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Short communication

Fuel utilization and fuel sensitivity of solid oxide fuel cells

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1. Introduction

One of the characteristic advantages of solid oxide fuel cell (SOFC) is its excellent fuel flexibility. Conventional fuels such as natural gas, hydrogen, coal gas, legacy fluid fuels, and biomass-derived renewable fuels all have the potential (or have been demonstrated) to be efficiently consumed by a SOFC in a direct or indirect manner. To facilitate the electrochemical conversion of these fuels into electricity in a SOFC, an electrode known as anode is the necessary working component. In developing high-performance anode materials for various fuels, properties such as high electro-catalytic activity, coking resistance and sulfur tolerance are highly preferred and particularly interested. The state-of-the-art Ni-based anode is excellent in catalyzing electrochemical oxidation of fuels but susceptible to coking and sulfur poisoning. Ceramic-based anode is one alternative class of materials that is currently being actively pursued by worldwide SOFC researchers to address issues of coking and sulfur poisoning [1-5]. A successful development of these highly active, anti-coking and sulfur tolerant anodes will substantially reduce the cost of a SOFC system by simplifying the fuel processing subsystems.

Of a practical viewpoint, regardless of the materials of anode, a comprehensive understanding of relationships between fuel flows, fuel utilization (or consumption), oxygen-leakage flux and fuel sensitivity is critically important to the operation of a SOFC. Since the in-stack temperature and current distributions can all be affected

ABSTRACT

Fuel utilization and fuel sensitivity are two important process variables widely used in operation of SOFC cells, stacks, and generators. To illustrate the technical values, the definitions of these two variables as well as practical examples are particularly given in this paper. It is explicitly shown that the oxygen-leakage has a substantial effect on the actual fuel utilization, fuel sensitivity and *V–I* characteristics. An underestimation of the leakage flux could potentially results in overly consuming fuel and oxidizing Ni-based anode. A fuel sensitivity model is also proposed to help extract the leakage flux information from a fuel sensitivity curve. Finally, the "bending-over" phenomenon observed in the low-current range of a *V–I* curve measured at constant fuel-utilization is quantitatively coupled with leakage flux.

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by the fuel utilization, a poor understanding of factors influencing the fuel utilization could lead to a failure of SOFC operation. Unfortunately, very little pertinent information is available in the literature [6]. In this paper, we present from the fundamentals of thermodynamics and electrochemistry the definitions of two important fuel-related process variables commonly used in the operation of SOFC cells, stacks and generators. In particular, fuel sensitivity is discussed in the perspectives of theoretical analysis and practical examples. Finally, the effect of oxygen-leakage flux on the *V*–*I* characteristic of a SOFC operating under a high constant fuel utilization is also examined.

2. Fuel utilization U_f

Fuel utilization represents a fraction of the total fuel input, which is electrochemically oxidized by the oxide-ion current. For the anodic reaction involving fuel-A:

$$A + \frac{z}{2}O^{2-} = AO_{z/2} + ze^{-}$$
(1)

a total of z electrons per A-molecule are transferred to the external circuit. A full utilization of the fuel-A with flux J^o (mol s⁻¹) or flow Q^o (standard l/min, slpm) requires by Faraday's law the total oxide-ion current $I_{O^{2-}}$

$$I_{\Omega^{2-}} = zFJ^0 \tag{2}$$

where *F* is the Faraday's constant, 96 485 C mol⁻¹. For a partial utilization of the fuel-A with flux *J* or flow *Q* at $I_{O^{2-}}$ to a degree of $U_{\rm f}$, the above equation can be changed to:

$$I_{0^{2-}} = zFJU_{\rm f} \tag{3}$$

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The division of Eq. (2) by Eq. (3) gives the mathematical definition of fuel utilization $U_{\rm f}$ at $I_{\rm O^{2-}}$

$$U_{\rm f}\Big|_{I_{0^{2-}}} = \frac{J^{\rm o}}{J}$$
 (4)

The importance of Eq. (3) is depicted by the basic relationship between J (or Q), U_f and $I_{Q^{2-}}$. Take H₂ as an example, the anodic reaction (1) is then replaced by:

$$H_2 + O^{2-} = H_2 O + 2e^-$$
(5)

