364-3.
10. R. K. Ahluwalia, E. D. Doss, and R. Kumar, “Performance of high-temperature polymer electrolyte fuel cell systems,” *J. Power Sources*, vol. 117, no. 1–2, pp. 45–60, 2003, doi: 10.1016/S0378-7753(03)00028-4.
11. R. Hahn, M. Krumm, and H. Reichl, “Thermal management of portable micro fuel cell stacks,” *Annu. IEEE Semicond. Therm. Meas. Manag. Symp.*, pp. 202–209, 2003, doi: 10.1109/stherm.2003.1194363.
12. J. W. Sheffield and Ç. dem Sheffield, *Assessment of Hydrogen Energy for Sustainable Development*, no. 1. Netherlands: Springer Netherlands, 2007.
13. S. Shiva Kumar and V. Himabindu, “Hydrogen production by PEM water electrolysis – A review,” *Mater. Sci. Energy Technol.*, vol. 2, no. 3, pp. 442–454, 2019, doi: 10.1016/j.mset.2019.03.002.