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Exergetic performance analysis of a direct ammonia-fed solid oxide fuel cell

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Abstract

To explore the performance limit of direct ammonia-fed solid oxide fuel cells based on oxygen-ion conductivity, a finite time thermodynamic model is developed. The finite time thermodynamic indexes including exergy efficiency, exergetic performance coefficient and entropy production rate are derived to evaluate the performance of direct ammonia-fed solid oxide fuel cells from multiple perspectives. Moreover, the effects of operating temperature, operating pressure, fuel utilization, electrolyte thickness and electrode porosity on exergy efficiency and exergetic performance coefficient of the studied direct ammonia-fed solid oxide fuel cells are numerically analyzed. The derived finite time thermodynamic model can be further employed to obtain optimal operating parameters and structural parameters under different application scenarios to guide engineering design and operation control.

Keywords: DA-SOFC; Finite time thermodynamics; Exergetic performance coefficient; Exergy efficiency

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1. Introduction

The overuse of fossil energy has brought serious energy crisis and environmental pollution to the world. Fuel cells are very attractive options because of their high efficiency, clean operation and fuel flexibility [1]. Since ammonia is safer, less expensive, easier to store and transport, and non-flammable at normal conditions, it has become a viable fuel for fuel cells [2]. Moreover, the technological foundation for the production of ammonia has been already established. Consequently, there is a growing interest in the use of ammonia in fuel cells, especially solid oxide fuel cells (SOFCs) [3–5].

Traditional methods of studying fuel cells are usually based on idealized assumptions. Finite time thermodynamics (FTT)

takes into account the irreversibility and actual operating conditions of the actual power and energy systems, and provides more realistic models for performance optimization [6]. One of the main purposes of FTT is to provide valuable means to optimize thermal and/or fluidic systems by seeking systems performance boundaries considering under finite time and finite size constraints [7].

The primary areas of study in finite-time thermodynamics include finite-time availability, optimum routes of thermodynamic processes, and novel assessment indices [8,9]. These include conventional thermodynamic devices such as power plants [10], heat pumps [11], fuel cells [12,13], and so on. Ge et al. [14] used finite time thermodynamic theory to optimize

Nomenclature

A_{cell}	– active cell area, m^2
ex	– exergy, J/mol
E_r	– cell reversible voltage, V
E^0	– voltage at standard pressure, V
EPC	– exergetic performance coefficient
Ex	– exergy, W
F	– Faraday's constant, C/mol
h	– molar enthalpy, J/mol
j	– current density, A/ m^2
\dot{m}	– mass flow rate, kg/h
$n_{H_2, equ}$	– equivalent molar flow rate of H_2 , mol/s
$n_{H_2, consumed}$	– actual consumed molar flow rate of H_2 , mol/s
n_{fuel}	– amount of fuel required, mol
p	– pressure, atm
p_i	– partial pressure of i -th component, atm
p_i^{TB}	– partial pressure of i -th component at three-phase boundary, atm
P	– power density, A/ m^2
\bar{R}	– gas constant, J/(mol · K)
R_{Ω}	– electrolyte resistivity, Ω m
s	– molar entropy, J/(mol K)
t_{ele}	– electrolyte thickness, mm
T	– operating temperature, K
T_e	– ambient temperature, K
T_L	– temperature of the low-temperature heat source, K
U_f	– fuel utilization, %
$V_{act, an}$	– anodic activation overpotential, V
$V_{act, ca}$	– cathodic activation overpotential, V
$V_{con, an}$	– anodic concentration overpotential, V

$V_{con, ca}$	– cathodic concentration overpotential, V
V_{ohm}	– Ohmic overpotential, V
W	– output power, W
x_i	– mole fraction

Greek symbols

ϕ	– exergy efficiency
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Subscripts and superscripts

0	– standard conditions
act	– activation
an	– anode
ca	– cathode
con	– concentration
fc	– fuel cell
H_2	– hydrogen
H_2O	– water
O_2	– oxygen
ohm	– Ohmic

