Reducing CO_2 Emissions from a Gas Turbine Power Plant by using a Molten Carbonate Fuel Cell

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***Abstract*—A Molten Carbonate Fuel Cell (MCFC) is shown to reduce CO_2 emissions from a Gas Turbine Power Plant (GTPP). The MCFC is placed in the flue gas stream of the gas turbine. The main advantages of this solution are: higher total electricity generated by a hybrid system and reduced CO_2 emissions with power generation efficiency remained the same. The model of the MCFC is given and described. The results obtained show that use of an MCFC could reduce CO_2 emissions by 73%, which gives a relative CO_2 emission rate of 135 kg_{CO_2} per MWh.**

***Index Terms*—fuel cells, molten carbonate fuel cell, CO_2 emissions.**

I. INTRODUCTION

THE European Union has placed limits on CO_2 emissions by Member States as part of its Emission Trading Scheme. This impacts fossil fuel power plants to a significant degree as their emissions are governed by the number of emission allowances they receive from the Member State allocation. Excess CO_2 emissions have to be covered by purchasing extra allowances, which is in effect a penalty (€15/Mg at present). In contrast, undershooting emission limits enables the emitter to sell CO_2 allowances. The selling price of a traded allowance is estimated at €20–30/Mg_{CO_2}.

There is a variety of methods available to remove CO_2 from a fossil fuel power plant system [1], [2]. The idea of adopting a molten carbonate fuel cell to reduce CO_2 emissions was developed by Campanari [3]. In this paper it is shown that an estimated reduction of 77% in CO_2 emissions can be achieved in a steam turbine power plant. Simalar investigations are performed recently by Sanches et al. [4].

Fuel cells generate electricity through electrochemical processes. There are many types of fuel cells, two of them – the Molten Carbonate Fuel Cell (MCFC) and the Solid Oxide Fuel Cell (SOFC) – are high temperature fuel cells [5]. They work at temperatures ranging from 600 to 1000°C. A combination of high temperature fuel cell with gas turbine gives a hybrid system with potentialy ultra-high efficiency [6].

Amorelli et al. [7] described an experimental investigation into the use of molten carbonate fuel cells to capture CO_2 from gas turbine exhaust gases. They obtained an emission reduction of 50%. Those experiments were performed using a singular cell.

Lusardi et al. [8] investigated the application of a fuel cell system for CO_2 sequestration from thermal plant exhaust. They found that, even without CO_2 separation, the relative emission of carbon dioxide could be reduced to below the Kyoto Protocol limit. If a separator is used, emissions could be reduced by 68%.

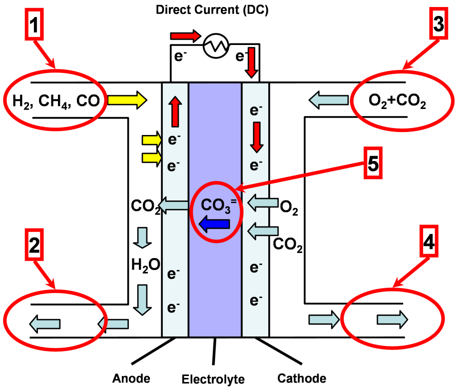

Fig. 1. Working principles of MCFC; 1) Fuel input, 2) Mixture of CO_2, H_2 and H_2O, 3) Oxidant input, 4) Exhaust, 5) Ions of CO_3^{2-}

The use of an MCFC as a carbon dioxide concentrator was investigated by Sugiura et al. [9]. In this work the experimental results of CO_2 sequestration by use of an MCFC are given. One key conclusion from this work is that the CO_2 removal rate can be obtained by making calculations using electrochemical theory.

Novel methods whereby carbonates were used as an electrochemical pump in carbon dioxide separation from gases were described by Granite et al. [10].

Based on the above review of literature, a reduction of at least 50% in CO_2 emissions could be expected.

Hydrogen, natural gas, methanol or biogas may be used as fuels for MCFCs. On the cathode side, a mixture of oxygen and carbon dioxide is required.

