Temperature regulation in an evaporatively cooled proton exchange membrane fuel cell stack

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ABSTRACT

Maintaining proton exchange membrane fuel cell (PEMFC) stack operating temperature across transient current profiles presents a significant challenge for fuel cell vehicles. Liquid cooled systems require active control of coolant temperature and flow rate to match heat rejection to heat generation. Evaporative cooling is an alternative to conventional liquid cooling in automotive sized PEMFC stacks. In an evaporatively cooled system, liquid water is injected directly into the cathode flow channels where it evaporates, both cooling and humidifying the stack. This paper uses a validated simulation to explore the inherent temperature regulation abilities of an evaporatively cooled PEMFC stack across a range of current profiles and drive cycles. Results show that throughout the normal operating current range, stack temperature varies by less than ±2.0 °C, this is comparable to liquid cooling but without the need for active temperature control. The introduction of variable operating pressure and cathode stoichiometry using proportional integral control, can further reduce temperature variation to ±1.0 °C and ±1.2 °C respectively for step increases in current demand. Variable operating pressure is also shown to improve warm up time and reduce heat loss at low operating loads.

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Introduction

Proton exchange membrane fuel cell (PEMFC) systems are seeing increased use as alternatives to internal combustion engines for motive power due to zero harmful tailpipe emissions and competitive driving range. In a conventional PEMFC, liquid water must be present to facilitate the transport of protons across the membrane, meaning the fuel cell stack temperature must remain below the boiling point of water to ensure high efficiency. The transient loads and vehicle speeds of automotive drive cycles, combined with the large thermal inertia of the fuel cell stack and low exhaust heat flow presents a challenge in regulating fuel cell stack temperature within acceptable limits, particularly at high ambient temperatures [1].

Traditionally waste heat from the electrochemical reaction in automotive stacks is removed from the fuel cell stack by passing coolant, usually water, through channels between the cells [2]. The heated coolant is then cooled in a conventional vehicle radiator; a diagram of a typical liquid cooled system is shown in Fig. 1. For accurate temperature regulation heat rejection from the stack must closely match the waste heat generated, this is achieved by controlling both the coolant inlet temperature (through a radiator by-pass) and coolant flow rate (variable speed pump) [3]. Thermal gradients across

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the fuel cell stack should also be kept small to reduce mechanical stress and degradation [4], limiting the coolant temperature change within the fuel cell stack. Temperature regulation in liquid cooled systems has been extensively studied in the literature by Refs. [3–8] among others, and a variety of control methods have been used; including pulse width modulation, proportional integral (PI), model predictive, optimal and fuzzy logic. Temperature variation across transient drive cycles for active liquid cooling in the literature is generally in the region of ±2.0 °C; however, of the referenced studies only [3] considers how changes in system parameters such as voltage degradation will affect controller performance.

Evaporative cooling is an alternative to liquid cooling where waste heat is removed through the evaporation of liquid water within the fuel cell flow channels, both cooling and humidifying the cells [2]. The evaporated water, along with some of the product water, is then condensed from the cathode exhaust using a heat exchanger and the liquid water can be re-used for cooling the stack. Because humidification and cooling happens within the flow channels of an evaporatively cooled system there is no need for an external humidifier or cooling plates within the fuel cell stack [9], a diagram of a typical evaporatively cooled system is shown in Fig. 2.

Evaporatively cooled PEMFC systems have been studied by [9–14]; [9–11] have produced commercial evaporatively cooled systems which have been used in several automotive applications. [13,14] have both produced models of evaporatively cooled systems [14], also investigated steady state heat transfer and required radiator frontal area. However none of the previous published work has studied temperature regulation of an evaporatively cooled fuel cell stack.

The amount of heat removed from the stack, and hence temperature, is directly related to the amount of liquid water which can be evaporated. It is well documented that for optimum performance a PEMFC should be run at high relative humidity, in the case of saturation the maximum mass flow rate of vapour is dictated by equation (1) [15].

\[
\dot{m}_{H_2O_v} = \frac{M_{H_2O}}{M_{air}} \frac{P_{sat}}{P - P_{sat}} \dot{m}_{air}
\]  

(1)

\[
Q = \Delta H_v \dot{m}_{H_2O_v} = \Delta H_v M_{H_2O} \frac{P_{sat}}{P - P_{sat}} \ln\frac{X_{O_2}}{x_{O_2}} \frac{4F}{2}
\]  

(2)

Assuming vapour flow into the stack is negligible, equation (1) demonstrates that the amount of water evaporated is dependant on the air mass flow rate, operating pressure and saturation pressure (which is a non-linear function of temperature). By expanding the air mass flow rate term in equation (1) and relating to thermal energy using the enthalpy of vapourisation of liquid water (\(\Delta H_v\)), equation (2) shows the heat removal through evaporative cooling. As air flow rate increases with current demand, so does heat rejection. An advantage of this relationship is that the stack temperature in an evaporatively cooled system is self-regulating, provided humidity is maintained. At constant stoichiometry and voltage, both heat rejection and heat generation will vary...
linearly with current demand, maintaining constant temperature. In reality, heat generation increase with current is non-linear due to voltage reductions and an increased amount of water vapour must be evaporated to maintain thermal balance, causing an increase in stack temperature; however this temperature increase is small due to the highly non-linear saturation pressure.

