

ARTICLE

Maximum Thermodynamic Electrical Efficiency of Fuel Cell System and Results for Hydrogen, Methane, and Propane Fuels

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The maximum electrical efficiency of fuel cell system, η_e^{\max} , is important for the understanding and development of the fuel cell technology. Attempt is made to build a theory for η_e^{\max} by considering the energy requirement of heating the fuel and air streams to the fuel cell operating temperature T . A general thermodynamic analysis is performed and the energy balances for the overall operating processes of a fuel cell system are established. Explicit expressions for the determination of η_e^{\max} are deduced. Unlike the Carnot efficiency, η_e^{\max} is found to be fuel specific. Except for hydrogen fuel, chemical equilibrium calculations are necessary to compute η_e^{\max} . Analytical solutions for the chemical equilibrium of alkane fuels are presented. The theoretical model is used to analyze the effects of T and the steam contents of CH_4 , C_3H_8 , and H_2 on η_e^{\max} for systems with various degrees of waste heat recovery. Contrary to the common perception concerning methane and propane fuels, η_e^{\max} decreases substantially with the increase of T . Moreover, η_e^{\max} of hydrogen fuel can be higher than that of methane and propane fuels for a system with a medium level of waste heat recovery and operated at $700\text{ }^\circ\text{C} \leq T \leq 900\text{ }^\circ\text{C}$.

Key words: Analytical theory, Energy balance, Nernst potential, Fuel utilization, Alkane, Chemical equilibrium

I. INTRODUCTION

The electrical power generation in a heat engine involves the processes of converting fuel chemical energy to heat, heat to mechanical energy, and mechanical energy to electricity. In comparison, the chemical energy of fuel is converted directly into electricity through electrochemical reactions in a fuel cell. Naturally, the electrical efficiency of a fuel cell is expected to be higher than that of a heat engine. The promising high electrical energy conversion efficiency is the major reason for the broad interest of the fuel cell technology. However, while the maximum efficiency of a heat engine has been clearly stated by the Carnot theorem, the maximum electrical efficiency of a fuel cell is only vaguely understood. A clear understanding of the maximum electrical efficiency of a fuel cell is of high scientific importance and technology implications.

Numerous efforts have been made towards the understanding of the fuel cell electrical efficiency [1–14]. Lutz *et al.* performed a thermodynamic analysis and concluded that the maximum fuel cell efficiency is

equivalent to the Carnot cycle with the high temperature reservoir set at the fuel combustion temperature [3]. The work of Lutz *et al.* is enlightening, but involves the unrealizable condition that the fuel cell operates with no load while the fuel is utilized 100%. Sidwell and Coors developed a numerical model to study the electrical efficiency of a solid oxide fuel cell (SOFC) fueled by hydrocarbon [6]. By setting the cell voltage at the Nernst potential of the consumed fuel and oxidant compositions, the dependence of the electrical efficiency on the cell voltage is determined and the maximum efficiency is then computed. Zhu and Kee carried out a general thermodynamic analysis that recovered the well known expression for the fuel cell electrical efficiency [7]. That is, the fuel cell electrical efficiency (η_{FC}) is the product of a fuel reversible efficiency (η_{Rev}), a cell voltage efficiency (η_V), and a fuel utilization ratio (η_{fuel}), *i.e.*, $\eta_{\text{FC}} = \eta_{\text{Rev}} \cdot \eta_V \cdot \eta_{\text{fuel}}$ [1, 6, 7]. Both the work of Sidwell and Coors and the work of Zhu and Kee did not consider the energy required by heating the fuel and air streams to the working temperature and by reforming the hydrocarbon fuel. Besides, the oxygen utilization ratio was assumed to be negligibly small. As is well known, ignoring the heat of reformation may result in the use of some unphysical value of $\eta_{\text{Rev}} > 1$ for fuels such as methane and methanol [1, 3]. The cell efficiency may thus be overestimated. In addition, the

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maximum cell efficiency cannot be reliably determined without considering the heating requirement of the fuel and air streams or with the assumption of a negligible oxygen utilization [8, 11]. The fuel and air streams heating requirement has been considered in a recent study [14], but the study is limited to hydrogen fuel and does not consider the waste heat reuse that is bound to underestimate the fuel cell efficiency.

This work aims at establishing a general analytical theory for the maximum thermodynamic electrical efficiency of a fuel cell system using a general fuel and considering the waste heat reuse. Based on the operations of a fuel cell system, a definition of the electrical efficiency of the fuel cell system is introduced. A general theoretical model for the maximum electrical efficiency of the fuel cell system is derived through the applications of thermodynamic principle. The general theory is further developed to yield analytical expressions specific to alkane and hydrogen fuels. Numerical examples about the maximum electrical efficiency are shown for methane, propane, and hydrogen fuels with representative steam contents and operating temperatures.

II. THEORY

A. Problem setting for the electrical efficiency of a fuel cell system

The electrical efficiency of a fuel cell system, η_e , is defined as:

$$\eta_e = \frac{IVt}{H_{\text{fuel}}} \quad (1)$$

where I is the current output at the fuel cell operating voltage of V , H_{fuel} is the heating value of the fuel input to the fuel cell system during the operating time period of t . A fuel cell system here means a complete power plant, including the fuel cell stack and all balance-of-plant components. For the study of the maximal electrical efficiency, the parasitic power losses in the balance-of-plant components are kept at the minimum. Nevertheless, the energy requirement of heating the fuel and air streams from T_0 (the room temperature) to T (the fuel cell working temperature) is considered as it is absolutely necessary in principle for the operation of a fuel cell system. Besides, the fuel cell stack is assumed to operate under the isobaric and isothermal conditions. The isobaric and isothermal conditions can be realized if the fuel and air streams flow very slowly. Moreover, the isobaric condition also eliminates the power requirement of the gas blower, consistent with the need of achieving the maximal electrical efficiency.

The overall thermodynamic processes of such a fuel cell system are schematically illustrated in FIG. 1. As shown in FIG. 1(a), the fuel and air streams are heated from T_0 to T . The amounts of heat required are ΔH_{fuel} and ΔH_{air} , respectively. Chemical reactions such as

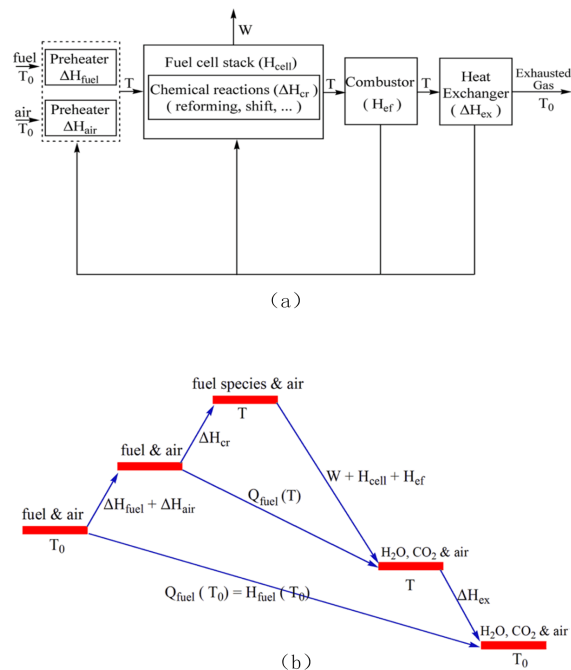


FIG. 1 Schematic of the thermodynamic processes of a fuel cell system: (a) the operating processes, (b) the energy balance diagram of the operation processes.

