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Effectiveness correlations for heat and mass transfer in membrane humidifiers

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ABSTRACT

The latent effectiveness and the latent number of transfer units (NTUs) for mass transfer in membrane humidity exchangers were applied to proton exchange membrane fuel cell (PEMFC) membrane humidifiers. We report on two limitations that cause deviations in the theoretical outlet conditions reported previously: (1) using a constant enthalpy of vaporization derived from the reference temperature in the Clausius–Clapeyron equation; and (2) simplifying the relationship between relative humidity and absolute humidity as linear. These limitations are alleviated by using an effective mass transfer coefficient U_{eff}. The constitutive equations are solved iteratively to find the flux of water through the membrane. The new procedure was applied to three types of membrane and compared to the curves of ε_L and NTU_L found using Zhang and Niu's method, which is normally applied to energy recovery ventilators (ERVs).

1. Introduction

While a PEM fuel cell may be operated with dry streams of air and hydrogen, Rajalakshmi et al. [1], among other researchers [2–4], have shown that the fuel cell power output increases if the reactant streams are humidified. Fig. 1 illustrates a typical implementation at the cathodes: dry air is pumped from a compressor or blower. As the dry stream passes over the membrane heat and mass are transferred from the wet stream at the fuel cell cathode exhaust.

The effectiveness-number of transfer units (ε-NTU) method is well known in heat exchanger design for determining the properties of the unknown outlet fluid streams, or for setting the geometrical and flow parameters to achieve the required composition at the outlets [5,6]. Heat transfer and mass transfer of water are coupled in an enthalpy exchanger for attaining the outlet conditions.

In this paper, the formulations by Zhang and Niu of latent effectiveness ε_L and number of transfer units for moisture transfer NTU_L [7] were extended for use in a membrane heat and humidity plate-and-frame exchanger. The effect of extended conditions, such as elevated temperatures, used in operating fuel cells was evaluated for the mathematical model used in energy recovery ventilator (ERV) systems. Some of the simplifications and assumptions made during the mathematical derivation by Zhang and Niu are analyzed for the situation in proton exchange membrane fuel cell (PEMFC) membrane heat and humidity exchangers. The results of an alternative approach were compared with results using the method proposed by Zhang and Niu.

2. Current latent effectiveness derivations

Niu and Zhang derived a latent effectiveness and number of transfer units (NTU_L) which closely resembles the sensible heat effectiveness and number of thermal units (NTUs) method commonly used in heat exchanger design. They show that the deduction of effectiveness correlations for moisture is of the same form as sensible effectiveness [7]. Heat and humidity exchangers, such as energy (or enthalpy) recovery ventilators (ERVs) commonly have their effectiveness measured with both sensible energy transfer and latent energy transfer. The same effectiveness measures can be applied to humidifiers used in fuel cell applications due to their similar configurations and operating principles.

The latent effectiveness ε_L can be defined as

$$\varepsilon_L = \frac{(\dot{m}c_p)_d(\omega_{di} - \omega_{do})}{(\dot{m}c_p)_{min}(\omega_{di} - \omega_{wi})} \quad (1)$$

The absolute humidity, ω, is used for latent transfer, where dry-bulb temperature is used in the form corresponding to sensible heat transfer. The outlet condition can then be determined by rearranging Eq. (1)

$$\omega_{do} = \omega_{di} - \varepsilon_L \frac{(\dot{m}c_p)_{min}}{(\dot{m}c_p)_d}(\omega_{di} - \omega_{wi}) \quad (2)$$

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Nomenclature

A	membrane surface area (m^2)
B	width of humidifier (m)
C	constant parameter for sorption curve equation
c_p	specific heat capacity at constant pressure (J kg^{-1} K^{-1})
d	channel depth (m)
D_{wm}	diffusivity of water in membrane (kg m^{-1} s^{-1})
h_M	convective mass transfer coefficient, or conductance (kg m^{-2} s^{-1})
Δh_{vap}	heat of vaporization (J kg^{-1})
J	water flux (kg s^{-1} m^{-2})
k	thermal conductivity (W m^{-1} K^{-1})
l	length of channel (m)
M	number of plates (levels) in humidifier
$\dot{m}$	mass flow rate (kg s^{-1})
n	number of channels in humidifier plate
P	pressure (Pa)
Q	volumetric flow rate (STP m^3/s)
R	universal gas constant (J kg^{-1} K^{-1})
R_L	ratio of mass capacity
T	temperature (K)
t_{mem}	membrane thickness (m)
U_L	overall mass transfer coefficient (kg m^{-2} s^{-1})
U_{eff}	effective mass transfer coefficient (kg m^{-2} s^{-1})
w	width of channel (m)

