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Modeling and characterization of molten carbonate fuel cell for electricity generation and carbon dioxide capture

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Abstract

The growing electricity request and more severe commitments on emissions led to the research of more and more efficient energy transformation processes. The use of Fuel Cell in order to improve energetic and exergetic efficiency is well-assessed and a number of advanced processes and highly functional materials are currently under investigation, advising a high potential of these systems for the future development of sustainable energy technologies. In particular, the capabilities of integrating high temperature fuel cells within energy conversion systems having medium and high grade thermal sources (flue gases) has resulted in a renewed interest in Molten Carbonate Fuel Cells (MCFC). In fact, they operate at temperatures in the range of 600-700°C and they could be fed by unreformed gas, internally integrating a methane-steam reforming section (direct or indirect).
In this paper, the Authors present a theoretical activity finalized to the design and characterization of the integration of a MCFC in a coal fired power plant: a physical model of the fuel cell has been developed, where the energy and chemical processes are represented for the cell stack and geometrical and electrical parameters have been taken into consideration. The model has been applied for system analysis with respect to multiple steady states, sensitivity and stability behavior. Both direct and indirect internal reforming cases have been compared each other, evaluating the energetic and environmental performances of the use of the MCFC as CO_2 remover.

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Keywords: MCFC, CO_2 capture, repowering, steam power plants, modeling

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1. Introduction

Fuel cells are high efficiency electricity-generating systems that convert chemical energy from oxidation of the fuel directly into electric energy. They have been widely researched and developed as a promising clean and high-efficiency energy-generating technology in several sectors: electrical energy production is surely the most important one, but also in transportation sector the interest in fuel cell powertrains is raising, in particular for marine [1] and automotive applications [2][3].

Molten carbonate fuel cells (MCFCs) are high-temperature fuel cells that operate in the range of 600-700 °C [4],[5]. The MCFC has different advantages with respect to other fuel cell systems: it offers high fuel conversion efficiency and can operate with alternative fuels and hydrocarbons [6][7], so widening the potential application: molten carbonates are very stable systems under a wide range of chemical conditions and temperature ranges, giving the possibility of designing ideal reactions and electrolyte media for advanced chemical/electrochemical processes related to production, storage, conversion and efficient energy performance [8].

MCFCs have two types of fuel processing. One is an external reforming MCFC (MCFC-ER) that produces hydrogen at the outside of the MCFC stack. In this case, the stack temperature can be controlled by convection heat transfer only. Therefore, the external reforming adopts a pressurized operation to supply a high rate of cooling gas flow. The other type is an internal reforming MCFC that directly generates hydrogen in the MCFC stack. The stack temperature is controlled within the stack to remove the heat generated from the endothermic reforming reactions. Because the MCFC with internal reforming does not require a high flow rate, this fuel cell can operate under atmospheric conditions [9]. Internal reforming processes can be further divided in direct (MCFC-DIR) and indirect (MCFC-IIR) one: in the first case, the reforming is made in the anode room itself, with benefit on thermal exchange efficiency, on the other hand, the internal reforming, can be also made just adjacent the anode. The two types can be also integrated in a two-step reforming of the anode inlet flow [10].

The application of Molten Carbonate Fuel Cell (MCFC) technology to CO_2 capture and separation from flue gases represents one of the most interesting technology [11],[12]. In fact, the electrochemical reactions that take place within the MCFC involve the migration of the CO_2 from the cathode to the anode. During this process, the MCFC can operate as power generator and, simultaneously, as CO_2 separator from an exhaust gaseous stream (produced by a combustion process) and sent to the cathode [13][14]. This opens the way to a wide applicability of MCFCs (and also other high temperature fuel cells, like SOFC [15],[16]) as CO_2 remover and energy conversion in thermoelectric power plants. In fact, they can be fed by the exhaust gases of a gas turbine power plant [17][19] or integrated in a gas-steamcombined cycle power plant [20],[21].

