

Optimization of a fuel cell system using process integration techniques

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Abstract

A model of a system including a proton exchange membrane (PEM) fuel cell and its fuel processing section has been developed. The goal was to investigate the process configurations to identify the optimal operating conditions and the optimal process structure of the system by applying modeling and process integration techniques.

A simulation model using commercial chemical process simulation software [Belsim-VALI III, Version 10: User Guide, Belsim s.a., Rue Georges Berotte 29A, B-4470 Saint-Georges-sur-Meuse, Belgium (2001)] has been developed. The model includes a steam methane reforming (SMR), a water gas shift (WGS) and a preferential oxidation (PROX) for the fuel processing feeding a proton exchange membrane cell stack (PEM) and a post-combustion unit. The energy and the material flows obtained by simulation have been used to compute the process composite curves of the system. This has been used to identify the best heat exchange opportunities and to define the optimal operating conditions of the reforming system in order to provide the best overall efficiency considering the balance of plant.

By improving the energy integration of the system and by using optimization of operating conditions, the efficiency can be raised from 35% for the reference system to up 49% in the optimized design. This is obtained both by process configuration modifications and by optimizing operating set-points. The model developed has been used for sensitivity analysis and will serve as a basis for further process design including the optimal structure determination and the thermo-economic optimization.

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1. Introduction

Proton exchange membrane (PEM) fuel cells for stationary applications are highly integrated systems including fuel processing, fuel cell itself and post-combustion units. The integration is not only expressed in terms of material flows but also in terms of energy balance. Process design has therefore a big impact on the performances of the total system [2]. Energy integration techniques have been widely applied in the chemical process industry to design and synthesize new processes. The combined use of process simulation packages with energy integration techniques allows today to design highly integrated systems. Our goal here has been to apply these methods to analyze and improve the performances of a total proton exchange membrane fuel cell (TPEMFC). As a starting point for the design, the ALSTOM BALLARD system [3] has been used as a reference case.

The adopted methodology is the following:

- (1) Develop a simulation model that allows computing the energy requirements defined as a list of hot and cold streams as a function of the major degree of freedom of the system, i.e. steam to carbon ratio, reforming temperature, operating temperature and pressure of the membrane, fuel utilization and air excess. This model may also be used later on to estimate equipment costs and compute operating constraints.
- (2) Compute the system integration using well established process integration techniques [4] into a specific optimization frame work [5]. This step allows targeting the integrated system performances without having to impose the heat exchange system design.
- (3) Optimize the system performance and analyze the sensitivity of major decision parameters in order to understand the major integration bottlenecks. This step aims also at understanding the influence of constraints and computing their importance with respect to the system performances.
- (4) Design the heat exchange network for the integrated system that is able to reach the targeted performance.

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Nomenclature	
A_{cell}	cell area (cm^2)
E_{cell}	cell power (W)
E_{FC}	fuel cell power (W)
$f_{\text{fuel}}^{\text{add}}$	fuel flow rate required in the post-combustion subsystem (kg/s)
$f_{\text{fuel}}^{\text{FC}}$	fuel flow rate processed in the fuel processing subsystem (kg/s)
f_i	total molar flow rate of stream i
F	Faraday number ($=96485 \text{ Cb mol}^{-1}$)
$h_i(T, P)$	molar total enthalpy (including enthalpy of formation) of stream i at a temperature T and a pressure P
i_{cell}	current density (A/cm^2)
I_{cell}	cell current (A)
LHV_{fuel}	lower heating value of the fuel (kJ/kg)
n_i^x	molar flow of component x in stream i
N	the number of the cells in the stack
P_{cell}	cell pressure (Pa)
PROX	preferential oxidation reactor
$Q_u^{-(+)}$	heat to be evaluated from (–), or supplied to (+) unit u (W)
R_{cell}	cell resistance ($\Omega \text{ cm}^2$)
SMR	steam methane reformer
T_i	temperature of the stream i (K)
U_{Nernst}	Nernst potential (V)
$U_{\text{Nernst}}^0(T_{\text{cell}})$	Nernst ideal voltage at T_{cell} (V)
V_{cell}	cell voltage (V)
WGS	water gas shift reactor
y_i^x	mole fraction of component x in stream i
Y_u^x	mean mole fraction of component x in the unit u : $Y_u^x = (y_{i,u}^x + y_{o,u}^x)/2$, where i, o refer to inlet, respective outlet of unit u
<i>Greek letter</i>	
η_u^x	conversion rate of component x in unit u : $\eta_u^x = (n_{i,u}^x - n_{o,u}^x)/n_{i,u}^x$

In this paper, we will present the methodology to realize the first three steps of the methodology.