An MCFC can work as a carbon dioxide separator/concentrator because the CO_2 is transported from the cathode side to the anode side through molten electrolyte.

The combination of GTPP with MCFC gives a hybrid system (HS) with increased efficiency and decreased carbon dioxide emission. The exhaust flue gas of gas turbine power plant consists mainly of nitrogen, oxygen, steam and carbon dioxide. This mixture can be used as oxidant in the MCFC (cathode feeding). The temperature of the exhaust gas and electric efficiency of GTPP are about 550°C and 35%, respectively. On the contrary fuel cells can achieve higher electric efficiency of 50–60%.

Negative ions are transferred through the molten electrolyte. Each ion is composed of one molecule of carbon dioxide, one atom of oxygen and two electrons. This means

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TABLE I
EXHAUST GAS COMPOSITION

Component	Mass fraction, %	Mole fraction, %
CO_2	5.2	3.4
H2O	4.1	6.6
O_2	15.3	13.6
N_2	74.0	75.4
Ar	1.4	1.0
CO_2/O_2	0.34	0.25

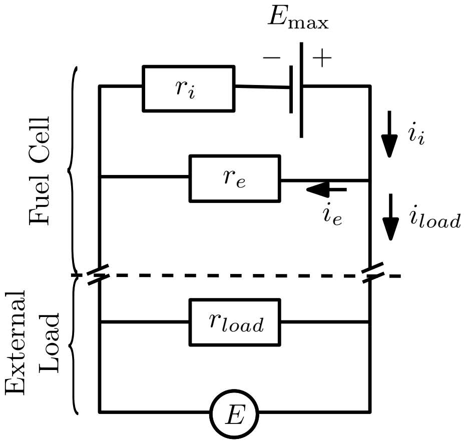

Fig. 2. Equivalent electric circuit of the cell [**?**]

that an adequate ratio of carbon dioxide to oxygen is 2.75 (mass based) or 2.0 (mole based).

The typical gas turbine flue gas composition is shown in Table~I. The ratio of CO_2 to oxygen is hence 0.25 (mole based) and 0.34 (mass based). This means that flue gas contains an insufficient quantity of oxygen to trap all CO_2.

II. THEORY

Mathematical modelling is now the basic method for analyzing systems incorporating fuel cells. A zero-dimensional approach is used for the modelling of system elements.

A. *MCFC*

The presented results are based on calculations made using an appropriate mathematical model. The governing equations of this model are presented in this section.

An MCFC consists of series and parallel connected cells. Series connected cells make a stack. The electricity generated by an MCFC is given by the following equation:

$$P_{MCFC} = \sum_{j=1}^{m} \left(I_j \cdot \sum_{i=1}^{n} E_{MCFC,i,j} \right) \tag{1}$$

where: i – cell number; n – number of cells; j – stack number; m – number of stacks; I – stack current; E_{MCFC} – singular cell voltage.

The stack current (I) is defined by the following equation:

$$I_3 = 2 \cdot F \cdot \dot{n}_{H_2,in} \cdot \eta_f \tag{2}$$

where: F – Faraday's constant, C/mol; $\dot{n}_{H2,in}$ – hydrogen inlet molar flow, kmol/s; E – cell voltage.

The equivalent electric circuit of a singular cell is shown in Fig. 2.

Two types of resistances are present in fuel cells: ionic resistance r_1 and electric resistance r_2. Resistance r_3 is the external load resistance of the fuel cell and is varied by the operator of the MCFC.

Voltage generated by a singular cell is given by the following equation [11]:

$$E_{MCFC} = \frac{E_{\max} - \eta_f \cdot i_{\max} \cdot r_1}{\frac{r_1}{r_2} \cdot (1 - \eta_f) + 1} \tag{3}$$

where: $E_{\max}$ – maximum voltage; η_f – fuel utilization factor; $i_{\max}$ – maximum current density; r_1 – internal ionic area specific resistance of the cell; r_2 – internal electronic area specific resistance of the cell.

