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Calculation procedure for mass transfer in fuel cells

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Abstract

An analysis of mass transfer losses, or concentration over-potentials in fuel cells is provided. An elementary theory, based on an equivalent film thickness, as proposed in some texts, is derived. This is followed by a more rigorous theoretical treatment of mass transfer theory, for which the mass transfer factor is obtained as a function of the driving force. The solution for the driving force is derived, for the well-known one-dimensional convection–diffusion problem. It is shown that mass transfer in planar and square geometries approximates this idealised situation. A linearised theory, appropriate for low mass flow rates is also presented. The methodology is illustrated using the simple example of a solid oxide fuel cell (SOFC). It is shown that the simplified theory is only applicable for very dilute binary mixtures. A step-by-step procedure for computing mass transfer in fuel cells is detailed, together with a discussion of the scope and range of application of the results.

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Keywords: Fuel cells; Transport phenomena; Mass transfer; Concentration polarisation; Over-potential; Diffusion

1. Introduction

When current is drawn from a fuel cell, the external voltage, V , may be written as

$$V = E - i''r - \sum \eta \quad (1)$$

where E is the ideal or Nernst potential, i'' the current density¹, r the resistance, and η are sometimes referred to as ‘over-potentials’ or ‘polarisations’.

The performance of fuel cells is generally accepted to be reduced by kinetics (activation), Ohmic resistance and mass transfer (concentration effects). At low current density, activation reduces the voltage, at intermediate voltages, Ohmic losses within the electrolyte are dominant, while at high current densities, necessary if fuel cells are to generate large sources of power, mass transfer effects predominate. Fig. 1 shows a current–voltage distribution curve typical of many hydrogen fuel cells. In this paper, we are concerned with the mass transfer effects, which predominate at high current densities. While numerous texts on fuel cells are available today, many of these are somewhat vague on the mathematical details needed to calculate mass transfer effects. The

purpose of this paper is to provide the reader with a reasonably simple yet comprehensive method by which he or she may estimate these factors.

Historically, chemists and chemical engineers, have considered mass transfer problems based on molar concentrations, x_i , see for example the book by Bird et al. [2]. Using this latter convention, diffusion losses may be introduced into Eq. (1) in the generic form

$$\eta = \pm \frac{RT}{n\nu F} \ln \left(\frac{x_b}{x_w} \right) \quad (2)$$

where x_b and x_w are the molar concentrations in the bulk of the fluid and near the wall, n an integer which is a function of the stoichiometry of the reaction(s), ν the valence of the ionic conductor, and F is the Faraday’s constant.

The theoretical foundation for the present manuscript follows the work of Spalding [3]. This is based on mass fraction, m_i and mass flux relative to mass-average velocity, j_i'' . A justification for the use of these widely adopted definitions is that in the conservation laws of continuum mechanics, it is mass (rather than mole numbers) momentum and energy which are conserved, and indeed mass-based formulations and methodologies and codes abound in the engineering community. Mass-based and mole-based approaches are easily inter-converted as follows:

$$m_i = x_i \frac{M_i}{M} \quad (3)$$

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¹ In this manuscript, the convention of Jacob [1] whereby a ‘dot’ represents a time derivative, and a ‘dash’ a space derivative, is adopted.

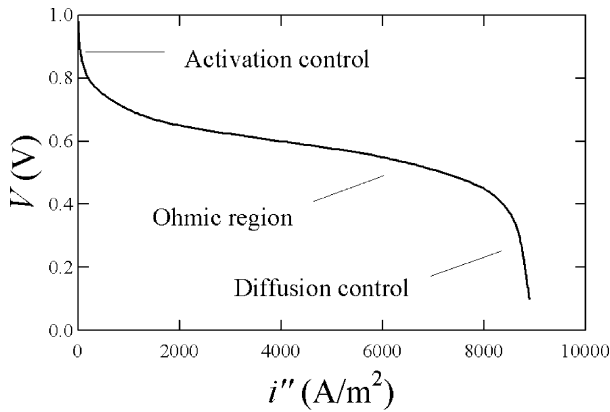


Fig. 1. Typical current–voltage distribution curve, for a hydrogen fuel cell.

