Interfacial Oxygen Transport Resistance in a Proton Exchange Membrane Fuel Cell Air Channel with an Array of Water Droplets

Mustafa Koz^a, Satish G. Kandlikar^{a, b, *}

^aMicrosystems Engineering, Rochester Institute of Technology, 168 Lomb Memorial Dr., Rochester, NY, 14623, U.S.A.

^bMechanical Engineering, Rochester Institute of Technology, 76 Lomb Memorial Dr., Rochester, NY, 14623, U.S.A.

E-mail Addresses:

Mustafa Koz: mxk7984@rit.edu

Satish G. Kandlikar: sgkeme@rit.edu

*Corresponding author:

Satish G. Kandlikar

sgkeme@rit.edu,

Mechanical Engineering, Rochester Institute of Technology, 76 Lomb Memorial Dr., Rochester, NY, 14623, U.S.A.

Telephone: +1-(585)-475-6728

Fax: +1-(585)-475-6879

Abstract

The oxygen (O_2) transport resistance at the gas diffusion layer (GDL) - air channel interface in a proton exchange membrane fuel cell (PEMFC) is numerically investigated. The interfacial O_2 resistance under the two-phase flow conditions is needed in modeling fuel cell performance. This work focuses on simulating the effect of water droplets on the GDL surface on the mass transfer resistance at the interface. Multiple droplets are placed at the center of channel width and spaced uniformly in the flow direction. The variation of interfacial O_2 transport resistance is characterized with the non-dimensional Sherwood number (Sh). The numerical technique is validated by comparing the fully developed Sh (in the absence of droplets) to the established values in the literature. Parametric simulations are performed for variable air Péclet number (varied due to changing superficial air velocity), non-dimensional droplet size and uniform spacing. Correlations are developed to express the Sh variation in terms of the aforementioned variables and the location in the channel. The maximum error of the correlations among numerically generated 999 data points is 11%.

Keywords:

Proton Exchange Membrane Fuel Cell; Interfacial Mass Transport; Droplet; Simulation; Sherwood number

Nomenclature

Abbreviations

BPP	bipolar plate
CL	catalyst layer
FD	fully developed
GDL	gas diffusion layer
PEMFC	proton exchange membrane fuel cell

Variables

a, b, c	coefficients of correlations
C	oxygen molar concentration
$D_{O_2\text{-air}}$	molar oxygen diffusivity in air
$D_{O_2\text{-GDL}}$	molar oxygen diffusivity in the gas diffusion layer
d_h	hydraulic diameter
F	air drag force on a droplet

F_c	Faraday's constant
g	gravitational acceleration
H	air channel height
h_M	mass transfer coefficient
$\mathbf{I}$	identity matrix
i	current density
j	magnitude of oxygen molar flux
L	simulated air channel length
N	number of droplets in the air channel
Nu	Nusselt number
Pe	Péclet number
Po	Poiseuille number
p	air pressure
r	droplet obstruction radius
R_u	universal gas constant
Sc	Schmidt number
Sh	Sherwood number
t	gas diffusion layer thickness
T	temperature
$\mathbf{u}$	velocity vector
u, v, w	velocity components
V	voltage
W	air channel width
We	Weber number
$\mathbf{x}$	spatial coordinate vector
x, y, z	spatial coordinate components
Δx	distance in the flow direction

Greek

Π	non-dimensional group
-------	-----------------------

θ	droplet static contact angle
μ	dynamic viscosity of air
ζ	droplet-blocked portion of the channel cross sectional area
ρ	air mass density
σ	air-water surface tension

Subscript

∞	asymptotic value
av	averaged
ch	channel based
d	droplet
eff	effective
FD	fully developed
int	interfacial
m	mean
n	droplet number in the flow direction
x	flow direction specific
wet	droplet-covered portion of the GDL-channel interface

Superscript

*	non-dimensionalized
T	transpose

1. Introduction

The performance of a proton exchange membrane fuel cell (PEMFC) is adversely affected by oxygen (O_2) concentration loss from the air flow channel to the cathode catalyst layer (CL). Oxygen transport resistance at the gas diffusion layer (GDL) and air channel interface is a non-negligible component of the O_2 concentration loss. The simplified PEMFC performance models in the available literature incorporate the O_2 resistance at the GDL-channel interface as an input parameter [1–8]. However, this parameter has been taken as a constant so far in the available literature and does not reflect variable PEMFC operating conditions and the effect of two-phase flow in the channels.

Sherwood number (Sh) is the non-dimensionalized mass transfer coefficient (h_M): $Sh = h_M \times d_h / D$ where d_h and D are the hydraulic diameter and diffusivity, respectively. O_2 transport resistance is inversely proportional to the mass transfer coefficient and thereby, Sherwood number. With a constant O_2 flux at the interface, the aforementioned proportionality can also be expressed by the interfacial concentration drop which is the difference between the mean concentration at a channel cross section and the average concentration at the respective interface. The interfacial concentration drop decreases with an increase in Sherwood number. This number depends on the aspect ratio of the channel cross section, boundary conditions, and local flow conditions. Local flow conditions can be characterized at cross sections with profiles of velocity and normalized concentration by using the mean value at the respective cross section. The aforementioned profiles stop varying in the direction of flow after enough distance traveled. Variations are observed only in axes normal to the flow direction. In laminar flow conditions of internal forced convection, variations of velocity and normalized concentration spans the entire channel cross section. These conditions are called fully developed (FD). In FD sections of a channel, the heat and mass transfer analogy is applicable, and Sh is the same as the Nusselt number (Nu). Hence, established Nu values can be used as Sh in PEMFC air channels with no limitations as long as the flow is fully developed. The heat and mass transfer analogy has been utilized by several researchers for the fully developed condition [9–12]. However, Nu-Sh mapping cannot always be easily performed in an air channel since liquid water features in the channel may not allow the flow to be fully developed. Since there is no equivalent heat transfer problem that resembles two-phase flow conditions in PEMFC channels, Sh needs to be simulated for channels with two-phase flow features.

Water features in the air channels can disrupt the air flow, prevent it from being fully developed, and lead to a local Sh value different than the fully developed one. Among the water features observed in PEMFC air

channels, droplets are the ones that emerge initially and then they transform into more complex shapes like films or slugs [13,14]. The droplet presence in the channel has been confirmed through visual observations by earlier researchers, and therefore deserves attention for research. Among the 3D and time-dependent numerical studies on the effects of water features on cell performance [15,16], Chen et al. focused on droplets and films [16]. They showed that a droplet constricts the cross section for the air flow and leads to a local high velocity field. The higher local velocity enhances the O_2 concentration at the interface and therefore increases the local cell performance. However, the effect of the droplet on Sh variation was not included in their analysis. By characterizing Sh in the vicinity of droplets, the interfacial O_2 transport resistance can be predicted for a GDL-channel interface known to be covered with droplets. Earlier researchers have used optical visualization of PEMFCs [17–26], electrochemical sensors in the flow channels [27], ex-situ [19,28–35] and numerical studies [16,19,30,36–38] to observe or predict droplet sizes and locations on the GDL-channel interface under a given PEMFC operating condition.

The droplet emergence location along the channel width alters the local flow conditions. It is known that droplets mostly emerge from under the bipolar plate (BPP) lands (ribs) [39,40]. However, there are studies suggesting that this does not always happen. Optical visualization studies documented that the droplets can emerge in the center of the channel width [17–22]. It was hypothesized that a GDL with a smaller porosity would lead to a droplet emergence more uniformly distributed along the channel width rather than concentrated underneath the BPP lands [23]. Moreover, it was proposed to make a groove in the GDL at the channel center width and along the flow direction to create a preferential droplet emergence location and hence increase cell performance [25].

The effect of droplets on Sh was studied numerically and shown to be important in the available literature. Koz and Kandlikar simulated a single droplet in an isolated 3D air channel by using a droplet-shaped obstruction [41]. The use of an obstruction allowed the steady-state solution of single-phase air flow and mass transport equations. This obstruction was centered along the channel width and local Sh was obtained along the flow direction as droplet size and superficial air velocity was varied. The variables in their analysis were selected in such a way that the numerically obtained drag force on the obstruction did not exceed the droplet-GDL adhesion force that was experimentally obtained by Das et al. [33]. Koz and Kandlikar numerically calculated Sh_{FD} to be 3.36 for a channel cross section of $0.40 \text{ mm} \times 0.70 \text{ mm}$. This Sh_{FD} is significantly lower than the ones previously recommended: $Sh_{FD}=5.274$ [42] (value readjusted for the characteristic length of hydraulic diameter),

6.0 [43], and 5.411 [44]. Moreover, it was shown that local Sh can be larger than Sh_{FD} downstream of the droplet [41]. As a result, a single droplet was found to decrease the interfacial O_2 resistance, a finding that is in agreement with the results of Chen et al. [16]. The increase in Sh by a single droplet leads to the question of how multiple droplets would alter Sh as a more representative case of a PEMFC air channel.

In a later numerical study by Koz and Kandlikar [45], single, two and multiple droplet cases were simulated over a more refined parametric range compared to their previous work [41]. Two and multiple droplet simulations revealed the effect of droplet spacing on Sh and how Sh varied at each consecutive droplet, respectively. An experimentally obtained correlation by Cho et al. [30,38] was used to ensure that no significant droplet deformation was present due to air flow. Multiple droplets with a uniform spacing of 2.00 mm led to a gradual increase in Sh that mostly disappeared after the sixth droplet. The value of Sh that showed no change in the flow direction was called as the asymptotic Sh .

