

Molten Carbonate Fuel Cells in Integrated Systems for the Exploitation of Poor Fuels and the Segregation of CO₂

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A multi-scale approach for the analysis of reactant systems is a very important mean for the improvement of the understanding of the phenomena occurring at different scales of space and time, and for the support it brings to the experimental and design activities in particular for scale-up purposes. In previous works, the authors developed kinetics, cell and stack models for molten carbonate fuel cells, integrating all these scales of simulation in a code called SIMFC, which has been successfully validated on experimental data. The aim of the present work is to show the benefits that the coupling of plant simulation and small scale models can bring to the study of innovative applications and the design of particular configurations. In the frame of this paper, the authors analysed the integration of the calcium-looping technology coupled with molten carbonate fuel cells for the capture and sequestration of carbon dioxide using a series of different sources of fuel gas to evaluate the best options. In particular, this work analyses the exploitation of poor fuels in fuel cell that would not be possible without enrichment. The availability of a small scale model allowed to identify critical operating conditions (of both local temperature and composition) that decrease the performance and that cannot be easily identified with the use of only large scale model commonly employed for plant design and simulations.

Key words: Molten carbonate fuel cell, calcium-looping, plant simulation, CO₂ sequestration

INTRODUCTION

The emissions of CO₂ in the atmosphere, especially due to energy production, are a major concern due to the effects that this greenhouse gas has on the climate. Carbon Capture, Utilization and Storage (CCU and CCS) are regarded as the main solution to mitigate these effects [1]. CCU and CCS consist of a number of technologies that can be used to separate and collect the CO₂ generated in industrial processes for its reuse in other chemical plants or for its storage in underground facilities to prevent emissions into the atmosphere.

Molten carbonate fuel cells (MCFCs) are a valid example of a technology that can be used for carbon capture thanks to its operating principles [2]. MCFCs are deeply studied by Electrochemical Impedance Spectroscopy [3,4], that is a valuable tool for the investigation of reactions and phenomena taking place in different materials [5-10]. In literature, different studies already exist concerning its application as mean for carbon capture [11-13]. Investigations on MCFCs vary

from studies to improve the used materials [14-21], to reduce the pollutant effects and to increase their resistance [22-26] and to realize robust computer simulations [27-29] in particular for integrated systems [12, 30, 31].

Computer simulation can be a useful instrument to support experimental and design activities. In particular, a multi-scale approach that integrates the modelling from the small to the large scale can highly improve the scale-up process.

In previous works, the authors developed a kinetics formulation to simulate the performance of MCFC [24, 32, 33]. This formulation has been integrated into a Fortran code coupled with a cell level simulation to accurately consider mass, energy and momentum balances. Through a series of experimental campaigns, the model has been tested and proved reliable for the simulation of the MCFC behaviour. In this work, the aim is to show the benefit of the integration of a local model into the overall plant simulation.

For this activity, an MCFC stack was integrated after a calcium-looping process for carbon capture and sequestration.

This integration is interesting because it allows the use of poor fuels in MCFC. In fact, for particular fuels such as the exhaust of a gas furnace

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or the product of the gasification of waste, the amount of fuel and the high content of CO₂ induce instability in the cell. If the calcium looping process is integrated upstream the cell, it removes CO₂, enriching the fuel. Moreover, performing the regeneration of the sorbent in the calcium looping

with air and not pure O₂, allows significant cost reduction and removal of CO₂ in the cell. In this way the two technologies brings mutual benefit [34]. A simple scheme that shows the integration of these two technologies is represented in figure 1.