The second type of internal resistance is electronic resistance – r_2 (see Fig. 2). The influences of temperature and matrix thickness on electronic internal resistance of electrolytes are not well known. The electronic conductivity values of molten carbonate electrolytes are probably spread across a very wide range. They do not have a major impact on calculated cell voltage for high fuel utilization factors. It is difficult to measure the electronic resistance of molten carbonate electrolytes because they have both conductivities, ionic and electronic simultaneously – which gives total electrical resistance. It should be noted that decreasing electrolyte matrix thickness reduces ionic resistance, but it also probably reduces electronic resistance. The electronic resistance has influence mainly on Open Circuit Voltage (OCV).

The value of electronic resistance of the cell can be estimated from available experimental results. Substituting η_f=0 into Eq. 3, the OCV can be defined by the following relationship:

$$E_{OCV} = \frac{E_{\max}}{\frac{r_1}{r_2} + 1}$$

For given r_1, $E_{\max}$ and E$_{OCV}$ (from experimental measurements) the value of electronic conductivity of the cell can be found from the following relationship:

$$\sigma_2 = \delta \cdot \frac{E_{\max} - E_{OCV}}{r_1 \cdot E_{OCV}} \tag{4}$$

where: where: σ_2 - electronic conductivity of the electrolyte.

$$r_2 = \frac{\delta}{\sigma_2} \tag{5}$$

The value of σ_2=3.5·10^{-3}S/cm, was taken from the researchers' own calculations, which were based on data presented by Arato et al [12]. It was assumed that this value is independent on temperature. It was assumed that the thickness of electrolyte matrix is 1 mm.

An MCFC installed at a gas turbine outlet requires a large active area due to the large amount of gas and low CO_2 content. The $i_{\max}$ of 0.6 A/cm^2 was determined by the researchers' own calculations and based on experimental data [12].

The maximum voltage of a singular cell is given by the following equation:

$$E_{\max} = \frac{RT}{4F} \ln \frac{p_{O_2,cathode} \cdot p^2_{CO_2,cathode}}{p_{O_2,anode} \cdot p^2_{CO_2,anode}} \tag{6}$$

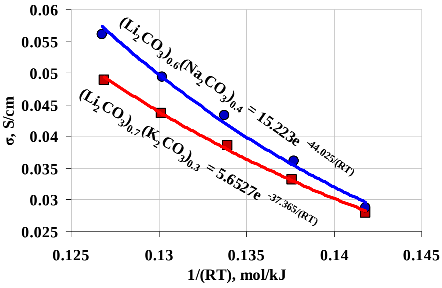

Fig. 3. Temperature dependence of ionic conductivity for molten carbonates

where: T – absolute temperature; R – universal gas constant; F – Faraday's constant; $p_{O_2,cathode}$ – oxygen partial pressure at cathode inlet; $p_{O_2,anode}$ – oxygen partial pressure at anode inlet; $p_{CO_2,cathode}$ – carbon dioxide partial pressure at cathode inlet; $p_{CO_2,anode}$ – carbon dioxide partial pressure at anode inlet.

The partial pressure of oxygen at the anode is very low and can be calculated using a chemical equilibrium constant:

$$K = \frac{p_{H_2O,anode} \cdot p_{ref}^{1/2}}{p_{H_2,anode} \cdot p_{O_2,anode}^{1/2}} \tag{7}$$

where: K – chemical equilibrium constant for hydrogen-oxygen reaction; $p_{H_2O,anode}$ – water partial pressure at anode inlet; $p_{H_2,anode}$ – hydrogen partial pressure at anode inlet; p_{ref} – reference pressure.

