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Comparison study on different ITM (Oxygen ion transfer membrane)-integrated molten carbonate fuel cell hybrid systems with CO_2 recovery using sweep gas

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Abstract:

ITM (Oxygen ion transfer membrane) is a kind of ceramic membrane which can separate oxygen from air with low energy consumption. Previous study shows the ITM-integrated MCFC (molten carbonate fuel cell) hybrid system with CO_2 recovery can maintain high efficiency when recovering 85% of CO_2 emission, however the oxygen partial pressure at the ITM permeate side is usually 1atm, which requires a very high pressure ratio of the air compressor for ITM without sweep gas to separate oxygen, using sweep gas can solve this problem. In this paper the ITM-integrated MCFC hybrid systems with CO_2 recovery using different sweep gases are studied. With the Aspen plus software, two systems with different sweep gases are established and their performances are compared with the benchmark system without sweep gas, the effects of key parameters on the optimum system performance are also investigated. Results show that compared with the benchmark system, the efficiencies of the systems with sweep gas are increased and the pressure ratio of air compressor is decreased, the system using pure CO_2 as sweep gas can improve the system efficiency by 1.25 percent points, which is superior to the system using the mixture gas of CO_2 and H_2O as sweep gas. Achievements from this paper will provide a valuable reference for CO_2 recovery from the MCFC hybrid power system with lower energy consumption.

Keywords:

ITM, MCFC hybrid system, Sweep gas, CO_2 recovery.

1.Introduction

Excessive emissions of greenhouse gases such as carbon dioxide and methane have caused the global temperature increase in recent years [1]. Reducing the emission of CO_2, one of the major greenhouse gases, would ease the trend of global warming. The main source of CO_2 emission is the burning of fossil fuels such as coal and natural gas [2], so all power generation plants that utilize fossil fuels, including coal-fired power plant, NGCC (natural gas combined cycle), IGCC (integrated gasification combined cycle), fuel cell, etc, would bring a large amount of CO_2 emissions.

Currently, the MCFC (molten carbonate fuel cell) has attracted more and more attentions because of its cleanliness and high efficiency, unlike traditional power generation methods, MCFC generates the electricity through an electrochemical reaction and its efficiency is not limited by Carnot cycle. Moreover, the temperature of MCFC exhaust gas is usually as high as 650℃, such high temperature makes it possible for MCFC to integrate with other power generation devices such as HRSG (heat recovery steam generator) and steam turbine system to constitute a more efficient hybrid power generation system. The power generation efficiency of MCFC is high, and the exhaust gas is clean with little pollutants for it is usually fueled by natural gas. However, the natural gas is also carbon-contained, and the CO_2 gas is still generated and discharged to the environment from MCFC. Although MCFC hasn't yet reached the stage of commercial operation with a large-scale capacity [3], and the amount of CO_2 emission is not comparable with that of traditional power generation methods, however, the development of MCFC is very fast in recent years and lots of related studies have emerged. Stefano Campanari [4] proposes an idea to place a MCFC system downstream a conventional "combustion fired" power plant, feeding the cathode with its exhaust gases with the aim of concentrating and then separating a fraction of the CO_2 otherwise vented, results show the overall net system efficiency including MCFC output power and gas treatment consumption slightly increases from the original 45% of the simple stream plant to 45.8% of the new plant when achieving a global CO_2 separation efficiency of 77%. Morris Brenna [5] proposes an innovative lay-out based on the use of MCFC applied to gas turbine exhaust gas to capture CO_2, results show the net output power of MCFC lay-out is about 20% higher than that of the reference power cycle with an efficiency reduction of 1%. Fumihiko Yoshiba [6] calculates the efficiency of an integrated coal gasification system equipped with a molten carbonate fuel cell, a gas turbine and a steam turbine (IG/MCFC), the calculation results reveal that a high efficiency system with CO_2 recovery is possible by applying the cathode gas in the IG/MCFC systems. C. Tomasi [7] examines a plant configuration based on a MCFC and a circulated fluidized-bed reactor which has been applied to the thermal conversion of many types of biomass, results demonstrate that the proposed coupling of a circulated fluidized-bed gasifier and an MCFC system presents high conversion efficiency (43-49%), which is better than that of the traditional fossil-fuel plants with the same size. G. Rinaldi [8] studies the separation of CO_2 in a biogas plant that co-produces electricity, hydrogen, and heat, and three potential CO_2 concentrating configurations are numerically simulated to evaluate potential CO_2 recovery rates, results show that carbon separation and hydrogen co-production processes are compatible and a series configuration of MCFC technology coupled with an ICE (internal combustion engine) achieves outstanding carbon recovery (exceeding 90%), with minimal parasitic load. It is obvious that MCFC technology has attracted a wide attention with so many related studies and its commercialization is coming soon. With the increase of MCFC capacity the amount of fuels needed would increase accordingly, which means more CO_2 emissions, thus it is also necessary to capture the CO_2 emitted from MCFC.

