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Vapor Concentration within the PEMFC Bipolar Plate over Long Term Operation

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Abstract – One of the primary technical challenges associated with proton exchange membrane fuel cells (PEMFCs) is ensuring their durability. This study introduces an experimental methodology to evaluate the thermal and water characteristics within PEMFC during continuous operation under the New European Driving Cycle (NEDC) mode. Specifically, fifty micro relative humidity and temperature sensors (micro-RH/T sensors) are integrated into flow field plate channels to measure the distribution and characteristics of temperature and water vapor within the PEMFC. The study tracks evolution of these parameters over a 100-hour NEDC durability test, revealing notable trends. In addition, an artificial neuron network-based model (ANN-based model) is developed to analyze water transport through the membrane at both the beginning and the end of the durability test. The findings demonstrate the significant impact of temperature and water characteristics on the performance and durability of PEMFC under prolonged operational conditions. This research provides valuable insights into PEMFC operation and establishes a foundation for further advancements in PEMFC design, aiming to enhance performance and extend the lifetime of automotive PEMFCs.

***Keywords*:** PEMFCs, Durability test, In-situ RH/T measurement, ANN-based model

Nomenclature:

A	Coefficient in dewpoint temperature equation ($A = 1732.7549$)
a_w	Water activity ($a_W = RH/100$)
B	Coefficient in dewpoint temperature equation ($B = 233.426$)
b_{HL}	Bias of ANN hidden layer
b_{out}	Bias of ANN output layer
DP	Dewpoint temperature
D_λ	Water diffusivity
F	Faraday constant
j	Current density
M_{mem}	Membrane molecular mass
n_{mem}	Membrane water transport flux
n_{drag}	Water electro-osmotic drag flux
n_{diff}	Water back diffusion flux
n_d	Electro-osmotic drag coefficient
RH	Relative humidity
T	Temperature
t_m	Membrane thickness
W_{HL}	Weights of ANN hidden layer
W_{out}	Weights of ANN output layer
λ	Membrane water content
ρ_{dry}	Membrane dry density

1. Introduction

With increasing environmental concerns, proton exchange membrane fuel cells (PEMFCs) have emerged as a promising solution for incorporating hydrogen-based green energy into transportation systems. While cost reduction remains important, enhancing durability represents the most critical issue that needs to be addressed to enable the successful commercialization of PEMFCs [1,2]. Although performance degradation is inevitable during long-term operation, the rate of degradation can be significantly reduced through a thorough understanding of the underlying mechanisms of degradation and failure [3].

Extensive experimental studies have been conducted to examine the degradation of PEMFC components, with findings emphasizing the critical influence of temperature and water on the failure of internal components, including catalyst layers dissolution [4], membrane thermal degradation [5], structural deterioration of gas diffusion layer [6], and corrosion of bipolar plates [7]. These investigations have utilized a variety of analytical techniques, such as performance testing, cyclic voltammetry (CV), scanning electron microscopy (SEM), and transmission electron microscopy (TEM) to evaluate performance degradation and component deterioration. However, despite these advancements, the effects of thermal and water characteristics within the flow field on PEMFC durability testing and deterioration assessment remain underexplored.

Thermal and water management are extremely important during the operation and performance of PEMFCs. The uneven temperature distribution can result in performance penalties, local dehydration, or flooding [8]. Consequently, analyzing temperature and water distribution is vital for assessing PEMFC degradation. Recent experimental studies have employed various techniques to measure water behavior within PEMFCs, including visualization [9], X-ray [10], neutron imaging [11]. In addition, in-situ measurements of relative humidity and temperature (RH/T) and their effects on PEMFC performance have been conducted using fibre Bradd grating sensors [12], micro-RH/T sensor [13]. Nevertheless, the simultaneous observation of temperature and water characteristics during long-term operation remains a significant challenge.