the steam reforming and water gas shift reactions may be involved to convert the fuel into chemical species suitable for the fuel cell electrochemical reactions. The amount of heat required by the overall chemical reactions is ΔH_{cr} . The fuel cell outputs an electrical energy of $W=IVt$ and releases an amount of heat, H_{cell} . The unused fuel exiting from the fuel cell stack can be burned to produce an amount of heat, H_{ef} , at the temperature of T . Cooling the exhausted fuel and air streams from T to T_0 yields an amount of residual heat, ΔH_{ex} . Through heat exchanger or other means, H_{cell} , H_{ef} and ΔH_{ex} are utilized to provide the required ΔH_{fuel} , ΔH_{air} and ΔH_{cr} . To be general, the amount of heat utilized is expressed as $H_{\text{cell}}+H_{\text{ef}}+\alpha\Delta H_{\text{ex}}$. Here α is an effective parameter denoting the capability of the fuel cell system of utilizing the waste heat. For an ideal fuel cell system, $\alpha=1$. To be practical, $\alpha<1$ is generally expected. It is noted that, as an effective parameter, $\alpha<0$ is allowed in principle. For example, $\alpha<0$ can be used to account for the effect of various kinds of heat losses, *e.g.*, thermal radiation of the fuel cell stack and/or incomplete combustion of the exiting fuel, on the system efficiency. A properly chosen value of $\alpha<0$ may also account for the effect of the energy consumed by the balance-of-plant components on the system efficiency.

Denoting the heat released by burning fuel when both reactants and products are at the temperature of T as $Q_{\text{fuel}}(T)$. When $Q_{\text{fuel}}(T)$ is at the standard state of $T=T_0$, it is equivalent to the conventional definition of the fuel heating value, *i.e.*, H_{fuel} in Eq.(1). To be

specific, H_{fuel} is denoted below as $H_{\text{fuel}}(T_0)$ that corresponds to $Q_{\text{fuel}}(T_0)$. Based on the nomenclature, the overall energy changes of the fuel cell operating processes described above can be summarized into FIG. 1(b).

B. Maximum thermodynamic electrical efficiency

To be thermally sustainable, the amount of usable heat should exceed the amount of heat necessary for heating fuel and air streams as well as maintaining the required chemical reactions, *i.e.*,

$$H_{\text{ef}} + H_{\text{cell}} + \alpha\Delta H_{\text{ex}} \geq \Delta H_{\text{fuel}} + \Delta H_{\text{air}} + \Delta H_{\text{cr}} \quad (2)$$

According to the energy balances shown in FIG. 1(b), one has:

$$\begin{aligned} H_{\text{ef}} + H_{\text{cell}} - \Delta H_{\text{cr}} &= Q_{\text{fuel}}(T) - W \\ &= Q_{\text{fuel}}(T) - IVt \end{aligned} \quad (3)$$

$$\Delta H_{\text{ex}} = H_{\text{fuel}}(T_0) - Q_{\text{fuel}}(T) + \Delta H_{\text{fuel}} + \Delta H_{\text{air}} \quad (4)$$

Combining Eq.(2)–Eq.(4) yields:

$$\begin{aligned} V &= \frac{Q_{\text{fuel}}(T) - (H_{\text{ef}} + H_{\text{cell}} - \Delta H_{\text{cr}})}{It} \\ &\leq \frac{Q_{\text{fuel}}(T) - (\Delta H_{\text{fuel}} + \Delta H_{\text{air}} - \alpha\Delta H_{\text{ex}})}{It} \\ &= \frac{(1 - \alpha)Q_{\text{fuel}}(T) + \alpha H_{\text{fuel}}(T_0)}{It} - \\ &\quad \frac{(1 - \alpha)(\Delta H_{\text{fuel}} + \Delta H_{\text{air}})}{It} \end{aligned} \quad (5)$$

The current production can be expressed in term of η_{fuel} and the oxygen utilization ratio, η_{O_2} , as:

$$It = mFN_{\text{fuel}}\eta_{\text{fuel}} = 4FN_{\text{O}_2}\eta_{\text{O}_2} \quad (6)$$

where m is the number of electrons released by fully oxidizing a fuel molecule, *e.g.*, $m=2$ for H_2 and $m=8$ for CH_4 . For a supplied fuel consisting of H_2 and CH_4 as well as other fuel species, m can be calculated by considering the molar fractions of the fuel species. N_{fuel} (N_{O_2}) is the moles of the fuel (O_2) input to the fuel cell during the time period of t , and F is the Faraday constant. Notice that no assumption is made here about the relationship between N_{fuel} and N_{O_2} , *i.e.*, N_{fuel} and N_{O_2} , or the corresponding η_{fuel} and η_{O_2} , are independent variables in the mathematical derivations.

With Eq.(6), only molar fuel and oxygen equivalent quantities are required for evaluating V of Eq.(5):

$$\begin{aligned} V &\leq \frac{(1 - \alpha)q_{\text{fuel}}(T) + \alpha h_{\text{fuel}}(T_0) - (1 - \alpha)\Delta h_{\text{fuel}}(T, T_0)}{mF\eta_{\text{fuel}}} \\ &\quad - \frac{(1 - \alpha)\Delta h_{\text{air}}(T, T_0)}{4F\eta_{\text{O}_2}} \end{aligned} \quad (7)$$

where $\Delta h_{\text{fuel}}(T, T_0)$ and $\Delta h_{\text{air}}(T, T_0)$ are the molar quantities of ΔH_{fuel} and ΔH_{air} , respectively. Notice that the content of non-fuel species should be considered in evaluating $\Delta h_{\text{fuel}}(T, T_0)$. For example, for wet fuel with steam fuel ratio of $y=N_{\text{H}_2\text{O}}/N_{\text{fuel}}$, one has:

$$\begin{aligned} \Delta h_{\text{fuel}}(T, T_0) &= (h_{\text{fuel}}(T) - h_{\text{fuel}}(T_0)) + \\ &\quad y(h_{\text{H}_2\text{O}}(T) - h_{\text{H}_2\text{O}}(T_0)) \end{aligned} \quad (8)$$

Similarly,

$$\begin{aligned} \Delta h_{\text{air}}(T, T_0) &= (h_{\text{O}_2}(T) - h_{\text{O}_2}(T_0)) + \\ &\quad \frac{N_{\text{N}_2}}{N_{\text{O}_2}}(h_{\text{N}_2}(T) - h_{\text{N}_2}(T_0)) \end{aligned} \quad (9)$$

Using Eq.(6) and Eq.(7), the electrical efficiency Eq.(1) is found to be:

$$\begin{aligned} \eta_e &= \frac{mFN_{\text{fuel}}\eta_{\text{fuel}}V}{H_{\text{fuel}}(T_0)} = \frac{mF\eta_{\text{fuel}}V}{h_{\text{fuel}}(T_0)} \\ &\leq \frac{(1 - \alpha)q_{\text{fuel}}(T) + \alpha h_{\text{fuel}}(T_0)}{h_{\text{fuel}}(T_0)} - \\ &\quad \frac{(1 - \alpha)\Delta h_{\text{fuel}}(T, T_0)}{h_{\text{fuel}}(T_0)} - \\ &\quad \frac{(1 - \alpha)m\Delta h_{\text{air}}(T, T_0)\frac{\eta_{\text{fuel}}}{4\eta_{\text{O}_2}}}{h_{\text{fuel}}(T_0)} \end{aligned} \quad (10)$$

