Water-Balance-Based Optimal Control for High-Current Operation of PEM Fuel Cells (Postprint)

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Abstract

Water management in Proton Exchange Membrane Fuel Cells (PEMFC) constitutes one of the primary factors influencing their performance. This paper proposes a temperature control method based on Membrane Electrode Assembly (MEA) water balance for PEM fuel cells operating at high current densities. Under specific operating pressures and stoichiometric ratios, the water balance temperature based on zero net electro-osmotic drag was determined for PEM cells and subsequently refined through three-dimensional simulation to obtain the operating temperature corresponding to optimal cell performance. The actual operating temperature control for the cell can be implemented based on the zero net electro-osmotic drag water balance temperature curve with the addition of a correction term, thereby enabling the cell to operate within a region of favorable performance. This control algorithm for cell operating temperature is straightforward and offers valuable reference for the design and operational optimization of fuel cells.

Full Text

Preamble

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Optimal Control of PEM Fuel Cells Operating at High Current Density Based on Water Balance

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Abstract: Water management is one of the primary factors affecting the performance of Proton Exchange Membrane Fuel Cells (PEMFC). This paper proposes a temperature control method based on Membrane Electrode Assembly (MEA) water balance for PEMFC operated at high current density. Under specific operating pressure and stoichiometric coefficients, the water-balance temperature of the PEMFC was obtained based on the condition of no net electro-osmotic drag. This temperature was then refined through three-dimensional simulation to determine the optimal operating temperature for cell performance. The actual operating temperature control can be implemented using the no-net-drag water-balance temperature as a baseline, plus a correction term, enabling the fuel cell to operate within a high-performance region. This temperature control algorithm is simple and provides a valuable reference for fuel cell design and operational optimization.

Keywords: PEM fuel cell; Membrane electrode; Water balance; Optimization; Temperature control

Introduction

Proton Exchange Membrane Fuel Cells (PEMFC) directly convert chemical energy stored in hydrogen into electrical energy through electrochemical reactions. They have attracted significant global attention due to their high energy conversion efficiency, low noise, zero pollution, rapid startup, and high power density. To meet the demanding requirements of automotive applications in confined spaces and under complex operating conditions, PEMFC performance, power density, and durability continue to improve while costs steadily decline.

In November 2013, Intelligent Energy in the UK announced the highest automotive fuel cell power density achieved to date, with volumetric and gravimetric power densities reaching 3.7 kW/L and 2.5 kW/kg, respectively [1]. In 2014, Toyota launched the MIRAI fuel cell vehicle, achieving stack gravimetric and volumetric power densities of 3.1 kW/L and 2.0 kW/kg [2]. High-power operation of fuel cell stacks generally requires increased back pressure to enhance reactant gas concentration. Simultaneously, current densities are often high, potentially reaching 1.0–2.0 A/cm^2, resulting in substantial water generation. If this product water can be effectively utilized, the humidification system can be simplified. Consequently, fuel cell stacks operating with increased pressure, unhumidified cathode inlet, and high current density have become a development trend [3, 4].

When applied in vehicle power systems, the control strategy is a critical factor determining whether the fuel cell can operate efficiently, durably, and stably.

Optimized control strategies depend on effective identification of the stack' s operating state, among which accurate identification of the MEA hydration state is extremely important [5] yet exceptionally challenging.

Researchers including Yuan [6], Jang [7], and Ding [8] have investigated optimal operating parameters such as temperature, gas humidity, operating pressure, and flow field structure through simulation and experimental methods. Kanani [9] and Damour [10] proposed control models based on Response Surface Methodology and Flatness-based Approach, respectively, to regulate operating conditions. Hu [11] and Cheng [12] employed model-based methods to control MEA flooding state (via anode purge) and operating temperature, applying these to actual bus fuel cell systems. However, the accuracy of these models, particularly the reliability of MEA flooding models, directly impacts whether the fuel cell can operate efficiently and stably.