This paper demonstrates the inherent temperature regulation abilities of an evaporatively cooled fuel cell stack using a validated simulation. It is also shown how small changes in stoichiometry and pressure can reduce temperature variation across transient drive cycles.

**PEMFC model**

A lumped parameter fuel cell stack model is used to study temperature regulation of evaporative cooling, the model consists of separate sub-systems for cell voltage, hydration, mass and energy balances. The major equations and model validation are detailed in this section.

**Voltage**

Cell voltage ($V_{cell}$) is calculated using the open circuit voltage and common irreversible voltages, shown in equation (3).

$$V_{cell} = V_{OCV} - V_{act} - V_{f} - V_{trans} - V_{ohm}$$

Open circuit voltage ($V_{OCV}$) is found for a hydrogen PEMFC with a liquid product, accounting for changes in temperature and pressure. The Tafel equation is used to find activation ($V_{act}$) and fuel crossover ($V_{f}$) overvoltages. Mass transport losses ($V_{trans}$) are determined based on an empirical exponential relationship and ohmic loss ($V_{ohm}$) from the membrane hydration model. Stack voltage is the sum of cell voltage assuming all cells are identical.

**Membrane**

Electro-osmotic drag and back diffusion across the membrane are both considered. The empirical model of Springer et al. [16] is used to calculate membrane water content, diffusivity and conductivity from anode and cathode relative humidity. Resistance is obtained assuming a linear water concentration between anode and cathode.

**Mass balance**

The anode and cathode volumes are modelled as separate lumped volumes, mass of each species are found from first order differential mass balances, equations (4)–(8) show each of the cathode and anode species.

**Cathode**

$$\frac{dm_{H_2O}}{dt} = m_{H_2O_{in}} - m_{H_2O_{out}}$$

**Anode**

$$\frac{dm_{H_2}}{dt} = m_{H_2_{in}} - m_{H_2_{out}} - m_{H_2_{react}}$$

Water into the system comes in vapour form from ambient air and liquid injected into the stack for humidification and cooling. It is assumed liquid water will evaporate where possible until saturation, flooding effects are ignored. Removal of liquid water from the flow channels is modelled using the entrainment factor shown in Ref. [13] (equation (9)) where $\delta$ is a constant relating liquid mass flow rate to accumulated liquid mass and gas flow rate.

$$m_{H_2_{reoct}} = \delta m_{H_2_{in}} (m_{H_2_{out}} + m_{H_2_{out}} + m_{H_2_{out}})$$

Pressure is found as the sum of gas species partial pressures, exhaust flow is determined from the non-choked nozzle equation [17]. Pressure loss within the stack is modelled as a linear function of inlet air flow rate; at maximum flow (1A/cm² 3.0 cathode stoichiometry) it is assumed to be 15 kPa. Cathode exhaust properties are evaluated at the stack exit; all cathode pressures discussed in this study refer to stack exit pressure (after pressure loss).

An additional lumped volume for mass accumulation is included to simulate manifold filling effects between compressor and cathode, the manifold is assumed to be adiabatic.

The stack which has been modelled has a dead-ended anode which is periodically purged to remove impurities and accumulated water. Periodic anode purging in the simulation is time averaged using a low hydrogen stoichiometry (1.05 in the current work), pressure is regulated using a non-choked back pressure valve in the same method as the cathode.

