# An Analytical Model of a Hollow Fiber Membrane Humidifier in Hydrogen Fuel Cell Systems Using Response Surface Method

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**Abstract** - Hydrogen fuel cell is a potential alternative power source for vehicles, which has a significant role in decarbonizing the future transport sector. Proton exchange membrane fuel cell is widely used because of its suitable temperature and power density. Performance and durability of stacks are important factors in the development of hydrogen fuel cell-powered vehicles. As a key subsystem, a hollow fiber membrane humidifier is investigated in this study to manage the water entering fuel cell electrodes. Parametric experiments of water transport through the membrane were done before applying the response surface method to establish a regression model based on fundamental operating parameters. The reliable regression equation of water transport performance ( $R^2 = 0.988$ ) was used to develop an analytical model of a hollow fiber membrane humidifier. The performance of the humidifier including the water transfer rate and outlet relative humidity were evaluated and scrutinized to process a better system for hydrogen vehicles. Fluid flow and transfer process were investigated under the isothermal conditions and cross-counter flow arrangements. The proposed Simulink-based model was properly validated with data from a practical humidifier, meaning that the model can be used to design humidification subsystems and further develop fuel cell systems.

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

## 1. Introduction

An increasing demand for fuel consumption over the years has accelerated a great problem of fossil fuel depletion. Therefore, it is required to develop green and clean alternative energy sources to reduce pollutant emissions and energy shortage. Proton exchange membrane fuel cells (PEMFC) were developed to be clean and sustainable-converting devices that use hydrogen as a fuel, which have a critical role in the vehicle industry because of its low operating temperature and high power density [1-2]. However, water management in fuel cell systems remains a crucial challenge in dealing with stack performance and durability. Excess water in the fuel cell can obstruct the membrane electrode assembly and decrease its active area, while a dry membrane can lead to reduced proton conductivity and potential membrane damage [3-4]. Even though the generated water from electrochemical reactants has a profound effect on the working performance of the Membrane Electrode Assembly (MEA), the water source supplied from the inlet reactant gases is also essential to investigation and development. This study focuses on an external membrane humidifier that controls the inlet water content of electrodes based on the exhaust wet stream from the cell.

Membrane humidifiers in automotive applications are commonly designed to use flat sheet and hollow fiber membrane modules. The focus of this research lies on the hollow fiber membrane module due to its various advantages, as mentioned in previous works [5,6,7]. In a hollow fiber membrane module, the gas streams flow along the fiber length on either side of the membrane wall. The flow configuration can be either in co-, counter- or cross-current flow depending on exchange performance requirements.

Numerous models have been developed for mass transfer analysis to investigate the water transport in hollow fiber membrane humidifiers under varying operating conditions and geometry design. Chen et. al [8] developed a thermodynamic model for a membrane humidifier using Nafion N117. The model was matched with experimental data in both steady and dynamic states, meaning it can be used in a fuel cell system humidification control. Yu et.al [9] and Afshari & Houreh [10] constructed a heat and mass transfer model for a planar membrane humidifier, analyzing the effect of parametric and geometric parameters on its performance. These models can also be used for a hollow fiber module with an equivalent membrane area and material. McCarthy et. al [11] used the response surface method to design an optimal membrane humidifier for a 10 kW PEMFC system. This method improved the accuracy of the empirical model with second-order interactions of the inlet factors. Li-Zhi Zhang [12,13] investigated heat and mass transfer in a hollow fiber membrane contactor with counter and cross-flow arrangements using a finite difference solution to solve the differential equations. A conclusion can be drawn from those is that the packing fraction has a dominant effect on the humidifier performance. Generally, models from the literature were developed for only one style of flow arrangement. However, a common shell-tube configuration in practice is designed with both counter and cross-flow. Therefore, the gap should be filled with an investigation.

In this work, an analytical model is proposed that accounts for mass transfer in a cross-counter humidifier at various operating conditions. A response surface method (RSM) was applied to generate a statistical correlation based on the data from water transport tests. The model was validated against experimental data measured with a real configuration of the membrane humidifier for fuel cell systems. The humidifier's performance can be evaluated using this model to further design fuel cell systems for vehicles.

#### 2. Model Description

This study focuses on the model development of a humidifier using hollow fiber membranes to evaluate the performance of a water management device in vehicles. The common shell-tube exchanger configuration includes both counter-flow and cross-flow arrangements. Therefore, this model does not assume that fluid flows with only counter arrangement for simplification as in many studies. Fig. 1 shows the analysis of flow arrangements and moisture transport mechanisms from section to section. The transport mechanism includes convection mass transfer on both sides of the membrane and diffusion mass transfer through the membrane. The Simulink-based model was developed using the  $\varepsilon$ -NTU approach to analyze mass transfer in a hollow fiber membrane module.



