IN-PLANE DIFFUSION IN FLAT-PLATE SOLID OXIDE FUEL CELLS

Asbjørn Solheim

SINTEF Metallurgy, N-7034 Trondheim, Norway

ABSTRACT

Flat-plate SOFCs are supplied with interconnector plates with channels for the gas transport and ribs for the electrical contact. Utilization of the area underneath the rib is depending on in-plane diffusion of reactants through the electrode. The diffusion-limited current density underneath the rib was modelled, and the results are given in dimensionless form.

INTRODUCTION

In flat-plate SOFC concepts, the electrical contact between the individual cells is provided by interconnector plates supplied with channels for gas transport and ribs for electrical contact. Since the interconnector material is gas tight, the current-producing efficiency of the region underneath the interconnector rib is depending upon in-plane diffusion of gaseous reactants and products through the porous electrodes, as illustrated in Figure 1. The reaction site at or near the electrolyte/electrode boundary "sees" a lower reactant concentration underneath the interconnector rib than in the gas channel, and hence, the current density becomes lower in the rib area. This problem has received little attention in the open literature.

The purpose of the present work was to derive some simple guidelines for estimating the efficiency of the interconnector rib area as a function of the different physical and geometrical parameters involved, in order to produce a more complete basis for optimizing the unit cell geometry - especially the shape and dimensions of the interconnector. So far, only diffusion in binary gas systems has been treated (hydrogen cells), due to the problems encountered in computing diffusional fluxes in a multicomponent gas mixture.

THEORY

Assuming that the activation overvoltages can be linearized and expressed as apparent resistances, and neglecting the crossplane diffusion overvoltage as well as current density variations due to in-plane resistance in the electrodes, the current density in the gas channel (outside the interconnector rib) is given by (please refer to the symbol list at end of paper)
whereas the current density at any point underneath the rib can be expressed as
\[ i_x = \frac{E_0^{\text{rev}} - U}{\sum r} \]

Neglecting the flow in the z- and y- directions (Figure 1), the in-plane flow of oxygen through the cathode at a position x becomes
\[ j_{O_2(x)} = -\frac{D_{\text{eff}}(x)}{RT} \cdot \frac{dY_{O_2}}{dx} + j_{O_2(x)}Y_{O_2} \text{ [mol m}^{-2}\text{s}^{-1}] \]

The last term of Eq. [3] can be neglected, as it will always give less than about 20 percent error, which is probably less than the uncertainty in estimating the effective diffusion coefficients.

The effective diffusion coefficient is composed of the ordinary diffusion coefficient (\(D_{12}\)) and the Knudsen diffusion coefficient (\(D_{1K}\)), corrected for the electrode porosity (\(\theta\)) and the tortuosity factor (\(\tau\)),
\[ D_{\text{eff}} = \frac{D_{12}D_{1K}}{D_{12} + D_{1K}} \cdot \frac{\theta}{\tau} \]

In SOFC, the effective diffusion coefficient will typically be about one order of magnitude lower than the ordinary diffusion coefficient.

The flow is related to the current density underneath the rib by
\[ j_{O_2(x)} = \frac{1}{4F\delta_c} \int_i w_a \, dx \]

Eqs. [3] and [5] are related through Eq. [2]. This system of equations cannot be solved analytically. However, a useful parameter can easily be derived assuming constant current density underneath the rib, as shown in the following.
Let the theoretical "penetration distance" \( L_{p(c)} \) be defined as the maximum distance the oxygen can diffuse from the edge of the interconnector rib towards its centre before it is consumed by the cell reaction, assuming constant current density \( i_0 \) (Eq. [1]). The oxygen flow at a given position \( x \) see (Eq. [5]) becomes

\[
J_{o_2(x)} = \frac{i_0 (L_{p(c)} - x)}{4F \delta_c}
\]

which can be combined with Eq. [3] to give

\[
\frac{i_0}{4F \delta_c} \int_0^{L_{p(c)}} (L_{p(c)} - x) \, dx = - \frac{D_{eff(c)} P}{RT} \int_0^0 dY_{o_2}
\]

Upon integration we arrive at

\[
L_{p(c)} = \sqrt{\frac{8FD_{eff(c)} P Y_{o_2(0)} \delta_c}{i_0 RT}}
\]

A similar calculation for the anode side gives

\[
L_{p(a)} = \sqrt{\frac{4FD_{eff(a)} P Y_{H_2(0)} \delta_a}{i_0 RT}}
\]

The rib area efficiency (RAE) can be defined as

\[
RAE = \frac{1}{W_{rib}} \int_0 W_a \, dx
\]

Obviously, RAE is a function of \( W_{rib}/L_{p} \). This can not be the only parameter, however, since the current density underneath the rib is not constant. Depending on the gas composition and the cell voltage, the local current density may be more or less sensitive to a change in the gas composition (Eq. [2]).