Shaul [13] suggested that operating PEMFC without external humidification of reactant gases, relying solely on product water to humidify the membrane, is feasible and proposed corresponding criteria. Janssen [14] experimentally studied the effects of operating conditions on the net electro-osmotic drag coefficient, concluding that it is minimally affected by current density and remains nearly constant under unhumidified cathode and humidified anode conditions, with electrochemically produced water being almost entirely removed by the cathode reactant gas. Based on this, Berning [15, 16] explored the ideal water balance state where both anode and cathode inlet gases are unhumidified and outlet gases are just saturated, proposing a control method using cathode and anode outlet dew point temperatures. However, they found that this control approach could achieve MEA water balance but not optimal cell performance, with strict constraints on gas stoichiometric coefficients.

In light of these limitations, this paper focuses on PEMFC operating at high current density with unhumidified cathode and pressurized anode/cathode. A zero-dimensional water balance model is established to obtain the water-balance temperature under no net electro-osmotic drag for various back pressure conditions and stoichiometric coefficients. Subsequently, considering small net electro-osmotic drag coefficients, the water-balance temperature is refined. Finally, three-dimensional simulation is employed to account for electro-osmotic drag and liquid water effects, optimizing the water-balance temperature for optimal cell performance under corresponding operating conditions.

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1. Zero-Dimensional Model of Water Transport in Fuel Cells

[Figure 1: see original paper] illustrates the water transport process in a PEM fuel cell, showing both overall cell water transport and membrane water transport. Under steady-state conditions, the water entering and exiting the anode and cathode flow channels relates to the water produced by reaction as follows:

Cathode inlet water flow:
Anode inlet water flow:

Cathode outlet water flow:
Anode outlet water flow:

Defining water flow from anode to cathode as positive, the net electro-osmotic drag flux in the membrane is:

This maintains water balance between anode and cathode while ensuring membrane hydration. The variables represent:
- wq^-: cathode inlet water flow
- $in\,anwq^-$: anode inlet water flow
- wq^-: water generated in cathode MEA
- $out\,wq^-$: cathode outlet water flow
- $out\,anwq^-$: anode outlet water flow
- $mem\,wq^-$: net water flux through membrane
- $backq$: back-diffusion flux
- $EODq$: electro-osmotic drag flux
- $press$: net electro-osmotic drag coefficient
- dr: pressure-driven diffusion flux
- hum: air humidity ratio
- $(hum\,2Hd)$: air humidity ratio
- $cellT$: cell temperature
- bp: operating pressure
- cS: cathode air stoichiometric coefficient
- aS: anode hydrogen stoichiometric coefficient
- $2ox$: oxygen volume fraction in air (typically 1/5)
- i: current density (A/cm²)
- F: Faraday constant (96487 C/mol)
- $airM$: molar mass of air (28.8 kg/kmol)
- $2HM$: molar mass of hydrogen (2 kg/kmol)
- wq^-: molar mass of water (18 kg/kmol)

The water-balance temperature value is independent of current density and depends only on pressure and stoichiometric coefficient. When operating temperature is below the balance point temperature, $q\Delta$ is positive, indicating that after product water is removed by saturated air, excess water remains in the MEA as liquid water, increasing with current density. When operating temperature exceeds the balance point temperature, $q\Delta$ becomes negative, indicating that after product water is completely removed, additional water is required, causing MEA dehydration. In actual fuel cell operation, this additional water primarily comes from humidification water and anode electro-osmotic drag water, with the required amount increasing with current density.

[Figure 4: see original paper] shows the relationship between $q\Delta$ and current density/operating pressure at a stoichiometric coefficient of $S_c = 2.5$ and temperature of 70°C. The figure demonstrates that $q\Delta$ increases with pressure, indicating that elevated pressure reduces the gas water-carrying capacity, leaving more residual water in the MEA. At 200 kPa, $q\Delta$ is nearly zero and independent of current density, consistent with [Figure 2: see original paper] where the balance temperature is 69.59°C at 200 kPa.

