A model based investigation of evaporative cooling for polymer electrolyte fuel cells – Stack level analysis

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HIGHLIGHTS

- Potentials and limits of evaporative cooling for PEFC are analyzed.
- A zero-dimensional fuel cell stack model is developed.
- Optimal operating conditions for evaporative cooling are quantified.
- An operating window for evaporative cooling is proposed.
- The high mass, volume and cost saving potential of evaporative cooling is proven.

ABSTRACT

Conventional cooling systems of polymer electrolyte fuel cells are responsible for a significant share of stack and system volume, mass and cost. Evaporative cooling shows the potential to overcome these hurdles by simplifying the design of bipolar plates and eliminating the need for an external humidifier. Thus, evaporative cooling can significantly contribute towards reaching the DOE fuel cell system power density target of 850 W/L.

This paper investigates the potentials and limits of evaporative cooling at stack level. For this, a zero-dimensional model has been developed, incorporating mass and energy balances as well as electrochemistry and evaporation.

Main findings show that evaporative cooling is feasible over a wide range of operating conditions. The cooling performance is a function of temperature, gas pressures and stoichiometric ratios, where the temperature shows the largest leverage.

A feasible operating window is proposed, which is slightly shifted towards higher temperatures (80–95 °C), lower pressures (100–200 kPa) and higher cathode stoichiometric ratios (>1.5) compared to conventional fuel cells. A slight decrease in electrochemical performance (ca. 3% at 1.5 A/cm\(^2\)) is easily compensated by the volume and weight saving potential of up to 30% and thus substantially reduced cost.

1. Introduction

Fuel cell electric vehicles (FCEV) \([1–4]\) are a promising alternative to conventional internal combustion engine vehicles (ICEV) and battery electric vehicles (BEV), particularly for heavy-duty transport applications, since they allow an efficient and emission-free conversion of hydrogen, provide high mileage \([5]\) and short refueling times \([6]\). Polymer electrolyte fuel cells (PEFC) are a favorable propulsion system for automotive applications, since they reach peak system efficiencies above 60% \([3]\), system power densities of more than 640 W/L \([7]\), respectively 659 W/kg \([8]\) and allow start-up times below 10 s at 20 °C and 20 s at –20 °C (to 50% of rated power) \([2]\) as well as freeze-start capabilities at temperatures below –30 °C \([9]\). However, several key issues regarding durability, power density, efficiency, dynamic response, heat rejection and cost have to be solved to achieve an increasing market penetration \([10]\).

State-of-the-art volumetric and gravimetric power densities of fuel cell systems are almost in accordance with U.S. Department of Energy...
2020 targets (650 W/L, 650 W/kg), however, to reach the ultimate goal of 850 W/L, a decrease in system volume of about 25% is required (at same performance). Further, fuel cell system costs need to be reduced from the 2017 benchmark of 45 USD/kW (projected for 500,000 units/year) [11] in order to achieve the ultimate target of 30 USD/kW [12].

In a conventional PEFC, the cooling system contributes significantly to stack and system volume as well as cost. The state-of-the-art cooling solution for transport applications aims at transferring the waste heat to a liquid coolant that flows through separate cooling channels in the bipolar plates. The shape of these cooling channels determines the cooling performance as well as the pressure drop significantly [13]. Therefore, a complex multi-layer design of bipolar plates is required [14] which contributes to about 75% of stack volume and approximately 80% of its mass [15]. The coolant is subsequently chilled in an external heat exchanger which requires a high surface area due to the small temperature difference between coolant and the ambient air and thus shows a high volume demand. In order to facilitate the heat transfer, stack operating temperatures in the range of 70–90 °C are necessary. However, to ensure a proper humidification and thus ionic conductivity of the proton exchange membrane [16] at elevated temperatures, an external humidifier is often required which adds to the system volume.

Several alternative cooling approaches such as heat spreaders as well as active or passive air cooling have been presented on single cells or short stacks [14]. However, these solutions have in common that the cooling power density is low compared to liquid cooling which limits their suitability for automotive applications.

Evaporative cooling is a promising concept to reduce the size, weight and cost of the cooling system. In general, a liquid (typically water) is injected into the fuel cell which evaporates and thereby cools the cell. Published and patented concepts include water injection into the gas supply [17–20], porous bipolar plates or flow fields [21–25], as well as into additional layers or devices [26–29]. All concepts have in common that the stack and/or system complexity is increased.

In the concept of Cochet et al. [30], in contrast, liquid water is fed to the cells through dedicated water channels that are directly located in the anode flow field, which requires no additional layer. Subsequently, the water is wicked into a specially designed gas diffusion layer (GDL) with a mixed hydrophilic and hydrophobic pattern [31] and distributed through the entire cross section. Consequently, the water in the hydrophilic lines evaporates, cools the cell by taking up the heat of evaporation and is released as water vapor. Moreover, the water evaporates close to the membrane and therefore contributes to a better humidification and thus higher ionic conductivity, which enables higher operating temperatures without the need for external humidification. Thereby, evaporative cooling shows the potential to reduce the system volume, mass, complexity and cost by up to 30% by simplifying the design of bipolar plates and balance of plant [8], which contributes towards reaching the U.S. Department of Energy ultimate power density target.