where the mixture molecular weight is just

$$M = \sum x_i M_i = \frac{1}{\sum (m_i / M_i)} \quad (4)$$

In the present formulation, the diffusion losses may, thus be expressed as

$$\eta = \frac{RT}{nvF} \ln \left(\frac{m_b}{m_w} \right) \quad (5)$$

for reactants, and

$$\eta = \frac{RT}{nvF} \ln \left(\frac{m_w}{m_b} \right) \quad (6)$$

for products, where m_b and m_w are bulk and wall values for mass fraction. The purpose of this paper is to derive expressions which allow for Eqs. (5) and (6) to be expressed analytically. The terms ‘over-potential’ or ‘polarisation’ are widely used and not entirely unambiguous. Moreover, the term RT/nvF , serves very little purpose for a fundamental analysis. Therefore, we shall define a mass transfer factor as $\ln(m_b/m_w)$, for reactants, and $\ln(m_w/m_b)$, for products. Determination of the mass transfer factors for the components of the fuel and air mixtures, is the goal of this work.

1.1. Simplified analysis based on equivalent film theory

Some authors [4,5] have argued that diffusion effects in fuel cells may be modelled using terms of the form:

$$\eta = \frac{RT}{nvF} \ln \left(1 - \frac{i''}{i''_{\max}} \right) \quad (7)$$

where i''_{\max} is a ‘limiting current density’.

A typical argument proceeds as follows [6]: the rate of mass transfer may be represented by Fick’s law

$$\dot{m}'' = \rho u = \Gamma \frac{dm}{dy} \quad (8)$$

where $\Gamma = \rho D$ is an exchange coefficient. It is generally useful to introduce the concept of a mass transfer conductance, g , by the following rate equation:

$$\dot{m}'' = g(m_w - m_b) \quad (9)$$

Faraday’s law may be written as

$$\dot{m}'' = \pm \frac{Mi''}{vF} \quad (10)$$

The current density is, therefore, given by

$$i'' = \pm \frac{Fv}{M} g(m_w - m_b) \quad (11)$$

For wall suction, the maximum possible current density is obtained when $m_w \rightarrow 0$

$$i''_{\max} = \mp \frac{Fv}{M} g m_b \quad (12)$$

Elementary mass transfer analyses employ an ‘equivalent-film-theory’ to obtain

$$g = \frac{\Gamma}{\delta} \quad (13)$$

where δ is the thickness (length-scale) of an imaginary film. Since by Eq. (13), the conductance, g , is constant

$$\frac{m_w}{m_b} = 1 - \frac{i''}{i''_{\max}} \quad (14)$$

and Eq. (7) is obtained, identically. We shall refer to Eq. (7) as the ‘simplified approach’ in the remainder of the text.

This analysis is not recommended by the present writer for a number reasons: (i) for high mass transfer/current densities, the conductance, g , may be a function of i'' , and not constant; (ii) Fick’s law has been written in terms of absolute rather than relative velocity; (iii) the notion of a conductance is generally introduced in a form such that the diffusion flux, $j'' = \Gamma(dm/dy)$, may be replaced by $g(m_w - m_b)$, an expression for the mass flux, $\dot{m}'' = \rho u$, requires the introduction a mass transfer driving force, or equivalent. This is discussed further below. On a more practical note, the equivalent film model does not, in any way, correspond to the physical reality of flow and mass transfer within the passages of a fuel cell, which are typically ducts of rectangular, or other, cross-section.

2. Elements of mass transfer theory

2.1. Definition of the driving force and blowing parameter

The present section follows that in [3,7], the latter of which contain reviews of Bird et al. [2], Treybal [8], Sherwood et al. [9], and other works. The methodology has been advanced in a number of more recent texts [10,11]. In this paper, the theory is further refined for application to fuel cells.

