THERMODYNAMIC BASIS OF FUEL CELL SYSTEMS

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Abstract

Power maximization approach is applied for fuel cells treated as flow engines driven by fluxes of chemical reagents and electrochemical mechanism of electric current generation. Analyzed are performance curves of a SOFC system and the effect of typical design and operating parameters on the cell performance. The theory combines a recent formalism worked out for chemical machines with the Faraday's law which determines the intensity of the electric current generation. Steady-state model of a high-temperature SOFC is considered, which refers to constant chemical potentials of incoming hydrogen fuel and oxidant. Lowering of the cell voltage below its reversible value is attributed to polarizations and imperfect conversions of reactions. The power formula summarizes the effect of transport laws, irreversible polarizations and efficiency of power yield. The reversible electrochemical theory is extended to the case with dissipative chemical reactions; this case includes systems with incomplete conversions, characterized by "reduced affinities" and an idle run voltage. The efficiency decrease is linked with thermodynamic and electrochemical irreversibilities expressed in terms of polarizations (activation, concentration and ohmic). Effect of incomplete conversions is modelled in a novel way assuming that substrates can be remained after the reaction and that side reactions may occur. Optimum and feasibility conditions are obtained and discussed for some important input parameters such as the effciency, power output, and electric current density of the cell. Calculations of the maximum power show that the data differ for power generated and consumed, and depend on parameters of the system, e.g., current intensity, number of mass transfer units, polarizations, electrode surface area, average chemical rate, etc.. These data provide bounds for SOFC energy generators, which are more exact and informative than reversible bounds for electrochemical transformation.

Key words

Power limits, entropy, engines, thermal efficiency, fuel cells.

1 Introduction

Fuel cells (FC) are electrochemical engines. Their role for environmental protection cannot be underestimated. The main advantage of fuel cells in comparison to heat engines is that their efficiency is not a major function of device size. A fuel cell continuously transforms a part of chemical energy into electrical energy by consuming fuel and oxidant. In the present paper power maximization approach is applied for the purpose of determining power limits in imperfect fuel cells, where for sufficiently large electric currents power decreases with current because of prevailing effect of loss phenomena

In a previous work [Sieniutycz, 2003] we modeled power production and its limits in purely thermal systems with finite rates. In particular, radiation engines were analyzed as nonlinear systems governed by laws of thermodynamics and transport. Temperatures T of resource media were only necessary variables to describe these systems. However, fuel cells are more general systems in which both temperatures T and chemical potentials mk are essential. They are electrochemical engines propelled by fluxes of both energy and substances.

Basic structure of fuel cells includes electrolyte layer in contact with a porous anode and cathode on either side. Gaseous fuels are fed continuously to the anode (negative electrode) compartment and an oxidant (i. e., oxygen from air) is fed continuously to the cathode (positive electrode) compartment. Electrochemical reactions take place at the electrodes to produce an electric current. Basic reaction is the electrochemical oxidation of fuel, usually hydrogen, and the reduction of the oxidant, usually oxygen. This principle makes a fuel cell similar to a chemical engine. In a FC process in Fig. 1 streams of fuel (H2) and oxidant (O2) interact; the process is propelled by diffusive and/or convective fluxes of heat and mass, transferred through the cell 'conductances' or boundary layers. The energy flux (power) is created in the cell generator which exploits the fuel stream contacting with the anode and the oxidant stream contacting with the catode. Both electrodes are separated by the electrolyte. As in typ-
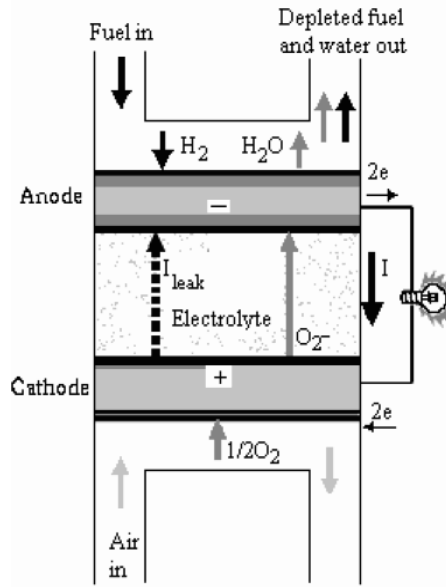
Figure 1.   Principle of a solid oxide fuel cell.

ical thermal machines and radiation engines [Curzon and Ahlborn, 1975], [De Vos, 1994], [Sieniutycz and Kuran, 2005], [Sieniutycz and Kuran, 2006], [Sieniutycz, 2009a], [Kuran, 2006] both transfer mechanisms and properties of conducting layers influence the rate of power production.