There are two electrons transferred per H_2 molecule. From Eq. (3), the H_2 flow, Q_{H_2} , in slpm, is given by:

$$Q_{\rm H_2} = \frac{22.4 \times 60 \times I_{\rm O^{2-}}}{2 \times 96485 \times U_{\rm f}} = 0.00697 \times \frac{I_{\rm O^{2-}}}{U_{\rm f}} \quad (\rm{slpm}) \tag{6}$$

The physical meaning of Eq. (6) is that 0.00697 slpm of H₂ will be consumed for every ampere of $I_{0^{2-}}$ involved in the anodic reaction. Such an equivalency is clearly the same for CO because of the same two electrons transferred, but is reduced to one quarter of 0.00697 slpm for CH₄ due to a transfer of eight electrons in the pertinent anodic reaction.

It is important to know that it is $I_{O^{2-}}$ not external load current I_L that should be used in Eq. (4). The factors that determine $I_{O^{2-}}$ include Nernst potential (or EMF) and polarization losses of the cell, load current I_L and oxygen-leakage current I_{lk} . For an oxygen-leakage-free cell, $I_{O^{2-}} = I_L$ whereas for an oxygen-leakage-present cell, I_L is reduced from $I_{O^{2-}}$ by the amount of leakage current I_{lk} , i. e., $I_L = I_{O^{2-}} - I_{lk}$. The nature of oxygen-leakage can be physical, chemical or both. The physical leakage refers to common molecular oxygen transport through physical pores and cracks whereas the chemical leakage relates to ionic oxygen transport by mixed oxide-ion and electron conduction in dense layers of the electrolyte and the interconnection. More importantly, these oxygen-leakages consume fuels without contributing to the external current I_L . Therefore, oxygen-leakage is considered as one of the primary causes for lowered electrical efficiency of a SOFC.

During a practical operation of SOFC generators, it is more convenient to use $I_{\rm L}$ not $I_{\rm Q^{2-}}$ as the process parameter to control fuel flows and fuel utilization. However, it is practically difficult to satisfy the "leakage-free" condition for any SOFC generator in order to validate $I_{\rm Q^{2-}} = I_{\rm L}$. Under this condition, the fuel utilization $U_{\rm f}$ can be modified to correct the presence of a leakage flux $Q_{\rm LK}$ (on the basis of H₂ flow rate) under a constant $I_{\rm L}$ in the form of:

$$U_{\rm f}\big|_{I_{\rm L}} = \frac{J^{\rm o} + Q_{\rm LK}}{J} \tag{7}$$

where Q_{LK} (mol s⁻¹) is directly related to I_{lk} . Theoretically speaking, Q_{LK} decreases with I_L as a result of the decreased P_{O_2} gradient across the electrolyte (and ceramic interconnection) by an applied I_L . In practice, Q_{LK} is conservatively taken as a constant value measured from the same cell under the open-circuit condition where the leakage flux is at the maximum. The experimental procedures of measuring Q_{lk} under the open-circuit condition have been described in detail in [7]. Clearly, the addition of Q_{LK} to Eq. (7) increases the actual U_f for the same J shown in Eq. (4). This information is important to the operation of a SOFC generator because overly consuming fuels by the presence of Q_{LK} could result in a severe oxidation of the Ni-based anode. To give an example, assuming a H₂-fueled SOFC operating at $I_L = I_{O^{2-}} = 20$ A and $U_f = 0.85$, the calculated H₂ flow from Eq. (6) is 0.164 slpm if no leakage is considered. However, if the same H₂ flow were kept for the SOFC with a leakage flux of 0.020 slpm H_2 , the actual U_f would be raised to 97% for the same 20 A current, dangerously close to the boundary at which the Ni-anode is oxidized. To correct the leakage flux Q_{LK} , Eq. (6) can be modified into:

$$Q_{\rm H_2} = \frac{0.00697 \times I_{\rm L} + Q_{\rm LK}}{U_{\rm f}\big|_{I_{\rm I}}} \quad (\rm{slpm})$$
(8)

Taking the same example again, the H₂-flow should be set as 0.188 slpm in order to compensate $Q_{LK} = 0.020$ slpm H₂ so as to yield $U_f = 0.85$ at $I_L = 20$ A. Clearly, $Q_{LK} = 0.00697 I_{lk}$ is assumed for H₂ as a fuel in Eq. (8).