The main method for controlling CO_2 emissions is CCS (CO_2 capture and storage) technology [9-11], which could help to realize the CO_2 reduction goal before the end of this century according to IPCC 2005 report [12]. The CO_2 capture technology could be categorized into three kinds of technical routes [13]: pre-combustion capture, post-combustion capture and oxy-fuel combustion. The oxy-fuel combustion method could capture 100% CO_2 theoretically without producing nitrogen oxides pollution, however the process of producing pure oxygen consumes large amount of energy which would decrease the system efficiency greatly [14]. To solve this problem, the author of this paper proposes an ITM (oxygen ion transfer membrane)-integrated MCFC hybrid system with CO_2 recovery based on oxy-fuel combustion method, the most important feature of this system is that the unreacted MCFC fuel gas is sent to a afterburner in which oxy-fuel combustion is taken place, so the outlet gas of afterburner is a mixture of CO_2 and H_2O which is easy to separate. In this system, the energy consumption for oxygen production is low because only a little oxygen is needed to make the unreacted fuel gas completely combusted, moreover, the integrated ITM technology could further reduce the energy consumption of producing oxygen.

However, the hybrid system mentioned above has a problem that the outlet pressure of the air compressor at the ITM feed side needs to be very high due to the relatively high oxygen partial pressure at the permeate side. Guiding the sweep gas into ITM permeate side can reduce the oxygen partial pressure which in turn will reduce the required outlet pressure of air compressor at the ITM feed side [15]. So the main purpose of this paper is to study the effects of using different sweep gases on the performance of ITM-integrated MCFC hybrid system with CO_2 recovery. The optimum system with the most suitable sweep gas is obtained through comparison with the benchmark system without using sweep gas both in system efficiency and exergy destruction distribution, and then, sensitive analyses are conducted on the optimum system to examine the effects of different parameters on the system performance.

2. System descriptions

2.1 Benchmark system without sweep gas

In this paper the ITM-integrated MCFC hybrid system with CO_2 recovery and without sweep gas is chosen as the benchmark system, the system layout is shown in Fig. 1. The fuel and part of circulated anode exhaust are mixed in mixer1 and then sent to the pre-reformer for reforming, the reformed gas then enters the MCFC anode. The air1 is preheated in the heater4 and heater3 and then enters into the mixer2 together with some afterburner exhaust gas and the cathode circulated exhaust gas, the mixed gas is heated in heater2 and then enters into the MCFC cathode. Electrochemical reactions take place in MCFC and the electricity is generated and output through the DC/AC converter and generator1. The cathode exhaust gas is divided into two parts by the splitter3: one part is circulated to the mixer2 to preheat the air and the other part is sent to the HRSG for waste heat recovery. The MCFC anode exhaust gas is divided into two parts by the splitter1: one part is circulated to the mixer1 to preheat fuel and the other part is sent to the afterburner for oxy-fuel combustion. The oxygen required for the afterburner combustion is provided by the air2, which is sent to the ITM feed side after being compressed to 30atm in the air compressor and exchanging heat in the heater1. In the ITM unit the air is separated into two parts: one part is pure oxygen which is sent to the afterburner for combustion and the other part is the oxygen-depleted air which expands in the air expander to produce power, the expanded air preheats the air1 in the heater3 and then is vented to the atmosphere. The afterburner exhaust gas is cooled in the heater1 and heater2, and then is divided into two parts: one part is sent to the mixer2 to preheat the air1 and provides the CO_2 required for the MCFC cathode reaction, the other part is sent to the HRSG, the exhaust gas of HRSG is a mixture of CO_2 and H_2O, so it is easy to obtain the high-purity CO_2 after cooling in the heater4 and removing H_2O in the condenser, the CO_2 is then compressed in the three-stage CO_2 compressor with intercoolers and liquefied for storage.

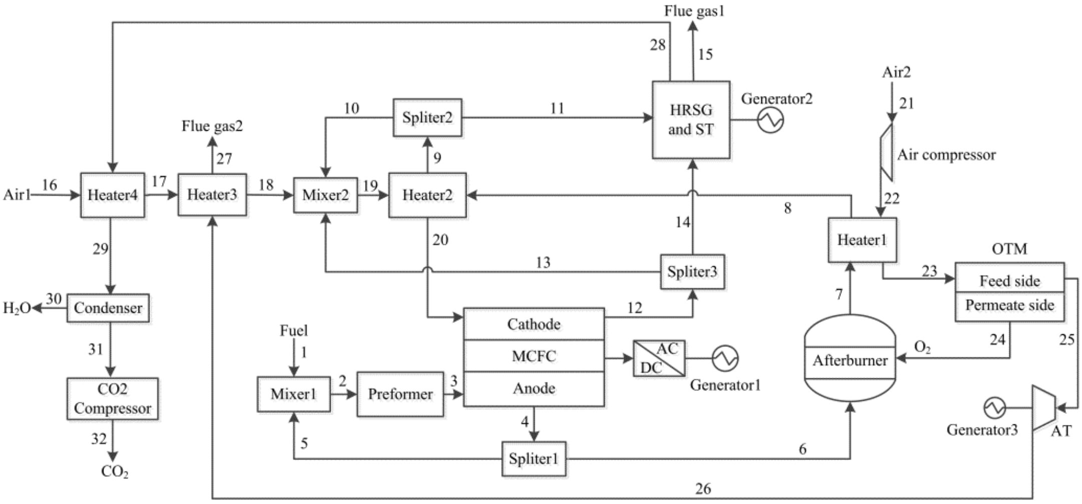


Fig. 1. Benchmark system.