Greek symbols

γ	moisture diffusive resistance (m^2 s kg^{-1})
ε	effectiveness [0,1]
θ	water uptake (kg H_2O/kg dry membrane)
θ_{max}	maximum water uptake capacity (kg H_2O/kg dry membrane)
ϕ	relative humidity
ω	absolute humidity (humidity ratio) (kg H_2O/kg dry air)

Subscripts

air	air species
d	referring to the dry (or sweep) side
di	dry-side channel inlet
do	dry-side channel outlet
H_2O	water
L	latent or moisture
mem, m	membrane
min	minimum
ref	reference state
sat	value at saturation
v	vapor
w	referring to the wet (or feed) side
wi	wet-side channel inlet
wo	wet-side channel outlet

Analogous to the expression for number of thermal units used for heat transfer in heat exchangers, a total number of transfer units for latent heat with overall mass transfer coefficient U_L is defined as

$$\mathrm{NTU_L} = \frac{AU_L}{\dot{m}_{min}} \tag{3}$$

for the total area of transfer A being equal on both sides. As is done for sensible heat, the latent effectiveness can be determined as a function of $\mathrm{NTU_L}$ and another dimensionless parameter, $R_L = \dot{m}_{min}/\dot{m}_{max}$. For unmixed cross-flows considered in the present work [6]:

$$\varepsilon_L = 1 - \exp\left[\frac{\exp\left(-\mathrm{NTU_L^{0.78}} R_L\right) - 1}{\mathrm{NTU_L^{-0.22}}} R_L\right] \tag{4}$$

Other effectiveness correlations are used for different exchanger configurations [5,6]. The latent effectiveness can therefore be substituted into Eq. (2) to determine the outlet moisture content. The total moisture transfer conductance U_L has been calculated by Niu and Zhang.

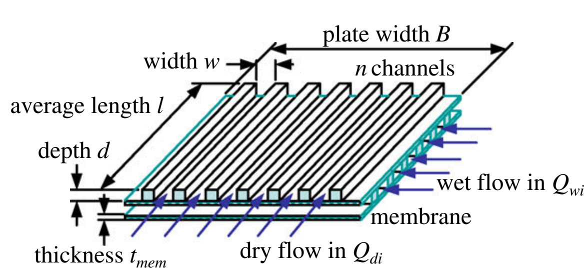

Fig. 1. Schematic of layered humidifier plates in cross-flow arrangement.

Water flux through the membrane at steady state was modeled with Fick's first law and incorporating membrane water uptake characteristics:

$$\dot{m}_{H_2O} = \frac{D_{wm}}{t_{mem}} \frac{\partial \theta}{\partial \phi}\bigg|_{mw} (\phi_{mw} - \phi_{md}) \tag{5}$$

To obtain the overall mass transfer coefficient U_L, the relative humidities must be changed into the driving force of absolute humidity ω, from a linear relation between the two parameters. Substituting the Clausius–Clapeyron equation into the relationship between relative humidity and absolute humidity based on vapor partial pressure, Zhang and Niu (from Simonson and Besant [8]) arrive at the following relation after substituting for the pressure at standard atmosphere:

$$\frac{\phi}{\omega} = \frac{e^{5294/T}}{10^6} - 1.61\phi \tag{6}$$

The second term on the right-hand side is ignored in order to simplify the equation to a linear relationship, assumed to have an effect of less than 5%

$$\phi = \frac{e^{5294/T}}{10^6} \omega \tag{7}$$

Therefore, Eq. (5) can now be written in terms of the driving force of absolute humidity

$$\dot{m}_{H_2O} = \frac{D_{wm}}{t_{mem}} \frac{\partial \theta}{\partial \phi}\bigg|_{mw} \frac{e^{5294/T}}{10^6} (\omega_{mw} - \omega_{md}) \tag{8}$$

