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Study of gas-steam combined cycle power plants integrated with MCFC for carbon dioxide capture

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Abstract

In the field of fossil-fuel based technologies, natural gas combined cycle (NGCC) power plants are currently the best option for electricity generation, having an efficiency close to 60%. However, they produce significant CO_2 emissions, amounting to around 0.4 tonne/MWh for new installations. Among the carbon capture and sequestration (CCS) technologies, the process based on chemical absorption is a well-established technology, but markedly reduces the NGCC performances. On the other side, the integration of molten carbonate fuel cells (MCFCs) is recognized as an attractive option to overcome the main drawbacks of traditional CCS technologies. If the cathode side is fed by NGCC exhaust gases, the MCFC operates as a CO_2 concentrator, beside providing an additional generating capacity.

In this paper the integration of MCFC into a two pressure levels combined cycle is investigated through an energy analysis. To improve the efficiency of MCFC and its integration within the NGCC, plant configurations based on two different gas recirculation options are analyzed. The first is a traditional recirculation of exhaust gases at the compressor inlet; the second, mainly involving the MCFC stack, is based on recirculating a fraction of anode exhaust gases at the cathode inlet. Effects of MCFC operating conditions on energy and environmental performances of the integrated system are evaluated.

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Keywords: MCFC, NGCC, CCS.

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1. Introduction

The installed capacity of natural gas combined-cycle (NGCC) has risen rapidly in the last two decades, because of their relatively low-cost, high efficiency and operating flexibility. Although natural gas is a low-carbon fossil fuel,

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the specific CO_2 emission of NGCCs are not negligible, varying in the range 300-500 kg/MWh [1]. According to the projections of the International Energy Agency (EIA), the efficiency of NGCC is expected to increase in the near future, passing from the current value of 60% to about 64% in 2020 [2]. Hence, the use of carbon capture and sequestration technologies is considered to be essential for a deeper cut of CO_2 emissions.

Nowadays chemical absorption is recognized as a well-established process for the CO_2 post-combustion capture, allowing to remove more than 90% of CO_2 from fossil-fuel power plants. Their main drawbacks are undoubtedly represented by the high energy requirement for the adsorbent regeneration and the low CO_2 concentration of flue gas to be treated. In fact, these energy penalties lower the net plant efficiency, up to about 8-10 percentage points in the case of natural gas combined cycles [3].

Recently the research efforts have focused the attention on carbon capture based on Molten Carbonate Fuel Cells (MCFCs). Indeed, when the cathode side of the MCFC is fed by flue gas of NGCC, CO_2 is transported via carbonate ions to the anode side and finally concentrated in the anode exhaust. With respect to conventional CCS technologies, MCFC is regarded as an "active" device, as it allows to improve the power plant performances while it acts as a CO_2 concentrator [4].

Several studies have examined the potential advantages of capturing CO_2 from fossil fuel power plants by means of MCFCs. Some of them examined the performances of the MCFC, evaluating the effects of temperature, pressure and electrolyte type [5], as well as CO_2 concentration at cathode inlet [6]. The investigations results highlighted that the cell potential is strongly affected by operating temperature below 625°C, while it retains a steady asymptotic profile above this limit; moreover CO_2 concentration should not fall below 6%, in order to prevent an excessive voltage degradation. The performances of MCFC as a CO_2 separator have also been investigated in [7], comparing configurations based on internal and external reforming of natural gas.

Other studies have focused on the performance evaluation of fossil-fuel power plants integrated with MCFCs. As regard to the application on NGCCs, simulations results revealed an efficiency penalty of few percentage points, together with net electrical power increase of 20-30% [4,8]. The behavior of a MCFC separating the CO_2 emissions from CHP plant has been analyzed in [9], evaluating the effects of anode recirculation on fuel consumption and cathode recirculation on CO_2 removal factor.

This paper aims to analyze the energy performances of a two pressure level NGCC, integrated with a molten carbonate fuel cell. Effects of different modifications to the baseline plant layout are analyzed, with the aim to increase CO_2 concentration at the cathode inlet. Two different types of gas recirculation are investigated: in the first, a fraction of exhaust gas at the HRSG outlet is recirculated and mixed to the air at compressor inlet (exhaust gas recirculation); in the second, mainly involving the MCFC stack, a fraction of the anode exhaust gas is directly recirculated at the cathode inlet (anode exhaust gas recirculation).

