

Reliability Improvement in Modelling a Hollow Fiber Membrane Humidifier Using the Response Surface Method

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Abstract – Water transport through the membrane represents the performance of a membrane humidifier, which is widely employed in fuel cell applications. Precise evaluation of this process is a foundation for system simulation, contributing to cost minimization. Conducting experiments for parametric analysis of water transport is a fundamental step in measuring reliable data to establish a correlation between the humidifier performance and operating conditions. This correlation should be proposed with minimal deviation to improve the reliability of the entire system. This study presents humidifier lumped models with different definitions of water diffusion through the hollow fiber membrane. The assumption of constant diffusivity in the membrane causes an overestimation in predicting the humidifier performance, with an average deviation of 10.75%. The selected model for system simulation is based on the response surface fitting function for the number of transfer units (NTU). This function helped minimize the deviation between simulation results and experimental data on water transport in a hollow fiber membrane humidifier. The average deviation in this case is 5.68%.

Keywords: Hydrogen fuel cell; Water management; Hollow fiber membrane humidifier; Response surface method.

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1. Introduction

Proton exchange membrane fuel cells have gained significant attention as a viable power source for automotive and stationary applications due to their high-power density, zero-emission characteristics, and low operating temperatures [1], [2]. In these systems, water management is a critical process that determines the efficiency, performance, and durability of the fuel cell stack [3]. The proton conductivity of the electrolyte membrane is heavily dependent on its hydration state. Protons migrate across the membrane; thus, insufficient moisture causes a sharp decline in conductivity, leading to performance degradation and potential mechanical damage. Conversely, excessive water can lead to cathode flooding, which blocks gas diffusion and limits the transport of reactants to the catalyst surface [4].

To maintain an optimal state of hydration, the air stream is usually humidified before being supplied to the stack. Among various technologies, such as bubblers, enthalpy wheels, and direct water injections, external membrane humidifiers are preferred for automotive applications [5]. These devices are passive electrochemical reactors that selectively transfer water vapor from the humid cathode exhaust to the dry intake

air across a permeable membrane. They offer advantages such as high mass transfer efficiency, compactness, and zero parasitic power loss since they do not require moving parts or external energy. In current commercial applications, these humidifiers utilize either flat-sheet planar modules or hollow-fiber membrane bundles. The latter provides superior packing density and a larger active surface area per unit volume [6].

The performance of a membrane humidifier should be determined by experiments to measure exactly the mass transfer characteristics. From this point, model development is necessary to integrate into a fuel cell system for further investigation and design. The reliability of a humidifier model often depends on its basis in experimental data fitting. A frequently used approach is the Springer model, which utilizes empirical formulas and constants obtained through curve-fitting experimental data from Nafion 117 [7]. This model relates membrane resistance and water content. Recent studies have explored more sophisticated statistical approaches. McCarthy et al [8] utilized Response Surface Methodology (RSM) to evaluate planar humidifier performance by employing a central composite design. They demonstrated that RSM is an effective solution for predicting output metrics, such as relative humidity and water transfer rate, across a full range of operating conditions. Nguyen et al. [9] focused on establishing empirical correlations for water diffusivity in hollow-fiber membranes. They utilized a latent Effectiveness-Number of Transfer Units ($\epsilon - NTU$) approach to derive water diffusivity from experimental data measured under various temperatures, pressures, and flow rates. Their findings showed that water diffusivity in the membrane is highly sensitive to operating parameters, especially temperature, and that established correlations could predict performance in large-scale humidifiers with a mean deviation as low as 7.72%. Furthermore, Tinz et al. [1] generated a comprehensive dataset of over 1200 points to develop a Gaussian Process Regression model to improve the accuracy of water transfer and outlet temperatures prediction with high coefficients of determination ($R^2 > 0.99$).

