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Comparative study of solid oxide fuel cell combined heat and power system with Multi-Stage Exhaust Chemical Energy Recycling: Modeling, experiment and optimization

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In this paper, a novel Multi-Stage Exhaust Energy Recycling strategy was proposed and optimized to maximize the system efficiency and performance of solid oxide fuel cell-combined heating and power (SOFC-CHP). Both process modeling and experiment work based on 1 kW SOFC-CHP systems were carried out to prove the concept and optimize the system. It is found that the system with multi-stage exhaust gas combustion (MS-EGC) will reduce the system operating temperature from 1149 °C to 830 °C, which significantly increases the safety of system operation and reduces the material requirement. The system combining MS-EGC with anode off-gas recovery (MS-AOGR & EGC) leads to highest overall co-generation efficiency up to 92%. A coupled reactor integrating MS-EGC modules was developed and tested for a 1 kW SOFC system to realize the proposed strategy. The results showed that in MS-EGC, recycling of thermal energy at first stage rarely affects the chemical energy utilization in subsequent stages and the overall system performance, further confirming the advantages of the innovative multi-stage energy recycling strategy.

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1. Introduction

Fuel cells offer an efficient and clean way to produce energy due primarily to the direct conversion of chemical energy of the fuel into electricity and thermal energy comparing to conventional power generation methods. Among various distributed power generation technologies, solid oxide fuel cells (SOFC) are widely regarded to be the most promising candidate for small and medium power applications (up to 1 MW) as they feature extremely high efficiency, low pollutant emissions, no moving parts, high reliability, low maintenance and fuel flexibility [1–4]. Generally, residential SOFC system discharges high-temperature exhaust gas, which can be utilized for space heating, process steam and domestic hot water demands because of its high-grade waste heat and partial unreacted fuel [5].

Currently, the recycling of stack exhaust gas includes (i) direct exhaust heat exchange and (ii) exhaust chemical energy recycling (e.g. exhaust gas combustion (EGC), anode off gas recovery (AOGR), and cathode off gas recovery (COGR)). Conventionally, recirculation of SOFC system exhaust gas is largely based on direct heat exchange, which cannot make full use of the energy in exhaust, leading to lower fuel utilization. Therefore, developing new and more efficient ways for the substantial utilization of chemical energy in exhaust gas are globally encouraged.

EGC model has been integrated in a SOFC-CHP system to use the chemical energy in exhaust gas [6–12]. Partial anode exhaust heat generated by combustion was used to vaporize water, preheat air, reform raw material [13–15]. Consequently, thermal efficiency of the system can be improved significantly by EGC model. Based on existing research, fuel utilization, cathode off-gas temperature and excess air ratio had exercised a great influence on EGC model [16]. Lower fuel rate or higher fuel inlet temperature is favorable for the improvement of the electrical efficiency in system with EGC model. However, it would lead to a significant reduction of the system thermal efficiency [17]. AOGR model is a derivative system concept whereby depleted exhaust gases from the anode outlet are recirculated to the reformer [18–22]. It could significantly improve the reforming efficiency and effectively inhibited carbon deposition by means of recovering anode off gas [23]. What’s more, the recycling of anode off-gas is of high importance to the overall system efficiency and maintaining the designed steam to carbon
ratio (S/C) [24]. Works in literature demonstrated that improvement of steam reforming efficiency facilitate the increase of system efficiency monotonically [25]. Additionally, recovery of anode off-gas would dramatically enhance the comprehensive efficiency in systems with steam reforming process compared to the systems with partial oxidation process [26].

