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A Critical Review of Thermal Management of Proton-Exchange Membrane Fuel Cells (PEMFC)

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Abstract *Fuel cells are advanced electrochemical devices that possess the remarkable ability to convert electrochemical reactions directly into electricity. Given their high efficiency, they offer a promising means of harnessing energy from renewable sources, particularly clean hydrogen, thereby presenting a viable solution for sustainable power generation. Proton exchange membrane fuel cells (PEMFCs) use a polymer membrane as electrolyte. The membrane has selective channels for the transport of hydrogen ions. Externally, the membrane is covered, both in the anode and the cathode sides, by a catalyst layer and a gas diffusion layer, forming the membrane-electrode assembly (MEA). Despite significant research efforts, operational challenges still require further resolution, such as maintaining the ideal temperature and humidity for optimal operation under large loads. Recent work in the literature addresses this issue on several fronts: (1) using differential models to understand the phenomena associated with heat generation at specific locations in the fuel cell; (2) using integral models to focus on the relationships between the various subsystems that make up the energetic matrix (fuel generation, storage, fuel cell, etc.); and (3) through experimental characterization of fuel cells and waste heat recovery systems for increased energy efficiency. Regarding thermal management, new strategies are emerging, ranging from techniques to enhance heat transfer (e.g., spray cooling and microchannels) to thermoelectric devices that can scale with fuel cell size and power. This paper reviews and discusses the state of the art in fuel cell thermal management, including numerical models and various cooling techniques suitable for different fuel cell applications.*

Keywords: *hydrogen fuel cell, PEMFC, thermal management, renewable fuel, cooling solutions*

1. INTRODUCTION

Amid the growing demand for low-environmental impact fuels, hydrogen (H_2) proves to be a reliable alternative with an increasing number of applications in a wide variety of industries (namely transport, steel, oil refining, and power generation) (IEA, 2022). Due to the push for decarbonization, the demand for hydrogen is expected to increase substantially in the near future (IEA, 2019), bringing about novel challenges and opportunities for scientific and technological progress.

The use of hydrogen for energy generation in fuel cells – devices that harness the electrochemical energy from the oxidation-reduction of H_2 with only electricity, water, and heat as products – is a suitable option to replace high-emission fuels in both mobile and stationary applications (Abe *et al.*, 2019; Pramuanjaroenkij and Kakaç, 2023). The widespread adoption of fuel cells, and hydrogen fuel as a whole, will require a complete overhaul of the current installed infrastructure, therefore technological advancements are tied to developments in hydrogen production, storage, and transportation (Al Ghafri *et al.*, 2022).

Proton exchange membrane fuel cells (PEMFCs) are a particular subset of fuel cells that employ a selectively-

permeable polymer membrane to control the transport of protons between the two electrodes, namely the cathode and the anode. Figure 1 schematically illustrates a PEMFC highlighting its different domains. Although Nafion is the most common polymeric material used to create these membranes, the development and search for new materials is still a research challenge being investigated by several groups (Zhang and Shen, 2012; Prykhodko *et al.*, 2021; Karimi *et al.*, 2019; Zhu *et al.*, 2022).