On the other hand, water transport mechanism plays a critical role in PEMFC operation, influencing key processes such as proton conductivity, hydrogen dissolution, and diffusion within the membrane [14]. Springer et al. (1991) developed a model to describe water transport in membranes, defining water flux as the combined effect of electro-osmotic drag and back diffusion fluxes [15]. A fundamental parameter required for calculating water flux is the membrane's water content. Several studies have focused on modeling water content in Nafion membranes such as N117 [15], N115 [16], and N212 [17]. However, challenges remain in accurately modeling water content in the N211 membrane, a thinner and potentially more efficient alternative to the thicker membranes currently in use.

Artificial neural networks (ANNs) have demonstrated their effectiveness as a robust machine learning tool for modeling and solving complex, nonlinear physical problems [18]. Numerous studies have employed ANN-based models to address heat and mass transfer challenges, highlighting their strong predictive capacities [19-21]. These findings suggest that applying ANN to model phenomena associated with PEMFC operation is both feasible and promising.

A review of the literature reveals that the mechanisms of temperature and water transport, along with their evolution in PEMFC during long-term operation, remain incompletely understood. This study addresses this gap by presenting the results of in-situ RH/T measurements and employing an ANN-based model to investigate thermal and water characteristics in PEMFC during a 100-hour durability test. The findings contribute to a deeper understanding of fuel cell operation and degradation processes, offering valuable insights for future advancements in PEMFC technology.

2. Methods

2.1. In-situ RH/T measurement in PEMFC flow field during durability test

The authors designed and fabricated a specialized PEMFC unit optimized for measuring temperature and water characteristics by integrating SENSIRION SHT31 micro-RH/T sensors into the flow field plates [22]. Figure 1a illustrates the numerical indexing of the sensor arrangement and the method used to embed the micro-RH/T sensors within the parallel-type flow field plate. The recorded RH/T data are utilized to calculate the dewpoint temperature and the water transported through the membrane.

The PEMFC was operated under controlled conditions, including a temperature of 70 ^{o}C and an inlet reactant humidity of 70% RH. The inlet reactant flow rates were adjusted to maintain a constant stoichiometry of 2.5, and the experiments were conducted at atmospheric pressure. The NEDC was employed as the dynamic loading protocol for the

100-hour PEMFC durability test. Measurements of temperature, water distribution, and water transport within the active area were performed at both the beginning of the test (BOT) and at the end of the 100-hour test (EOT-100h).

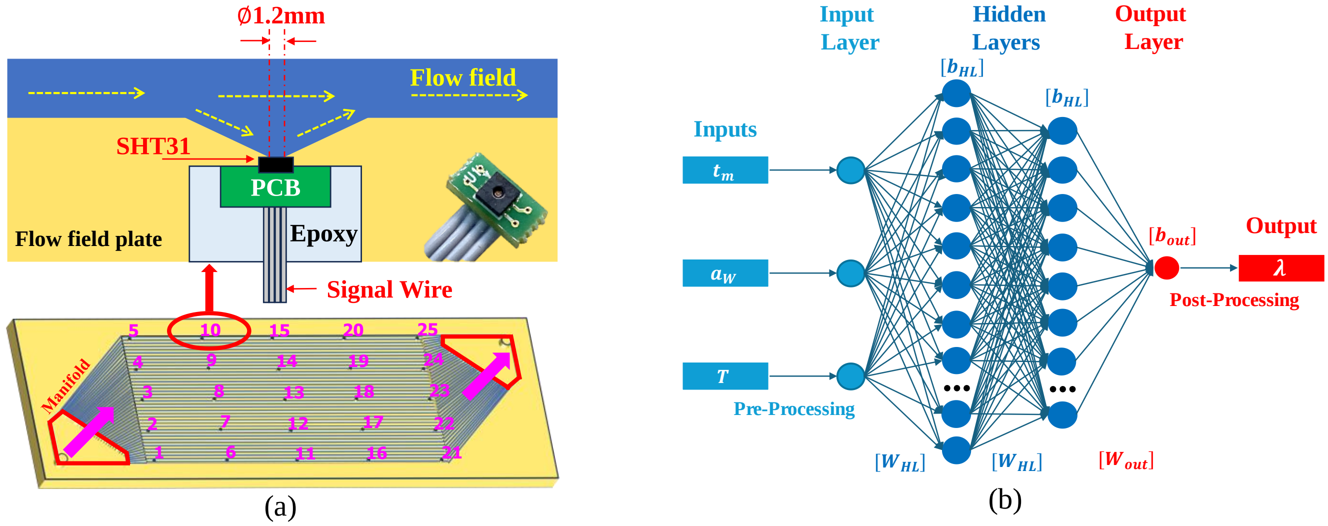

Fig. 1: Methodology of research (a) Flow field plate with micro-RH/T sensors, (b) ANN-based model