The early contributions on exhaust chemical energy recycling development illustrated high potential to increase system efficiency and fuel utilization of SOFC-CHP. However, there are still several major technological limitations. Firstly, all the current EGC system are based on single stage (SS) design, in which the exhaust gas from fuel cell is directly fed to the combustor. In SS-EGC, the extremely high gas temperature (up to 1200 °C) induced by exhaust combustion will adversely affect the system stability, safety and material cost. Therefore, advanced operational strategy with lower gas temperature in recirculation need to be developed. Secondly, there is still large room to improve the CHP efficiency by system optimization. This requires a thorough understanding on the design and operational parametric effects, such as the effect of specific AOGR ratio on system efficiency, which is also lack in literature. Moreover, a number of exhaust chemical energy recycling designs (such as EGC, AOGR, AOGR & COGR, AOGR & EGC) have been proposed, but were studied in different conditions with different software/experimental setups, leading to the ineffective comparison of various options and determination of preferred design. Thus, a systematic and comprehensive comparative study on different recirculation modules for exhaust chemical energy recycling is highly desired.

To fill the research gaps, in this paper, an innovative multi-stage EGC (MS-EGC) system coupled with an additional air preheater was built up to reduce the combustion gas temperature, which makes the system safer and more secure. In the MS-EGC, the thermal energy and chemical energy are recycled in different stages. At the first stage, the thermal energy in anode exhaust is recycled to preheat the intake air, and, then at the consequent stages, the combustion of exhaust makes a further utilization of the exhaust chemical energy. Based on the MS-EGC design, the influence of AOGR ratio on system electrical efficiency was analyzed and reasonable AOGR ratio were derived for different conditions. With the optimized design, we then compare the electrical and thermal efficiency calculated from four derivative systems working at same condition, leading to the conclusion that the AOGR & EGC system was the most efficient. According to the simulation results, a reactor integrating MS-AOGR & EGC modules for a 1 kW SOFC-CHP system was developed to realize the conceptual design and prove the technology feasibility.

2. Methodologies

2.1. Process design

To make full use of the energy in exhaust gas of SOFC-CHP, four systems with different Multi-Stage Exhaust Energy Recycling model (EGC, AOGR, AOGR & COGR, and AOGR & EGC) are designed in Fig. 1. A SS-EGC model is also included in the study as benchmark (Fig. 1(a)).

2.1.1. EGC SOFC-CHP system design

EGC module transfers chemical energy of exhaust gas into thermal energy in the afterburner, which located immediately downstream of the stack thus increasing exhaust’s emission temperature. However, ultra high temperature in the burner may cause higher grade material requirement, lower operation safety and service life of the equipment. Fig. 1(a) shows a traditional system with SS-EGC model. Here, we propose and apply a MS-EGC process to address this problem. Fig. 1(b) depicts an optimized process flow diagram for a conceptual methane-fueled SOFC system operating near atmospheric pressure with MS-EGC module. With the multi-stage design, temperature in anode exhaust can be reduced by heat exchange with the intake air, and then the chemical energy in the anode exhaust was used by combustion. Based on the design philosophy mentioned above, three derivative system variations (AOGR, AOGR & COGR and AOGR & EGC) were further proposed to make full use of exhaust chemical energy.

2.1.2. AOGR SOFC-CHP system design

Anode off gas recycle (AOGR) is a system concept whereby depleted exhaust gases from the anode outlet are recirculated to the reformer providing water vapor and heat to assist in methane reforming and inhibit carbon deposition. A gas ejector is used to accomplish the anode off recycle, and system complexity decreases due to the saving of additional water and external thermal energy supply. The system diagram is represented in Fig. 1(c).
2.1.3. AOGR & COGR SOFC-CHP system design

Cathode off gas recycle is a prospective system concept whereby cathode exhaust gases are recirculated to the fuel cell inlet to reduce the consumption of air compressor by reducing the demand for air. Gas ejectors are used to accomplish the anode and cathode off recycle. Fig. 1(d) delineates a SOFC-CHP system with AOGR & COGR module where anode off gas is recycled to reformer and cathode off gas is recycled to cathode chamber simultaneously.

2.1.4. AOGR & EGC SOFC-CHP system design

In this design, the anode off gas is partially recycled by directly leading it to reformer and the rest was lead to the burner. The combination of AOGR & EGC may be a potential approach for a higher enhancement of system efficiency. Subsequently, Fig. 1(d) displays such a complementary SOFC-CHP system with AOGR & EGC module.