A change in $U_{\rm f}$ will result in a change in the average concentration of fuels over the entire active area of a SOFC. This will further alter the Nernst potential and the concentration-dependent anode polarizations. In general, the voltage of a healthy SOFC decreases with increasing $U_{\rm f}$ due to a reduced Nernst potential and increased anode polarizations, but the resulting electrical efficiency still increases with $U_{\rm f}$. However, if the reduction in cell voltage is too sensitive to the increase in $U_{\rm f}$, the characteristic of higher electrical efficiency at a higher $U_{\rm f}$ could be lost. Therefore, the sensitivity of cell voltage to the change of $U_{\rm f}$ has a great impact on the achievable upper limit of electrical efficiency. In what follows, the definition and applications of fuel sensitivity are particularly described.

3. Fuel sensitivity β

Fuel sensitivity, β , of an operating SOFC is defined as the change in cell voltage, V_c , with respect to the change in U_f when cell temperature T, i_L (current density is used here instead of I_L for convenience) and oxygen utilization U_o are held constant. Mathematically, $\beta = (\partial V_c / \partial U_f)_{T, I_L, U_o}$. It is conceivable that β varies with U_f to a lesser degree at lower U_f but to a higher degree at higher U_f , mainly as a result of logarithmic variation of the Nernst potential with P_{O_2} . Therefore, a typical β – U_f curve has a characteristic of a low but flat β at low end of U_f , and a high but sharp β at high end of U_f . The presence of oxygen-leakage to the cell would make β value even greater and possibly change the shape of a β – U_f curve. Careful examination of these characteristic changes in a β – U_f curve could potentially reveal the hidden Q_{LK} information. A methodology to extract Q_{LK} from a measured β – U_f curve is presented in the following.

3.1. Basic equations

The derivation of one-dimensional (*x*-axis) β -*U*_f equations starts from the basic equation of energy conservation within a SOFC:

$$V_{\rm c}(i_{\rm L},x) = E(i_{\rm L},x) - i_{\rm L}R_{\rm ohm}(x) - \int_{o}^{i_{\rm L}} R_{\rm c}(i_{\rm L},x)di_{\rm L} - \int_{o}^{i_{\rm L}} R_{\rm a}(i_{\rm L},x)di_{\rm L}$$
(9)

where cell voltage $V_c(i_L, x)$, Nernst potential $E(i_L, x)$, areas specific resistance $R_c(i_L, x)$ of the cathode and area specific resistance $R_a(i_L, x)$ of the anode are all i_L - and x-dependent. Ohmic area specific resistance $R_{ohm}(x)$ is only x-dependent. Obviously, $(\partial V_c/\partial U_f)_{T,i_L,U_0}$ depends on how $E(i_L, x)$ and $R_a(i_L, x)$ will vary with i_L and x. For an ideal SOFC, it is reasonable to assume that all area specific resistances are x-independent.

The understanding of the $E(i_L, x)-U_f$ relationship is based on the $E(i_L, x)-(i_L, x)$ profiles. An example of such a profile is shown in Fig. 1 for a 50-cm long and 270-cm² active area cylindrical tubular SOFC at 1000 °C and $U_f = 0.85$. A general trend is that the $E(i_L, x)-x$ profile at a given current is predominated by the Nernst potential E_{ex} of the exit fuel composition at the lowest current, and it gradually becomes linear over the length of the cell as the current density i_L increases (e.g. >250 mA cm⁻²). The actual average Nernst potential $\bar{E}(i_L)$ at each i_L can be mathematically calculated by integrating $E(i_L)$.