The thermodynamic analysis of the integrated system is performed using the software Gate-Cycle [10] for the NGCC and the software CycleTempo [11] for the MCFC stack, properly integrated into Excel environment using macros developed in Visual Basic. The simulation results allow to highlight effects of the most significant operating parameters, such as the fraction of exhaust gas or anode gas recirculated and the stack operating voltage, on power plant energy performances.

Nomenclature

Symbols

E	Cell voltage, V
M	Mass flow rate, kg/s
p	Pressure, bar
P	Power, MW
Q	Temperature, °C
η	Efficiency, %

Subscripts

a	Anode
c	Cathode

EXH	Exhaust
LP	Low pressure
HP	High pressure
rev	Reversible
S	Steam
SH	Superheated
Acronyms	
CC	Combined cycle
CCS	Carbon Capture & Sequestration
CP	Compressor
GT	Gas turbine
MCFC	Molten Carbonate Fuel Cell
NGCC	Natural Gas Combined Cycle
Configurations studied	
Case 0	NGCC baseline power plant (w/o MCFC integration and w/o CO_2 capture)
Case 1	NGCC integrated with MCFC (w/o CO_2 capture)
Case 2	NGCC integrated with MCFC using exhaust gas recirculation (w/o CO_2 capture)
Case 3	NGCC integrated with MCFC using anode exhaust gas recirculation (w/o CO_2 capture)
Case 4	NGCC integrated with MCFC using anode gas recirculation (as Case 3) and CO_2 capture system

2. Post-combustion carbon capture with molten carbonate fuel cells

A molten carbonate fuel cell is an electrochemical device that directly converts into electrical energy the chemical energy of a fuel, that is usually hydrogen resulting from a steam methane reforming process:

$$CH_4 + 2H_2O \longrightarrow 4H_2 + CO_2 \tag{1}$$

The operating principle of a MCFC is shown in Fig. 1. At the anode side, H_2 combines with carbonate ions (CO_3^{2-}) generating steam, carbon dioxide and electrons:

$$H_2 + CO_3^{2-} \longrightarrow H_2O + CO_2 + 2e^- \tag{2}$$

At the cathode side, the reaction between the oxygen and the carbon dioxide produces carbonate ions (CO_3^{2-})

$$\frac{1}{2}O_2 + CO_2 + 2e^- \longrightarrow CO_3^{2-} \tag{3}$$

Hence, the overall reaction

$$H_2 + \frac{1}{2}O_2 + CO_2 \longrightarrow H_2O + CO_2 \tag{4}$$

enables the steam production, together with the transfer of CO_2 from the cathode to the anode. This is obtained through the migration of carbonate ions (CO_3^{2-}) across the carbonate salts at molten state and the electrons flow through the external circuit, generating DC electricity.

The anodic exhaust includes unreacted H_2, steam and concentrated CO_2, resulting from the cathode transfer and from the reforming of natural gas; after an oxy-combustion, the CO_2-rich flow is sent to the drying and compression system, to condensate the steam and capture the carbon dioxide.

Thus, if the cathode side is fed by flue gas from a fossil-fuel power plant, the MCFC acts as an active device able to concentrate CO_2 and increase the plant generating capacity, with limited layout modifications.

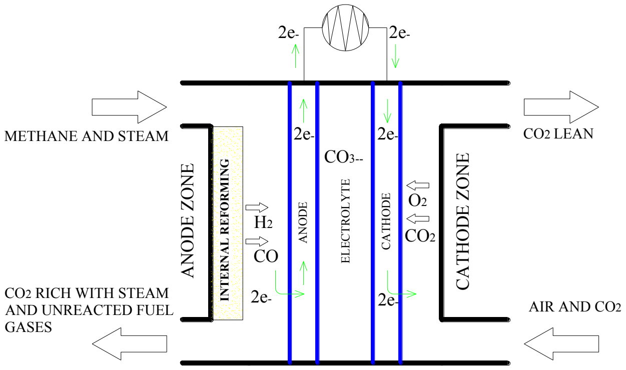

Fig. 1. Operating principle of a MCFC with internal reforming

3. Operating conditions of MCFC

The maximum theoretical cell voltage or reversible potential is given by Nernst equation

$$E_{rev} = E^0 + \frac{RT}{2F}\ln\frac{p_{H_2}p_{O_2}^{0.5}}{p_{H_2O}} + \frac{RT}{2F}\ln\frac{p_{CO_2,c}}{p_{CO_2,a}} \tag{5}$$

that depends on operating pressure and temperature, as well as on gas composition and reactants utilization [12].