Abbreviations and Acronyms

DA-SOFC	– direct ammonia-fed solid oxide fuel cells
EDI	– exergy destruction index
ESI	– exergy sustainability index
FTT	– finite time thermodynamic (model)
GA	– genetic algorithm
HT-PEMFC	– high temperature proton exchange membrane fuel cell
MSR	– methanol steam reforming
ORC	– organic Rankine cycle
PEMFC	– proton exchange membrane fuel cell

the performance by setting work, power and efficiency as optimization targets and configuration of an internal combustion engine. Chen et al. [15] applied the finite time thermodynamic theory to build a novel solar driven system and analysed the effects of solar irradiance on the thermodynamic performance of the system. Li et al. [13] developed a finite-time irreversible thermodynamic model of proton exchange membrane fuel cell (PEMFC) and produced the ecological performance coefficient and the finite-time ecological objective function. Guo et al. [16] derived finite-time thermodynamic indexes such as exergy efficiency, exergy destruction index (EDI) and exergy sustainability index (ESI) to better evaluate the performance of the fuel cell. Li et al. [17] applied finite time thermodynamic theory to analyse and optimize the power system of a vehicle mounted high temperature proton exchange membrane fuel cell (HT-PEMFC) integrating methanol steam reforming (MSR) and organic Rankine cycle (ORC). The results showed that higher operating temperature and anode pressures are beneficial to improve overall performance and that systems operating at optimum parameters have better thermodynamic, economic and environmental performance.

To improve the performance of fuel cells in economic and ecological terms, many optimization studies have been done for different operating parameters [18]. Hajabdollahi and Fu [19] set up a SOFC cogeneration system optimized with the objective function of maximum exergy efficiency and minimum total cost

rate, and the optimal exergy economic configuration of the system was obtained. Mert et al. [20] carried out multi-objective optimization work on a vehicle-mounted proton exchange membrane fuel cell system using a genetic algorithm (GA) and optimized the system with the goals of maximizing power output, energy efficiency and minimizing power generation costs. You et al. [21] used the NSGA-II method to perform two sets of multi-objective optimizations on the SOFC system with five key objectives: energy efficiency (maximization), system energy efficiency (maximization), system net output power (maximization), freshwater production (maximization), and system total cost (minimization), achieving optimal system performance in three typical scenarios. Forough and Roshandel [22] used genetic algorithms to perform multi-objective optimization of SOFC stacks with minimum energy cost, maximum output power, minimum energy cost and maximum power efficiency as objective functions. Mojaver et al. [23] used a genetic algorithm for the multi-objective optimization of the SOFC power generation system. The current density and the inlet temperature of the stack were used as input variables when the sum of energy efficiency and product unit cost were taken as the objective function, comparing the performance of SOFC-O based and SOFC-H based power generation systems under optimal operating conditions.

In this paper, we first analyse the irreversibility of DA-SOFC from the point of view of finite time thermodynamics, and establish a mathematical model of exergetic efficiency ϕ and

EPC considering irreversible polarizations. Secondly, the performance of DA-SOFC is analysed, and the effects of operating temperature, operating pressure, fuel utilisation, electrolyte thickness and electrode porosity on the exergetic efficiency and *EPC* of DA-SOFC are studied. The results obtained can provide theoretical guidance for the optimization design and practical application of DA-SOFCs.

2. Exergetic performance analysis model of DA-SOFC

Conventional SOFCs typically use oxygen ions as the conducting electrolyte (SOFC-O), such as the commonly used yttria stabilised zirconia (YSZ). In addition, SOFC can also use protons as conductive electrolytes (SOFC-H). In solid oxide fuel cells based on oxygen ion conductivity, there are still problems to be solved such as low cell efficiency due to higher anode concentration overpotential [24]. This paper will focus on the redox reaction in solid oxide fuel cells based on oxygen ion conduction, and study the problems such as reducing the concentration overpotential at the anode, so as to improve the oxygen ion conduction and cell efficiency.

O^{2-} ions must flow through the electrolyte in solid oxide fuel cells based on oxygen ion conducting electrolytes. Air or pure oxygen gas should be supplied to the fuel cell's cathode side. Oxygen is converted to oxygen ions at the cathode-electrolyte junction. At the interface between the anode and electrolyte, these ions go through the electrolyte and react electrochemically with hydrogen gas to form water vapour. Ammonia fuel thermally breaks down to make hydrogen. At the anode side of the fuel cell, the ammonia fuel is fed in and thermally decomposes with the help of a catalyst to produce H_2 and N_2 .