2.2 New systems with sweep gas

2.2.1 Case a (using CO_2 and H_2O as sweep gas)

Layout of the new ITM-integrated MCFC hybrid system using CO_2 and H_2O as sweep gas (case a) is shown in Fig. 2. The difference with the benchmark system is that a splitter4 is added between the HRSG and the heater4, which is used to recycle part of CO_2 and H_2O to the ITM permeate side as sweep gas, besides, a heater5 is added between the heater1 and heater2, which is used to heat the sweep gas to the required temperature.

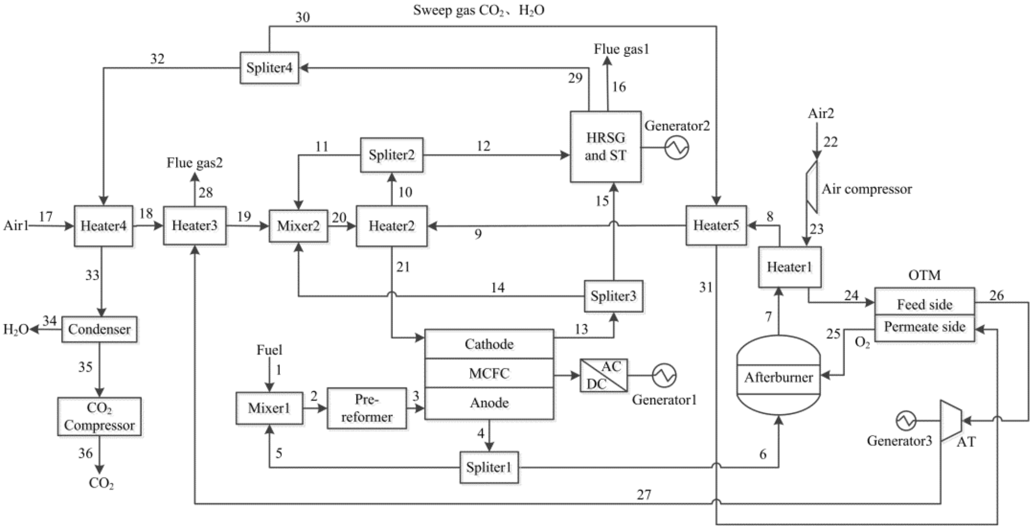


Fig. 2. case a（using CO_2 and H_2O as sweep gas）.

2.2.2 Case b (using CO_2 as sweep gas)

Layout of the new ITM-integrated MCFC hybrid system using CO_2 as sweep gas (case b) is shown in Fig. 3. Unlike case a, the position of the splitter4 is placed between the condenser and the CO_2 compressor, through which part of the high-purity CO_2 is recycled to the ITM permeate side as sweep gas.

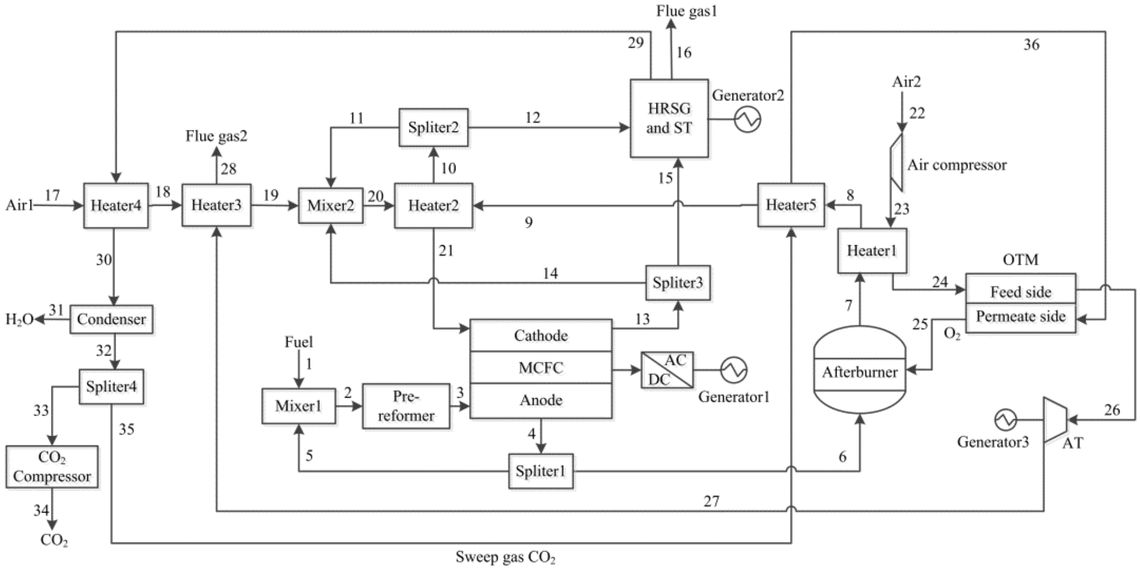


Fig. 3. case b（using CO_2 as sweep gas）.

