ABSTRACT: A solid oxide fuel cell (SOFC) has wide stationary and mobile application prospects due to its high efficiency and fuel flexibility. The SOFC system’s performance depends on the reforming option and system design. In this paper, we designed a novel SOFC auxiliary power unit (APU) system with ethanol onboard reforming aiming at vehicle application. The thermodynamic analysis is employed to evaluate the ethanol-fueled SOFC performance of different reforming options with a metal-supported SOFC working at 600 °C and a 0.3 A/cm² current density. The electrical efficiency of the SOFC can reach a maximum of 50% with ethanol autothermal reforming. Under the optimal reforming option and operating conditions, the conceptual SOFC-APU system design is identified with the trade-off between system efficiency and ethanol flow from the startup and stable operation phase. The results show that the system efficiency of 44.4% can be achieved with a 0.42 g/s ethanol flow at the startup phase. During the stable operation, the electrical efficiency and exergy efficiency of the SOFC-APU system can reach 55.4 and 77.1% with a 70% anode gas recirculation ratio, respectively.

1. INTRODUCTION

With the rapid economic development, the demand for fossil fuels has also increased. The consumption of fossil energy not only faces a sharp decline in reserves but also serious environmental pollution. The number of internal combustion engine vehicles fueled by petroleum and other primary energy sources has been increasing year by year, and the fixed-source power generation dominated by coal has severely increased the world energy crisis. Therefore, vigorously developing new energy has become one of the important strategies for countries to transform their energy structure. A fuel cell is a high-efficiency and environmentally friendly power generation device with a higher energy density than traditional chemical energy facilities. Among them, the solid oxide fuel cell (SOFC) has developed into a mature technology with wide fuel adaptability. High-grade heat energy while generating electricity has attracted more interest in mobile applications.

The plate SOFC can be divided into electrolyte-supported (over 800 °C), anode-supported (650–800 °C), and metal-supported (450–600 °C). The metal-supported SOFC uses cerium gadolinium oxide as the electrolyte and a porous anode layer with the metal substrate. Ceres Power company reveals that its metal-supported SOFC uses gadolinium-doped ceria as the electrolyte, a ceria–nickel cermet as the anode, lanthanum perovskite as the cathode, and laser-drilled ferritic steel as the substrate. Fast startup capability and high mechanical robustness make it a promising candidate for mobile applications, especially the SOFC-battery hybrid power system can lower the vehicle weight and extend the driving range with fast refueling.

In mobile applications, liquid fuel can operate the SOFC system directly with easy storage and transportation, reducing the system size and complexity. Without depending on the hydrogen infrastructure, the SOFC system has safer and cheaper refueling than the hydrogen fuel cell system. In addition, the waste heat generated by the SOFC can be used in the endothermic steam-reforming process and cabin heating, avoiding the trouble of cold start in winter and proton membrane humidity maintenance in hot climates. Compared with the traditional internal combustion engine (ICE) vehicles, the SOFC-APU system has higher efficiency and lower noise and vibration with silent running. Less emission (including NOx and SOx) makes it environmentally friendly. The pure electric vehicle is restricted to the battery energy density with a short driving range. The SOFC-APU system can improve the driving range due to the fuel adaptability and replenishment convenience. However, there are still big challenges ahead for

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the SOFC-APU system. A high operating temperature and stack heat capacity increase the startup time. Another significant obstacle that hinders achieving commercialization is the high production cost with the complicated preparation process. The system durability and thermal management need to be enhanced further.5

Fuel flexibility is an appealing advantage for SOFC vehicles. Bioethanol is considered to be the most potential substitute for gasoline in the transport sector, contributing to the reduction of carbon emissions and energy agriculture development.10,11 Bioethanol, as an effective hydrogen carrier, is mainly produced from biomass fermentation and can be directly fueled for SOFC. However, the SOFC internal reforming system is prone to a pyrolysis reaction with carbon deposition at high temperatures.12 In addition, the endothermic reforming reaction can break the electrode sheet due to a large temperature gradient, affecting the SOFC performance and service life.13 An external reformer is widely used to reduce the SOFC carbon deposition and temperature gradient without affecting the original structure, ensuring stable operation. The performance of a SOFC system with an external reformer depends on the fuel reforming process and operating conditions.

Ethanol reforming to produce hydrogen for a SOFC is regarded as a significant technology routine, including steam wet reforming, carbon dioxide dry reforming, partial oxidation reforming, and autothermal reforming. Among them, steam wet reforming technology is a mature and efficient way.14 A thermodynamic analysis of the steam ethanol reforming-SOFC system was conducted, showing the highest hydrogen production rate at 973 K with a steam to carbon ratio of 1.15 A SOFC system simulation with steam—ethanol reforming was performed and it reached the highest efficiency of 83.9% theoretically over 1100 K.16 The carbon dioxide dry reforming technology can exploit CO2 to produce hydrogen without additional water intake, improving the living environment of human beings. A thermodynamic analysis of a SOFC system operating with ethanol dry reforming was carried out.17 The results showed that ethanol dry reforming can avoid coking at 973 K and 28.7% fuel utilization with high ratios of CO2/ethanol. The partial oxidation reforming reaction of ethanol is a slightly endothermic reaction with a less hydrogen production rate than steam reforming. However, the problem of pure oxygen supply needs to be solved. The nickel–zirconium–cerium composite ceramic material for the catalytic partial oxidation of ethanol in a SOFC system can reach 88.9% hydrogen selectivity at 700 °C and 692 mW cm−2 peak power density.18 The energy analysis of ethanol partial oxidation reforming was performed with 55.60% SOFC electrical efficiency (LHV) at 1150 K and a 0.6 oxygen/ethanol ratio.19 The autothermal reforming technology can provide heat for the startup and load requirement operation without additional heat sources, reducing the system’s energy consumption and shortening the startup time. Especially for fast-start mobile applications, ethanol autothermal reforming is an excellent route for on-board hydrogen production. Nissan has announced the world’s first SOFC vehicle with bioethanol autothermal reforming in June 2016 and completed the first period of testing in Brazil.20-22