In fuel cells, heterogeneous chemical reactions occur on electrode surfaces. These lead to sources and sinks in the continuity and species (mass fraction) equations. The mass flux, \dot{m}'' , is given by

$$\dot{m}'' = gB \quad (15)$$

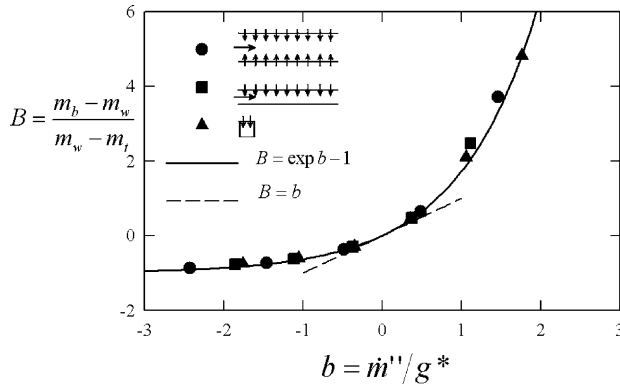


Fig. 3. Driving force vs. normalised mass flux.

The compression of these data onto a single curve is due to the choice of non-dimensional parameter(s), and would not occur if the abscissa were non-dimensionalised in the form of a Sherwood number

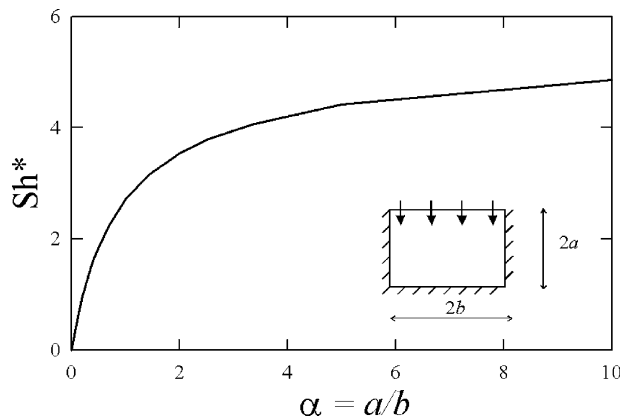
$$Sh = \frac{gD_h}{\Gamma} \quad (26)$$

where D_h is a hydraulic diameter [10]. The reader will appreciate that although, the results of the numerical data are in reasonably close agreement with Eq. (25), values of Sh^* are quite different for the three cases; with Sh^* equal to (a) 8.23, (b) 5.38, (c) 2.84 [13]. Fig. 4 shows values of Sh^* for rectangular ducts, $2a \times 2b$, typical of many fuel cell designs as a function of aspect ratio, $\alpha = a/b$. This was obtained from Table 44 in the book by Shah and London [13]. Eq. (25) combined with Eqs. (21) and (22) is the recommended method for computing mass transfer effects in fuel cells. It is referred to below as the ‘present method’.

2.3. Linearised method for low mass flux

Fig. 3 also shows the low mass flow-rate solution, i.e. $\lim_{|m''| \rightarrow 0} B = 0$, for which it may readily be shown that

$$\frac{g}{g^*} = 1 \quad (27)$$

Fig. 4. Sh^* for a rectangular duct. Adapted from Shah and London [13].

$$B = b \quad (28)$$

This is referred to below as a ‘linearised solution’, and may only be applied to situations for which $b \ll 1$, a situation which occurs in many fuel cells operating at present-day current densities.

3. Example: a solid oxide fuel cell with H_2 as fuel

3.1. Problem

The proposed methodology will be presented by means of a quantitative example. The simple case of a solid oxide fuel cell (SOFC), with H_2 as fuel is considered. Oxidation takes place at the anode surface:



and reduction takes place at the cathode surface:



The Nernst equation for a solid oxide fuel cell may be written in terms of molar fractions as

$$E = E^0 + \frac{RT}{2F} \ln \left(\frac{x_{H_2} x_{O_2}^{0.5}}{x_{H_2O}} \right) + \frac{RT}{4F} \ln P_a \quad (31)$$