Decomposition-produced hydrogen diffuses to the anode-electrolyte interface, where it electrochemically interacts with oxygen ions to make water vapour. The fuel cell releases hydrogen, nitrogen and unreacted ammonia fuel, and produces water vapour. The hydrogen concentration is diluted as a result of creation of nitrogen during the thermal breakdown of ammonia, which lowers the fuel cell's reversible potential. At low temperatures and without a catalyst, the decomposition rate is slower, but at high temperatures and with a catalyst, the decomposition rate is faster.

Based on series process assumptions [15,25,26]:

- ammonia fully participates in gasification reactions,
- by volume, air is composed of 79% non- O_2 fraction and 21% O_2 ,
- every gas is deemed ideal and gas leakage is negligible,
- all reaction processes are steady-state within the stack,
- the temperature distribution of the stack is uniform,
- using environmental conditions (temperature and pressure of 25°C and 1 atm \approx 1.013 bar, respectively) as reference conditions, exergy analysis is conducted under design conditions,

the mathematical expressions for the output power, output efficiency, ecological objective function and ecological coefficient of performance of the direct ammonia-fed solid oxide fuel cell were derived in [27].

The single DA-SOFC voltage can be obtained [27]:

$$V = E_r - V_{act,an} - V_{act,ca} - V_{ohm} - V_{con,an} - V_{con,ca} \quad (1)$$

The output power density of DA-SOFC is [27]:

$$\begin{aligned} P = \frac{P_{cell}}{A_{cell}} &= jV_{cell} = j(E_r - V_{act,an} - V_{act,ca} + \\ &+ V_{con,an} - V_{con,ca}) = j(E^0 + \frac{\bar{R}T}{2F} \ln(\frac{p_{H_2}(p_{O_2})^{\frac{1}{2}}}{p_{H_2O}}) + \\ &- \frac{\bar{R}T}{F} \sinh^{-1}(\frac{j}{zJ_{ca}}) - \frac{\bar{R}T}{F} \sinh^{-1}(\frac{j}{zJ_{an}}) - jt_{ele}R_{\Omega} + \\ &- \frac{\bar{R}T}{2F} \ln(\frac{p_{H_2}^{TB} p_{H_2O}}{p_{H_2} p_{H_2O}^{TB}}) - \frac{\bar{R}T}{4F} \ln(\frac{p_{O_2}}{p_{O_2}^{TB}})). \end{aligned} \quad (2)$$

For a more detailed derivation and working scheme chart the reader can be referred to reference [27].

Energy loss and deterioration in output performance can result from heat loss and friction between the gas and the channel during DA-SOFC operation. The second law of thermodynamics requires that exergy analysis be carried out in order to thoroughly assess the thermodynamic performance of DA-SOFC. The potential and kinetic energies of the steady state system are negligible. The logistical exergy mainly includes physical exergy and chemical exergy [28], which are calculated by the formula:

$$Ex = \dot{m} \cdot ex = \dot{m}(ex^{ch} + ex^{ph}), \quad (3)$$

$$ex^{ch} = \sum x_i ex_{0,i}^{ch} + RT_0 \sum x_i \ln x_i, \quad (4)$$

$$ex^{ph} = (h - h_0) - T_0(s - s_0), \quad (5)$$

where x_i and $ex_{0,i}^{ch}$ are the mole fraction of the substance and the standardized chemical exergy, respectively; T_0 is the ambient temperature; h_0 and s_0 are the specific enthalpy and entropy of the reference state, respectively; $h-h_0$ represents the change in molar enthalpy and $s-s_0$ the change in molar entropy.

In the direct ammonia fuel cell system, the total energy input is:

$$Ex_{in}^{fc} = Ex_{NH_3,in} + Ex_{O_2,in}. \quad (6)$$

The efficient use of energy is reflected in the exergy efficiency of DA-SOFC, which is expressed as follows:

$$\phi = \frac{P}{Ex_{in}^{fc}} = \frac{j(E_r - V_{act,an} - V_{act,ca} - V_{ohm} - V_{con,an} - V_{con,ca})A}{Ex_{H_2,in} + Ex_{O_2,in}}. \quad (7)$$

The exergy balance equation of the steady-state system is expressed as:

$$Ex_D = \sum Q(1 - \frac{T_0}{T}) - W + \sum Ex_{in}^{fc} - \sum Ex_{out}^{fc}, \quad (8)$$

where Ex_D is the exergy loss rate.