Many fitting algorithms can be employed to generalize experimental data to create a model used for water transport prediction. In this study, humidifier models are developed based on several data fitting methods to determine a highly reliable model. Three approaches to describe the diffusion in the membrane were applied in humidifier modelling: (1) constant water diffusivity; (2) diffusivity explained by physical

nonlinear correlation; (3) diffusivity explained by RSM-based correlation. The simulation results were compared with experimental data in terms of the water transfer rate through the membrane to determine the optimal solution to humidifier modelling.

2. Model description

This study presents a lumped-mass model of a humidifier using hollow fiber membranes to evaluate the performance of a water management device in fuel cell vehicles. Figure 1 shows the schematic diagram of a humidifier using hollow fiber membranes. The transport mechanism includes convection mass transfer on both sides of the membrane and diffusion mass transfer through the membrane. The diffusion mechanism is represented by water diffusivity through the membrane, determined by experiments. This coefficient can be assumed to be consistent with varying operating conditions [10] or be a function of various input parameters [9]. In this study, different models of diffusion are established and integrated into a humidifier model to compare their reliability based on a validation with experimental results of the water transfer rate.

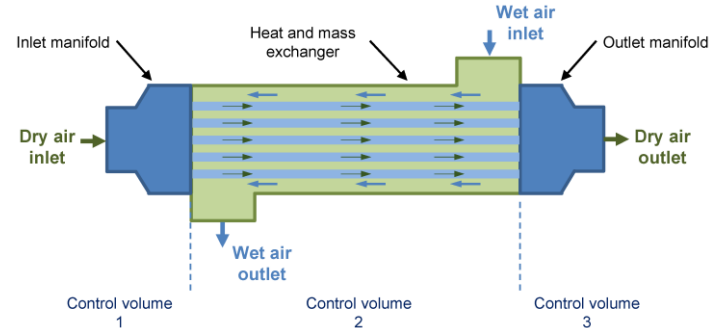


Figure 1. Diagram of a hollow fiber membrane humidifier [10]

Several assumptions are applied for modelling the hollow fiber membrane humidifier:

- Air flow behaviors are determined as the ideal gas.
- The diffusion mechanism is dominant in the membrane, and the convection mechanism is dominant on both sides.
- The dry and wet air streams are arranged by the counter direction.
- Isothermal condition is assumed for the humidifier operation.
- The air flows are radially uniform, and the concentration variation is only in the axial direction.

The overall mass transfer coefficients can be calculated based on the sum of resistance:

$$\frac{1}{k_{overall}\pi d_o L} = \frac{1}{k_i\pi d_i L} + \frac{\ln(d_o/d_i)}{2D_{vm}\pi L} + \frac{1}{k_o\pi d_o L} \quad (1)$$

where, k is convection mass transfer coefficient calculated via a relation with the Sherwood number:

$$Sh = \frac{kL}{D_h} \quad (2)$$

Empirical correlations are employed to predict the mass transfer coefficients for the tube and shell sides [3].

Tube side:

$$Sh = 1.62 \left(\frac{d_i^2 u_i}{LD_{va}} \right)^{1/3} \quad (3)$$

Shell side:

$$Sh_w = (0.3045\phi^2 - 0.3421\phi + 0.0015)Re^{0.9}Sc^{0.33} \quad (4)$$

For the membrane side, the diffusivity of water vapor is determined as follows:

Case 1: $D_{vm} = \text{constant}$;

Case 2: $D_{vm} = f(Re, Sc, T)$ based on correlation fitting;

Case 3: $NTU = f(T, p, m, RH)$ based on the response surface method.