2.2. Numerical modeling

2.2.1. Electrochemical model

Single cell electrochemical model is developed in FORTRAN program and integrated in the commercial software Aspen Plus™, and subsequently converted into a subroutine capable to work as an Aspen Plus™ Unit Operation [25]. Parameters in simulation are subsequently converted into a subroutine capable to work as an

\[
E = \frac{-AG(T_i)}{2F} + R_i \frac{T_i}{2F} \ln \left( \frac{P_{H_2}/P_{H_2O} \cdot P_{H_2O}^{0.5}}{P_{H_2O}^{atm}} \right) \quad (1)
\]

\[
V = E - \eta_{act} - \eta_{ohm} - \eta_{ion} \quad (2)
\]

where \(E\) is the open circuit voltage (V), \(V\) is the single cell voltage (V) and \(\eta\) indicates the voltage loss (V) due respectively to activation polarization \(\eta_{act}\) (V), ohmic resistance \(\eta_{ohm}\) (V) and concentration polarization \(\eta_{ion}\) (V).

The activation over-potentials are obtained via the Butler–Volmer equation, where \(i_0\) is the exchange current density (A m\(^{-2}\)), and \(i\) is the current density (A m\(^{-2}\)).

\[
\eta_{act} = \frac{R_i T_i}{F} \sinh^{-1} \left( \frac{i}{2i_0} \right) + \frac{R_i T_i}{F} \sinh^{-1} \left( \frac{i}{2i_0} \right) \quad (3)
\]

The anodic and cathodic exchange current density (A m\(^{-2}\)) can be defined as Eqs. (4) and (5) [25]. The value of \(\gamma_a, \gamma_c, E_{act,a}, E_{act,c}\) are given in Table 1.

\[
\eta_{ohm} = \frac{1}{v} \sum \frac{A_i \delta_i \exp \left( \frac{B_i}{T_i} \right)}{i_0} \quad (6)
\]

where \(A_i\) is the ohmic loss coefficients of each layer (anode, cathode and electrolyte) and \(\delta_i\) is the corresponding thickness (m).

| Coefficient | Anode | Cathode | Electrolyte |
|-------------|-------|---------|-------------|
| A/(Ωm)     | 1.05 \times 10^{-5} | 2.38 \times 10^{-5} | 6.029 \times 94 T^{-1} |
| b/K         | 1150  | 1200    | 10,300      |
| \delta/m    | 5 \times 10^{-4} | 5 \times 10^{-5} | 2 \times 10^{-5} |
| c/mol      | 70,000 | 90,000  | 90,000      |
| \gamma/(A cm²) | 79,000 | 79,000  | 79,000      |
| D_{eff}(cm² s⁻¹) | 2.1 \times 10^{-3} | 5.4 \times 10^{-6} |
Concentration losses are calculated using Eq. (7) widely adopted in literature with proven accuracy [25,29,30], where \( i_{\text{L,A}} \) and \( i_{\text{L,C}} \) are limiting current (A m\(^{-2}\)) densities for anode and cathode, respectively and shown in Eqs. (8) and (9), and \( D_{\text{eff}} \) is the effective gaseous diffusivity through the electrode [30].

\[
\dot{n}_{\text{H}_2} = \frac{\frac{1}{A} - 2 \frac{\dot{n}_{\text{H}_2 \text{eq}} \cdot F}{A}}{1} - (\text{H}_2 + \text{CO} + 4\text{CH}_4) \tag{10}
\]