RESULTS AND DISCUSSION

The calculations were performed on a PC. The electrodes were divided into 5 x 80 elements in the z- and x directions, respectively. The diffusion fluxes in the x- and z-directions were calculated (Eq. [3], neglecting the last term) as well as the concentrations and the fluxes at the boundaries (the reactions were assumed to take
place at the electrolyte/electrode boundaries). The computation was stopped when the crossplane flux from the gas channel to the electrode, the inplane flux across the projection of the rib edge, and the crossplane flux at the electrode/electrolyte boundary were within 0.05 percent. A "standard case" was defined (see Table I below), and the different parameters were varied one at a time, keeping the other parameters constant. The necessary thermodynamic data were taken from JANAF (1).

Table I: Physical and geometrical data used in simulations. The "standard" case was chosen arbitrarily. The parameters were varied one at a time, keeping the other parameters at "standard" magnitude. The two last columns give the symbols used in Figs. 4 and 5 when the actual parameter was varied.

| Parameter                              | "Standard" | Fig. 4 | Fig. 5 |
|----------------------------------------|------------|--------|--------|
| Anode thickness \( \delta_a \) [\( \mu \text{m} \)] | 20         | +      |        |
| Cathode thickness \( \delta_c \) [\( \mu \text{m} \)] | 20         | +      |        |
| Rib half-width \( \mathcal{W}_{rh} \) [\( \text{mm} \)] | 0.5        | \( \Delta \) | \( \Delta \) |
| Diff. coeff., anode \( \mathcal{D}_{\text{eff}(a)} \) [\( \text{m}^2\text{s}^{-1} \)] | \( 2 \cdot 10^{-5} \) | \( \Delta \) | \( \Delta \) |
| Diff. coeff., cathode \( \mathcal{D}_{\text{eff}(c)} \) [\( \text{m}^2\text{s}^{-1} \)] | \( 4 \cdot 10^{-5} \) | \( \Delta \) | \( \Delta \) |
| Internal resistance \( \Sigma_r \) [\( \text{ohm m}^2 \)] | \( 7 \cdot 10^{-5} \) | \( \Diamond \) | \( \Diamond \) |
| Temperature \( T \) [\( \text{K} \)] | 1200       |        |        |
| Total pressure \( P \) [\( \text{Pa} \)] | \( 1.013 \cdot 10^5 \) |        |        |
| \( E_0^{\text{rev}} - U \) [\( \text{V} \)] | 0.2        | Parameter |
| Oxygen molar frac. \( Y_{O_2(0)} \) [\( 1 \)] | 0.2        |        |
| Hydrogen molar frac. \( Y_{H_2(0)} \) [\( 1 \)] | 0.5        |        |

Staggered Gas Channels

Cathode side. With staggered gas channels, the anode/electrolyte/cathode plate "sees" a gas channel on one side and an interconnector rib on the other side. Hence, in the calculations for the cathode, the partial pressure of hydrogen at the anode could be kept constant. Figure 2 shows the local molar fraction of oxygen and the normalized local current density at different values of \( E_0^{\text{rev}} - U \), keeping the parameter \( \mathcal{W}_{rh}/L_{p(c)} \) constant = 1.2. Some normalized current density profiles at different values of \( \mathcal{W}_{rh}/L_{p(c)} \) are given in Figure 3.
Figure 4 shows the rib area efficiency (RAE, Eq. [10]) as a function of the parameter $W_{rh}/L_{p(O)}$. As can be observed, the RAE decreases with decreasing $E_0^{rev-U}$, keeping the factor $W_{rh}/L_p$ constant.

**Anode side.** In the calculations for the anode side, the molar fraction of oxygen was kept constant (0.2). The RAE is given as function of $W_{rh}/L_{p(a)}$ in Figure 5. In this figure, the molar fraction of $H_2$ in the gas channel was kept constant. The effect of changing the hydrogen pressure is shown in Figure 6.

The results in Figs. 4 - 6 could be reasonably well fitted by the equation

$$\text{RAE} = a \left[ a^b + \frac{W_{rh}}{L_p} \right]^{-\frac{1}{b}}$$

$$a = 0.4 + 0.58 \exp \left[ \frac{-1.3}{\ln (Y_0/Y_{eq})} \right] \quad \text{and} \quad b = 2 + (2.2 + 0.375 \varphi) \left( \frac{W_{rh}}{L_{p(O)}} \right)^{1.5}$$

where $Y_{eq}$ is the equilibrium molar fraction of either $H_2$ or $O_2$, i.e., the value of $Y_x$ in Eq.[2] which gives zero current density. The quantity $\varphi$ is defined as

$$\varphi_c = \ln \left( \frac{Y_{O_2(0)}}{Y_{O_2(eq)}} \right) \quad \text{and} \quad \varphi_a = 0.7 \ln \left( \frac{Y_{H_2(0)}}{Y_{H_2(eq)}} \right) + 3.3 \ln (1 - Y_{H_2(0)})$$

for the cathode and the anode, respectively. Eq. [11] is represented by the solid lines in Figs. 4-6.