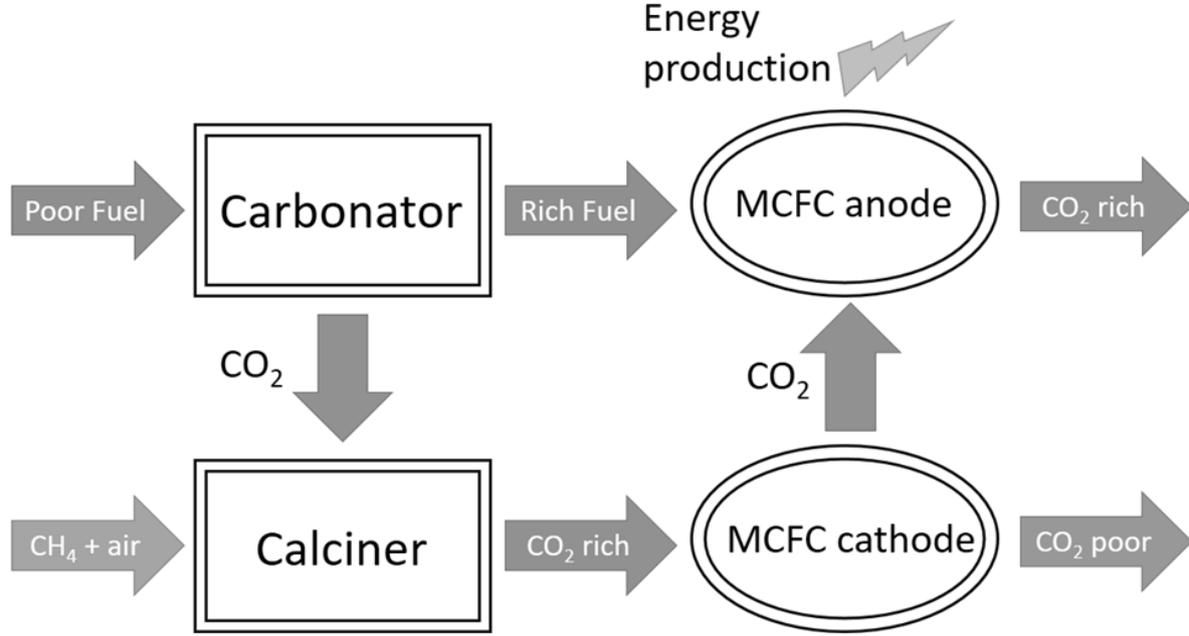
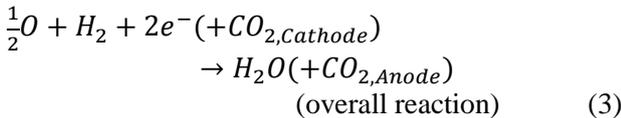
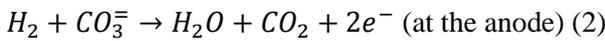
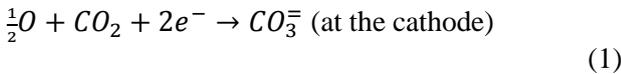


Fig. 1. Schematic representation of the integration between calcium looping process and MCFC.

MOLTEN CARBONATE FUEL CELL & SIMFC CODE

A molten carbonate fuel cell is an electrochemical device that directly converts the chemical energy presents in fuel to electrical energy. The electrochemical reactions that allow this conversion are the following:



An important characteristic of MCFC that can be seen from the overall equation (eq. 3) is that the CO₂ at the cathode moves to the anode side where is released. This process allows the depletion of CO₂ from the cathode feed and the subsequent enrichment of the anode stream. At the end of the process, ideally, the cathode outlet will have a low content or none CO₂, while the anode one will consist of a mixture of CO₂ and water that can be

separated with less energy consumption compared to the CO₂ and N₂ mixture of the cathode feed. For this reason and for the surplus energy that they produce, MCFCs are deemed as a valid instrument for carbon capture and segregation.

The performance of an MCFC can be expressed through the following equation:

$$\Delta V = \Delta E - J R_{Tot} \quad (4)$$

where ΔV is the measurable voltage (V), ΔE the voltage at the thermodynamic equilibrium given by the Nernst equation (V), J the total current density (A m⁻²) and R_{TOT} the overall cell resistance (Ω m²) which includes all the voltage losses of the cell which are Ohmic, concentration and activation losses.

In the model developed by the authors [24, 32, 33, 35] and named SIMFC, the resistance term has been expressed as:

$$\begin{aligned} R_{Tot} = P_1 e^{\frac{P_2}{T}} + \frac{P_3 T e^{\frac{P_4}{T}}}{p \ln \left[1 - \frac{1.5}{1+\delta} (y_{CO_2} + y_{H_2O}) \right]^{-1}} + \\ P_5 T e^{\frac{P_6}{T}} p_{CO_2}^{0.5} p_{O_2}^{-0.75} + \frac{P_7 T e^{\frac{P_8}{T}}}{p \ln(1+y_{H_2})} \end{aligned} \quad (5)$$

where T is the operating temperature (K), p is the operating pressure (atm), P_i s and ϑ are empirical parameters, p_i s are the partial pressure of the i -th gas (atm) and y_{H_2} is the molar fraction of H_2 . In the formulation, each addend represents a particular aspect of the resistance: the first is the ohmic term, while the others are the polarization resistances due to respectively CO_2 , O_2 and H_2 .