The DA-SOFC thermodynamic performance is analysed using the exergetic performance coefficient (*EPC*). It is defined as the output power to exergy destruction rate ratio, combining thermodynamic and exergy performance. *EPC* may be described as follows:

$$EPC = \frac{P}{Ex_D} = \frac{j(E_r - V_{act,an} - V_{act,ca} - V_{ohm} - V_{con,an} - V_{con,ca})A}{\sum Q(1 - \frac{T_0}{T}) - P + \sum Ex_{in}^{fc} - \sum Ex_{out}^{fc}}. \quad (9)$$

3. Results and discussion

To numerically analyse and compare studied DA-SOFC performance based derived models, the same data sets of a DA-SOFC single cell as in [27] are employed.

3.1. Effects of operating temperature on DA-SOFC performance

The operating temperature of DA-SOFC is crucial for its performance. In a suitable high-temperature environment, the electrochemical reaction rate is accelerated and the electrode activity is improved, which helps to reduce the polarization loss inside the fuel cell stack and thereby increase the efficiency and power density of the fuel cell. Increasing the temperature is beneficial for the rapid conduction of electrons and oxygen ions and enhances the electrical conductivity of DA-SOFC. However, at excessively high operating temperatures, fuel cell components may be affected by corrosion and its life can be reduced. As well, inconsistent thermal expansion coefficients of components may lead to cracks and failures inside the components. In addition, high-temperature operation usually requires expensive special materials such as high-temperature alloys and ceramics, increasing manufacturing costs and also requiring the introduction of more complex thermal management systems. Therefore, accurately selecting and controlling the operating temperature is vital for ensuring the efficient and stable operation of DA-SOFC.

Figures 1 and 2 present the exergy efficiency ϕ and EPC varying with current density at different operating temperatures, respectively. One can see that the operating temperature of DA-SOFC has a significant impact on exergy evaluation indicators. The exergy efficiency ϕ and exergy performance coefficient EPC of DA-SOFC both increase with the increase of operating temperature. In the high current density range, as the operating temperature increases, the electrochemical reaction rate significantly increases, thereby improving the fuel utilization efficiency, reducing fuel waste, and effectively reducing the exergy loss of the system. At the same time, increasing the temperature also leads to an increase in ionic conductivity, significantly reducing the ionic conduction resistance and slowing down the waste heat generation caused by ionic transmission resistance. Therefore, during the operation of DA-SOFC, the unrecoverable exergy loss is significantly reduced. As shown in Fig. 2, in the low current density range, the improvement degree of EPC by increasing temperature is much smaller than that in the high current density range. This is mainly because under low current density conditions, the electrochemical reaction rate is relatively slow, so the influence of operating temperature on the activity of electrons and ions is relatively limited. However, when the current of DA-SOFC is 15 000 A/m² and the operating temperature is increased from 1043 K to 1103 K, EPC increases by 61%. Therefore, increasing the operating temperature can significantly improve the exergy performance of DA-SOFC.

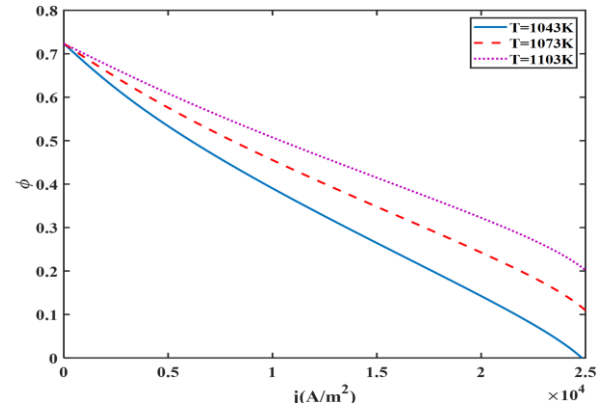


Fig. 1. ϕ varying with current density at different T .