The major problems of these published reports are the following: (1) the performance of the metal-supported SOFC with different ethanol-reforming options is not discussed in detail. (2) The complete mobile SOFC-APU system is not designed in detail. (3) The overall performance of the mobile SOFC-APU system is not discussed in detail to balance the system efficiency and operating conditions.

The paper is organized as follows: In Section 2, process modeling including the metal-supported SOFC and ethanol reforming process are provided. In Section 3, the SOFC system with different ethanol-reforming options and a conceptual SOFC-APU system design are demonstrated. Then, in Section 4, the metal-supported SOFC model is validated first, and the performance comparison of different reforming processes is discussed. The overall performance of the designed SOFC-APU system is analyzed. Finally, the conclusions are drawn in Section 5.

2. DESCRIPTION OF THE METAL-SUPPORTED SOFC MODEL

2.1. Open-Circuit Voltage. According to the thermodynamics law, the electronic transmission amount is equal to the Gibbs free energy change ΔG in the chemical reaction. The electromotive force (UNernst) corresponding to the pressure (P) and temperature (T) is

\[
U_{\text{Nernst}} = \frac{-\Delta G^0(T)}{2F} + \frac{RT}{2F} \ln \left( \frac{P_{H_2}P_{O_2}^{1/2}}{P_{H_2O}} \right) = U_0^{\text{Nernst}} + \frac{RT}{4F} \ln \frac{P_{O_2}P_{H_2}^2}{P_{H_2O}^2}
\]

(1)

where \( R \) is the gas constant (J/mol-K), \( T \) is the reaction temperature (K), and \( P_{H_2} \), \( P_{O_2} \), and \( P_{H_2O} \) are the partial pressures of hydrogen and oxygen (bar), respectively. \( F \) is the Faraday constant, representing the charge per mole of an electron (C/mol), and \( F = 96 485 \text{ C/mol}^{-1} \). \( U_0^{\text{Nernst}} \) is the open-circuit voltage at standard pressure and is a function of the operating temperature.23

\[
U_0^{\text{Nernst}} = 1.2723 - 2.7645 \times 10^{-4}T
\]

(2)

The actual SOFC output voltage is not always equal to the electrochemical potential calculated by the Nernst equation due to the internal voltage losses encountered in real operation.24 In this paper, the influence of concentration overpotentials on the metal-supported SOFC voltage is ignored,25 and the output voltage is

\[
V_{\text{cell}} = U_{\text{Nernst}} - E_{\text{ohm}} - E_{\text{act}}
\]

(3)

where \( E_{\text{ohm}} \) is the ohmic overpotentials and \( E_{\text{act}} \) is the activation overpotentials.

2.2. Ohmic Overpotentials. Ohmic overpotentials arise from the conduction resistance of materials to ions or electrons. Specifically, the ionic conductance in the electrolyte, the electronic resistance in the electrode and the connecting body, and the contact resistance between the cell elements can lead to polarization. Ohmic overpotentials can be expressed as

\[
R_{\text{ohm}} = \frac{r_{\text{anode}}}{\sigma_{\text{anode}}} + \frac{r_{\text{cathode}}}{\sigma_{\text{cathode}}} + \frac{r_{\text{el}}}{\sigma_{\text{el}}} + R_{\text{cont}}
\]

(4)

where \( r_{\text{anode}}, r_{\text{cathode}}, \) and \( r_{\text{el}} \) represent the thickness of the anode, electrolyte, and cathode layers; \( \sigma_{\text{anode}} \) and \( \sigma_{\text{cathode}} \) are the electronic conductivities of the anode and cathode, respectively, and \( \sigma_{\text{el}} \) is the ionic conductivity of the electrolyte. \( R_{\text{cont}} \) is the contact resistance between the cell elements. The physical parameters of the SOFC components are listed in Table 1.
The material of the anode functional layer is similar to the cathode and anode exchange current can be expressed as\textsuperscript{26} the activation overpotentials $E_{\text{act},i}$ can be derived as follows

$$E_{\text{act},i} = \frac{2 \cdot R \cdot T}{n_{i} F} \sinh^{-1} \left( \frac{i}{2 \cdot j_{0,i}} \right)$$

where $n_{i}$ is the number of electrons transferred during the reaction, and the value is 2 on the anode side and 4 on the cathode side; if $j_{0,i}$ is the exchange current density (A/cm\textsuperscript{2}), then the cathode and anode exchange current can be expressed as\textsuperscript{26}

$$j_{0,c} = K_{C} \frac{R \cdot T}{2F} \exp \left( \frac{-E_{AC}}{R \cdot T} \right)$$

$$j_{0,a} = (P_{H_{2}})^{0.5} \frac{K_{A}}{R \cdot T^2} \exp \left( \frac{-E_{AA}}{R \cdot T} \right)$$

The metal-supported SOFC anode mainly comprises the metal support body and anode functional layer, as shown in Figure 1. The porous metal substrate can support the cell structure and adjust the thickness according to the mechanical requirements, providing a fuel transmission channel and increasing the current conduction efficiency due to the metal materials’ conductivity. The material of the anode functional layer is similar to the anode-supported SOFC, adopting a porous nickel–cerium gadolinium oxide (CGO) composite cermet. The electrolyte layer is deposited on the anode with a dense CGO film, and the cathode layer is deposited over the electrolyte, consisting of a thin lanthanum strontium cobalt ferrite (LSCF)/CGO composite layer.\textsuperscript{25} The thinner film with smaller material particles increases the area of the three-phase active reaction zone significantly and reduces the polarization loss, improving the output characteristics and oxidation–reduction reaction stability greatly.