Hence, the MCFC potential and its efficiency increase at increasing of carbon dioxide concentration at cathode side with respect to anode side, while reduce at increasing of reactants utilization. The actual cell potential is lower than the ideal value because of irreversible losses, mainly due to activation, ohmic and gas concentration polarization phenomena.

Table 1. Operating conditions of MCFC in the base case

Parameter	Value
Operating pressure, bar	1.05
Operating temperature, °C	625
Steam-to-methane molar ratio, %	3
H_2 Utilization factor, %	78
O_2 Utilization factor, %	11
Conversion efficiency DC/AC, %mol	94
Current density, A/m^2	1500
Cell Voltage, V	0.701
Cell area, m^2	10118
Cell resistance, Ω m^2	9.53E-05
Power electrical AC, MW	10
Efficiency, %	49.4

The MCFC operating conditions are defined by setting the current density and the polarization curve, that describes the relationship between the cell potential and the current density, for fixed values of temperature, pressure and reactant stoichiometry [12]. A high current density allows a high transfer rate of the carbonate ions and, consequently, a more effective CO_2 separation. However, the increase of current density reduces the fuel cell potential and the efficiency due to higher polarization losses. On the other hand, for a fixed rated power, a low current density improves the efficiency, but requires a larger cell area. In this study, the MCFC stack operates at atmospheric pressure and 625°C, with a current density of 1500 A/m^2, offering a good compromise between costs and efficiency.

In order to evaluate the effects of CO_2 concentration at the cathode inlet on the cell potential, a parametric study has been performed using the software CycleTempo in off-design mode [11]. Table 1 summarizes the MCFC main operating parameters, while Table 2 reports thermodynamic properties and gas composition of MCFC streams. Results refer to the base case with a CO_2 concentration of 3.53% at the cathode inlet, typical of NGCC flue gases without exhaust gas recirculation (EGR), and the steam-to-methane molar ratio of 3 at the anode inlet, to avoid the reduction of fuel cell efficiency due to the formation and deposition of carbon particles on anode active surface [12]. In the parametric study, the CO_2 concentration at cathode inlet has been varied in a range compatible with various NGCC operating conditions (with EGR), assuming a CO_2 utilization factor of 85% in all simulations. This requires values of the O_2 utilization factor lower than 20%, due to the high O_2 content in the flue gas with respect to the CO_2 content.

Table 2. Main properties of MCFC streams in the base case

Parameter	Anode inlet	Cathode inlet	Anode outlet	Cathode outlet
Mass flow rate, kg/s	1.77	78.00	6.49	73.28
Pressure, bar	1.05	1.05	1.02	1.02
Temperature, °C	625	625	664	664
Molar fraction, %				
CH_4	25.0	-	-	-
H_2O	75.0	7.96	47.84	8.32
H_2	-	-	6.98	-
O_2	-	13.02	-	12.11
N_2	-	74.59	-	77.94
CO_2	-	3.53	42.52	0.70
CO	-	-	2.65	-
Ar	-	0.90	-	0.94

Figure 2 depicts the effects of CO_2 concentration on MCFC voltage and performances. As shown in Fig. 2a, the MCFC voltage increases with the CO_2 concentration, reaching about 0.74 V at CO_2 molar fraction of 11% .

Interpolating data resulted from the parametric analysis, a correlation between the cell voltage and CO_2 concentration at cathode inlet has been derived. This relationship allows to properly evaluate the MCFC performances varying the CO_2 concentration at the cathode inlet. Increasing the CO_2 molar fraction from 3 to 10%, as shown in Fig. 2b, the MCFC rated power rises from 10 MW to about 30 MW, while the MCFC efficiency gains 4 percentage points.