Stack output power \( P_{\text{DC}} \) (W), electrical efficiency of the stack \( \eta_{\text{SOFC}} \), net system electric efficiency \( \eta_{\text{sys.e}} \), system thermal efficiency \( \eta_{\text{heat}} \), and system CHP efficiency \( \eta_{\text{SOFC-CHP}} \) are described by the following equations:

\[
P_{\text{DC}} = V \cdot i \cdot A \tag{12}
\]

\[
\eta_{\text{SOFC}} = \frac{P_{\text{DC}}}{P_{\text{in,SOFC}}} \tag{13}
\]

\[
\eta_{\text{sys.e}} = \frac{P_{\text{AC,net}}}{P_{\text{in,SOFC}} \cdot \text{LHV}_{\text{fuel}}} \tag{14}
\]

\[
\eta_{\text{heat}} = \frac{Q_{\text{rec}}}{Q_{\text{in,SOFC}} \cdot \text{LHV}_{\text{fuel}}} \tag{15}
\]

\[
\eta_{\text{SOFC-CHP}} = \frac{P_{\text{AC,net}} + Q_{\text{rec}}}{P_{\text{in,SOFC}} \cdot \text{LHV}_{\text{fuel}}} \tag{16}
\]

where \( P_{\text{DC}} \) is DC power generated (W) by the stack, \( P_{\text{AC,net}} \) is net AC power generated (W) by the system, \( Q_{\text{rec}} \) (W) is heat which can be used by other blocks, \( (\text{HHV}_{\text{fuel}} \cdot \text{fuel}_{\text{in,SOFC}}) \) is lower heating value of fuel from anode (W), and \( (\text{HHV}_{\text{fuel}} \cdot \text{fuel}_{\text{in,system}}) \) is lower heating value of fuel from system (W). The geometric and material properties obtained from literature are given in Table 1 [31,32].

### 2.2.2. Process flow model

A typical SOFC-CHP system includes several components, including blowers, pumps, heat exchangers, combustors, water tanks, and others. Operating conditions and their characteristics exhibit different patterns [8]. In regard to the complexity of the SOFC stack, the following assumptions and considerations are applied to the present model:

1. It is a zero dimensional steady state isothermal model.
2. Temperature at anode outlet and cathode outlet are identical [25,29].
3. All working fluids are assumed as ideal gas with no pressure drop. This assumption has been proven reasonable in many literatures for SOFC stack and system modeling [17,30].

There are no direct electrochemical modules in Aspen Plus\textsuperscript{TM}, so a Gibbs reactor, ‘Anode’, a separator module, ‘Cathode’, and a heater module, ‘Heater’ are used to simulate the electrochemical reaction.

As Aspen Plus\textsuperscript{TM} is not capable of simulating the cell half reactions e.g. Eqs. (17) and (18), the overall electrochemical reaction (Eq. (19)) [25] is used in ‘Anode’:

Cathode reaction: \( \frac{1}{2} \text{O}_2 + 2e^- \rightarrow \text{O}^{2-} \) \tag{17}

Anode reaction: \( \text{H}_2 + \text{O}^{2-} \rightarrow \text{H}_2\text{O} + 2e^- \) \tag{18}

Overall reactions: \( \text{H}_2 + \frac{1}{2} \text{O}_2 \rightarrow \text{H}_2\text{O} \) \tag{19}

The function of the ‘Cathode’ block is to separate oxygen from air, based on the fuel utilization at cell stack. In the simulation, the fuel of CO and CH\(_4\) in SOFC stack have been converted to Hydrogen Equivalent (H\(_2\),eqv) to account for their contribution to the overall electrochemical reactions. Such simplification has also been widely used in SOFC process modeling [25,29]. Accordingly, the split ratio of O\(_2\) is used to calculate the amount of O\(_2\) consumed in ‘Anode’ by using the Aspen Plus calculator block and Eqs. (20)–(23) [33].

\[ n_{\text{H}_2\text{eq.in}} = n_{\text{H}_2\text{syngas}} + n_{\text{COsyngas}} + 4n_{\text{CH}_4\text{syngas}} \] \tag{20}

\[ n_{\text{H}_2\text{consumed}} = U_{\text{f}} \cdot n_{\text{H}_2\text{eq.in}} \] \tag{21}

\[ n_{\text{O}_2\text{consumed}} = 0.5 \cdot n_{\text{H}_2\text{consumed}} \] \tag{22}

\[ n_{\text{O}_2\text{fuel}} = \frac{n_{\text{O}_2\text{consumed}}}{n_{\text{O}_2\text{in}}} \] \tag{23}