Some algebraic manipulations lead to the water transfer in terms of the difference in absolute humidity as the driving force [9]:

$$\dot{m}_{H_2O} = \left(\frac{1}{h_{Mw}} + \gamma_m + \frac{1}{h_{Md}}\right)^{-1} (\omega_w - \omega_d) \tag{9}$$

Therefore, the overall mass transfer coefficient U_L to be used in Eq. (3) has been found as

$$U_L = \left(\frac{1}{h_{Mw}} + \gamma_m + \frac{1}{h_{Md}}\right)^{-1} \quad (10)$$

The first and third terms on the right side are the convective mass transfer resistances as produced with the heat-mass transfer analogy, while the middle term, the moisture diffusive resistance in the membrane, is analogous to the conduction resistance in heat transfer:

$$\gamma_m = \frac{t_{mem}}{D_{wm}\frac{\partial\theta}{\partial\phi}\Big|_{mw}\frac{e^{5294/T}}{10^6}} \quad (11)$$

3. Theory: limitations of ε-NTU method for heat and mass transfer

This section discusses the limitations of the ε-NTU method for heat and mass transfer in membrane humidity exchangers as proposed by Zhang in [7], specifically in the context of applying the same model to PEMFC heat and humidity exchangers. Two key observations can be made regarding the derivations that call into question the validity of the ε-NTU method being applied to fuel cell humidifiers:

1. The Clausius–Clapeyron reference temperature (used in the value for constant enthalpy of vaporization) is a constant and may be far from the actual operating temperature.
2. At higher temperatures, the absolute humidity calculated without the 1.61ϕ term in Eq. (6) will diverge substantially due to the non-linear nature of the saturated water vapor pressure curve.

3.1. Use of the Clausius–Clapeyron saturation vapor pressure equation

The Clausius–Clapeyron equation is a theoretical expression for the saturation vapor pressure of most liquids [10]:

$$P_{sat} = P_{ref}\exp\left(\frac{\Delta H_{vap}}{RT_{ref}}\right)\exp\left(-\frac{\Delta H_{vap}}{RT}\right) \quad (12)$$

The form of Clausius–Clapeyron equation used by Zhang (after Simonson–Besant [8]) employs a reference state of 3007 Pa and 297.3 K (24.1 °C) and assumes a constant heat of vaporization at that state:

$$P_{sat} = 1.629 \times 10^{11}\,e^{-5294/T} \quad (13)$$

A result of assuming a constant heat of vaporization is that the error in the equation increases with larger deviations from the reference conditions. Fig. 2 shows the percent deviation of Eq. (13) compared to the Hyland–Wexler equation. The well-known Goff–Gratch equation is also shown for a third reference correlation.

From a survey of fuel cell system manufacturers' requirements for backup and portable power applications, a reference temperature of 45 °C is more in line with the operating conditions experienced in PEM fuel cell humidification, and the corresponding heat of vaporization results in a saturation vapor pressure equation in the Clausius–Clapeyron form of:

$$P_{sat} = 1.163 \times 10^{11}\,e^{-5189/T} \quad (14)$$

The Goff–Gratch and Hyland–Wexler equations show very little deviation from each other and are taken to be most accurate [11], where Goff–Gratch is generally considered the reference equation. The percent deviation of the Clausius–Clapeyron equation used by Zhang, Eq. (13), is within 2% for most atmospheric temperatures, but increases rapidly at either higher or lower temperatures. The Clausius–Clapeyron equation derived for fuel cell applications, Eq. (14), has also been plotted, and it can be seen that the deviation is within 2% for most fuel cell applications from 21 to 80 °C.