2.2. Calculation method for water transport using ANN-based model

The dewpoint temperature (DP) is calculated using RH/T values and is employed to represent the water distribution within the PEMFC.

$$DP[°\mathrm{C}] = \frac{A \times T + B(T + B) \times \log(RH/100)}{A - (T + B) \times \log(RH/100)} \tag{1}$$

where A=1732.7549, B=233.426, and T is the gas temperature in the range of 1-100 °C [22].

In a previous study, the experimental database (comprising 554 data points) on Nafion membrane's water content from the literature were gathered to develop a predictive model [23]. Our approach utilizes a data-driven method based on ANN, following the ANN structural optimization framework outline in our prior work [24]. The optimal ANN configuration resulted in a model where water content (λ) is represented as a function of membrane thickness (t_m), water activity (a_w), and temperature (T), with the relationships determined through a matrix of weights and biases (Figure 1b).

$$\lambda = \lambda(t_m, a_w, T) \tag{2}$$

The water flux transport in membrane (n_{mem}) can be described as follows [15]:

$$n_{mem} = n_{drag} - n_{diff} \tag{3}$$

The electro-osmotic drag water flux (n_{drag}) is formulated in terms of current density (j) as follows [15]:

$$n_{drag} = 2.5 \frac{j}{F} \frac{\lambda}{22} \tag{4}$$

From the Fick's law, the back diffusion water flux (n_{diff}) is a function of water content (λ):

$$n_{diff} = \frac{\rho_{dry}}{M_{mem}} D_\lambda \frac{\lambda_{ca} - \lambda_{an}}{t_m} \tag{5}$$

The water diffusivity (D_λ) is defined by [17]

$$D_\lambda = 13.3 \times 10^{-4} \times \lambda\left(161 e^{-\lambda} + 1\right) \exp\left(-\frac{2436}{T}\right) \tag{6}$$

3. Results and discussion

Figure 2 illustrates the distribution of temperature and water at both the beginning of the durability test (BOT) and the end of the 100-hour test (EOT-100h), under a current density load condition of 0.5A/cm^2. The water distribution is depicted through the dewpoint temperature distribution, which is calculated using Equation (1). The arrows in the figures indicate the direction of inlet and outlet reactant flow at each electrode for anode and cathode.