Koz and Kandlikar found that the increase in the Sherwood number was a function of the number of droplets [45]. It was shown that the local Sh downstream of a droplet can increase at the most 21% by the first droplet, 78% by the second droplet, and 122% by the sixth and consecutive droplets with respect to Sh_{FD} . These results suggested an increase in the PEMFC performance with the presence of multiple droplets. Heidary and Kermani proposed a similar performance increase by artificially placing indentations in the channel wall across the GDL [10]. The multiple droplet study by Koz and Kandlikar [45] presented only four cases of multiple droplets with variable droplet size and air velocity, and constant droplet spacing. The asymptotic Sh due to multiple droplets needs to be characterized for a wider range of droplet size and air velocity, and variable droplet spacing.

The present work focuses on the numerical evaluation of Sh in an air channel that houses uniformly spaced multiple droplets on the GDL-channel interface. Compared to the previously published work [45], the present work is distinguished with a validation for droplet adhesion prediction, a significantly extended data set of Sh for multiple droplets (111 cases compared to 4), and correlations of Sh (tested against 999 data points). As a fundamental insight, the present study shows a critical condition that distinguishes an increase and decrease in Sh with respect to the fully developed value. Moreover, this study is conducted with a non-dimensional approach. This allows the results from this paper to be applicable to different length scales. A verification of this approach is provided later in this paper. The Péclet number (in this work, it was varied due to changes in superficial air velocity), non-dimensional droplet radius and uniform spacing were varied. Sets of these

parameters were chosen in such a way to ensure that the droplets remained spherical and adhered to the interface. Local variations of Sh in the flow direction and the asymptotic Sh are reported as figures of merit.

2. Methodology

Dynamic simulations [16,36,37] and experiments [17–20,22] in the available literature captured instances at which droplets are attached to the GDL surface and are still growing. This growth process was documented to last 3 minutes [17], much longer than the removal process occurring over milliseconds [16]. The present work is aimed at investigating the interfacial mass transport resistance under the effect of droplets with long attachment times, as identified in the available literature. In order to pursue this goal, droplet shaped obstructions were used similar to the work reported by Cho et al. [38] and Venkatraman et al. [46]. The droplets were considered to remain spherical and attached to the GDL. These conditions were selected based on experimentally obtained correlations for droplet deformation due to air flow [30,38] and surface adhesion force [33].

2.1 Introduction to the Problem Geometry and Physics

The numerical study was conducted for an air channel with single-phase air flow and droplet-shaped obstructions placed on its bottom surface. The channel width ($W = 0.70$ mm) and height ($H = 0.40$ mm) were taken from the parallel flow field design study [47] to meet the U.S. Department of Energy performance targets. The corresponding hydraulic diameter (d_h) is 0.51 mm.

Fig. 1a shows multiple droplets placed in a row in the channel and numbered sequentially in the flow direction. The number of droplets (N) varied based on the uniform droplet spacing (Δx_d) and this led to a variable simulated channel length (L). In order to comply with the computational limitations, N was selected high enough for the results not to differ significantly in the flow direction by the last droplet. The value of N was 11 for $\Delta x_d = 1.00$ and 2.00 mm, and 5 for $\Delta x_d = 5.00$ mm. The first droplet was placed 63.00 mm downstream of the channel inlet ($x_{d,1} = 63.00$ mm) to obtain a fully developed flow at the first droplet. The distance between the last droplet and the channel exit was kept 10.00 mm to allow the secondary flows in the wake of the last droplet to be simulated.

The droplets were centered along the channel width ($y_{d,n} = W/2$) and were represented by spheres of radius (r). The spheres were truncated to obtain the desired contact angle, $\theta = 147^\circ$. The contact angle was selected as a representative value of the GDL characterized by Gopalan and Kandlikar [48] and Das et al. [33]. As a result of the selected r and θ , a portion of the GDL-channel interface width (W_{wet}) was covered by the droplet.

Air properties were calculated at a temperature (T) of 80 °C and 100% relative humidity. Under these conditions the molar fraction of oxygen at the channel inlet was 0.11. The consumption of oxygen does not alter the air properties significantly. Hence, the properties in the channel were kept constant in the simulations. These properties are density ($\rho = 0.828 \text{ kg m}^{-3}$), dynamic viscosity ($\mu = 16.253 \text{ }\mu\text{Pa s}$), oxygen diffusivity in the air ($D_{\text{O}_2\text{-air}} = 0.310 \text{ cm}^2 \text{ s}^{-1}$), and the corresponding Schmidt number ($Sc = \mu / (\rho D_{\text{O}_2\text{-air}}) = 0.633$). The procedure for calculating these properties was presented earlier [45].

The superficial air velocity (u_m) imposed at the channel inlet was calculated based on the rate of O_2 consumption for the corresponding active area and the stoichiometric ratio. The rate of O_2 consumption was calculated as a function of average current density (i) for a range of 0.05 - 1.5 A cm^{-2} . The current density was assumed to be constant over the flow direction. The available literature provides the extent of current density variation along the channel width. The numerical work by Um and Wang showed that the current density variation is 23% at 1.01 A cm^{-2} [49]. The experimental work by Wang and Liu demonstrated that current density variations are less than 18% for average current densities larger than 0.30 A cm^{-2} [50]. Since the current work mostly covers current densities larger than 0.30 A cm^{-2} , the aforementioned findings are relevant to this work. As a result, the current density was assumed to be uniform along the channel width. This assumption was translated to the GDL-channel interface as constant oxygen flux since channel width was larger than the land width.

The stoichiometry was taken as 2 for the entire range of current density, except 0.05 and 0.1 A cm^{-2} , for which the stoichiometry was taken as 3 to maintain a reasonable flow velocity. As shown in Fig. 1b, the active area corresponding to the channel was in a repeating pattern that covers the width of the GDL-channel interface and the bipolar plate land (rib). The active area length and bipolar plate land width were taken to be 183.00 mm and 0.50 mm, respectively [47]. The range of superficial air velocity corresponding to the aforementioned current density range was calculated to be 0.80 - 15.89 m s^{-1} .

2.2 Simulation Matrix

Certain sets of droplet size and superficial air velocity were excluded from the simulation matrix as being unrealistic cases. The minimum droplet radius was chosen to be 0.10 mm as governed by the maximum GDL pore size of 0.10 mm [19]. The maximum droplet radius was dependent on the specific superficial air velocity since a set of these parameters had to comply with the following conditions: 1) the droplets should remain in the form of a sphere, and 2) they should remain attached to the GDL-channel interface. When these conditions were

satisfied, the use of droplet-shaped obstructions could be justified. Koz and Kandlikar showed earlier that the droplets considered in this study were unaffected by the gravitational forces [45].

The air flow can deform a droplet from a perfect spherical shape. The deformation of the droplet was experimentally characterized by Cho et al. [30,38]. The deformed shape can be expressed as an ellipsoid with a semi-major axis of r_x and a semi-minor axis r_y . The mismatch between the two semi-principal axes is normalized with the average droplet radius $(r_y - r_x) / r$. The authors correlated these normalized deformations with the Weber number (We) and channel blockage ratio (ξ). The Weber number expresses how dominant the air flow inertia is over the surface tension ($\sigma = 62.6 \text{ mN m}^{-1}$). The channel blockage ratio takes the increased air flow velocity into account when a droplet constricts the channel. Eq. (1) provides the aforementioned correlation in which θ is in radians.

$$\frac{(r_y - r_x)}{r} = \frac{We}{4} \xi^2 = \left(\frac{\rho u_m^2 r}{4\sigma} \right) \left(\frac{WH}{WH - r^2(\theta - \cos \theta \sin \theta)} \right)^2 \quad (1)$$

Table 1 shows the calculated normalized deformation values for a set of droplet radii and superficial air velocities. The air velocity values were selected to present the higher end of the range where significant deformations happen. The emboldened values indicate cases with normalized deformations larger than 0.105. These cases were not simulated as the spherical droplet assumption was not valid.

Koz and Kandlikar investigated the droplet adhesion to the GDL-channel interface [45]. They took a hybrid approach of comparing the numerically calculated air drag force on a droplet to an experimentally obtained adhesion force [33]. However, they did not use the superficial air velocity of 8.48 m s^{-1} in their analysis. Therefore, the same technique was applied in this work for the droplet radius of 0.18 mm. Although the drag force (13.16 mN) exceeded the adhesion force (12.32 mN) only 7%, the aforementioned condition was still taken as an adhered droplet for the richness of the data presented here. Based on the earlier [45] and current results, it can be concluded that the cases in Table 1 with a negligible deformation will adhere to the interface. Moreover, all droplet radii with air velocities lower than 4.24 m s^{-1} also lead to adhesion.

The uniform spacing between the droplets was varied in between the values that were documented to exist in an operational fuel cell and also led to meaningful changes in Sh. Visualization studies of operational PEMFCs reported in the available literature were used to extract a representative value for the minimum droplet spacing. The droplet images provided by Yang et al. [17] and Zhang et al. [18] suggest that 1.00 mm is a reasonable droplet spacing. The maximum droplet spacing needs to be chosen in such a way that two consecutive large

droplets should be close enough to interact even at low superficial air velocities. When the velocity was 4.24 m s⁻¹ and the droplet radius was 0.20 mm, Sh was only marginally affected 5.00 mm downstream of the droplet [45]. Therefore, the maximum droplet spacing was chosen to be 5.00 mm. As an intermediate value of droplet spacing, 2.00 mm was selected since most of the maximum local Sh values were seen 2.00 mm downstream of a droplet [45].

