

Flowsheet calculation of a combined heat and power fuel cell plant with a conceptual molten carbonate fuel cell with separate CO₂ supply

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Abstract

A new type of MCFC with a separate CO₂ supply (improved or i-MCFC) is previously presented, which has the potential for reducing NiO cathode dissolution and system enhancement by CO₂ removal from fuel gas. This article presents the first flowsheet calculations of an i-MCFC system that utilizes the potential of reducing NiO dissolution. A submodel that simulates energy and massflows of the i-MCFC is built using standard flowsheeting components. The performance of the i-MCFC is assumed to be equal to the MCFC and differences in Nernst potentials and irreversible losses are neglected. To compare the differences in concept, a MCFC combined heat and power (CHP) system flowsheet is modified and the MCFC model substituted by the i-MCFC submodel. The overall efficiencies of both fuel cell systems are calculated using a flowsheeting program. The calculated results are compared and the differences analyzed. The overall system performance of this i-MCFC CHP system is slightly lower than the MCFC CHP reference system (about 0.1% point in average). The difference in performance is ascribed to the change in gas composition and heat capacity of the cathode gas. The change in heat capacity increases the total massflow through the i-MCFC resulting to an increase in overall auxiliary power consumption. The low CO₂ content of the cathode gas should reduce the NiO cathode dissolution to a negligible level.

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Keywords: Fuel cell; MCFC; Cathode; System modeling; Flowsheet; Design

1. Introduction

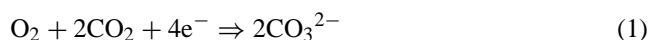
The improved molten carbonate fuel cell or i-MCFC [1] is a new fuel cell design that is developed at the Faculty of Applied Science of Delft University of Technology. In contrast to conventional fuel cells that features two gas inlets and outlets, the i-MCFC features three separate gas inlets and outlets, which is shown by Fig. 1. Although this new concept has only been demonstrated on laboratory scale, it is worthwhile studying the benefits first by flowsheet simulations before starting an expensive development program on the i-MCFC. This paper presents the first study of system concept by flowsheet calculations in which the i-MCFC is implemented. Using standard flowsheeting components, a submodel is built that simulates the energy and mass balance of the i-MCFC. This submodel is implemented in a 250 kW class conventional MCFC combined heat and power

(CHP) plant by modifying the fuel cell's inputs and outputs to match the requirements of the i-MCFCs. The CO₂ that is needed for the electrochemical reaction of the cathode is here supplied solely by the matrix flow. The cathode is fed with air having low CO₂ content (about 0.03 mol%). This low CO₂ content should sufficiently suppress cathode dissolution that at present severely limits endurance of the MCFC. The change in the overall system performance is assessed by flowsheet calculations using the flowsheeting program Cycle-Tempo [2].

2. Theory

2.1. Principle

At the MCFC's cathode, O₂ molecules react electrochemically with CO₂ molecules to form a CO₃²⁻ ion:



There is a general agreement that this reaction can be divided into three sequential reaction steps. Using the peroxide

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Nomenclature

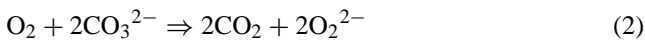
A_{cell}	active cell area (m^2)
F	Faraday's constant (C/mol)
$h_{\text{dc-ac}}$	dc to ac inverter efficiency
i_{cell}	fuel cell current density (A/m^2)
n	number of electrons involved in the cell half-reaction
p	pressure (bar)
p_i	partial pressure of specie i (bar)
Δp	pressure loss (bar)
P	power delivered (kW)
Q	heat transfer (kW)
T	temperature ($^{\circ}\text{C}$)
T_{cell}	fuel cell temperature ($^{\circ}\text{C}$)
ΔT_{low}	low end temperature difference of heat exchanger (K)
u_f	total fuel utilization
V_{cell}	cell voltage (V)

Greek letters

η_i	intrinsic efficiency
η_{EX}	exergy efficiency based exergy of fuel input and $T_0 = 25^{\circ}\text{C}$
η_{th}	thermal efficiency based on lower heating value of fuel input
Φ	massflow (kg/s)

mechanism¹ [3] as an example, we can distinguish these following steps (see Fig. 2).