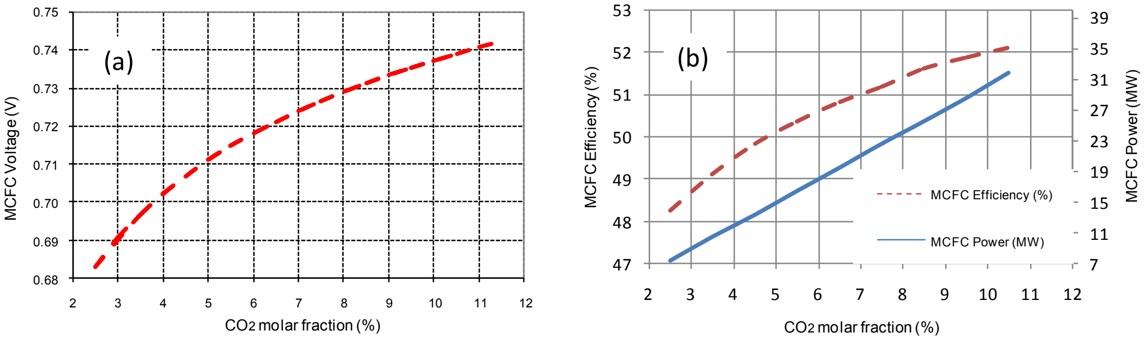

Fig. 2. Effect of CO_2 concentration on MCFC voltage (a) and energy performances (b)

4. Energy analysis of a combined cycle power plant with MCFC as post-combustion capture system

In this paper the capabilities of integrating a MCFC in a combined cycle power plant have been analyzed, by comparing the energy performances of different system configurations. The reference combined cycle power plant is based on a heavy-duty gas turbine (ABB GT10), with a rated capacity of 23.8 MW and a LHV efficiency of 33.3%. The steam cycle comprises high and low pressure steam turbines, a condensing system and a two-pressure level heat recovery steam generator (HRSG). In the base configuration without MCFC, the whole flow exhausted by the gas turbine, having a low CO_2 molar concentration (3.3%), is sent to the HRSG. It allows to produce 9.44 kg/s of HP superheated steam that evolves in the bottoming steam cycle. Thus, the natural gas combined cycle (NGCC) produces a rated capacity of 35.8 MW, with a LHV efficiency of 50.2% and specific CO_2 emissions of 397 kg/MWh. Table 3 summarizes the main performance parameters of the baseline NGCC without MCFC.

Table 3. Performance parameters of the baseline NGCC

Gas turbine	
Air mass flow rate, kg/s	76.5
Methane mass flow rate, kg/s	1.43
Combustor exit temperature, °C	1219
Turbine exit pressure, kPa	105.1
Turbine exit temperature, °C	541
Turbine exit CO_2 molar fraction, %	3.28
GT net efficiency, %	33.3
GT power output, MW	23.8
Gas-steam combined cycle	
LP SH temperature (S40), °C	265
HP SH temperature (S34), °C	501
LP SH pressure, bar	3.5
HP SH pressure, bar	70.7
LP SH mass flow rate, kg/s	2.48
HP SH mass flow rate, kg/s	9.44
Exhaust gas temperature , °C	90
CC net efficiency, %	50.2
CC power output, MW	35.8
Specific CO_2 emissions, kg/MWh_{el}	397

Figure 3 shows the layout of the NGCC power plant integrated with the MCFC. This layout allows to simulate all different configurations analyzed in this study. They range from the baseline NGCC configuration, where all added sections (MCFC, gas recirculation options, CO_2 capture system) are by-passed, to the configuration with MCFC and CO_2 capture. In the study, five configurations have been evaluated; they are indicated as:

- ✓ **Case 0**: NGCC baseline power plant (w/o MCFC integration and w/o CO_2 capture);
- ✓ **Case 1**: NGCC integrated with MCFC (w/o CO_2 capture);
- ✓ **Case 2**: NGCC integrated with MCFC using exhaust gas recirculation (w/o CO_2 capture);
- ✓ **Case 3**: NGCC integrated with MCFC using anode exhaust gas recirculation (w/o CO_2 capture);
- ✓ **Case 4**: NGCC integrated with MCFC using anode exhaust gas recirculation (as Case 3) and CO_2 capture.

As shown in the overall plant layout (Fig. 3), in **Case 2** a fraction of the exhaust gas is recirculated at the compressor inlet (green line), in **Case 3** a fraction of the anode exhaust gas is recirculated at the MCFC cathode inlet (red line). Both the recirculation options aim to increase the CO_2 concentration at the cathode inlet, to improve performance of the MCFC and enhance effectiveness of the CO_2 capture.

The thermodynamic behaviour of various configurations is discussed in the following subparagraphs, while their operating conditions and main performance parameters are summarized in Table 4 and 5.