The model has been tested with different operating conditions (temperature and composition) and electrode materials (with opportune parameters re-evaluation), and showed always a satisfying fit [24,32,33].

The SIMFC code uses equations 4 and 5 to describe the performance of an MCFC and is completed with the addition of energy, mass and momentum balances. Through the code it is possible to obtain good fitting results of a single cell or a stack performance, composition and temperature maps and other kind of electrochemical information.

This model can be helpful if integrated in a plant simulation because it can provide information about parameters that usually plant scale design models ignore, such as local compositions, current and temperatures.

CALCIUM-LOOPING

The calcium looping is a carbon capture technology that is based on the reversibility of the lime carbonation [36]. The process can be divided into two main phases: carbonation and calcination.

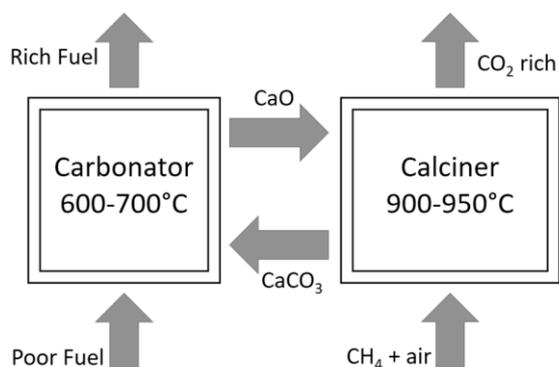
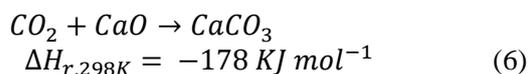


Fig. 2. Calcium looping process scheme.

The carbonation consist in an exothermic step where a flue gas containing CO_2 enters a carbonator reactor where the CO_2 reacts with CaO to form $CaCO_3$ (usually at an operating temperature between 870 and 970 K), according to the following reaction [37].



Through this first phase the feeding is depleted of CO_2 and can be used for different scopes as gas with a higher heating value.

Subsequently, the formed carbonate is sent to a calciner where, at temperature higher than 1170 K, it dissociate to CO_2 and CaO . The gaseous CO_2 can be collected, while the solid can be recycled to the carbonator reactor. This step is endothermic and heat is usually provided with combustion with pure O_2 in order to obtain a highly concentrated stream of CO_2 [37]. This represents one of the main disadvantages of this process [38] due to the high cost of pure oxygen. A possible solution considers the use of air, but it would require a subsequent separation of N_2 and CO_2 for carbon sequestration. This separation can be carried out exploiting an MCFC where the CO_2 rich stream can be used as cathode feed while the previously enriched fuel can be used as the anode one.

DESCRIPTION OF THE SYSTEM

The studied system consists in the integration of the technologies of calcium looping and MCFC for the segregation of carbon dioxide and the exploitation of poor fuels. The development of the calcium looping process was based on the ZECOMIX high efficiency hydrogen power plant currently operating at the ENEA research centre Cassaccia (Italy) [39], where the process is positioned downstream of a gasifier for the decarbonisation of raw syngas.

A fuel gas reach in CO_2 and that cannot be directly used in MCFC is sent to the calcium looping system, after a treatment to remove possible poisonous gases as sulphur compounds (This aspect has not been treated in this work). In the carbonator the fuel is depleted of CO_2 thanks to reaction 6 and can be used as anode inlet for an MCFC. The $CaCO_3$ moves to the calciner where the thermal regeneration of the solid sorbent occurs. Combustion of methane in air is utilized to ensure an adequate temperature rise to facilitate the regeneration. The integration of the MCFC downstream allows for methane combustion in the presence of air instead of pure O_2 , reducing the energy and cost penalty associated with producing pure oxygen in an air separation unit. The regenerated CaO is recycled back to the carbonator while the CO_2 rich gas is used as the cathode inlet to the MCFC, allowing for further CO_2 concentration and the generation of useable electrical power. The utilization of the decarbonized syngas for electricity generation in an MCFC is preferable due to the higher electrical efficiencies observed in MCFCs

compared to the commonly utilized gas turbines. At the end of the process the enriched CO₂ should undergo a further separation to remove steam or other impurities. In this work three different possibilities have been investigated: simple condensation, burner and condensation to remove

possible unreacted O₂, and membranes [40, 41]. A scheme of the proposed solution with the integration of the burner and condensation separation process as used in the simulation in Aspen is presented in figure 3.