However, the interactions between operating conditions, evaporation rates, cooling power as well as stack performance are complex and manifold. Therefore, numerical models can help to foster the understanding of the relevant underlying processes.

A substantial contribution to evaporative cooling modelling on stack and system level has been made by Fly and Thring. In 2013 [32] they presented a fuel cell vehicle model with electrochemical and radiator subsystems in order to determine the thermal and water balance of an evaporatively cooled PEFC system. The impact of the operating pressure on the waste heat and thus on the radiator frontal area was studied. They showed that an increase in operating pressure reduces the radiator area by 30% due to the elevated operating temperature which is required to maintain the thermal balance. In 2015, Fly and Thring [33] investigated the transient temperature regulation in an evaporatively cooled PEFC stack at different current profiles and drive cycles using a lumped parameter fuel cell model. They showed that the operating temperatures varies below ±2 °C at typical operating conditions without active temperature control. In addition, they present a PI control strategy that can further reduce the temperature variation (±1 to ±1.2 °C) by adapting the pressure and cathode stoichiometry which can also contribute to reduce heat loss at low operating loads. In 2016, Fly and Thring [34] conducted a comparison of evaporative and liquid cooling methods for fuel cell vehicles. In addition to their evaporative cooling model (described above) they presented a detailed 1-D model for heat exchangers and condensing radiators. Their main finding shows that the frontal area of a vehicle radiator with evaporative cooling can be reduced by up to 27% as compared to conventional cooling techniques due to the phase change in the condensing radiator.

Furthermore, Schulzle and Horn [35] developed a control-oriented simulation model for evaporatively cooled PEFC systems consisting of the fuel cell stack, a heat exchanger, a water separation cyclone, a cooling water tank and three water pumps. The model captures the system behavior well and can be used for future controller design.

Weber and Darling [36] simulated the effect of porous flow field plates in comparison to conventional flow fields and showed an increased performance at low RH conditions due to a more uniform membrane hydration throughout the cell.

Several experimental studies have been conducted at cell, stack and system levels. Choi et al. [17] as well as Hwang and Kim [18] studied the effect of liquid water injection into the air stream supplied to the cathode. They analyzed a cathode humidification and evaporative cooling system using an external-mixing air-assist atomizer and showed that the direct water injection method was effective for cathode humidification and stack cooling. Cochet and co-workers [30] have investigated an evaporative cooling concept, based on above mentioned GDL with patterned wettability, in differential single cells. They have visualized water transport with neutron radiography and measured heat transport with integrated heat flux sensors. In situ experiments were conducted and a simplistic 1-D model was developed to determine evaporation rates into the gas channel. They have shown that the contact surface between the water and the gas flow is an important parameter and presented that the evaporation can be controlled by the mass flow rates, temperatures, pressures of gases, and the geometry of the hydrophilic lines. Additionally, the impact of cathode and anode gas flow rates on cooling power and the limitations due to saturation of the gases at low flow rates and vapor transport at higher flow rates were shown.

Further, they presented an operando study [37], supported by neutron imaging that showed the beneficial effects of evaporative cooling on cell humidification and measured only a slight reduction in peak power density compared to conventional operation.

Additionally, the evaporation of water in fuel cell GDLs has been studied extensively, both, numerically [38–41] and experimentally [40, 42–44] by several groups.

Despite the extensive numerical and experimental studies performed in this field, literature does not provide sufficient information on the interactions between operating conditions and evaporation rate and thus available cooling power yet. Furthermore, the required operating conditions to remove the entire waste heat by evaporative cooling as well as the impact of evaporative cooling on the fuel cell performance have not been elaborated in detail so far.

This work aims at closing the above mentioned research gaps. Therefore, a zero-dimensional simulation model is introduced in order to conduct fast parameter studies and judge the suitability of the evaporative cooling concept for transport applications. The developed model is generic in nature and therefore applicable to all evaporative cooling concepts. Potentials and limits of the evaporative cooling approach are elucidated and overall trends are identified. Appropriate operating conditions, required evaporation rates, as well as the impact of evaporative cooling on the fuel cell performance and system complexity are discussed. Finally, a feasible operating window for evaporative cooling is proposed.
2. Numerical model

Experimental investigations of fuel cell stacks and systems require a highly integrated testing environment, including a complex hydrogen and air supply, gas conditioning infrastructure as well as cooling supply, electric load and a sophisticated control system [45,46]. Moreover, the actual test campaigns are cumbersome, time consuming and expensive due to the hydrogen consumption of full-size fuel cell stacks. Furthermore, the comprehensive development of a prototype stack would be required, which is costly and time consuming.

In order to quantify the potentials, trends and limits of evaporative cooling in an early development stage, a numerical model, which allows fast parameter studies, is beneficial. Therefore a zero-dimensional fuel cell system model has been developed, focusing on the fuel cell stack. Still, to prove the suitability of evaporative cooling for fuel cell applications, empirical verification will be ultimately required.

2.1. Overview and assumptions

The model presented here is zero-dimensional (i.e. it considers no spatial extent), semi-empirical and quasi-stationary. In the context of this work, semi-empirical means that the electrochemical performance model is based on parameters, gained in prior measurement campaigns with differential single cells [30,37]. Quasi-stationary, on the other hand, indicates that thermodynamic equilibrium is assumed in each time step and dynamic fuel cell behavior (i.e. capacitive as well as inductive effects) are neglected. Nevertheless, variables can change in time. Therefore the equations presented in the following chapters yield a solution that is time-averaged.