3.2. Correlation between absolute humidity and relative humidity

From the definitions of relative humidity and absolute humidity, the following relation can be derived:

$$\frac{\phi}{\omega} = \frac{P}{0.622P_{sat}} - \frac{\phi}{0.622} \quad (15)$$

The numeric constant 0.622 is the molecular weight ratio of water to dry air composition. Substituting in Eq. (13) for a standard atmospheric pressure of 101,325 Pa this time, Zhang (after Simonson and Besant [8]) arrived at:

$$\frac{\phi}{\omega} = \frac{e^{5294/T}}{10^6} - \frac{\phi}{0.622} \quad (16)$$

Zhang (and Simonson and Besant) then neglected the second term on the right-hand side on the grounds that it is generally less than 5% of the first term on the right-hand side. This approximation can be made if the vapor pressure is much less than the air pressure, and can be arrived at by starting with a simplified version of the definition of humidity ratio

$$\omega = 0.622\frac{P_v}{P - P_v} \approx 0.622\frac{P_v}{P}, \quad P_v \ll P_{air} \quad (17)$$

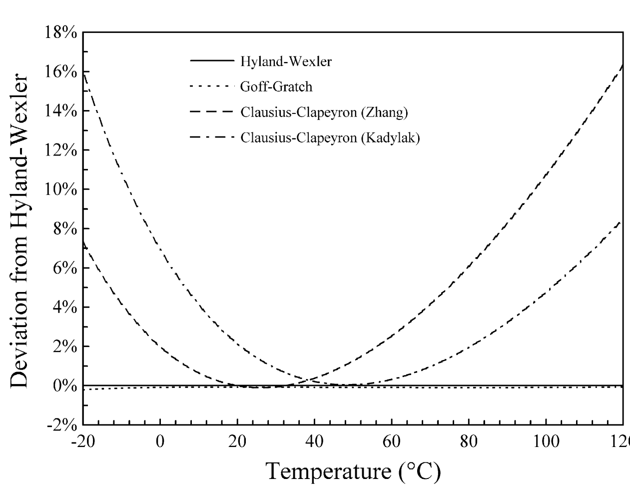

Fig. 2. Comparison of saturated vapor pressure from four different equations.

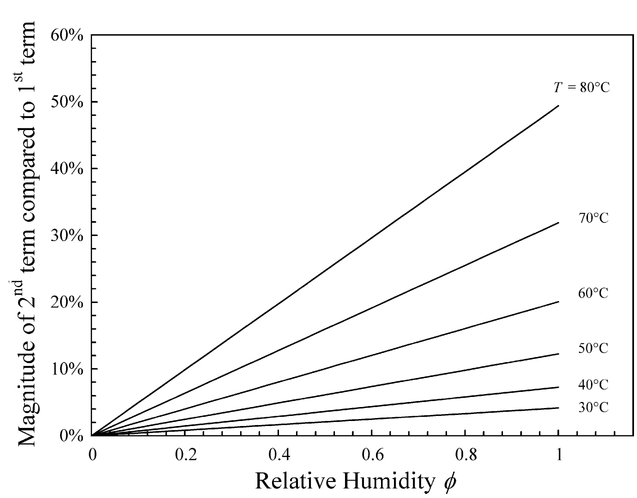

Fig. 3. Magnitude of second term compared to first on right-hand side of Eq. (16).

While this is a reasonable assumption at atmospheric pressures ($\sim$1 atm) and temperatures (20–40 °C), it is not the case at the higher temperatures that are found in fuel cell humidifiers (see Fig. 3). With errors up to 20% at 60 °C and 100% relative humidity, the second term on the right-hand side of Eq. (16) cannot be neglected.

4. Model

This section discusses recommendations to address the limitations presented above and to propose a solution for application in PEMFC membrane heat and humidity exchangers. Firstly, the Clausius–Clapeyron equation, Eq. (12), can be left as is. However, the enthalpy of vaporization and reference states shall be calculated at the operating conditions, and substituted directly into Eq. (15), as well as keeping the pressure P in Eq. (15) a variable parameter

$$\frac{\phi}{\omega} = \frac{Pe^{\Delta H_{\mathrm{vap}}/RT}}{0.622 P_{\mathrm{ref}}\, e^{\Delta H_{\mathrm{vap}}/RT_{\mathrm{ref}}}} - \frac{\phi}{0.622} \tag{18}$$