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2. Water Balance in MEA with Unhumidified Cathode

For the operating conditions studied in this paper, the cathode gas is unhumidified ($0 = in\,c\,q$). Based on Janssen' s research, the total water flux through the membrane caused by electro-osmotic drag, back-diffusion, and pressure-driven diffusion (i.e., net electro-osmotic drag) is much smaller than the water production rate [14]. Therefore, it is possible to utilize cathode gas to remove product water while achieving saturated outlet conditions and ensuring no excess water accumulates in the MEA [13]. However, this water balance condition tends to leave the membrane slightly dry, resulting in suboptimal cell performance [15]. When cell performance is optimal, a small amount of liquid water typically exists on the cathode side of the MEA, providing ideal membrane hydration [6]. This liquid water forms from condensation of product water and electro-osmotic drag water, subsequently driven into the flow channels by capillary pressure, resulting in supersaturated gas at the cathode outlet.

2.1 Water Balance in MEA with No Net Electro-Osmotic Drag

Assuming zero net electro-osmotic drag between anode and cathode (no water transport between electrodes), the difference between product water and cathode outlet water flow is defined as $q\Delta$:

According to the Clausius-Clapeyron equation, the relationship between saturated water vapor pressure and temperature is [17]:

where $satp$ is the saturated water vapor pressure at temperature T.

[Figure 2: see original paper] and [Figure 3: see original paper] show the relationship between $q\Delta$ and temperature/current density at a cathode stoichiometric coefficient of 2.5 and operating pressures of 200 kPa and 100 kPa, respectively. For a given operating pressure, $q\Delta$ decreases to zero with increasing temperature and becomes negative thereafter, with its absolute value increasing with current density. The point where $q\Delta = 0$ represents the condition where all product water is removed by dry cathode gas and reaches saturation at the outlet, defined as the MEA water-balance point, with the corresponding temperature termed the water-balance temperature. At 100 kPa, the balance temperature is 54.3°C; at 200 kPa, it is 69.59°C; and at 300 kPa, it is 79.33°C. Notably, the balance temperature is independent of current density.

[Figure 5: see original paper] demonstrates that the water-balance temperature increases with operating pressure but decreases with increasing cathode stoichiometric coefficient. This indicates that for a given pressure, increasing the stoichiometric coefficient enhances water vapor capacity without requiring higher temperature, while for a given stoichiometric coefficient, increasing pressure reduces the gas water-carrying capacity.

These equations provide a relatively simple method to determine the balance temperature under specific operating conditions. However, operating along the balance temperature line leaves the membrane dry, resulting in suboptimal cell performance [15].

2.2 Water Balance Considering Net Electro-Osmotic Drag

Under unhumidified cathode and humidified anode conditions, the net electro-osmotic drag coefficient is small [14], meaning most water dragged from the anode returns via back-diffusion. This is particularly true for the 15-micron Nafion membrane considered in this study, providing the theoretical basis for using the balance temperature (dew point temperature) to control cell water balance as shown in [Figure 5: see original paper]. However, as previously noted, balance temperature control leads to membrane dryness [15]. To achieve optimal membrane hydration and cell performance, an appropriate amount of liquid water should exist in the MEA, requiring correction of the balance temperature in [Figure 5: see original paper] to account for small net electro-osmotic drag and condensation effects.

To incorporate net electro-osmotic drag effects, let $mem\,wq^{-}$ represent the net electro-osmotic drag water. The sum of net electro-osmotic drag water and product water is removed by cathode outlet gas, modifying equation (10) to:

[Figure 6: see original paper] shows $q\Delta$ variation with net electro-osmotic drag coefficient at $S_c = 2.5$ and $bp = 300$ kPa, with net electro-osmotic drag coefficients of 0.05 and 0.1 based on Janssen' s research [14]. Since net electro-osmotic drag is positive (water dragged from anode to cathode), higher temperatures are required to remove this additional water along with product water. The water-balance temperatures are 81.36°C and 83.2°C for net electro-osmotic drag coefficients of 0.05 and 0.1, respectively.

At higher back pressures, the optimal performance temperature and water-balance temperature become very close. When operating at elevated back pressure, the cathode gas water-carrying capacity decreases, allowing good membrane hydration even when cell temperature is slightly below the balance temperature, consistent with the trend shown in [Figure 4: see original paper].