The developed model describes the thermodynamic and electrochemical behavior of a polymer electrolyte fuel cell stack operated with hydrogen and air. Therefore the mass and energy balances as well as the chemical reaction as well as the operating temperature are considered for the electrochemical performance, is averaged between inlet and outlet.

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Fig. 1. Modelling domain for mass and energy balance, showing molar flow rates \(n_i\) with their respective molar enthalpy \(H_{m,i}\) across the system boundaries, sink and source terms for hydrogen and oxygen consumption \((\dot{n}_{H_2}^{\text{con}}, \dot{n}_{O_2}^{\text{con}})\) as well as water production \((\dot{n}_{H_2O}^{\text{el}})\) due to the chemical reaction. White arrows indicate energy transfer as electricity \(P_{el}\) and heat \(Q_{surf}\), black solid line indicates the system boundaries.
xii. Identical performance of all cells in the stack is assumed.

The model consists of three sub-modules for mass balance, energy balance and electrochemistry. In order to enhance the readability, the mathematical model description is reduced to a minimum here. For further details please consult the provided supplementary information.

2.2. Mass balance

The steady-state mass balance of the system as depicted in Fig. 1 can be written for each species \(i\) in molar notation as follows:

\[
\sum n_i \dot{n}_i = 0
\]  

(1)

Where \(\dot{n}_i\) denotes the molar flow rates of the species \(i\) across the system boundaries and \(n_i\) is the respective sink or source term inside the control volume. Since no gas crossover is assumed, hydrogen only occurs at the anode whereas oxygen only occurs at the cathode. Nitrogen is considered separately for anode and cathode whereas the water balance on the other hand is written for the entire control volume as follows:

\[
\sum n_i \dot{n}_i = \dot{n}_{liq,i}^{in} + \dot{n}_{liq,i}^{out} + \dot{n}_{g,i}^{in} + \dot{n}_{g,i}^{out} - \dot{n}_{liq,i}^{out} - \dot{n}_{g,i}^{out} = 0
\]  

(2)

The superscripts \(a\) and \(c\) denote for anode and cathode respectively. \(\dot{n}_{liq,i}^{in}\) represents the liquid water supply. Assuming dry gas supply, fully saturated outlets and ideal water supply, the maximal evaporation rate \(\dot{n}_{liq,i}^{max}\) can be calculated as follows:

\[
\dot{n}_{liq,i}^{max} = \dot{n}_{liq,i}^{in} - \left( 1 - \frac{\rho_{sat,i}}{\rho_i} \right) \dot{n}_{g,i}^{in} - \dot{n}_{g,i}^{out}
\]  

(3)

Which can be further written as a function of temperature \(T\), pressure \(p_{act}\), stoichiometry \(\lambda^a\), \(\lambda^c\), dry gas composition of cathode and anode supply \(x_{H_2}^{in}, x_{O_2}^{in}, x_{N_2}^{in}\) as well as current density \(j\), active area per cell \(A_{act}\), number of cells \(k\), saturation pressure \(p_{sat,i}\) (as a function of temperature \(T\)) and Faraday constant \(F\). See supplementary information for detailed derivation.

\[
\dot{n}_{liq,i}^{max} = \dot{n}_{liq,i}^{in} - \frac{2}{2} F \left[ \lambda^a \left( \frac{x_{H_2,act}^{in}}{x_{H_2,act}^{in} + 1} \right) + \lambda^c \left( \frac{x_{O_2,act}^{in}}{x_{O_2,act}^{in} + 1} \right) - 1 \right] - \dot{n}_{g,i}^{out}
\]  

(4)

With the molar enthalpy of evaporation of water \(\Delta H_{vap,i}^{ev}\), the corresponding cooling power can be calculated as follows:

\[
\dot{Q}_{cool} = \dot{n}_{liq,i}^{max} \Delta H_{vap,i}^{ev}
\]  

(5)

2.3. Energy balance

For a zero-dimensional system as depicted in Fig. 1, the energy balance can be written as the first principle of thermodynamics for an open system, equation (6). The transfer of heat \(Q_{surf}\) and electrical power \(P_d\) as well as enthalpy and external energy of mass flow \(m_i(h_i + e_i)\) across the system boundary changes the internal energy \(\frac{dU}{dt}\) of the system. For steady-state conditions, the change in internal energy equals zero.

\[
Q_{surf} + P_d + \sum m_i(h_i + e_i) = \frac{dU}{dt} = 0
\]  

(6)

Heat \(Q_{surf}\) is transferred from the system to the environment by free convection over the surface of the fuel cell stack, where \(k_g\) is the heat transfer coefficient for free convection, \(T_{cell}\) is the cell temperature, \(T_{env}\) is the environmental temperature and \(A_{surf}\) is the surface area of the fuel cell stack.

\[
Q_{surf} = k_g A_{surf}(T_{cell} - T_{env})
\]  

(7)

Electric Power \(P_d\), can be written as a function of current density \(j\), active area per cell \(A_{act}\), actual cell voltage \(E_{cell}\) and number of cells \(k\).

\[
P_d = j A_{act} E_{cell} k
\]  

(8)

Neglecting the external energy of the mass flows across the system boundary \((e_i = 0)\) and assuming ideal gas behavior, the enthalpy flow rates across the system boundary can be written in molar notation as follows:

\[
\sum n_i(h_i + e_i) \approx \sum n_i H_m = \sum n_i \left( H_m^0 + \frac{\gamma}{2} \epsilon_{m,act} \Delta T \right)
\]  

(9)

The molar heat capacity at constant pressure \(\epsilon_{m,act}\) can be approximated for an ideal gas with a polynomial. The coefficients as well as the molar standard enthalpy of formation \(H_m^0\) have been taken from Ref. [49]. See supplementary information for details.