Thus, $\nu = 2$ (for O_2), and $n = 1, 2, 1$ for H_2, O_2, H_2O , respectively. The Nernst equation may easily be converted to a mass fraction form:

$$E = E^0 + \frac{RT}{2F} \left[\ln \left(\frac{m_{H_2} m_{O_2}^{0.5}}{m_{H_2O}} \right) - \frac{1}{2} \ln(M_a) - 0.4643 \right] + \frac{RT}{4F} \ln P_a \quad (32)$$

where

$$\ln \left(\frac{M_{H_2} M_{O_2}^{0.5}}{M_{H_2O}} \right) \approx \ln(2) + \frac{1}{2} \ln(32) - \ln(18) = -0.4643 \quad (33)$$

Assume the oxidant to be a mixture of O_2 and atmospheric nitrogen. Since only O_2 is transferred at the cathode, $m_t = 1$, Fig. 5. Fig. 6 shows the corresponding situation at the anode, if the fuel is pure hydrogen, the reaction may be treated as a ‘simple chemical reaction’



If the weight of the electrons is neglected, $r = 8$. Thus, from Eq. (17) $m_t = -1/8$ for H_2 , and for H_2O , $m_t = 9/8$. On the anode side, for H_2 , Eq. (21) applies whereas for H_2O it is Eq. (22) which must be used.

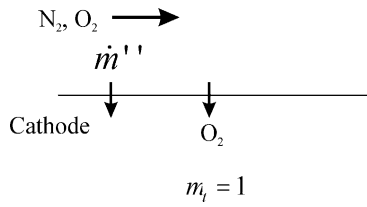


Fig. 5. Mass transfer at cathode.

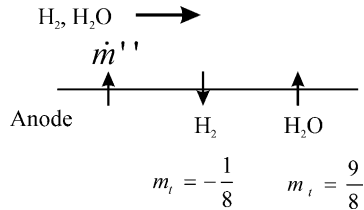


Fig. 6. Mass transfer at anode.

3.2. Solution

Figs. 7 and 8 are plots of the mass transfer factor versus blowing parameter, b , for O_2 on the cathode side. Fig. 7 is a comparison of the present and simplified methods, for suction b is negative and for O_2 $m_b > m_w$ and $m_w > m_t$. The simplified method, Eq. (7) has been plotted in a functionally

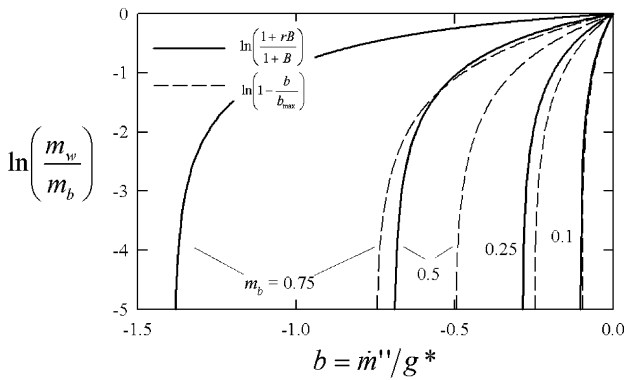


Fig. 7. Present method compared with simplified method for oxygen (cathode).

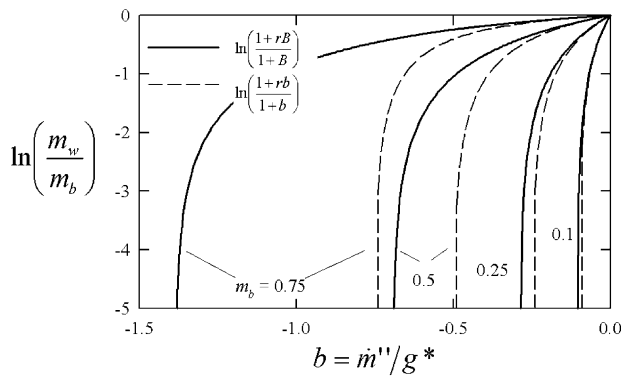


Fig. 8. Present method compared with linearised method for oxygen (cathode).