In this paper, a physical model for the design of a MCFC is presented, considering both direct and indirect internal reforming. The model has been applied on the integration of a MCFC with a coal fired power plant [22]. The integration is realized using the MCFC as active unit for the CO_2 removal and a novel thermal recovery section has been introduced, using the residual heat, after the steam production needed for methane reforming, to partially preheat the feedwater in high- and low-pressure regenerative lines of the SPP. MCFC has been sized in order to capture the CO_2 from flue gases of the thermoelectric power plant and performances of the integrated system have been investigated. Great environmental and energetic improvements are achieved in terms of plant efficiencies and specific primary consumption for CO_2 avoided (SPECCA index).

Nomenclature

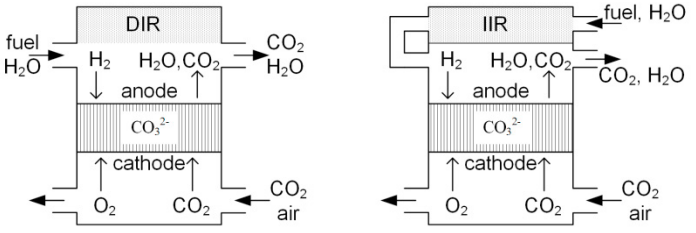
an	anode	*IIR*	Indirect Internal Reforming	*ref*	reference
ASU	Air Separation Unit	*in*	inlet	*SOFC*	Solid Oxide Fuel Cell
cat	cathode	*IPP*	Integrated Power Plant	*SPECCA*	Specific Primary Energy Consumption for CO_2 Avoided
comp	compression	*LHV*	Lower Heating Value		
DIR	Direct Internal Reforming	*M*	Molar mass	*SPP*	Steam Power Plant
ER	External Reforming ṁ	*ṁ*	mass flow rate	*U*	Utilization factor
F	Faraday constant	*MCFC*	Molten Carbonate Fuel Cell	*y*	molar fraction
h	enthalpy	*P*	Power	η	efficiency
HX	Heat Exchanger	*out*	outlet	φ	CO_2 removal factor
I	Current	*Q*	heat flux		

2. MCFC modeling

In high-temperature FC technology, fuel (usually natural gas), mixed with steam, can feed directly the anode of the FC stack. In MCFC, the reforming stage can be integrated within the fuel cell in an indirect (IIR) or direct way (DIR). In the IIR case, a reforming unit is placed in the stack, converting natural gas and steam into a hydrogen-rich gas before entering into the cell anode. In MCFC-DIR, instead, the catalyst bed is located in the same chamber of the anode, providing better thermal and steam management. It is assumed that only H_2 is directly converted in the cell electro-chemical reaction. Other components in the fuel, such as CO, CH_4 and higher hydrocarbons are converted through reforming processes and shift reactions: in IIR case, the fuel reaches an equilibrium in the integrated reformer at a specified temperature before it can be transmitted to the anode, while, when the DIR is considered, this reforming temperature is the same of the fuel cell one [23] and the methane conversion is affected by mutual interaction of reforming and oxidation reactions at the anode side.

As a result of steam reforming, H_2 is introduced at the porous anode electrode. At the cathode electrode, the oxygen and carbon dioxide in the oxidant stream realize a reaction to form carbonate ions (CO_3^{2-}). At anode side, the hydrogen is transformed into steam and carbon dioxide thanks to the carbonate ions which migrate through the molten salt electrolyte. The model is isothermal: the calculated chemical balances on the active cell area and the current density are based on the average cell temperature, which is equal to 640°C in the application considered. Moreover, the (electrical) cells connected in series that constitute a fuel cell stack, have identical performance.