Apparently, the maximum electrical efficiency is found when “ \leq ” in Eq.(10) is changed to “ $=$ ”. However, the maximum V , V_{max} , for current production is limited by the reversible potential of the fuel and air streams exiting from the fuel cell [6, 7]. The reversible potential, E_{Nernst} , is calculated with the equilibrium contents of the species in the exiting fuel stream. Notice that, according to the thermodynamics principle, E_{Nernst} is the same for all oxidation species at the equilibrium. For example, the reversible potentials for both H_2 and CO oxidations are the same. When H_2 or CO oxidation and other possible oxidation processes are involved, it suffices to compute either $E_{\text{Nernst}}(\text{H}_2 + 0.5\text{O}_2 \rightarrow \text{H}_2\text{O})$ or $E_{\text{Nernst}}(\text{CO} + 0.5\text{O}_2 \rightarrow \text{CO}_2)$, with the contents of fuel species determined by the equilibrium condition. To be general, the reversible potential of the exiting gas streams is denoted as $E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2})$. Using the ideal gas assumption, the maximum electrical efficiency is found for:

$$\begin{aligned} V_{\text{max}} &= \frac{(1 - \alpha)q_{\text{fuel}}(T) + \alpha h_{\text{fuel}}(T_0)}{mF\eta_{\text{fuel}}} - \\ &\quad \frac{(1 - \alpha)\Delta h_{\text{fuel}}(T, T_0)}{mF\eta_{\text{fuel}}} - \frac{(1 - \alpha)\Delta h_{\text{air}}(T, T_0)}{4F\eta_{\text{O}_2}} \\ &= E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2}) \\ &= -\frac{\Delta G_0^{\text{H}_2}(T)}{2F} + \frac{RT}{2F} \ln \frac{P'_{\text{H}_2} P'_{\text{O}_2}{}^{1/2}}{P'_{\text{H}_2\text{O}}} \\ &= -\frac{\Delta G_0^{\text{CO}}(T)}{2F} + \frac{RT}{2F} \ln \frac{P'_{\text{CO}} P'_{\text{O}_2}{}^{1/2}}{P'_{\text{CO}_2}} \end{aligned} \quad (11)$$

Above P'_{H_2} , $P'_{\text{H}_2\text{O}}$, P'_{CO} , and P'_{CO_2} are respectively the equilibrium partial pressures of H_2 , H_2O , CO , and CO_2 at the exiting fuel stream that are dependent on the input fuel compositions and η_{fuel} . For a wet hydrogen fuel, $P'_{\text{H}_2}=(1-\eta_{\text{H}_2})P_{\text{H}_2}$ and $P'_{\text{H}_2\text{O}}=(y+\eta_{\text{H}_2})P_{\text{H}_2}$, where P_{H_2} is the pressure of H_2 in the fuel supply. For a general fuel, P'_{H_2} and $P'_{\text{H}_2\text{O}}$ (P'_{CO} and P'_{CO_2}) should be determined by the equilibrium condition. $P'_{\text{O}_2}=(1-\eta_{\text{O}_2})P_{\text{O}_2}$ is the partial pressure of O_2 at the exiting air stream when the partial pressure of O_2 in the air supply is P_{O_2} . $\Delta G_0^{\text{H}_2}(T)$ ($\Delta G_0^{\text{CO}}(T)$) is the change in the standard-state Gibbs free energy for the reaction $\text{H}_2+0.5\text{O}_2\rightarrow\text{H}_2\text{O}$ ($\text{CO}+0.5\text{O}_2\rightarrow\text{CO}_2$). To be explicit, $E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2})$ is often specified below in terms of $E_{\text{Nernst}}(\text{H}_2+0.5\text{O}_2\rightarrow\text{H}_2\text{O})$. A change to $E_{\text{Nernst}}(\text{CO}+0.5\text{O}_2\rightarrow\text{CO}_2)$ is trivial, as indicated in Eq.(11). Notice that the use of $E_{\text{Nernst}}(\text{CO}+0.5\text{O}_2\rightarrow\text{CO}_2)$ is mandatory in some cases, *e.g.*, when fueled by dry gasified carbon.

To summarize, there are only two steps in the computation of the maximum electrical efficiency of a fuel cell system. First, for a given η_{fuel} , the value of η_{O_2} is obtained by solving the maximum operating voltage equation:

$$\begin{aligned} & \frac{(1-\alpha)q_{\text{fuel}}(T) + \alpha h_{\text{fuel}}(T_0) - (1-\alpha)\Delta h_{\text{fuel}}(T, T_0)}{mF\eta_{\text{fuel}}} \\ & - \frac{(1-\alpha)\Delta h_{\text{air}}(T, T_0)}{4F\eta_{\text{O}_2}} \\ & = E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2}) \\ & = -\frac{\Delta G_0^{\text{H}_2}(T)}{2F} + \frac{RT}{2F} \ln \frac{P'_{\text{H}_2}[(1-\eta_{\text{O}_2})P_{\text{O}_2}]^{1/2}}{P'_{\text{H}_2\text{O}}} \quad (12) \end{aligned}$$

Second, the maximum electrical efficiency, η_e^{max} , is then found with the maximum $\eta_{\text{fuel}}E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2})$ or the minimum $\eta_{\text{fuel}}/\eta_{\text{O}_2}$ obtained through Eq. (12),

$$\begin{aligned} \eta_e^{\text{max}} &= \frac{mF}{h_{\text{fuel}}(T_0)} [\eta_{\text{fuel}}E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2})]_{\text{max}} \\ &= \frac{(1-\alpha)q_{\text{fuel}}(T) + \alpha h_{\text{fuel}}(T_0)}{h_{\text{fuel}}(T_0)} - \\ & \quad \frac{(1-\alpha)\Delta h_{\text{fuel}}(T, T_0)}{h_{\text{fuel}}(T_0)} - \\ & \quad \frac{\Delta h_{\text{air}}(T, T_0) \left[\frac{(1-\alpha)m\eta_{\text{fuel}}}{4\eta_{\text{O}_2}} \right]_{\text{min}}}{h_{\text{fuel}}(T_0)} \quad (13) \end{aligned}$$

The rightmost hand side of Eq.(13) is useful and informative by showing explicitly the connection between η_e^{max} and the thermal characters of the fuel cell system and the fuel, α , $h_{\text{fuel}}(T_0)$, $\Delta h_{\text{fuel}}(T, T_0)$, *etc.*

Notice that, according to Eq.(12), η_{O_2} should approach zero when α approaches 1, with $(1-\alpha)/\eta_{\text{O}_2}$ approaching a finite value:

$$\frac{(1-\alpha)}{\eta_{\text{O}_2}} = \frac{4F}{\Delta h_{\text{air}}(T, T_0)} \left[\frac{h_{\text{fuel}}(T_0)}{mF\eta_{\text{fuel}}} - E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2} = 0) \right] \quad (14)$$

Consequently, the determination of η_e^{max} for $\alpha \approx 1$ is simpler than the two-step approach described above. η_e^{max} for an ideal fuel cell system involves only one variable, η_{fuel} , and can be determined directly by:

$$\eta_e^{\text{max}} = \frac{mF}{h_{\text{fuel}}(T_0)} [\eta_{\text{fuel}}E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2} = 0)]_{\text{max}} \quad (15)$$