Substituting Eq. 7 into Eq. 6, the Nernst equation is obtained:

$$\begin{aligned} E_{\max} &= \frac{RT}{2F} \ln K \\ &+ \frac{RT}{2F} \ln \frac{p_{H_2,anode} \cdot p_{O_2,cathode}^{1/2}}{p_{H_2O,anode} \cdot p_{ref}^{1/2}} \\ &+ + \frac{RT}{4F} \ln \frac{p_{CO_2,cathode}^2}{p_{CO_2,anode}^2} \end{aligned} \tag{8}$$

Adequate partial pressures have been calculated through the use of software. Those calculations are based on Peng-Robinson thermodynamic functions and minimization of Gibbs' free energy [13].

The ionic resistance of molten carbonate electrolytes as a function of electrolyte matrix thickness and temperature is shown in Fig. 3. This diagram contains values obtained by the researchers' own calculations, which were based on data published by Morita et al. [14].

The internal area specific ionic resistance can be described by the following relationship:

$$r_1 = \frac{\delta}{\sigma} \tag{9}$$

where: δ – electrolyte matrix thickness; σ – ionic conductivity of molten carbonate.

The ionic conductivity of the carbonate is defined as follows:

$$\sigma = \sigma_0 \cdot e^{\frac{-E}{RT}} \tag{10}$$

where: σ_0, S/cm; E, kJ/mol, – factors depended on used carbonate.

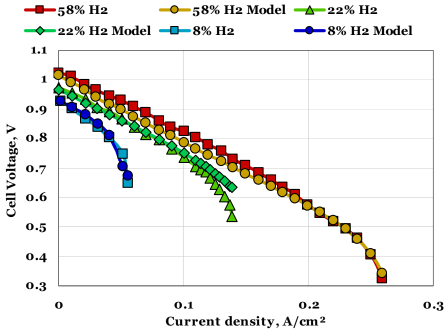

Fig. 4. Experimental and simulations data at different H_2 molar fractions, experimental data taken from [14]

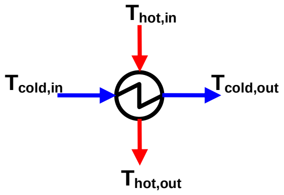

Fig. 5. Heat exchanger

B. Heat Exchanger

Heat exchanger efficiency is defined by the following equation:

$$\eta_{HX} = \frac{T_{hot,out} - T_{hot,in}}{T_{cold,in} - T_{hot,in}} \tag{11}$$

where: T – temperature, *hot* – hot side of heat exchanger, *cold* – cold side of heat exchanger, *in* – inlet, *out* – outlet.

C. Gas Turbine Power Plant

GTPP consists of the following elements:

1) Air compressor
2) Gas turbine
3) Combustion chamber

Compressed air is delivered to the combustion chamber where fuel (natural gas) is oxidized. Hot gas expands through the gas turbine and escapes to the atmosphere.

Mathematical model of the GTPP was created based on the following assumptions:

- air compressor isentropic efficiency: 79%
- gas turbine isentropic efficiency: 88%
- no pressure drops across the combustion chamber

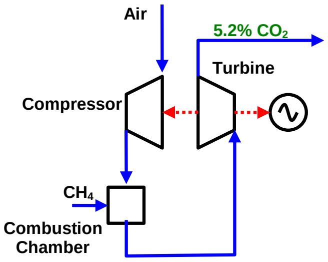

Fig. 6. GTPP system

TABLE II
NOMINAL PARAMETERS OF GT POWER PLANT [15]

Name	Value
Air compressor inlet pressure, MPa	0.1
Air compressor inlet temperature, °C	15
Pressure ratio	17.1
Fuel	Natural Gas
Fuel massflow, kg/s	4.0
Turbine inlet temperature, °C	1210
Exhaust gas massflow, kg/s	213
Turbine outlet temperature, °C	587
GT Power, MW	65
GT Efficiency (LHV), %	33
CO2 annually emission, Gg/a	250
Relative emission of CO_2, kg/MWh	609
CO_2 mass flow, kg/s	11

A commercial gas turbine unit was chosen to analyze [15]. Nominal parameters of the GTPP and exhaust gas composition are shown in Tables III and I, respectively.