The design of the Molten Carbonate Fuel Cell starts from the knowledge of all conditions at the inlet (pressure, temperature and composition) of both the anode and the cathode sides. In design calculations, cell voltage, current density and fuel utilization are specified. On the basis of this data, the corresponding cell resistance and cell area can be estimated; the effective power of the fuel cell is calculated on the basis of the fuel mass flow to the anode. The mass balance is represented in eqn. 2, where also the mass transfer from cathode to anode has been considered.



$$\begin{cases} \frac{1}{2}O_2 + CO_2 + 2e^- \xrightarrow{cathode} CO_3^{2-} \\ H_2 + CO_3^{2-} \xrightarrow{anode} H_2O + CO_2 + 2e^- \end{cases} \quad (1)$$

$$\begin{cases} \dot{m}_{an,in} + \dot{m}_{cat,in} = \dot{m}_{an,out} + \dot{m}_{cat,out} \\ \dot{m}_{an,out} - \dot{m}_{an,in} = \dot{m}_{O_2,cat\to an} + \dot{m}_{CO_2,cat\to an} \end{cases} \quad (2)$$

Figure 1. MCFC with internal direct (DIR) and indirect (IIR) reforming

This flow is constituted by CO_2 and O_2 that migrate from cathode to anode. The two contributes can be calculated according to eqn. 3 and they are strictly related to the electrical current produced by the fuel cell. O_2 utilization factor U_{O_2} is defined as the ratio between the migrated O_2 and the inlet cathode fluxes.

$$\begin{cases} \dot{m}_{O_2,cat\to an} = M_{O_2}\frac{I}{4F} = U_{O_2}\cdot\dot{m}_{O_2,cat} \\ \dot{m}_{CO_2,cat\to an} = M_{CO_2}\frac{I}{2F} = \varphi\cdot\dot{m}_{CO_2,cat} \end{cases} \quad (3)$$

At the same moment, the current produced is calculated from the fuel utilization level U_{fuel} (that affects the fuel conversion efficiency) and its chemical composition (considering only CH_4, CO and H_2):

$$I = 2\cdot F\cdot U_{fuel}\cdot\frac{\dot{m}_{an,in}}{M_{an,in}}\left(4y_{CH_4} + y_{CO} + y_{H_2}\right)_{an,in} \quad (4)$$

Once current is determined, the fuel cell power can be calculated from the design voltage and its area from the current density. In general, cell voltage and current depend on the polarization curve of the fuel cell. However, in

design case, they are considered constant in a fixed design working point, which is the suitable one for the operation of the fuel cell (i.e maximum efficiency). Finally, energy balance is used to calculate the temperatures of the exiting anode and cathode flows, imposing that they have the same outlet temperature.

$$\dot{m}_{an,in}(h_{an,in}+LHV_{an,in})+\dot{m}_{cat,in}h_{cat,in}-\dot{m}_{an,out}(h_{an,out}+LHV_{an,out})-\dot{m}_{cat,out}h_{cat,out}=P-Q \quad (5)$$

In the cases studied (MCFC DIR and IIR) the fuel cell has been considered adiabatic. This can be done thanks to the limited temperature increase of the gases: most part of the thermal power associated to irreversibility is used in the endothermic process of the fuel reforming.

3. Application on a coal fired power plant

The model has been used in order to study a possible integration between an existing coal-fired power plant and a MCFC. The steam power plant (SPP), simulated using Gatecycle software [24], comprises a subcritical fossil boiler, high, intermediate and low pressure steam turbines, a deaerator and six regenerative heat exchangers, three in the LP line and three in the HP line (Figure 2). It has a net power output of 160 MW and all its fundamental characteristic parameters are summarized in Table 1. The fuel cell can be used as a CO_2 capture device for the flue gases.

