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Some Aspects of Mass Transfer within the Passages of Fuel Cells

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SOME ASPECTS OF MASS TRANSFER WITHIN THE PASSAGES OF FUEL CELLS

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ABSTRACT

This paper describes a numerical heat/mass transfer analysis for planar and square duct geometries, found in certain fuel cells. Both developing and fully-developed scalar transport are considered. The solution to the heat/mass transfer problem is presented in terms of normalized conductance as a function of the driving force and wall Reynolds/Peclet numbers.

INTRODUCTION

Motivation for the present work

At the present time there is substantial interest in the development of computer programs to perform calculations of the electrochemical and thermomechanical performance of fuel cells. Several groups are now developing computational fluid dynamics (CFD) codes. The present author has analysed planar solid-oxide fuel cells. Figure 1 is a schematic of the problem; the air passages are in the form of rectangular ducts while the fuel passages are such that they may be treated as planar (2-D) ducts. A problem associated with the use of CFD codes to design stacks relates to the excessive mesh sizes required to capture fine detail within fluid regions for large-scale industrial units. One solution proposed, by the author and his co-workers (Beale et al., 2003) is a distributed resistance analogy, whereby diffusion terms are replaced by terms of the form $\dot{q}'' = \alpha \Delta \phi$, where α is a volumetric conductance (Patankar and Spalding, 1972) corresponding to the better-known area conductance, g ,

$$\dot{q}'' = -\Gamma \frac{\partial \phi}{\partial y} = g(\phi_w - \phi_b) \quad (1)$$

where ϕ_w and ϕ_b are the value of enthalpy or mass fraction, at the wall and in the bulk of the fluid. The adoption of a rate equation for the diffusion flux allows for a very substantial reduction in required computational effort, owing to much coarser meshes being sufficient for the purpose-at-

Figure 1. Schematic of fuel cell under consideration.

hand. In addition numerous other 'presumed flow schemes', based on non-CFD approaches also require that heat/mass transfer coefficients be prescribed.

Values of the conductance, g , may be obtained from (a) theoretical analysis, (b) experimental data or appropriate empirical correlation, or (c) from the results of fine-scale numerical calculations. Preliminary efforts were based on assuming constant conductance from the standard analyses for fully-developed flow and heat transfer with negligible mass transfer, g^* . Shah (1978) provides a review of the results for ducts of various cross-sectional areas. In this paper the implications of such an assumption is investigated in detail.

In fuel cells, heterogeneous chemical reactions on the electrode surfaces lead to sources and sinks in the continuity and species (mass fraction) equations. Under the circumstances there is a driving force, B , so that the mass flux, $\dot{m}''$, is given by

$$\dot{m}'' = gB \quad (2)$$

with

$$B = \frac{\phi_b - \phi_w}{\phi_w - \phi_t} \quad (3)$$

where ϕ_t is the value at the so-called transferred substance state, or T-state (Spalding, 1963; Kays and Crawford, 1980; Mills, 2001). The heat/mass transfer problem may be considered as characterized by the normalized conductance g/g^* expressed as a function of B . An alternative

representation is sometimes found in terms of a blowing parameter, b , such that

$$\dot{m}'' = g^* b \quad (4)$$

In the present work, heat and mass transfer in planar and square ducts, found in fuel cells such as illustrated in Fig. 1 is analysed using numerical means. The extension to other aspect ratios and geometries is straightforward. The scope of the problem is confined to Fickian/Fourier type diffusion, for laminar flow with constant properties, negligible dissipation, and $Le = 1$. Thermo-diffusion (Soret and Dufour) effects are neglected.

PROBLEM DESCRIPTION

Previous Work

Fluid Mechanics. Most of the available analyses are for plane ducts. Berman (1953) was among the first to obtain an approximate analytical solution for laminar flow with blowing/suction at the wall, and reported velocity and pressure distributions for fully-developed flow in a plane channel with injection/suction at two walls, Fig. 2(a). The case of injection at a single wall, Fig. 2(b), has been considered by Jorne (1982).

Heat/mass transfer. Sherwood et al. (1965) performed machine calculations for mass transfer of brine solutions by reverse osmosis, based on Berman's velocity profile for planar geometry, Fig. 2(a). They correlated a concentration polarization, defined as $\phi_w/\phi_b - 1$ as a function of non-dimensional distance. One of the co-authors (Dressner, 1964) also obtained a correlation for fully-developed mass transfer.