Neglecting the enthalpy change of the excess gases (the heat transferred to the excess gases is small compared to the other heat fluxes), the steady-state energy balance can be simplified as follows:

\[
j A_{act} k \left( -\frac{\Delta H_e^{ev}}{2} - E_{cell} \right) = k_g A_{surf}(T_{cell} - T_{env}) + \dot{n}_{liq,i}^{max} \Delta H_{vap,i}^{ev}
\]  

(10)

With \(\dot{n}_{liq,i}^{max}\) being the evaporation rate as a function of current density, temperature, pressure and stoichiometry according to equation (4), \(\Delta H_{vap,i}^{ev}\) is the molar heat of evaporation of water and \(\Delta H_e^{ev}\) is the molar enthalpy of reaction. The only unknown in equation (10) is the cell voltage \(E_{cell}\), which will be discussed in the following section.

2.4. Electrochemical performance

The actual cell voltage \(E_{cell}\) can be calculated as the reversible voltage \(E_{rev}\) minus the kinetic overpotential of the oxygen reduction reaction (ORR) \(\eta_{ORR}\), the ohmic losses due to the ionic resistance of the membrane \(j R_{mem}\) and the contact resistance between catalyst layer, gas diffusion layer and flow field \(j R_{cont}\) as well as the protonic resistance in the cathode catalyst layer \(j R_{H^+}\) and mass transport resistance \(\eta_{mix,\Delta O_2}\). The kinetic overpotential of the hydrogen oxidation reaction (HOR) as well as the protonic resistance of the anode catalyst layer have been neglected [50]. Neyerlin et al. [51] have shown that this assumption is justified for high RH conditions.

\[
E_{cell} = E_{rev} - \eta_{ORR} - j R_{mem} - j R_{cont} - j R_{H^+} - \eta_{mix,\Delta O_2}
\]  

(11)

Reversible cell voltage: The reversible cell voltage \(E_{rev}\) can be calculated according to equation (12), which is derived from Nernst
equation under the assumption of proton and water activity of unity, which is only given at relative humidities $\geq 100\%$. However, Neyerlin and co-workers [51] have shown, that the error is negligible for RH $> 50–60\%$.  

$$E_{\text{rev}} = E^0(T) + \frac{2.303 \, R \, T}{4 \, F} \log \left[ \frac{p_{H_2}}{p_{H_2, \text{in}}} \right]$$

(12)

Where $R$ is the universal gas constant, $T$ is the temperature at the catalyst layer, which is approximated with the cell temperature $T_{\text{cell}}$, $F$ is Faraday constant, $p_{H_2}$ and $p_{H_2, \text{in}}$ are the partial pressure of hydrogen and oxygen in the gas channel, $p_{H_2, \text{in}}$ is the reference partial pressure of 101.3 kPa and $E^0(T)$ is the standard potential. For details on the approximation of the temperature dependent standard potential consult the supplementary information.

**Kinetic overpotential:** The kinetic overpotential of the ORR, $\eta_{\text{kin}}$ is modeled according to equation (13) (i.e. Tafel kinetics with one constant Tafel slope over the entire current regime).

$$\eta_{\text{kin}} = \frac{2.303 \, R \, T}{\alpha \, F} \log \left[ \frac{j + j_0}{10 \, L_s n_p T_{\text{kin}}} \right]$$

(13)

$$j_0 = j_{\text{kin}} \frac{p_{O_2}}{p_{O_2, \text{kin}}}^\gamma \exp \left[ -\frac{E^*_{\text{ORR}}}{RT} (1 - \frac{T}{T_{\text{kin}}}) \right]$$

(14)

Where $\alpha$ is the cathodic transfer coefficient, $L_s$ is the cathode Pt loading, $n_p$ is the available Pt surface area per gram platinum, and $j_0$ is the exchange current density. $j_{\text{kin}}$ is the catalyst specific exchange current density normalized to reference oxygen partial pressure $p_{O_2, \text{kin}}$ of 101.3 kPa and a reference temperature $T_{\text{kin}}$ of 80 °C, $\gamma$ is the reaction order and $E^*_{\text{ORR}}$ is the activation energy of the ORR at the reversible cell potential. Further, $j_0$ is the hydrogen crossover current [52].