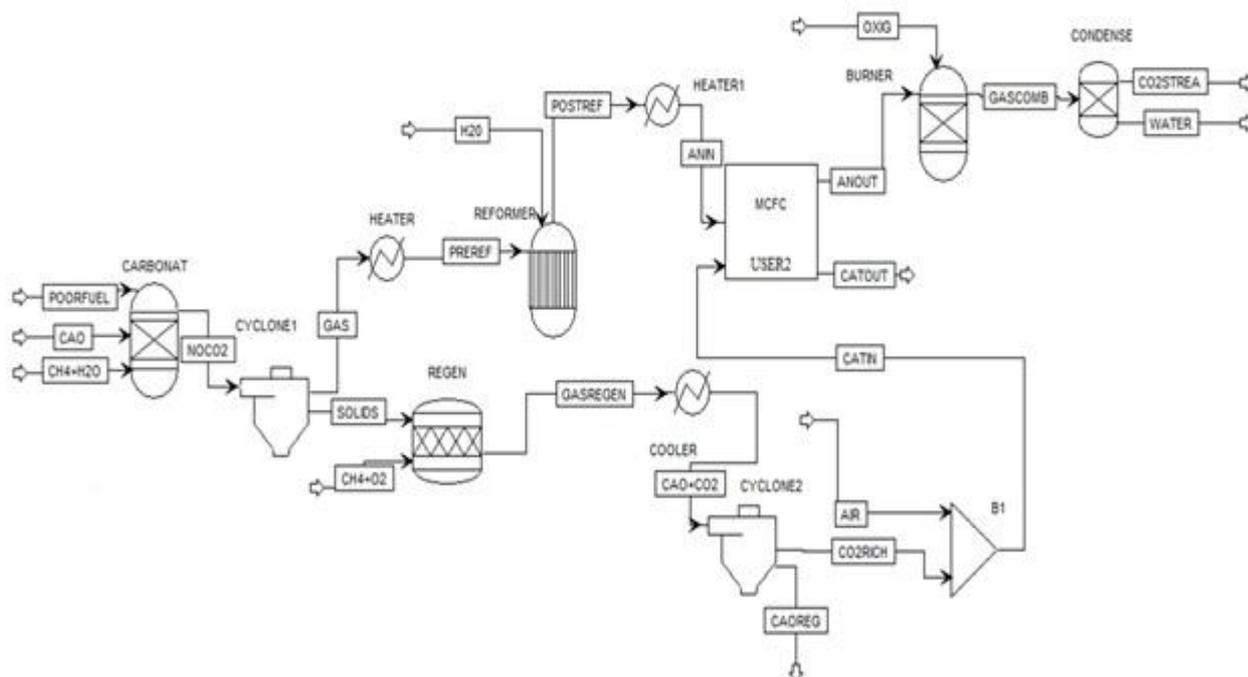


Fig. 3. Aspen representation of the calcium looping and MCFC integration.

The following assumptions have been made for the simulation of the process:

1. The removal of pollutants happens before the beginning of the studied process.
2. The carbonator works at 973 K. The carbonation reaction is at the thermodynamic equilibrium, while the conversion of CH₄ is fixed at 80% as literature suggests [42]. CH₄ and CaO feed flow rates are optimized as function of the operating conditions.
3. The regeneration works at 1173 K at the calciner. The regeneration reaction is at the thermodynamic equilibrium, while the conversion of CH₄ is total. The amount of the “CH₄+O₂” stream has been calculated in order to ensure an adiabatic reaction.
4. The solid phases after the carbonator and the regenerator are completely separated from the gas phases in downstream cyclones.
5. A reformer is placed before the anode inlet enters the cell to convert all the CH₄ in H₂. Prior to this reaction the stream is preheated up to 1123 K. Also, steam is added to increase the content up to 10 mol% since it is necessary to avoid carbon deposition.

6. The cathode inlet stream is mixed with air to ensure an amount of O₂ equal to 20% and the thermal management of the cell.

7. In the cell the fuel utilization factor (at the anode) is assumed to be 70%, while the CO₂ utilization factor (at the cathode) is assumed to be 90%.