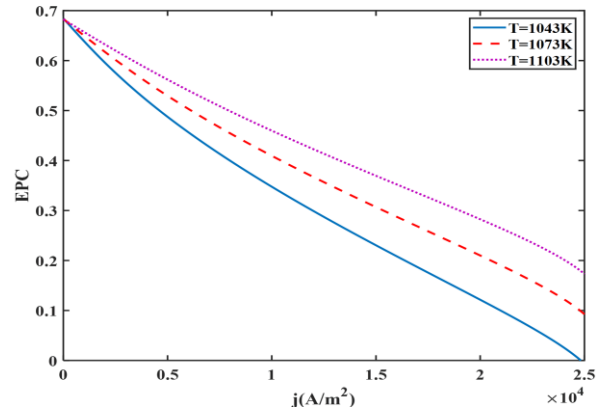


Fig. 2. EPC varying with current density at different T .

3.2. Effects of operating pressure on DA-SOFC performance

As is known to all, increasing the operating pressure can significantly improve the thermodynamic performance of DA-SOFC. First of all, increasing the operating pressure helps to increase the gas diffusion rate and makes it easier for reactants to be transported inside the fuel cell flow field, so that fuel and oxygen can be used more effectively, thereby improving the overall efficiency. Secondly, a higher operating pressure is conducive to reducing the flow resistance and improving the response speed of the fuel cell. However, increasing the operating pressure will consume more power from auxiliary equipment such as compressors, requiring higher costs, and the manufacturing and maintenance costs of the system may also increase. Therefore, when considering increasing the operating pressure, multiple factors such as performance improvement, system cost, and material strength must be comprehensively considered to find the best operating parameters to meet the needs of specific application scenarios. In general, optimizing the operating pressure is one of the important strategies to improve the performance of fuel cell systems, but various factors need a careful trade-off.

Figures 3 and 4 reflect the changes of exergy efficiency ϕ and exergy performance coefficient EPC with the current density under different operating pressures p . As can be seen from

the figures, the exergy efficiency ϕ and exergy performance coefficient EPC of DA-SOFC both increase with the increase of operating temperature. When the current density is $15\,000\text{ A/m}^2$ and the operating pressure changes from 1 atm to 2 atm, the exergy efficiency ϕ and exergy performance coefficient EPC increase by 11.1% and 22.5%, respectively. Therefore, when the current density and operating temperature are constant, increasing the operating pressure can significantly improve the exergy performance indicators of DA-SOFC.

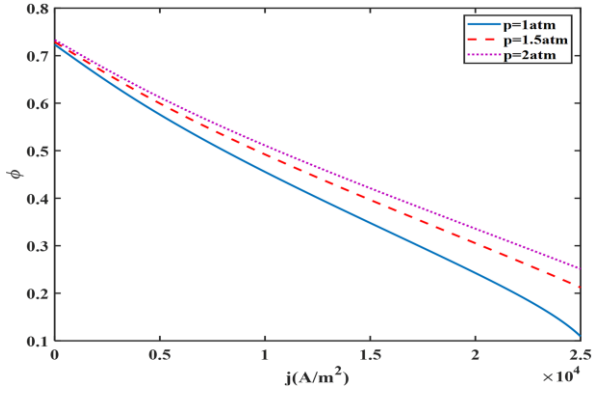


Fig. 3. ϕ varying with current density at different p .

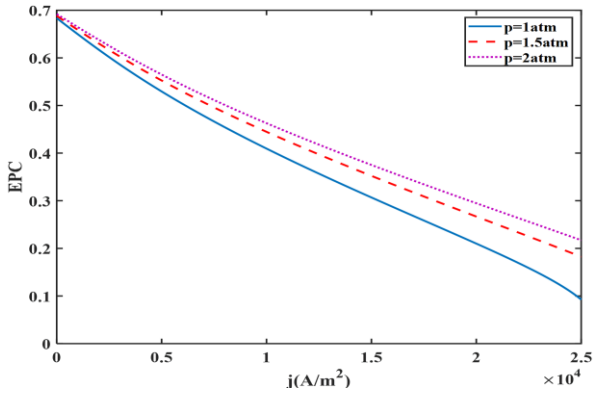


Fig. 4. EPC varying with current density at different p .