3 System model

3.1 MCFC model

The main reactions that take place inside MCFC are as follows:

$$C_nH_{2n+2} + nH_2O = nCO + (2n+1)H_2 \quad \text{(Reform reaction)} \tag{1}$$

$$CO + H_2O = CO_2 + H_2 \quad \text{(Substitution reaction)} \tag{2}$$

$$O_2 + 2CO_2 + 4e^- = 2CO_3^{2-} \quad \text{(Cathode reaction)} \tag{3}$$

$$H_2 + CO_3^{2-} = H_2O + CO_2 + 2e^- \quad \text{(Anode reaction)} \tag{4}$$

The MCFC Nernst potential E_{Nernst} could be calculated as follows [16-17]:

$$E_{Nernst} = \frac{\Delta G}{nF} + \frac{RT}{nF}\ln\left(\frac{P_{H_2,an} \times \left(P_{O_2,ca}\right)^{1/2} \times P_{CO_2,ca}}{P_{H_2\mathrm{O},an} \times P_{CO_2,an}}\right) \tag{5}$$

$$\Delta G = 24,2000 - 45.8 \times T \tag{6}$$

When calculating the actual voltage V_{cell}, voltage losses caused by irreversible resistance losses need to be considered. The resistances are composed of anode, cathode and ohmic resistance, represented by R_{an}, R_{ca} and R_{ohm} respectively. The actual voltage is then obtained [18-19]:

$$V_{cell} = E_{Nernst} - \left(R_{an} + R_{ca} + R_{ohm}\right) \times i_C \tag{7}$$

The current density i_c is calculated as follows:

$$i_C = \frac{I}{S} \tag{8}$$

Where n is the number of electrons released in the dissociation of a H_2 molecule (equal to 2); F is the Faraday's constant (96,487 C/mol); R is the gas constant; T is the MCFC operating temperature, K; P_k is the partial pressure of the k specie, atm, the subscript *an* and *ca* refer to the anode and cathode of MCFC, respectively; I is the MCFC current, A; S is the MCFC active area, m^2.

3.2 ITM model

The ITM is a kind of nonporous, mixed-conducting, ceramic membrane that has both electronic and ionic conductivity when operating at high temperature, usually 800-900℃. Its simplified schematic is shown in Fig. 4. The driving force of oxygen separation is the oxygen partial pressure difference, so the oxygen partial pressure on the feed side (P'_{O2}) should be higher than the oxygen partial pressure on the permeate side (P''_{O2}). The ratio of P'_{O2} to P''_{O2} is defined as the oxygen partial pressure ratio (X).

$$X = P'_{O_2} / P''_{O_2} \tag{9}$$

Generally the value of X is from 5-7, in this paper the value 6.3 is chosen for the calculation.

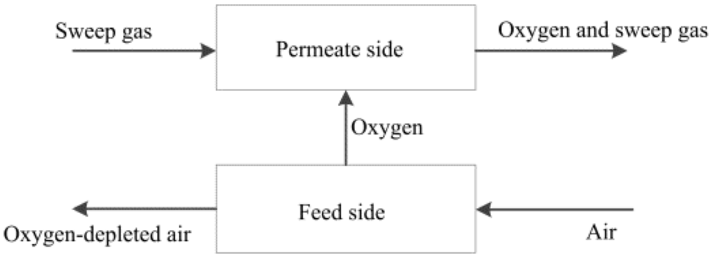


Fig. 4. ITM simplified schematic.

Based on the partial pressure difference, the separation performance of ITM can be estimated using the following equation [20]:

$$Y_{O_2} = \eta Y_{th} = \eta\left(1 - \left(\frac{1 - X_{feed}}{X_{feed}}\right)\left(\frac{P''_{O_2}}{P_{feed} - P''_{O_2}}\right)\right) \tag{10}$$

Where the Y_{O_2} is the actual oxygen separation rate; Y_{th} is the theoretical oxygen separation rate; η is a ratio of Y_{O_2} to Y_{th} (usually from 25% to 85%). In this paper η is 80%; X_{feed} is the mole fraction of oxygen in the feed gas; P_{feed} is the total pressure on the feed side, atm.

4 Results and discussions

4.1 Simulation assumptions

The input fuels of the new systems with sweep gas are the same with the benchmark system, the fuel components are 93.6% CH_4, 4.9% C_2H_6, 0.4% C_3H_8, 0.2% C_4H_{10} and 0.9% CO [21]. The operating parameters of MCFC and ITM units are shown in Table 1 and Table 2 respectively.

Table 1. MCFC unit parameters.

Parameters	Value
Fuel flow, kmol/s	0.226
Temperature，℃	650
Pressure，atm	1
Fuel utilization ratio，%	85
CO_2 utilization ratio，%	85
Steam/carbon ratio	3.5
Current density，A/m^2	1500
DC/AC converter efficiency, %	92

Table 2. ITM unit parameters.

Parameters	Value
Air compressor isentropic efficiency，%	85
Air turbine isentropic efficiency，%	88
Temperature，℃	900
Pressure，atm	30
Pressure drop，%	4
Oxygen partial pressure ratio	6.3
Outlet pressure of permeate side，atm	1

4.2 Performance comparisons of different systems

Table 3. Results comparison of different systems.