Fuel cell systems working in the power yield mode are electrochemical flow engines propelled by chemical reactions. Their performance is determined by magnitudes and directions of participating streams and by mechanism of electric current generation. Voltage lowering in a cell below its reversible value is a good measure of the cell imperfection.

The goals of the present paper include: (a) formulation of a thermo-electro-chemical model for imperfect fuel cells, especially for those with incomplete chemical conversions, (b) implementation of the model to simulate the behavior of high-T solid oxide fuel cells, (c) prediction of various losses of the voltage and their effect on the cell performance, and (d) application of fuel cell characteristics for the purpose of determining power limits.

2   Thermodynamics of power generation

Knowledge of operational voltage is required to define a cell efficiency as the ratio $\chi = V/E$, where E is the reversible cell voltage or the equilibrium cell potential. For the power density in terms of χ one has $p = iE\chi$ or $p = p^{rev}$, which means that this efficiency is equal to the ratio of the actual power to the maximum reversible power. This definition links the fuel cell efficiency with the second law, and stresses substantial role of the operational voltage.

Assume that all incoming streams (those with "higher" Gibbs flux $G_{in} = G_{1'}$) represent a common phase of "substrates" (all system's components in

the state before the chemical transformation, index $1'$). All outgoing streams (those with "lower" Gibbs flux $G_{out} = G_{2'}$ represent the common phase of "products" (all system components in the state after the transformation, index $2'$). The power expression follows from entropy conservation and energy balance in the reversible part of the system. For an isothermal reactor power yield is

$$p = \mu_{1_{1'}}\dot{n}_{1_{1'}} + \mu_{2_{1'}}\dot{n}_{2_{1'}} + ...\mu_{i_{1'}}\dot{n}_{i_{1'}} .... + \mu_{m_{1'}}\dot{n}_{m_{1'}}$$

$$(1)$$

$$-\mu_{1_{2'}}\dot{n}_{1_{2'}} - \mu_{2_{2'}}\dot{n}_{2_{2'}} .... - \mu_{i_{2'}}\dot{n}_{i_{2'}} .... - \mu_{m_{2'}}\dot{n}_{m_{2'}}.$$

This formula shows that, in a steady and isothermal process, power yield of a chemical engine system is the difference between the input and output flux of the Gibb's function [Sieniutycz and Jeowski, 2009], [Sieniutycz, 2008], [Tsirlin et al, 1998], [Sieniutycz, 2009b]. We can transform Eq. (1) to a pronouncing form of Eq. (2) below, specific to the case of the complete conversion. In this case the components are numbered such that species $1, 2, \ldots, i$ are substrates and species $i + 1, i + 2, \ldots, m$ are products. Total power yield of an isothermal multi-reaction process takes the form

$$p = \sum_{j=1}^{R}\{p_j\} = \sum_{j=1}^{R} -\{\mu_{1_{1'}}\nu_{1j} + \mu_{2_{1'}}\nu_{2j} + ...\mu_{i_{1'}}\nu_{ij}$$

$$(2)$$

$$+\mu_{i+1_{2'}}\nu_{i+1j} + ...\mu_{m-1_{2'}}\nu_{m-1j} + \mu_{m_{2'}}\nu_{mj})\}\dot{n}_j.$$

Quantities $\dot{n}_j$ are molar chemical fluxes of reagents, i. e. products of the electrode surface area F and heterogeneous rates, r_j. In the case of complete conversion, power yield from the unit electrode area equals the sum of products of the affinity driving forces and the reaction rates

$$p = \sum_{j=1}^{R}\{\tilde{A}_j\dot{n}_j\} = F\sum_{j=1}^{R}\{\tilde{A}_j r_j\}. \qquad (3)$$