The $\varepsilon - NTU$ approach is applied for mass transfer modelling, as the following relationship:

$$NTU = \frac{\rho_a k_{overall} A}{m_{min}} \quad (5)$$

$$\varepsilon = \frac{1 - \exp[-NTU(1 - C_{Lat})]}{1 - C_{Lat}\exp[-NTU(1 - C_{Lat})]} \quad (6)$$

According to the definition of mass transfer effectiveness:

$$\varepsilon = \frac{m_d(\omega_{do} - \omega_{di})}{m_{min}(\omega_{wi} - \omega_{di})} \quad (7)$$

The water transfer rate (\dot{m}_{tr}) through the membrane and outlet relative humidity (RH_o) of dry air can be calculated to show the humidifier's performance:

$$WTR = m_{v,do} - m_{v,di} = m_d(\omega_{do} - \omega_{di}) \quad (8)$$

$$RH_o = \frac{\omega_{do} p_t}{(0.622 + \omega_{do}) p_s} \quad (9)$$

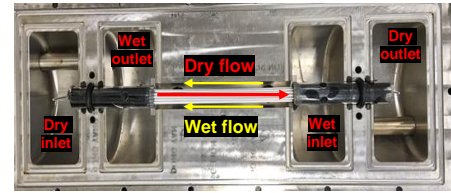
3. Experimental Analysis

3.1. Apparatus and measuring process

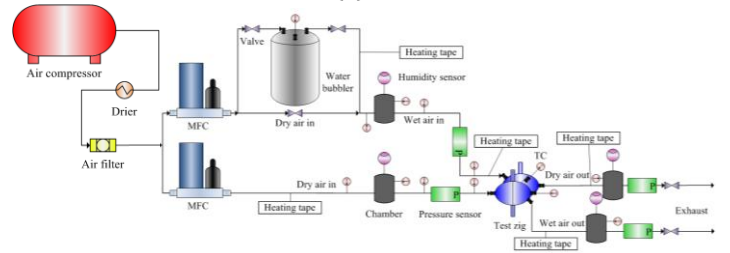
Experiments of water transport in a hollow fiber membrane humidifier were conducted to evaluate the humidification performance, creating a database for model development. A small membrane module was employed to measure the mass transfer characteristics and create an empirical correlation to further adapt to the model. Subsequently, a larger module was used for model validation. The membrane properties are shown in Table 1.

Table 1. Membrane module properties.

Parameter	Small module	Large module
	Fundamental test	Validation test
Inner diameter (mm)	0.9	0.9
Thickness (mm)	0.1	0.1
Length (mm)	110	254
The number of fibers	21	4800



(a)



(b)

Figure 2. (a) Configuration of test jig; (b) Diagram of water transport measurement

Water vapor transport is significantly influenced by vapor concentration, which is determined by the partial pressure of air-vapor mixtures. Since the temperature variation in the flow direction complicates the physical problem, experiments on vapor transport through the hollow fiber tube were conducted under isothermal conditions. Figure 2 illustrates a test jig configuration using a hollow fiber membrane module and provides a diagram of the experimental apparatus. Air is supplied by a compressor and divided into two pathways: dry air and wet air. The dry air flows directly into the dry channel (tube side), while a bubbler humidifies the wet air before it enters the wet channel of the test jig (shell side). Fluid streams primarily move

along the membranes' length, resulting in a convection effect on moisture exchange from the wet to the dry air. Additionally, the diffusion process in the membrane also contributes to mass transfer. Vapor transport characteristics were measured using sensors (T-thermocouple, pressure transmitter P126, Vaisala HTM337) to evaluate the humidifier's performance, including effectiveness, NTU, water transfer rate (WTR), and outlet relative humidity.

The fundamental experiments were conducted under conditions of temperature from 60°C to 80°C, pressure from 150 kPa to 250 kPa, flow rate from 10 slpm to 30 slpm, and inlet air humidity from 50% to 90%. Regarding the validation tests, a dataset of 20 random cases is selected to compare with the simulated results. Conditions of the validation experiment are 2250 slpm to 5050 slpm for the air flow rate, 60°C to 80°C for system temperature, 160 kPa to 230 kPa for pressure, and 70% to 95% for relative humidity.