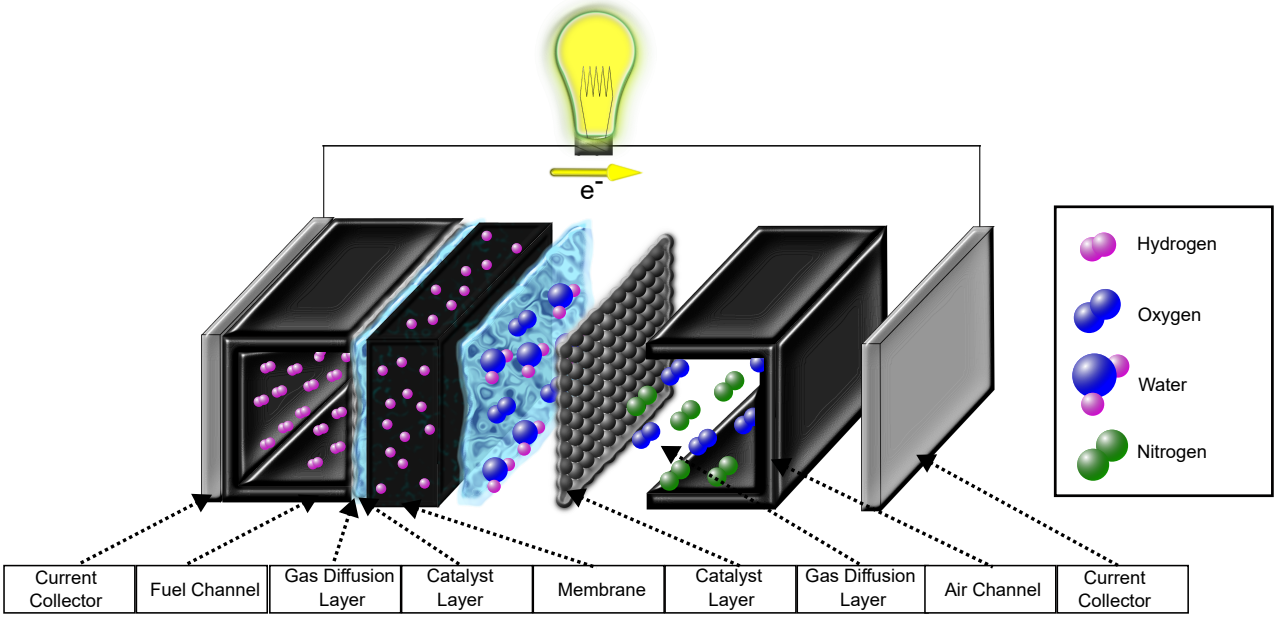

Figure 1. Schematic representation of PEMFC with exploded view on the cathode side.

The working principle of a PEMFC can be summarized as follows: fuel (H_2) flows in a channel within the anode while the oxidizing agent (pure O_2 or air) flows in a channel inside the cathode. On the anode side, the polymeric membrane is covered by a thin layer of catalyst responsible for splitting the fuel into protons and electrons, given by the following half-cell oxidation reaction (Bernardi and Verbrugge, 1991; Rowe and Li, 2001):

$$\mathrm{H_2} \rightarrow 2\mathrm{H^+} + 2\mathrm{e^-}. \tag{1}$$

The protons are transported through the membrane into the cathode side, while the electrons flow through the metallic structure of the fuel cell, producing the voltage output. On the cathode side, the newly formed protons that traversed the membrane react with the oxygen from the stream in a half-cell reduction reaction over the thin catalyst layer releasing thermal energy and forming liquid water (Bernardi and Verbrugge, 1991; Rowe and Li, 2001)

$$\frac{1}{2}\mathrm{O_2} + 2\mathrm{H^+} + 2\mathrm{e^-} \rightarrow \mathrm{H_2O}. \tag{2}$$

The transport of protons and water across the catalyst layer and membrane has been the subject of several modeling efforts in recent years (Sui *et al.*, 2019).

In order to ensure the optimal operating condition of the fuel cell, the relative humidity from the gaseous streams and the water content in the membrane must be properly dealt with. Thermal management is another key aspect since elevated temperatures may hinder the fuel cell performance. Several authors have resorted to different mechanistic and empirical modeling strategies to describe transport and polarization phenomena in the cell, as discussed below.

2. MODELING

PEMFC modeling is a complex task as it combines conservation of species, potential, and energy, as well as phase change and energy conversion terms (Barbir, 2005). Several authors have reviewed PEMFC modeling. Wu (2016) offers a review of 2- and 3-D modeling strategies, including the fluid flow in the plate flow channels. These higher-dimension models allow for accurate predictions of both temperature and water content distributions, providing invaluable insights for the thermal management of the fuel cell.

Andersson *et al.* (2016) reviewed the flow channel and gas diffusion layer modeling in polymeric electrolyte fuel cells considering the multiphase flow and species transport at different characteristic scales. They highlighted the importance of hybrid, multiscale models that can provide a broader solution to the fuel cell.

Similarly, Chen *et al.* (2022) also indicate the need for hybrid, multiscale models for a more robust fuel cell representation. The authors proposed an extensive review on the modeling of polymeric membrane through a mechanistic multiphysics approach. They suggested that machine learning techniques could be used to generate novel and optimal polymeric membrane structures.