The maximum electrical efficiency for an ideal fuel cell system fueled by H_2 is particularly simple and given by:

$$\eta_e^{\text{max}} = \frac{2F}{h_{\text{H}_2}(T_0)} \left\{ \eta_{\text{H}_2} \left[-\frac{\Delta G_0^{\text{H}_2}(T)}{2F} + \frac{RT}{2F} \ln \frac{(1-\eta_{\text{H}_2})P_{\text{O}_2}^{1/2}}{y + \eta_{\text{H}_2}} \right] \right\}_{\text{max}} \quad (16)$$

Eq.(16) is the closest analogue of the Carnot theorem for the fuel cell system. The analogue of the Carnot theorem for an ideal fuel cell system using a general fuel is given by Eq.(15). Unlike the Carnot efficiency that depends on T only (or T/T_0 , to be precise), η_e^{max} given by Eq.(15) depends also on the fuel type and the H_2O content in the fuel supplied to the fuel cells.

Except for H_2 fuel, the determination of η_e^{max} for any α requires the evaluation of $E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2})$ based on the fuel equilibrium compositions. Generally, the equilibrium compositions of a fuel mixture can be computed numerically by using some free or commercial codes.

C. Maximum electrical efficiency for hydrogen or alkane fuel

Alkane, $\text{C}_n\text{H}_{2n+2}$, is a class of widely used fuel. Explicit result on the maximum electrical efficiency of alkane fuel is of practical significance. Attempt is made here to derive an analytical expression for $E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2})$. For simplicity, the fuel stream is assumed to consist of H_2 , H_2O , CO , CO_2 , and $\text{C}_n\text{H}_{2n+2}$ only. The approximation should be accurate in practice due to: (i) coke formation is detrimental to the fuel cell operation and should be minimized by a choice of steam fuel ratio [15]; (ii) the equilibrium contents of other fuel species encountered when $n > 1$ are usually quite low and have a limited effect on the contents of H_2 and H_2O . Nevertheless, it is cautioned that the equilibrium theory below is built upon the assumption that can be problematic under undesirable conditions. The results are rigorous for H_2 fuel and CH_4 fuel when the coke formation is prevented thermodynamically.

TABLE I Fuel compositions of the initial and final states and the composition changes due to the SR and WGS reactions and the overall electrochemical reaction.

Species	Initial	Change	Final
C_nH_{2n+2}	1	$-x_1$	$1 - x_1$
CO	c_1	$nx_1 - x_2$	$c_1 + nx_1 - x_2$
CO ₂	c_2	x_2	$c_2 + x_2$
H ₂	c_3	$(2n + 1)x_1 + x_2 - \frac{m}{2}\eta_{\text{fuel}}$	$c_3 + (2n + 1)x_1 + x_2 - \frac{m}{2}\eta_{\text{fuel}}$
H ₂ O	c_4	$-nx_1 - x_2 + \frac{m}{2}\eta_{\text{fuel}}$	$c_4 - nx_1 - x_2 + \frac{m}{2}\eta_{\text{fuel}}$

The equilibrium contents of fuel species can be determined by the equilibrium conditions of two chemical reactions: (i) the steam reforming (SR) reaction, $C_nH_{2n+2} + nH_2O = nCO + (2n+1)H_2$; (ii) the water gas shift (WGS) reaction, $CO + H_2O = CO_2 + H_2$.

Assuming the species mole ratio of an input fuel is $C_nH_{2n+2}:CO:CO_2:H_2:H_2O = 1:c_1:c_2:c_3:c_4$. For 1 mol C_nH_{2n+2} , x_1 mol C_nH_{2n+2} is reformed by the SR reaction, x_2 mol CO is shifted by the WGS reaction. In addition, a fuel utilization of η_{fuel} corresponds to a consumption of $\frac{m}{2}\eta_{\text{fuel}}$ mol H₂ for the electricity production. The fuel compositions for the initial and final states and the composition changes due to the SR and WGS reactions and the current production can be summarized into Table I.

The equilibrium SR and WGS reactions yield respectively:

$$\exp\left(-\frac{\Delta G_{\text{SR}}(T)}{RT}\right) = \frac{P'_{\text{CO}} P'^{2n+1}_{\text{H}_2}}{P'_{C_nH_{2n+2}} P'^n_{\text{H}_2O}} = \frac{\mathcal{A}}{\mathcal{B}} \quad (17)$$

$$\mathcal{A} = (c_1 + nx_1 - x_2)^n [c_3 + (2n + 1)x_1 + x_2 - \frac{m}{2}\eta_{\text{fuel}}]^{2n+1} P^{2n}$$

$$\mathcal{B} = (1 - x_1) \left[c_4 - nx_1 - x_2 + \frac{m}{2}\eta_{\text{fuel}} \right]^n (1 + c_1 + c_2 + c_3 + c_4 + 2nx_1)^{2n}$$

$$\exp\left(-\frac{\Delta G_{\text{WGS}}(T)}{RT}\right) = \frac{P'_{\text{CO}_2} P'_{\text{H}_2}}{P'_{\text{CO}} P'_{\text{H}_2O}} = \frac{(c_2 + x_2) \left[c_3 + (2n + 1)x_1 + x_2 - \frac{m}{2}\eta_{\text{fuel}} \right]}{(c_1 + nx_1 - x_2) \left[c_4 - nx_1 - x_2 + \frac{m}{2}\eta_{\text{fuel}} \right]} \quad (18)$$

where $\Delta G_{\text{SR}}(T)$ and $\Delta G_{\text{WGS}}(T)$ are the Gibbs free energy changes of the SR and WGS reactions, respectively. P'_i is the partial pressure of reacting species i ($i = H_2, H_2O, CO, CO_2$ or C_nH_{2n+2}) and P is the total pressure of the fuel stream. Eq.(17) and Eq.(18) are used to determine the results of the two variables, x_1 and x_2 .

With x_1 and x_2 obtained by solving Eq.(17) and Eq.(18), $E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2})$ can be calculated as:

$$E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2}) = -\frac{\Delta G_0^{\text{H}_2}(T)}{2F} + \frac{RT}{2F} \ln \frac{\mathcal{C}}{\mathcal{D}} \quad (19)$$

$$\mathcal{C} = [c_3 + (2n + 1)x_1 + x_2 - \frac{m}{2}\eta_{\text{fuel}}] \cdot [P_{\text{O}_2}(1 - \eta_{\text{O}_2})]^{1/2}$$

$$\mathcal{D} = c_4 - nx_1 - x_2 + \frac{m}{2}\eta_{\text{fuel}}$$

Inserting Eq.(19) into Eq.(12) and Eq.(13), the maximum electrical efficiency for alkane fuel can be determined. Eq.(19) is also valid for H₂ fuel that corresponds to $n=0, x_1=1$ and $c_1=c_2=c_3=x_2=0$.