Fig. 1. *E*(*x*)-profiles at various current densities.

x) over the entire length *L* of the cell:

$$\overline{E}(i_{\rm L}) = \frac{\int_0^L E(i_{\rm L}, x) dx}{\int_0^L dx}$$
(10)

The observed linear relationship at approximately >250 mA cm⁻² allows the use of a simple arithmetic average of E_{in} (of the inlet fuel) and E_{ex} to obtain the i_L -independent \overline{E} :

$$\bar{E} \approx \frac{1}{2}(E_{\rm in} + E_{\rm ex}) \tag{11}$$

 E_{in} and E_{ex} are both i_L -independent and are only dependent of temperature, composition and type of fuel. With H₂-H₂O fuel, for example, they follow with temperature *T* and inlet gas flow rates Q° of H₂ and H₂O (assuming the system pressure of 1 atm) in the form of:

$$E_{\rm in} = 1275 - 0.3171T - 0.0430T \ln\left(\frac{Q_{\rm H_2O}^{\rm o}}{Q_{\rm H_2}^{\rm o}}\right) \,\mathrm{mV} \tag{12}$$

$$E_{\text{ex}} = 1275 - 0.3171T - 0.0430T \ln \left(\frac{Q_{\text{H}_2\text{O}}^{\circ} + U_f Q_{\text{H}_2}^{\circ} + Q_{\text{LK}}}{Q_{\text{H}_2}^{\circ} (1 - U_f) - Q_{\text{LK}}} \right) \quad (13)$$

Differentiating Eq. (11) with respective to $U_{\rm f}$ and substituting Eqs. (12) and (13) lead to:

$$\left(\frac{\partial \tilde{E}}{\partial U_{\rm f}}\right)_{T,i_{\rm L},Q_{\rm L}} = -0.0215T \times \left(\left(r^{\rm o} \times \left(1 + \frac{Q_{\rm LK}}{0.00697 \times i_{\rm L} \times A_{\rm c}}\right)^{-1} + U_{\rm f}\right)^{-1} + \left(\left(1 + \frac{Q_{\rm LK}}{0.00697 \times i_{\rm L} \times A_{\rm c}}\right)^{-1} - U_{\rm f}\right)^{-1}\right)$$
(14)

where \bar{E} has a unit of mV. R is the universal gas constant, 8.314 J mol⁻¹ K⁻¹; $r^{o} = Q_{H_{2}O}^{o}/Q_{H_{2}}^{o}$; A_{c} is the active area of the cell. The variations of $Q_{H_{2}}^{o}$ (and therefore $Q_{H_{2}O}^{o}$ for a fixed r^{o}) with U_{f} are considered during the differentiation.

For a cathode-supported SOFC, the total contribution from R_a is marginal comparing to R_c . Therefore, the differentiation of R_a with respective to U_f can be reasonably ignored in the following discussion. Eq. (14) then becomes the definition of fuel sensitivity β :

$$\beta = -0.0215T \times \left(\left(r^{o} \times \left(1 + \frac{Q_{LK}}{0.00697 \times I_{L}} \right)^{-1} + U_{f} \right)^{-1} + \left(\left(1 + \frac{Q_{LK}}{0.00697 \times I_{L}} \right)^{-1} - U_{f} \right)^{-1} \right) (mV)\%$$
(15)



Fig. 2. Fuel sensitivity β as a function of U_t of a SOFC consisting of plasma sprayed electrolyte and ceramic interconnect.

where $i_L A_c$ is substituted by I_L . β has a unit of mV/%. For the cases where R_a is dependent of U_f , Ref. [7] gives more general solutions.

One of the utilities of Eq. (15) is that it provides a methodology to acquire Q_{LK} from a measured β – U_f curve. To be specific, a non-linear least-square fitting program can be used to best fit the experimental data (e.g., β – U_f curve), using Eq. (15) as the model and Q_{LK} as the only adjustable parameter. One example is given to illustrate the use of the method in the following.

3.2. Application to cathode-supported tubular SOFCs

To use the linear relationship expressed in Eq. (11), fuelsensitivity measurements need to be conducted at higher current densities (e.g., $i_L > 250 \text{ mA cm}^{-2}$) to ensure a better accuracy. The measurement of fuel sensitivity is rather simple. When *T*, i_L , and U_o are held constant in a SOFC, U_f of the cell is gradually varied by changing fuel flow rates while the corresponding V_c is recorded. U_f is typically varied from 0.70 to 0.95 in a step of 0.05. The resultant V_c at each U_f is then mathematically treated to give numerical approximations of the derivative $\beta = (\partial V_c / \partial U_f)_{T, i_L, Q_L}$. The latter is then plotted against U_f to produce a β - U_f curve.