In general, modeling of PEMFCs can be divided into two separate approaches: Gas Transport Models (GTM) or differential models and Polarization Curve Models (PCM) or integral models (George *et al.*, 2022). These and other relevant modeling aspects are discussed in the following sections.

2.1 Integral Models

Integral models or polarization curve models focus on the steady-state voltage output of the cell considering the available reversible potential, given by the Nernst-Planck equation, and the potential losses. It is represented by the following equation:

$$V_{\text{cell}} = E_{\text{Nernst}} - E_{\text{act}} - E_{\text{ohm}} - E_{\text{con}}, \tag{3}$$

where V_{cell} is the resulting cell output voltage and the subscripts on the RHS represent the reversible potential and losses due to (i) activation over potential, (ii) charge transport and (iii) species transport, respectively (EG&G Technical Services, 2004). The maximum cell output voltage is given by the Nernst-Planck equation Barbir (2005); Newman and Balsara (2020)

$$E_{\text{Nernst}} = -\frac{\Delta G^o}{nF} + \frac{\Delta S}{nF}\left(T - T_{\text{ref}}\right) + \frac{RT}{nF}\ln\left(\frac{p_{H_2} p_{O_2}^{0.5}}{p_{H_2O}} p_{\text{ref}}^{-0.5}\right), \tag{4}$$

where ΔG^o is the Gibbs free energy of formation, n is the stoichiometric coefficient, F is the Faraday's constant, ΔS is the entropy change, $T_{\text{ref}} = 298.15$ K is the reference temperature, R is the universal gas constant and p_i are the partial pressures of species i, and $p_{\text{ref}} = 100$ kPa.

Activation losses correspond to the over potential required to overcome the activation energy of catalytic reaction. Ohmic or resistive losses are attributed to the resistance to the flow of ions in the electrolyte and the flow of electrons in the current-collecting plates from the catalyst sites to the external circuit. Similarly, concentration losses are attributed to the deficit in reactant mass flow at the catalyst surface, leading to a smaller current density, resulting in voltage losses (Barbir, 2005; Ramalingam and Mathur, 2011). The result from Eq. (3) is the polarization curve, the output voltage response for a given current density output. These losses and the polarization curve of a theoretical fuel cell are illustrated in Fig. 2, adapted from Jouin *et al.* (2013).

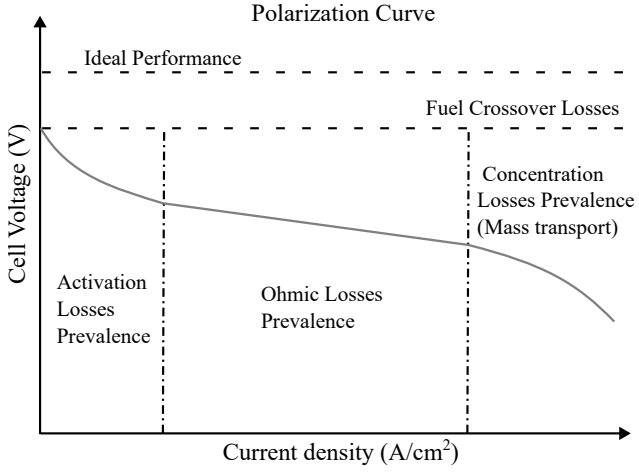

Figure 2. Fuel cell polarization curve and the different voltage losses. [Adapted from Jouin *et al.* (2013)]

Integral models are a convenient way to estimate the polarization curve of a fuel cell from a computational point of view. The major drawbacks are the large number of parametric coefficients and the reliance on macroscopic values, a

byproduct of the model's inability to resolve the profile of physical quantities along the fuel cell. Despite the aforementioned limitations, integral models can still be used for dynamic analyses and provide useful results for fuel cell analysis (George *et al.*, 2022).

Ramalingam and Mathur (2011) used an integral model to investigate both the steady-state and transient performance responses of a PEMFC subject to various changing input parameters (current density, anode gas pressure, cell temperature, etc.). The authors argue that operating the fuel cell at large values of input parameters (high temperature or high gas pressure, for instance) can lead to a reduction in voltage losses. Their model was also able to reasonably predict the transient response of the cell, which can be used to design real-time control systems.