Step 1. Chemical dissolution of O_2 molecules:



Step 2. Reduction of O_2^{2-} peroxide ion:



Step 3. Recombination reaction:



The first step releases CO_2 while the third step consumes CO_2 . From that, we can see that step 1 is favored by a low partial pressure of CO_2 (p_{CO_2}) of the cathode gas or oxidant while step 3 is favored by a high p_{CO_2} . These two conflicting demands can be satisfied if steps 1 and 3 can take place at different locations. The principle of the i-MCFC is based on the assumption that steps 1 and 3 will take place at different locations inside the cell, if the O_2 and CO_2 are supplied at different locations. Both reaction steps can then be enhanced

¹ The peroxide mechanism is named to the peroxide ion which is assumed to be an intermediate specie. Others propose the superoxide as the other possible intermediate specie. Many years of study on the reaction mechanism has not let to a generally accepted mechanism but the principle of i-MCFC is valid for both species.

by choosing the p_{CO_2} accordingly. In the i-MCFC, the O_2 is supplied with the cathode gas and step 1 can take place at the gas electrolyte interface. CO_2 is supplied by an additional channel that is made in the matrix support tile that contains the molten carbonate electrolyte (see Fig. 1). Step 3 can take place anywhere between this additional matrix channel and the cathode where the oxide ions are produced. Both gasses are now supplied separately and their concentration can be chosen individually to meet the conflicting demands of p_{CO_2} .

2.2. Advantages and disadvantages of the i-MCFC

Starting with the disadvantages, we note that making a gas channel in the matrix tile is very difficult. Present MCFC already suffers from a low mechanical stability of the matrix while the i-MCFC is tested in laboratory scale only [1,4]. In addition, increased effects of diffusion polarization are expected due to the diffusion distance that the O^{2-} and CO_2 need to overcome.

The advantages of the i-MCFC are based on the extra degree of freedom in the choice of gas compositions of the cathode gas. Since CO_2 is supplied separately, the typical restriction in a minimum of CO_2 concentration of the cathode gas is no longer required. Intricate connections between the anode output and cathode input are no longer required giving additional degrees of freedom in system design.

Removing the CO_2 restriction opens a unique opportunity to bring down the dissolution of the porous NiO cathode electrode used in present MCFC. The NiO cathode dissolution severely limits the endurance of the MCFC [5] and this problem needs to be solved in order to make the MCFC competitive to other fuel cell types and other power production technologies [6]. The importance of this problem can be seen from the numerous research projects that are presently taking place all over the world. [7–12]. Present research projects focus on finding alternative materials or material enhancements for the MCFC cathode. The i-MCFC concept however creates a different and unique opportunity to lower the NiO-cathode dissolution by changing cathode gas composition. The NiO dissolution process depends strongly on the p_{CO_2} and it shows a minimum at p_{CO_2} of around 10^{-2} to 10^{-3} bar with a rapid increase with increasing p_{CO_2} [13–15]. It is not possible to operate the MCFC at this minimum due to the electrochemical cathode reaction that requires CO_2 . Hence, alternative cathode material is needed in order to improve endurance of the MCFC [7–12]. On the other hand, the i-MCFC with its separate CO_2 supply does not require CO_2 in the cathode gas. Hence, in the i-MCFC, NiO cathode dissolution can be reduced to a negligible level when low CO_2 containing cathode gas (e.g. air) is used. Therefore, the i-MCFC creates a unique possibility to reduce this NiO cathode dissolution to enhance endurance and thus reducing operating costs of the fuel cell power plants.

Next to these opportunities in enhancing endurance, the extra degree of freedom in the choice of gas compositions