The partial pressure of hydrogen $P_{H_{2}}^{\text{TPB}}$ at the interface can be expressed as\textsuperscript{25}

$$P_{H_{2}}^{\text{TPB}} = \frac{P_{H_{2}} \cdot (\tau_{\text{holepitch}} + \tau_{\text{substrate}})}{2 \cdot F \cdot D_{\text{anode}}^{\text{diffusion}}}$$

### 2.3. Activation Overpotentials.

The electrode reaction proceeds at a certain rate, which is determined by the rate-determining step. The reaction energy barrier of the rate-determining step needs to be overcome by additional energy. The necessary potential energy is defined as the activation potential, which is related to the speed of the electrochemical reaction. The activation overpotential becomes the most important reason for the metal-supported voltage drop with the decreased operating temperature.\textsuperscript{26} The activation overpotentials $E_{\text{act},i}$ can be derived as follows

$$E_{\text{act},i} = \frac{2 \cdot R \cdot T}{n_{i} F} \sinh^{-1} \left( \frac{j}{2 \cdot j_{0,i}} \right)$$

### 2.4. Key Parameters.

The inlet flow of the SOFC affects the whole system efficiency, which can be deduced from eq \textsuperscript{11}\textsuperscript{27}

$$FH_{2,\text{eq}} = \left( \frac{j_{\text{cell}} \cdot A_{\text{cell}} \cdot N_{\text{cell}}}{F \cdot n_{\text{cell}} \cdot UFF} \right)$$

where $A_{\text{cell}}$ is the SOFC active surface area, $N_{\text{cell}}$ is the number of SOFCs, and $UFF$ is the factor of fuel utilization.

The anode off-gas recirculation ratio is defined as

$$RR = \frac{n_{\text{rec}}}{n_{\text{off}}}$$

where $n_{\text{rec}}$ is the molar flow rate recirculated toward the fuel processing block and $n_{\text{off}}$ is the anode exhaust molar flow rate.

### 2.5. Ethanol Reforming Theory.

Ethanol can be converted to hydrogen through different reforming processes: steam ethanol reforming (SER), carbon dioxide dry ethanol reforming (DER), partial oxidation ethanol reforming (OER), and autothermal ethanol reforming (AER), featured by a proper operational method and energy demand.\textsuperscript{28–30}

Steam ethanol reforming

$$C_{2}H_{5}OH + H_{2}O \rightarrow 2CO + 4H_{2} ; \Delta H = +278.5 \text{ kJ/mol}$$

Carbon dioxide dry ethanol reforming

$$C_{2}H_{5}OH + CO_{2} \rightarrow 2CO + 3H_{2} ; \Delta H = +296.8 \text{ kJ/mol}$$

Partial oxidation ethanol reforming

$$C_{2}H_{5}OH + \frac{1}{2}O_{2} \rightarrow 2CO + 3H_{2} ; \Delta H = +30.6 \text{ kJ/mol}$$

Autothermal ethanol reforming

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**Table 1. Physical Parameters of Cell Components**\textsuperscript{25}

| Parameter                | Description                  | Value     | Unit  |
|--------------------------|------------------------------|-----------|-------|
| $R$                      | Ideal gas constant           | 8.314     | J mol\textsuperscript{-1} K\textsuperscript{-1} |
| $t_{\text{anode}}$       | Anode thickness              | 1.5 \times 10\textsuperscript{-5} | m     |
| $t_{\text{cathode}}$     | Cathode thickness            | 5.0 \times 10\textsuperscript{-5} | m     |
| $t_{\text{el}}$          | Electrolyte thickness        | 1.5 \times 10\textsuperscript{-5} | m     |
| $\sigma_{\text{anode}}$  | Anode electrical conductivity | 8.0 \times 10\textsuperscript{4}  | S/m   |
| $\sigma_{\text{cathode}}$| Cathode electrical conductivity| 8.4 \times 10\textsuperscript{3} | S/m   |
| $R_{\text{cont}}$        | Contact resistance           | 8.46 \times 10\textsuperscript{-6} | $\Omega \cdot$ m\textsuperscript{2} |
| $K_{C}$                  | Ionic conductivity pre-exponential factor | 2.706 \times 10\textsuperscript{6} | S/K/m |
| $K_{A}$                  | Cathode pre-exponential factor | 7 \times 10\textsuperscript{11} | S/m\textsuperscript{2} |
| $E_{\text{AC}}$          | Cathode activation energy    | 1.309 \times 10\textsuperscript{3} | J/mol |
| $K_{A}$                  | Anode pre-exponential factor | 3.2 \times 10\textsuperscript{15} | S/m-bar\textsuperscript{0.5} |
| $E_{\text{AA}}$          | Anode activation energy      | 1.294 \times 10\textsuperscript{3} | J/mol |
| $\tau_{\text{holepitch}}$| Substrate hole spacing       | 1.25 \times 10\textsuperscript{-4} | m     |
| $\tau_{\text{substrate}}$| Substrate thickness         | 3.0 \times 10\textsuperscript{-4} | m     |
| $D_{\text{anode}}^{\text{diffusion}}$ | Anode activation diffusivity | 1.495 \times 10\textsuperscript{-6} | m\textsuperscript{2}/S |