Integral models are also useful when combining different subsystems into a single dynamic model. Saeed and Warkozek (2015) investigated a fuel cell system comprised by a fuel cell stack, electrolyser, photovoltaic modules and hydrogen storage tank set combined into a single global model to simulate a stand-alone energy-generating system capable of continuous operation during sunny days. Each subsystem is then characterized using state variables to obtain the resulting polarization curve. The authors discussed the economical viability of the system, concluding that its competitiveness relies on a reduction of the price of hydrogen.

Omran *et al.* (2021) implemented and validated their polarization curve model with experiments from an installed fuel cell stack. The authors considered a varying external load and obtained results with less than 5% deviation for the polarization curve and less than 6% for the output power when compared with the experiments for a set operating condition. No further investigation regarding operation parameters was conducted and although the encountered stack efficiency was 47%, no discussion regarding the thermal management was presented.

In Ding *et al.* (2023), the authors considered the effect of different flow regime in the gas flow channels during operation of the fuel cell in the polarization curve using non-dimensional numbers (Dankhöler and Reynolds numbers) to determine the characteristics of the fluid flow (Sherwood number).

2.2 Differential Models

Differential models or gas transport models describe the species concentration, electric potential, and energy transport across the different fuel cell domains illustrated in Fig. 1. They provide the local distribution of several variables of interest (e.g. temperature, mole fractions, electric potential) by solving a series of coupled differential equations (conservation equations), making its implementation process more computationally expensive and considerably more complex. Among the common assumptions for fuel cell modeling, the following hypotheses can be highlighted: ideal gas properties and ideal gas mixtures; laminar incompressible flow; isotropic and homogeneous membrane and electrode; negligible potential drop in the current-collecting plates (Barbir, 2005).

In their pioneering work, Bernardi and Verbrugge (1991) modelled the cathode side of an isothermal fuel cell including the diffusive transport of oxygen across the porous electrode, membrane, and a catalyst layer (1-D). The authors made a distinction between gaseous, electronic, and water transport. The latter considered a balance between pressure and electro-osmotic forces (the Schlögel equation). The model considers three phenomenological closure relationships that have been adopted by several authors. The first one is the Nernst-Planck (Taylor and Krishna, 1993; Newman and Balsara, 2020) equation for the transport of ions, given by:

$$N_i = -z_i F D_i \frac{c_i}{RT} \nabla V_m - D_i \nabla c_i + c_i v, \tag{5}$$

where N_i is the molar flux, z_i the valence of the ionic species, D_i the diffusive coefficient of the species in the membrane and c_i the species molar concentration. The second relationship is the Butler-Volmer (Bockris *et al.*, 2002) equation for the current density-voltage relation at the electrodes:

$$j(x) = a i_0 \left\{ \exp\left[\frac{-\alpha_a F \left(V_s - V_m\right)}{RT} \right] - \exp\left[\frac{-\alpha_c F \left(V_s - V_m\right)}{RT} \right] \right\}, \tag{6}$$

where $j(x)$ is the current density distribution, a the catalyst reactive area per unit volume, i_0 the current density per unit of catalyst reactive area, α_i a transfer coefficients in the anode and cathode sides, and $V_s - V_m$ the voltage drop across the membrane. Finally, the Stefan-Maxwell (Taylor and Krishna, 1993) equation for species transport in the gas diffusion layers is given by:

$$\nabla y_i = RT \sum_i \frac{y_i N_j - y_j N_i}{p D_{ij}^{\text{eff}}}, \tag{7}$$

where y is the molar fraction, and D_{ij}^{eff} is the effective diffusive coefficient.

Springer *et al.* (1991) modeled the water transport across the fuel cell (isothermal, 1-D), correlating properties to the water content in the polymeric membrane. Their results were validated against experimental measurements. As a product of the oxidation-reduction reaction, water content must be managed to ensure the proper operation of the polymeric

membrane and the fuel cell as a whole. The authors concluded that thinner membranes are preferable due to the large negative contribution of the membrane resistance. The authors point to a key limitation in the model regarding thermal management: the experiments show that the water produced by the reaction is incapable of compensating for the liquid water evaporation by the generated heat, which leads to membrane dryout.