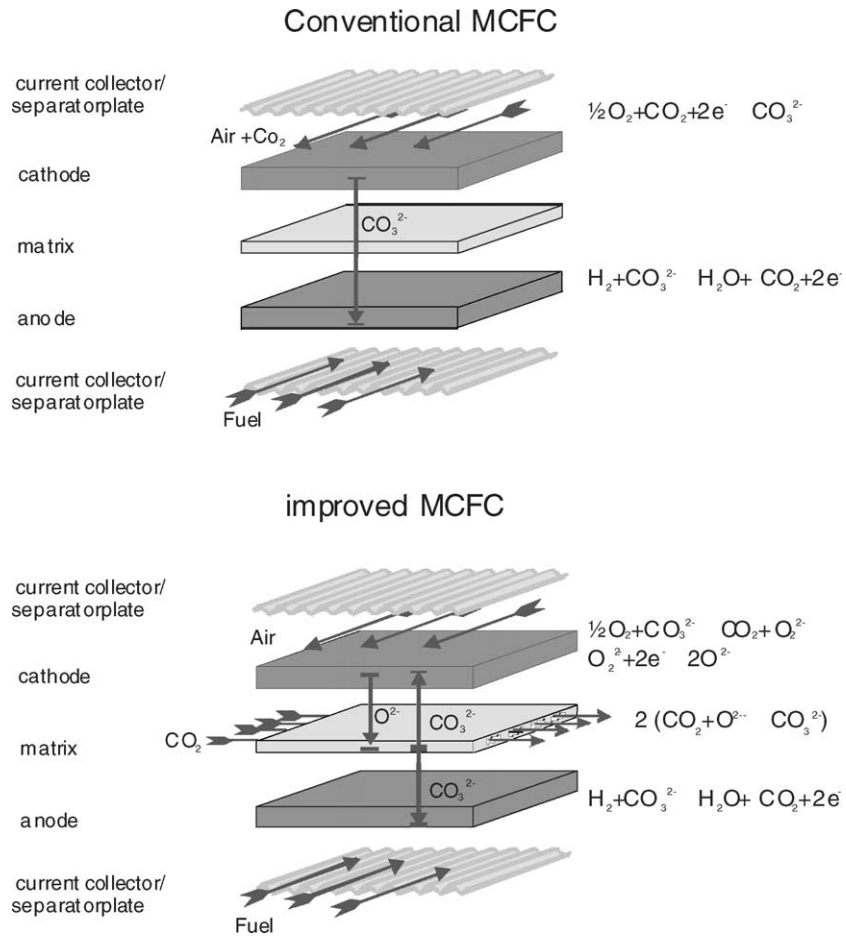


Fig. 1. Schematic representations of the conventional MCFC and the improved MCFC.

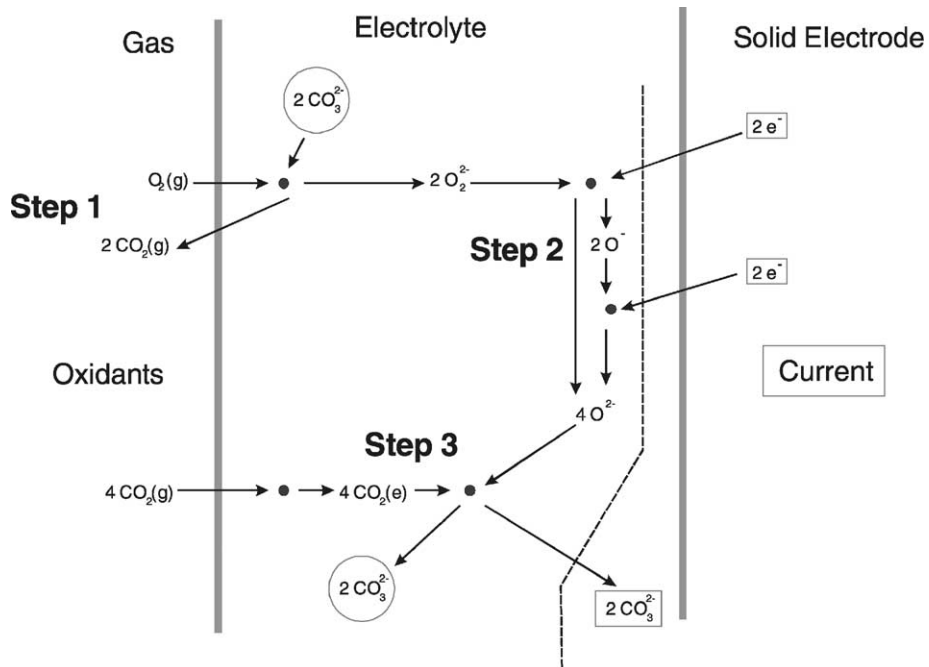


Fig. 2. MCFC peroxide cathode mechanism, taken from [3].

creates opportunities to reduce cathode polarization, which is the main cause of the MCFCs irreversible losses. For example, heated air can be fed directly to the cathode and its high O₂ and low CO₂ concentration enhances step 1 of the cathode reaction mechanism. Concentrated CO₂ gas can be supplied to the matrix thus enhancing step 3 of the reaction mechanism.

Another opportunity for the i-MCFC is to utilize the active CO₂ separation process of the matrix gas stream. This feature can be used in new system designs: e.g. removing CO₂ from low caloric fuel gas (e.g. landfill gas or biogas) thus enriching the fuel gas prior feeding it to the anode. For this option, the fuel gas should contain sufficient CO₂, since each mol of H₂ that is converted in the anode requires the transport of an equal amount of CO₂ from the matrix stream. Hence, the H₂/CO₂ ratio of the fuel gas should be equal to or less than one. Landfill gas and biogas should be suited for this option since they both contains relatively large amount of CO₂. Reformed natural gas (containing mostly methane) on the other hand is not suited due to its low CO₂ content relatively to the H₂ content. Reforming methane produces ideally three H₂ molecules together with only one CO₂ molecule, hence, the H₂/CO₂ ratio is too high and therefore methane is not suited as fuel for this specific system configuration.

2.3. Modeling the i-MCFC

Studying the influence of the i-MCFC concept on system level requires models that fully describe the performance and energy and massflows. They can be distinguished by two submodels:

1. A submodel that describes the cell voltage of the i-MCFC at various load conditions.
2. A submodel that calculates the energy and mass balance of the i-MCFC.