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**Figure 1.** Schematic of the diffusion path of a metal-supported SOFC cell.
The autothermal reforming process becomes less endothermic in the range of 0–0.6 by increasing the oxygen amount \((AH = 4.4 \text{ kJ/mol})\), while the reaction reaches thermoneutral conditions at \(x = 0.6\) with a and the external reformer are shown steam/ethanol ratio of 1.8.\(^{31}\)

\[
\text{C}_2\text{H}_5\text{OH} + x\text{O}_2 + (3 - 2x)\text{H}_2\text{O} \\
\quad \rightarrow (6 - 2x)\text{H}_2 + 2\text{CO}_2 \quad (16)
\]

The autothermal reforming process becomes less endothermic in the range of 0–0.6 by increasing the oxygen amount \((AH = 4.4 \text{ kJ/mol})\), while the reaction reaches thermoneutral conditions at \(x = 0.6\) with a and the external reformer are shown steam/ethanol ratio of 1.8.\(^{31}\)

### 3. Description and Technology

#### Specifications of the SOFC Power System

**3.1. Ethanol-SOFC System with Different Reforming Processes.** The ethanol-SOFC system with different reforming processes is modeled through a steady-state, lumped parameter approach implemented in Aspen Plus. The state equation in the calculation is based on the Peng–Robinson (PEN–ROB) method to predict the behavior of hydrocarbons, and the chemical reactions involved are based on the RGibbs reactor to predict the composition under chemical equilibrium.\(^{23}\) The dry and steam ethanol reforming and water–gas shift reactions are considered. The external reformer is regarded as an isothermal reactor modeled by a Requil reactor to simulate the gas composition under chemical equilibrium.\(^{32}\) The afterburner boiler is modeled by an RGibbs model under isothermal and equilibrium conditions.\(^{33}\) The equilibrium composition is determined by the minimization of the Gibbs free energy, and the gas thermodynamic properties are solved by the Peng–Robinson equation of state.\(^{19}\) The work required by auxiliaries is calculated from theenthalpy difference and the isentropic efficiency, which is 0.8 for pumps and compressors.\(^{33}\)

The ethanol and gas flow mix before entering the external reformer and produce syngas when the mixture reaches the external reformer temperature. Then, the syngas reaches the anode inlet temperature and enters the stack, as shown in Figure 2. At the anodes, both chemical reactions and electrochemical reactions take place. Two reactors are placed to represent these reactions with the first handling the chemical reactions of internal reforming and water–gas shift reaction, and the second handling the electrochemical reactions of steam splitting. The number of a single cell \((N_{cell})\) is set to 1650 to ensure the nominal 5 kW output power. The current density is set to 0.3 A/cm\(^2\), which is a typical operating parameter.\(^{25}\) The rated operating conditions of the SOFC and the external reformer are shown in Tables 2 and 3.

#### 3.2. Concept of the Ethanol-Feed SOFC-APU Demonstration.** The SOFC-APU power system is mainly composed of a heat exchanger, start-burner, reformer, SOFC, after-burner, and other auxiliary components, as shown in Figure 3. In a practical SOFC system, the hot components are supposed to be insulated. The reformer, SOFC stack, start-burner, heat exchanger, and after-burner are the heat exchange equipment for the entire system and can be placed in an insulated box, known as the hot box. Other necessary pieces of equipment, such as fuel pumps and compressors, have less impact on the system heat management, which are called the cold box.

Three flows mix before entering the external reformer: (i) ethanol–water, (ii) air supply, and (iii) the recirculated hot anode exhaust gases. The mixture enters into the SOFC anode after autothermal reforming. The anode exhaust (line 7) divides into two parts: The first fraction is recirculated, while the second fraction is burnt in the after-burner. The air is heated and enters the cathode to serve as the reactant for the cathode reduction reaction. The hot air discharged from the cathode goes to the after-burner and contains the oxygen necessary to burn the anode exhaust in the after-burner. The after-burner exhaust is recycled to heat other components and leave the power system. Particularly, the fuel and air are transported into the start-burner for the system heating at the start phase. The ethanol-blended water can lower the ethanol production cost from an economic point of view\(^{26}\) and makes the combustion reactions occur in the liquid and gas phase with a lower temperature as well as NOx emissions and combustion noise,\(^{25,36}\) which has been tested at the AVL company.\(^{37}\)

#### 3.3. Energy and Heat Evaluation.** The SOFC electrical efficiency \((\eta_{elec})\), system’s electrical efficiency \((\eta_{elec,sys})\), and system’s thermal efficiency \((\eta_{t,sys})\) are used to evaluate the system design points, which are defined as follows

\[
\eta_{elec} = \frac{W_{\text{SOFC}}}{m_{\text{EthOH}}^{\text{in}} \cdot \text{LHV}_{\text{EthOH}}} \quad (17)
\]

where \(W_{\text{SOFC}}\) is the electrical power, which is produced from the fuel cell. \(m_{\text{EthOH}}^{\text{in}}\) is the mass flow of ethanol consumed by the system with the lower heating value (LHV) of ethanol equal to 26.7 MJ/kg.\(^{33}\)

| Parameter                  | Value | Unit |
|----------------------------|-------|------|
| SOFC operating pressure    | 1.05  | bar  |
| SOFC operating temperature | 600   | °C   |
| Utilization factor of fuel | 80%   |      |
| Utilization factor of oxygen | 30% |      |
| Current density            | 0.3   | A/cm\(^2\) |
| Fuel inlet temperature     | 25    | °C   |
| Fuel inlet pressure        | 1.05  | bar  |
| Air, water inlet temperature | 25   | °C   |
| Air, water inlet pressure  | 1.05  | bar  |