The use of the Clausius–Clapeyron equation and neglecting the $\phi/0.622$ term in Eq. (16) allow Zhang to provide a simplified expression for overall moisture transfer resistance across the membrane analogous to the overall heat transfer coefficient. Without these assumptions, the problem cannot be simplified as readily. The proposed solution is to maintain the $\phi/0.622$ term, thereby making the solution iterative. One has the following system of three flux equations with five unknowns (J, ω_{mw}, θ_{mw}, ω_{md}, and θ_{md}):

$$\begin{aligned} J &= h_{\mathrm{Mw}}(\omega_{\mathrm{w}} - \omega_{\mathrm{mw}}) \\ J &= \frac{D_{\mathrm{wm}}}{t_{\mathrm{mem}}}(\theta_{\mathrm{mw}} - \theta_{\mathrm{md}} t) \\ J &= h_{\mathrm{Md}}(\omega_{\mathrm{md}} - \omega_{\mathrm{d}}) \end{aligned} \tag{19}$$

Introducing two more unknowns (ϕ_{md} and ϕ_{mw}) and four equations of relation close the problem:

$$\begin{aligned} \frac{\phi_{\mathrm{mw}}}{\omega_{\mathrm{mw}}} &= \frac{P}{0.622 P_{\mathrm{sat,w}}} - \frac{\phi_{\mathrm{mw}}}{0.622} \\ \frac{\phi_{\mathrm{md}}}{\omega_{\mathrm{md}}} &= \frac{P}{0.622 P_{\mathrm{sat,d}}} - \frac{\phi_{\mathrm{md}}}{0.622} \end{aligned} \tag{20}$$

and the two sorption curves for the membrane under consideration, relating water uptake θ to relative humidity at the membrane interface, can be represented parametrically:

$$\begin{aligned} \theta_{\mathrm{mw}} &= \frac{\theta_{\max}}{1 - C + C/\phi_{\mathrm{mw}}} \\ \theta_{\mathrm{md}} &= \frac{\theta_{\max}}{1 - C + C/\phi_{\mathrm{md}}} \end{aligned} \tag{21}$$

where C is a variable denoting the type of membrane being used affecting the shape of the sorption curve. A value of $C = 1$ denotes a linear membrane, usually employing a silica gel desiccant. A Type-I membrane such as a molecular sieve has a value of $C < 1$, and Type-III membranes such as those containing polymer desiccants have a value of $C > 1$ [9].

This provides a set of seven equations, two of which are non-linear. Since the problem remained non-linear an iterative approach was required. The proposed solution further requires determining an effective overall mass transfer coefficient U_{eff}

$$J = U_{\mathrm{eff}}(\omega_{\mathrm{w}} - \omega_{\mathrm{d}}) \tag{22}$$

instead of the overall mass transfer coefficient U_{L} found through mass transfer resistances in Eq. (10).

The method of determining the humidifier outputs of absolute humidity is as follows:

- Solve the seven non-linear equations (19)–(21) simultaneously to find the flux J.
- Find the effective mass transfer coefficient U_{eff} using Eq. (22).
- Use U_{eff} in place of U_{L} in the equation used to find $\mathrm{NTU_L}$, Eq. (3).
- The latent effectiveness is now found from the correlation of Eq. (4) or similar.
- The outputs can now be found from Eq. (2).

An outside iterative loop involving a first estimate for the outlet values of absolute humidity, using subsequent approximations, must be implemented along with the inlet values of absolute humidity to obtain values to use for the absolute humidities in Eq. (22).

5. Results

The solution proposed in the previous section was implemented and compared to the one given by Zhang and Niu. The inputs used for this comparison, which were arrived at by considering typical PEM fuel cell conditions but also closely following the parameters used by Zhang and Niu in previous work, are summarized in Table 1.

Fig. 4 compares the proposed method to that used by Zhang and Niu of $\mathrm{NTU_L}$ when varying the dry inlet relative humidities from 0% to 90% for three types of membrane. While the new proposed method follows similar trends to that of Zhang and Niu for the corresponding type of membrane, the values in $\mathrm{NTU_L}$ vary considerably.