Specifically, in Eqs. (20), the Hydrogen Equivalent conversion of fuel was calculated based on the follow equations according to the mole flow of H\(_2\):

\[ \text{Steam reforming reaction:} \]

\[ \text{CH}_4 + \text{H}_2\text{O} \leftrightarrow 3\text{H}_2 + \text{CO} \] \tag{24}

\[ \text{Water-gas shift reaction:} \]

\[ \text{CO} + \text{H}_2\text{O} \leftrightarrow \text{CO}_2 + \text{H}_2 \] \tag{25}

The function of heat exchanger module is to maintain a constant stack temperature by removing heat from the stack to cooler air in heat-matched flux. Energy released from stack consists of electrical energy and thermal energy. ‘Q\(_{\text{elec}}\)’ based on voltage formula represents output power and ‘Q\(_{\text{th}}\)’ represents heat transferred by the air. Calculator block and voltage formula facilitates the determination of the desired air flux.

In this study, steam reforming reaction is the predominant process for the realization of fuel pretreating. ‘Reformer’ is a Gibbs reactor module which simulates the adiabatic steam reforming of fuel by utilizing the incoming steam and the fuel stream mixture. By setting ‘Q\(_{\text{th}}\)’ as zero, temperature of reformer is calculated using calculator block. Eqs. (24) and (25) present the general equations of steam reforming (SR) and WGS reactions, which both contribute in hydrogen production apart from hydrocarbons simultaneously.

### 2.3. Experiment

As confirmed by our modeling analysis, to be described in Section 3.2, system with multi-stage exhaust gas combustion (MS-EGC) will reduce the system operating temperature from 1149 °C to 830 °C, which significantly increases the safety of system operation and reduces the material requirement. The MS-EGC is the fundamental process. Consequently, a reactor
integrating catalytic combustion, heat exchange and steam reforming was developed for MS-EGC modules to match the 1 kW SOFC-CHP system. The stack exhaust gas was combusted to provide heat for reforming. Experiments were carried out to investigate the effect of fuel utilization and inlet temperature of combustion gas on properties of the reactor.

2.3.1. Reactor configuration
The experimental system process is shown in Fig. 2. Stainless steel 316 was used to fabricate the reactor consisting of two covers, where steam reforming catalyst and catalytic combustion catalyst were placed. The thickness of the heat exchange surface was 0.2 mm and its area was designed as 0.05 m² (width: 100 mm, length: 500 mm) to adapt substantially heat transfer demand of approximately 700 W in 1 kW SOFC-CHP system. The combustion chamber had a volume of 0.75 L, and groove depth was 15 mm according to the 10,000⁻¹ GHSV of the catalytic combustion catalyst. The reforming chamber volume was 1.5 L, and groove depth was 30 mm in terms of the 500–20,000⁻¹ GHSV of the steam reforming catalyst. Temperature measuring points were set up every 80 mm on both sides of the reactor. 12 K-type thermocouples were used to the simultaneous exploration of the temperature along with the flow direction in the two channels.

2.3.2. Experiment condition
The reactor was designed to match the 1 kW SOFC-CHP system. In order to effectively evaluate the performance of coupled reactor and to exclude the impact of fluctuations from SOFC-CHP system, the combustion inlet parameters were set up in line with the actual operation conditions in 1 kW stack (e.g. temperature, gas composition, flow rate of anode off gas). Air flow rate dependent to the stack fuel utilization was determined by the flux of cathode off gas. The initial experimental conditions based on stack operation temperature of 750 °C and pressure of 1.05 bar are as follows. The inlet temperature of combustion gas is 300 °C, air-to-fuel ratio is 10:1, fuel utilization of the stack is 65% and water-to-carbon

Fig. 2. Schematic diagram of experimental setup: (a) system process, and (b) reactor configuration; (c) digital photograph of the coupled reactor.
ratio is 3:1. The outlet temperature of steam generator is 230 °C, preheating temperature of mixture including steam and methane is 300 °C, and gas inlet pressure is 1.05 bar. The accuracy of mass flux controllers which control mass flux of CH₄, H₂, N₂ and air in system is ±1% FS. The accuracy of thermocouples and high pressure infusion pump is ±1.5 °C and ±0.25%, respectively.