**Energy balance**

The fuel cell stack is modelled as a single lumped thermal capacitance, temperature is found using a balance of energy flow into and out of the stack, equation (10).

$$mC_p \frac{dT_s}{dt} = Q_{in} - Q_{out} - Q_{elec} - Q_{heat}$$

$Q_{in}$ is the heat released by the electrochemical reaction, based on the higher heating value (HHV) of hydrogen (liquid product), $Q_{elec}$ is the electrical power produced by the stack. $Q_{in}$ and $Q_{out}$ are the heat flows into and out of the stack respectively, which are the sum of species enthalpies ($H_2$, $N_2$, $O_2$, $H_2O_2$, $H_2O$) where the first term in equation (11) accounts for the enthalpy of vapourisation of water. Exit flow is assumed to be at stack temperature, variation in material properties with temperature are accounted for using look-up tables.
\[
Q = \dot{m}_{\text{H}_2O} \Delta H_v + \sum_{j=1}^{n} \dot{m}_j C_p(T - T^c) \tag{11}
\]

\[Q_{\text{loss}}\] represents the small amount of energy lost from the stack surface through natural convection, assuming a constant heat transfer coefficient, shown in equation (12).

\[Q_{\text{loss}} = h_i A_i(T_s - T_a) \tag{12}\]

**Balance of plant**

The compressor is represented in the model using the non-isentropic compressor equation \[15\], assuming a constant efficiency of 75\%, this is used to calculate parasitic load and stack inlet conditions. It is assumed that the compressor can maintain a desired stoichiometry independent of operating pressure down to a minimum flow rate of 5 g/s (equivalent to a 2.5 stoichiometric flow rate at 60 mA/cm\(^2\) for the system in this study); transients are represented using a first order transfer function with a 0.75 s time constant.

To regulate back pressure in both the anode and cathode back pressure valves are simulated by varying the cross sectional area in the non-choked nozzle equation. A proportional integral (PI) controller is used to maintain desired pressure. Liquid water is injected into the stack at a rate sufficient to maintain a desired cathode humidity using a PI controller, in the current work the target cathode humidity is 100%.

**Vehicle model**

To simulate transient drive cycles a simple vehicle model has been used to calculate the power requirement for a velocity (V) time history. The model, which considers acceleration, aerodynamic and friction loads, is shown in equation (13).

\[P_v = V \left( m \frac{dV}{dt} + \frac{1}{2} \rho C_D A_p V^2 + m g a_t + V b_t \right) \tag{13}\]

The fuel cell is operated so that net power output matches the power demand of the vehicle, parameters for a typical passenger vehicle are used, these are shown in Table 1.

**Validation**

The model was implemented using MATLAB Simulink. The fuel cell voltage sub-model has been validated using an experimental polarization curve for a 12 kW evaporatively cooled PEMFC stack from Ref. [9]. Mean voltage difference between experimental and simulation from 0 to 150 A was 7.3 mV (<1\%). Mass and energy balance sub-models were validated using experimental data from a 15 kW commercial evaporatively cooled fuel cell stack. Across a range of operating currents, cathode stoichiometries and membrane thicknesses the mean absolute temperature difference between simulated and experimental stack temperature was 1.5 °C. These results imply the model provides a good representation of an evaporatively cooled fuel cell at a stack level, and is suitable for the study of temperature dynamics.

**Results and discussion**

The validated model was scaled and used to simulate stack temperature across a range of steady state and transient conditions, with both active and passive temperature control, key parameters used in the model are shown in Table 1.

**Steady state**

Figs. 3 and 4 show the steady state temperature variation with current for different cathode exit pressures and cathode inlet stoichiometries respectively with no active temperature control. Across the normal Ohmic loss operating region, mean stack temperature variation from 0.2 to 0.8 A/cm\(^2\) is 3.9 °C, demonstrating the inherent temperature regulation ability of an evaporatively cooled system. As discussed in section two, the increase in temperature is partially due to the decrease in voltage and partially due to pressure drop within the stack. Below 0.1 A/cm\(^2\) temperature is seen to reduce significantly, especially in Fig. 4, this is due to both the compressor minimum set-point causing high stoichiometries and reduced heat generation through increased efficiency.

![Fig. 3 – Steady state temperature at different pressures (\(\lambda_\text{c}=2.5\)).](image-url)
Unlike a liquid cooled system, pressure effects vapour flow and hence temperature, the maximum pressure at which the stack can operate is limited by the cell operating temperature. The maximum cathode exit pressure which can be used in the current simulation before stack temperature exceeds 100 °C is 1.8 bar.a, this can be increased by improving electrical efficiency, increasing stoichiometry or reducing pressure loss.

Transient

Temperature profile for a step current from 0.3 to 0.8 A/cm² at 1.2 bar.a exhaust pressure and 2.5 cathode stoichiometry is shown in Fig. 5. During the step current, heat generation within the stack increases by 330%, yet the observed temperature increase is only 3.5 °C without the need for any active temperature control or any overshoot. Current and temperature profiles for a non-hybridised Federal Test Procedure (FTP) drive cycle from a warm start are shown in Fig. 6. Temperature variation across the drive cycle is 6.0 °C, the increase compared to the steady state results are caused by frequent operation at low current densities when the vehicle is either braking or stationary, cooling the stack. Hybrid operation would decrease temperature variation by reducing time spent at low current densities. The influence of hybrid control strategy on stack temperature is beyond the scope of the current study.