Raithby (1972) considered heat transfer under conditions of both constant wall temperature and constant wall flux for the planar geometry of Fig. 2(a). Several other authors have conducted related studies. For rectangular ducts, Shah (1978) has reviewed the data for both fluid flow and heat transfer under conditions of zero mass transfer. Yuan et al. (2001) performed heat transfer calculations for rectangular and trapezoidal cross-sections under various boundary conditions.

Present Scheme

In this study, numerical calculations were performed for mass transfer in ducts corresponding to the cases shown in Fig. 2 (a-c). The equations solved are,

$$\frac{\partial \rho}{\partial t} + \text{div } \rho \vec{u} = 0 \quad (5)$$

$$\frac{\partial (\rho \vec{u})}{\partial t} + \text{div}(\rho \vec{u}; \vec{u}) = -\text{grad } p + \text{div } \mu \text{ grad } \vec{u} \quad (6)$$

$$\frac{\partial (\rho \phi)}{\partial t} + \text{div } \rho \vec{u} \phi = \text{div } \Gamma \text{ grad } \phi \quad (7)$$

Figure 2. Three problems considered in the present study.

A rectilinear mesh was concentrated towards the wall(s) using a geometric progression. The inlet flow conditions were either: (i) presumed constant $u = u_0$, $v = v_w$, $\phi = \phi_0$ etc.: or else (ii) downstream values at $x = L/2$ were back-substituted, as outlined in Beale and Spalding (1998), to provide fully-developed inlet profiles; u -values were scaled by the bulk velocity ratio $\bar{u}_0/\bar{u}(x)$. For fully-developed scalar transport, it is presumed that the non-dimensional quantity $\tilde{\phi} = (\phi - \phi_w)/(\phi_w - \phi_t)$ and hence the driving force $B = \tilde{\phi}_b$ is constant, thus inlet values of ϕ (including the wall value ϕ_w which is not constant for this problem) may be computed from those at $x = L/2$. and scaled to yield the desired bulk inlet value. Downstream, at $x = L$, a constant pressure was prescribed.

At the wall, the mass fraction at the T-state was presumed to be zero for the considered phase (i.e. unity for the transferred substance). For heat transfer the T-state enthalpy was set to a reasonable value; (while the choice of ϕ_t affects the magnitude of $\dot{q}''$ it does not change Nu for a given $\dot{m}''$). The computer code PHOENICS was selected to perform the calculations described below.

Figure 3. Pressure coefficient for case (a).

Figure 4. Velocity profiles for case (a), $Re = 100, Re_w = 1$.

Figure 5. Normalized friction coefficient types (a) and (b).

Figure 6. Scalar polarisation for case (a).

Figure 7. Transfer factor as a function of driving force.

Figure 8. Transfer factor as a function of wall Peclet number.

RESULTS

Figure 3 shows results for pressure coefficient, $c_p = (p_0 - p)/\frac{1}{2}\rho u_0^2$, compared to Berman's (1953) equation. Figure 4 is a similar comparison for cross-wise and stream-wise velocity profiles. It can be seen that agreement is excellent. Figure 5 shows normalized friction coefficient as a function of a wall Reynolds number, defined according to Berman's definition, $Re_w = \rho D_h v_w / 4\mu$. Also shown is the straight line,

$$\frac{c_f}{c_f^*} = 1 - \frac{Re_w}{35} \quad (8)$$

Equation (8) was obtained by differentiating Berman's u -velocity equation. The local Reynolds number, $Re = \rho D_h \bar{u} / \mu$ varies as a function of x , even when the flow is fully-developed since the bulk velocity $\bar{u}(x)$ increases for injection and decreases for suction, however for fully-developed flow c_f , as normalized by $\frac{1}{2}\rho \bar{u}^2$ is constant for a given Re_w .

Figure 6 is a comparison of the present author's work with the calculations of Sherwood et al. (1965) for wall Peclet number, $Pe_w = \rho D_h v_w / \Gamma$ of -2 and -3.7. The results are presented in terms of a 'polarisation', $\hat{\phi} = (\phi_w - \phi_b)/(\phi_b - \phi_t)$, discussed below, as a function of non-dimensional distance, $\xi = \frac{1}{3}(Pe_w^3/Re_0 Sc)x/H$. It can be seen that agreement is good. The reader will note that ξ is negative because of the definition of v_w as being positive for injection. Note that although Sherwood's definition of ξ removes Schmidt number dependence, $Sc = 1$, was presumed throughout this study.