4.1. NGCC power plant integrated with MCFC (Case 1)

As shown in Fig. 3, the MCFC stack is interposed between the gas turbine and the HRSG. The flue gas at the turbine outlet (S25) is entirely sent to the cathode inlet (XS13), after being heated to the operating temperature of fuel cell stack (625°C) by the post-combustor (XDB1). Hence, the cathode exhaust gases (S17) pass across the two pressure HRSG and are cooled up to 90°C (EXH). The anode side of the MCFC is fed by the fuel flow (XS17) and the steam needed for the internal reforming of the natural gas. Input parameters used for simulating the MCFC are those reported in Table 1. As shown in Table 5, the efficiency of the NGCC power plant integrated with the MCFC (**Case 1**) is slightly lower (-1.4%) than the baseline NGCC (**Case 0**), while the rated power increases of about 39%, reaching 49.8 MW; the gain of rated capacity is attributable to the additional MCFC stack for 75% and to the steam cycle for the remainder 25%, due to the higher gas temperature at the HRSG inlet. Moreover the specific CO_2 emissions without capture decreases by approximately 4%, passing from 397 to 381 kg/MWh.

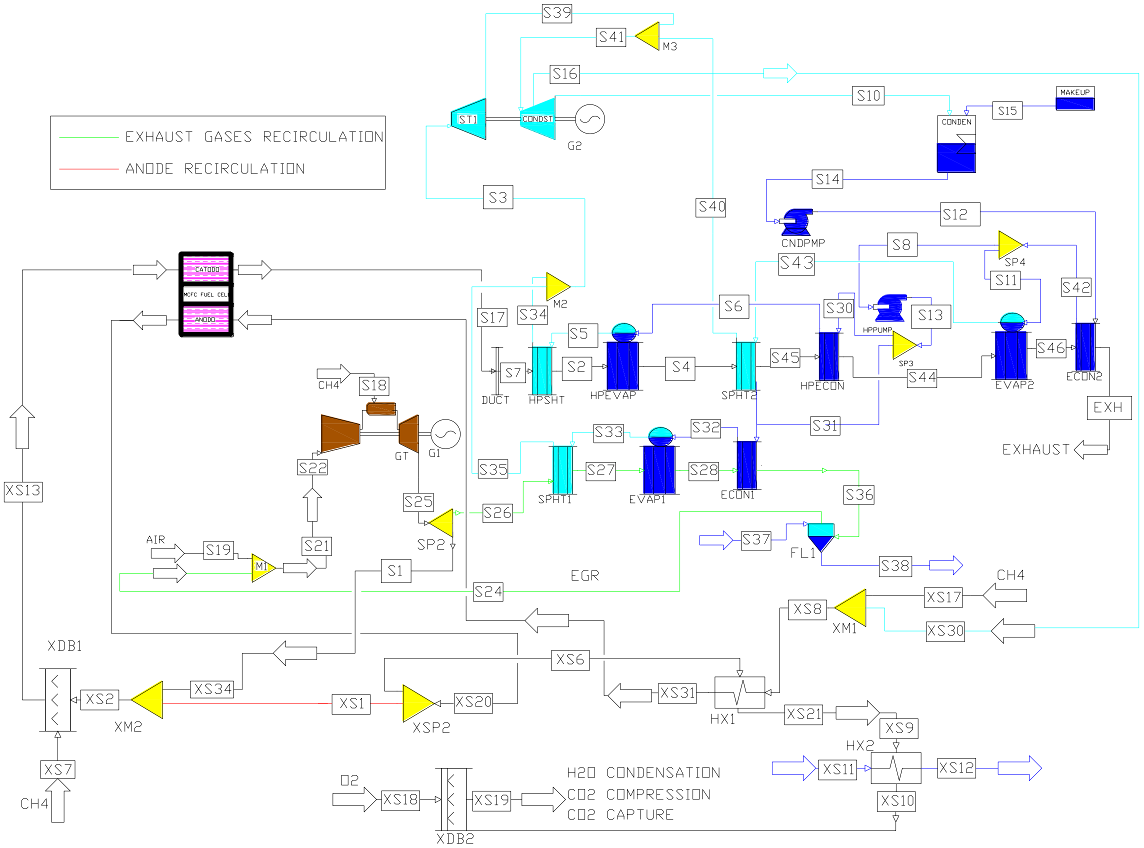

Fig. 3. Layout of the dual pressure natural gas combined cycle integrated with MCFC