	Benchmark system	Case a	Case b
Fuel flow, kmol/s	0.226	0.226	0.226
MCFC air flow，kmol/s	4.1254	3.781	3.7832
ITM air flow，kmol/s	0.5404	0.5404	0.5404
MCFC voltage，V	0.6675	0.6639	0.67
MCFC power，MW	93.111	92.608	93.45
MCFC efficiency，%	49.21	48.94	49.39
ST(steam turbine) power，MW	28.236	28.036	28.198
Air compressor power，MW	-9.184	-4.573	-4.573
Air turbine power，MW	8.753	6.198	6.198
ITM power，MW	-0.431	1.625	1.625
CO_2 compressor power，MW	-3.544	-3.544	-3.544
System net power，MW	117.372	118.725	119.729
System efficiency，%	62.03	62.74	63.28
CO_2 capture ratio，%	85.8	85.8	85.8
Outlet pressure of air compressor，atm	30	8.04	8.04

As shown in Table 3, for the benchmark system the air for ITM unit needs to be compressed to

30atm which would cause great power consumption of the air compressor. In contrast, the new systems using sweep gas require much lower outlet pressure of air compressor under the premise of keeping the oxygen partial pressure ratio constant at 6.3. In this paper the air compressor outlet pressure of the new systems is 8.04 atm. Results show that the efficiency of the benchmark system is 62.03%, in comparison the efficiency of the new system case a is 62.74%, which is 0.71 percent point higher than that of the benchmark system, the reason is that the ITM unit can generate instead of consuming power thanks to the decrease of the air compressor outlet pressure; the efficiency of the new system case b is 63.28%, which is 1.25 percent point higher than that of the benchmark system, the reason is similar with that of the new system case a, moreover the powers generated by both MCFC and ST in the new system case b are slightly increased, which contribute to the increase of system efficiency.

4.3 Comparisons of the exergy destruction distribution

The exergy analysis is conducted for all the systems proposed in this paper, each system is divided into various subsystems and the exergy destruction of each subsystem ($E_{x,i}$) is calculated with the help of Aspen plus software, the proportion of the $E_{x,i}$ to the input fuel exergy (E_{fuel}) is defined as the exergy destruction coefficient (σ_i) as follows.

$$\sigma_i = \frac{E_{X,i}}{E_{fuel}} \times 100\% \tag{11}$$

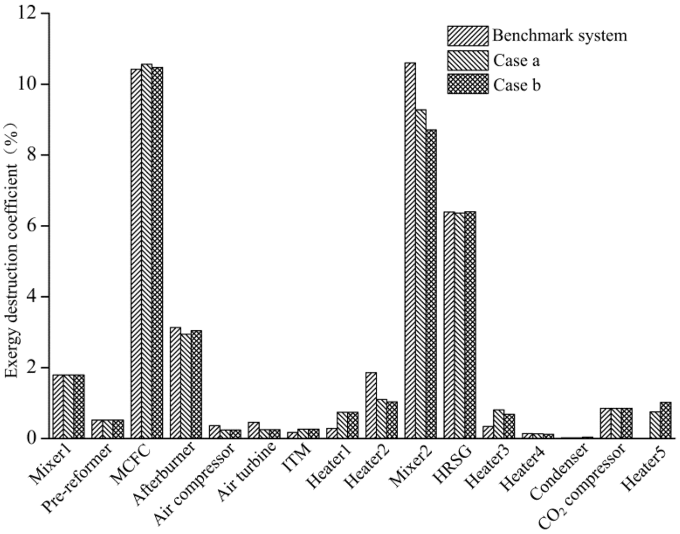


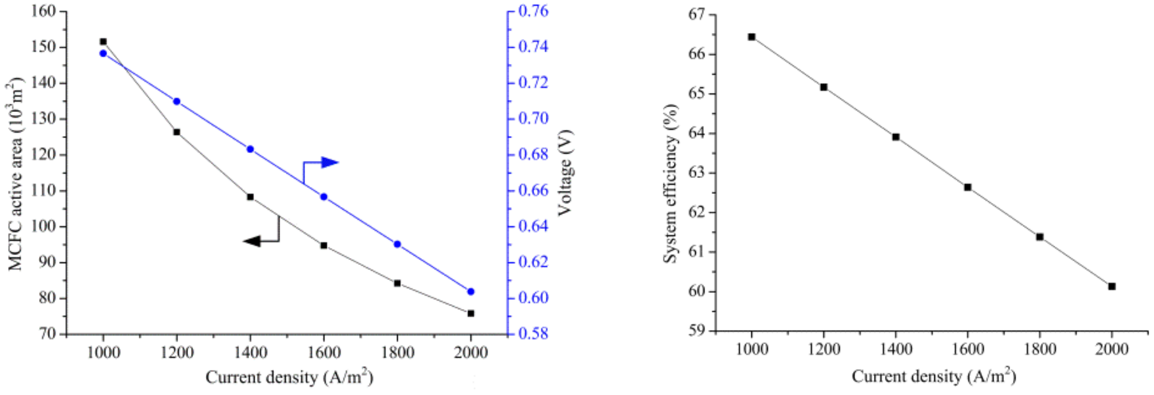
Fig. 5. Exergy destruction distributions of different systems.