Table 1. Operating parameters and energy performances of coal-fired steam power plant at design conditions

Net power output (P_{ref})	160 MW
Net LHV efficiency (η_{ref})	41.0 %
Condenser operating pressure	0.05 bar
Temperature at HP steam turbine inlet	540 °C
Pressure at HP steam turbine inlet	180 bar
Temperature at IP steam turbine inlet	540 °C
Pressure at IP steam turbine inlet	34 bar
Temperature at LP steam turbine inlet	320.5 °C
Pressure at LP steam turbine inlet	7 bar
Coal lower heating value (LHV_{coal})	25.4 MJ/kg
Fuel mass flow rate ($\dot{m}_{coal,SPP}$)	15.4 kg/s
Steam mass flow rate at condenser inlet	88.6 kg/s
Water mass flow rate at deaerator inlet	105.0 kg/s
Superheated steam mass flow rate	128.3 kg/s
Water temperature at fossil-boiler inlet	242.4 °C
Water temperature at deaerator inlet	130.8 °C
Thermal power to feedwater in LP regenerative line	39.9 MW
Thermal power to feedwater in HP regenerative line	42.1 MW
CO_2 molar fraction in the flue gas	13.7 %
O_2 molar fraction in the flue gas	3.2 %
Specific CO_2 emissions	768.5 kg/MWh

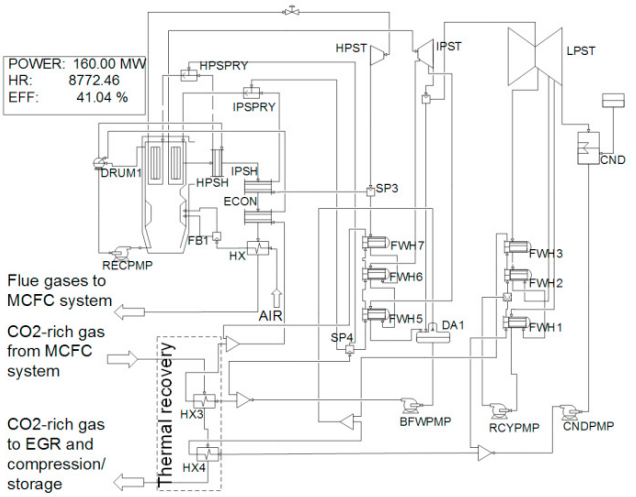


Figure 2. Coal fired power plant with thermal recovery section fed by CO_2-rich gas from MCFC system

Flue gas from the steam power plant is rich of CO_2 (13.7 % on molar basis, Table 1), but the residual oxygen is low (3.2%), due to the limited air excess in combustion side of the boiler (20%). Therefore, fresh air has to be added to the flue gas in order to feed the cathode of the MCFC unit (Figure 3) and to reach a significant CO_2 removal factor. If fresh air was not added, maximum CO_2 conversion in the MCFC anode is about 33% and resulted MCFC size is 30 MW (having considered a fuel utilization of 80% and an O_2 utilization of 70%). A compressor is needed to energize the flow of this mixture of gases. On the other hand, the anode side is fed by methane and steam. The latter is produced by using the thermal power coming from the combustion of anode exhaust gases with oxygen: this produces a hot CO_2-rich gas which can be used to produce steam for the anode inlet and pre-heat a fraction of the feedwater in the regenerative line of the steam power plant (thermal recovery section, Figure 2 and Figure 3), both at low and high pressure (HX3 and HX4 in Figure 2). The thermal power recovered from the MCFC system, sized to

obtain a fixed CO_2 removal factor from the SPP, allows to reduce the steam extractions along the turbines, producing an overload of the LP turbine and condenser. The behaviour and performance of the modified SPP have been evaluated in off-design conditions [24]. Thus, the condenser overload with respect to design case has been estimated and, if required, reduced through a boiler derating, obtained lowering the coal feeding and the superheated steam generation. Downstream the heat recovery (HX4 in Figure 2), the CO_2-rich gas is cooled and compressed in order to separate and store the carbon dioxide in liquid phase. A fraction of the exhaust gases flowrate is also recirculated (EGR) to the combustor to control the maximum gas temperature at the combustor exit and reduce the amount of oxygen to be produced by the Air Separation Unit (ASU). Furthermore, anode and cathode outlet gases can be used to pre-heat the homologous inlet flows in HX1 and HX2 of Figure 3. It shows, also, the novel thermal recovery, after having produced the steam needed for methane reforming, which further integrates the MCFC system with the coal fired power plant. This is allowed because the thermal energy produced inside the combustor is greater than what is required to produce the steam for methane reforming.