4.2. NGCC power plant integrated with MCFC using exhaust gas recirculation (Case 2)

With the aim to increase the CO_2 concentration at the cathode inlet, the recirculation of a fraction of exhaust gas at the compressor inlet has been investigated. In this case, about 60% of exhaust gas at turbine outlet is heated up to 625°C by the duct burner and the remainder 40% is cooled up to 35°C, passing through a one pressure HRSG and a flash tank that separates the excess condensed water, and then mixed to the air at compressor inlet. Hence, the CO_2 concentration at cathode inlet rises to 5.71%, thus increasing the MCFC efficiency up to 50.5% . The thermal power of the cathode exhaust gases, depleted of CO_2, is recovered by the two pressure level HRSG, whereas anode exhaust (XS20) are cooled in HX1 at a temperature of 457°C, by-passing all equipment required for the CO_2 capture.

The simulation results show that, despite the greater complexity of the plant layout, the rated capacity reduces to 45.7 MW, as the exhaust gas recirculation slightly reduces the power output of the gas turbine (-5%), and the reduction of the cathode exhaust flow rate negatively affects the power output of the steam cycle (-14%) and the MCFC stack (-5%). As a results, despite the higher efficiency of MCFC (+1.2 percentage points), the efficiency of the integrated system reduces to 49.1% and the specific CO_2 emissions increases to 383 kg/MWh. Moreover, a further thermal power of 3.8 MW at 457°C could be recovered from the anode exhaust gases.

Table 4. MCFC operating parameters for the integrated system configurations

	Case 1	**Case 2**	**Case 3**	**Case 4**
MCFC	yes	yes	yes	yes
Exhaust gas recirculation	-	yes	-	-
Anode exhaust gas recirculation	-	-	yes	yes
CO_2 capture system	-	-	-	yes
MCFC input parameters				
CO_2 cathode input,%mol	3.63	5.71	6.6	6.6
O_2 cathode input,%mol	12.79	8.73	12.1	12.1
H_2O cathode input,%mol	8.18	9.75	11.3	11.3
N_2 cathode input,%mol	74.51	74.91	69.2	69.2
Ar cathode input,%mol	0.89	0.90	0.8	0.8
Cathode input flow rate, kg/s	78.0	45.9	84.4	84.4
Utilization O_2 factor, %	11.65	26.8	24.3	24.3
MCFC voltage, V	0.698	0.716	0.722	0.722
MCFC output parameters				
MCFC net power, MW	10.4	9.84	22.8	22.5
MCFC efficiency, %	49.3	50.5	50.9	50.9
CH_4 flow rate XDB1, (XS7) kg/s	0.16	0.08	0.03	0.03
Power consumption for CO_2 compression, MW	-	-	-	1.83
Power consumption for O_2 production, MW	-	-	-	0.45
O_2 flow rate, kg/s	-	-	-	0.43

Table 5. Main performance parameters of the integrated system configurations

	Case 0	**Case 1**	**Case 2**	**Case 3**	**Case 4**
MCFC	-	yes	yes	yes	yes
Exhaust gas recirculation	-	-	yes	-	-
Anode exhaust gas recirculation	-	-	-	yes	yes
CO_2 capture system	-	-	-	-	yes
Mass flow rate at CP inlet (S22), kg/s	76.5	76.5	73.7	76.5	76.5
Molar CO_2 fraction at GT exit (S25) %	3.28	3.28	5.42	3.28	3.28
Exhaust GT temperature (EXH), °C	90	90	90	90	90
Net gas turbine power, MW	23.8	23.8	22.6	23.8	23.8
Net steam cycle power, MW	12.3	15.9	13.6	17.0	17.0
Net MCFC power, MW	-	10.4	9.84	22.8	22.5
Net integrated plant power, MW	35.8	49.8	45.7	63.2	60.6
Net integrated plant efficiency, %	50.2	49.5	49.1	53.8	51.8
Specific CO_2 emissions, kg/MWh_{el}	397	381	383	350	58

4.3. NGCC power plant integrated with MCFC using anode exhaust gas recirculation (Case 3)

In this configuration, about 45% of anode exhaust gases (XS20) are recycled to cathode input (XS13) and mixed to the flue gas at the turbine exit. As a result, the CO_2 concentration at cathode inlet rises to 6.6%, thus increasing the MCFC efficiency to 50.9% and its rated power to about 23 MW. Moreover, the anodic recirculation allows to partially exploit the energy content of the anode exhaust, thus increasing the temperature at cathode inlet. Hence, the fuel flow supplied to the duct burner is considerably reduced, passing from 0.16 to 0.03 kg/s. At the same time, the thermal power of the anode exhaust gases reduces its temperature level (2.6 MW at 271°C).