To compose a $CO_3{}^{2-}$ ion, it is needed to split a half mole of O_2 with one mole of CO_2. Adequate mass and molar ratios of CO_2 to O_2 (for capture all carbon dioxide) are 1.38 and 2, respectively. From data given in Table I seems that, theoretically, all CO_2 could be captured.

D. Optimizing process

All analyzed cases were optimized with the objective function being total power generation efficiency. Nevertheless, there is room for discussion as to the choice of this as the objective function of the optimizing process. While the main task of an MCFC is to capture CO_2 from flue gas, it also increases total power generation efficiency due to its higher efficiency compared with that of the steam cycle (44% vs. 30%).

The BOX method was used for optimizing all systems [13]. The procedure is loosely based on the "Complex" method of Box [16]; the Downhill Simplex algorithm of Press et al. [17] and the BOX algorithm of Kuester and Mize [18].

This method is a sequential search technique which solves problems with non-linear objective functions, subject to non-linear inequality constraints. No derivatives are required. It handles inequality but not equality constraints. This method is not very efficient in terms of the required number of function evaluations. It generally requires a large number of iterations to converge on the solution.

The size of the MCFC installed at the flue gas can be varied in wide range. From the other hand the same fuel utilization ratio can be realized by different size fuel cells. There are three main parameters which determine the MCFC size: fuel utilization factor, maximum current density and inlet fuel flow. Two from these three parameters set the size of the MCFC. The stack fuel utilization factor was chosen at constant level of 90%. The maximum current density and fuel mass flow were taken as primary variables of the optimizing process.

Optimized parameters:

- MCFC fuel mass flow
- The value of i_{max} in the range 0.06 A/cm^2 to 0.3 A/cm^2
- Heat Exchanger efficiency in the range 0 to 85%

The optimizing process was carried out with the temperature inside the stack below 750°C.

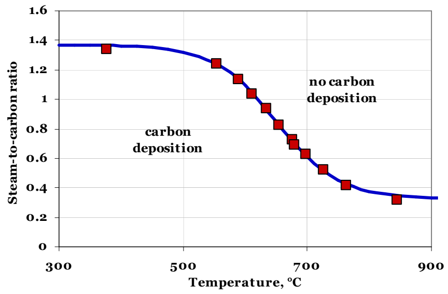

Fig. 7. Minimum temperature and required ratio of steam-to-carbon (s/c) above which no carbon deposition takes place [19]

III. GAS TURBINE POWER PLANT WITH MCFC

The CO_2 reduction emission factor is defined as follows:

$$\eta_{CO_2} = 1 - \frac{\dot{m}_{CO_2,out}}{\dot{m}_{CO_2,in}} \tag{12}$$

where: $\dot{m}$ – mass flow, kg/s; out – MCFC outlet cathode stream; in – MCFC inlet cathode stream.

Two cases of gas turbine power plant with the MCFC were investigated. Case 1 concerns a situation when there is no intervention in GTTP cycle. It means that MCFC is added at GTPP outlet stream. Case 2 concerns the situation when heat exchangers before combustion chamber are added to the GTPP. These heat exchangers are fed by MCFC exhaust streams. Relatively low CO_2 content in flue gas results low MCFC efficiency. The MCFC efficiency is about 34% (based on Lower Heating Value, LHV). It seems to be unreasonable to compose low efficient MCFC with high efficient Combined Cycle Gas Turbine (with efficiency about 55%) and this case was not investigated.

Proper objective function of the optimizing process is not obviously. The MCFC is installed to capture the CO_2, from this point of view the quantity of captured CO_2 should be maximized. But from other side, the MCFC uses the same fuel as gas turbine to produce the electricity. Both analyzed cases were optimized to obtain maximum system efficiency.

In both cases apart from MCFC, following devices should be installed:

1) CO_2 separator
2) Catalytic burner
3) DC/AC converter with efficiency of 95%

The CO_2 separator is cooled by water. When steam condenses, water is taken away from the carbon dioxide stream.