The effect that a change in $\mathrm{NTU_L}$ has on the latent effectiveness is portrayed graphically in Fig. 5. As expected, the curves of effectiveness follow the same shapes as those for $\mathrm{NTU_L}$ for each membrane type. The deviations between the two methods employed are also of similar magnitude in the ε_{L} curves as in the $\mathrm{NTU_L}$ curves. According to Eq. (2) the difference in effectiveness between the two methods will also determine how severely the simplifications made by Zhang and Niu affect the outputs, namely the outlet absolute humidity when compared to the inlet absolute humidity. Therefore, for a linear membrane operating with a 90% relative humidity at the dry inlet, the error caused by the simplifications at the 70 °C isothermal case investigated would translate to a 40% over-prediction in outlet absolute humidity as compared to the inlet absolute humidity.

When dealing with PEMFC humidification, the dry inlet stream will generally be supplied by a compressor or blower very dry at a dew point of −40 °C or less, or nearly zero relative humidity. Thus,

Table 1
Summary of parameters used in humidifier model comparisons

Configurations	Cross-flow
l	500 mm
B	500 mm
d	5 mm
M	30 layers
D_{wm}	2.16×10^{-8} kg/m s
$\dot{m}$	0.05 kg/s
Q	0.039 m^3/s
ϕ_{di}	0–0.9 RH
ϕ_{wi}	1 RH
w	5 mm
n	83 channels
Membrane	PVC
t_{mem}	0.02 mm
k_{mem}	0.18 W/m K
$\theta_{\max}$	0.23 kg/kg
T_{di}	343.15 K
P_{di}	101,325 Pa
T_{wi}	343.15 K
P_{wi}	101,325 Pa