---

![Figure 2. Schematic of the SOFC system with the ethanol reforming process.](https://doi.org/10.1021/acsomega.1c04698)
Here, the $W_{\text{el,net}}$ is the net electrical power, calculated by subtracting the power required by the auxiliaries in the system from the electrical power produced from the SOFC.

\[ n_{\text{ele,sys}} = \frac{W_{\text{el,net}}}{m_{\text{EtOH,in}} \cdot LHV_{\text{EtOH}}} \] (18)

where $W_{\text{th,net}}$ is the net thermal power, calculated by subtracting the heat consumption (steam generation, reforming, etc.) from the heat production of the SOFC.

\[ n_{\text{th,sys}} = \frac{W_{\text{th,net}}}{m_{\text{EtOH,in}} \cdot LHV_{\text{EtOH}}} \] (19)

3.4. Exergy Analysis. The total exergy $E_k$ of a material stream $k$ includes physical ($E_k^{PH}$) and chemical ($E_k^{CH}$) exergies. The physical exergy denotes the maximum theoretical useful work from the stream to the temperature and pressure of the thermodynamic reference environment without changing the chemical composition. The chemical exergy denotes the useful work obtainable by realizing the substances' conversion by mixing and separation and the chemical reaction.\(^{(18)}\)

\[ E_k = E_k^{\text{CH}} + E_k^{\text{PH}} = n (E_k^{\text{CH}} + E_k^{\text{PH}}) \] (20)

\[ E_k^{\text{PH}} = E_k^{\text{TH}} + E_k^{\text{ME}} = E_k (T, P, x \rightarrow T_0, P_0, x_0) = (h_{T,p,x,k} - h_{T_0,p_0,x_0,k}) - T_0 (s_{T,p,x,k} - s_{T_0,p_0,x_0,k}) \] (21)

where $h$ and $s$ represent the enthalpy and entropy at the operating temperature and pressure, and the $h_0$ and $s_0$ represent the enthalpy and entropy under the reference states ($T_0 = 298.15$ K and $p_0 = 101.325$ kPa), respectively. $x$ is the molar fraction and $i$ represents each chemical composition. The standard chemical exergies of pure substances involved in the proposed system are shown in Table 4.(1) Component level Exergy is always destroyed and not conserved when the work is done. Lower exergy destruction can improve the system performance and efficiency.

### Table 3. External Reformer Operating Parameters

| reforming technology          | temperature (°C) | pressure (bar) | steam–ethanol ratio | $O_2$–ethanol ratio | $CO_2$–ethanol ratio |
|-------------------------------|------------------|----------------|---------------------|---------------------|----------------------|
| CO$_2$ dry reforming          | 400–600          | 1.05           |                     | [1,5]               |                      |
| steam reforming               | 400–600          | 1.05           |                     | [1,5]               |                      |
| partial oxidation reforming   | 400–600          | 1.05           |                     | [0.1,2.1]           |                      |
| autothermal reforming         | 400–600          | 1.05           |                     | [0.78,2.78]         | [0.11,1.11]          |

### Table 4. Standard Molar Chemical Exergy at Reference States

| substance | state     | exergy [kJ/mol] |
|-----------|-----------|-----------------|
| C$_2$H$_5$OH | gas       | 1363.9          |
|           | liquid    | 1357.7          |
| CH$_4$    | gas       | 831.65          |
| CO        | gas       | 275.1           |
| CO$_2$    | gas       | 19.87           |
| H$_2$     | gas       | 236.1           |
| H$_2$O    | gas       | 9.5             |
|           | liquid    | 0.9             |
| O$_2$     | gas       | 3.97            |
| N$_2$     | gas       | 0.72            |
reduce operating costs. At the component level, the exergy balance of a component \( j \) is defined as
\[
\dot{E}_{\text{D}j} = \dot{E}_{\text{P}j} - \dot{E}_{\text{F}j}
\]  
(23)
where the \( \dot{E}_{\text{D}j} \) is the exergy destruction, illustrating the inherent process irreversibility. \( \dot{E}_{\text{P}j} \) is the product exergy, representing the useful and desirable effect it delivers, whereas, \( \dot{E}_{\text{F}j} \) is the fuel exergy, representing the amount of exergy consumed by the same component to generate the product. The effect of energy conversion and the effective utilization degree can be determined through the exergy efficiency analysis. The available exergy efficiency \( \varepsilon_{j} \) is defined as
\[
\varepsilon_{j} = \frac{\dot{E}_{\text{P}j}}{\dot{E}_{\text{F}j}}
\]  
(24)

(2) System level

Exergy flows can leave a system as losses without a productive purpose, interacting with the thermodynamic reference environment via the material or heat/power flows. The system’s exergy efficiency \( \varepsilon_{\text{sys}} \) is written as
\[
\varepsilon_{\text{sys}} = \frac{\dot{E}_{\text{P,sys}}}{\dot{E}_{\text{F,sys}}}
\]  
(25)
The exergy destruction ratio \( \gamma_{j} \) is calculated as
\[
\gamma_{j} = \frac{\dot{E}_{\text{D}j}}{\dot{E}_{\text{F,sys}}}
\]  
(26)
The exergy destruction contribution ratio \( \gamma_{c} \) is evaluated as
\[
\gamma_{c} = \frac{\dot{E}_{\text{D}j}}{\dot{E}_{\text{D,sys}}}
\]  
(27)
The exergy product and exergy fuel of the system compositions are summarized in Table 5. Chemical and physical exergies are considered separately for the reformer, after-burner, and SOFC stack, reflecting the operation characteristics of the system.