For the special case of $c_1=c_2=c_3=0$ (and $c_4=y$ and $m=2(3n+1)$), *i.e.*, the input fuel consists of C_nH_{2n+2} and H₂O only, Eq.(17)–Eq.(19) are reduced to:

$$\exp\left(-\frac{\Delta G_{\text{SR}}(T)}{RT}\right) = \frac{\mathcal{E}}{\mathcal{F}} \quad (20)$$

$$\mathcal{E} = (nx_1 - x_2)^n [(2n + 1)x_1 + x_2 - (3n + 1)\eta_{\text{fuel}}]^{2n+1} P^{2n}$$

$$\mathcal{F} = (1 - x_1) [y - nx_1 - x_2 + (3n + 1)\eta_{\text{fuel}}]^n (1 + y + 2nx_1)^{2n}$$

$$\exp\left(-\frac{\Delta G_{\text{WGS}}(T)}{RT}\right) = \frac{\mathcal{G}}{\mathcal{H}} \quad (21)$$

$$\mathcal{G} = x_2 [(2n + 1)x_1 + x_2 - (3n + 1)\eta_{\text{fuel}}]$$

$$\mathcal{H} = (nx_1 - x_2) [y - nx_1 - x_2 + (3n + 1)\eta_{\text{fuel}}]$$

$$E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2}) = -\frac{\Delta G_0^{\text{H}_2}(T)}{2F} + \frac{RT}{2F} \ln \frac{\mathcal{I}}{\mathcal{J}} \quad (22)$$

$$\mathcal{I} = [(2nx_1 + x_1 + x_2) - (3n + 1)\eta_{\text{fuel}}] \cdot [P_{\text{O}_2}(1 - \eta_{\text{O}_2})]^{1/2}$$

$$\mathcal{J} = y - nx_1 - x_2 + (3n + 1)\eta_{\text{fuel}}$$

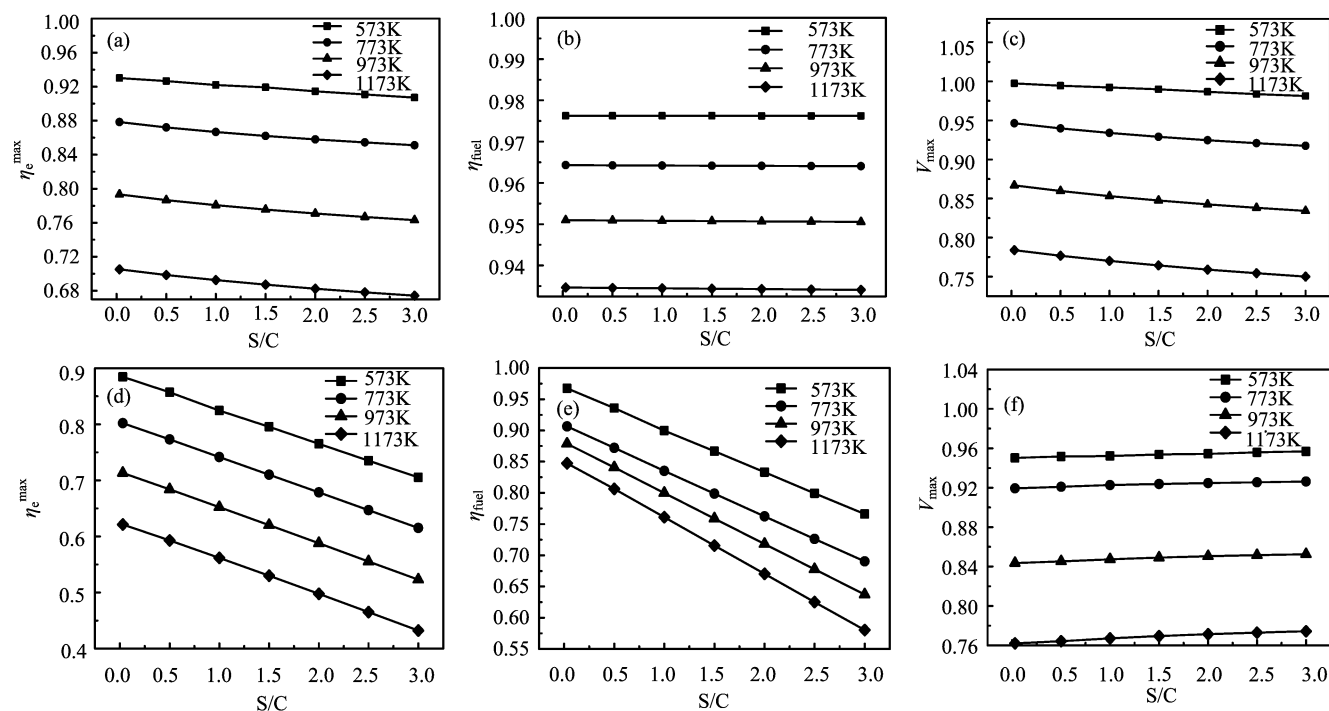


FIG. 2 η_e^{\max} and the corresponding η_{fuel} and V_{\max} as functions of S/C of methane fuel at different temperature T : (a) η_e^{\max} for $\alpha=1$, (b) η_{fuel} for $\alpha=1$, (c) V_{\max} for $\alpha=1$, (d) η_e^{\max} for $\alpha=0$, (e) η_{fuel} for $\alpha=0$, (f) V_{\max} for $\alpha=0$.

III. NUMERICAL RESULTS

The maximum electrical efficiency theory developed above is applied to fuel cell systems fueled by methane, propane, and hydrogen, assuming both the total pressures of fuel and air are both 1 atm. In most literatures the low heating value (LHV) of a fuel for the fuel cell efficiency was used [3, 6, 7, 13], the numerical results presented here are also based on $h_{\text{fuel}}(T_0)=\text{LHV}$. Conversion to high heating value (HHV) efficiency only requires a scaling factor of LHV/HHV . (LHV, HHV) in kJ/mol are (802, 890), (2043, 2219), and (241, 285) for CH_4 , C_3H_8 , and H_2 , respectively [16].

A. Maximum electrical efficiency of methane fuel

FIG. 2 shows the variations of the maximum electrical efficiency, the corresponding optimal fuel utilization and maximal cell operating voltage with the steam carbon ratio of S/C ranging from 0 to 3, of methane fuel for fuel cell systems of $\alpha=1$ and $\alpha=0$. The results are obtained based on Eqs. (12), (13), (20), (21), (22) using the thermodynamic property data in Ref.[16]. Low S/C methane is prone to coke formation, but the results are shown to indicate the potential benefit of reducing S/C to the fuel cell efficiency. Meanwhile, the results may be meaningful in practice as there are reports that the coke formation may be prevented when S/C is as low as 3% [17–20]. Similarly, results for low tempera-

ture are presented to illustrate the effect of operating temperature on the fuel cell efficiency.

FIG. 2(a) shows that η_e^{\max} decreases slowly and approximately linearly with the increase of S/C . The weak dependence of η_e^{\max} on S/C for $\alpha=1$ is due to that the residual heat of the exiting fuel stream is utilized for heating the input fuel. That is, an increase of S/C does not necessarily mean an increase of the heating requirement. Correspondingly, η_{fuel} is nearly independent of S/C , as shown in FIG. 2(b). Due to the constant η_{fuel} and the adverse effect of the steam content on the Nernst potential, V_{\max} decreases with the increase of S/C (FIG. 2(c)).

For the temperatures examined, η_e^{\max} decreases by about 1% when S/C is increased by 1. The benefit of increasing η_e^{\max} by decreasing S/C is rather limited for an ideal fuel cell system, which suffers the drawback of high propensity of coke formation. Therefore, a relatively high S/C methane that safely inhibits the coke formation is a natural choice of fuel for fuel systems with $\alpha \approx 1$.