**Fluctuating Gas Channels**

With fluctuating gas channels, the current is limited by in-plane diffusion at both sides of the anode/electrolyte/cathode layer. It was found that the best way of representing the data was a plot of RAE versus $W_{rh} \left[ L_{p(c)}^{-4} + L_{p(a)}^{-4} \right]^{1/4}$. As shown in Figure 7, the points are close to the staggered channel-data for oxygen diffusion if $L_{p(c)} < L_{p(a)}$ and close to the staggered channels-data for hydrogen diffusion if $L_{p(c)} > L_{p(a)}$. It is assumed that the RAE at 50:50 hydrogen and water can be estimated at a reasonable degree of accuracy from Figure 7.

The effect of variable hydrogen molar fraction is shown in Figure 8. As can be observed, there appears to be a maximum in the RAE at medium molar fractions, which should be taken into account.
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REFERENCE

(1) JANAF Thermochemical Tables, 3rd Ed. (J. Phys. Chem. Ref. Data, Vol.14, Suppl. 1, 1985)

NOMENCLATURE

Roman

$D_{\text{eff}}$ Effective diffusion coefficient [$m^2 s^{-1}$]  
$E_{\text{rev}}$ Reversible cell voltage [V]  
$F$ Faraday's constant [96487 As equiv.$^{-1}$]  
$i$ Current density [$A m^{-2}$]  
$j$ Molar flux [mol $m^{-2}s^{-1}$]  
$L_p$ "Penetration distance" at constant current density (Eqs. [8] and [9]) [m]  
P Total pressure [Pa]  
$R$ Universal gas constant [8.3143 J mol$^{-1}$ K$^{-1}$]  
$R_{AE}$ Rib area efficiency (Eq. [10])  
$T$ Temperature [K]  
$U$ Cell voltage [V]  
$W_{rh}$ Half-width of interconnector rib [m]  
x Distance from edge of interconnector rib [m]  
$Y$ Molar fraction

Greek

$\delta$ Thickness of electrode [m]  
$\varphi$ Dimensionless quantity defined in Eq. [11]  
$\Sigma_r$ Crossplane resistance, including activation overvoltage [ohm $m^2$]  
$\theta$ Electrode porosity  
$\tau$ Tortuosity factor

Sub- and superscripts

$0$ In gas channel (outside rib area)  
a Anode  
c Cathode  
eq At equilibrium (zero current density)  
x At a distance $x$ from edge of rib
Figure 1: In-plane oxygen diffusion path in flat-plate SOFC. A - anode, E - electrolyte, C - cathode.

Figure 2: Normalized current density underneath interconnector rib as a function of $x/L_{p(c)}$ at $W_{rh}/L_{p(c)} = 1.2$. Parameter: $E_o^{rev} - U$ (see text). Oxygen diffusion, staggered gas channels.
Figure 3: Normalized current density underneath interconnector rib as a function of $x/W_{rh}$ at $E_{o}^{rev} - U = 0.2$ V. Parameter: $W_{rh}/L_{p(c)}$ (see text). Oxygen diffusion, staggered gas channels.

Figure 4: Rib area efficiency as a function of $W_{rh}/L_{p(c)}$ for oxygen diffusion, staggered gas channels. Parameter: $E_{o}^{rev} - U$. See Table I for further data.
Figure 5: Rib area efficiency as a function of $W_{rh}/L_{p(a)}$ for hydrogen diffusion, staggered gas channels. Parameter: $E_0^{rev} - U$. See Table I for further data.

Figure 6: Rib area efficiency as a function of the hydrogen molar fraction in the gas channel. Parameter: $W_{rh}/L_{p(a)}$. $E_0^{rev} - U = 0.2$ V.
1.0

$W_{rh}/L_{p}(c) = 2W_{rh}/L_{p}(a)$

$W_{rh}/L_{p}(c) = W_{rh}/L_{p}(a)$

$W_{rh}/L_{p}(c) = 0.5W_{rh}/L_{p}(a)$

$W_{rh}/L_{p}(a) = 0$

$W_{rh}/L_{p}(c) = 0$

Figure 7: Rib area efficiency with fluctuating gas channels.

Figure 8: Rib area efficiency and $W_{rh}/L_{p}$ as a function of the hydrogen molar fraction in the gas channel. "Standard" data (Table I), fluctuating gas channels.