1 kW SOFC power generation system operated normally when hydrogen concentration varied between 50% and 100% and flow rate was approximately at 11 L·min⁻¹. In this study, the concentration and flow rate of hydrogen in our experiment are as follows: hydrogen concentration was 66.2–73% and flow rate was 11.1–13.1 L·min⁻¹ when Uᵢ changed from 0.5 to 0.7. Hydrogen concentration was 66.2–68.7% and flow rate was 10.7–11.8 L·min⁻¹ when inlet temperature of combustion gas in the range of 150–350 °C. As described above, with an appropriate value of process parameters, it was validated that the concentration and flow rate of hydrogen were all in line with the requirements for 1 kW SOFC power generation system.

3. Results and discussion

3.1. Model validation

To validate our numerical model, a SOFC-CHP system in literature [34] was simulated. This experimental system in literature [34] includes SOFC with reformer, recuperator, blowers, heat exchangers, combustors, condenser and others, which is similar with the ones in this study. The simulation results were compared with experimental results (literature [34]) with variation of fuel flow rate as shown in Fig. 3.

The results obtained by Aspen Plus in the current study showed a high conformity with experimental results in literature, which explained the reliability of the process model. The error between experimental and numerical values in output power and electrical efficiency was acceptable within our investigation conditions. The slight error was resulting from simulation process hypothesis a constant stack temperature, in practical operation, temperature of SOFC stack varies in a positive correlation with constant fuel efficiency in cell stack. What’s more, it is shown that even after first-stage thermal energy recycling, the combustion gas from the second-stage chemical energy recycling contains sufficient energy to support the fuel processing reactions such as reforming and gasification in the MS-EGC. As a result, in the new design, the thermal efficiency and electrical efficiency reaches 47.4% and 39.4%, respectively, compared to the traditional SS-EGC system with thermal efficiency and electrical efficiency reaches 41.5% and 42.3% respectively. It clearly demonstrated that the new design of MS-EGC increases the system operation safety and reduces requirement for equipment and materials by significantly lowering down the system operating temperature, without compromising the system performance and efficiency.

3.2. Numerical analysis

Four different system processes are built in Aspen Plus as shown in Fig. 4. Table 2 illustrates the parameters for the case under basic conditions in numerical simulation process. The output powers of four systems are guaranteed at 1 kW by adjusting the fuel flux while keeping the decisive design parameters (Table 2) constant in simulation. Comparison of the efficiency helps to delineate merits and demerits of the four different system flows.

3.2.1. Multi-stage EGC SOFC-CHP system process

A new system design with multi-stage EGC model is represented in Fig. 4(a). According to the principle of graded utilization of energy, thermal energy in anode exhaust was used to preheat air at the first step. Excess temperature was decreased to appropriate range at the same time. At the second step, combustion of unreacted component in anode exhaust achieves full use of anode exhaust chemical energy. The thermal efficiency, electrical efficiency and afterburner temperature of the newly developed multi-stage and conventional single stage systems are compared in Fig. 5. For the newly design MS-EGC operating with air preheated and exhaust gas cooled, the gas temperature after combustion reaches 830.8 °C, slightly higher than the stack temperature. However, in traditional SS-EGC system, the temperature in afterburner is up to 1149 °C, due to direct combustion of exhaust gas.