Yet, the assumption about the complete transformation of substrates into products can be relaxed, and the present paper shows how this can be done for fuel cells. By considering the chemistry of systems with power production and transport phenomena one can quantitatively estimate effects of incomplete conversions. The related formula resembles the one which describes the effect of the internal entropy production within these systems [Sieniutycz, 2009b]. For a single isothermal chemical reaction the corresponding power formula which generalizes Eq. (3) to include effect of incomplete conversions can be written in the form

$$p = (\Pi_{1'} - \Xi\Pi_{2'})\dot{n}_{1'}$$
$$= -\frac{iA}{n_eF}\Delta g^{eff}(T, p) = -\Delta\dot{G}^{eff}, \qquad (4)$$

where primed quantities refer to the inputs and outputs of the chemically active zone and $\dot{Q}_{1'}$ is the total heat flux (involving the sensible heat flux, $q_{1'}$, and the sum of products of partial entropies and fluxes of species multiplied by the temperature T), $\Pi_{1'}$ is "one-way chemical affinity" attributed to reactants with known chemical potentials [Sieniutycz and Jeowski, 2009], [Sieniutycz, 2009b], $\dot{n}_{1'}$ is the (positive) chemical flux defined as the product of the heterogeneous reaction rate and the electrode area. Internal imperfection functions, Φ and Ξ, are respectively related to internal entropy production and incomplete conversion. The fraction Ξ is the reciprocity of coefficient Ψ introduced in [Sieniutycz, 2009b]; they both characterize detrimental increase of chemical potentials of reaction products caused by their dilution by remaining reactants.

Power formula of Eq. (4) generalizes the idealized power of an "endoreversible" system (with $\Xi = 1$) in which case difference $\Pi_{1'} - \Pi_{2'}$ is chemical affinity or $-\Delta g$. This is the chemical component of power, which describes power yield caused by chemical flux $n_{1'}$. Electrochemical power is generated with non-ideal chemical efficiency $\xi = \Pi_{1'} - \Xi\Pi_{2'}$. For the simplest reaction, 1⇔2, $\xi = \mu_{1'} - \Xi\mu_{2'}$ which is lower than $\mu_{1'} - \mu_{2'}$. Effectively, in the engine mode where $\Xi \leq 1$, the system with internal imperfections, behalves as it would operate with a decreased affinity of an effective value $\Pi_{1'} - \Xi\Pi_{2'}$. Of course, power production is decreased by this imperfection.

3 Effect of transport phenomena

For brevity we limit this section to the case of a simple isomerisation reaction $A_1 - A_2 = 0$, which drives the fuel cell generator. Transported energy and components also drive power generation in fuel cells.

Interestingly, there exists a formal link between the mathematics of thermal engines and fuel cells. To display this link let us first begin with a simnplest chemical engine without electrochemical effects. We note that the power producing force in an endoreversible thermal engine equals $T_{1'} - T_{2'}$. Whereas the propelling force in the simplest chemical engine is $\mu_{1'} - \mu_{2'}$. For the bulks of the streams or reservoirs the related differences of temperature and chemical potential are $T_1 - T_2$ and $\mu_1 - \mu_2$. Since the deviations of $T_{1'}$ and $\mu_{1'}$ from T_1 and μ_1 are of purely dissipative origin and the bulk differences $T_1 - T_2$, and $\mu_1 - \mu_2$ are identical with the "open circuit" (Carnot) values for the "active" differences $T_{1'} - T_{2'}$, and $\mu_{1'} - \mu_{2'}$, we may write

$$T_{1'} - T_{2'} = T_1 - T_2 - I_s(R_{1s} + R_{2s}) \qquad (5)$$

$$\mu_{1'} - \mu_{2'} = \mu_1 - \mu_2 - I_n(R_{1n} + R_{2n}), \qquad (6)$$

where I_s and I_n are the conserved currents of entropy and matter flowing through the energy-generating zone

of the system. The indices 1 and 2 refer, respectively, to the resistances in the "upper" and "lower" part of the engine system.