Figure 7 shows normalized conductance, g/g^* plotted as a function of the driving force, B , while Fig. 8 is a similar plot of g/g^* as a function of Pe_w . It can be seen that the form of the graph is similar to that of Fig. 5 but the scale of the variation is much larger than for c_f . The solid line in Fig. 7 is obtained from a Couette flow analysis, for which,

$$\frac{g}{g^*} = \frac{\ln(1+B)}{B} = \frac{b}{\exp(b)-1} \quad (9)$$

Agreement is remarkably good. For the solid lines in Fig. 8, the further assumption has been made that,

$$b = aPe_w \quad (10)$$

Values of a obtained using a regression analysis are given in Table 1. These were fitted to the three sets of data shown in Fig. 8, which are also reproduced in Table 1. The solid lines in Fig. 8 are the results of the regression analysis and it can be seen that they correlate well with the numerical data.

Table 1. Heat/mass transfer data.

Pe_w	Fig. 2(a) $a = 0.5187$		Fig. 2(b) $a = 0.4357$		Fig. 2(c) $a = 0.4034$	
	B	g/g^*	B	g/g^*	B	g/g^*
-5	-0.871	2.790	-0.763	2.435	-0.754	2.334
-3	-0.736	1.979	-0.620	1.797	-0.610	1.732
-1	-0.377	1.287	-0.300	1.238	-0.292	1.205
1	0.644	0.754	0.474	0.784	0.448	0.786
3	3.707	0.393	2.473	0.451	2.093	0.505
5	13.036	0.186	7.858	0.236	4.818	0.365

Numerical Accuracy

The computational grid was refined until the following limits were achieved: For $Re_w = 0$, a value of $c_f^* = 24/Re$ was obtained for plane geometry case (a). For $Pe_w = 0.0001$, values of $Nu/Sh = 8.23$ and 5.38 fully-developed flow, constant wall heat flux, and zero blowing (Kays and Crawford, 1980) were obtained for cases (a) and (b) respectively. For case (c) a value of 2.84 was observed. Convergence proved to be readily obtainable for the most part. For large negative values of Pe , however the scalar gradient becomes highly concentrated at the wall necessitating the use of highly concentrated grid in this region. Under these conditions departures from Berman's velocity/pressure profiles were observed.

DISCUSSION

Figure 3 shows that the fluid pressure is affected by inertia and friction. For zero mass transfer, the linear profile $Re_w = 0$ indicates that pressure gradient drives the flow to overcome the skin friction at the wall. For $Re_w > 0$ (injection) the pressure gradient increases; Because $\frac{1}{2}\rho \bar{u}^2$ increases with x , the local pressure decreases and hence the pressure gradient is larger than for $Re_w = 0$. Pressure losses due to mass efflux are important considerations in the headers of fuel cell stacks where they can contribute to flow maldistributions if they are large compared to drag within the cells.

Note from Fig. 5 that the skin friction coefficient actually decreases for injection, due to the slight changes in the parabolic-shaped velocity profile, $u/\bar{u}$, in Fig. 4, as a result of blowing at the wall. (The shear stress τ_w increases with $\bar{u}$ and hence $\partial u/\partial y$, but $\tau_w/\frac{1}{2}\rho \bar{u}^2$ does not). For suction the converse is true, the pressure gradient is reduced, and c_f increases. In this case, unlike for injection, if the duct is sufficiently long an extremum occurs with $dp/dx = 0$, at which point all matter has been removed at the wall. For the current boundary-value problem, the flow was then seen to reverse at this location with in-flow occurring at the 'outlet'. Some authors have observed multiple solutions under these circumstances. In practice such a limit should never be reached, and is avoided numerically by increasing the main-flow Reynolds number. The reader will note that for

strongly negative Re_w , it is actually possible to get negative values of c_p (not shown in Fig. 3) so that the inertial effects are actually larger than the opposing frictional effects and there is an adverse pressure gradient.