2.3 Theoretical Background for the Interfacial Transport Resistance

The objective of this work is to find the oxygen concentration drop (ΔC) for a given magnitude oxygen flux (j) at the GDL-air channel interface. Non-dimensional variables are used to reduce the number of simulations and obtain correlations that are applicable to a wider range of conditions. All dependent and independent parameters for this problem with a constant channel aspect ratio are ρ , μ , Δx_d , $D_{O_2\text{-air}}$, r , u_m , d_h , ΔC , and j . These parameters were non-dimensionalized by using the Buckingham Pi Theorem [51]. It was aimed to obtain Sherwood number as one of the Π groups. This was possible by choosing the following repeating parameters: d_h , ΔC , $D_{O_2\text{-air}}$, and ρ . The resulting set of Π groups were Schmidt number ($Sc = \mu / (\rho D_{O_2\text{-air}})$), non-dimensional droplet radius ($r^* = r / d_h$), non-dimensional droplet spacing ($\Delta x_d^* = \Delta x_d / d_h$), Péclet number ($Pe = d_h u_m / D_{O_2\text{-air}}$), and Sherwood number ($Sh = j d_h / (\Delta C D_{O_2\text{-air}})$).

As a result of non-dimensionalization, the parameters r , Δx_d , and u_m were mapped to r^* , Δx_d^* , and Pe , respectively. The values of droplet radius $r = 0.10, 0.12, 0.15, 0.18$, and 0.20 mm are referred to as $r^* = 0.20, 0.24, 0.29, 0.35$, and 0.39 , respectively. The values of uniform droplet spacing $\Delta x_d = 1.00, 2.00$, and 5.00 mm are mapped to Δx_d^* as $1.96, 3.92$, and 9.80 , respectively. The mapping from i to u_m and Pe is given in Table 2.

For the numerical evaluation of Sh, the local oxygen transport resistance is calculated at every point on the GDL-air channel interface. For a given magnitude of O₂ molar flux at the GDL-channel interface (j), the corresponding transport resistance is ΔC which is the difference between the O₂ concentration at interface $C_{\text{int}} = C(z=0)$ and the mean value at a channel cross section (C_m). This resistance is expressed with the mass transfer coefficient (h_M) which is a component of the Sherwood number definition.

$$h_M(x, y) = \frac{j}{\Delta C} = \frac{j}{(C_m - C_{\text{int}})} \quad (2)$$

$$C_m(x) = \frac{\int_{\text{cross section}}^{\text{Channel}} C u dA}{\int_{\text{cross section}}^{\text{Channel}} u dA} \quad (3)$$

In simulations, h_M is zero at the portion of the GDL-channel interface covered by the droplet: $W_{\text{wet}}(x)$. An effective mass transfer coefficient ($h_{M,\text{eff}}$) was defined in Eq. (4) to have an assigned value of h_M everywhere on the interface ($z = 0$).

$$h_{M,\text{eff}}(x, y) = \begin{cases} \text{Dry Interface} & h_M \\ \text{Wet Interface} & 0 \end{cases} \quad (4)$$

In Eq. (5), the variations of $h_{M,\text{eff}}(x, y)$ were averaged along the dry GDL-channel interface width ($W - W_{\text{wet}}(x)$) into the dry-region-averaged mass transfer coefficient ($\overline{h_M}(x)$) to express its variation in the flow direction.

$$\overline{h_M}(x) = \frac{1}{W - W_{\text{wet}}} \int_{y=0}^W h_{M,\text{eff}} dy \quad (5)$$

The dependency of $\overline{h_M}$ on d_h and $D_{\text{O}_2\text{-air}}$ was eliminated with the use of Sherwood number ($\text{Sh} = \overline{h_M} d_h / D_{\text{O}_2\text{-air}}$). For expressing the variations of Sh along the flow direction in a more compact manner, Sh was averaged in between two consecutive droplets (n and $n+1$ in the row) into down-the-channel-averaged Sh ($\text{Sh}_{\text{av},n}$) as shown in Eq. (6).

$$\text{Sh}_{\text{av},n} = \frac{1}{\Delta x_d} \int_{x=x_{d,n}}^{x_{d,n} + \Delta x_d} \text{Sh} dx \quad (6)$$

3. Numerical Technique

In this numerical study, convective mass transport was modeled with steady-state and 3D formulation of Navier-Stokes, conservation of mass and species equations given by Eqs. (7) - (9), respectively. The air flow was assumed to be incompressible. The oxygen transport was governed by Fick's law as presented in Eq. (9). This is a simplified formulation of Maxwell-Stefan equation. This simplification is justified for a condition of transported species being dilute in the mixture as stated by Krishna and Wesselingh [52]. Since, oxygen had a molar fraction of 0.11 at the channel inlet, Fick's law could be utilized.

$$\rho(\mathbf{u} \cdot \nabla) \mathbf{u} = \nabla \cdot [-p\mathbf{I} + \mu(\nabla \mathbf{u} + (\nabla \mathbf{u})^T)] \quad (7)$$

$$\rho \nabla \cdot \mathbf{u} = 0 \quad (8)$$

$$\mathbf{u} \cdot \nabla C = \nabla \cdot (D_{\text{O}_2\text{-air}} \nabla C) \quad (9)$$

In order to reduce the computational cost of the numerical solution, the governing equations were solved separately, the entire length of the domain was treated in shorter and separate components, and a symmetry boundary condition was utilized. Firstly, constant air properties allowed the separate solutions of the governing

equations. Eqs. (7) and (8) were initially solved together, and then followed by Eq. (9). Secondly, the entire length of the channel was divided into three sections and the resulting shorter channel sections were simulated in sequence along the flow direction. The resulting flow profiles at the exit of the first and second sections were mapped as inlet conditions for the second and third sections, respectively. The third section housed the droplets that were placed in a row starting from 3.00 mm away from the channel inlet. For convenience, $x = 0$ and 3.00 mm will refer to the third channel inlet and the location of the first droplet, respectively in the following text. This technique was verified by comparing two approaches that simulated the entire channel either as a single domain and three separate domains. As the final technique to reduce the computational requirements, a symmetry boundary condition was imposed at the $y = W/2$ plane.

Fig. 2 shows the mesh for the symmetric half of the channel that is housing hollow droplet-shaped obstructions with a radius of 0.15 mm. Along with tetrahedral mesh elements, boundary layer mesh elements were used on the channel walls and droplet boundary to increase accuracy. The maximum mesh element size in this domain is $58.7 \mu\text{m}$ that was established as a result of a mesh independency study as presented in Section 4.1.

The boundary conditions for the Navier-Stokes equations included uniform air velocity at the inlet, zero gauge pressure at the outlet, and zero-velocity boundary condition at all other boundaries. The rationale for considering the zero-velocity boundary condition on the droplet surface and GDL-channel interface was presented earlier [45].

Constant O_2 concentration (3.83 mol m^{-3}) was imposed at the inlet, and only convection was allowed at the outlet. Since the O_2 diffusion through the droplet is negligible compared to the convection around the droplet in reality, the magnitude of O_2 flux (j) was imposed to be zero on the droplet surface. Moreover, all walls of the channel except the GDL had zero j .

The magnitude of O_2 flux was imposed on the GDL-channel interface in such a way that the net mass transfer rate through the entire available interfacial width for a given x (down the channel position) remained constant. The constant flux assumption should nevertheless serve as the base for future studies with varying flux. The details about imposing this boundary condition was presented earlier [45].

The numerical problem was solved with a commercially available software package COMSOL Multiphysics® 4.3. The unknown variables $\mathbf{u}$, T and C were represented with second order shape functions while the variable p was represented with a first order one. The error residuals were automatically scaled and required to converge to

10^{-6} . An iterative solver, Generalized Minimum Residual Method, solved the numerical problem while utilizing a geometric multigrid scheme. A typical domain with droplets had 1.4 M tetrahedral mesh elements and these elements corresponded to 6.2 M degrees of freedom during the solution of Navier-Stokes equations.

4. Results and Discussion

In this section, the numerical technique is first validated. The validation is followed by parametric Sherwood number (Sh) results along with correlations. The obtained results of Sh are valid for the specified channel cross sectional aspect ratio, GDL contact angle, adequately small droplet size and superficial air velocity (for ensuring spherical droplet shape and droplet adhesion to the interface), constant oxygen flux boundary condition on the interface, and the Schmidt number. Although, the exact values of the aforementioned parameters may differ in fuel cells, the values used in this work are representative of the mostly encountered conditions. The results are presented in a non-dimensional form to make them useful in different channel sizes and the results have been verified. Towards the end of the section, the applicability of the results at different air properties (expressed by the Schmidt number) and the significance of Sherwood number variations in terms of fuel cell performance are discussed.

4.1 Mesh Independency and Validation Study

The numerical technique was checked for mesh independency for the figure of merits that were unique to this study and have not been established in the literature. Following the mesh independency, well established figures of merit from the literature were compared to the numerical results from the fully developed flow region. The mesh independency study was performed on a case that led to the strongest flow disruption affiliated to high Sh and drag force on a droplet (F_x). This case was identified by the results of Koz and Kandlikar as $u_m = 10.59 \text{ m s}^{-1}$, $r = 0.15 \text{ mm}$ and $\Delta x_d = 2.00 \text{ mm}$ [45].