8. The cells of the stack are considered to have an area of 1 m² each working with a current density of 1000 A cm⁻².

9. The performance of the cell is calculated on the basis of the local SIMFC model.

10. At the anode outlet a burner is used for the combustion of excess H₂ to obtain only CO₂ as gas outlet and water which is separated thanks to a condensation. The O₂ flow rate is optimized as a function of the operating conditions.

11. In case a membrane is considered downstream, it is assumed that the CO₂ removal efficiency is of 90% and that it produces a 95% pure CO₂ stream [43].

For this analysis, three different feeds have been studied to describe three different possible applications: blast furnace exhaust [44], gasified waste [45] and syngas

Table 1. Compositions (in molar fractions) of the three kinds of fuel gas used for the simulation.

Type of fuel	H ₂	CO	CO ₂	H ₂ O	N ₂	H ₂ S	HCN	NH ₃
Blast furnace exhaust	0.048	0.228	0.190	0.062	0.472	1.97 10 ⁻⁵	1.66 10 ⁻⁴	1.32 10 ⁻⁵
Gasified waste	0.292	0.425	0.161	0.068	0.054	0.0036	9.95 10 ⁻⁵	0.0025
Syngas	0.282	0.350	0.139	0.198	0.031	0.0068	-	0.0015

SIMULATION RESULTS

The capture of CO₂

The main interest of this integrated application is its applicability to the capture of CO₂ to reduce emissions in the atmosphere and the simultaneous exploitation of poor fuels. For this reason the main results presented will be focused on the flow rate and the molar fraction of CO₂ released and captured during the process as well as the energy efficiency.

Before showing the results of the integrated process, the results of the single units (fuel cell and calcium looping separately) should be presented for comparison. However, since poor gases are considered, they cannot be used directly in a fuel cell stack efficiently, and therefore only the calcium looping results are shown in table 2. The use of the calcium looping allows to ideally obtain a pure stream of CO₂, but the emissions in atmosphere still contain CO₂ in the order of 5 mol%.

In table 3 and 4, the main results concerning the CO₂ captured and released in atmosphere of the simulation for the integrated process are presented for the three different types of fuel gas with three different methods for the final downstream separation.

Table 2. Main results concerning the CO₂ captured and released in atmosphere for the simulation of the calcium looping process only.

Type of fuel	CO ₂ stream	Molar fraction	CO ₂ Flowrate/ CO ₂ Flowrate in the fuel	CO ₂ Flowrate/ MCFC Power Output (kmol_CO ₂ MW ⁻¹)
Blast furnace	CO ₂ released	0.060	0.288	6.635
	CO ₂ captured	1.000	1.208	27.830
Gasified waste	CO ₂ released	0.034	0.183	1.680
	CO ₂ captured	1.000	0.979	8.980
Syngas	CO ₂ released	0.050	0.344	2.628
	CO ₂ captured	1.000	1.785	13.618

In table 3, the column “CO₂ molar fraction” represents the values in the final enriched stream of CO₂ after the calcium looping process, the fuel cell operation (anode outlet) and one of the three processes of separation considered. In all the analysed cases, it appears clear that the use of the condensation only is not sufficient to obtain a stream of CO₂ suitable for sequestration or other possible uses since it is still rich in other compounds (mainly N₂ and unreacted H₂). The addition of a burner will favour the process, but the best solutions is to add a further separation by means of membranes. This kind of separation does not only allow to obtain a stream very rich in CO₂, but also to reduce the final flow rate.

Table 4 refers to the sum of all CO₂ presents in all the streams that are released into the atmosphere during the overall process. In particular the column “CO₂ molar fraction” indicates the molar fraction of CO₂ if all the emissions were grouped in one stream. As expected, for all the three fuel gas cases, the lowest value of CO₂ emission is obtained when membrane separation is used. In particular it appears that there is no actual difference between the use of only condensation and burner coupled with condensation.

Table 3. Simulation results for the three studied types of fuel gas.

Type of Fuel	Downstream separation	CO ₂ Molar fraction	Flowrate/ CO ₂ Flowrate in the fuel	Flowrate/ MCFC Power Output (kmol MW ⁻¹)
Blast furnace exhaust	With Condensation Only	0.351	1.767	421
	Burner + Condensation	0.444	1.985	27.446
	CO ₂ Selective Membrane	0.950	1.590	21.979
	With Condensation Only	0.647	3.166	24.872
Gasified waste	Burner + Condensation	0.909	3.554	27.916
	CO ₂ Selective Membrane	0.950	2.850	22.385
	With Condensation Only	0.687	3.510	28.048
	Burner + Condensation	0.941	3.886	31.052
Syngas	CO ₂ Selective Membrane	0.950	3.159	25.243

Table 4. Simulation results for the three studied types of gas concerning the captured CO₂.