Fig. 2 shows a measured β – U_f curve (dots) of a cathodesupported tubular cell with a plasma-sprayed electrolyte and ceramic interconnection. The simulation using Eq. (15) as represented by the line was obtained by fitting the experimental data with a commercial non-linear least-square program DataFit[®]. The best-fit Q_{LK} value is found to be 18.17 ± 1.88 sccm (standard cubic centimeter per minute) H₂ for the 50-cm long cell under a condition of *T* = 1000 °C and *i*_L = 0.30 A cm⁻².

4. Effect of leakage flux on the *V–J* curve – "bending-over" phenomenon at low current

It is commonly seen in electrical testing of tubular cathodesupported SOFCs that *V*–*I* characteristic, when measured at high temperature and high $U_{\rm f}$, bends over at low-current–density regime. Fig. 3 shows a representation of this "bending-over" phenomenon at different temperatures. The fact that the "bendingover" becomes less pronounced at lower temperatures and lower $U_{\rm f}$ (not shown in Fig. 3) strongly suggests that the mixed-conduction induced oxygen-leakage is the predominant cause for the observed lower voltage at low current density. In the following, a mathematic expression is given to illustrate the effect of leakage flux on the degree of the "bending-over".

In theory, the degree to which V_c is varied with Q_{LK} is primarily determined by how much Nernst potential is influenced by Q_{LK} because the partial pressures of H₂ and H₂O can be altered by Q_{LK} .



Fig. 3. A typical *V*–*I* characteristic measured from an oxygen-leakage present cathode-supported SOFC at *T* = 1000, 940 and 900 °C, U_f = 85% and U_o = 16%.

Therefore, the Nernst potential in Eq. (12) of the inlet H_2 - H_2O fuel is changed by Q_{LK} .

$$E = 1275 - 0.3171T - 0.0430T \ln\left(\frac{Q_{H_2O}^{0} + Q_{LK}}{Q_{H_2}^{0} - Q_{LK}}\right) (mV)$$
(16)

The difference between Eqs. (12) and (16), ΔE , represents the reduction in Nernst potential E_{in} of the inlet fuel by Q_{LK} .

$$\Delta E = 0.0430T \ln \left(\frac{Q_{H_2O}^0 + Q_{LK}}{Q_{H_2}^0 Q_{LK}} \times \frac{Q_{H_2}^0}{Q_{H_2O}^0} \right)$$

= 0.0430T ln $\left(\frac{((0.00697 \times I_L)/U_f) + (Q_{LK}/r^o)}{((0.00697 \times I_L)/U_f) - Q_{LK}} \right) (mV)$ (17)

where *I*_L is the load current in amperes and *Q*_{LK} is the leakage flux in slpm.

As aforementioned, the variations of E with i_L and x are the result of a progressive electro-oxidation of the fuel as it travels over the entire cell surface area from the entrance to the exit. Since the linear approximation is only valid at high current density, dependence of the average \overline{E} with I_L is needed for low-current range. An empirical equation that correlates the average $\overline{E}(I_L)$ of a leakage-free cell with I_L over a broad current regime is given by:

$$\bar{E}(I_{\rm L}) = 34.3 \times \ln(I_{\rm L}) + 702 \,({\rm mV})$$
 (18)

The expression was obtained under the conditions of $r^{o} = 11/89$, $U_{\rm f} = 0.85$, $T = 1000 \,^{\circ}$ C, $A_{\rm c} = 270 \,\rm{cm}^{2}$ and total cell resistance $R_{\rm cell} = 4 \,\rm{m}\Omega$. Substitution of Eq. (18) into Eq. (17) yields $Q_{\rm LK}$ -dependent *V*–*I* equation:

$$V_{\rm c} = 34.3 \ln I_{\rm L} + 702 - I_{\rm L} \times R_{\rm cell} -0.0430T \ln \left(\frac{((0.00697 \times I_{\rm L})/U_{\rm f}) + (Q_{\rm LK}/r^{\rm o})}{((0.00697 \times I_{\rm L})/U_{\rm f}) - Q_{\rm LK}} \right) (\rm mV)$$
(19)