Fig. 1: Fluid flow and mass transfer process in a hollow fiber membrane humidifier.

For moisture exchange in Section 1 and Section 3, the cross-flow arrangement is applied:

$$NTU_{cross} = \frac{\rho_a k_{cross} A_{cross}}{(\dot{m})_{min}} \tag{1}$$

$$\varepsilon_{1(3)} = \left(\frac{1}{C_{Lat}}\right) (1 - exp\{-C_{Lat}[1 - exp(-NTU_{cross})]\})$$
<sup>(2)</sup>

In Section 2 of the humidifier, the counter flow is arranged for fluid flows, analyzed by the following relations:

$$NTU_{coun} = \frac{\rho_a k_{coun} A_{coun}}{(\dot{m})} \tag{3}$$

$$\varepsilon_2 = \frac{1 - exp[-NTU_{coun}(1 - C_{Lat})]}{1 - C_{Lat}exp[-NTU_{coun}(1 - C_{Lat})]}$$
(4)

Mass transfer coefficients can be calculated using the resistance concept and empirical correlations: 1 1  $ln(d_o/d_i)$  1

$$\frac{1}{k_{total}\pi d_o L} = \frac{1}{k_i \pi d_i L} + \frac{ln(d_o/d_i)}{2D_{vm}\pi L} + \frac{1}{k_o \pi d_o L}$$
(5)

Tube side:

$$Sh = 1.62 \left(\frac{d_i^2 u_i}{LD_{va}}\right)^{1/3}$$
 (6)

Shell side with the counter-flow:

$$Sh_w = (0.3045\phi^2 - 0.3421\phi + 0.0015)Re^{0.9}Sc^{0.33}$$
<sup>(7)</sup>

Shell side with cross flow:

$$Sh_w = Nu_w \left(\frac{Sc}{Pr}\right)^{1/3} \tag{8}$$

$$Nu = 0.3 + \frac{0.62Re^{1/2}Pr^{1/3}}{[1 + (0.4/Pr)^{2/3}]^{1/4}} \left[ 1 + \left(\frac{Re}{282000}\right)^{5/8} \right]^{4/5}$$
(9)

Moisture transfer in this study was analyzed in each section via latent effectiveness to evaluate exactly the performance of a practical membrane humidifier.

Effectiveness and relation:

$$\varepsilon = \frac{\dot{m}_d(\omega_{i4} - \omega_{i1})}{\dot{m}_{min}(\omega_{o1} - \omega_{i1})} \tag{10}$$

$$NTU = \frac{1}{C-1} \ln\left(\frac{\varepsilon - 1}{\varepsilon C - 1}\right) \tag{11}$$

Mass balance:

$$\dot{m}_{do} - \dot{m}_{di} = \dot{m}_{wi} - \dot{m}_{wo} \tag{12}$$

Deriving for three sections using Eq. 10 and Eq. 12, the unknown absolute humidity can be determined by solving the following matrix:

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

$$\dot{m}_{tr} = \dot{m}_{v,do} - \dot{m}_{v,di} = (\omega_{i4} - \omega_{i1})\dot{m}_d \tag{13}$$

$$RH_o = \frac{1}{(0.622 + \omega)p_s} \tag{14}$$

## 3. Experimental Analysis

# 3.1. Experimental setup and measuring process

The effectiveness and number of transfer units (NTU) were determined in this study via water vapor transport experiments. Fundamental tests were performed to establish the correlation between operating parameters (variable) and NTU (response) for model development. A larger module was used for the validation test, as shown in Table 1.

Donomotor	Module 1	Module 2	
Farameter	Fundamental test	Validation test	
Inner diameter (mm)	0.9	0.9	
Thickness (mm)	0.1	0.1	
Length (mm)	110	236	
Packing fraction (%)	8.3	52.7	

Table	1: I	Memb	orane	module	pro	perties.
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Fig. 2: (a) Configuration of test jig; (b) Diagram of water transport measurement

Water vapor transport is significantly affected by vapor concentration, determined by the relative humidity of gasvapor mixtures. Since the temperature variation in flow direction makes the physical problem more complicated, the experiments of vapor transport through the hollow fiber tube were done with isothermal conditions. Fig. 2 depicts a test jig configuration using a hollow fiber membrane module and a diagram of the experimental apparatus. The 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 moistens the wet air before entering the wet channel of the test jig (shell side). Fluid streams move dominantly along the membranes' length, resulting in the convection effect on the moisture exchange from the wet to the dry air. In addition, the diffusion process in the membrane also contributes to the mass transfer. Vapor transport characteristics were captured by sensors (T-thermocouple, pressure transmitter P126, Vaisala HTM337) to determine the humidifier's performance including effectiveness, NTU, water transfer rate (WTR), and outlet relative humidity.