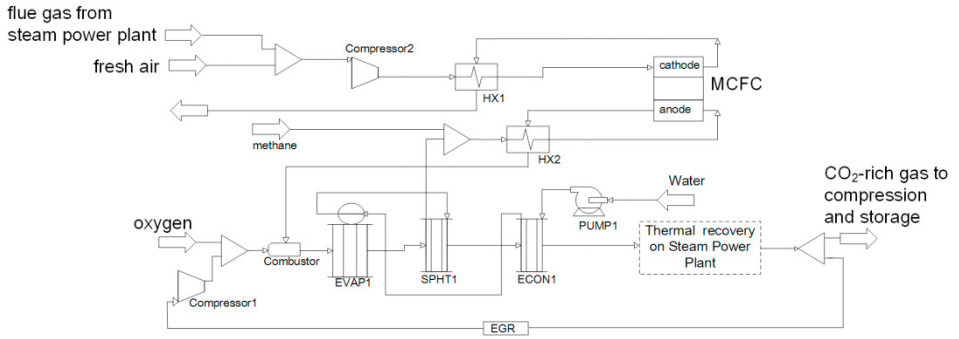


Figure 3. MCFC system

In the configuration proposed, the flue gas temperature is well-matched with the internal reforming of the MCFC. Hence, an indirect internal reforming (IIR) has been firstly considered and the whole performances have been analyzed imposing a fixed desired percentage of CO_2 removal factor (φ) from 70% to 95%. With these high values, important rates of fresh air are needed and this produces high sizes of MCFC (40-65 MW, Figure 4). Therefore, oxygen utilization must be adequate to CO_2 removal factor: high O_2 utilization produces a reduction of the fuel cell size but leads to lower fuel cell performance due to polarization losses: a value of 70% seems to be suitable in this application. At anode side, for IIR case, a steam-to-methane molar ratio higher than the stoichiometric value [18] has been considered (3), in order to promote methane conversion and avoid carbon deposition. The analyses have been conducted admitting a certain value of condenser overload: this is maximum when the steam mass flow rate produced in the boiler is the same of the original plant (i.e. no boiler derating), while, whenever the condenser overload would be limited, the steam mass flow rate has to be reduced and, so, the boiler has to be derated.

Figure 4 shows the ratio of the mass flow rate of the feedwater deviation needed to dispose the thermal power from the MCFC system. Both the deviations at high pressure and low pressure regenerative line have the same value. In particular, it is about 50% when a CO_2 removal factor equal to 70% is considered and MCFC thermal power is in the range between 40-47 MW. It reaches values up to 70% when a maximum CO_2 removal factor of 95% is considered and the MCFC thermal power ranges from 55 to 67 MW (Figure 4). However, in the higher cases, the condenser overload resulted in values higher than 20%. This is not really sustainable for the condenser. Lower condenser overloads can be achieved only with a lower steam mass flow rate production and, so, a boiler derating (Figure 5). In a bordering case, when no condenser overload is admitted, the boiler derating ranges from 16% to 21% for CO_2 removal factor from 70% to 95% and MCFC electrical power from 61 to 80 MW. If a suitable maximum value of about 15% is assumed for the condenser overload, the boiler derating ranges from 2% to 8% and MCFC net AC power from 68 to 89 MW.

Figure 6 shows the efficiency of the coal fired steam power plant (η_{SPP}, eqn. 6) when the thermal power recovered from the MCFC system is integrated within it. The efficiency is always higher (than the initial value of 41%) because the partial pre-heating of the feedwater done allows to reduce the steam extractions along the

expansion line: a maximum increase till to 45.3% is achievable. Higher values are reached without overloading the condenser. The most valuable result is that this efficiency increases at higher CO_2 removal factor, due to the active role of the MCFC system.