In the mesh independency study, Sh and F_x showed the largest variation with the mesh at the third droplet. The mesh with a maximum element size of $58.7 \text{ }\mu\text{m}$ was compared against another one with a maximum element size of $87.6 \text{ }\mu\text{m}$. Sh and F_x varied 0.5% and 2.2%, respectively in between two mesh configurations. A conservative approach was taken and the mesh with a maximum element size of $58.7 \text{ }\mu\text{m}$ was used for all simulations in this work. The total number tetrahedral mesh elements was 1,725,467.

Following the mesh independency study, the numerical method was validated against fully developed values of Sh and Poiseuille (Po) numbers. Since Sh is equal to Nu in the fully developed region, the numerical study on Nu by Dharaiya and Kandlikar [53] was utilized to validate Sh_{FD} . The correlation provided by this study [53]

was developed for constant heat flux boundary condition that is equivalent to the constant O_2 flux. The values of $Nu_{FD} = 3.584$ [53] and $Sh_{FD} = 3.349$ deviate within 7.0% and hence, agree well. The value of $Po_{FD} = 15.127$ was provided by Shah and Sekulic [54]. This value agrees with the numerically obtained value $Po_{FD} = 15.155$ within 0.2% error.

As a final validation, the droplet adhesion prediction technique that is presented in this work was tested against the experimental data by Kumbur et al. [35]. These researchers observed droplet attachment and detachment conditions for a set of droplet size and channel-based Reynolds number ($Re_{ch} = \rho d_h u_m \mu^{-1}$). The droplet size was expressed through the ratio of droplet height-to-cord length (width of droplet footprint) at a constant cord length of 1.90 mm.

The ex-situ setup [35] was numerically modeled with a mesh equivalent to the one resulting from the mesh-independency study. The simulated cases were a subset of the reported conditions [35]. Among the selected data points of attachment, the set of Re_{ch} and droplet height-to-chord length ratio was selected as large as possible. For detachment condition, a set of aforementioned parameters was selected with values as low as possible. Any condition with a larger droplet size and Re_{ch} would lead to detachment as well. The experimental and numerical conditions are summarized in Table 3. The attachment conditions were simulated with the exact droplet height-to-cord length ratio and Re_{ch} . The detachment condition was simulated with a lower Re_{ch} than the reported one [35]. This change has been done to obtain a steady state numerical solution with the assumption of a spherical droplet shape. Despite the lower simulated Re_{ch} , detachment was still predicted and matched the simulated experimental condition.

The droplet was expressed as a truncated sphere as presented in Section 2.1. The spherical droplet representation could be used because the largest normalized droplet deformation was calculated to be 0.15 for the droplet height-to-chord length ratio of 1.33 and Re_{ch} of 710. In order to have droplets with a given height and cord length, the contact angle was varied within the range of $130.0^\circ < \theta < 139.0^\circ$ as presented in Table 3.

In the experiment, a droplet was injected from the center width of the channel through an artificial pore of SGL[®] carbon paper with 20% PTFE loading. The adhesion force of a droplet is affected by the droplet-pore interaction [33]. Das et al. experimentally obtained the adhesion force for the same GDL and droplet-pore interaction [33]. For a fresh sample of SGL[®] 24DA GDL with 20% PTFE loading, the adhesion-force-per-droplet-footprint-perimeter was reported to be $12 \mu N \text{ mm}^{-1}$. The resulting net adhesion force for the given cord length is $35.9 \mu N$. The numerically obtained drag values in Table 3 were compared against this adhesion force value to predict

attachment or detachment. The numerical prediction agreed with all experimental observations [35]. Hence, the numerical technique was validated and proven to be mesh-independent.

4.2 The Sherwood Number Results

This section presents variations of Sherwood number (Sh) in down the channel direction due to droplets. These variations are categorized based on the non-dimensional parameters affecting the results: Péclet number (Pe), non-dimensional uniform droplet spacing (Δx_d^*) and radius (r^*). The effect of Pe on Sh is presented in terms of Sh evolving into a value that is higher or lower than the fully developed value (Sh_{FD}) due to droplets. The effect of Δx_d^* is presented by showing how fast Sh changes down the channel direction with different values of Δx_d^* . Lastly, the effect of r^* is presented by depicting linear and quadratic dependences of Sh on r^* at high and low Pe values, respectively.

4.2.1 Sherwood Number Trends Based on the Critical Péclet Number

The effect of multiple droplets was characterized with the down-the-channel-averaged Sherwood number (Sh_{av}). The value of Sh_{av} can be larger or smaller than the fully developed value ($Sh_{FD}=3.349$) depending on the Péclet number (Pe) with respect to the critical Pe . The critical value of Pe changes slightly with the set of droplet size and spacing. The values of Sh_{av} among the droplets were found to be lower than Sh_{FD} at a Pe lower than the critical value.

Values of Sh_{av} for a non-dimensionalized droplet radius of $r^* = 0.39$ were compared against each other as the Péclet number was varied from 26.16 to 69.75 at each given uniform droplet spacing. Fig. 3 shows the Sh_{av} profiles at $r^* = 0.39$, $\Delta x_d^* = 1.96$, and $Pe = 26.16$ and 69.75. The case of $Pe = 26.16$ led to a decrease in Sh_{av} with respect to the fully developed value. Contrarily, the case $Pe = 69.75$ led to an increase. The variations of Sh_{av} were reduced considerably by the fourth droplet. In between the last two droplets, $Sh_{av,10}$ ($Sh_{av,N-1}$) was 24.6% higher at $Pe = 69.75$ compared to $Pe = 26.16$. At other values of non-dimensional droplet spacing ($\Delta x_d^* = 3.92$ and 9.80), the $Sh_{av,N-1}$ variation between of the corresponding Pe values was less than the reported one for $\Delta x_d^* = 1.96$.

Fig. 4 provides insight about how the local Sh increases with the introduction of droplets at high Pe . The conditions in Fig. 4 are $Pe = 261.42$, $r^* = 0.20$, and $\Delta x_d^* = 3.92$. Fig. 4a shows an air streamline in the symmetric half of the channel. For providing distinguishable images of droplets, one droplet is skipped to be shown for every other shown droplet. The streamline follows a straight line path upstream of the droplets.

Downstream of the droplets, the streamline follows a helical path. In down the channel direction, air rotates around the $+x$ axis on this helical path. This helical path of the air flow alters the local Sh. This alteration can be understood with the normalized oxygen concentration profile in the channel. Fig. 4b shows two yz cross sections taken at the channel inlet and in between the fifth and sixth droplets. The O_2 concentration is normalized with the mean concentration at the respective cross sections (C/C_m). Down the channel, the helical air flow path brings the high O_2 concentration from the vicinity of the top wall towards the GDL-channel interface as shown in Fig. 4b. In this particular case, the Sherwood number is increased since the O_2 concentration at the interface is increased.

Fig. 5 provides an explanation for the decrease of Sh below the Sh_{FD} value at low Pe values. As a representative condition, Pe, r^* , and Δx_d are selected to be 13.16, 0.39, and 1.96, respectively in Fig. 5. Fig. 5a shows streamlines that are placed at $z = 0.19$ mm that is slightly below the half-height of the channel ($H/2 = 0.20$ mm) and evenly distributed along the channel width. The streamlines creep over the top of the droplets since their originating height is slightly above the xy plane that crosses the droplet from its widest cross section ($z = 0.17$ mm). Among the consecutive droplets, no streamlines are directed towards the GDL-air channel interface. The effect of this flow pattern on the normalized O_2 profile is shown in Fig. 5b. The fully developed concentration profile is compared against the one in the wake of the 9th droplet at $x = 11.50$ mm. It can be seen that the normalized O_2 concentration at the interface is reduced due to droplets, a condition opposite to the one in high Pe.

4.2.2 The Effect of Droplet Spacing on the Sherwood Number

Fig. 6 shows the effect of uniform droplet spacing on Sh and Sh_{av} while r^* and Pe are 0.29 and 69.75, respectively. The number of simulated droplets (N) was 11 for $\Delta x_d^* = 1.96$ and 3.92, and 5 for $\Delta x_d^* = 9.80$. Fig. 6a uses a non-dimensional distance parameter to show variations of Sh for all droplet spacing values in a common domain. The non-dimensional distance parameter (x^*) expresses the first droplet position to be zero and the distance relative to the first droplet is normalized with Δx_d : $x^* = (x - x_{d,1}) / \Delta x_d$.

Local variations of Sh in Fig. 6a are averaged in down the channel direction into $Sh_{av,n}$. Fig. 6b shows values of $Sh_{av,n}$ corresponding to the averaged values of Sh between the n and $n+1$ st droplets. The values of $Sh_{av,n}$ reach an asymptotic value ($Sh_{av,\infty}$) towards the channel exit. In this particular case, $Sh_{av,\infty}$ is inversely proportional to Δx_d^* . However, it will later be shown that no such linear proportionality exists for all cases. The rate of Sh_{av}

convergence to the respective asymptotic value was observed to be higher with Δx_d^* . The analysis of the entire data set will show in Section 4.3 that this conclusion is applicable to all cases.