Inspection of Fig. 5 reveals that for planar geometry, in the range $-2 \leq Re_w \leq 2$ the deviation from the zero mass transfer form is less than 5%. Thus frictional losses within fuel cells may be reasonably approximated by the zero-blowing form $c_f \propto 1/Re$, provided Re is based on the local velocity $\bar{u}(x)$, and the blowing/suction velocity is not too high. It can also be seen that the results of Berman's linear perturbation theory represent a tangent to the present results at $Re_w = 0$, which lend credence to the results of this work.

Figure 6 is a comparison between the results of the present author and the work of Sherwood et al. for developing scalar transport with fully-developed flow. Also shown are the author's calculations based on the back-substitution process described above, i.e. full-developed scalar transport. It can be seen that the concentration polarisation

$$\hat{\phi} = \frac{\phi_w - \phi_b}{\phi_b - \phi_t} \quad (11)$$

increases linearly up to a maximum corresponding to the full-developed profile. For small (negative values) of Pe_w , the fully-developed condition is rapidly reached whereas at higher values (for suction) a fully-developed condition may not be reached prior to the mass fraction of the transferred substance becoming zero (and that of the considered phase unity).

Sherwood et al. defined a concentration polarisation by the quantity $\phi_w/\phi_b - 1$. The present writer has replaced this with the modified form, Eq. (11) for the results of Fig. 6: For mass transfer with $\phi_t = 0$ (which is the case here) these are to be considered identically equivalent. However there are many situations involving mass transfer, for example reverse-osmosis with incomplete rejection, where $\phi_t > 0$. Moreover for heat transfer, the value of ϕ_t is a function of the magnitudes and signs of $\dot{m}''$ and $\dot{q}''$ (and vice-versa). Under the circumstances the presently-defined polarisation is invariant, whereas that of Sherwood et al. is not (Brian, 1965). Tests showed that regardless of the choice of ϕ_b , identical B and $\hat{\phi}$ characteristics were obtained. The heat and mass transfer problems are to be considered entirely equivalent, as was found to be the case. The solution for normalised enthalpy and concentration polarisation are identical for both developing and fully-developed cases. The scalar boundary values do not affect the g/g^* profile, although the mass flow rate, $\dot{m}''$ does. For cases Fig. 2(b-c), however, the boundary conditions for mass transfer are considered as different for heat transfer, since although there is zero mass flux at all but one boundary, heat transfer would occur. Only the results for the former are presented here in this paper.

It can readily be shown that.

$$B = \frac{\hat{\phi}}{\hat{\phi} + 1} \quad (12)$$

so that the use of a modified scalar polarisation may be considered as functionally equivalent to a driving force in mass transfer theory.

Figure 7 shows values of the normalized conductance g/g^* , as a function of the mass transfer driving force B for fully-developed scalar transport. While from a formal point-of-view g/g^* may be considered a function of B or b , from a practical position it is probably more useful to correlate the conductances with the wall Peclet number (or Reynolds number for skin friction). It is clear from the latter that variation in the heat/mass transfer factor is much larger than is the case for skin friction. This must be accounted for when considering transport phenomena in fuel cells.

Standard mass transfer techniques appear well suited to this class of internal-flow problem, despite the pressure variations. There is a remarkable agreement between the Couette flow analysis and the numerical results. This is perhaps not entirely surprising: The Couette flow analysis is just the solution of a one-dimensional convection-diffusion equation,

$$\dot{m}'' \phi(y) - \Gamma \frac{d\phi}{dy} = \dot{m}'' \phi_t \quad (13)$$

This is the same equation that forms a basis for the finite-volume method. It is discussed in detail in the book by Patankar (1980). The result that $b = 2Pe_w$ corresponds to the notion that the bulk value is the arithmetic mean of the wall and the centre-line values, (rather than at the edge of a boundary layer for a standard analysis). Inspection of the parabolic v -velocity profile in Fig. 4 shows departures from linearity. Thus values of the coefficient a provided in Table 1 are a function of geometry. If these are not available, however, a quite reasonable assumption would appear to be to compute the heat/mass transfer factor, g , from the zero mass-transfer value based on the Couette flow model with $a = 0.5$.