Thus, with the same gas turbine rated power (23.8 MW), the steam cycle capacity increases of about 7% with respect to the NGCC configuration integrated with MCFC (**Case 1**), due to the higher cathodic exhaust flow rate; as a result, the rated power of the integrated system reaches 63.2 MW (+27%), the additional MCFC accounting for about 92% and the steam cycle for the remainder 8%. Moreover, due to a net efficiency gain of 4.3 percentage points (53.8%), the specific CO_2 emission without carbon capture reduces from 397 to 350 kg/MWh.

4.4. NGCC power plant integrated with MCFC using anode exhaust gas recirculation and CO_2 capture (Case 4)

The energy performances of the system configuration with anode gas recirculation (**Case 3**) has been further investigated, considering the CO_2 capture and storage.

Anode exhaust gases, containing about 42.5% CO_2, 7.0% H_2 and 2.6% CO, are cooled in the heat exchangers HX1 and HX2 and then sent to duct-burner XDB2. Downstream the oxy-combustion of unreacted fuel gases, the steam is separated via condensation from the anode exhaust, composed by 45.2% CO_2 and 54.8% H_2O. The resulting CO_2 flow is sent to a 5-stages compression system with intermediate refrigeration and stored at 110 bar.

Taking into account for energy requirements of CO_2 compression system, the rated power reduces to 60.6 MW, with a net efficiency of 51.8%, that is 1.6 percentage points higher than the baseline combined cycle. Moreover, the specific CO_2 emission drastically reduces, passing from 397 to 58 kg/MWh.

Finally, it should be noted that the efficiency of the integrated system could be further increased by partly recovering the significant thermal power of anode exhaust gases, 9.2 MW from a temperature of 665°C.

Conclusions

The aim of this study was to examine the capabilities of MCFC to act as a CO_2 separator in natural gas combined cycles. The reference NGCC is based on a dual pressure HRSG, with a rated power of 35.8 MW, a LHV efficiency of 50.2%, and CO_2 specific emissions of 397 kg/MWh.

The simulation results have shown that the addition of a MCFC fed by gas turbine exhaust gases markedly increases the rated plant capacity, that reaches about 49.8 MW (+39%); although the efficiency remains almost unchanged, the specific CO_2 emissions without capture system reduces from 397 to 381 kg/MWh.

In order to increase the CO_2 concentration at the cathode inlet and then the MCFC performances, different configurations based on two gas recirculation options have been investigated.

In the first configuration (**Case 2**), 40% of the exhaust gas at the turbine outlet is cooled up to 35°C and recirculated at the compressor inlet. As a result, the CO_2 concentration at the cathode inlet increases to 5.7%, enabling an efficiency gain of MCFC of about 1.2 percentage points (50.5%). However, with respect to NGCC integrated with MCFC, the exhaust gas recirculation adversely affects the rated capacity of the gas (-5%) and steam (-14%) cycles, as well as the size of MCFC (-5%). Hence the net plant rated power and efficiency reduce to 45.7 MW and 49.1% respectively, while the specific CO_2 emissions increases to 383 kg/MWh.

In the second configuration (**Case 3**), 45% of the anode exhaust gas is recirculated at the cathode inlet, thus increasing the MCFC efficiency up to 50.9%. Hence, due to the additional capacity of the MCFC and to the increase of steam cycle power, the rated capacity of the integrated system reaches 63.2 MW, while the efficiency gains 3.6% percentage points with respect to the baseline NGCC. Hence, the specific CO_2 emissions without carbon capture reduces from 397 to 350 kg/MWh.

The same configuration, if integrated with a carbon capture system (**Case 4**), has a slightly lower rated power (60.6 MW), due to the energy requirement for CO_2 compression and pays a net efficiency decrease of 2 percentage points (51.8%). However, the specific CO_2 emissions reduces to about 58 kg/MWh, with a carbon capture ratio of about 85%.

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