4.2.3 The Asymptotic Sherwood Number

Among all the simulated cases, the maximum change of Sh_{av} in between the last two sets of consecutive droplets was less than 2.0% except the case $r^* = 0.20$, $Pe = 261.42$, and $\Delta x_d^* = 9.80$ with a change of 3.4%. The reported change in Sh_{av} values show that the selected number of droplets was high enough to achieve Sh_{av} convergence for any uniform droplet spacing. The converged value is called the asymptotic Sh_{av} ($Sh_{av,\infty}$).

Table 4-6 show values of $Sh_{av,\infty}$ for all simulated cases while each table is given for a Δx_d^* . There are sets of r^* and Pe values leading to $Sh_{av,\infty} < Sh_{FD}$. The remaining sets of r^* and Pe lead to the opposite condition of $Sh_{av,\infty} > Sh_{FD}$. There is no critical value of r^* that can clearly distinguish $Sh_{av,\infty}$ values with respect to Sh_{FD} . However, the range of $Pe = 52.32 - 69.75$ can be used to separate the entire Pe range into three.

In the range of $Pe < 52.32$, $Sh_{av,\infty}$ values are always smaller than Sh_{FD} . These values can be as low as $0.87Sh_{FD}$. As shown by Fig. 7 and supported by Tables 4-6, $Sh_{av,\infty}$ increases with Δx_d^* . This is due to the air flow partially going over droplets and not having enough droplet spacing to be directed towards the GDL-air channel interface (as shown in Fig. 5). Fig. 7 also shows the quadratic relation between $Sh_{av,\infty}$ and r^* at three Δx_d^* values. Values of $Sh_{av,\infty}$ first decrease with r^* , reach a minimum value, and then increase. In Fig. 7a, the family of plots at $Pe = 13.16$ shows one symmetric half of a quadratic function. In Fig. 7b, data points exist at both sides of the minimum $Sh_{av,\infty}$ at $Pe = 26.16$. This is due to the position of the $Sh_{av,\infty}$ minimum shifting towards smaller values of r^* with increasing Pe . The same shift happens consistently as Pe is increased to 34.88.

In the range of $Pe > 69.75$, $Sh_{av,\infty}$ values are always larger than Sh_{FD} . Values can be as high as $2.22Sh_{FD}$. Family of plots in Fig. 8 and Tables 4-6 show that $Sh_{av,\infty}$ is always the lowest at $\Delta x_d^* = 9.80$ compared to the other two Δx_d^* . However, $Sh_{av,\infty}$ can reach its highest value at a different Δx_d^* value (1.96 or 3.92) based on the remaining set of parameters: r^* and Pe . There is also a strong decrease of $Sh_{av,\infty}$ observed with Δx_d^* approaching 9.80. Values of $Sh_{av,\infty}$ are linearly dependent on r^* . This dependency is valid for the entire range of $Pe > 69.75$. Moreover, $Sh_{av,\infty}$ increases consistently with Pe . This relationship manifests itself by increasing the slope of $Sh_{av,\infty} - r^*$ plots with Pe .

In the range of $52.32 \leq Pe \leq 69.75$, a transition happens from the trend at $Pe < 52.32$ to the one at $Pe > 69.75$. For instance, values of $Sh_{av,\infty}$ inconsistently decrease when r^* makes a transition from 0.29 to 0.35 at $Pe =$

69.75. This inconsistency had been observed at a single droplet and explained by Koz and Kandlikar with the use of vortices in the droplet wake and around the flow direction (x) [45]. They correlated the local decrease in Sh with the vortices changing their sense of rotation in the aforementioned transitional case [45]. Under these transitions, it is not possible to express the changes in $Sh_{av,\infty}$ with consistent relationships. The values of $Sh_{av,\infty}$ deviate from Sh_{FD} at most 11% in this Pe range. Consequently, a constant value of Sh_{FD} can be assumed in an application within this Pe range.

4.3 Correlations for the Down-the-Channel Averaged Sherwood Number

For each spacing between two consecutive droplets (n^{th} and $n+1^{st}$), $Sh_{av,n}$ can be predicted through the use of correlations in this section. The input variables are Pe , r^* , and Δx_d^* . Based on the number of simulated droplets, $Sh_{av,n}$ can be predicted with a maximum error of 11% at 999 data points corresponding to the interfacial area left in between two droplets.

As it was seen in Figs. 3 and 6b, Sh_{av} starts with an initial value ($Sh_{av,1}$) and converges to an asymptotic value in between the last two droplets ($Sh_{av,\infty}$) with a variable convergence rate (α). Eq. (11) mathematically expresses the down the channel variation of $Sh_{av,n}$. The three parameters that constitute the Eq. (11) were correlated separately to the input parameters.

$$Sh_{av,n} = Sh_{av,1} + (Sh_{av,\infty} - Sh_{av,1}) \left(1 - e^{-(n-1)/\alpha}\right) \quad (11)$$

The values of $Sh_{av,1}$ are correlated separately at $Pe \leq 34.88$ and $Pe \geq 121.91$. However, extrapolating the use of these correlations respectively up to $Pe \leq 52.32$ and $Pe \geq 69.75$ is also possible while remaining within 11% error. For the remaining range of Pe ($52.32 < Pe < 69.75$), $Sh_{av,1}$ can be assumed to be Sh_{FD} . The errors still remain below 11%. Irrespective of the Pe value, $Sh_{av,1}$ correlations behave quadratically with r^* as shown in Eq. (12). Coefficients of Eq. (12) are defined in Eq. (13) as a function of the matrix $\mathbf{M}$ and Δx_d^* . Eq. (13) reveals that $Sh_{av,1}$ decreases with Δx_d^* in the entire Pe range.

$$Sh_{av,1} = a_1 r^{*2} + b_1 r^* + c_1 \quad (12)$$

$$\begin{bmatrix} a_1 \\ b_1 \\ c_1 \end{bmatrix} = \mathbf{M} \begin{bmatrix} (\Delta x_d^*)^{-1} \\ 1 \end{bmatrix} \quad (13)$$

The matrix $\mathbf{M}$ in Eq. (13) is a function of Pe and its dependency of Pe changes at different ranges of Pe . The correlations for $\mathbf{M}$ are given in Eq. (14) and operate at two ranges of Pe ($Pe \leq 52.32$ and $Pe \geq 69.75$).

$$\mathbf{M} = \begin{cases} \text{Pe} \leq 52.32 & \begin{bmatrix} 10.481 & \frac{\text{Pe}^{2.13}}{558.24} \\ 0.0565\text{Pe} - 8.6652 & -\frac{\text{Pe}^{2.094}}{1015.2} \\ -0.0119\text{Pe} + 0.8786 & \frac{\text{Pe}^{0.043}}{0.3313} \end{bmatrix} \\ \text{Pe} \geq 69.75 & \begin{bmatrix} 0.6877\text{Pe} + 28.471 & 0.9231\text{Pe} - 120.64 \\ -0.2235\text{Pe} - 17.431 & -0.3925\text{Pe} + 54.264 \\ 0.02\text{Pe} + 1.8361 & 0.0424\text{Pe} - 2.7638 \end{bmatrix} \end{cases} \quad (14)$$

Values of $\text{Sh}_{\text{av},\infty}$ takes the same value with $\text{Sh}_{\text{av},1}$ at the conditions of $\text{Pe} < 69.75$ since $\text{Sh}_{\text{av},\infty}$ and $\text{Sh}_{\text{av},1}$ are very close to each other in this Pe range. For conditions of $\text{Pe} \geq 69.75$, Eqs. (15) – (18) express $\text{Sh}_{\text{av},\infty}$. Eq. (15) shows that $\text{Sh}_{\text{av},\infty}$ increases linearly with r^* . The coefficients for Eq. (15) are dependent on the matrices $\mathbf{K}$ and $\mathbf{L}$, and Δx_d^* . With the help of Eqs. (16)-(18), it can be stated that the coefficients a_2 and b_2 are quadratically dependent on Δx_d^* and Pe .

$$\text{Sh}_{\text{av},\infty} = a_2 r^* + b_2 \quad (15)$$

$$\begin{bmatrix} a_2 \\ b_2 \end{bmatrix} = \begin{bmatrix} \mathbf{K} \\ \mathbf{L} \end{bmatrix} \begin{bmatrix} (\Delta x_d^*)^2 \\ (\Delta x_d^*) \\ 1 \end{bmatrix} \quad (16)$$

$$\mathbf{K}^T = \begin{bmatrix} -2.224 \times 10^{-4} & 5.371 \times 10^{-2} & -3.3943 \\ 2.732 \times 10^{-3} & -0.6604 & 40.5 \\ -3.792 \times 10^{-3} & 1.042 & -56.55 \end{bmatrix} \begin{bmatrix} \text{Pe}^2 \\ \text{Pe} \\ 1 \end{bmatrix} \quad (17)$$

$$\mathbf{L}^T = \begin{bmatrix} 4.333 \times 10^{-5} & -1.075 \times 10^{-2} & 0.7054 \\ -5.216 \times 10^{-4} & 0.1272 & -8.051 \\ 6.422 \times 10^{-4} & -0.162 & 11.67 \end{bmatrix} \begin{bmatrix} \text{Pe}^2 \\ \text{Pe} \\ 1 \end{bmatrix} \quad (18)$$

The parameter for the rate of $\text{Sh}_{\text{av},n}$ convergence (α) is expressed through Eqs. (19) - (21). The value of α is independent of Pe and decreases with r^* and Δx_d^* . For a given r^* , α becomes less dependent of the droplet spacing as the spacing increases.

$$\alpha = \alpha_{\max} - (\alpha_{\max} - \alpha_{\min}) \left(1 - e^{\frac{(1.96 - \Delta x_d^*)}{1.8}} \right) \quad (19)$$

$$\alpha_{\max} = 0.0735 r^{*-2.6} \quad (20)$$

$$\alpha_{\min} = 0.1708 r^{*-1.036} \quad (21)$$

As introduced earlier in Section 2.3, Sherwood number is a function of Pe , r^* , Δx_d^* , and Sc . The present study provides Sh correlations for the first three parameters. In order to showcase the applicability of these correlations to different length scales, two simulations were performed. Both of them had the same Pe , r^* , Δx_d^* ,

and Sc that correspond to 174.22, 0.29, 3.92, and 0.633 respectively. One of the simulations had dimensions that were scaled-up by a factor of 2 based on the dimensions of the other simulation. Hence, these simulations can be differentiated with their hydraulic diameters: $d_h = 0.51$ and 1.02 mm. In order to keep the non-dimensional parameters of r^* and Δx_d^* the same, r and Δx_d were changed proportional to d_h . Contrarily, u_m was changed inversely proportional to d_h to keep Pe the same.