**Ohmic losses:** The Ohmic losses are modeled empirically, based on the measured high frequency resistance (HFR):

$$\eta_{\text{ohm}} = j \cdot R_{\text{ohm}} + \frac{j \cdot R_{\text{act}}}{1 + j} = j \cdot \text{HFR}$$

(15)

In order to account for different operating conditions, only the temperature dependency of the membrane resistance is taken into account since a sufficiently high humidification is assumed at all operating conditions for evaporative cooling. Kusoglu and Weber ([16], Figure 34d) summarize several studies that show logarithmic behavior of membrane conductivity $\kappa = \left(1/R_{\text{mem}}\right)$ with 1/temperature. The contact resistance is assumed to be constant.

$$\kappa(T) = 10^{-10^2}$$

(16)

**Mass transport losses:** Due to the limited mass transport through the gas diffusion layer, the oxygen concentration at the catalyst layer is lower than the concentration in the gas channel. This lower concentration affects the Nernst potential on the one hand as well as the ORR overpotential on the other hand. This change in cell voltage can be calculated according to equation (17). For the detailed derivation see Ref. [53].

$$\eta_{\text{O}_2, \text{ch}} = \frac{RT}{F} \left( \frac{1}{4} + \frac{1}{a} \right) \ln \left( \frac{p_{O_2, \text{ch}} - \frac{RT}{F} R_T j}{p_{O_2}} \right)$$

(17)

Where $R_T$ is the total oxygen transport resistance which can be determined experimentally by limiting current measurements [54].

In order to further improve the quality of the model and assure its applicability to evaporatively cooled cells, an experimentally determined polarization curve is used as a basis for the electrochemical performance. Subsequently, the changes in overpotentials between modelling conditions and measurement conditions are computed and thus the change in cell voltage can be obtained, see Fig. 2. It is assumed, that the changes of contact resistance, and protonic resistance of the cathode catalyst layer between modelling and measuring conditions are negligible.

$$\Delta E_{\text{cell}} = \Delta E_{\text{rev}} - \Delta \eta_{\text{kin}} - j \cdot \Delta R_{\text{mem}} - \Delta \eta_{\text{O}_2}$$

(18)

3. Simulation conditions and parameters

Simulations with variations of operating conditions were carried out for two different test cases. Test case 1 (differential cell test case) is used to map the measurement conditions used by Cochet et al. [37] in order to parametrize the model. Test case 2 (optimized stack test case) simulates extrapolations for a stack with an active area of 300 cm$^2$ per cell and 450 cells, corresponding to a nominal power of approx. 120 kW. Table 1 gives an overview of general simulation conditions and applied parameters.

<table>
<thead>
<tr>
<th>Differential cell test case (1)</th>
<th>Optimized stack test case (2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Active area per cell ($A_{\text{act}}$)</td>
<td>4.4 cm$^2$ [37]</td>
</tr>
<tr>
<td>No. of cells ($k$)</td>
<td>1</td>
</tr>
<tr>
<td>HFR at 80 °C and 100% RH</td>
<td>50 m2 cm$^{-2}$ [37]</td>
</tr>
<tr>
<td>Transport resistance ($R_t$)</td>
<td>1.5–2.75 s/cm [55]</td>
</tr>
<tr>
<td>Stack surface area ($A_s$)</td>
<td>0.72 m$^2$</td>
</tr>
<tr>
<td>Contact resistance ($R_{\text{cont}}$)</td>
<td>5 m2 cm$^{-2}$</td>
</tr>
<tr>
<td>Environmental temperature ($T_{\text{env}}$)</td>
<td>25 °C</td>
</tr>
<tr>
<td>Anode stoichiometry ($x^2$)</td>
<td>1.0</td>
</tr>
<tr>
<td>Anode dry gas composition</td>
<td>100 vol% hydrogen</td>
</tr>
<tr>
<td>Cathode dry gas composition</td>
<td>21 vol% oxygen, 79 vol% nitrogen (synthetic air)</td>
</tr>
<tr>
<td>RH at anode and cathode inlet ($R_{\text{in}}$)</td>
<td>0</td>
</tr>
<tr>
<td>Heat transfer coefficient ($h_{\text{in}}$)</td>
<td>5 W/(cm$^2$ K)</td>
</tr>
<tr>
<td>Cathode Pt loading ($L_s$)</td>
<td>0.4 mg/cm$^2$</td>
</tr>
<tr>
<td>Specific Pt surface area ($A_{\text{Pt}}$)</td>
<td>50 m$^2$/g [152] (rounded)</td>
</tr>
<tr>
<td>ORR activation energy ($E^*_{\text{ORR}}$)</td>
<td>67 kJ/mol [52]</td>
</tr>
<tr>
<td>ORR transfer coefficient ($\alpha$)</td>
<td>1 [53]</td>
</tr>
<tr>
<td>ORR reaction order ($\gamma$)</td>
<td>0.54 [52]</td>
</tr>
<tr>
<td>Reference exchange current density ($j_{\text{kin}}$)</td>
<td>2.5E-8 A/cm$^2$ [52]</td>
</tr>
</tbody>
</table>
Empirical polarization data: Fig. 2 shows the empirical polarization data that has been used as a baseline for simulations. The polarization curve has been determined in an evaporatively cooled, differential cell setup with an active area of 4.4 cm², at 80 °C, 200 kPa, dry gas supply and flow rates of 400 SCCM hydrogen at the anode and 1600 SCCM air at the cathode. It can be seen exemplarily how the simulated cell performance changes with operating pressure. Starting from experimental results (200 kPa, black squares) the performance improves with higher pressure (300 kPa, green line) and is decreasing with lower pressure (150 kPa, red line).