At present, we are still investigating the i-MCFC mechanism in detail but already the basic thermodynamics felt short in defining the Nernst voltage of the cell. With the present knowledge, it is not possible yet to build a model that uses electrochemical kinetics to determine the cell performance at load conditions. We therefore need to rely on experimental results in order to estimate the cell performance of the i-MCFC. Peelen et. al. [4] showed that the performance of the i-MCFC is comparable to the conventional MCFC. Hence, the present study assumes the same performance for both the i-MCFC and the MCFC in order to study this new concept. This means that the cell voltage of a MCFC operating under similar condition is used here as the cell voltage of the i-MCFC. In other words, a MCFC's design point of operation under similar operating condition is used for the i-MCFC. The differences between the two types of cells in open cell voltage (OCV), Nernst loss and irreversible losses are therefore neglected. All possible differences in system performances are direct results of differences in system layout.

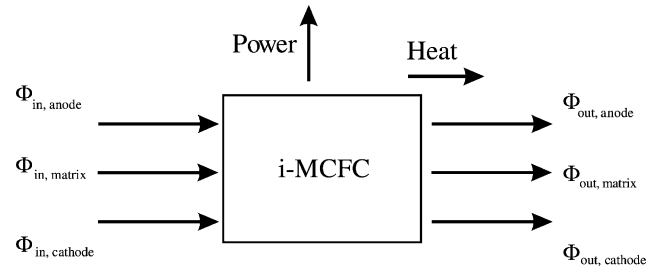
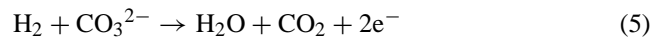


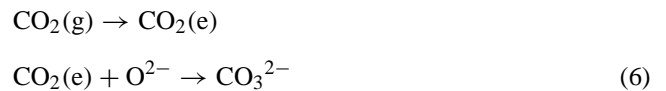
Fig. 3. Massflows and energy flows of the i-MCFC.

Once the performance of the i-MCFC is determined, this data is used as input value for the submodel that calculates the energy and mass balance of the i-MCFC as required for the flowsheet calculations. This submodel is built with the standard components of the flowsheeting program Cycle-Tempo [2]. This submodel simulates i-MCFC's process and energy flows, which are given by Fig. 3. The following three reactions take place inside the cell.

At the anode, the normal MCFC anode reaction takes place:



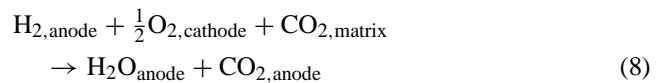
In the matrix, CO₂ dissolves into the electrolyte and reacts with O²⁻ ions:



At the cathode, oxygen is reduced to O²⁻ ions:



The overall reaction is the same as that of the MCFC: H₂ from the anode inlet massflow $\Phi_{\text{in,anode}}$ reacts with O₂ from the cathode inlet massflow $\Phi_{\text{in,cathode}}$ to form H₂O that is released by the anode outlet massflow $\Phi_{\text{out,anode}}$. The difference here is that CO₂ is transported from the matrix to the anode:



The energy that is released by this reaction is released partly as electric power and partly as heat that is used for heating-up the three massflows. The unique feature of the i-MCFC is that CO₂ is introduced by the matrix inlet massflow $\Phi_{\text{in,matrix}}$ and part of this CO₂ is transported to the anode and released by the anode outlet massflow $\Phi_{\text{out,anode}}$. The objective of the flowsheet submodel is to simulate the process and energy flows given by Fig. 3.

A submodel is built that simulates all the features of the i-MCFC (see Fig. 4). This model features:

- matrix stream with CO₂ separation that is transferred to the anode;