Fig. 9 shows local Sh results at different length scales $d_h = 0.51$ and 1.02 mm with the same non-dimensional parameters (Pe, r^* , and Δx_d^*) that was specified in the paragraph above. Since two simulations had two different channel lengths, down the channel position (x) was non-dimensionalized with the respective value of d_h . This plot depicts the same case presented in Section 4.1 for mesh independency study. The non-dimensional position $x/d_h = 21.56$ refers to the position of the fifth droplet. The close-up view in Fig. 9 is shown with the intent of showing how well two results match. This plot proves that the non-dimensionalization was performed correctly since results match at the same non-dimensional parameters despite different length scales.

The parametric investigation of Sh dependence on Sc is kept out of the scope of this work. In order to provide a rough estimate for the Sh-Sc dependency, two simulations were performed. Both simulations had the conditions of $Pe = 139.13$, $r^* = 0.29$, and $\Delta x_d^* = 3.92$. However, they had different Sc values of 0.633 and 0.722. The former value corresponds to conditions that all other simulations in this work had (fully humidified air at 80 °C). The latter value corresponds to fully humidified conditions at 50 °C. The aforementioned conditions led to similar $Sh_{av,n}$ profiles with 7% difference. Further studies can map the effect Sc at different values of Pe, r^* , and Δx_d^* .

4.4 Effect of Sherwood Number Variations on Fuel Cell Performance

The effect of multiple droplets on the cell performance is a function of the O_2 concentration drop from the channel mean (C_m) to the average interfacial value ($C_{int,av} = W^{-1} \int_{y=0}^W C_{int} dy$). The difference between C_m and $C_{int,av}$ adversely affects the cell voltage for a given current density. Fig. 10 shows C_m and $C_{int,av}$ at the current density of 1.5 A cm⁻² and, Pe of 26.16 and 174.22. C_m varies from 3.83 to 1.92 mol m⁻³ at the inlet and outlet, respectively. This variation does not depend on Pe since the stoichiometry is kept at 2. Fig. 10 focuses on the first 21.00 mm of the channel. Values of $C_{int,av}$ vary with Pe while r^* and Δx_d^* are 0.29 (0.15 mm) and 3.92 (2.00 mm), respectively. Both plots of $C_{int,av}$ have the same fully developed concentration drop upstream of the first droplet ($x < 3.00$ mm) and the average slope of C_m , $C_{int,av}$ at $Pe = 26.16$ follows the trend of the fully developed region in between the droplets. In the immediate proximity of droplets, $C_{int,av}$ decreases with respect

to the fully developed line. $C_{\text{int,av}}$ at $Pe = 174.22$ shows an increase with respect to the fully developed line. This results in a smaller concentration drop at the interface and thereby, an increase in the performance of the fuel cell.

The varying O_2 concentration drop at the GDL-channel interface affects the voltage drop in the fuel cell. Eq. (22) determines the decrease in voltage due to a decrease in O_2 concentration in the cathode catalyst layer (CL). The calculations focus on the channel outlet since the transport losses will be more severe there. The O_2 concentration at the CL was calculated by applying reductions to the O_2 concentration at the channel outlet (1.92 mol m^{-3}) at the interface and within the GDL. Eq. (22) uses Faraday's (F_c) and universal gas (R_u) constants with respective values of $96,485 \text{ C mol}^{-1}$ and $8.314 \text{ J mol}^{-1} \text{ K}^{-1}$. C_1 and C_2 represent two values of O_2 concentration in the cathode CL. The resulting voltage change due to this concentration difference is expressed with ΔV_{1-2} .

$$\Delta V_{1-2} = \frac{R_u T}{F_c} \ln \left[\frac{C_2}{C_1} \right] \quad (22)$$

For the calculations of O_2 concentration loss within the GDL, the PEMFC was assumed to have a dry, $200 \mu\text{m}$ thick GDL with an O_2 diffusivity of $D_{O_2\text{-GDL}} = D_{O_2\text{-air}}/1.5$ [55]. The Fick's diffusion of O_2 in the GDL causes 0.65 mol m^{-3} of an O_2 concentration drop at the current density of 1.5 A cm^{-2} .

For the calculation of interfacial O_2 concentration drop, three values of Sh are used: the fully developed: $Sh_{\text{FD}} = 3.349$, the minimum: $Sh_{\text{av},\infty} = 2.925$, and the maximum: $Sh_{\text{av},\infty} = 7.438$. The minimum and maximum values of $Sh_{\text{av},\infty}$ were not obtained at the Pe value equivalent to the current density of 1.5 A cm^{-2} . However, due to possible flow maldistribution in parallel flow channels, it is safe to consider the possibility that those Sh values may exist at the aforementioned current density. Values of Sh were transformed into the interfacial O_2 concentration drop (ΔC) with the following dimensionalization process: $\Delta C = j (Sh D_{O_2\text{-air}} d_h^{-1})^{-1}$ in which j was $6.663 \times 10^{-2} \text{ mol m}^{-2} \text{ s}^{-1}$.

The voltage drop due to the inclusion of fully developed interfacial O_2 resistance is 9.0 mV . Since the minimum $Sh_{\text{av},\infty}$ is lower than the fully developed value, this case creates an additional voltage drop of 1.6 mV . The maximum $Sh_{\text{av},\infty}$ reduced the original voltage drop 5.3 mV which is a 58.9% reduction in the voltage drop. The findings suggest that the variations in Sh can lead to non-negligible variations in cell voltage at the current density of 1.5 A cm^{-2} . This effect can be stronger at higher current densities.

CONCLUSIONS

The oxygen (O_2) transport resistance at the gas diffusion layer (GDL) – air flow channel interface was investigated numerically. The numerical model was built for steady convective mass transport in a three dimensional air flow channel that was isolated from the neighboring channels. Multiple droplet-shaped obstructions were placed in a row within the channel and centered along the channel width to simulate the droplet-emergence aspect of two-phase flow in air channels. The effect of droplets on the interfacial transport resistance was parametrically characterized locally in the flow direction using a non-dimensional approach. This allowed the applicability of results to different length scales and conditions. The input parameters in this non-dimensional problem were Péclet number (varied due to changes in superficial air velocity), non-dimensional droplet size and uniform spacing. The values for these parameters were selected in such a way that the droplet adhesion and spherical shape assumptions were not violated.

The interfacial O_2 transport resistance was expressed by the non-dimensional Sherwood number (Sh). The values of Sh were averaged on the interfacial region between two consecutive droplets. The result provides the average Sh value within the droplet spacing (Sh_{av}). The local values of Sh_{av} were compared against the fully developed value (3.349) that corresponds to a channel without any droplets. The following are the main findings from the current work:

- A critical range of Péclet number was found ($52.32 < Pe < 69.75$) to identify asymptotic Sh_{av} values ($Sh_{av,\infty}$) switching from being lower than the fully developed value to being higher.
- Sherwood number values do not change significantly (less than 11%) throughout droplets in a row when Péclet number is below about 69.
- A set of correlations were built to provide Sh_{av} between any two consecutive droplets as a subset of a multiple droplet array. The correlations are a function of droplet number, Péclet number, non-dimensional droplet size and uniform droplet spacing. The maximum error of these correlations among 999 data points is 11%. Since these correlations are using non-dimensional parameters, they can be used at different length scales.
- The maximum $Sh_{av,\infty}$ was obtained to be 7.438 at the Péclet number, non-dimensional droplet radius and uniform spacing of 174.22, 0.29, and 3.92, respectively. The minimum $Sh_{av,\infty}$ was obtained to be

2.925 at the Péclet number, non-dimensional droplet radius and uniform spacing of 13.16, 0.39, and 1.96, respectively.

- In modeling the performance of a PEMFC, inclusion of interfacial O_2 transport resistance was predicted to decrease the calculated voltage by 9.0 mV at a cell current density of 1.5 A cm^{-2} . It was also noted that the voltage can additionally decrease by 1.6 mV due to the minimum Sh and relatively increase by 5.3 mV due to the maximum Sh as compared to a channel with no droplets.
- Film flow is another important flow pattern that occurs in the fuel cell channels. It will be investigated in the future for its impact on the interfacial O_2 transport resistance.