3.2. Uncertainty analysis

Measurement uncertainty must always be performed to ensure the reliability of experimental work. The water transport measurement is affected by each measurement of conditional parameters. Based on the root-sum-squared method, the overall uncertainty of the WTR, 3.97%, can be estimated using the following equation [9]:

$$WTR = f(T, p, RH, m) \quad (10)$$

$$\Delta(WTR) = \sqrt{\left(\frac{\partial(WTR)}{\partial T} \Delta T\right)^2 + \left(\frac{\partial(WTR)}{\partial p} \Delta p\right)^2 + \left(\frac{\partial(WTR)}{\partial RH} \Delta RH\right)^2 + \left(\frac{\partial(WTR)}{\partial m} \Delta m\right)^2} \quad (11)$$

$$e = \frac{\Delta(WTR)}{WTR} \times 100\% \quad (12)$$

3.3. Response surface method

Response Surface Method (RSM) is a statistical technique used to examine and develop a mathematical model that clarifies the correlation between inputs and outputs. The objective of the response surface method is to determine the correlation between variables by accurately fitting mathematical models to experimental data. These models can assist in determining the optimal designs for input variables to achieve target outputs, improve processes, and comprehend the interactions between various components [8].

In this study, the output is NTU, which is calculated using vapor transport experimental data. This output depends on four independent variables: temperature, relative humidity, mass flow rate, and pressure. The relationship between the response function and input variables is described by Eq. 13 as follows:

$$y = \beta_0 + \sum_{i=1}^4 \beta_i x_i + \sum_{i=1}^4 \beta_{ii} x_i^2 + \sum_{i=1}^4 \sum_{j=i+1}^4 \beta_{ij} x_i x_j \quad (13)$$

Herein, y is the response, β_0 is the constant, and β_i, β_j and β_{ij} are the linear, squared, and interaction coefficients, respectively, x_i and x_j are the variables. These coefficients can be determined using MINITAB software based on experimental data.

4. Results and Discussion

4.1. Empirical correlation development for water transport

According to the previous study [3], a water diffusivity correlation was developed based on the experimental data, as follows:

$$D_{vm} = 8.25 \times 10^{-7} Re^{-0.535} Sc^{1.826} \exp\left(\frac{115.51}{T}\right) \quad (14)$$

Using the proposed correlation with R-squared value of 0.92, as shown in Figure 3, to describe water diffusion in the membrane and published correlations to describe fluid convection on the tube/shell side, a 1-D humidifier model was developed using MATLAB/Simulink platform.

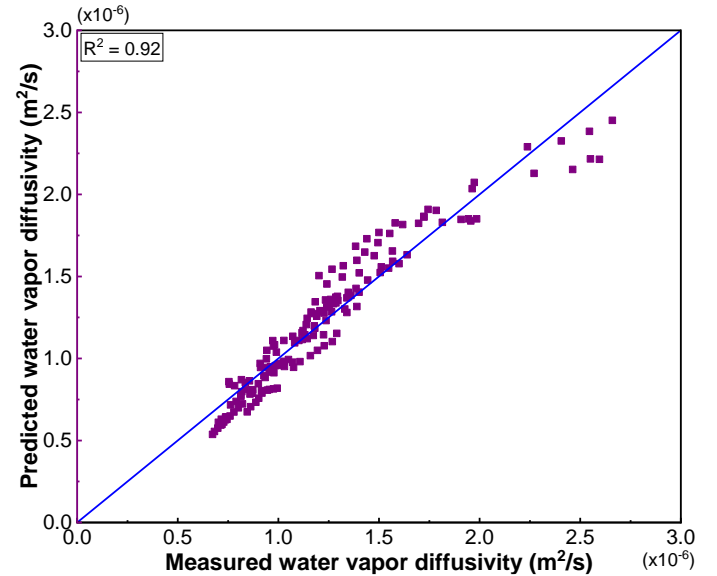


Figure 3. Comparison of water diffusivity estimated by the correlation and experimental data