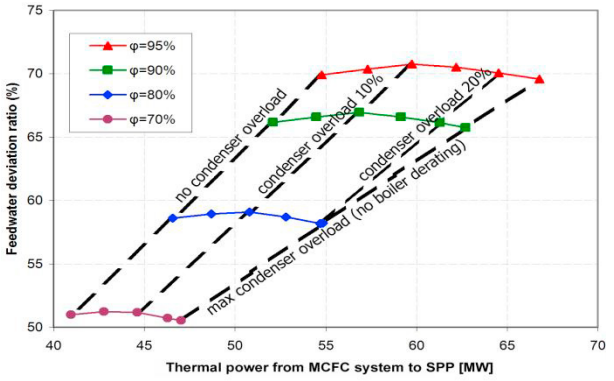


Figure 4. MCFC-IIR: feedwater deviation ratio varying CO_2 removal factor and condenser overload

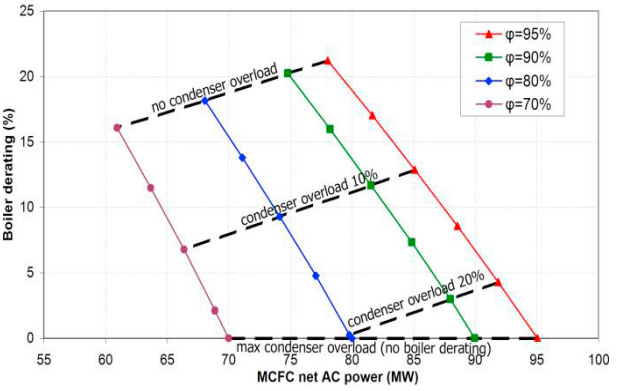


Figure 5. MCFC-IIR: boiler derating varying CO_2 removal factor and condenser overload

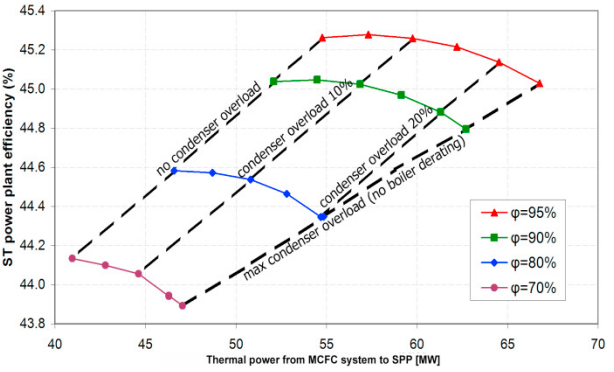


Figure 6. MCFC-IIR: steam power plant efficiency (η_{SPP}) varying CO_2 removal factor and condenser overload

$$\eta_{SPP} = \frac{P_{SPP}}{\dot{m}_{coal} LHV_{coal,SPP}} \tag{6}$$

$$\eta_{IPP} = \frac{P_{SPP} + P_{MCFC} - P_{comp,MCFC} - P_{ASU} - P_{comp,CO_2}}{\dot{m}_{coal} LHV_{coal,SPP} + \dot{m}_{CH_4} LHV_{CH_4,MCFC}} \tag{7}$$

$$SPECCA = \frac{\left(\eta_{SPP}^{-1} - \eta_{ref}^{-1}\right)}{\frac{\dot{m}_{CO_2,ref}}{P_{ref}} - \frac{\dot{m}_{CO_2,SPP}}{P_{SPP}}} \tag{8}$$

Same trends are showed for the integrated power plant (IPP) efficiency: it has been calculated as in eqn. 7, where the net overall power is the sum between the steam power plant one (P_{SPP}) and the MCFC one (P_{MCFC}), properly decurted by the power needed by the compressors in the MCFC unit, the one absorbed by the ASU (P_{ASU}, estimated in 880 MJ/t_{O_2}) and the one given to the compression stages of the CO_2 separation and storage section (P_{comp,CO_2}, assumed to be 350 MJ/t_{CO_2}). Coal for the SPP and methane for MCFC have been considered as inlet fuels.