The catalytic burner is fed by pure oxygen to utilize the rest of methane, hydrogen and carbon oxide. An oxygen extraction (e.g. from air) requires energy. The production of one kilogram of oxygen at atmospheric pressure requires from 200 to 300 kJ. The value of 250 kJ was taken into calculations, which decreases the system efficiency depending on the amount of consumed oxygen.

The methane is mixed with steam to avoid a carbon deposition. It was assumed that adequate steam-to-carbon ratio to prevent the carbon deposition is 1.4 (see Fig. 7). Steam-to-carbon ratio (s/c ratio) defines steam molar flow in relation to methane and carbon monoxide molar flows delivered to the reformer.

TABLE III
NOMINAL PARAMETERS OF GTPP-MCFC, CASE 1

Name	Value
GTPP-MCFC power (total power), MW	80
GTPP power/total power, %	81
MCFC power/total power, %	19
GTPP-MCFC efficiency (LHV), %	33
CO_2 emission reduction factor, %	73
Annual CO_2 emission, Gg/a	67
Relative CO_2 emission, kg/MWh	132
MCFC efficiency (LHV), %	34
GTPP efficiency (LHV), %	32
Fuel utilization factor, %	90
Average cell voltage, mV	513
Current density, mA/cm^2	29.5
Oxygen mass flow, kg/s	0.2
MCFC/GTPP fuel ratio	0.52

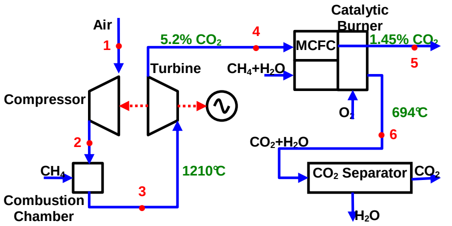

Fig. 8. GTPP-MCFC system – Case 1

$$s/c = \frac{\dot{n}_{H_2O}}{\dot{n}_{CH_4} + \dot{n}_{CO}} \qquad (13)$$

where: $\dot{n}$ – molar flow, kmol/s; H_2O – steam; CO – carbon oxide; CH_4 – methane.

The installation of MCFC at gas turbine outlet means back pressure drop of about 1%. It decreases the efficiency of the GTPP from 33% to 32%.

A. Case 1

The MCFC is fed by two streams: GTPP exhaust gas at cathode side and a mixture of methane and steam at anode side.

The MCFC anode outlet stream is directly delivered to the CO_2 separator.

The system was optimized to obtain maximum system efficiency. Primary (adjusted) variables of the optimizing process were:

1) cell current density;
2) and MCFC/GTPP fuel ratio.

Parameters obtained during the optimizing process are given in Table III.

A simple combination of the MCFC with GTPP gives:

1) CO_2 emission reduction of 73%,
2) Electric efficiency remains the same,
3) Power generation increase of 23%.

B. Case 2

The MCFC-GTPP in Case 2 was created by adding two heat exchangers (see Fig. 9). The heat exchangers have a role to recover exhaust heat from MCFC outlet streams. Note that the GTPP efficiency would increase with a recuperative heat exchanger when no MCFC is installed as well.

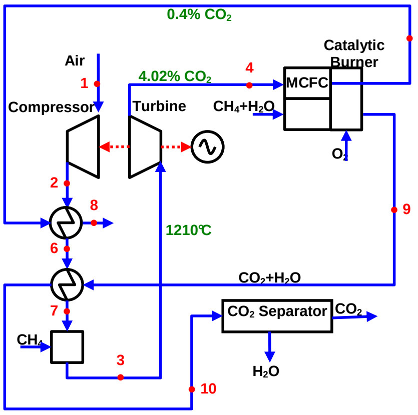

Fig. 9. GTPP-MCFC system – Case 2

TABLE IV
NOMINAL PARAMETERS OF GTPP-MCFC, CASE 2

Name	Value
GTPP-MCFC power (total power), MW	77
GTPP power/total power, %	82
MCFC power/total power, %	18
GTPP-MCFC efficiency (LHV), %	40
CO_2 emission reduction factor, %	91
Annual CO_2 emission, Gg/a	18
Relative CO_2 emission, kg/MWh	37
MCFC efficiency (LHV), %	36
GTPP efficiency (LHV), %	41
MCFC fuel utilization factor, %	90
Average cell voltage, mV	486
Average current density, mA/cm^2	29.6
Oxygen massflow, kg/s	0.2
MCFC/GTPP fuel ratio	0.65

The system was optimized with the same conditions like Case 1. Nominal parameters of Case 2 of GTPP-MCFC system are given in Table IV.