Oxygen transport resistance: The oxygen transport resistance values were taken from C. Simon [36]. He performed limiting current experiments with differential cells with an active area of 5 cm². The values were taken for Toray 030T gas diffusion substrate (hydrophobically treated with 20 wt% PTFE) with homemade carbon black MPL, 50 °C and 80 °C, 300 kPa, RH = 100%. Values of the oxygen transport resistance range between 1 s/cm at low current densities and are plateauing at ca. 2.75 s/cm at high current densities. To account for the actual thickness of the diffusion medium as well as for the actual operating conditions, corrections have been made according to Baker and co-workers [54]. More details are given in the supplementary information.

4. Results and discussion

In the following subsections, the results of the parameter study are presented. Suitable operating conditions, required evaporation rates, as well as the impact of evaporative cooling on the fuel cell performance are discussed. Eventually, a feasible operating window for evaporative cooling is proposed.

4.1. Impact of operating conditions on cooling performance

In order to determine the optimal operating conditions for evaporative cooling, the impact of different operating conditions on the evaporation rate and thus the cooling power has been studied. Fig. 3 shows simulations for a fuel cell stack with 450 cells and an active area of 300 cm², operated at 1 A/cm² as a function of operating pressure, temperature and cathode stoichiometry. The anode stoichiometry is held constant at 1.0 to mimic the conditions in a fuel cell system (i.e. recirculation of humid hydrogen and stoichiometric supply of dry hydrogen). The anode recirculation rate is not considered in this model, since the amount of supplied dry gas is not affected and thus, the evaporation rate and cooling performance will remain unchanged.

First, it can be seen that the evaporation rate increases with increasing temperature. This can be explained by the fact that the water saturation pressure also increases with temperature and thus more water can evaporate. Second, the evaporation rate increases with cathode stoichiometry because at higher stoichiometry the supply of dry air is increased. Lastly, the evaporation decreases with increasing gas pressure, the reason being that the volume flow rate decreases with higher pressure at constant stoichiometry. Thus, the dry gas volume flow decreases, which in turn reduces the water removal capability of the gas stream. Furthermore, Fig. 3 shows that a sufficiently high cooling performance (>100 kWth) can be achieved to reject the entire waste heat of the stack at beginning and end of life.

Fig. 4 shows the sensitivity of the cooling power towards different levels of the input parameters temperature, pressure and cathode stoichiometry. It can be seen that a 10% change in temperature shows the highest leverage, followed by the pressure and the stoichiometry. However, there are strong correlations between the input quantities. It can be seen, that a 10% change in the aforementioned parameters has a higher impact at high temperature (90 °C) and low pressure (150 kPa), whereas it is less pronounced at lower temperature (80 °C) and higher pressure (200 kPa).

Based on the results, an evaporatively cooled stack shall be operated at increased temperature, increased cathode stoichiometry and decreased pressure, from a thermodynamic perspective.

4.2. Impact of operating conditions on thermal steady state

The energy balance of the fuel cell stack has been investigated in order to determine feasible combinations of the operating parameters (temperature, pressure, stoichiometry and current density) that allow a complete removal of the waste heat (i.e. thermal steady state) by evaporative cooling.

Fig. 5a shows an evaluation of the energy flow rates across the system boundaries as a function of current density at 87 °C (solid) and 89 °C (dashed), 200 kPa and a cathode stoichiometry of 2. The enthalpy of the reactants (black curve) increases linearly with current density due to the constant stoichiometry. However, the electric power output (red curve) shows a non-linear increase with current density, since the cell voltage (Ecell) decreases with increasing current (see eq. (11)). The heat transport due to surface cooling (blue curve) is comparably small but it has to be taken into account to model the thermal stack behavior at low current densities correctly. Lastly, the heat flow rate due to evaporation (solid green curve) increases linearly with current density, due to the increased gas flow rates with increasing current densities (assuming constant stoichiometry). Note: In the model presented here, the evaporation is only limited by the saturation of the gas flows. It can be further seen that the largest portion of the heat is removed by evaporative cooling, whereas the heat transfer to the environment by surface cooling is comparably insignificant. The green dashed line indicates the increase in heat removal by evaporation due to a temperature increase by 2 °C.

Fig. 5b shows the corresponding change in internal energy (dU/dt) of the system (i.e. the sum of all heat fluxes in Fig. 5a). If the value is equal to zero, the system is thermally balanced and the temperature remains constant. At a positive value, however, the stack temperature will increase whereas it will decrease at a negative value. The change in internal energy is slightly negative at OCV due to the surface cooling effect. At low current densities it decreases due to the prevailing effect of evaporative cooling, whereas it increases with higher current density due to an over-proportional increase of waste heat. It can be seen, that the steady-state condition (dU/dt = 0) is only given at one current density. Therefore, the stack will heat up or cool down at different current densities. In the presented case the thermal equilibrium at 87 °C is reached at about 0.3 A/cm². A load increase to about 0.9 A/cm² will lead to an increase in stack temperature (positive dU/dt). However, when the stack heats up, the evaporation rate increases as well (due to increased temperature, see Fig. 3) which will in turn increase the heat removal. Thus, a new steady state is reached at a slightly increased temperature of 89 °C.
More generally speaking, a defined combination of temperature, pressure, stoichiometry and current density is required to reach a thermal steady state (see equation (10)). However, there exist multiple feasible combinations in the 4-dimensional parameter space that allow a complete heat removal by evaporation. Fig. 6 shows steady-state operating conditions at 1 A/cm\(^2\) (black curves) and 2 A/cm\(^2\) (red curves) and an anode stoichiometry of unity. Following the iso-stoichiometry lines allows the removal of the entire waste heat by evaporative cooling.