3.2.2. Effect of AOGR ratio on system efficiency

In order to take full advantage of anode exhaust chemical energy, Fig. 4(b) depicts a multi-staged system with AOGR model. Recirculation of partial anode off-gas is favorable for more effective utilization of unreacted component in exhaust gas delivered to the reformer. In addition, water vapor in anode off-gas has contributed to the reforming reaction. This system provides an electrical efficiency gain of 11.9% and a thermal efficiency reduction of 16.1%, and air-fuel ratio is approximately improved to 9.8:1 for larger air flow. Moreover, cathode off-gas with higher temperature is used for preheating the coming air involved in the stack to take full advantage of thermal energy in system. By this way, Overall fuel utilization is enhanced advantageously to 88.2%, thus increasing the electrical efficiency of the whole system with remaining constant fuel efficiency in cell stack.
System performance changes with the different AOGR ratios. The effect of AOGR ratios on system performance in the numerical simulation process are plotted in Fig. 6. Adopting higher AOGR ratios initially increases the stack output power, resulting from higher amount of H₂, CO and CH₄ into the anode. However, the output power starts to decrease when AOGR ratios exceed 0.7. It is
because the system power consumption will increase with the increase in AOGR ratios. Consequently, electrical efficiency increases first and then decreases under the combinational influence of above two factors. Additionally, thermal efficiency monotonically increases with the improvement of AOGR ratios. In another word, more amount of chemical energy is converted to thermal energy. The results indicate that higher system electrical efficiency is achieved at lower AOGR ratio, while higher system thermal efficiency is achieved at higher AOGR ratio, which provides a new control strategy to obtain a variation of thermos-electric output proportion of SOFC-CHP on demand by adjusting the AOGR ratio.

### 3.2.3. Efficiency comparison

Analyzing and optimizing the 1 kW SOFC-CHP system via different methods of multi-stage exhaust recovery were conducted. Process efficiencies of the four derivative systems in the numerical simulation process are shown in Fig. 7. It is found that both MS-EGC and MS-AOGR modules lead to a better system integration and higher efficiency by further recovery of exhaust chemical energy. The MS-EGC module has a considerably positive impact on system thermal efficiency because of the combustion of stack exhaust gas, while the MS-AOGR module is beneficial to the enhancement of system electrical efficiency due to the utilization of unreacted component in anode off-gas. The MS-COGR achieves a significant reduction on the air demand and compressor power consumption. However, it will decrease the oxygen partial pressure, causing decline in output power. Thus, the MS-AOGR & COGR system shows a negligible enhancement in net electrical efficiency compared to the MS-AOGR system. On the other hand, the MS-AOGR & EGC system demonstrated significant advantages by combining the benefits from both EGC and AOGR. The MS-AOGR & EGC system shows 7% increase in thermal efficiency compared to the MS-AOGR module, and 12% increase in electrical efficiency compared to the MS-EGC module. As a result, the MS-AOGR & EGC module is capable of an overall cogeneration efficiency of 92%, which leads to the best comprehensive performance among the four systems.

---

**Table 2**

| Key parameters                  | Value       | Key parameters                  | Value       |
|---------------------------------|-------------|---------------------------------|-------------|
| Stack temperature/°C            | 750         | Cell active area/m²             | 0.137       |
| Air and fuel inlet temperature/°C | 25          | Steam to carbon mole ratio      | 3:1         |
| Cooling water inlet temperature/°C | 25          | AOGR ratio/%                   | 60          |
| Exhaust emission temperature/°C | 60          | Fuel utilization U_s (single step)/% | 60          |
| Anode outlet temperature/°C     | 750         | Air volume fraction (O₂:N₂)/%    | 21:79       |
| Cathode outlet temperature/°C   | 750         | Fuel compressor isentropic efficiency/% | 85          |
| Cell pressure/bar               | 1.05        |                                 |             |

---

**Fig. 5.** Thermal efficiency, electrical efficiency and afterburner temperature of the new MS-EGC and conventional SS-EGC SOFC-CHP systems.

**Fig. 6.** (a) Effect of AOGR rate on stack voltage, system efficiency and fuel utilization in numerical simulation process. (b) Effect of AOGR rate on stack output power and compressor dissipation power in numerical simulation process.

**Fig. 7.** System efficiency of the four processes in numerical simulation process.
3.3. Experimental optimization

A reactor integrating catalytic combustion, heat exchange and steam reforming was developed for MS-EGC modules to match the 1 kW SOFC-CHP system. Experimental results on performance of MS-EGC process were presented as follow.