As shown in Fig. 5, in all systems the exergy destruction of MCFC is the most significant, mainly because the internal electrochemical reaction of MCFC would cause great irreversible losses. The exergy destruction in the mixer2 is also very big for the reason that the temperature differences of the three gas streams entering into the mixer2 are large when mixing with each other, which would cause great irreversible losses of heat exchange. However, when compared with the benchmark system, the exergy destructions of the mixer2 are much smaller in the new systems with sweep gases, mainly because the low temperature of air1 is preheated to a higher temperature before entering into the mixer2, so the temperature differences of the mixing gases are reduced. The exergy destruction of the heater2 of the new systems is also smaller than that of the benchmark system for the same reason as the mixer2. Through comparisons of both thermal efficiency and exergy destruction distribution, the case b system using CO_2 as sweep gas is finally chosen as the optimum system which is superior to the benchmark system as well as the case a system using CO_2 and H_2O as sweep gas.

5 Sensitivity analysis of the optimum system (case b)

The sensitivity analyses of the key parameters such as the current density, the oxygen partial pressure ratio, the sweep gas recycle ratio, the cathode exhaust recycle ratio and the fuel utilization ratio on the optimum system (case b) performance are investigated.

5.1 Effects of the current density



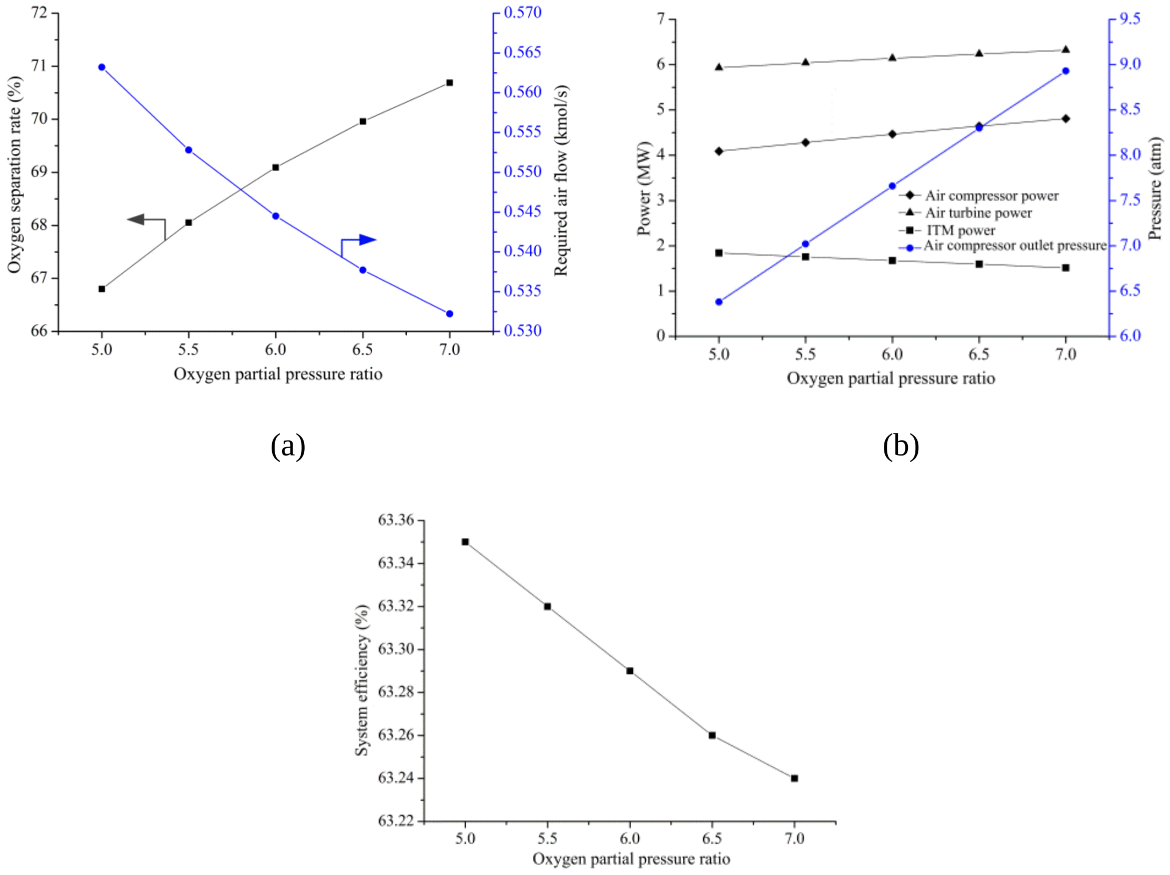
(a) (b)

Fig. 6. Effects of the current density on: a) MCFC active area and voltage, b) System efficiency.

The MCFC current is fixed when keeping the fuel flow and fuel utilization ratio at constant, with the increase of current density, the required MCFC active area is reduced, according to the equation (7) the MCFC voltage will also decrease, as shown in Fig. 6(a). As the current is unchanged, the decrease of voltage will make the MCFC power decrease, which will also lead to the decrease of the system net power, as a result, the system efficiency is decreased with the increase of the current density, as shown in Fig. 6(b).