Active (power producing) driving forces corresponding with Eqs. (5) and (6) are the temperature difference $T_{1'} - T_{2'}$ and the chemical affinity $\mu_{1'} - \mu_{2'}$. Total power yield is then

$$
\begin{aligned}
p &= (T_{1'} - T_{2'})I_s + (\mu_{1'} - \mu_{2'})I_n \\
&= (T_1 - T_2)I_s + (\mu_1 - \mu_2)I_n \\
&\quad - (R_{1s} + R_{2s})I_s^2 - (R_{1n} + R_{2n})I_n^2.
\end{aligned} \qquad (7)
$$

By introducing the electric current I_e and the electric potential V into this equation we can generalize it to electrochemical systems and fuel cells. In terms of the definitions of total resistances the so-generalized Eq. (7) can be written in the form

$$
\begin{aligned}
p &= (T_{1'} - T_{2'})I_s + (\mu_{1'} - \mu_{2'})I_n + \\
&\qquad\qquad (\phi_{1'} - \phi_{2'})I_e \\
&= (T_1 - T_2)I_s + (\mu_1 - \mu_2)I_n + (\phi_1 - \phi_2)I_e \\
&\qquad - R_{ss}I_s^2 - R_{nn}I_n^2 - R_{ee}I_e^2.
\end{aligned} \qquad (8)
$$

Equation (8) can still be generalized to the case of coupled heat and charge transfer in the dissipative conductors of the system. Such a generalization will lead us to a general result for power limits in linear systems. The generalization has the form

$$
\begin{aligned}
p &= (T_{1'} - T_{2'})I_s + (\mu_{1'} - \mu_{2'})I_n + \\
&\qquad\qquad (\phi_{1'} - \phi_{2'})I_e \\
&= (T_1 - T_2)I_s + (\mu_1 - \mu_2)I_n + (\phi_1 - \phi_2)I_e \\
&\qquad - R_{ss}I_s^2 - R_{nn}I_n^2 - R_{ee}I_e^2 \\
&\qquad - R_{sn}I_sI_n - R_{se}I_sI_e - R_{ne}I_nI_e
\end{aligned} \qquad (9)
$$

After introducing the enlarged flux vector $\tilde{\mathbf{I}} = (I_s, I_n, I_e)$, the enlarged thermal potential vector $\tilde{\mu} = (T, m, V)$ and the overall resistance tensor $\tilde{\mathbf{R}}$, Eq. (9) can be written in a simple and concise matrix-vector form

$$p = (\tilde{\mu}_1 - \tilde{\mu}_2).\tilde{\mathbf{I}} - \tilde{\mathbf{R}} : \tilde{\mathbf{I}}\tilde{\mathbf{I}}. \qquad (10)$$

4 Power limits in thermal power systems and fuel cells

While for a complex driving reaction the dimensionality of the potential vector $\tilde{\mu}$ will certainly be much larger than that of the original vector, in real systems the structure of Eq. (10) can be preserved whenever this equation will be considered in the vector form. Maximum power corresponds with the vanishing partial derivatives

$$\partial p / \partial \tilde{\mathbf{I}} = \tilde{\mu}_1 - \tilde{\mu}_2 - 2\tilde{\mathbf{R}}.\tilde{\mathbf{I}} = 0. \qquad (11)$$

Therefore, the optimal (power-maximizing) vector of currents at the maximum point of the system can be written in the form

$$\tilde{\mathbf{I}}_{mp} = \frac{1}{2}\tilde{\mathbf{R}}^{-1}.(\tilde{\mu}_1 - \tilde{\mu}_2) \equiv \frac{1}{2}\tilde{\mathbf{I}}_F \qquad (12)$$

This result means that in the strictly linear systems the power-maximizing current vector $\tilde{I}_{mp}$ is equal to one half of the purely dissipative current at the Fourier-Onsager point, $\tilde{\mathbf{I}}_{mp}$. The latter point refers to the system's state at which no power production occurs. Consistently, Eqs. (10) and (12) yield the following result for the maximum power limit of the system

$$p_{mp} = \frac{1}{4}(\tilde{\mu}_1 - \tilde{\mu}_2).\tilde{\mathbf{R}}^{-1}.(\tilde{\mu}_1 - \tilde{\mu}_2). \qquad (13)$$

In terms of the purely dissipative flux vector at the Fourier-Onsager point, $\tilde{\mathbf{I}}_F$, the above limit of maximum power is represented by an equation

$$p_{mp} = \frac{1}{4}\tilde{\mathbf{R}} : \tilde{\mathbf{I}}_F\tilde{\mathbf{I}}_F \qquad (14)$$