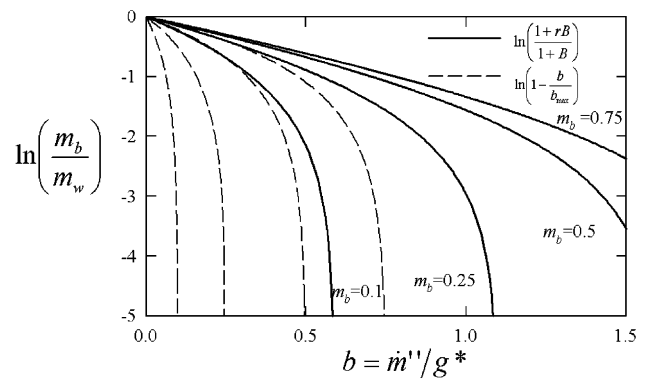


Fig. 9. Present method compared with simplified method for hydrogen (anode).

similar form by substituting for the independent variable according to

$$\frac{b}{b_{\max}} = \frac{\dot{m}''}{\dot{m}''_{\max}} = \frac{i''}{i''_{\max}} \quad (35)$$

where $b_{\max} = -gm_b$ (g occurs in both numerator and denominator of Eq. (35), and is assumed constant in Eq. (13) for the equivalent film thickness theory; therefore it does not enter the calculations). Fig. 8 shows a comparison of the present analysis with that obtained using the linearised method.

Figs. 9–11 show anode side results. Fig. 9 shows a comparison of the simplified and present methods for H_2 on the anode side, for the fuel-side there is blowing and b is positive, with $m_t > m_w > m_b$ for H_2 . Fig. 10 shows a comparison for H_2 between the present and linearised analyses. Fig. 11 shows a similar comparison for H_2O .

3.3. Discussion

The results of Fig. 7 suggest that the simplified approach agrees with the present procedure only when m_b is very small. Inspection of Fig. 7 reveals that unless the O_2 mass fraction is very small, the simplified methodology is not in agreement with the present method, even at very low values

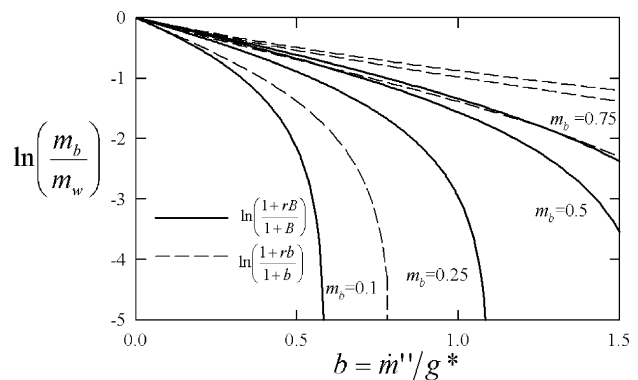


Fig. 10. Present method compared with linearised method for hydrogen (anode).

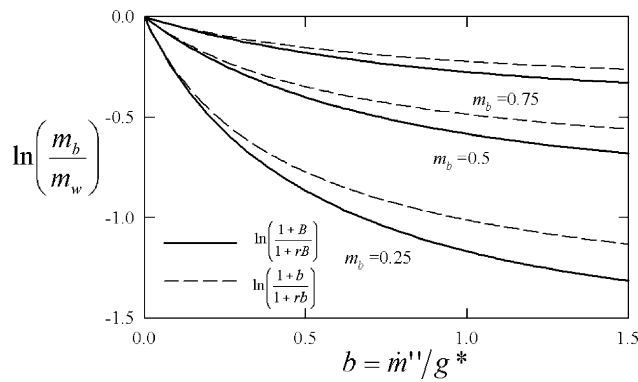


Fig. 11. Present method compared with linearised method for water (anode).

of the mass flux, \dot{m}'' , corresponding to low current densities. Thus, this approach would appear only to be beneficial if the air were highly oxygen-lean, a situation which is unlikely to be found in modern fuel cells, which typically employ atmospheric or enriched air. Of course, it is true that the conductance may be redefined as $\tilde{g} = g/(m_t - m_w)$, so that $\dot{m}'' = \tilde{g}/(m_w - m_b)$. That however, does not fundamentally change the argument.