4.2. Improvement of water transport prediction by response surface method

A correlation of NTU is developed using data from fundamental tests to predict the humidification performance of the membrane module. Utilizing MINITAB software, a response surface was designed to improve the reliability of the regression model. The correlation is shown with $R^2 = 0.988$:

$$NTU = 0.503 - 0.00413T - 0.042m + 0.0019 + 0.004RH + 0.000828m^2 + 0.000005p^2 + 0.000233T \times m - 0.000051T \times RH - 0.000113m \times p \quad (15)$$

The reliability of the model was determined by analyzing the residual plot, which is typically used in statistics and regression analysis. It allows for a visual assessment of whether the model assumptions are met and ensures that the patterns and variations in the data are accurately represented. Figure 4a illustrates the distribution of the residuals. The residuals, represented by the red data points, exhibit a significant alignment with the normal probability plot, as indicated by the black line. The red points are evenly distributed around the black line, suggesting that the regression model is appropriate.

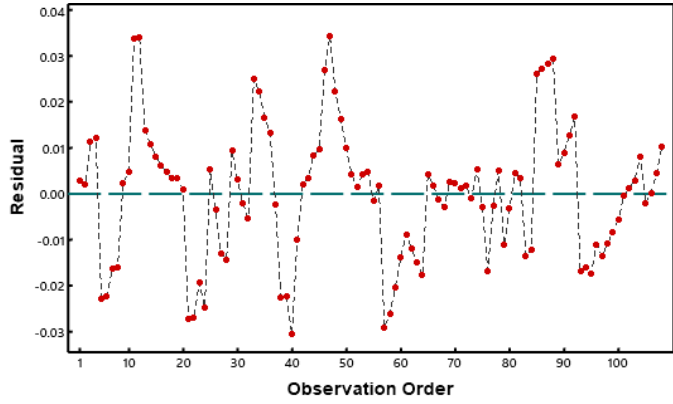
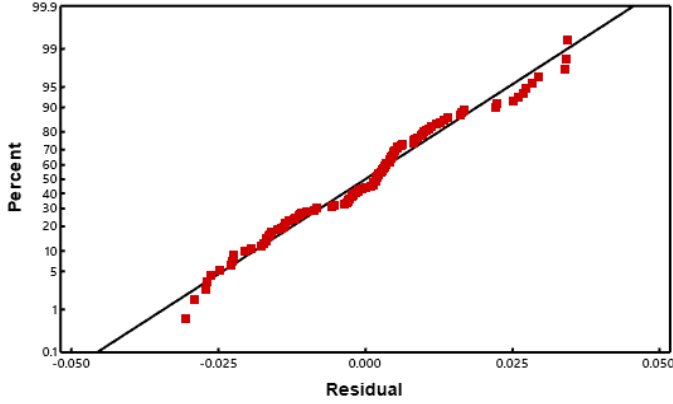


Figure 4. Probability and residual plot of the NTU

4.3. Deviation of different humidifier models

Three different humidifier models are developed based on the water diffusivity through the membrane. Based on the three mentioned methods to obtain the diffusivity data, the minimal deviation model should be selected for further modeling of a fuel cell system. The deviation between simulation and experiment in terms of the water transfer rate was calculated by Eq. 16.

$$d\% = \left| \frac{\text{Measured data} - \text{Predicted data}}{\text{Measured data}} \right| \times 100\% \quad (16)$$