On the other hand, power dissipated at the Fourier-Onsager point equals

$$p_F = \tilde{\mathbf{R}} : \tilde{\mathbf{I}}_F\tilde{\mathbf{I}}_F \qquad (15)$$

Comparison of Eqs. (14) and (15) proves that, in linear thermo-electro-chemical systems, only at most 25% of power which is dissipated in the natural transfer process, can be transformed into the noble form of the mechanical power. This is a general result which, probably, cannot be easily generalized to the nonlinear transfer systems where significant deviations from Eq. (14) may appear depending on the nature of diverse nonlinearities.

It may be noted that the ratios of Eqs. (14) and (15) can be regarded as some efficiency measures. Yet, they should not be mixed with commonly used, popular efficiencies. There is a number of definitions of FC efficiencies proposed for measuring and comparing performances of electrochemical processes. Clearly, the popular fuel cell efficiencies defined as $\eta_{FC} = \Delta G/\Delta H$, which are commonly applied to many fuel cell systems, can achieve numerical values much higher than $\frac{1}{4}$ very easily because they are first-law efficiencies, not equivalent to power ratios P_{mp}/P_F involving Eqs. (14) and (15). In fact, the latter power ratios represent the second-law efficiencies of the overall thermo-electro-chemical process. Only second-law efficiencies are correct efficiency measures in every case. Efficiencies based on the first law (often found in the literature) can generate efficiency values greater than 100% for certain systems. One of the most correct, simplest, and practical definition of efficiency for a fuel cell operating near ambient temperature is the ratio of the actual voltage to the reversible voltage.

5 Some experimental data

In fact, the forms of Eqs. (9) and (10) are sufficient to claim that the thermal force formula and the power formula for the thermal engine are similar to the voltage and power formulae in the fuel cell system. This observation proves that a link exists between the mathematics of the thermal engines and fuel cells, and also that the theory of fuel cells can be unified with the theory of thermal engines. In this spirit, we present a brief analysis of some experimental data.

Voltage lowering in fuel cells below the reversible voltage is a good measure of their imperfection. Yet we need to distinguish between Nernst ideal voltage E^0 or and idle run voltage E_0. It is the latter quantity from which all rate dependent losses of voltage should be subtracted. A number of approaches for calculating these polarization losses have been reviewed in literature by Zhao, Ou and Chen [Zhao et al, 2008]. The details of calculations of the idle run voltage E_0 are thoroughly discussed by Wierzbicki [Wierzbicki, 2009] who has implemented the $Aspen\ Plus^{TM}$ software to investigate the behavior of SOFC based energy system using his own theoretical model of power yield kinetics. The model was based on Eqs. (16)–(19) of the present paper and some associated relationships. His calculations were compared with the experimental findings of the voltage and power in a laboratory FC system.

In some situations difference between E^0 and E_0 is a current independent loss which may be described by a fraction Ξ characterizing the detrimental increase of chemical potentials of products caused by their dilution by unreacted substrates. With the concept of effective nonlinear resistances operating voltage can be represented as the departure from the idle run voltage E_0 (the quantity which replaces the reversible voltage E^0 in more involved situations)

$$V = E_0 - V_{int} = E_0 - V_{act} - V_{conc} - V_{ohm}$$
$$= E_0 - I(R_{act} + R_{conc} + R_{ohm}). \quad (16)$$