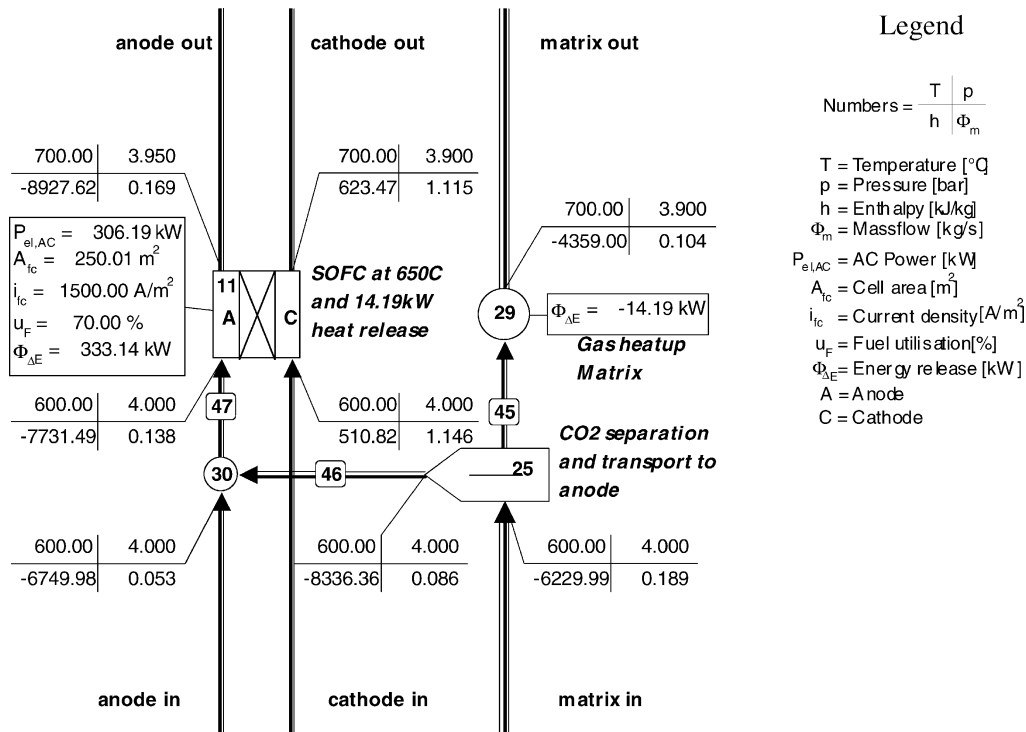


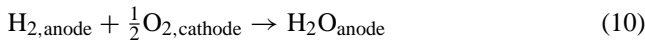
Fig. 4. i-MCFC energy and massflow model in Cycle-Tempo.

- O^{2-} ion conducting high temperature fuel cell simulated by the Cycle-Tempo model for the solid oxide fuel cell (SOFC) but operated at 650°C with additional heat bypass to matrix stream;
- some heat from the fuel cell is transferred to the matrix stream.

In this simulation of the i-MCFC, the CO_2 from the matrix stream is separated from the matrix stream and added to the anode inlet massflow (Fig. 4, apparatus #25, pipe #46 and node #30):



This separation process partly fulfills reaction (6) and the release of CO_2 in the anode as given by reaction (5). The electrochemical reaction from H_2 with $(1/2)\text{O}_2$ to H_2O and the transport processes are simulated by the SOFC model (apparatus #11):



It is clear that the i-MCFC overall reaction (reaction (8)) is obtained by combining reactions (9) and (10).

In this case, the SOFC model only calculates the enthalpy change² of the massflows, and with pre defined power re-

lease, it calculates the heat release that determines the cooling requirement of the cell. Part of the heat release is subtracted and transferred to the matrix stream in order to heat up this stream. This heat up process is simulated here by means of a heat source (apparatus #7 of Fig. 4) by defining an outlet temperature of 700°C . This standard flowsheeting apparatus calculates the amount of heat that is required for this process and the amount of heat is used as additional cooling or energy release defined for the fuel cell. In addition, apparatus 7 has the function of imposing pressure loss to the matrix channels, and this pressure loss is here assumed to be the same as the cathode pressure loss.

The molar flow of CO_2 (Φ_{CO_2}) from the matrix channel to the anode is given by Faraday's law:

$$\Phi_{\text{CO}_2} = \frac{A_{\text{cell}} i_{\text{cell}}}{nF} \quad (11)$$

where A_{cell} is the total cell area, i_{cell} the current density, n the number of charge (here 2), and F is the Faraday's number. Exactly, this molar flow of CO_2 is separated from the matrix stream and send to the anode inlet.

Note here that the CO_2 is fed to the anode inlet massflow prior to feeding it to the cell. This significantly changes the inlet gas composition, which in theory will alter the Nernst voltage and thus the performance of the cell. Nevertheless, this change of the inlet gas composition has no influence on the overall results since the performance of the cell (i.e. power output) is here an external given input and *not* calculated by means of the Nernst equation and the internal kinetical model [18] of the SOFC model.

² In principle, the SOFC model of the flowsheeting software also calculates a cell resistance based on internal kinetical model [18]. In this case, this calculation is not correct and has no practical value since it uses Nernst voltage calculation based on the SOFC electrode reactions, which are different from the i-MCFC electrode reactions.

The purpose of feeding the CO₂ to the inlet is to obtain chemical equilibrium at the outlet for the hydrogen shift reaction equilibrium:



At high temperature, equilibrium of this fast chemical reaction is assumed and this reaction is considered by the MCFC model of the references system. Changes in gas composition by this equilibrium influences the energy balance and cooling requirement of the fuel cell. By feeding the CO₂ at the anode inlet of the fuel cell, the SOFC model used in the i-MCFC model also considers this hydrogen shift equilibrium and compensates changes in the gas composition. It therefore calculates the composition at the anode outlet according to this equilibrium and adjusts the cooling requirement for the cell, as done by the MCFC model of the reference system.