Unlike the weak effect of S/C , the influences of T on η_e^{\max} , η_{fuel} , and V_{\max} are much more substantial, as seen in FIG. 2 (a)–(c). V_{\max} decreases with the increase of T due to the term $\Delta G_0^{\text{H}_2}(T)$ (Eq.(11)). As $E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2}=0)$ decreases with η_{fuel} faster for higher T (Eq.(21)), η_{fuel} that maximizes $\eta_{\text{fuel}} E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2}=0)$ is lower for higher T . The combined effect of the reduced $E_{\text{Nernst}}(T, \eta_{\text{fuel}}, \eta_{\text{O}_2}=0)$ and η_{fuel} results in a sharp decline of η_e^{\max} with the increase of T . For

TABLE II η_e^{\max} of methane and propane fuels for representative operating parameters of the fuel cell system.

T/°C	$\eta_e^{\max}(\text{CH}_4)/\%$				$\eta_e^{\max}(\text{C}_3\text{H}_8)/\%$			
	S/C=0.03		S/C=2		S/C=0.03		S/C=2	
	$\alpha=1$	$\alpha=0$	$\alpha=1$	$\alpha=0$	$\alpha=1$	$\alpha=0$	$\alpha=1$	$\alpha=0$
700	79.3	71.3	77.1	58.8	78.0	72.5	75.5	57.4
900	70.5	62.1	68.2	49.8	69.2	63.6	66.7	48.9

S/C=2, η_e^{\max} is 91.5%, 85.8%, 77.1%, and 68.2% for $T=300, 500, 700,$ and 900 °C, respectively.

Contrary to the common perception concerning methane fuel, reducing the fuel cell operating temperature is highly beneficial for improving the electrical efficiency of an ideal fuel cell system. For a realistic working temperature of 700–900 °C, η_e^{\max} is in the range of 68%–77%.

For $\alpha=0$, *i.e.*, the residual heat of the exhausted gases at the working temperature is not utilized, η_e^{\max} exhibits a strong dependence on S/C, as seen in FIG. 2(d). On average, η_e^{\max} decreases by about 6% with an increase of S/C by 1. Novel anodes designed to prevent the coke formation so that a low S/C methane fuel may be used [16, 17] are highly desirable for obtaining high η_e^{\max} . The decrease of η_e^{\max} with the increase of S/C is primarily associated with the decrease of η_{fuel} (FIG. 2(e)) so that the increased need of heating the fuel stream can be met by H_{ef} . Due to the sharp decline of η_{fuel} , V_{max} shows a slight increase with the increase of S/C (FIG. 2(f)).

Expectably, η_e^{\max} is strongly dependent on T for $\alpha=0$. Interestingly, for a given S/C, the temperature dependence of η_e^{\max} for $\alpha=0$ is only slightly stronger than that for $\alpha=1$. For example, for S/C=0.03, η_e^{\max} for $\alpha=0$ decreases from 71.3% to 62.1% when T is increased from 700 °C to 900 °C, *i.e.*, a decrease of 9.2%. In comparison, the corresponding change for $\alpha=1$ is 8.8%.

The combined effect of S/C and T on η_e^{\max} of $\alpha=0$ is very significant. For a practical operating condition of S/C=2 and $T=700$ – 900 °C, $\eta_e^{\max}(\alpha=0)$ is in the range of 50%–59%. The result of $\eta_e^{\max}(\alpha=1) - \eta_e^{\max}(\alpha=0) \approx 18\%$ for the operating condition is startling, indicating the extreme importance of the thermal management for the electrical efficiency of the fuel cell system.

FIG. 3 shows the effect of α on η_e^{\max} for S/C=2 and $T=500, 700,$ and 900 °C. η_e^{\max} increases rapidly with α for $\alpha \leq 0.5$, but the rate of increase slows down substantially thereafter. This is attributed to the fact that once more than half of ΔH_{ex} is utilized, the heating requirement can be met without a much loss of η_{fuel} . For S/C=2 and $T=700, 900$ °C, $\eta_e^{\max}(\alpha=1) - \eta_e^{\max}(\alpha=0.5) \approx 3\%$ is observed. That is, over 80% of the loss in η_e^{\max} with $\alpha=0$ is recovered with $\alpha=0.5$. Compared to the impossible system design of $\alpha=1$, $\alpha=0.5$ appears to be a worthwhile design goal.

While $\alpha=1$ is too idealistic, $\alpha \geq 0$ might be achiev-

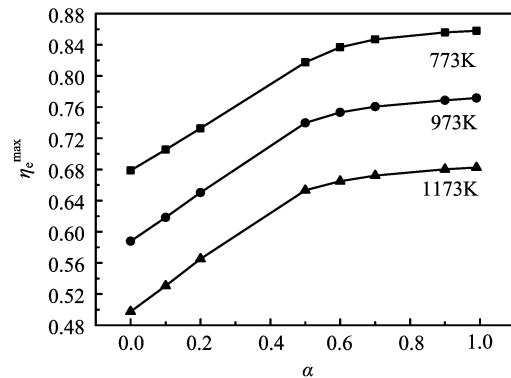


FIG. 3 η_e^{\max} for methane of S/C=2 as a function of α for $T=773, 973$ and 1173 K.

able in practice. For S/C=2, $T=700$ °C, and $\alpha=0$, $\eta_e^{\max}=59\%$, $\eta_{\text{fuel}}=72\%$, and $V_{\text{max}}=0.85$ V are found. $\eta_{\text{fuel}}=72\%$ is quite practical. Together with a realistic operating voltage of $V_{\text{op}}=0.75$ V, η_e is found to be 52%. As H_{cell} is higher for $V_{\text{op}}=0.75$ V than that for $V_{\text{max}}=0.85$ V, the heating requirement met with V_{max} is automatically met with V_{op} . It is therefore concluded that an overall electrical efficiency of more than 50% is realistic for a fuel cell system.

B. Maximum electrical efficiency of propane fuel

FIG. 4 shows the variations of η_e^{\max} , η_{fuel} , and V_{max} with S/C of propane fuel at different temperature T for $\alpha=1$ and $\alpha=0$. Comparison of FIG. 2 and FIG. 4 shows that the variation patterns for methane and propane fuels are basically the same. The discussion made above for methane applies equally well to propane. To see their subtle differences more clearly, representative data of η_e^{\max} for methane and propane fuels are displayed in Table II.

As can be seen in Table II, η_e^{\max} of methane is larger than that of propane in most cases. For S/C=2 and 700 °C $\leq T \leq 900$ °C, η_e^{\max} of CH_4 is higher than that of C_3H_8 by 0.9%–1.6%. However, η_e^{\max} of CH_4 with low S/C shows a stronger α dependence than that of C_3H_8 counterpart. For S/C=0.03, $\alpha=0$ and 700 °C $\leq T \leq 900$ °C, η_e^{\max} of CH_4 is lower than that of C_3H_8 by 1.2%–1.5%. As S/C ≈ 2 is more practical, however, it is roughly correct to say that η_e^{\max} of CH_4 is about 1% higher than that of C_3H_8 .