Acknowledgements

This work was conducted in the Thermal Analysis, Microfluidics, and Fuel Cell Laboratory in the Mechanical Engineering Department at Rochester Institute of Technology. Support for this project was provided by the U.S. Department of Energy under award number: DE-EE0000470.

REFERENCES

- [1] U. Pasaogullari, C.Y. Wang, *J. Electrochem. Soc.* 151 (2004) A399. DOI: 10.1149/1.1646148
- [2] U. Pasaogullari, C.-Y. Wang, K.S. Chen, *J. Electrochem. Soc.* 152 (2005) A1574. DOI: 10.1149/1.1938067
- [3] K. Tüber, D. Póca, C. Hebling, *J. Power Sources* 124 (2003) 403. DOI: 10.1016/S0378-7753(03)00797-3
- [4] *Handbook of Fuel Cells: Fundamentals, Technology, and Applications*, Wiley, Chichester, England; New York, 2003.
- [5] H. Liu, P. Li, K. Wang, *Int. J. Hydrog. Energy* 38 (2013) 9835. DOI: 10.1016/j.ijhydene.2013.05.159
- [6] H. Wu, X. Li, P. Berg, *Int. J. Hydrog. Energy* 32 (2007) 2022. DOI: 10.1016/j.ijhydene.2006.09.046
- [7] P.K. Das, X. Li, Z.-S. Liu, *Int. J. Hydrog. Energy* 35 (2010) 2403. DOI: 10.1016/j.ijhydene.2009.12.160
- [8] G.-S. Kim, P.C. Sui, A.A. Shah, N. Djilali, *J. Power Sources* 195 (2010) 3240. DOI: 10.1016/j.jpowsour.2009.11.110
- [9] J. Park, K. Min, *J. Power Sources* 216 (2012) 152. DOI: 10.1016/j.jpowsour.2012.05.054
- [10] H. Heidary, M.J. Kermani, *Int. J. Hydrog. Energy* 38 (2013) 5485. DOI: 10.1016/j.ijhydene.2012.10.020
- [11] A. Casalegno, F. Bresciani, G. Groppi, R. Marchesi, *J. Power Sources* 196 (2011) 10632. DOI: 10.1016/j.jpowsour.2011.08.094
- [12] R.M. Darling, S. Khateeb, *J. Power Sources* 243 (2013) 328. DOI: 10.1016/j.jpowsour.2013.06.015
- [13] J.M. Sergi, S.G. Kandlikar, *Int. J. Hydrog. Energy* 36 (2011) 12381. DOI: 10.1016/j.ijhydene.2011.06.092
- [14] R. Banerjee, S.G. Kandlikar, *J. Power Sources* 247 (2014) 9. DOI: 10.1016/j.jpowsour.2013.08.016
- [15] Y. Ding, X. Bi, D.P. Wilkinson, *Chem. Eng. Sci.* 100 (2013) 445. DOI: 10.1016/j.ces.2012.11.007
- [16] L. Chen, T.F. Cao, Z.H. Li, Y.L. He, *Int. J. Hydrog. Energy* 37 (2012) 9155. DOI: 10.1016/j.ijhydene.2012.01.101
- [17] X.G. Yang, F.Y. Zhang, A.L. Lubawy, C.Y. Wang, *Electrochem. Solid-State Lett.* 7 (2004) A408. DOI: 10.1149/1.1803051
- [18] F.Y. Zhang, X.G. Yang, C.Y. Wang, *J. Electrochem. Soc.* 153 (2006) A225. DOI: 10.1149/1.2138675
- [19] A. Theodorakakos, T. Ous, M. Gavaises, J.M. Nouri, N. Nikolopoulos, H. Yanagihara, *J. Colloid Interface Sci.* 300 (2006) 673. DOI: 10.1016/j.jcis.2006.04.021
- [20] I.S. Hussaini, C.-Y. Wang, *J. Power Sources* 187 (2009) 444. DOI: 10.1016/j.jpowsour.2008.11.030
- [21] K. Nishida, S. Tanaka, S. Tsushima, S. Hirai, in: *ECS Trans.* (2011) 419. DOI: 10.1149/1.3635576
- [22] J.M. Sergi, Z. Lu, S.G. Kandlikar, in: *Proceeding Seventh Int. Conf. Nanochannels Microchannels Minichannels.* (2009) 303. DOI: 10.1115/ICNMM2009-82140
- [23] K. Takada, Y. Ishigami, S. Hirakata, M. Uchida, Y. Nagumo, J. Inukai, H. Nishide, M. Watanabe, *Electrochemistry* 79 (2011) 388. DOI: 10.5796/electrochemistry.79.388
- [24] M.I. Rosli, D.J. Borman, D.B. Ingham, M.S. Ismail, L. Ma, M. Pourkashanian, *J. Fuel Cell Sci. Technol.* 7 (2010) 061015. DOI: 10.1115/1.4001353
- [25] K. Nishida, T. Murakami, S. Tsushima, S. Hirai, *J. Power Sources* 195 (2010) 3365. DOI: 10.1016/j.jpowsour.2009.12.073
- [26] D. Spornjak, A.K. Prasad, S.G. Advani, *J. Power Sources* 170 (2007) 334. DOI: 10.1016/j.jpowsour.2007.04.020
- [27] D. Contreau, C. Bonnet, D. Funfschilling, M. Weber, S. Didierjean, F. Lapicque, *Fuel Cells* 10 (2010) 520. DOI: 10.1002/face.200900167
- [28] K.S. Chen, M.A. Hickner, D.R. Noble, *Int. J. Energy Res.* 29 (2005) 1113. DOI: 10.1002/er.1143
- [29] A.D. Le, B. Zhou, H.-R. Shiu, C.-I. Lee, W.-C. Chang, *J. Power Sources* 195 (2010) 7302. DOI: 10.1016/j.jpowsour.2010.05.045
- [30] S.C. Cho, Y. Wang, K.S. Chen, *J. Power Sources* 210 (2012) 191. DOI: 10.1016/j.jpowsour.2012.03.033
- [31] T.C. Wu, N. Djilali, *J. Power Sources* 208 (2012) 248. DOI: 10.1016/j.jpowsour.2012.02.026
- [32] E. Gauthier, T. Hellstern, I.G. Kevrekidis, J. Benziger, *ACS Appl. Mater. Interfaces* 4 (2012) 761. DOI: 10.1021/am201408t
- [33] P.K. Das, A. Grippin, A. Kwong, A.Z. Weber, *J. Electrochem. Soc.* 159 (2012) B489. DOI: 10.1149/2.052205jes
- [34] A. Esposito, A.D. Montello, Y.G. Guezennec, C. Pianese, *J. Power Sources* 195 (2010) 2691. DOI: 10.1016/j.jpowsour.2009.11.078
- [35] E.C. Kumbur, K.V. Sharp, M.M. Mench, *J. Power Sources* 161 (2006) 333. DOI: 10.1016/j.jpowsour.2006.04.093
- [36] L. Chen, Y.-L. He, W.-Q. Tao, *Int. J. Heat Mass Transf.* 60 (2013) 252. DOI: 10.1016/j.ijheatmasstransfer.2012.11.024
- [37] C. Qin, D. Rensink, S.M. Hassanizadeh, S. Fell, *J. Electrochem. Soc.* 159 (2012) B434. DOI: 10.1149/2.004205jes
- [38] S.C. Cho, Y. Wang, K.S. Chen, *J. Power Sources* 206 (2012) 119. DOI: 10.1016/j.jpowsour.2012.01.057

- [39] K. Seidenberger, F. Wilhelm, J. Haußmann, H. Markötter, I. Manke, J. Scholta, J. Power Sources (2013) 628. DOI: 10.1016/j.jpowsour.2013.02.006
- [40] A. Turhan, K. Heller, J.S. Brenizer, M.M. Mench, J. Power Sources 160 (2006) 1195. DOI: 10.1016/j.jpowsour.2006.03.027
- [41] M. Koz, S.G. Kandlikar, ECS Trans. 50 (2012) 183. DOI: 10.1149/05002.0183ecst
- [42] C.Y. Wang, Z.H. Wang, K.S. Chen, J. Power Sources 94 (2001) 40. DOI: 10.1016/S0378-7753(00)00662-5
- [43] K.T. Jeng, C.Y. Wen, L.D. Anh, J. Mech. 23 (2007) 275. DOI: 10.1017/S1727719100001325
- [44] H. Hassanzadeh, X.G. Li, J.J. Baschuk, S.H. Mansouri, Int. J. Energy Res. 35 (2011) 670. DOI: 10.1002/er.1717
- [45] M. Koz, S.G. Kandlikar, J. Power Sources 243 (2013) 946. DOI: 10.1016/j.jpowsour.2013.06.075
- [46] M. Venkatraman, S. Shimpalee, J.W. Van Zee, S.I. Moon, C.W. Extrand, Int. J. Hydrog. Energy 34 (2009) 5522. DOI: 10.1016/j.ijhydene.2009.04.039
- [47] J.P. Owejan, J.J. Gagliardo, J.M. Sergi, S.G. Kandlikar, T.A. Trabold, Int. J. Hydrog. Energy 34 (2009) 3436. DOI: 10.1016/j.ijhydene.2008.12.100
- [48] P. Gopalan, S.G. Kandlikar, J. Electrochem. Soc. 160 (2013) F487. DOI: 10.1149/2.030306jes
- [49] S. Um, C.Y. Wang, J. Power Sources 125 (2004) 40. DOI: 10.1016/j.jpowsour.2003.07.007
- [50] L. Wang, H. Liu, J. Power Sources 180 (2008) 365. DOI: 10.1016/j.jpowsour.2008.01.091
- [51] P.J. Pritchard, Fox and McDonald's Introduction to Fluid Mechanics, 8th Edition, 8 edition, John Wiley & Sons, Inc., 2010.
- [52] R. Krishna, J.A. Wesselingh, Chem. Eng. Sci. 52 (1997) 861. DOI: 10.1016/S0009-2509(96)00458-7
- [53] V.V. Dharaiya, S.G. Kandlikar, J. Heat Transf. 134 (2012) 20911. DOI: 10.1115/1.4004934
- [54] R.K. Shah, Fundamentals of Heat Exchanger Design, John Wiley & Sons, Hoboken, NJ, 2003.
- [55] M.M. Mench, Fuel Cell Engines, John Wiley & Sons, 2008.