### 3.5. Carbon Deposition Judgment

Carbon deposition in the SOFC anode can block the electrode pores, inhibiting the species migration to the active site and electrochemical reactions. Carbon deposition depends on the composition, pressure, and temperature of the gas mixture, and can be determined by assuming a thermodynamic equilibrium at the given ratios of C, H, and O. The C–H–O ternary diagrams have been widely used to describe phase diagrams and carbon deposition boundary limits at various temperatures. Therefore, the carbon deposition risk of the SOFC entrance can be calculated with the C–H–O mole fraction of the gas mixture, and compared in the ternary diagram, determining whether the composition is outside the carbon deposition area.

![Figure 4. V–I (a) and P–I (b) curves obtained from the SOFC between simulation results and experimental data.](image-url)

Table 5. Definitions of the Exergy Fuel and the Product of Each Component on a Steady Operation Mode

| component                | exergy fuel | exergy product |
|--------------------------|-------------|----------------|
| pump                     | \( W_{P} \) | \( E_{1} - E_{1} \) |
| blower                   | \( W_{B} \) | \( E_{10} - E_{1} \) |
| anode gas recirculation blower | \( W_{XB} \) | \( E_{3} - E_{2} \) |
| heat exchanger           | \( E_{15} - E_{14} \) | \( E_{4} - E_{3} \) |
| evaporator               | \( E_{16} - E_{15} \) | \( E_{5} - E_{6} \) |
| reformer                 | \( E_{20}^{\text{H}} + E_{21}^{\text{H}} + E_{22}^{\text{H}} \) | \( E_{7} - E_{6} \) |
| afterburner              | \( E_{21}^{\text{H}} + E_{22}^{\text{H}} \) | \( E_{8} - E_{7} \) |
| SOFC stack               | \( E_{23}^{\text{H}} + E_{24}^{\text{H}} + E_{25}^{\text{H}} \) | \( E_{9} - E_{8} \) |

The exergy destruction ratio \( \gamma_{j} \) is calculated as
4. RESULTS AND DISCUSSION

4.1. Model and Validation of the Metal-Supported SOFC. The SOFC electrochemical model and parameters are presented in Section 2 and validated with the experimental data. The standard cell-active surface is 15.2 cm² with 0.45 L/min of air and the fuel inlet flow containing 97% H₂ and 3% H₂O. The voltage–current and power–current curves are tested at 873, 853, and 823 K. The comparison between the prediction model and the experimental data at different current densities and temperature is shown in Figure 4, showing a good agreement with the experimental data.

4.2. Parametric Study of the Metal-Supported SOFC.

4.2.1. Substrate Thickness and Hole Spacing Effect. Metal substrates can transfer energy fast with high thermal conductivity and mechanical stability, improving the rapid thermal cycling performance. The porous metal support structure can provide channels for fuel transmission and increase the current conduction efficiency by the conductivity of metal materials. The variations of substrate thickness with the current density of the metal-supported SOFC are presented in Figure 5a. The operating temperature is 873 K and other conditions remain the same as in Section 4.1. The activation overpotential increases with the increasing substrate thickness due to the larger barrier for hydrogen reaching the anode interface and steam diffusion. Substrate hole spacing is another important parameter for the gas transport characteristics in the porous metal support. The cell voltage shows a similar trend to that of the substrate thickness, as shown in Figure 5b. Hydrogen needs to diffuse through the substrate hole before reaching the reacting surface, and the average diffusion path consists of the substrate thickness and hole pitch as expressed in eq 10. Therefore, smaller hole spacing can decrease the activation overpotentials with a higher voltage, especially for a high current density.

4.2.2. Operating Pressure and Temperature Effects. The operating pressure effect with different current densities is illustrated in Figure 6a. The increase of operating pressure can
promote the equilibrium potential due to the Nernst equation. In addition, the activation overpotential decreases at higher pressures, as expressed in eqs 7–10. Therefore, operation at 1 bar is recommended due to more energy consumption for higher pressures.

A lower operating temperature with high thermal and electrical conductivity is the distinct advantage for the metal-supported SOFC. The cell voltage can reach 0.6–0.7 V at 600 °C for the typical operating current density, as shown in Figure 6b. However, the anode-supported SOFC can only achieve 0.3–0.5 V at 600 °C, and the electrolyte-supported SOFC is even lower than the previous two types. Therefore, the metal-supported SOFC allows the practical operating temperature below to 600 °C.