3.3. Effects of fuel utilization on DA-SOFC performance

Fuel utilization rate U_f has an important impact on the performance of fuel cells. A higher fuel utilization rate U_f means more efficient conversion of fuel into electrical energy, thereby improving the overall efficiency of the fuel cell. By maximizing the available portion of fuel chemical energy, the economy and sustainability of the fuel cell system can be significantly improved. This has positive significance for realizing clean energy conversion and mitigating climate change in the field of sustainable energy. However, multiple aspects such as fuel supply, reaction kinetics, and system design need to be comprehensively considered to achieve optimal fuel utilization rate U_f . Therefore, in the development and optimization of fuel cell technology and applications, maximizing the fuel utilization rate is one of the key considerations for achieving efficient, economical and sustainable energy conversion.

Figures 5 and 6 present the influence of fuel utilization rate U_f on the exergy performance indicators of DA-SOFC under different operating pressures when the inlet flow rate and the operating temperature are constant. The exergy efficiency ϕ and exergy performance coefficient EPC of DA-SOFC both increase with the increase of fuel utilization rate U_f . Increasing the fuel utilization rate U_f can reduce the consumption of underutilized fuel in the system through non-electrochemical pathways, thereby reducing unnecessary heat generation. A higher fuel utilization rate U_f also means a more effective power generation process, reducing the energy loss caused by electrochemical reactions inside the system and helping to slow down the temperature rise of the system and further reducing the release of waste heat. When the operating temperature is 1073 K , the current density is $15\,000\text{ A/m}^2$ and the fuel utilization rate is 0.9, the exergetic performance coefficient is increased by 22.2%. Therefore, improving fuel utilization can reduce cell heat loss and improve the exergetic performance coefficient. In summary, under certain conditions, increasing fuel utilization can improve the finite time thermodynamic performance of DA-SOFC.

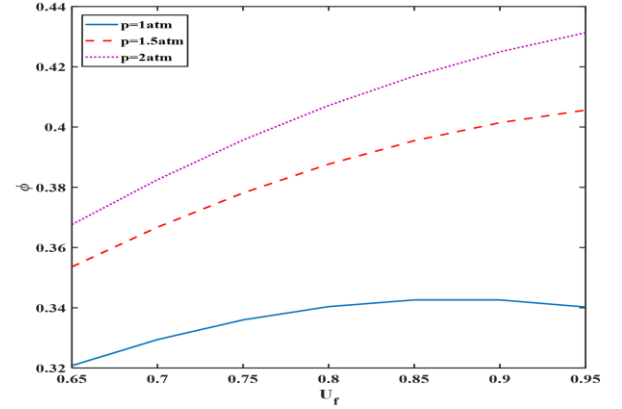


Fig. 5. ϕ varying with fuel utilization at different p .

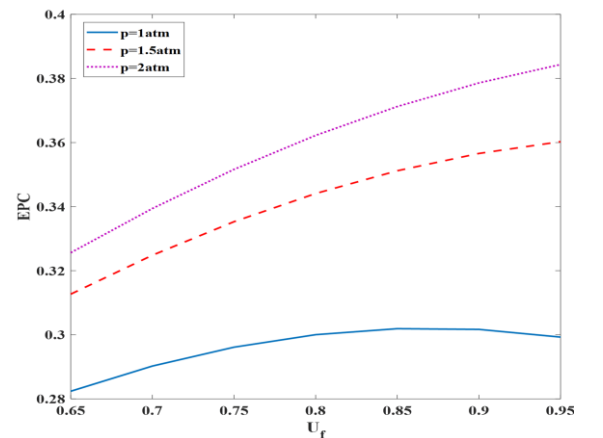


Fig. 6. EPC varying with fuel utilization at different p .

3.4. Effects of electrolyte thickness on DA-SOFC performance

Electrolyte thickness is a key design parameter for fuel cells and has an important impact on system performance. When the elec-

trolyte thickness is too large, the internal resistance of the fuel cell also increases due to the increase in the resistance of the ion transport path. This will increase the resistance of current transmission in the electrolyte and reduce the overall conductivity of the fuel cell, thus affecting the output power. A thicker electrolyte may also lead to an increase in the voltage drop between electrodes, thus affecting the fuel cell system efficiency. Secondly, a thicker electrolyte increases the gas diffusion path and hinders the effective transportation of fuel and oxygen. This affects the utilization efficiency of reactants and reduces the rate of electrochemical reactions. Therefore, the response speed of the fuel cell is slower, affecting its dynamic performance, which may be limited, especially in applications that require a quick response. Therefore, when designing a fuel cell, it is necessary to carefully balance the influence of electrolyte thickness on resistance and reaction rate.