GTPP-MCFC Case 2 generates slightly less power in comparison with Case 1. During the simulations a constant value of Turbine Inlet Temperature (TIT) was assumed. The implementation of heat exchangers means lower fuel massflow demanded by the combustion chamber.

A reduction of the CO_2 emission of 91% is obtained. Simultaneously, electric efficiency is increased to 40% (LHV) what gives the relative emission of CO_2 of 37 kg/MWh.

IV. DISCUSSION

The CO_2 emission reduction factor and CO_2 relative emission were used to compare the systems. These values for all analyzed cases are given in Table V. The MCFC could

TABLE V
MAIN PARAMETERS OF ANALYZED SYSTEMS

Name	GTPP	Case 1	Case 2
MCFC$_{fuelflow}$/GT$_{fuelflow}$, %	0	29	45
System Efficiency, %	33	33	40
CO_2 emission reduction factor, %	0	73	91
MCFC/GTPP fuel ratio	0	0.52	0.65
Relative CO_2 emission, kg/MWh	609	135	37
Annual CO_2 emission, Gg/a	250	68	18
Total system power, MW	65	80	77

reduce the CO_2 emission of above 70% from gas turbine power plant exhaust. The relative CO_2 emission decreases more significant because the MCFC produces additional power.

Relatively low efficiency of the MCFC is caused by low CO_2 content at gas turbine exhaust, which gives low maximum cell voltage (see Eq. 6).

The combination of MCFC with GTPP means higher investment costs. Other devices like CO_2 separator and heat exchangers increase the total investment cost as well. It should be noted that typical CO_2 separation methods also increase the investment costs.

Application of the MCFC in a Gas Turbine Power Plant gives a relatively high reduction in CO_2 emissions. The relative CO_2 emission of the GTPP is estimated at 609 kg_{CO_2}/MWh while in contrast the MCFC-GTPP hybrid system has an emission rate of 135 kg_{CO_2}/MWh. The quantity of CO_2 emitted by the MCFC-GTPP is 73% lower than is the case with the GTPP.

As mentioned earlier, all cases were optimized to achieve maximum power generation efficiency. However, this may be open to challenge if it is accepted that the main task of the MCFC is to limit CO_2 emissions, which would result in the CO_2 emission reduction factor being used as the objective function of the optimizing process. If this factor is optimized the cell voltage at last cell can fall below zero and the MCFC will work as a CO_2 concentrator. At the very least, the MCFC would generate no power, and might even consume some. However, the main task of a power plant is power generation; hence hybrid system efficiency was chosen as the objective function for optimization.

It should be borne in mind that prices of tradeable CO_2 allowances are relatively constant at present, which affords opportunity to realize profits from carbon trading.

Important technical issues such as sulphur or dust resistances of the MCFC fell outside the remit of this paper, although they can evidently limit the application of MCFCs in gas turbine power plants.

MCFCs could be profitably used in existing power plants which have been given CO_2 limits. MCFCs could potentially decrease CO_2 emissions, leaving the power generation capacity of the system at least the same, if not greater.

V. CONCLUSIONS

1) There is no unequivocal choice of objective function of the optimizing process for this type of hybrid system
2) MCFCs could reduce CO_2 emissions by a factor of 73%
3) A relative emission of 135 kg/MWh appears achievable for the hybrid system
4) Current price levels on the CO_2 emissions trading market in the EU are at €15/Mg_{CO_2}, wchich allows the realization of carbon trading profits from the use of MCFCs.

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