4.3. Comparison of Different Reforming Options.

4.3.1. Thermodynamic Analysis. The SOFC electrical efficiency increases with the increase of the reforming temperature and decreases with the increase of the steam–ethanol ratio, CO₂–ethanol ratio, and O₂–ethanol ratio, as shown in Figure 7. The ethanol reforming reaction is enhanced due to the endothermic nature of reforming, and more hydrogen is produced largely with the increase of the reforming temperature. Therefore, the voltage increases gradually according to the Nernst equation (eq 1). With the increase of the steam–ethanol ratio, CO₂–ethanol ratio, and O₂–ethanol ratio, the fuel concentration is diluted by the steam- and CO₂-rich inlet flows, leading to a decrease in the cell voltage.

The 36% peak efficiency is always reached for the steam reforming at a SOFC operating temperature of 600 °C. However, the anode-supported SOFC electrical efficiency can only achieve 20% with steam reforming at 600 °C, showing the low-temperature advantage of metal-supported SOFC. The SOFC electrical efficiency can reach the maximum of 50% with autothermal reforming at a SOFC operating temperature of 600 °C, showing higher performance than the 34.4% electrical efficiency with the AER process at an anode-supported SOFC operating temperature of 650 °C. The ethanol-fueled SOFC with a 26% efficiency with the OER process is higher than that of the previous research (20% electrical efficiency) due to a wise heat integration design. The peak efficiency of 28% is always reached with an external reformer temperature of 600 °C for dry reforming.
According to the ethanol reforming theory, the hydrogen production of the SER and AER is higher than that of DER and OER processes, leading to the decrease of the Nernst voltage and electrical efficiency. Specifically, the AER can provide a higher hydrogen yield than other reforming processes with an oxygen–ethanol ratio of 0.11 and a steam–ethanol ratio of 2.78.

4.3.2. Carbon Deposition Analysis. The risk of carbon deposition is mainly concentrated toward a low steam–(oxygen–)ethanol ratio below 600 °C for the SER and OER process, consistent with the thermodynamics of carbon deposition, as shown in Figure 7. The higher steam–(oxygen–)ethanol ratio (at least over 3) will lower the carbon fraction of the mixture entering the anode inlet, thus reducing the risk of carbon deposition during reforming. The simulation result is consistent with the research of Tippawan.19 The DER process can exploit the CO₂ to produce hydrogen without additional water, but more CO₂ is added to the fuel side across the electrolytes, increasing the carbon fraction of the anode side. Therefore, the risk of carbon deposition always exists for the whole operating condition.

The ATR process has the highest SOFC electrical efficiency, but carbon deposition always exists at nominal operating points due to the lower steam–(oxygen–)ethanol ratio. Therefore, the SOFC-APU system can compromise electrical efficiency by adding more water into the fuel and decreasing the risk of carbon deposition.

4.4. System Performance Analysis. 4.4.1. Startup Stage. During the startup stage, the SOFC-APU system components are heated up until the reformer and SOFC reaches the minimum operating temperature. The startup burner provides the heat for the reformer and SOFC stack, and the minimum ethanol flow for combustion is determined by the heat balance between the external and internal reforming, combustion, and electrochemical reaction undergoing several iterations. The total ethanol flow comprises the consumption of startup combustion and the SOFC system operation. The additional water supply is identified by the water input for reforming and the recirculation flow.

The ethanol flow decreases first and then increases with the increase of the recirculation ratio, as shown in Figure 8b. More recirculated flow comprising unreacted methane and carbon monoxide enters into the reformer, absorbing more heat due to the endothermic reaction. Therefore, more ethanol for the combustion reaction is needed to release heat. At the same time, the system’s electrical efficiency shows the opposite trend, and the peak efficiency of 44.4% is always reached with a recirculation ratio equal to 30%, which is consistent with the conclusion of a minimum ethanol flow and can be explained by eq 18.

The higher the recirculation ratio, the less additional water will be needed, up to 0.34 g/s. The recirculation flow containing unreacted water reduces the intake of extra water based on the water management strategy. For a very high recirculation ratio of 60%, carbon deposition can be avoided without additional water fed due to the increased carbon tolerance.

4.4.2. Steady Operating Stage. 4.4.2.1. Energy Analysis. For the same external reforming temperature of 550 °C, the system’s electrical efficiency increases first and then decreases with the increase of recirculation ratio. The peak efficiency above 55.4% is reached with a 70% recirculation ratio, as shown in Figure 9a. For the previous ethanol-fueled SOFC system design, the electrical efficiency of 46.16% could only be achieved without exploring the recirculation ratio effect.24 The increase in the recirculation ratio brings more unreacted fuel molecules back to the anode, which brings the following two effects: (1) at the same current density, the ethanol feed and additional water for reforming will be reduced, as shown in Figure 9b, and (2) the fuel concentration is diluted by the steam- and CO₂-rich recirculated flow, which will lead to a decrease in the cell voltage, as shown in Figure 9c,d. The profile of the system efficiency versus the increase of the recirculation ratio is due to the trade-off between the reduction of the fuel and the decrease in the cell voltage. With the increase of the recirculation ratio, the increase of the system efficiency is first dominated by effect (1). Once additional water is not needed for reforming, the effect of the recirculation ratio increase will be dominated by effect (2).

From the perspective of fuel economy, ethanol flow is an important parameter. Ethanol decreases with the increase of the recirculation ratio, reaching a minimum flow of 0.318 g/s. The anode recirculation technology shows the advantages of fuel
saving. The higher the recirculation ratio, the less additional water will be needed, as shown in Figure 9b. The carbon deposition risk can be eliminated over a 60% recirculation ratio without adding external water.

The system’s thermal efficiency also increases first and then decreases with the increase of the recirculation ratio, reaching 37.3% at a 50% recirculation ratio. A larger recirculation flow requires more heat for steam generation and gas reforming and causes higher heat to be released by the SOFC. Therefore, there is a trade-off between heat absorption and release.