Rowe and Li (2001) presented a complete 1-D model for a PEMFC, including thermal response and water content tracking with a source term for the membrane water evaporation. Their model attempts to accurately represent species and energy transport across all cell domains (gas-diffusion layer and catalyst in both anode and cathode, and membrane). This type of modeling lends itself to design and assessment of fuel cell operation. Figure 3 illustrates both the polarization curve and the temperature profile across the different domains of the PEMFC. The model results are compared against experimental data yielding satisfactory results. Among their findings is highlighted the need to ensure membrane humidity: the importance of accounting for the phase change of water and the fuel cell performance greater dependence on the anode stream humidity (when compared to the cathode stream) at higher current densities, indicating this to be one of the most important parameters. As observed by Rowe and Li (2001) (and illustrated in Fig. 3 for an operating temperature of 353 K), the temperature increase in a PEMFC could reach 5 K, which could lead to a significant increase for a fuel cell stack. For this reason, the thermal management of the fuel cell (and stack) should be addressed.

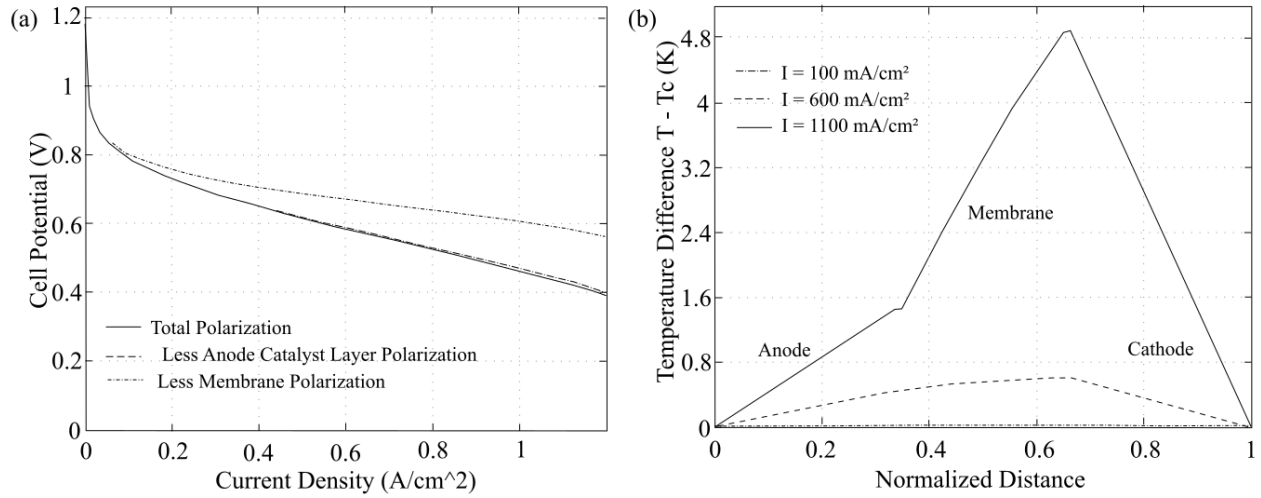

Figure 3. Results from the differential model described in Rowe and Li (2001). (a) Polarization curve. (b) Variation of fuel cell temperature with normalized position across the different domains.

Despite the totality of models discussed in this section being limited to the continuum (macroscopic) scale, the transport phenomena in the catalyst and membrane occur at a smaller (microscopic) scale. Modelling approaches at this scale consider the liquid flow movement and include Lattice-Boltzmann (Molaeimanesh *et al.*, 2016; Yang *et al.*, 2022), Volume of Fluid (Wu *et al.*, 2020; Shi *et al.*, 2022), and Pore-Network methods (Zapardiel and García-Salaberri, 2022; Li *et al.*, 2021). They are very useful and provide several insights into the mechanistic nature of transport at the core of the PEMFC, but their use is not advised due to the very large computational cost (Affonso Nóbrega, 2023).

2.3 Thermal Management

Since the oxidation-reduction fuel cell reaction is exothermal, the resulting heat generation must be accounted for. This heat generation causes the cell temperature to increase, steering the operating condition away from the optimal catalyst performance point. Conversely, lower temperatures may hinder the reaction kinetics and lead to flooding due to lower water saturation pressure (Zhang and Shen, 2012). Hence, thermal management is a key aspect to ensure the proper operation of a single PEMFC or a stack (Nguyen and White, 1993).