### 3.2. Uncertainty analysis

Measuring fluid flow characteristics, including temperature, pressure, relative humidity and flow rate, is crucial in determining the corresponding WTR. However, the accuracy of these measurements can be affected by various factors, such as the use of measuring devices and experimental fluctuations in the conditions under which the measurements are taken. To address these uncertainties, the idea of uncertainty propagation is utilized, as exemplified in Eqs. (15), (16),

(17). Through this analysis, the potential impact of such uncertainties on the overall accuracy of the WTR can be assessed with 3.97% [6]:

$$\dot{m}_{tr} = f(T, p, RH, m) \tag{15}$$

$$\Delta(\dot{m}_{tr}) = \sqrt{\left(\frac{\partial(\dot{m}_{tr})}{\partial T}\Delta T\right)^2 + \left(\frac{\partial(\dot{m}_{tr})}{\partial p}\Delta p\right)^2 + \left(\frac{\partial(\dot{m}_{tr})}{\partial RH}\Delta RH\right)^2 + \left(\frac{\partial(\dot{m}_{tr})}{\partial m}\Delta m\right)^2}$$
(16)

$$e = \frac{\Delta(\dot{m}_{tr})}{\dot{m}_{tr}} \times 100\% \tag{17}$$

#### 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 several inputs (known as factor or independent variable) and one or more outputs (known as responses or dependent variable). 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 most effective designs for input variables to accomplish target outputs, improve processes, and comprehend the interactions between various components [14].

In this study, the output variable is NTU which was calculated using vapor transport experimental data. This output depends on four input variables: temperature, relative humidity, mass flow rate, and pressure. The relationship between the response function and input variables is described by Eq. 18 as the following:

$$y = \beta_0 + \sum_{i=1}^4 \beta_i x_i + \sum_{i=1}^4 \beta_{ii} x_i^2 \sum_{i=1}^4 \sum_{j=i+1}^4 \beta_{ij} x_i x_j$$
(18)

Here, y is the response,  $\beta_0$  is the constant, and  $\beta_i$ ,  $\beta_j$ , and  $\beta_{ij}$  are the linear, squared, and interaction coefficients, respectively,  $x_i$  and  $x_j$  are the variables. Based on the experimental data, fitting values can be determined using the MINITAB software.

## 4. Results and Discussion

NTU =

#### 4.1. Response surface method analysis

This research used model reduction to simplify the model and improve the accuracy of the predictions. The model is improved by removing the negligible impact of some statistically insignificant variables, resulting in a more optimized and higher-performing model. The final model with  $R^2 = 0.988$  is:

$$0.503 - 0.00413T - 0.042m + 0.00197p + 0.004RH + 0.000828m^{2} + 0.000005p^{2} + 0.000233T \times m - 0.000051T \times RH - 0.000113m \times p$$
<sup>(19)</sup>

The reliability of the model was determined by analyzing the residual plot. A residual plot is a graphical technique frequently utilized in statistics and regression analysis. It enables the visual analysis of whether the model assumptions are achieved and ensures that the patterns and variations of the data are accurately represented. Fig. 3a shows the distribution of the residuals. The residuals, shown as the red data points, have a significant relationship with the normal probability plot, as demonstrated by the black line. The red points are evenly dispersed around the black line. This indicates that the regression model is suitable, demonstrating a linear distribution without any indications of non-normality or unknown variables.



#### 4.2. Model Validation

The 1-D model was validated with the experimental data of water transfer rate through a 4800-tube module (52.7% of packing fraction), shown in Table 2. The deviation between the simulation and experiment was calculated by Eq. 20, reaching the highest value at 12.81%.

$$d\% = \left|\frac{Measured \, data - Simulated \, data}{Measured \, data}\right| \times 100\% \tag{20}$$

Flow (kg/h)	T (°C)	P (kPa)	RH (%)	Measured $\dot{m}_{tr}$ (kg/s)	Predicted $\dot{m}_{tr}$ (kg/s)	d%
168.1	59.9	144.5	80.09	0.00119	0.00135	11.53
168.1	79.95	145	78.74	0.00302	0.00325	7.07
293.6	70.2	223.6	88.15	0.00207	0.00229	9.41
294	69.94	225.7	70.18	0.00151	0.00173	12.81
360	80.6	224.5	87.4	0.00346	0.00355	2.58
361.6	70.09	225.3	78.33	0.00188	0.00200	6.09

Table 2: Comparison of simulation and experiment.