Environmental performance has been evaluated according to the specific primary energy consumption for CO_2 avoided (SPECCA). It is estimated as in eq. 8 and its negative values mean for active CO_2 capture. Figure 7 shows environmental and energetic results of the integrated power plant with MCFC-IIR. SPECCA index is in the range of -0.30 to -0.36 for MCFC electrical AC rated power from 60 to 95 MW: better performance are reached with higher CO_2 removal factor and without admitting condenser overload. Integrated plant energetic efficiency, on the other hand, ranges from 41.9% to 42.35%. Same considerations apply when a MCFC-DIR is considered (Figure 8). In this case, a steam-to-methane molar ratio lower than the stoichiometric value can be assumed (1.5), thanks to a favourable steam management in the anode side. With the DIR fuel cell, SPECCA index is more than doubled and IPP efficiency is 1.5% higher than in IIR case.

Finally, the comparison between the MCFC-IIR and MCFC-DIR in terms of water/steam flows (in the MCFC system) is shown in Figure 9 and 10: in IIR case, the steam needed to feed the anode ranges from 9 to 11 kg/s, while in the DIR case it is lower (4-5 kg/s). Hence, water recovered (condensate) within the carbon dioxide compression and separation section is 15-18 kg/s in the IIR case and 9-12 kg/s in the DIR one.

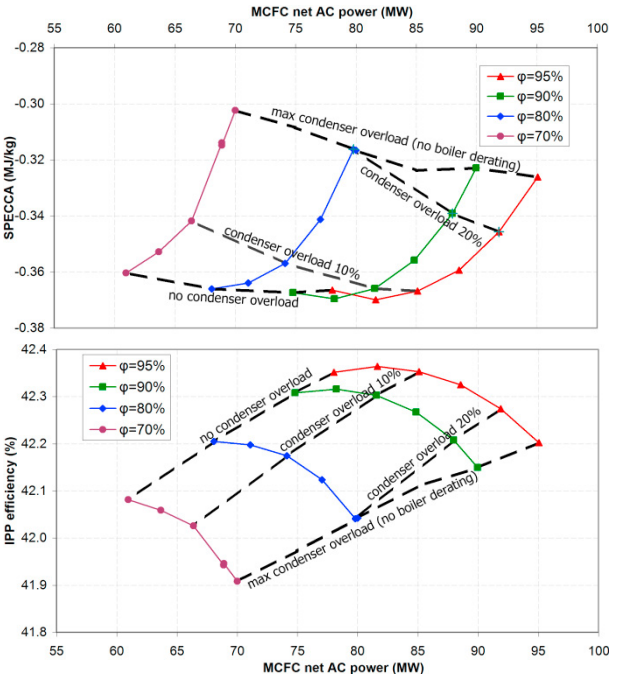


Figure 7. SPECCA index and IPP efficiency for the MCFC-IIR unit integrated with SPP

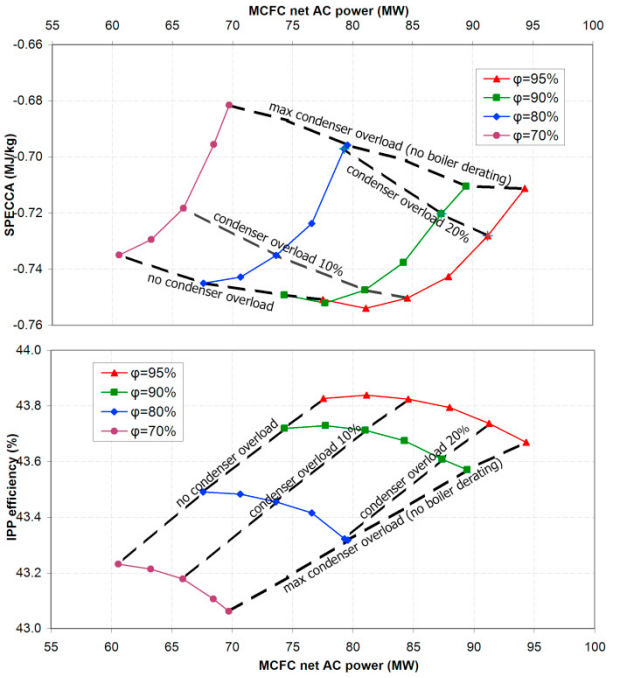


Figure 8. SPECCA index and IPP efficiency for the MCFC-DIR unit integrated with SPP