Note the analogy between this equation and Eqs. (8) and (9)) especially after its multiplication by the electric current (its presentation in the power terms). The losses, called polarization, include three main sources: activation polarization (V_{act}), ohmic polarization (V_{ohm}), and concentration polarization (V_{conc}). They refer to the equivalent activation resistance (R_{act}), equivalent ohmic resistance (R_{ohm}), and equivalent concentration resistance (R_{conc}). Activation and concentration polarization occur at both anode and cathode locations, while the resistive polarization represents ohmic losses throughout the fuel cell. Activation polarization V_{act} is neglected in the model of ref. [Wierzbicki, 2009], nonetheless the power curve is typical. As the voltage losses increase with current, the initially increasing power begins finally to decrease for sufficiently large currents, so that maxima of power are
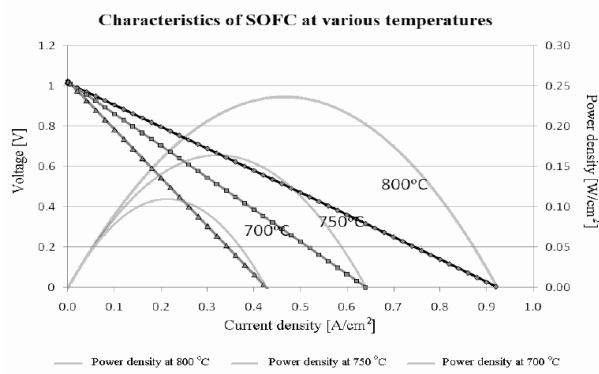
Figure 2. Voltage-current density and power-current density characteristics of the SOFC for various temperatures. Continuous lines represent the Aspen PlusTM calculations testing the model versus the experiments. The lines were obtained in Wierzbicki's MsD thesis supervised by S. Sieniutycz and J. Jewulski [13]. Points refer to experiments of Wierzbicki and Jewulski in Warsaw Institute of Energetics ([Wierzbicki, 2009], and his ref. 18).

observed (Fig. 2). The data include the losses of the idle run attributed to the flaws in electrode constructions and other imperfections which cause that the open circuit voltage is in reality lower than E^0.

Moreover, in the literature there are many other experimental and theoretical examples showing power maxima in fuel cells and proving the suitability of the thermal machine theory to chemical and electrochemical systems.

The voltage equation used in Wierzbicki's SOFC calculations is:

$$V = E_0(T, p_{H_2}) - iA_R(p_{H_2}) \exp\left(\frac{\Delta E}{RT}\right)$$
$$+ B \ln\left(1 - \frac{i}{i_L(T, p_{H_2})}\right) \quad (17)$$

where a limiting current is introduced defined by the equation

$$i_L = C_1 T^{-1} \exp\left(\frac{-E_a}{RT}\right) p_{H_2} \quad (18)$$

in which $C1$ is a experimentally determined parameter. Power density is simply the product of voltage V and current density i

$$p = Vi. \quad (19)$$

In an ideal situation (no losses) the cell voltage is defined by the Nernst equation. Yet, while the first term of Eq. (17) defines the voltage without load, it nonetheless takes into account the losses of the idle run, which are the effect of flaws in electrode constructions and other imperfections which cause that the open circuit voltage in reality to be lower than the theoretical value. The losses include ohmic polarization and concentration polarization. The second term of Eq. (17) quantifies ohmic losses associated with electric resistance

of electrodes and flow resistance of ions through the electrolyte. The third term refers to mass transport losses. Quantity i_L is the particular current density arising when the fuel is consumed in the reaction with the maximum possible feed rate.

6 Final remarks

The FC model developed in this paper describes physical and chemical performance of the irreversible fuel cells at various operating conditions. Lowering of SOFC efficiency is linked with polarizations (activation, concentration and ohmic) and incomplete conversions.Effect of incomplete conversion has been modeled assuming that substrates can be remained after the reaction and that side reactions may occur. Optimum and feasibility conditions have been obtained for a fuel cell, and discussed for some input parameters such as effciency, power output, and electric current density of the cell. Calculations of optimal power show that the data differ for power generated and consumed, and depend on parameters of the system, e. g., current intensity, number of mass transfer units, polarizations, electrode surface area, average chemical rate, etc.. These data provide bounds for SOFC energy generators, which are more exact and informative than reversible bounds for electrochemical transformation.

Power production bounds (limits) obtained in this paper are enhanced in comparison with those predicted by classical thermodynamics. As opposed to classical thermodynamics, our power bounds depend not only on changes of the thermodynamic state of participating resources but also on process irreversibilities, process direction and mechanism of heat and mass transfer. In fact, our research provides enhanced bounds. In thermostatics the bound on the work produced coincides with that on the work consumed. The generalized thermo-kinetic bounds, obtained here, are stronger than those predicted by thermostatics. Only for infinitely long durations or for processes with excellent transfer (an infinite number of transfer units) the thermokinetic bounds reduce to the classical thermostatic bounds.

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