The overall thermal management steady-state equation is:

$$\dot{Q}_{\mathrm{FC}} = UA\Delta T, \tag{8}$$

where $\dot{Q}_{\mathrm{FC}}$ is the heat generated by the oxidation-reduction reaction, U is the overall thermal conductance, A the heat-exchange area, and $\Delta T = T_{\mathrm{FC}} - T_{\infty}$ the temperature difference between the PEMFC and the heat sink.

Examining Eq. (8), it can be noted that the cell operating temperature is usually set to some upper limit (often 358.15 K due to catalyst operation) while the heat sink temperature is also fixed (e.g., reservoir temperature, free-stream temperature, phase-change temperature), so ΔT is an input parameter. The remaining terms can be individually addressed.

First, the generated heat, on the LHS of Eq. (8), can be reduced by ensuring the PEFMC operates close to its maximum efficiency, since

$$\dot{Q}_{\mathrm{FC}} = (1-\eta_{\mathrm{FC}})\dot{m}_{\mathrm{H_2}}\Delta h_{\mathrm{H_2}} = \left(1 - \frac{\dot{W}_{\mathrm{ele}}}{\dot{m}_{\mathrm{H_2}}\Delta h_{\mathrm{H_2}}}\right)\dot{m}_{\mathrm{H_2}}\Delta h_{\mathrm{H_2}} = \dot{m}_{\mathrm{H_2}}\Delta h_{\mathrm{H_2}} - \dot{W}_{\mathrm{ele}}, \tag{9}$$

where η_{FC} is the fuel cell efficiency, $\dot{m}_{\mathrm{H_2}}$ the mass flow rate of hydrogen, $\Delta h_{\mathrm{H_2}}$ the enthalpy of combustion, and $\dot{W}_{\mathrm{ele}}$ is the generated electrical power.

Next, UA is a function of the adopted cooling method. Cold plate and liquid cooling are most commonly selected for these types of application (Chen *et al.*, 2021). The major drawbacks are the need for an antifreezing agent for below 0°C applications, the need for liquid storage and the pumping power requirements.

A major challenge in the cooling of automotive PEMFC is the small temperature difference between the fuel cell and the ambient, when compared to internal combustion engines, especially for locations with high ambient temperatures.

Due to its compact nature, PEMFC cooling may be well suited to the use of microchannels, which substantially increase the heat transfer area per volume, improving the heat transfer coefficient (Kandlikar *et al.*, 2006). Microchannels also allow for both single-phase and phase-change flows (Poachaiyapoom *et al.*, 2019; Hoang *et al.*, 2021b). Although the use of microchannels in PEMFCs to handle the mass transport is topic of crescent interest, the available literature still lacks more in-depth study on the use of microchannel heat exchangers for thermal management. Recent studies have attempted to combine direct cooling (sprays) with microchannels in novel thermal management architectures (Hoang *et al.*, 2021a).

While the majority of the discussion in this section is limited to steady-state modeling and operation of PEMFC, the transient phenomena including start-up conditions comprise a very active research topic (Huang *et al.*, 2016; Pourrahmani *et al.*, 2022).

Thermal management and water content management of PEMFCs often require higher-dimension modeling in order to accurately predict the temperature distribution in order to identify hotspots, as well as the water content distribution to predict dryout/flooding of the membrane. For this reason, authors have resorted to a commercial softwares with fuel cell add-on modules such as ANSYS Fluent and COMSOL multiphysics to handle the bulk of the calculations (Liu *et al.*, 2023; Farsi and Rosen, 2023).

Figure 4 shows the expected behavior for the PEMFC power outputs – electric work and heat flux dissipation –, as well as the efficiency for various current densities in an isothermal fuel cell. The results presented in Fig. 4 were simulated with COMSOL multiphysics fuel cell module, showing good agreement with results from the literature for a single fuel cell (Zhang and Shen, 2012). It is important to notice the thermal load strong dependence on the fuel cell stack. For this reason, the chosen thermal management strategy can vary with a wide range of factors.