Figures 7 and 8 demonstrate the influence of electrolyte thickness t_{ele} on the exergy performance indicators of DA-SOFC when the operating temperature is constant.

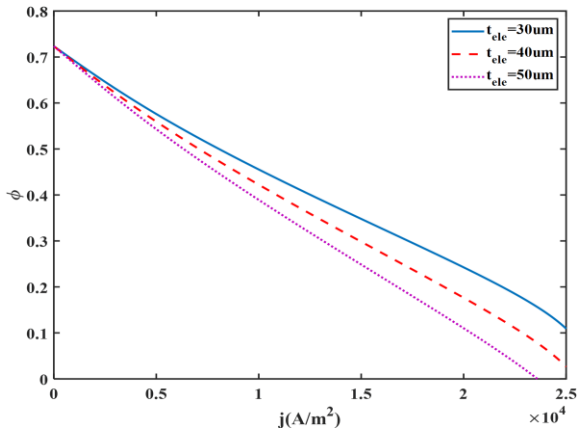


Fig. 7. ϕ varying with current density at different t_{ele} .

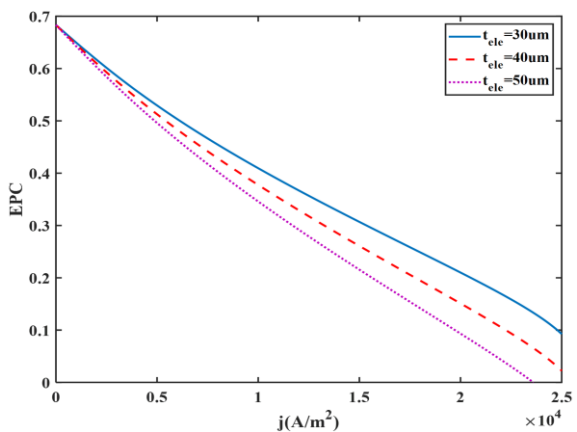


Fig. 8. EPC varying with current density at different t_{ele} .

The exergy efficiency ϕ and exergy performance coefficient EPC of DA-SOFC both decrease with the increase of electrolyte thickness. Increasing the electrolyte thickness of DA-SOFC has a significant impact on waste heat generation. There are multiple reasons for the increase in waste heat generation in DA-SOFC

due to an increase in electrolyte thickness. First, increasing the electrolyte thickness will significantly increase the internal resistance of the fuel cell, leading to greater Ohmic losses. Secondly, due to the increase in electrolyte thickness, the diffusion path of fuel and oxygen in the fuel cell becomes longer, increasing the resistance to mass transfer. This leads to more diffusion limitations, reducing the utilization efficiency of fuel, thereby increasing the proportion of chemical energy that is not converted into electrical energy and is converted into thermal energy. Therefore, although an increase in electrolyte thickness is beneficial to some aspects of performance, it simultaneously brings greater resistance and diffusion limitations, resulting in additional waste heat generation. All these reasons will lead to an increase in exergy loss, resulting in a decrease in exergy efficiency ϕ and exergy performance coefficient EPC .

3.5. Effects of porosity on DA-SOFC performance

The porosity of fuel cell electrodes greatly affects DA-SOFC performance. Moderate porosity can improve gas diffusion and electron conduction, and promote the effective transmission of reactants in the electrode. Higher porosity helps to increase gas passage in the electrode, provides more active surfaces and promotes catalytic reactions. However, too high porosity may also lead to a loose electrode structure and reduce the effective electrode surface area, thereby affecting the reaction rate. Too high or too low porosity may lead to the concentration of mechanical stress and affect the stability of the electrode structure, thus affecting DA-SOFC life. An increase in porosity may lead to an extension of the gas flow path and an increase in gas diffusion resistance. Therefore, when optimizing the porosity value of fuel cell electrodes, factors such as gas diffusion, electron conduction and reaction rate need to be comprehensively considered to achieve the appropriate performance.