#### 4.3. Humidifier performance evaluation

In fuel cell systems, a membrane humidifier is designed to control the inlet humidity of the cathode air. This humidity comes from the outlet of the humidifier, affected by operating parameters and also the water transfer rate through the membrane. The sensitivity analysis can be conducted using the validated model to evaluate the effect of various parameters on the system's performance. The water transfer rate and outlet relative humidity were predicted as shown in Fig. 4 and Fig. 5. In Fig. 4, the water transfer rate and outlet relative humidity of the dry air are obtained with variations in operating temperature and pressure. While the WTR exponentially increases with temperature, the outlet relative humidity shows a negligible decline from 60°C to 80°C. The temperature rise is a cause of concentration increase for the wet air, but does not affect the dry air because this stream does not include water vapor. When pressure increases from 150 kPa to 250 kPa, there is an opposite trend for the performance merits. Specifically, the WTR decreases following the tendency of the air absolute humidity as pressure increases. However, the outlet relative humidity is proportional to the operating pressure as a relationship from Eq. 14. Vapor transports due to the concentration gradient between two streams, significantly depending on the inlet relative humidity.

The dry air always includes very small moisture, so the higher inlet relative humidity of the wet air raises the WTR as shown in Fig. 5. Flow rate is an important factor that needs to be controlled frequently in a fuel cell system. In this case, the WTR at the flow rate of 0.1 kg/s is higher than at 0.02 kg/s even though there is a slight reduction after a plateau from 0.06 kg/s to 0.08 kg/s. The decrease in WTR at a very high air flow rate is caused by the less resident time due to the higher air velocity. On the other hand, the outlet relative humidity significantly reduces with increasing the air flow rate. The reason is that the increase of water vapor transport in the membrane is much less than in the dry air component in the inlet streams (a mixture of dry air and water vapor).

### 4. Conclusion

A Simulink-based analytical model was developed to scrutinize the water transport performance of a cross-counter hollow fiber membrane humidifier used for a fuel cell's water management system. The model was started from an empirical NTU correlation established by a statistical response surface method with  $R^2$ =0.988. The latent effectiveness was also evaluated for each section of the humidifier to improve the reliability of the prediction model. The model was validated with experimental results of the water transfer rate, showing a deviation of less than 13%. The verified model can be used to predict the humidification performance in the water management system, and then improve the humidity control strategy to deal with the degradation process in hydrogen fuel cell systems.



# Nomenclature

А	Membrane contact area	Sc	Schmidt number
$C_{Lat}$	Latent heat capacity ratio	Sh	Sherwood number
$d_i, d_o$	Membrane-tube inner/outer diameter	$u_i$	Air velocity inside the membrane tubes
$D_o$	Shell diameter	$\omega_i, \omega_o$	Absolute humidity of dry-side air and
			wet-side air
$D_{va}$	Diffusivity of vapor in air	$\phi = N d_o^2 / D_o^2$	packing fraction

Diffusivity of vapor in membrane	Е	Latent effectiveness
Inside, outside, total mass transfer coefficient	$ ho_a$	Air density
Tube length	Subscripts	
Dry air flow rate	d	dry
The number of tubes	i	inlet/ inside
Nusselt number	0	outlet/ outside
Numbers of transfer unit	v	vapor
Total and saturation pressure	W	wet
Prandtl number		
Reynold number		
	Diffusivity of vapor in membrane Inside, outside, total mass transfer coefficient Tube length Dry air flow rate The number of tubes Nusselt number Numbers of transfer unit Total and saturation pressure Prandtl number Reynold number	Diffusivity of vapor in membrane $\varepsilon$ Inside, outside, total mass transfer coefficient $\rho_a$ Tube lengthSubscriptsDry air flow rate $d$ The number of tubes $i$ Nusselt number $o$ Numbers of transfer unit $v$ Total and saturation pressure $w$ Prandtl number $e$ Reynold number $e$

## Acknowledgments

This work was supported by the Technology Innovation Program (20015756) and (20024961) funded By the Ministry of Trade, Industry & Energy (MOTIE, Korea).

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