2.3 Water Balance Based on Optimal Performance

The aforementioned dew-point-based MEA water-balance temperature control, whether considering net electro-osmotic drag or not, tends to result in dry proton exchange membranes that do not achieve optimal hydration, leading to suboptimal cell performance. Therefore, this paper employs computational fluid dynamics software (Fluent) to perform three-dimensional simulations and determine the optimal operating temperature for cell performance.

The main structural parameters and operating conditions for the simulation are listed in . Due to space limitations, the heat and mass transfer equations, electrochemical equations, and specific physical and electrochemical parameters can be found in [6].

[Figure 7: see original paper] and [Figure 8: see original paper] show the optimal performance temperatures (dashed lines on the left) obtained from simulation at a stoichiometric coefficient of 2.5 and back pressures of 100 kPa and 300 kPa, respectively. In [Figure 7: see original paper], the cell balance temperature is 54.3°C, independent of current density. At this temperature, product water is just removed by cathode gas while maintaining saturated outlet conditions. However, the actual optimal operating temperature is approximately 47.7°C (varying slightly with current density). At this temperature, a small amount of liquid water exists on the MEA cathode side, but the membrane is well-hydrated. Considering both oxygen transport and membrane conductivity, cell performance is optimal, although the cathode outlet gas is supersaturated. Operating below this temperature causes severe MEA flooding, while operating above it gradually dries the membrane. [Figure 8: see original paper] shows a similar partitioning, though the optimal performance temperature and water-balance temperature are closer at higher pressure.

Berning et al. proposed controlling stack operating temperature using the balance temperature [14-16], but this approach results in dry membranes regardless of whether net electro-osmotic drag is considered. This paper seeks optimal performance conditions based on existing mass transfer and electrochemical models, while acknowledging that: (1) current models are imperfect and results may deviate from reality; (2) physical quantities such as temperature are non-uniformly distributed within the stack, making strict control to a single temperature value impractical; (3) changes in stack and MEA materials and structure will shift the optimal performance line; and (4) control system errors exist. Therefore, this paper proposes maintaining operation within a high-performance region rather than strictly controlling to a calculated optimal performance line.

[Figure 9: see original paper] compares the simulation-refined optimal performance temperature with the no-net-drag water-balance temperature at a cathode gas stoichiometric coefficient of 2.5. The optimal operating temperature is lower than the water-balance temperature. This defines the fuel cell operating temperature range: operating below the optimal temperature line causes flooding; operating between the optimal and water-balance temperature lines yields good performance; operating above the water-balance temperature line results in membrane dryness. Actual cell temperature control can use the no-net-drag water-balance temperature as a baseline plus a correction term to maintain operation in the high-performance region, providing a simple control algorithm.

Conclusions

This study investigated water transport and balance characteristics in PEMFC operating at high current density, proposing a temperature control method based on MEA water balance. Utilizing product water, the system employs unhumidified cathode inlet, humidified anode inlet (simulating recirculation humidification), and pressurized operation. The main conclusions are:

1. Under determined operating pressure and stoichiometric coefficients, a water-balance temperature exists for the MEA based on no net electro-osmotic drag, independent of current density. At this temperature, product water is just removed by cathode outlet air. Operating above this temperature causes MEA dehydration, while operating below it may cause partial flooding. The balance temperature increases with back pressure and decreases with cathode gas stoichiometric coefficient.
2. The no-net-drag water-balance temperature algorithm is simple, but controlling cell operation at this temperature results in dry membranes.
3. Three-dimensional simulation refinement of the no-net-drag balance temperature yields the optimal operating temperature for cell performance. As operating pressure increases, the optimal performance temperature and no-net-drag balance temperature gradually converge. Operating at the optimal temperature line results in slight MEA flooding, but considering the combined effects of membrane hydration and oxygen transport, cell performance is optimal.
4. Actual cell temperature control can use the no-net-drag water-balance temperature line as a baseline plus a correction term to maintain operation in the high-performance region, providing a simple control algorithm.

The optimal operating temperature obtained through simulation refinement of the water-balance temperature will vary with MEA structure and material composition. However, the identified trends and the proposed temperature control methodology provide valuable references for fuel cell design and operational optimization.

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Note: Figure translations are in progress. See original paper for figures.