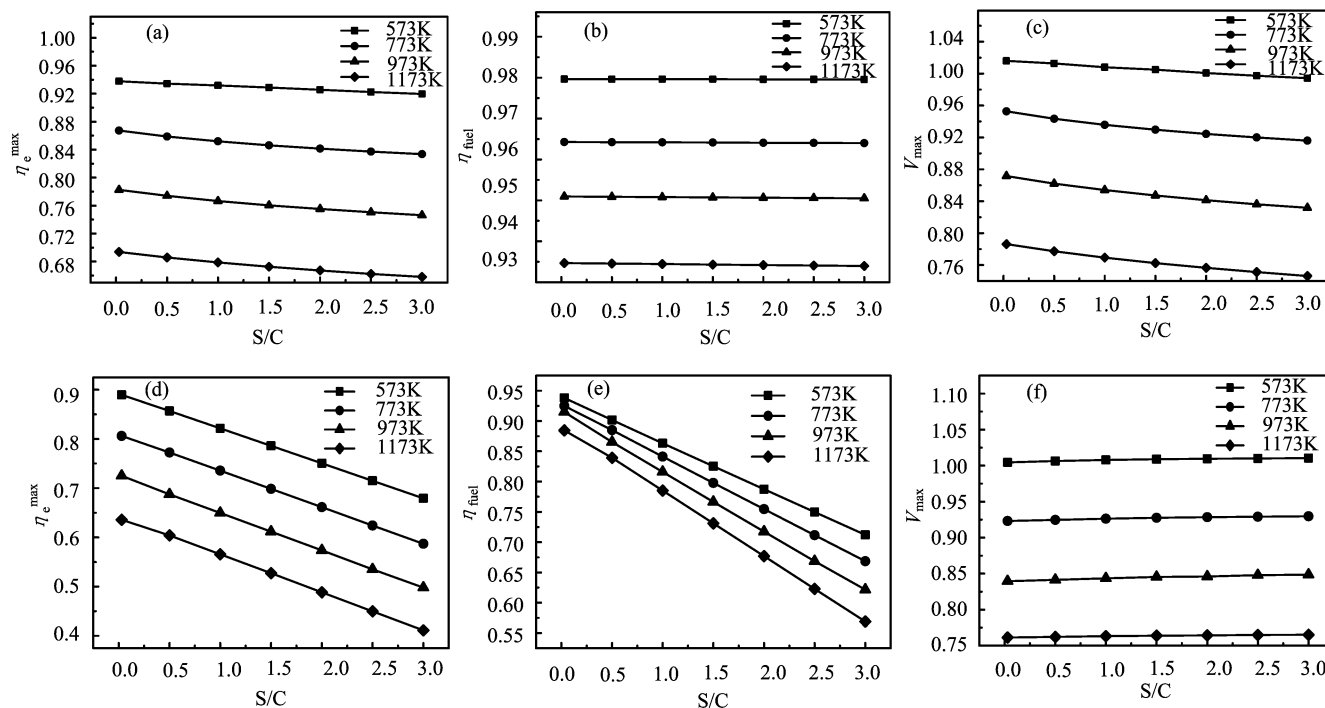


FIG. 4 η_e^{\max} and the corresponding η_{fuel} and V_{max} as functions of S/C of propane fuel at different temperature T : (a) η_e^{\max} for $\alpha=1$, (b) η_{fuel} for $\alpha=1$, (c) V_{max} for $\alpha=1$, (d) η_e^{\max} for $\alpha=0$, (e) η_{fuel} for $\alpha=0$, (f) V_{max} for $\alpha=0$.

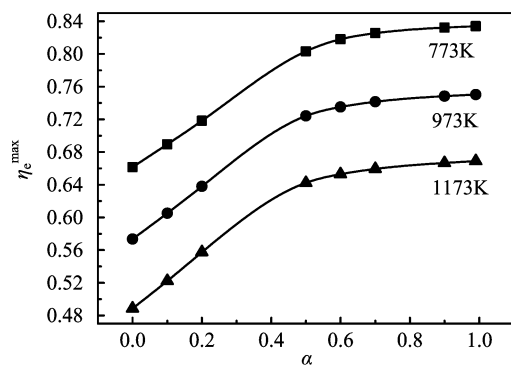


FIG. 5 η_e^{\max} for propane fuel of S/C=2 as a function of α for $T=773$, 973, and 1173 K.

FIG. 5 shows the effect of α on η_e^{\max} of propane with S/C=2 and $T=500$, 700 and 900 °C. As can be seen by comparing FIG. 5 and FIG. 3, the α dependences of η_e^{\max} of propane and methane are basically the same and over 80% of the loss in η_e^{\max} with $\alpha=0$ is recovered with $\alpha=0.5$. Similarly, it is found that an overall electrical efficiency of over 50% is expectable for a propane fueled fuel cell system.

C. Maximum electrical efficiency of hydrogen fuel

Hydrogen is the simplest fuel suitable for all fuel cell operating temperatures. FIG. 6 shows the variations

of η_e^{\max} and the corresponding η_{fuel} as functions of H_2 molar fraction β , $\beta=N_{\text{H}_2}/(N_{\text{H}_2}+N_{\text{H}_2\text{O}})$ for $\alpha=1$ and $\alpha=0$ and 100, 300, 500, 700, and 900 °C.

Considering $T=100$ °C, FIG. 6 (a) and (c) show that the effect of the steam content on η_e^{\max} is very limited. For $\alpha=1$, η_e^{\max} for $\beta=97\%$ is only 0.6% higher than that for $\beta=60\%$. For $\alpha=0$, the corresponding difference in η_e^{\max} increases to 1.4%, still a relatively small amount. The difference between $\eta_e^{\max}(\alpha=1)$ and $\eta_e^{\max}(\alpha=0)$ is a negligible amount of 0.1% for $\beta=97\%$. For $T<100$ °C, as encountered in PEM fuel cell operations, neither the water content nor the thermal management is important factor affecting η_e^{\max} .

Expectably, the effect of β on η_e^{\max} increases with the temperature T . For $\alpha=0$ and $T=900$ °C, $\eta_e^{\max}(\beta=97\%)-\eta_e^{\max}(\beta=60\%)=8\%$ is observed. However, the result is likely purely theoretical as only high β ($\sim 97\%$) H_2 fuel may be used when $T\gg 100$ °C [1, 21]. For $\beta=97\%$, the difference between $\eta_e^{\max}(\alpha=1)$ and $\eta_e^{\max}(\alpha=0)$ increases with the temperature T , but remains relatively small. For example, $\eta_e^{\max}(\alpha=1)-\eta_e^{\max}(\alpha=0)$ is 1.6% and 2.3% for $T=700$ and $T=900$ °C, respectively. Overall, the difference between $\eta_e^{\max}(\alpha=1)$ and $\eta_e^{\max}(\alpha=0)$ is limited for H_2 fuel operating at all temperatures.