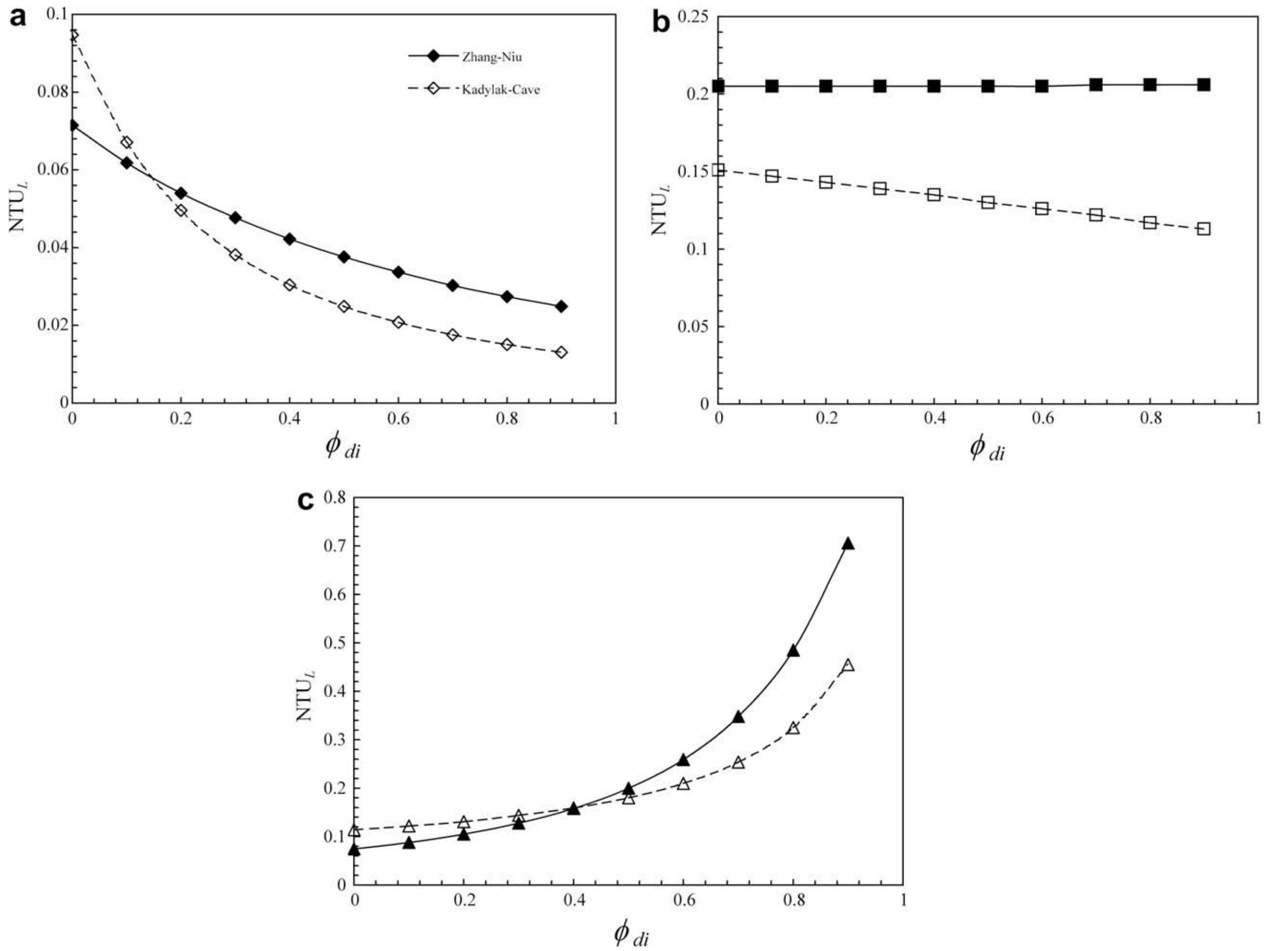

Fig. 4. Variation of NTUL with inlet relative humidity for constant NTU: (a) Type-I membrane ($C = 0.1$); (b) linear-type membrane ($C = 1$); and (c) Type-III membrane ($C = 10$).

the points of greatest interest on the curves are at the far left, at zero dry inlet relative humidity. Table 2 summarizes the NTU_L and latent effectiveness deviations when the simplifications made by Zhang and Niu are used as compared to the proposed solution method of this paper. When the two limitations have been addressed, improvement is made to the calculated outputs, ranging from 23% to 46% for the Type-III membrane when determining the latent effectiveness for the 70 °C isothermal case.

As the conditions for PEMFC operation show larger deviations from those of ERVs, it is expected that the difference between the two methods will grow larger, and it will become more important to implement the approach outlined in this paper. It is also beneficial to incorporate the proposed solution for rectifying the limitations to ERV systems to attain more accurate predictions for the outlet conditions.

6. Conclusions

Zhang and Niu have developed a method for determining the latent effectiveness and latent number of transfer units from an analogy to heat transfer. However, limitations were discovered when applying the technique from ERVs to PEMFC plate-and-frame membrane humidifiers. The two limitations that cause the predicted outlet conditions to deviate from the true conditions are:

1. The use of a constant enthalpy of vaporization taken from a low reference temperature in the Clausius–Clapeyron equation.
2. The simplification used in order to make a linear relationship between relative humidity and absolute humidity.

Using the Clausius–Clapeyron equation with the parameters that Zhang and Niu use will create a 4% deviation from the Hyland–Wexler equation for the water saturation pressure curve. With the simplification of the vapor pressure being much less than the air pressure to make the relation between relative humidity and absolute humidity linear, the value in using the relation can be underreported by over a third at 70 °C.

Due to the elevated temperatures used in PEM fuel cells as compared to ERV systems and the non-linear dependence of the water saturation curve on temperature, the aforementioned limitations must be addressed in order to use the latent effectiveness method for PEMFC membrane humidifiers. This was accomplished by finding an effective mass transfer coefficient U_{eff} instead of the U_L proposed by Zhang and Niu. The U_{eff} coefficient is calculated by first iteratively solving the relevant constitutive equations to find the flux of water through the membrane. This approach requires the use of a computer code, while the model by Niu and Zhang results in a simple algebraic formula accurate enough for analyzing the performance in most climate conditions for ERVs.

The new procedure was applied to three membrane types and compared to the curves of latent effectiveness and latent NTU found using Zhang and Niu's method. In fuel cell operation, the most likely conditions for the incoming wet stream will be 100% relative humidity and close to 0% relative humidity for the incoming dry stream. For a 70 °C isothermal case, the technique yielded an enhancement as compared to Zhang and Niu's method in latent effectiveness of 29% for Type-I membranes, 23% for linear-type membranes, and 46% for Type-III membranes.