**Active temperature control**

Whilst the evaporatively cooled system exhibits excellent temperature regulation across normal operating currents, the temperature variation can be further reduced by actively changing the operating pressure and stoichiometry. Improved temperature regulation allows higher operating temperatures, giving better system efficiency and heat exchanger performance [8]. In the current study pressure control is simulated using cascade proportional integral (PI) control. An initial PI controller determines the desired cathode pressure based on temperature error, the cascaded PI controller aims to achieve the requested pressure by controlling the back pressure valve position. Stoichiometry request uses a simple PI controller based on temperature error; the control diagrams.
for both methods are shown in Fig. 7. Cathode pressure request was limited in the range 1.1–2.5 bar.a, stoichiometry request was limited between 1.8 and 3.5.

The PI controllers were simulated individually to prevent an over actuated system. Results for the previous step current profile at 1.3 bar.a nominal pressure, 82 °C target temperature with and without active control are shown in Fig. 8. Both actuation methods demonstrate an improvement in temperature regulation compared to the passive case. Temperature variation is reduced to 1.4 °C and 1.85 °C for pressure and stoichiometry variation respectively with improved warm up time. The lower plot demonstrates the actuation taken by each method to achieve the target temperature separately. During high current, pressure was reduced and stoichiometry increased, with the opposite occurring at low currents. Across the step current profile this equates to a 0.9% increase in net work output for pressure regulated temperature and a 1.7% decrease for stoichiometry regulated temperature.

During automotive drive cycles active temperature control can be used to reduce temperature variation caused by low current densities, this is demonstrated using the New European Drive Cycle (NEDC) in Fig. 9. Stack temperature variation is 13.4 °C without active control, 3.7 °C with pressure based active control and 9.5 °C with stoichiometry based active control. In the passive case, temperature is lost during the urban driving section and regained during the extra-urban section. By operating at higher pressure during low currents, pressure based temperature regulation is able to maintain temperature better than the stoichiometry controlled case which has limited effect due to the compressor minimum set point. These results indicate better temperature regulation can be achieved from pressure variation compared to stoichiometry variation across a wide operating range. The higher operating pressure however increased hydrogen consumption by 1.7% across the NEDC.

**Conclusion**

A validated model for an evaporatively cooled fuel cell stack has been presented and used to study stack temperature dynamics. Simulated results show that the evaporatively cooled system has an excellent ability to self-regulate temperature within ±2 °C during normal operating conditions, including step load changes. This is comparable to existing work on liquid cooled systems but without the need for active control. However operation at low current densities such as when a vehicle is stationary reduces temperature due to the minimum compressor set-point.
By changing the operating pressure, stack temperature can be effectively regulated to a set point, maintaining temperature better during periods of low loads and increasing warm up time. Across the tests conducted variable pressure was seen to outperform variable stoichiometry in achieving desired temperature.

Further work will look at how energy storage and hybridization influence stack temperature as well as system level water balance of evaporatively cooled systems.

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Nomenclature

| Symbol | Description                                      |
|--------|--------------------------------------------------|
| $A_f$  | vehicle frontal area, m$^2$                     |
| $A_s$  | stack external surface area, m$^2$              |
| $\alpha, \beta$ | rolling resistance constants                |
| $C_d$  | vehicle drag coefficient                       |
| $C_p$  | specific heat, J/kgK                           |
| $F$    | Faraday constant, C/mol                         |
| $h$    | heat transfer coefficient, W/m$^2$K             |
| $I$    | current, A                                      |
| $M$    | molar mass, kg/mol                              |
| $m$    | mass, kg                                        |
| $n$    | number cells in stack                           |
| $P$    | pressure, Pa                                     |
| $Q$    | Thermal energy, W                               |
| $T$    | temperature, K                                  |
| $V$    | voltage, V/velocity, m/s                       |
| $x_{O_2}$ | volume fraction Oxygen in air                 |

Subscript

| Symbol | Description                                      |
|--------|--------------------------------------------------|
| reac   | electrochemical reaction                         |
| trans  | transport across membrane                        |
| s      | fuel cell stack                                  |
| a      | ambient                                          |
| v      | water vapour                                    |
| l      | liquid water                                     |

Superscript

| Symbol | Description                                      |
|--------|--------------------------------------------------|
| 0      | value at STP                                     |

Greek

| Symbol | Description                                      |
|--------|--------------------------------------------------|
| $\Delta H_v$ | enthalpy of vapourisation, J/kg     |

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