5.2Effects of the oxygen partial pressure ratio

The oxygen partial pressure ratio, as illustrated in Fig. 3, is defined as the ratio of the oxygen partial pressure in stream 24 to the oxygen partial pressure in stream 25. As shown in Figure 7(a), the change of oxygen partial pressure ratio will influence the oxygen separation rate and the required air flow on ITM feed side. When keeping the oxygen partial pressure on ITM permeate side at constant, with the increase of the oxygen partial pressure ratio, the oxygen separation rate (which is defined as the ratio of the oxygen mass flow in stream 25 to the oxygen mass flow in stream 24 as illustrated in Fig. 3) is improved and the required air mass flow is reduced. In addition, with the increase of the oxygen partial pressure ratio, the required air compressor outlet pressure will increase, which will lead to the increase of the air compressor power as well as the turbine output power, as the increase of the air compressor power is greater than the increase of the turbine output power, the ITM power is decreased slightly, as shown in Fig. 7(b). With the decrease of ITM power, the system efficiency is decreased, as shown in Fig. 7(c).

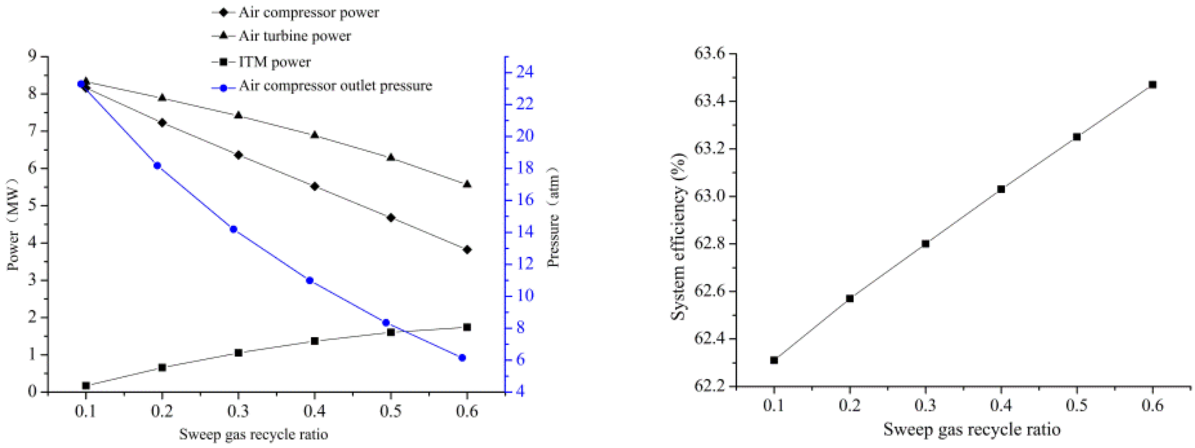


(a) (b)

(c)

Fig. 7. Effects of the oxygen partial pressure ratio on: a) oxygen separation rate and required air flow, b) ITM power and air compressor outlet pressure, c) system efficiency.

5.3 Effects of the sweep gas recycle ratio



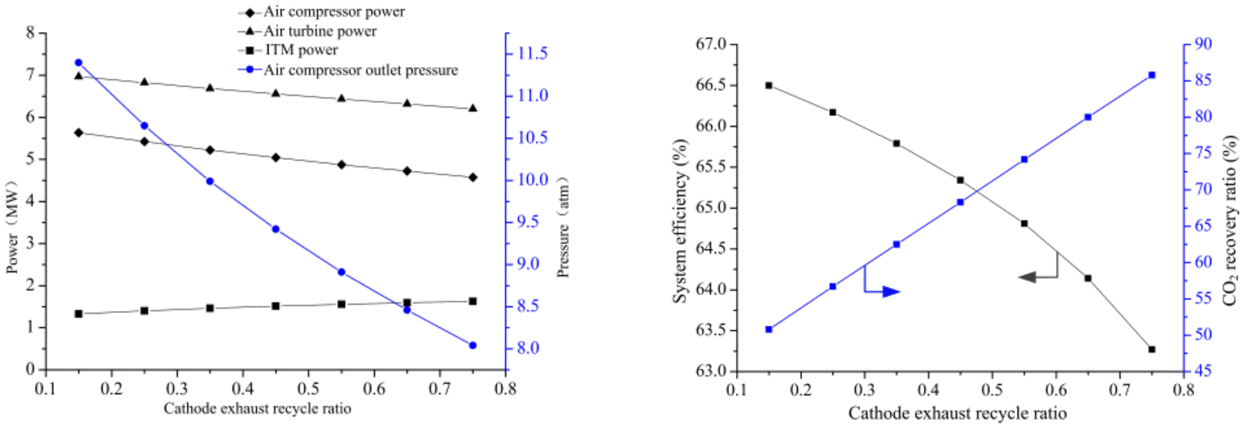
(a) (b)

Fig. 8. Effects of the sweep gas recycle ratio on: a) ITM power and air compressor outlet pressure, b) system efficiency.

The sweep gas recycle ratio, as illustrated in Fig. 3, is defined as the ratio of the mass flow of stream 35 to the mass flow of stream 32. As shown in Fig. 8(a), when keeping the oxygen partial pressure ratio at constant (which means that the oxygen separation rate is unchanged), with the increase of the sweep gas recycle ratio, the oxygen partial pressure on ITM permeate side is decreased and the required air compressor outlet pressure is decreased, which will lead to the decrease of the air compressor power as well as the turbine output power, as the decrease of the air compressor power is greater than the decrease of the turbine output power, the ITM power is increased. With the increase of ITM power, the system efficiency is increased, as shown in Fig. 8(b).