In this way, all characteristics of the i-MCFC are simulated by the subsystem given by Fig. 4, with which the system calculations can be performed. Note again, that only the energy and massflows of the i-MCFC are simulated while the power output of the i-MCFC is defined based on currently available knowledge.

3. System calculations

3.1. Reference system and adjustments to match the i-MCFC

A MCFC CHP system is selected as a benchmark for this study of concept. This system has the following main features:

- 250 kW class fuel cell stack operating at 650 °C;
- waste heat utilization at two levels (steam at $T = 180^\circ\text{C}$ and hot water at $T = 80^\circ\text{C}$);
- natural gas as primary fuel (equivalent to 557.57 kW LHV);
- fuel gas is externally reformed;
- pressurized system operating at 4 bar.

The system is previously used for two other studies [16,17] and a detailed description of this system, including all input parameters, is given in [16].

Using this system as reference, the MCFC stack is substituted by an i-MCFC submodel as shown by Fig. 5. In the reference system, the required CO₂ for the cathode reaction is supplied by the flue gas of the heat exchange reformer

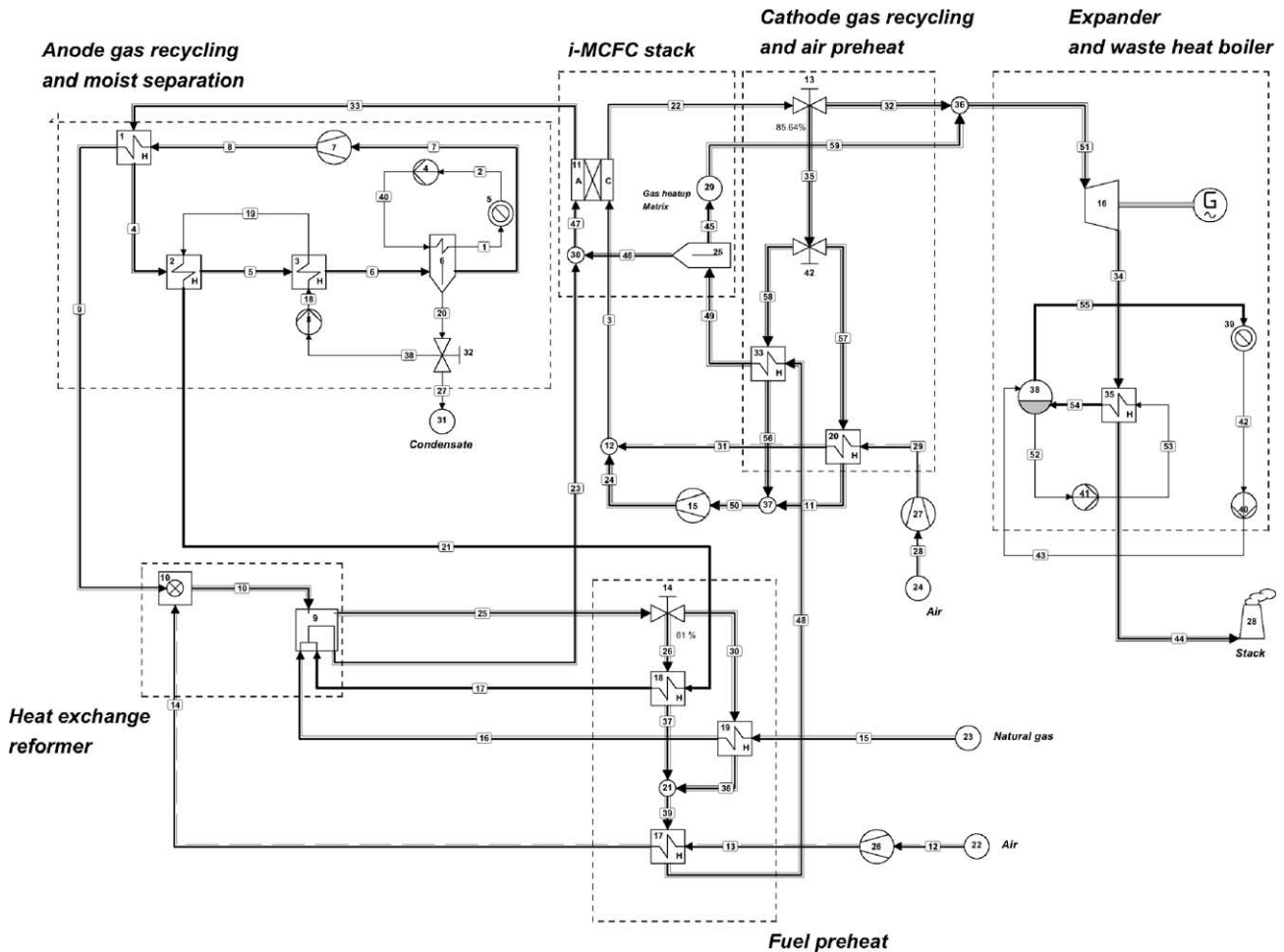


Fig. 5. Flowsheet of the 250 kW-class i-MCFC CHP plant.