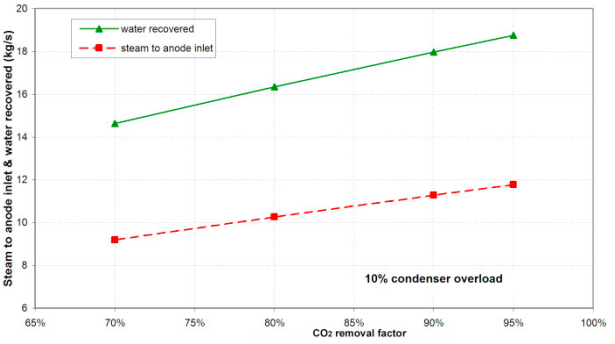


Figure 9. MCFC-IIR system: steam to anode inlet and water recovered in the carbon dioxide separation and storage section

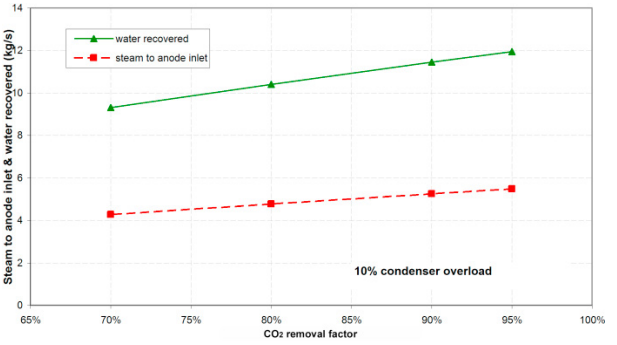


Figure 10. MCFC-DIR system: steam to anode inlet and water recovered in the carbon dioxide separation and storage section

4. Conclusion

In this work, a theoretical activity has been done on the use of a MCFC as an active device for electricity generation and carbon dioxide capture. A physical model of the MCFC has been developed and applied to the design and characterization of the fuel cell, when it is integrated in a coal fired steam power plant. This has been done feeding the MCFC's cathode with the exhaust gas of a coal fired power plant and recovering the exceeding heat from the MCFC system to partially preheat the feed water in the coal fired steam power plant.

A complex system has been conceived around the MCFC: fresh air is needed to improve O_2 concentration at the cathode inlet and move CO_2 to anode where it is mixed with H_2O and easily separated. H_2 and steam are required to feed the anode, being the H_2 the result of a CH_4 steam reforming. The steam is produced thanks to the unreacted fuel gases out from the anode which are burned in a combustor. This energy exceeds what is required by the steam needed for the CH_4 reforming: a further heat recovery is, therefore, possible on the feedwater in the regenerative line of the coal fired power plant representing a novel integration of the MCFC system.

This integration has a great impact in the single SPP plant, improving its efficiency from 41% up to 45.3%. In particular, performance is higher when CO_2 removal factor of the MCFC is higher. This is mainly due to the thermal recovery done on the feedwater: if the boiler was not derated (i.e. lower steam mass flow rate), the condenser would be overloaded. A suitable value of 15% for the condenser overload has been identified. MCFC resulted electrical power is in the range of 60-95 MW, which is surely a value worthy of interest.

Finally, considering the whole IPP, overall energetic efficiency is always higher than the original SPP: it ranges from 41.9 to 42.35% with a MCFC-IIR and it reaches 43.8% with a MCFC-DIR. Environmental performance is also very promising: negative SPECCA has been always achieved, with higher absolute value for the MCFC-DIR case.

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