5.4 Effects of the cathode exhaust recycle ratio

The cathode exhaust recycle ratio, as illustrated in Fig. 3, is defined as the ratio of the mass flow of stream 14 to the mass flow of stream 13. As shown in Fig. 9(a), when keeping the oxygen partial pressure ratio at constant, with the increase of the cathode exhaust recycle ratio. the oxygen partial pressure on ITM permeate side is decreased and the required air compressor outlet pressure is decreased, which will lead to the decrease of the air compressor power as well as the turbine output power, as the decrease of the air compressor power is greater than the decrease of the turbine output power, the ITM power is increased slightly. In addition, with the increase of the cathode exhaust recycle ratio, the system CO_2 recovery ratio (which is defined as the ratio of carbon content in stream 34 to carbon content in fuel, as illustrated in Fig. 4) is increased, as a result the CO_2 compressor power is increased, as the increase of the CO_2 compressor power is greater than the increase of the ITM power, the system efficiency is decreased, as shown in Fig. 9(b).

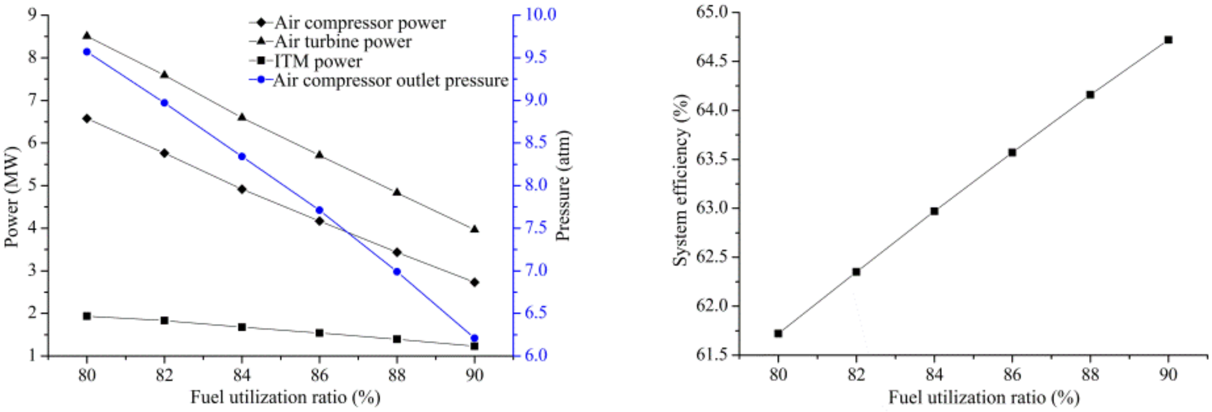


(a) (b)

Fig. 9. Effects of the cathode exhaust recycle ratio on: a) ITM power and air compressor outlet pressure, b) system efficiency and CO_2 recovery ratio.

5.5 Effects of the fuel utilization ratio

When keeping the fuel flow at constant, with the increase of the fuel utilization ratio, both the unreacted fuel and the required oxygen for the afterburner combustion are reduced, as a result the oxygen partial pressure is decreased. When keeping the oxygen partial pressure ratio at constant, both the required air flow and the air compressor outlet pressure are decreased, which will lead to the decrease of the air compressor power as well as the turbine output power, as the decrease of the turbine output power is greater than the decrease of the air compressor power, the ITM power is decreased, as shown in Fig. 10(a). However, with the increase of the fuel utilization ratio, the MCFC current is increased which will contribute to the increase of the MCFC power, as the increase of the MCFC power is greater than the decrease of the ITM power, the system efficiency is increased, as shown in Fig. 10(b).



(a) (b)

Fig. 10. Effects of the fuel utilization ratio on: a) ITM power and air compressor outlet pressure, b) System efficiency.

6 Conclusions

In this paper two ITM-integrated MCFC hybrid systems with CO_2 recovery using sweep gas are proposed and compared with the benchmark system without sweep gas. Results show that using sweep gas could reduce the oxygen partial pressure at the ITM permeate side, thus reducing the required air compressor outlet pressure when keeping the oxygen partial pressure ratio constant. Also using the different sweep gases may have different effects on system performance, under the condition of keeping the air compressor outlet pressure at 8.04atm, the efficiency of case a system using the mixture of CO_2 and H_2O as sweep gas and case b system using CO_2 as sweep gas are 0.71 and 1.25 percent points higher than that of the benchmark system, respectively. The exergy analyses show that among all systems proposed in this paper the case b system has the minimum total exergy destruction and is superior to other systems in thermal performance. Through the sensitive analysis of the optimum case b system, results show that the increase of both the sweep gas recycle ratio and fuel utilization ratio are beneficial to improving the system efficiency, however increasing the MCFC current density, the oxygen partial pressure ratio and the cathode exhaust recycle ratio would all lead to the decrease of the overall system efficiency, besides, the system CO_2 recovery ratio would increase with the increase of the cathode exhaust recycle ratio.

Acknowledgments

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