Larminie and Dicks [4] noted problems with the simplified analysis, and recommended the use of an empirical relation of the form

$$\eta = A \ln \left(1 - \frac{i''}{i''_{\max}} \right) \quad (36)$$

where values of the constant A differ from RT/nvF by almost an order of magnitude. There should be no need to employ empirical relations, the existing theory is sufficient to adequately describe the situation. For H_2 on the anode side, Fig. 9, the effect of the T-state value not being unity is such that the simplified method is in complete disagreement with the present approach, even when m_b is very small.

The results of Figs. 8 and 10 do, however, suggest that there is good agreement between the present method and the linearised method for all values of m_b , provided the mass flux \dot{m}'' (i.e. b) is sufficiently small, it can be seen that the two sets of O_2 curves are in agreement as $b \rightarrow 0$, for large b there are significant departures. The trend is repeated in Fig. 11, for H_2O . Water unlike H_2 and O_2 is being produced rather than consumed by the reaction, and hence there is no limiting current/mass flux. Rather the build-up of water has decreasing impact of the mass transfer-limiting effects on the analysis. Of course, this does not mean the production of H_2O is limitless, since the current density is limited by the mass transfer of H_2 and O_2 . When computing the cell potential in Eq. (1) the sum of the three contributions, H_2 , O_2 and H_2O must be considered.

For many fuel cells, the linearised approach will suffice, but this will not be the case as the limiting current (mass flux) is approached and \dot{m}'' increases. Thus, the linearised analysis will not be applicable at high current densities, and the user

should always perform sample calculations to obtain trial values of b prior to using the linearised method. The present analysis generates meaningful results at all mass transfer rates. It is, therefore, recommended. While it is possible to construct more complex problems, this simple example has served to illustrate the important principles of the analysis.

4. Method for calculating rate of mass transfer in fuel cells

The analysis can readily be used to estimate mass transfer in fuel cells, at all mass flow rates (current densities), low or high. The following approach is suggested.

- (1) Estimate g^* for both the anode and cathode geometries under consideration (this is required to compute b for both air and fuel sides). For many duct geometries, under conditions of fully-developed flow, these are widely published in the form of zero mass transfer Sherwood (Nusselt) numbers, see for example Fig. 4. Hence, compute g^* and $b = \dot{M}i''/\nu F g^*$.
- (2) Compute the driving force $B(b)$. Best practice would be for this to be obtained from measured experimental data, or detailed numerical calculations for the particular geometry/flow conditions under consideration. If this is not available, Eq. (25), should be used as an estimate.
- (3) Compute the T-state value from the stoichiometry, and hence compute the mass transfer factors (i.e. polarisations, η) given the bulk value, using Eq. (21) or (22).
- (4) Repeat the above for all participating species for the given working fluid (fuel or air) and sum the terms as per Eq. (1).

5. General discussion

The mass-based conductance, g , is by and large, equivalent to the molar conductance the symbol, $k^\bullet = g/M$, in Bird et al. [2] (the radical is to remind the reader that k is not constant). Reference [7] notes the mole numbers are not preserved in many chemical reactions, and suggests this as a reason for the use of a mass-based formulation. The present author's position is that provided the mixture-molecular weight at the wall and in the bulk are nominally equal, $M_w = M_b$, molar-based and mass-based formulations should be entirely equivalent. If, however, $M_w \neq M_b$, the issue of replacing local values for the diffusion fluxes by global (i.e. integrated) values, $g(m_w - m_b)$ and $k^\bullet(x_w - x_b)$, may not render identical results.