List of Figures

Fig. 1: a) A schematic of the simulated channel with uniformly spaced droplets; b) The dimensions for the channel and bipolar plate cross section. The bipolar plate was not simulated.

Fig. 2: The meshed domain as the result of mesh independency study. The droplet radius is $r = 0.15$ mm and uniform droplet spacing $\Delta x_d = 1.00$ mm. The first droplet is moved closer to the inlet for easier demonstration.

Fig. 3: The down-the-channel-averaged Sherwood number ($Sh_{av,n}$) for the non-dimensional droplet radius $r^* = 0.39$, uniform droplet spacing $\Delta x_d^* = 1.96$, and Péclet number $Pe = 26.16$ and 69.75 .

Fig. 4: The effect of helical air flow path on local O_2 concentration profile: The Péclet number $Pe = 261.42$, non-dimensional droplet radius $r^* = 0.20$, non-dimensional uniform droplet spacing $\Delta x_d^* = 3.92$. a) The streamline originated at $x = 0$, $y = 0.10$ mm, $z = 0.15$ mm; b) Oxygen concentration normalized with the mean concentration (C/C_m) at a yz cross section from the symmetric half of the channel. C_m is 3.520 and 3.394 mol m^{-3} at $x = 0$ and 12.00 mm, respectively.

Fig. 5: The effect of creeping air flow over the droplets on local O_2 concentration profile: The Péclet number $Pe = 13.16$, non-dimensional droplet radius $r^* = 0.39$, and non-dimensional uniform droplet spacing $\Delta x_d^* = 1.96$. a) Streamlines originated at $x = 0$, $0 < y < 0.70$ mm, $z = 0.19$ mm; b) Oxygen concentration normalized with the mean concentration (C/C_m) at a yz cross section from the symmetric half of the channel. C_m is 3.626 and 3.464 mol m^{-3} at $x = 0$ and 11.50 mm, respectively.

Fig. 6: The effect of uniform non-dimensional droplet spacing (Δx_d^*) on the Sherwood number (Sh) and down-the-channel-averaged Sh ($Sh_{av,n}$) at the Péclet number, $Pe = 69.75$, non-dimensional droplet radius, $r^* = 0.29$: a) Sh against the non-dimensional distance parameter ($x^* = (x - x_{d,1}) / \Delta x_d$); b) $Sh_{av,n}$ against the droplet number in the flow direction (n).

Fig. 7: The quadratic behavior of $Sh_{av,\infty}$ with the non-dimensional radius (r^*) at the Péclet number of a) $Pe = 13.16$ and b) $Pe = 26.16$.

Fig. 8: The linear behavior of $Sh_{av,\infty}$ with the non-dimensional radius (r^*) at the Péclet number of $Pe = 121.91$.

Fig. 9: The applicability of Sherwood number (Sh) correlations at different length scales. The Péclet number (Pe), non-dimensional droplet radius (r^*) and uniform spacing (Δx_d^*) are 174.22 , 0.29 , and 3.92 , respectively. Results of Sh exactly match at different hydraulic diameters (d_h) since non-dimensional variables are the same.

Fig. 10: The comparison between O_2 concentration drop from the channel cross sectional mean value (C_m) to interfacial average one ($C_{int,av}$) at two different Péclet numbers: 26.16 and 174.22. Non-dimensional droplet radius and uniform spacing are 0.29 (0.15 mm) and 3.92 (2.00 mm), respectively.

List of Tables

Table 1: The normalized deformation of a droplet characterized as the normalized mismatch between the semi-principal axes $(r_y - r_x) / r$, and tabulated for sets of droplet radius (r) and superficial air velocity (u_m). The emboldened values indicate cases that were not simulated due to normalized deformations larger than 0.105.

Table 2: Mapping from current density (i) to superficial air velocity (u_m) and Péclet number (Pe) for fully humidified air at 80 °C.

Table 3: Comparison of experimental observation [35] and numerical prediction for droplet attachment/detachment. The required drag force to break adhesion was calculated to be 35.9 μ N based on the data by Das et al. [33].

Table 4: Asymptotic values of down-the-channel-averaged Sherwood number ($Sh_{av,\infty}$) at the uniform non-dimensional droplet spacing of $\Delta x_d^* = 1.96$ as a function of non-dimensional droplet radius (r^*) and Péclet number (Pe).

Table 5: Asymptotic values of down-the-channel-averaged Sherwood number ($Sh_{av,\infty}$) at the uniform non-dimensional droplet spacing of $\Delta x_d^* = 3.92$ as a function of non-dimensional droplet radius (r^*) and Péclet number (Pe).

Tables 6: Asymptotic values of down-the-channel-averaged Sherwood number ($Sh_{av,\infty}$) at the uniform non-dimensional droplet spacing of $\Delta x_d^* = 9.80$ as a function of non-dimensional droplet radius (r^*) and Péclet number (Pe).

Table1

<i>r</i> [mm]	<i>u_m</i> [m s ⁻¹]				
	4.24	7.41	8.48	10.59	15.89
0.10	0.007	0.023	0.030	0.047	0.105
0.12	0.010	0.031	0.040	0.063	0.141
0.15	0.016	0.048	0.062	0.097	0.219
0.18	0.025	0.077	0.101	0.158	0.356
0.20	0.037	0.113	0.148	0.230	0.518

Table2

i [A cm ⁻²]	0.05	0.10	0.20	0.30	0.40	0.70	0.80	1.00	1.50
u_m [m s ⁻¹]	0.80	1.59	2.12	3.18	4.24	7.41	8.48	10.59	15.89
Pe	13.16	26.16	34.88	52.32	69.75	121.91	139.51	174.22	261.42

Table3

Droplet Height/Cord	Experimental Re_{ch}	Experimental Condition	Simulated θ [°]	Simulated Re_{ch}	Numerical Drag [μN]	Predicted Condition
1.16	513	Attach	133.0	513	13.3	Attach
1.19	411	Attach	134.5	411	10.2	Attach
1.22	206	Attach	135.4	206	3.6	Attach
1.09	718	Attach	130.1	718	19.6	Attach
1.08	920	Attach	130.0	920	29.8	Attach
1.33	1136	Detach	139.0	710	36.2	Detach

Table4

<i>r</i> [*]	Pe								
	13.16	26.16	34.88	52.32	69.75	121.91	139.51	174.22	261.42
0.20	3.118	3.110	3.114	3.152	3.236	3.794	4.200	4.536	5.292
0.24	3.050	3.045	3.062	3.153	3.335	4.409	4.830	5.525	
0.29	2.967	2.983	3.023	3.232	3.638	5.601	6.198	6.995	
0.35	2.927	2.973	3.007	3.164	3.590	6.106	6.792		
0.39	2.925	3.005	3.085	3.335	3.743				

Table5

<i>r</i> [*]	Pe								
	13.16	26.16	34.88	52.32	69.75	121.91	139.51	174.22	261.42
0.20	3.235	3.227	3.227	3.242	3.281	3.628	3.838	4.391	6.113
0.24	3.199	3.198	3.205	3.249	3.349	4.191	4.680	5.861	
0.29	3.162	3.171	3.190	3.300	3.550	5.356	6.130	7.438	
0.35	3.150	3.186	3.207	3.268	3.511	5.759	6.498		
0.39	3.153	3.217	3.271	3.392	3.547				

Table6

<i>r</i> [*]	Pe								
	13.16	26.16	34.88	52.32	69.75	121.91	139.51	174.22	261.42
0.20	3.303	3.301	3.301	3.305	3.316	3.396	3.444	3.573	4.074
0.24	3.290	3.289	3.291	3.305	3.380	3.540	3.663	4.021	
0.29	3.274	3.279	3.286	3.321	3.393	3.968	4.299	5.171	
0.35	3.270	3.289	3.302	3.327	3.397	4.225	4.681		
0.39	3.270	3.301	3.329	3.388	3.443				

Figure 1a
a)

Figure 2

Figure3

Figure 4a
a)

Figure 4b

$x = 0$ mm

$x = 12.00$ mm

Figure 5a
a)

Figure 5b $x = 0$ mm

$x = 11.50$ mm

Figure6a

a)

Figure6b

Figure7a

a)

Figure7b

Figure8

Figure9

Figure10