Although the effect of α on η_e^{\max} is moderate for all temperatures, the effect of T on η_e^{\max} is much more substantial, due primarily to the decrease of $|\Delta G_0^{\text{H}_2}(T)|$ with the increase of T . For $\beta=97\%$, η_e^{\max} decreases linearly with the increase of T . For an increase of T of

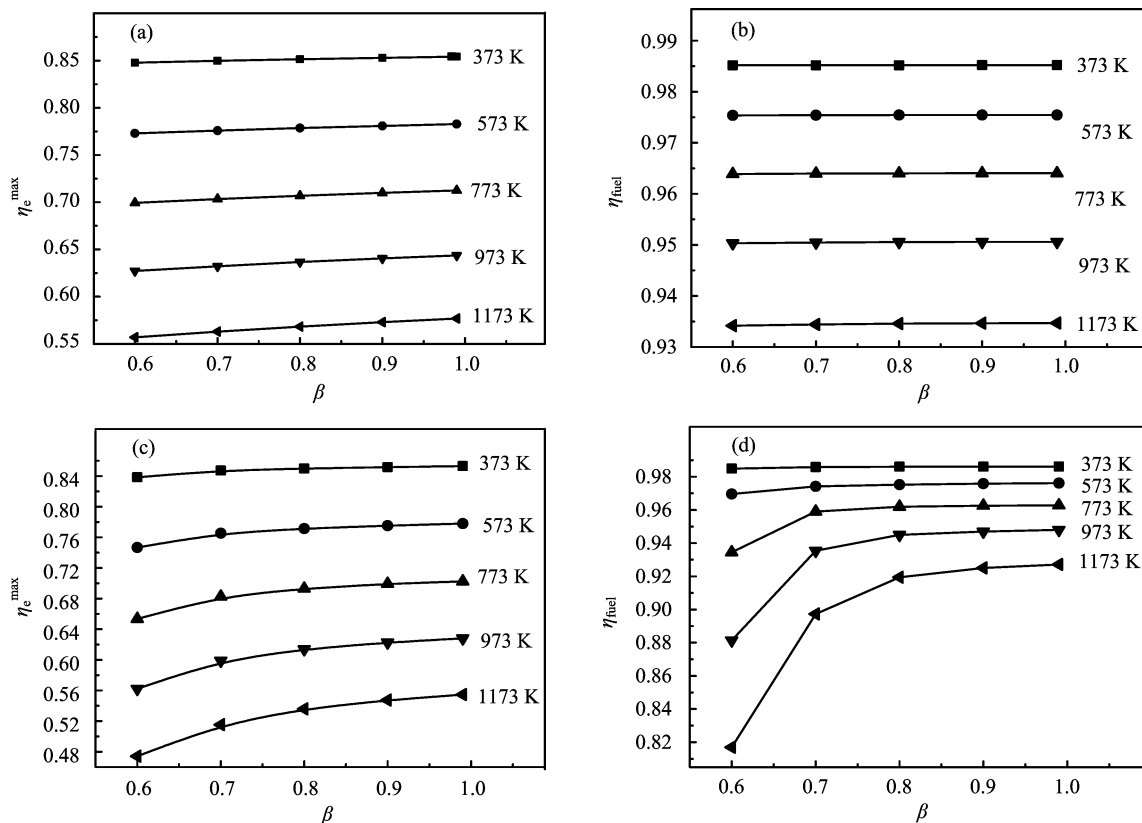


FIG. 6 η_e^{\max} and the corresponding η_{fuel} as functions of the hydrogen molar fraction β in hydrogen fuel at different temperature T : (a) η_e^{\max} for $\alpha=1$, (b) η_{fuel} for $\alpha=1$, (c) η_e^{\max} for $\alpha=0$, (d) η_{fuel} for $\alpha=0$.

100 K, η_e^{\max} decreases by 3.5% for $\alpha=1$ and 3.7% for $\alpha=0$.

Considering $700\text{ }^\circ\text{C} \leq T \leq 900\text{ }^\circ\text{C}$, η_e^{\max} for H_2 fuel with $\beta=97\%$ and $\alpha=1$ is in the range of 58%–64%. In comparison, η_e^{\max} for CH_4 fuel with $\text{S/C}=2$ and $\alpha=1$ is in the range of 68%–77%. That is, $\eta_e^{\max}(\alpha=1, \text{H}_2, \beta=97\%)$ is less than $\eta_e^{\max}(\alpha=1, \text{CH}_4, \text{S/C}=2)$. However, $\eta_e^{\max}(\alpha=0, \text{H}_2, \beta=97\%)$ is in the range of 55%–63% and is more than $\eta_e^{\max}(\alpha=1, \text{CH}_4, \text{S/C}=2)$ that is in the range of 50%–59%.

Notice that $\alpha=0$ means that the heat value of $H_{\text{cell}}+H_{\text{ef}}$ has been utilized, though ΔH_{ex} is wasted. That is, $\alpha=0$ corresponds to a reasonable level of thermal management in a fuel cell system. Therefore, except for a fuel cell system with a high quality thermal design, H_2 fuel is expected to have a higher η_e^{\max} than that of CH_4 fuel for a system operated at $700\text{ }^\circ\text{C} \leq T \leq 900\text{ }^\circ\text{C}$.

IV. CONCLUSION

Explicit expressions for the maximum electrical efficiency of a fuel cell system are derived by applying the fundamental thermodynamics principle. The result for an ideal fuel cell system is analogous to the Carnot theorem for an ideal heat engine. Unlike the Carnot

efficiency, η_e^{\max} is fuel specific.

For an ideal fuel cell system fueled by hydrogen, η_e^{\max} is given by a single equation. For a realistic fuel cell system characterized by an effective heat exchange parameter, η_e^{\max} for hydrogen fuel is given by solving a set of two equations. Chemical equilibrium calculations are required to determine η_e^{\max} for a general fuel. Other than the equilibrium calculations, η_e^{\max} for a general fuel is similar to the case of hydrogen fuel and given by one or two equations for an ideal or non-ideal fuel cell system, respectively. Chemical equilibrium is considered for a class of widely available fuel sources, alkane. Analytical solutions of the chemical equilibrium are obtained by assuming the fuel stream only consisting of $\text{C}_n\text{H}_{2n+2}$, H_2 , H_2O , CO , and CO_2 .

The theoretical models can be used to conveniently analyze the impacts of various factors on the maximum electrical efficiency. Numerical results are presented for methane, propane, and hydrogen fuels. η_e^{\max} decreases with the increase of T for all fuels examined and for both $\alpha=1$ and $\alpha=0$. For an ideal system, the steam content does not affect η_e^{\max} appreciably. For $\alpha=0$, however, η_e^{\max} decreases substantially with the increase of S/C of the alkane fuel. The thermal management is critically important for the obtainable electrical efficiency of a fuel cell system fueled by methane

or propane. Nevertheless, most of the efficiency loss with $\alpha=0$ can be recovered with $\alpha=0.5$ and a system design with $\alpha\approx 0.5$ is therefore recommended. For H_2 fuel, however, the thermal management is less critical and $\alpha=0$ is quite acceptable.

For $700\text{ }^\circ\text{C}\leq T\leq 900\text{ }^\circ\text{C}$ and a representative steam content, $S/C=2$ for CH_4 and C_3H_8 and $\beta=97\%$ for H_2 , η_e^{\max} for CH_4 , C_3H_8 , and H_2 with $\alpha=1$ ($\alpha=0$) are in the range of $68\%–77\%$ ($50\%–59\%$), $66\%–76\%$ ($49\%–57\%$) and $58\%–64\%$ ($55\%–63\%$), respectively. That is, η_e^{\max} for CH_4 and C_3H_8 are higher than that of H_2 for an ideal system. For a realistic system of $\alpha=0$, however, hydrogen fuel can have a higher efficiency than that of methane and propane for the operating temperature of SOFCs.

V. ACKNOWLEDGMENTS

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