In this paper, the example of a SOFC with H_2 as fuel was given. The present method may readily be extended to other fuel cells, involving different fuels, more complex heterogeneous and/or homogeneous reactions, thermal radiation, etc. Kays and Crawford [10] suggest that the present analysis is appropriate for non-dilute, non-binary mixtures provided all

diffusion coefficients are equal

$$\Gamma_{12} = \Gamma_{13} = \dots \Gamma_{ij} = \Gamma = \rho D \quad (37)$$

This situation would appear to be reasonably true for the high-temperature gases in SOFCs. It may also be approximately true for lower temperature PEMFC's. However, the analysis is not valid for liquid-based electrolytic solutions and other possible scenarios. Wilke [15] provides an approximate method for computing Γ_{ij} for non-binary mixtures, for a given driving force, which is exact for dilute mixtures, and has enjoyed widespread use.

It might be argued, that there are situations for which the present analysis does not hold, and for which there is no other recourse than to solve the Stefan–Maxwell equations for the multi-component system. Knuth [14] considers circumstances under which it is prudent to substitute ordinary diffusion for the Stefan–Maxwell system of equations for non-binary non-dilute systems where $\Gamma_{ij} \neq \Gamma_{ik}$. Coffee and Heimerl [16] considered five approximate solutions to the Stefan–Maxwell equations, for laminar flames, including [15] and noted surprisingly minor differences in computed values. Amali et al. [17] considered Stefan–Maxwell and Fickian diffusion in porous media and observed differences of 4–10%. Thus, while the use of Fick's law, and the important additional step of replacing the gradient diffusion term with a rate term, may not necessarily be 'correct' in the strict sense for non-dilute multi-component mixtures, it does offer a reasonable balance between mathematical rigour, and computing effort.

There are also a number of other important factors to consider when analysing mass transfer in fuel cells. For example the present approach must be modified if large property variations occur; for example the Schmidt number may vary as a result of temperature gradients in the passages of the fuel cell. Surface diffusion and Knudsen diffusion may be present, and 'ordinary' diffusion is not sufficient to characterise these processes. While the importance of Stefan–Maxwell, surface, and Knudsen diffusion is not to be understated, this should not be used as an excuse to justify the use of over-simplified methods, of questionable value in place of the present approach. There is clearly range of applications, where the present method will give a good estimate of behaviour. The goal of this work was to provide a reasonably simple, yet versatile methodology by which the fuel cell designer could estimate the impact of mass transfer on their design.

In this paper, the focus has been on the analysis of mass transfer in gas-flow channels of fuel cells. Many fuel cells also contain gas diffusion and electrode layers in the form of consolidated porous media. Many methods consider diffusion by defining an effective diffusion coefficient, with

$$\Gamma_{\text{eff}} = \Gamma \frac{\varepsilon}{\tau} \quad (38)$$

where ε is the void fraction and τ is a tortuosity or length-scale (normalised). While, such an approach may

well approximate within-phase diffusion in gas diffusion layers; in electrode layers it is also necessary to compute bulk-to-wall mass transfer, by calculating a driving force, B , as described above, since mass transfer can only occur in the presence of a concentration gradient between the bulk of the fluid and the wall. Thus, it is again necessary to obtain Sh^* (or Nu^*), for the porous media and also, if possible, the B versus b characteristic curve, prior to applying the methodology above.

6. Conclusions

An analysis based on the well-established concepts of mass transfer driving force, B , and transferred substance state was adapted to provide a method for estimating the mass transfer factors (concentration polarisations) in fuel cells. The method is based on knowledge of B as a function of the non-dimensional mass flux (i.e. current density), b , together with data for the zero mass transfer conductance, g^* , obtained from an appropriate Sherwood number correlation, Sh^* , for the geometry under consideration.

It was demonstrated that the use of a simple equivalent falling-film method, to estimate mass transfer is unreliable, even for situations where the rate of mass transfer is very small. A linearised analysis does provide meaningful results for small current densities, corresponding to $|b| \ll 1$, although, it does not appear to offer any major advantages over the present analysis, which is valid for both low and high mass flow rates. A methodology whereby calculations on mass transfer factors in practical fuel cell geometries may be performed. This has the advantage of being in a form compatible with methods used elsewhere in the analysis of transport phenomena in engineering equipment.

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