Type of Fuel	Downstream Separation	CO ₂ Molar Fraction	Flowrate/ CO ₂ Flowrate in Fuel	Flowrate / MCFC Power Output (Kmol MWh ⁻¹)
Blast furnace exhaust	With Condensation Only	0.005	0.202	2.791
	Burner + Condensation	0.005	0.202	2.791
	CO ₂ Selective Membrane	0.0009	0.379	5.233
	With Condensation Only	0.005	0.360	2.826
Gasified waste	Burner + Condensation	0.005	0.360	2.826
	CO ₂ Selective Membrane	0.0011	0.676	5.313
	With Condensation Only	0.005	0.355	2.834
	Burner + Condensation	0.005	0.355	2.834
Syngas	CO ₂ Selective Membrane	0.0010	0.706	5.639

Finally in table 5 the global efficiency of the integrated solutions in terms of electrical energy and CO₂ capture are reported for the three type of analysed fuels. For all the gases the efficiencies are similar and in particular the segregation values reached are significantly high.

Compared to the calcium looping only solution, it is clear that the molar fraction of CO₂ emitted

into the atmosphere is much lower, allowing larger savings on CO₂ emissions tax.

On the basis of this analysis it is possible to say that the burner coupled with a condensation step and the membrane separation are the best scenario; the difference between the two would require a further economic analysis which is beyond the scope of this work.

Table 5. Electrical energy and CO₂ segregation global efficiency of the integrated solution.

Type of Fuel	Electrical energy efficiency	CO ₂ segregation efficiency
Blast furnace exhaust	0.287	0.933
Gasified waste	0.286	0.922
Syngas	0.285	0.922

The benefits of a small scale model

An important aspect of a multi-scale approach is the possibility to rely on low level analysis to verify the reliability of larger scale processes. In particular the use of both a detailed kinetics model and a cell level model allows to verify the stability of each element of the MCFC stack in each point in the possible operating conditions.

In figure 4 the maps that describe the molar fraction of CO₂ on the cell plane at the anode and cathode side are presented. Relying on these type of maps can be useful to determine the best operating

conditions. For example, they can help determine whether it is necessary to increase the surface and therefore increase the capture rate or decrease the surface because the cell is working in very unstable conditions (in this case the analysis of the H₂ or O₂ maps is also necessary).

In figure 5 the maps that represent the temperature of the solid structure (a), the cathode gas (b) and the anode gas (c) are presented. The use of these maps is important to verify that there are no hotspots that can compromise the cell performance and stability in any point of the cell.

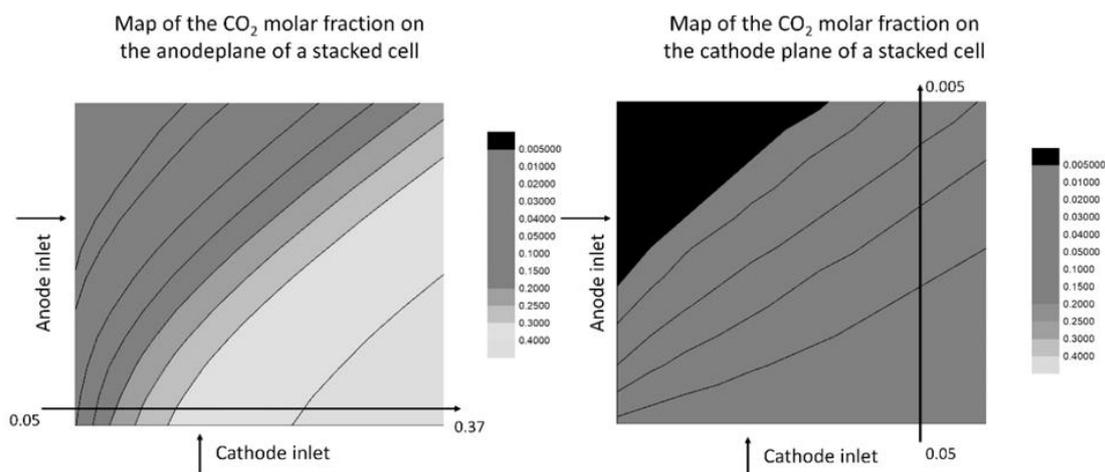


Fig. 4. Example of maps of the CO₂ molar fraction on a cell plane at the anode and cathode side of the gas deriving from the treatment of the blast furnace exhaust with the cell operating at operating at 1000 A m⁻²