Some writers refer to mass transfer wall boundary conditions with the right-side of Eq. (13) set to zero. For strong suction $v_w \ll 0$, this condition is approximately true, due the highly non-linear nature of the convection-diffusion equations (Patanekar, 1980). What should be understood is that a flux $\rho v_w (\phi_t - \phi_p)$, where ϕ_p is the in-cell value, is prescribed for both injection and suction; in the former case as a linearised source term, and in the latter case as a fixed source (in order to avoid the creation of negative coefficients). Many authors presume 'no-slip' boundary conditions to prevail for the stream-wise u -velocity, i.e. $\tau_w = \mu \partial u / \partial y|_w$. (NB: For constant v_w , $\partial v / \partial x|_w = 0$). Saffman (1971) discusses the conditions for slip boundary conditions to occur for porous media related mass transfer,

and many fuel cells involve porous electrode assemblies. In the light of the 1-D convection-diffusion analysis a potentially superior momentum-equation prescription would be to presume an effective shear stress of the form, $\tau_w = C\mu\partial u/\partial y$ where C is computed as a function of Pe_w from the exact solution for 1-D convection-diffusion, the so-called exponential scheme, Patankar (1980). This would preserve the diffusive no slip condition in the limit $Re_w \rightarrow 0$, while correctly accounting for the non-linearity of the missing convection and diffusion fluxes at the wall, eg. for the case of strong suction.. Because of these factors, it is also worth entertaining the future use of higher-order schemes than the hybrid scheme which forms the basis for the code used to perform the calculations presented in this study.

The back-substitution process for the inlet values, allows for fully-developed flow for arbitrary geometry to be prescribed upstream. Previous authors were confined to using Berman's velocity profile to generate a fully-developed flow field. This clearly would not be appropriate for the cases shown in Figs. 2(b-c).

CONCLUSIONS

Numerical studies of fluid flow, heat and mass transfer in the passages of fuel cells have identified a number transport phenomena. The effects of injection are to decrease non-dimensional friction, heat and mass transfer conductances, while increasing the pressure gradient. Suction has the opposite effect. Under most conditions a fully-developed situation is attained; although for large negative wall Peclet numbers it is possible that fully-developed condition will never be achieved. Heat and mass transfer conductances are significantly altered by injection/suction at the wall(s), the influence on the friction coefficient is less major. For many ducts it would appear that a reasonable engineering approximation for the heat/mass transfer conductance can be obtained from a simple Couette-flow analysis.

NOMENCLATURE

D_h	Hydraulic diameter (m)
g	Conductance $\frac{\Gamma\partial\phi/\partial y _{y=H}}{(\phi_b - \phi_w)}$
L	Length (m)
u	Stream-wise velocity (m/s)
v	Cross-wise velocity (m/s)
H	Height, half-height (m)
$\dot{m}''$	Rate of mass transfer (kg/m ² s)
p	Pressure (Pa)
$\dot{q}''$	Rate of transfer of ϕ /m ²

$\dot{q}'''$ Rate of transfer of ϕ /m³

Greek letter symbols

ϕ	Scalar variable
Γ	Exchange coefficient (kg/ms)
μ	Viscosity (kg/ms)
ρ	Density (kg/m ³)
τ	Shear stress (N/m ²)

Non-dimensional numbers

B	Driving force $\frac{\phi_b - \phi_w}{\phi_w - \phi_T}$
b	Blowing parameter $\frac{\dot{m}''}{g^*}$
c_f	Friction factor $\frac{\mu\partial u/\partial y _{y=H}}{\frac{1}{2}\rho u^2}$
c_p	Pressure coefficient $\frac{p_0 - p}{\frac{1}{2}\rho u_0^2}$
Re	Reynolds number $4\frac{\rho D_h u}{\mu}$
Pe_w	Wall Peclet number $\frac{\rho D_h v_w}{\Gamma}$
Le	Lewis number $\frac{\Gamma_h}{\Gamma_m}$
$\hat{\phi}$	Modified polarisation $\frac{\phi_w - \phi_b}{\phi_b - \phi_t}$
$\tilde{\phi}$	Non-dimensional scalar $\frac{\phi - \phi_w}{\phi_w - \phi_t}$
ξ	Non-dimensional distance $\frac{1}{3} \frac{Pe_w^3}{Re_0 Sc} \frac{x}{H}$

Superscripts

* Zero mass transfer

Subscripts

0	Inlet condition
b	Bulk
f	Fluid
h	Heat transfer
m	Mass transfer
t	Transferred substance state
w	Wall

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