(HER). This flue gas has the highest CO₂ concentration and therefore it is used here for feeding the matrix channel. Removing the connection to the cathode recycling results in a low CO₂ content of the cathode gas and should bring down the NiO cathode dissolution to a negligible level [13–15]. Before entering the matrix channel, the flue gas first goes to the fuel pre-heating stage (see Fig. 5) and the heat of the flue gas is used for preheating fuel and air for the HER. As a result, the flue gas is cooled to a temperature of about 430 °C and it needs to be heated-up again to the fuel cell entry temperature of 600 °C. The existing heat exchanger that is used for preheating air for the fuel cell's cathode is therefore extended to incorporate a heating stage for the matrix gas. This heat exchanger is modeled here by two parallel-connected heat exchangers. In practice, both heat exchangers can be integrated into a single unit, thus reducing investment costs. The matrix outlet is connected to the inlet of the expander. This expander recovers part of the exergy of this flue gas and part of cathode outgas. The following waste heat boiler recovers other residual heat.

3.2. Input parameters for the i-MCFC

The same input parameters of the reference MCFC stack are used here for the i-MCFC stack. The cell area (A_{cell}), fuel utilization (u_f) and current density i_{cell} are the same as the reference MCFC stack. Temperature difference between the inlets and outlets ($T_{\text{out}} - T_{\text{in}}$) are also set equal to the reference. Following the experimental results of Peelen et al. [4], the cell performance of the reference MCFC stack is used for the i-MCFC as well. In this flowsheeting program, this is done by defining the total power output ($P_{\text{el,ac}}$) and cell voltage (V_{cell}). With the current density as the other required input, the program calculates the required cell area, which obviously is here the same as the reference stack. Inverter efficiency ($\eta_{\text{dc-ac}}$) and pressure losses (Δp_i) are also defined according to the reference system. Gas compositions and massflows are calculated by the program.

Three input parameters that are specific for this i-MCFC model are the CO₂ molar flow from matrix to anode (Φ_{CO_2}), heating the matrix stream (Q_{matrix}) and pressure loss in the matrix channel (Δp_{matrix}). The CO₂ flow (Φ_{CO_2}) from the matrix flow to the anode inlet is calculated separately using Eq. (11) and set at the separator. After the calculations, the accuracy of this flow is checked by comparing the gas compositions and massflows of the anode outlet of the i-MCFC with the reference. Both must be the same. Q_{matrix} is calculated by the program (by apparatus #29, Fig. 4) and this heat amount is set to the fuel cell as heat dissipation. The pressure drop Δp_{matrix} is set at the same heat source that transfers heat to the matrix flow.

Table 1 summarizes the input parameters of the i-MCFC. The input parameters of the rest of the i-MCFC system are the same as the reference system. A detailed list of these input parameters are given in [16].

Table 1
Operating conditions and input parameters of the i-MCFC

Cell temperature, T_{cell} (°C)	650
Operating pressure, p (bar)	4
Fuel utilization, u_f (%)	70
Current density, i_{cell} (A/m ²)	1500
Cell voltage, V_{cell} (V)	0.8505
Cell power output, P_{cell} (kW)	306.19
Inverter efficiency, $\eta_{\text{dc-ac}}$ (%)	96
Temperature raise, $T_{\text{out}} - T_{\text{in}}$ (°C)	100
Anode pressure loss, Δp_{anode} (bar)	0.05
Cathode pressure loss, $\Delta p_{\text{cathode}}$ (bar)	0.1
Matrix pressure loss, Δp_{matrix} (bar)	0.1
CO ₂ molar flow, Φ_{CO_2} (mol/s)	1.9433
Heat transfer to matrix, Q_{matrix} (kW)	14.19

4. Results and discussion

Table 2 gives the resulting CO₂ and O₂ partial pressures and massflow at the cathode inlet. It shows that the i-MCFC system concept has a much lower p_{CO_2} (0.001 bar) in the cathode and high p_{CO_2} (1.89 bar) in the matrix. The i-MCFC should be able to operate under these gas compositions. With a p_{CO_2} in the cathode of 10⁻³ bar, the NiO cathode dissolution should reduce to a negligible level.