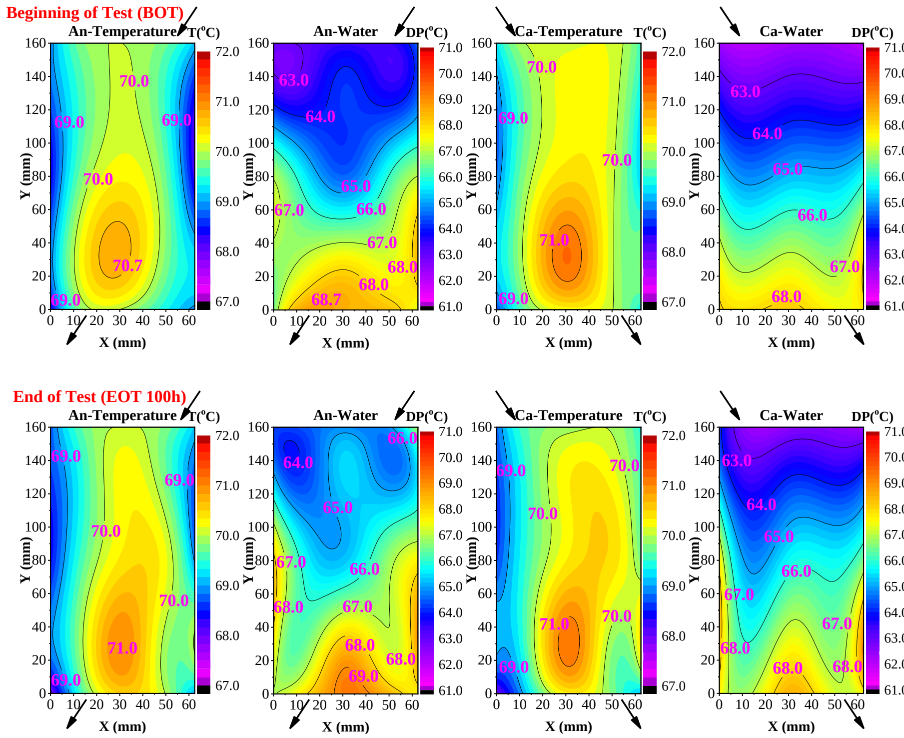

Fig. 2: Temperature and water distribution within PEMFC before and after durability test

At the BOT, the mechanisms for managing temperature and water within the PEMFC operated relatively effectively, maintaining an even temperature distribution and preventing flooding within the flow field. The high-temperature region was observed near the downstream section of the flow field. Water tended to accumulate in the downstream region due to both gravity and the gas flow within the flow field channels, resulting in a higher water concentration (higher DP value) in this area. At EOT-100h, water accumulation resulting from prolonged operation led to an increase in water concentration, causing flooding phenomena in the central downstream and lower edge regions. Additionally, a significant rise in temperature was observed at EOT-100h. These detrimental effects, including localized hot spots with elevated temperatures, expanded high-temperature zones, and the flooding from water accumulation, negatively impacted both the performance and durability of the fuel cell [25].

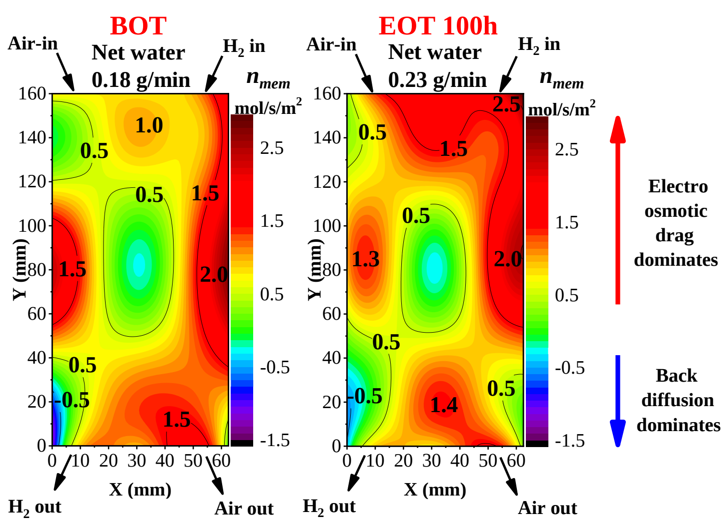

Fig. 3: Water transport flux distribution in active area of PEMFC before and after durability test

Figure 3 illustrates the distribution of water flux in the active area, comparing the results between the BOT and EOT-100h. The red regions indicate dominance of water electro-osmotic drag flux from anode to cathode, while the blue regions represent the back diffusion flux from cathode to anode. Notable changes in water transport were observed between BOT and EOT-100h, with a higher amount of water transport from anode to cathode after 100-hour durability test. This shift may reduce the water content on the anode side, leading to a decrease in membrane proton conductivity, which in turn contributes to performance degradation.

4. Conclusion

Some conclusions from this study are as follows:

- This study demonstrates the potential application of ANN to model the phenomena related to PEMFC operation.
- The evolution of temperature and water distribution within PEMFC flow field during long-term operation is analyzed, showing an increase in both average temperature and water content.
- The trend in water transport within PEMFC membrane is examined using experimental data and ANN-based model. After prolonged operation, the amount water transported from anode to cathode increases, leading to performance degradation due to the drying-out of the membrane on the anode side.

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