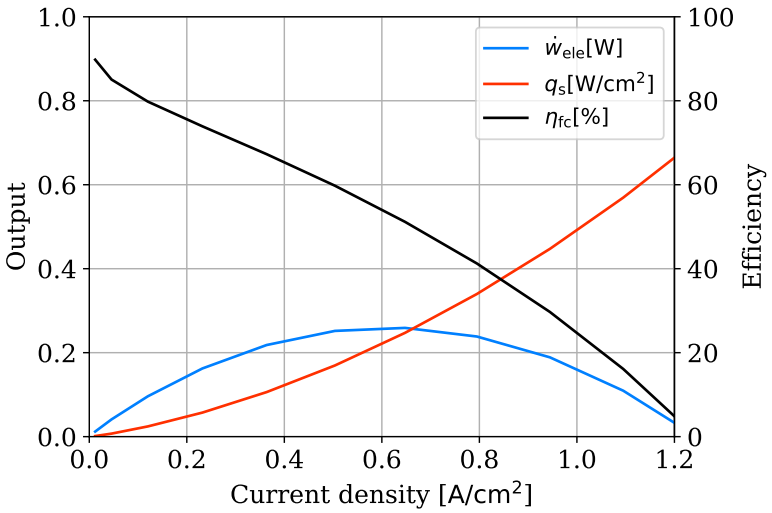

Figure 4. Isothermal fuel cell behavior for different current densities.

Figure 5 presents the range of heat removal capabilities from commonly adopted thermal management strategies. Additionally, important to note the geometric aspect effect when selecting a thermal management system since this kind of compact system usually comes with significant space constraints.

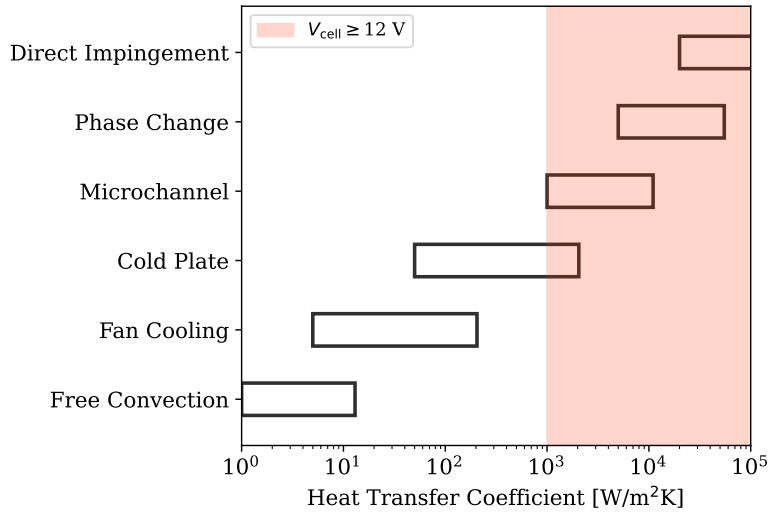

Figure 5. Range of heat transfer coefficient for different thermal management strategies ($T_{\text{FC,max}} = 85$ °C). Fuel cell data from present work and Zhang and Kandlikar (2012). Thermal management datasets taken from the literature (Rohsenow and Choi, 1961; Kandlikar *et al.*, 2006; Incropera *et al.*, 2007; Kaviany, 2011).

3. CONCLUSIONS

Proton-exchange membrane fuel cells are complex power-generating electrochemical devices which have observed a significantly increase in usage within the context of the hydrogen economy. For this reason, robust modeling techniques are required in order to confidently predict the PEMFC's performance under different operating conditions.

Polarization curve models consider macroscopic parameters to obtain the PEMFC electrical response. They can be easily integrated into dynamic analysis to predict the transient behavior of the fuel cell. Differential models, on the other hand, account for the transport of gaseous species and water production through conservation equations at the continuum scale.

Due to its complex nature, PEMFC performance is very sensitive to temperature and hydration. Both the reaction kinetics at the catalyst and the species transport at the membrane are bound to a narrow range of temperature and humidity. For this reason, the thermal management must be addressed to ensure optimal operating conditions. Although liquid cooling is the most widespread adopted cooling solution, novel techniques are under experimental and analytical investigation showing promising results.

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