In addition, Fig. 6 indicates that a load change towards higher values will increase the steady-state temperature, if all other operating conditions remain constant. To avoid thermal cycling with load, the temperature change could be counteracted by a load-dependent change in pressure and/or stoichiometry.

4.3. Electrochemical performance

As shown above (discussion of Fig. 3), the favorable operating conditions for evaporative cooling are shifted towards higher temperatures and lower pressures compared to conventional fuel cells. Thus, an impact on the fuel cell performance is expected.

Fig. 7 shows simulated polarization curves for conventional and evaporative cooling for a single cell in the optimized stack test case. It can be seen, that the performance of the evaporatively cooled cell is slightly lower (ca. \(-23.5\) mV at 1.5 A/cm\(^2\)), representing a volume increase of \(-3\)–\(-4\)\%, due to the lower operating pressure. Moreover, the limiting current is also shifted towards lower current densities due to the decreased oxygen partial pressure. However, the decrease in power density is small compared to the volume saving potential of evaporative cooling.

4.4. Operating window for evaporative cooling

The physical limitations of the parameter space and thus boundaries for the calculation domain can be summarized as follows:

**Temperature:** The maximal temperature is limited by the polymer electrolyte membrane. The material starts decomposing at about 280 °C \([57,58]\), however, glass transition sets in already at 140 °C \([59]\). More importantly, the membrane humidification and corresponding protonic resistance becomes already challenging at temperatures above 95 °C. The minimal temperature is physically limited at around 5 °C due to freezing risks. However, the achievable evaporation rates and fuel cell performance will limit the temperature at around 60 °C on the lower boundary. Nevertheless, the technology is freeze start capable, since no water would be injected at sub-zero temperatures to achieve a rapid heat up.

**Pressure:** The minimal pressure is limited at ambient pressure whereas the upper physical limit is determined by the mechanics of the components and the membrane. However, in a fuel cell system, the power consumption for the air blower might limit the pressure at significantly lower levels.

**Anode stoichiometry:** The lower limit is 1, whereas the upper limit is assumed 1.05 due to the hydrogen consumption. The recirculation rate shows no impact on evaporation since the amount of dry gas supplied is unchanged.

**Cathode stoichiometry:** The lower limit is 1 due to the electrochemical performance. There is no upper limit from the stack level perspective, however, in a fuel cell system the power consumption of the air blower might limit the stoichiometry.

**Current Density:** The lower limit is 0 and the upper limit is assumed at 2.5 A/cm\(^2\) for recent technology and technical cell size (see Fig. 7).

Taking into account the limitations mentioned above and considering the trade-off between cooling power and electrochemical performance, the operating window for evaporative cooling can be estimated. Fig. 8 summarizes the constraints and presents a possible operating window for evaporative cooling (red), which is slightly shifted towards higher temperatures (80–95 °C), lower pressures (100–200 kPa) and
higher cathode stoichiometries (>1.5) in comparison to a conventional fuel cell stacks (blue).

However, the lower temperature limitation as well as the upper limit of system pressure are determined by balance of plant (BoP) components. Therefore, a more detailed analysis at the system level, to define the operating window and system efficiency more precisely, is conducted in the second part of this study.

4.5. Challenges of the evaporative cooling concept

Evaporative cooling has the potential to reduce the stack volume by up to 30% and to allow the operation at increased temperature without external humidification, however, the technical application of the concept also entails several challenges:

Water recovery and quality: In order to avoid the storage and refilling of liquid water, the presented concept foresees the condensation of water vapor from the exhaust gas to ensure a closed water loop. The size of a condensing radiator depends strongly on the exhaust gas and ambient temperature. It has to be shown, if enough water can be condensed at the given operating conditions. Therefore, we will analyze the evaporative cooling concept in further detail on the system level in our future work. Furthermore, several questions related to the corrosion of the condenser, as well as the water quality (e.g. conductivity) have to be solved.

Inhomogeneous temperature and humidity distribution: Due to the microstructure of the GDL with patterned wettability, the evaporation and thus cooling and humidification take place locally at the hydrophilic lines. In addition, the relative humidity of hydrogen and air changes along the gas channel, which affects the local evaporation rates and thus may lead to non-uniform cooling and humidification along the channel. This can be mitigated by using a GDL line pattern with gradually decreasing distance between the hydrophilic lines along the
channel. However, this shows the potential of an uneven temperature distribution which might enhance local degradation phenomena. It has to be shown in further studies that the high thermal in-plane conductivity of the GDL can smooth out temperature peaks in the membrane and catalyst layer.