3.3.1. Effects of fuel utilization

In order to investigate the influence of fuel utilization on reforming conversion efficiency in MS-EGC system, the reactor is operated under $U_f$ between 50% and 90% with an interval of 10%. Fig. 8(a) shows the reactor temperature measured by experiment under different fuel utilizations.

Increasing stack $U_f$ results in an obvious decrease in overall temperature of the reactor. When $U_f$ increases from 50% to 90%, the maximum temperature of the combustion side falls from 1014.1 °C to 448.7 °C due to the drop of combustion gas concentration in exhaust. The reduced heat supply from combustion side leads to the decline of temperature in reforming reaction where the maximum temperature decreases from 778.5 °C to 328.4 °C. It then results in the sharp increase of CH₄ concentration in reformate because of the decline of reforming efficiency. Fig. 8(b) further shows the concentration of reforming products under various conditions. With the increase of stack $U_f$, the concentration of CH₄ at the outlet of the reformer raises dramatically from 2.6% to 89.8% while the concentration of H₂ reduces from 73.5% to 7.6%. Corresponding trend is more significant when stack $U_f$ is higher than 80%. Consequently, reforming reaction cannot take place adequately because of insufficient thermal energy in this condition.

3.3.2. Effects of inlet temperature

According to the process simulation, in SOFC operation, the temperature of exhaust gas is 297.6 °C after heat exchange with air while the stack temperature is 750 °C. Therefore, in the experiment, the inlet temperature of combustion channel is set between 150 °C and 350 °C with 50 °C interval. The tested temperature distribution and species concentration distribution in the reactor process are shown in Fig 8(c) and (d), respectively.

The temperature of combustion gas increases instantly above 800 °C at the entrance of channel and gradually decreased along the flow direction, indicating that the catalytic combustion reaction is so fast that it burns completely at the entrance. It is also found that the reforming process is not sensitive to the inlet temperature variation. With the increase in the inlet gas temperature from 150 to 350 °C at combustion side, the H₂ concentrations only rises slightly from 66.2% to 68.7% while the CH₄ concentration decreases from 13.4% to 6.2%. Since lower inlet gas temperature at the combustion side only shows minor effect on methane conversion, it is recommended that heat exchange between exhaust gas and intake air should be conducted prior to the catalytic combustion to improve system heat efficiency without obvious trade-
off to reforming reaction efficiency. It is worth noting that the recycling of thermal energy rarely affects the subsequent utilization of chemical energy in exhaust according to the experimental results, which further confirms the advantages of the MS-EGC design.

4. Conclusions

In this study, a SOFC-CHP system integrating multi-stage exhaust energy recycling model is optimized, which makes the maximum temperature of system decrease from 1149 °C to 830 °C. Therefore, it significantly increases the safety of system operation and reduces the material requirement. In the innovative MS-EGC system, thermal energy in exhaust was recycled to pre-heat the intake air, and the chemical energy was recycled by combustion according to the principle of graded utilization of energy. Consequently, both thermal and chemical energy in exhaust were used efficiently and intensively in MS-EGC system. Corresponding AOGPR ratio range was determined to match different requirements. Lower AOGPR ratio (<0.7) will contribute to the enhancement of electrical efficiency, achieving 62.1% maximum system electrical efficiency. Higher AOGPR ratio (>0.7) will help to the improvement of thermal efficiency up to 35.9%. Based on above research, four SOFC-CHP system processes, i.e. processes with EGC, AOGPR, AOGPR & COGR and AOGPR & EGC, are compared to each other in terms power efficiency, thermal efficiency and the efficiency of cogeneration. It is found that system with AOGPR & EGC modules leads to the best comprehensive performance, with electrical efficiency of 59.3%, thermal efficiency of 32.8%, and overall co-generation efficiency of 92%. Furthermore, according to the simulation results, an integrated reactor combing the EGC modules was developed to realize the proposed strategy. Optimized operation conditions and methodologies were obtained by experiment. The study will provide profound insights for future analysis of effective SOFC-CHP system.

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