The heat duty of the SOFC-APU system components at a 50% recirculation ratio is shown in Figure 10b. The cathode preheater is the main heat absorption component, and the exothermic reaction of the SOFC can supply the heat for the APU system. The available heat of the system is 3.5 kW, demonstrating that the SOFC-APU system is thermally self-sustainable in the steady-state operation.

Figure 9. System’s electrical efficiency (a), the feed mass flow (b), the SOFC inlet fuel mole fraction (c), and the SOFC voltage (d) with different recirculation ratios.

The local fuel utilization factor is the ratio of the reacted fuel flow and the anode inlet fuel flow. For a given optimal 70% recirculation ratio, the system’s electrical efficiency increases from 50 to 57% with the increase of the fuel utilization factor due to more hydrogen reaction and the reduction of the ethanol feed, as shown in Figure 11b. For the anode-supported ethanol-fueled SOFC system, the peak electrical efficiency can reach 45% with different fuel utilization factors at a 70% recirculation ratio. Therefore, the metal-supported SOFC system is more suitable for automobile applications with higher efficiency.

4.4.2.2. Exergy Analysis. The system’s exergy efficiency increases first and then decreases with the increase of the recirculation ratio, as shown in Figure 12a. The peak efficiency of up to 77.1% can be reached at a 70% recirculation ratio, which is consistent with the energy analysis. At the same time, the exergy destruction ratio reaches the lowest 22%, indicating the best energy utilization at a 70% recirculation ratio.
For the system components, the SOFC has the highest contribution rate of exergy destruction, followed by the afterburner and reformer, indicating more relevance in chemical exergy changes than physical exergy. The result is consistent with the research of Pegah and Casas. The exergy destruction contribution of the after-burner decreases with a higher recirculation ratio due to less exhaust for combustion and heat release, as shown in Figure 13.

### 5. CONCLUSIONS

Four ethanol reforming processes utilized on the metal-supported SOFC are compared with thermodynamic analysis. The autothermal ethanol reforming shows the highest electrical efficiency of 50%. The optimal conceptual designs are evaluated by the energy analysis from the startup and stable stages. The exergy of the fuel and product of all components have been defined and the exergetic efficiency has been analyzed. From the perspective of carbon deposition safety and fuel economy, the SOFC-APU system can operate with a minimum of 0.42 g/s ethanol flow satisfying the system startup phase. The carbon deposition risk can be eliminated with an over 60% recirculation ratio. The electrical efficiency and exergy efficiency of the SOFC-APU system can reach 55.4 and 77.1% with a 70% anode gas recirculation ratio during the stable running phase. This work could be the basis for SOFC systems suitable for automotive integration and provide valuable insights into the SOFC application in mobility. Compared with the pure electric and H₂ fuel cell power system, the onboard SOFC power system combines the attractiveness of electric vehicles with the convenience of the liquid fuel, achieving fast refueling, a higher

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**Figure 10.** System’s thermal efficiency with different recirculation ratios (a) and components’ heat duty (b).

**Figure 11.** System’s electrical efficiency (a) and ethanol flow (b) with different fuel utilization factors.
efficiency, and a longer driving range. The high efficiency of
electrochemical energy conversion directly corresponds to lower
CO₂ emissions and achieves almost particle-free exhaust gas.
The environmental impact could be lower compared with diesel
generator APUs. Therefore, the SOFC-APU system provides a
complementary solution to the advantages of new energy
vehicles at this stage. From the design to commercialization,
there are still some challenges. Lower technology maturity
including the specific energy density, thermal transient response,
and lifetime need to be addressed in future efforts. Moreover, the
SOFC production cost is the main cost driver, becoming
another great challenge. The maturity and scalability of SOFC
are accelerating fast, and these challenges can be solved with
technology development.

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Notes
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■ NOMENCLATURE

Abbreviations
SOFC solid oxide fuel cell
APU auxiliary power unit
UFF factor of fuel utilization
SER steam-ethanol reforming

Figure 12. System’s exergy efficiency (a) and destruction ratio (b) on different recirculation ratios.

Figure 13. Contribution (%) of each component to exergy destruction on different recirculation ratios.
MATHEMATICAL SYMBOLS

- $U_{Net}$: electromotive force
- $P$: pressure
- $T$: temperature
- $\Delta g$: Gibbs free energy change
- $R_g$: gas constant
- $F$: Faraday constant
- $R_{ohmic}$: ohmic overpotentials
- $i$: current density
- $p_{H2}$: partial pressure of hydrogen
- $F_{H2}$: inlet flow of SOFC
- $A_{diss}$: effective surface area
- $N_{SOFC}$: number of SOFC cells
- $\eta_{el}$: electrical efficiency
- $\eta_{sys}$: system’s efficiency
- $\eta_{ther}$: system’s thermal efficiency
- $E_i$: total exergy
- $E_{phys}$: physical exergy
- $E_{chem}$: chemical exergy
- $x_{mol}$: molar fraction
- $\xi$: enthalpy
- $\xi_{D,j}$: exergy destruction
- $\xi_{P,j}$: product exergy
- $\xi_{F,j}$: fuel exergy
- $\xi_{sys}$: system exergy efficiency
- $\xi_{sys}$: system exergy destruction ratio
- $\xi_{sys}$: system exergy destruction contribution ratio

SUPERSCRIPTS

- $i$, $j$, $k$: index

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