**Stability:** Studies on the material level have shown, that liquid water can be kept in the hydrophilic lines at capillary pressure (water pressure subtracted by the gas pressure) up to 40 mbar [60], however, the pressure drop along the anode gas channel is in the range of 100 mbar or more. Therefore, further material improvements are required to allow robust operation on a technical cell size. Furthermore, the dynamic operation of the fuel cell stack can lead to pressure transients which may contribute to the aforementioned limitation and be challenging for the development of suitable control strategies.

**Freeze start:** The freeze start capability of evaporative cooling has not been proven yet. Due to the increased amount of liquid water in the cell, a more sophisticated drying strategy will have to be applied during shutdown which might lead to increased shutdown durations at sub-zero conditions. Furthermore, the condenser has to be bypassed and or heated during startup to avoid condensation and freezing of the product water at temperatures below 0 °C. This might lead to an increased complexity of the system design and the control strategy.

**GDL degradation:** The long term stability of the hydrophilic pattern in the GDL under fuel cell operating conditions has to be analyzed in future work. A degradation of the hydrophilicity could lead to a limitation of the above mentioned capillary pressure window.

### 4.6. Limitations of the applied modelling approach

The zero-dimensional, steady-state modelling approach has been chosen to perform fast parameter studies and allows the integration into a fuel cell system model. However, to enable faster computation times, several trade-offs had to be made:

- **Spatial effects:** Due to the zero-dimensional nature of the model, spatial distributions of reactant concentration and properties like pressure and temperature as well as the current density and thus waste heat production are neglected. These effects are partly accounted for by the semi-empirical approach (use of an experimentally determined polarization curve as model input), however, depending on the operating conditions, this assumption might lead to a considerable error when extrapolating to a technical cell size. Nevertheless, the model reasonably captures the performance trends of evaporative cooling as a function of different operating conditions.

- **Hydrogen crossover:** Hydrogen crossover is indirectly accounted for by the semi-empirical nature of the model. However, the pressure dependency of the crossover current has been neglected since we assume, that especially at high current densities, the effect is small compared to other losses. Nevertheless, this simplification might lead to a slight overestimation of fuel cell performance and thus an underestimation of waste heat.

- **Degradation:** Degradation of the catalyst layer and the polymer electrolyte membrane over the lifetime of the stack has not been accounted for in the presented model. Degradation might lead to a decreased performance and thus increased waste heat production over the lifetime of the stack. The U.S. Department of Energy uses a 10% voltage drop compared to beginning of life performance as a measure for degradation [12]. It has been shown by Kurtz et al. [61] that the fleet averaged degradation of 10% is reached after 2000–3000 h in real life operation. Several vehicles have exceeded 5000 h. However, the DOE ultimate durability target of 6000 h [12] has not been met so far. A 10% decrease in voltage would eventually lead to a 10% increase in waste heat at end of life. According to Fig. 3 this increased heat production can be easily removed by evaporative cooling, however, the operating window will be slightly shifted towards higher temperature and stoichiometry as well as slightly lower pressure.

### 5. Conclusion

To analyze the suitability of the evaporative cooling concept for PEFC, a zero-dimensional fuel cell system model has been developed, enabling fast parameter studies. It has been shown that evaporative cooling works effectively over a broad range of operating conditions and a suitable operating windows has been quantified. The proposed operating window is not smaller compared to state-of-the-art liquid cooled PEFC stacks, however the optimal operating conditions are slightly shifted towards higher temperatures (80–90 °C), lower pressure (100–200 kPa) and higher cathode stoichiometric ratios (>1.5).

Higher operating temperatures are beneficial for fuel cell cooling in general but for evaporative cooling, an increase in temperature shows by far the highest leverage to increase the cooling power. However, the maximal operating temperature is limited by applied materials (e.g. PFSA membrane). Therefore, future material developments, such as more temperature-stable polymer electrolyte membranes will significantly increase the potential of evaporative cooling.

Furthermore, we have shown that an evaporatively cooled fuel cell stack is not operated isothermally in a self-controlled manner. In contrast, the temperature will follow the power demand (at constant operating conditions) or can be controlled by a model-based adjustment of operating pressure and/or stoichiometry. The presented model can contribute to the development of operating strategies or be applied in fuel cell control units due to its fast computing time.

The performance of an evaporatively cooled stack is comparable to conventional fuel cell stacks. A slightly lower specific performance, due to the lower operating pressure, is easily compensated by the achieved mass and volume savings. Moreover, evaporative cooling allows to decouple the water management from operating conditions by providing the flexibility of several suitable combinations thereof and allows an optimal membrane humidification at high temperature and low pressure.

Therefore, evaporative cooling can significantly contribute to achieving stack and system power density targets as well as substantial cost reductions without compromising fuel cell performance. However, system level constraints, such as the power consumption of the air blower as well as the requirements of an exhaust gas condenser, ensuring a closed water loop, have to be investigated in future work.

### CRediT authorship contribution statement

**Michael Striednig:** Methodology, Software, Visualization, Investigation, Writing – original draft. **Magali Cochet:** Formal analysis, Writing – review & editing. **Pierre Boillat:** Funding acquisition, Conceptualization, Writing – review & editing. **Thomas J. Schmidt:** Resources, Project administration, Writing – review & editing. **Felix N. Büchi:** Funding acquisition, Conceptualization, Writing – review & editing, Supervision.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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