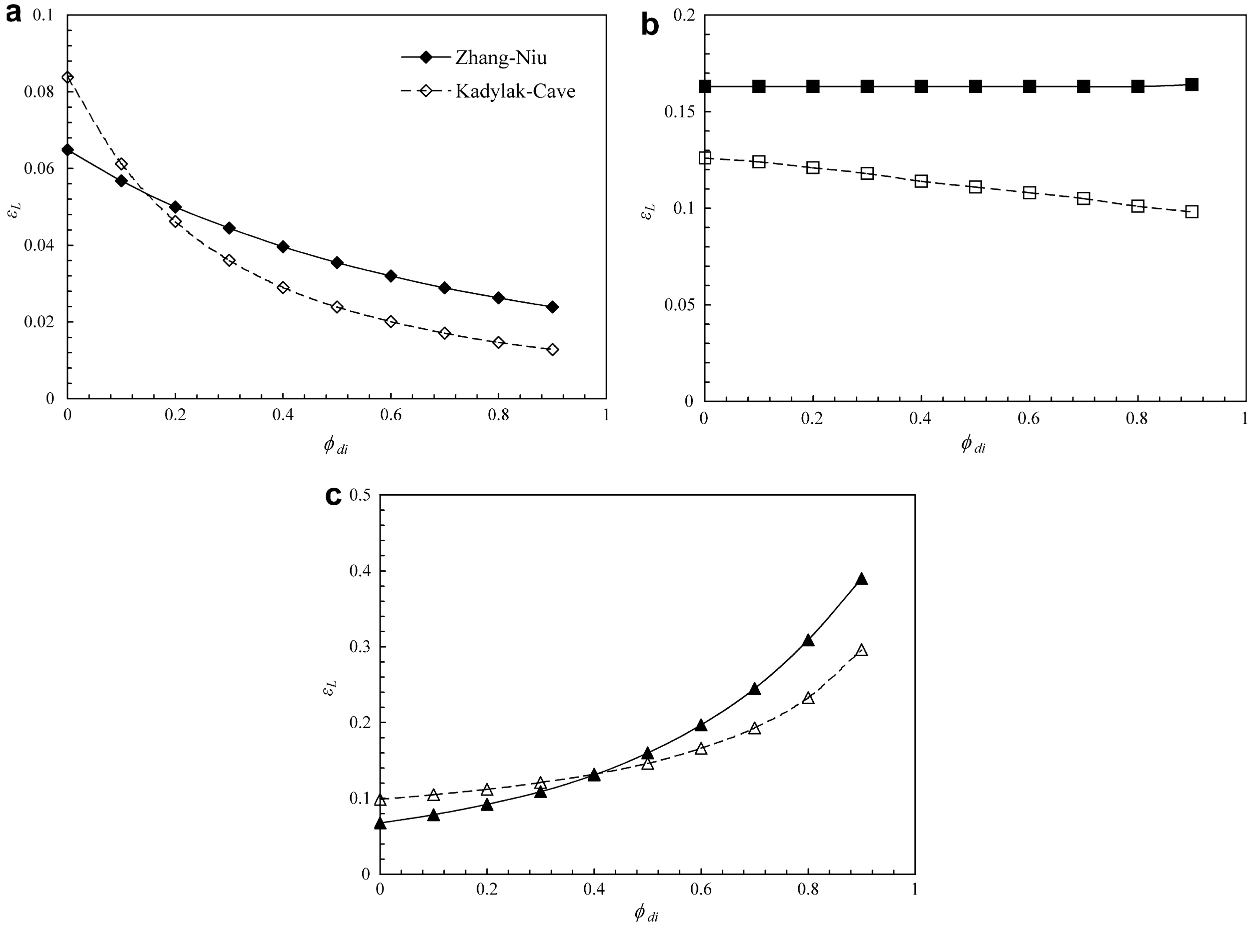

Fig. 5. Latent effectiveness for constant NTU: (a) Type-I membrane ($C = 0.1$); (b) linear-type membrane ($C = 1$); and (c) Type-III membrane ($C = 10$).

Table 2
Comparison based on methodology of latent NTU and latent effectiveness for 0% dry inlet RH

Membrane	Difference Kadylak–Cave vs. Zhang–Niu	
	NTU_L (%)	ε_L (%)
Type-I	32.4	29.1
Linear-type	−26.3	−22.7
Type-III	56.8	46.4

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