Table 3 gives the energy output and efficiencies of the two systems. It shows that the overall efficiency of this i-MCFC system is slightly lower than the MCFC. This is caused

Table 2
 p_{CO_2} and p_{O_2} of the cathode or matrix, and massflows through the fuel cell

	Reference	i-MCFC
p_{CO_2} (bar)	0.468 (cathode)	0.001 (cathode), 1.890 (matrix)
p_{O_2} (bar)	0.262 (cathode)	0.378 (cathode)
Φ_{expander} (kg/s)	0.261	0.262
Φ_{cathode} (kg/s)	1.257	1.146
Φ_{anode} (kg/s)	0.053	0.053
Φ_{matrix} (kg/s)	–	0.189
Φ_{total} (kg/s)	1.310	1.388

Table 3
Energy output and efficiencies of the reference and i-MCFC systems

	Reference	i-MCFC
FC stack output (kW)	306.19	306.19
Expander (kW)	60.05	60.34
Auxiliary (kW)	–77.58	–78.66
Net power (kW)	288.66	287.87
(η (%) LHV)	(51.77)	(51.63)
(η (%) EX)	(49.70)	(49.56)
Heat _{T=180°C} (kW)	91.59	91.98
Heat _{T=80°C} (kW)	109.49	109.49
Total output (kW)	489.74	489.34
(η (%) LHV)	(87.84)	(87.76)
(η (%) EX)	(58.46)	(58.34)

by the increased auxiliary power consumption. The cathode massflow of the i-MCFC is lower but nevertheless, the total massflow Φ_{total} (see Table 2) that is going through the cell is slightly higher than for the MCFC. This results in an increase of auxiliary power consumption. In theory, similar power output for both stacks should result in the same cooling requirements and the total massflow through the stacks should be about the same. The difference in total massflow can be ascribed to the difference in gas composition and thus heat capacity of the cathode gas. CO_2 has a high heat capacity, and in the reference system [16] part of the CO_2 in the flue gas from the HER is recycled by the cathode recycling. In the i-MCFC system, this CO_2 is not recycled in any way but leaves the system via the expander. Therefore, the amount of CO_2 that flows through the i-MCFC is smaller than the MCFC reference. The relative cooling capacity of the gas flow through the i-MCFC is hence lower, thus, the total gas flow is higher than the reference in order to meet the same cooling requirement.

The flowsheet calculation shows that the air input of the i-MCFC system is slightly higher than the reference resulting in a higher expander massflow (Φ_{expander} , see Table 2). This increases the compressor's power consumption, which is partly recovered by the expander. Finally, the i-MCFC system suffers from additional pressure loss from an extra heating stage that is required for heating-up the matrix gas to fuel cell entrance temperature. This gas is driven by the anode recycle blower and the additional pressure loss results to an increase in power consumption by this blower. The heat output of the i-MCFC system is slightly higher than the MCFC reference system. Production of hot water ($T = 80^\circ\text{C}$) is the same but the amount of steam ($T = 180^\circ\text{C}$) is slightly more (0.4 kW).

All together, the i-MCFC system performance is slightly lower than the reference system as shown by Table 3. The net power output is about 0.8 kW lower than the reference system resulting in a decrease of 0.15% point based on LHV, and a decrease of 0.14% point based on exergy. Due to the slightly higher steam output of the i-MCFC system, the decrease in overall CHP efficiency is only 0.08% point based on LHV, and 0.11% point based on exergy.

5. Conclusion

This present study shows that the i-MCFC concept with its unique three massflows feature can be implemented in a MCFC system layout with just minor modifications to the reference system. When keeping the fuel cell performance the same, the overall system performance does not suffer significantly when the MCFC stack is substituted by an i-MCFC stack. The net power efficiency of the system is about 0.15 and 0.14% lower than the reference MCFC system, based on respectively the lower heating value and exergy value of the fuel input. The decrease in overall CHP efficiency is just 0.08 and 0.11% based on LHV and exergy,

respectively. An advantage of this system as implemented here is the elimination of the lifetime limiting cathode dissolution. This is achieved by utilizing the unique feature of the i-MCFC of separating the CO_2 supply for the cell from the cathode gas. The p_{CO_2} of the cathode gas is here reduced from 0.468 bar for the reference MCFC system to 0.012 bar for this i-MCFC system. This should sufficiently suppress the cathode dissolution.

Finally, it should be mentioned that the i-MCFC system that is presented here does not utilize the unique feature of active CO_2 separation from fuel gas by the matrix channel. This option promises enhanced system performance. The reference MCFC system we used does not allow this without major system modification, which makes a proper comparison impossible. Other system studies are required in order to assess the possible improvement in system efficiency that fully exploits the features of the i-MCFC. Especially systems that are fueled by biogas and landfill gas utilizing are promising due to the low caloric value and high CO_2 content of these gasses.

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