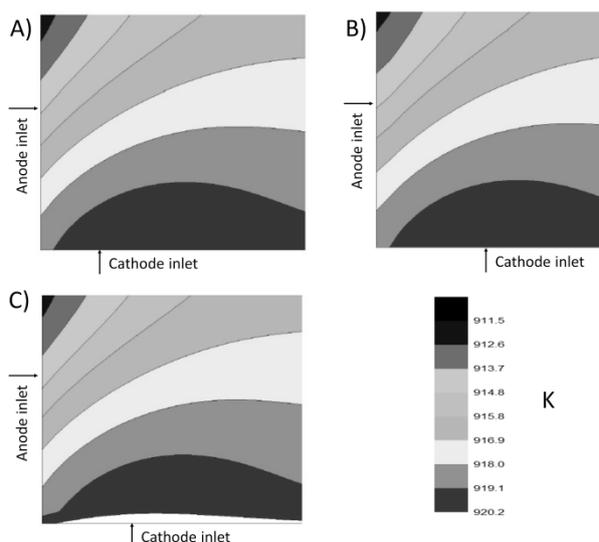


Fig. 5. Map of the temperature on a cell plane of the solid structure (A), the anode (B) and the cathode (C) gas of a cell operating at 1000 A m^{-2} .

CONCLUSION

The aim of this work is to show the benefits that the coupling of plant simulation and small scale models can bring to the design of plants and the study of particular configurations. In the frame of this work, the authors analysed the integration of the calcium-looping technology coupled with MCFCs for the capture and sequestration of CO_2 and the exploitation of a series of different sources of poor fuel gases.

The availability of a small scale model allows the identification of critical operating conditions (of both local temperature and composition) that can decrease the performance and that cannot be easily identified with the use of only large scale model that are mainly used for plant design and simulations.

A further feasibility analysis will be carried out taking into account the detailed plant battery limits and economical considerations in order to evaluate the benefit of the proposed system integration as a function of specific applications.

Symbols

ΔE	cell reversible potential, V
ΔH_{reaz}	enthalpy of reaction, kJ mol^{-1}
J	cell current density, A m^{-2}
p	pressure, atm
p_i	partial pressure of the i -th gas, atm
P_y	empirical parameters, variable
R_{TOT}	total cell resistance, $\Omega \text{ m}^{-2}$
T	temperature, K
ΔV	cell measurable voltage, V
y_i	molar fraction of the i -th gas
ϑ	empirical parameter

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Горивни клетки със стопен карбонат в интегрирани системи с използване на бедни горива и сегрегация на CO₂

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(Резюме)

Многомащабния подход за анализ на реагиращите системи е много важно средство за подобряване на разбирането за явленията, настъпили в различни мащаби на пространството и времето, както и за подпомагане на експерименталното и дизайнерското решение, специално при увеличение на мащаба. В предишни трудове авторите разработват кинетичен модел на клетката и на стека при горивни клетки със стопен карбонат, интегриращи всички тези мащаби на симулация в код, наречен СИМФК, който е успешно валидиран на базата на експериментални данни. Целта на настоящата работа е да покаже ползите, които могат да донесат свързването на симулацията на централи и на по-малко мащабни модели при изучаването на иновативни приложения и проектиране на определени конфигурации. В рамките на тази статия авторите анализират интеграцията на технологията Регенериращ цикъл на калций успоредно с горивни клетки със стопен карбонат за улавяне и отвеждане на въглероден двуокис, използвайки серия от различни източници на горивен газ за определяне на най-добрите варианти. По-специално, този труд прави анализ на употребата на бедни горива в горивните клетки, което не би било възможно без тяхното обогатяване. Наличието на малко-мащабни модели позволява да се идентифицират критичните работни условия (както на локалната температура, така и на състава), които понижават експлоатационните качества и които не могат да бъдат лесно идентифицирани с използването само на широкомащабен модел, използван обикновено при симулации и проектиране и на централи.