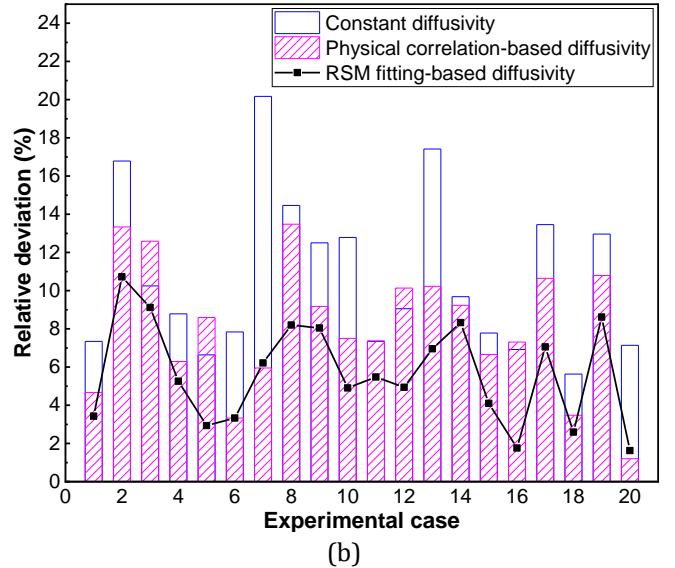
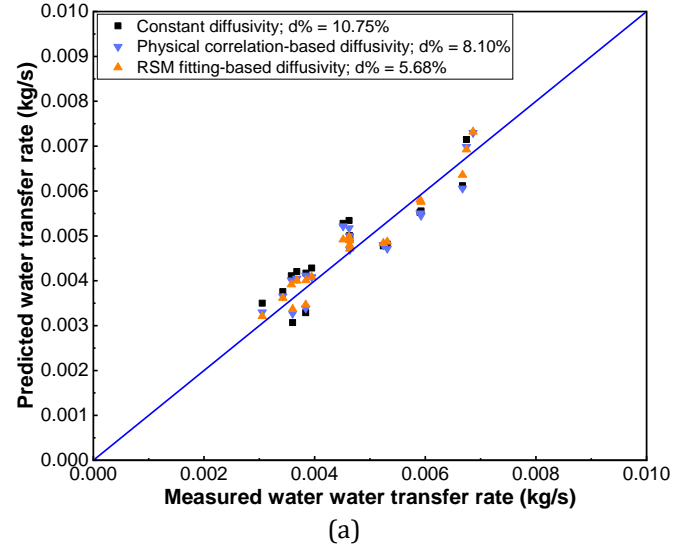


Figure 5. Validation of different humidifier models with experiments

Figure 5a illustrates the deviation in the water transfer rate estimated by three models and experimental data. It is exhibited that the response surface method significantly improves the quality of the

fitting function. The model based on RSM demonstrates the minimal average deviation, 5.68%, as the fitting algorithm accounts not only for the effect of individual input parameters but also for the interaction between those parameters.

Figure 5b presents details of the relative deviation under various conditions of water transport experiments. The assumption of constant diffusivity leads to an overestimation of the model, with the highest deviation being about 20.5%, whereas the RSM improves the model reliability in all conditions. The highest deviation is reduced to less than 11%. Consequently, the RSM-based fitting technique is suitable for developing a reliable membrane humidifier model.

5. Conclusion

Hollow fiber membrane humidifier models were developed with different assumptions for water vapor diffusion in the membrane. The results indicate that the response surface method is effective in obtaining a highly accurate empirical correlation of the NTU, as evidenced by an R-squared value of 0.988. The RSM-based fitting function contributes to minimizing the average deviation of the humidifier model compared to the experimental data, which is 5.68%. The selected model can be used to predict humidification performance in water management for hydrogen fuel cell systems.

Nomenclature

A	Membrane contact area
C_{Lat}	Latent heat capacity ratio
d_i, d_o	Membrane-tube inner/outer diameter
D_{va}	Diffusivity of vapor in air
D_{vm}	Diffusivity of vapor in membrane
k_i, k_o, k_t	Inside, outside, total mass transfer coefficient
L	Tube length
\dot{m}	Air flow rate
NTU	Number of transfer units
p_t, p_s	Total and saturation pressure
Re	Reynold number
Sc	Schmidt number
Sh	Sherwood number
u_i	Air velocity inside the membrane tubes
ω	Absolute humidity of the air
$\phi = Nd_o^2/D_o^2$	Packing fraction

ε	Latent effectiveness
ρ_a	Air density

Subscripts

d	dry
i	inlet/ inside
o	outlet/ outside
v	vapor
w	wet

Acknowledgments

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