The critical current I_c at which the maximum V_c occurs can be obtained by differentiating V_c with respect to I_L at a constant T and U_f :

$$\frac{\partial V_{\rm c}}{\partial I_{\rm L}} \mid = \frac{34.3}{I_{\rm L}} - R_{\rm cell} + 0.043T \times \frac{m(1+1/r^0)Q_{\rm LK}}{(mI_{\rm L}+Q_{\rm LK}/r^0)(mI_{\rm L}-Q_{\rm LK})}$$
(20)

where the constant $m = 0.00697/U_{\rm f}$. By setting Eq. (20) to zero, **I**_c can be solved.

To simulate the "bending-over" behavior of a V–I curve with Eq. (19), a SOFC with $R_{cell} = 4 \text{ m}\Omega$ and an active surface area of 270 cm² is virtually perceived to operate at 1000 °C with $U_f = 0.85$ of an inlet fuel containing 89% H₂ and 11% H₂O. Fig. 4 shows the simulated V–I curves under various leakage fluxes. Under the leakage-free condition, V_c gradually approaches to 0.750 V as I_L comes close to zero.



Fig. 4. Effect of leakage flux on the V-I characteristic of a leakage-present SOFC.



Fig. 5. Plot of the critical load current as a function of leakage flux.

 $V_{\rm C}$ = 0.750 is the exit Nernst potential at 1000 °C after 85% utilization of the 89% H_2 - H_2 O inlet fuel. In the presence of leakage flux, V_c bends over with $I_{\rm L}$ at the low-current region, an opposite characteristic of conventional V-I curve. The degree of the "bending-over" increases with Q_{LK}. The underlying reason for the "bending-over" is the decreasing Q/Q_{LK} ratio with decreasing current because Q_{LK} increases while fuel flow Q decreases with decreasing current for a constant fuel-utilization operation. The consequence of this combined effect is that the amount of fuel Q given to the cell is easily overwhelmed by the amount of leakage flux Q_{LK}. The cell voltage is lowered as a result of the reduced Nernst potential. To further illustrate the shift of the critical current I_c at which the peak voltage appears, Fig. 5 plots I_c as a function of Q_{LK} . Clearly, a higher Q_{LK} would force the V–I curve to bend over at higher current. The "bending-over" behavior should be prevented for any SOFCs because it has a negative impact on the maximum electrical efficiency. Therefore, suppression of fuel loss induced by oxygenleakage, particularly mixed-conduction induced one, is critical to maintaining high electrical efficiency offered by a SOFC. Lowering the operating temperature of a SOFC is an effective way to minimize the mixed-conduction induced oxygen-leakage.

5. Summary

Fuel utilization $U_{\rm f}$ represents a fraction of the fuel input that is electrochemically consumed by oxide-ion current. In the presence of an oxygen-leakage current, the relationship between fuel flow and $U_{\rm f}$ needs revision in order to properly compensate the leakage flux. A failure to do so could unknowingly result in overconsumption of fuel and possibly cause the oxidation of Ni-based anode. Fuel sensitivity β is the change in cell voltage with respect to *U*_f under a constant current, temperature and oxygen utilization. The shape of a β – U_f curve can be best characterized by low-value but flat-change at low *U*_f and high-value but sharp-change at high $U_{\rm f}$. The presence of a leakage flux could change the magnitude and the shape of a β - U_f curve. A mathematical expression including fuel sensitivity, fuel utilization, load current and more importantly leakage flux is established, based on which the leakage flux information can be extracted from a measured sensitivity curve using nonlinear least-square fitting. Finally, the observed "bending-over" phenomenon at the low-current range of a constant-*U*_f *V*–*I* curve is quantitatively coupled with the leakage flux. The greater the leakage-flux the lower the peak voltage and the higher the current at which the peak voltage appears. The oxygen-leakage in an SOFC, if not properly managed, will cause a reduced electrical efficiency.

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