Figures 9 and 10 present the influence of electrode porosity ε on the exergy performance indicators of DA-SOFC when the operating temperature is constant. The exergy efficiency ϕ and exergy performance coefficient EPC of DA-SOFC both decrease with the increase of electrode porosity ε . A moderate pore structure can improve the uniform distribution of fuel in the electrode, improve the utilization efficiency of fuel, help to maximize the conversion of chemical energy into electrical energy, and reduce the proportion of unused energy converted into thermal energy, thereby improving the thermal efficiency of the fuel cell. When increasing the porosity, EPC increases. When the current density is 15 000 A/m² and the porosity increases from 0.6 to 0.8, EPC increases by 6.8%. From this analysis, it can be seen that the exergy performance and ecological performance of the fuel cell can be improved by adjusting the microstructure of the electrode, thereby improving the finite time thermodynamic performance of DA-SOFC.

4. Conclusions

A novel finite time thermodynamic model formulating the exergy efficiency and EPC is derived for a DA-SOFC. The effects of parameters including the operating temperature, operating pressure, fuel utilization, electrolyte thickness and electrode porosity on the finite time thermodynamic performance of the

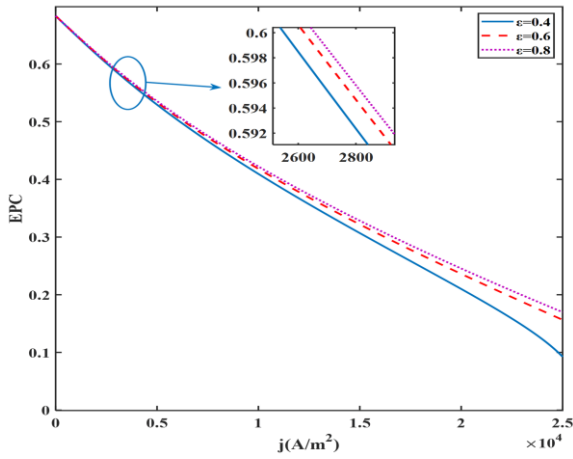


Fig. 9. ϕ varying with current density at different porosity ε .

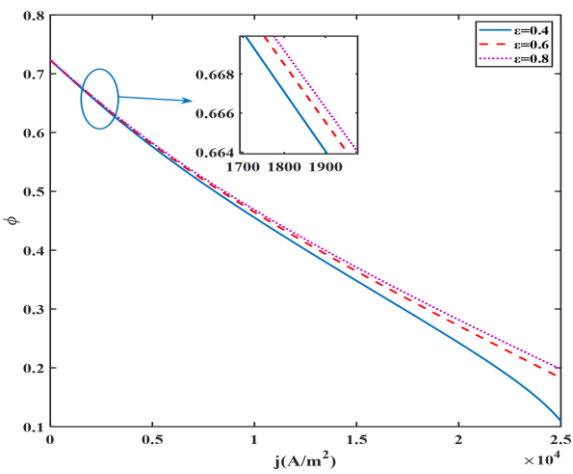


Fig. 10. EPC varying with current density at different porosity ε .

studied irreversible DA-SOFC are investigated. The main results of this work could be concluded as follows:

- 1) Increasing the operating temperature T , inlet pressure p , and fuel utilization rate U_f is beneficial for reducing waste heat generation of DA-SOFC, decreasing exergy loss and entropy production rate, and thereby improving performance indicators such as exergy efficiency and exergy performance coefficient of DA-SOFC. The increase in inlet pressure will consume additional external power, leading to a more complex system, increased cost and weight.
- 2) Reducing the electrolyte thickness (t_{ele}) can improve the thermodynamic performance of DA-SOFC. However, in practical work, a thinner electrolyte will make it difficult to effectively isolate the heat between the electrodes, which may lead to uneven temperature distribution and even thermal runaway, affecting the stack stability and lifespan. In the real design of DA-SOFC, a reasonable electrolyte thickness and electrode porosity need to be selected for DA-SOFC according to specific application scenarios.
- 3) The derived finite time thermodynamic model can be further employed to obtain optimal operating parameters and structural parameters under different application scenarios. These

optimization results can be used to guide engineering design and operation control.

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