2. The total proton exchange membrane fuel cell model

The total proton exchange membrane fuel cell (TPMEMFC) model has been built according to the ALSTOM BALLARD system [3]. It has been divided in three subsystems:

- (1) The fuel processing (FP) subsystem.
- (2) The proton exchange membrane fuel cell (PEMFC) subsystem.
- (3) The post-combustion subsystem.

The model has been developed using a commercial modular software (Belsim-VALI III, [1]) that is using an “equation

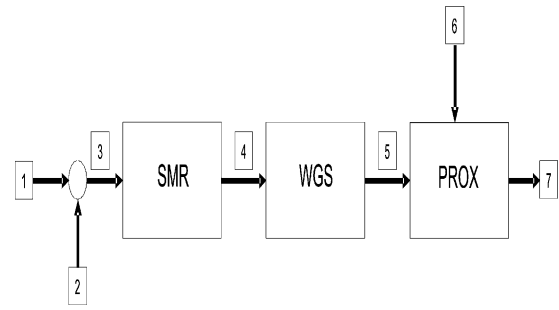


Fig. 1. Fuel processing process flow diagram.

solver” approach that reveals to be very powerful and useful for process synthesis. The calculation of the heat exchange system using process integration techniques allows avoiding the calculation of a complex integrated system with complex heat exchange links.

2.1. The fuel processing subsystem model

The fuel processing subsystem (Fig. 1) includes the following three main reactors:

- The steam methane reformer (SMR) that proceeds with the main reactions of steam reforming to produce hydrogen from the methane.
- The water gas shift (WGS) reactor that transforms the remaining CO into additional hydrogen at a lower operating temperature.
- The preferential oxidation reactor (PROX) whose aim is the conversion of the remaining CO into CO_2 to avoid poisoning the fuel cell membrane.

The inlet mass flows presented in Fig. 1 are the following:

- The stream #1 is the methane flow provided at 3 bar and 25 °C. Pure methane has been considered for the calculation but the model might be easily adapted when real fuel including other component will be considered.
- The stream #2 is the water flow required for the steam methane reformer whose flowrate is controlled by the steam to carbon ratio.
- The stream #6 is a pure oxygen flow required for the PROX.

As this model is developed for system design, the pressure drops have been neglected and the operating pressure has been fixed at 3 bar.

2.1.1. Steam methane reformer

The steam methane reformer performs the following reactions:

- The endothermic steam methane reforming:

$$\text{CH}_4 + \text{H}_2\text{O} \rightleftharpoons 3\text{H}_2 + \text{CO}, \quad \Delta H_r = 206.11 \text{ kJ/mol.}$$
- The exothermic water gas shift:

$$\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{CO}_2 + \text{H}_2, \quad \Delta H_r = -41.2 \text{ kJ/mol.}$$

The two reactions are supposed to be near equilibrium. A temperature difference with respect to the equilibrium is used to represent the imperfection of the catalyst. We assume that the size of the catalyst bed will be sufficient to approach equilibrium. The reactor is supposed to be isothermal, meaning that heat has to be supplied to maintain the temperature in the reactor from an external energy source. In order to define appropriately the energy requirement of the heat exchange, we have assumed that the inlet streams will be preheated up to the reaction temperature. This assumption is indeed conservative in the sense that it does not take account for the reactor temperature profile that will be below the temperature profile obtained.

In this study, the temperature of the reformer $T_{o,SMR}$ and the steam to carbon ratio have been considered as being the major decision variables for the optimization. Other parameters might be also chosen like the pressure or the size of the catalyst beds for further thermo-economic optimizations.

2.1.2. Water gas shift reactor

We consider a WGS system configuration with only one water gas shift reactor that proceeds with the exothermal WGS reaction ($\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{CO}_2 + \text{H}_2$) that is supposed to be near the equilibrium. In order to proceed with only one reactor the temperature of the system as been fixed at 250 °C using new catalyst as mentioned in [6]. The isothermal conditions will be maintained by heat exchange in the reactor. This means a special design for the reactor that will differ from the conventional adiabatic configurations. The isothermal conditions are assumed in order to avoid energy losses in the heat source definition and guarantee the lower possible equilibrium temperature. This approach is similar to the approach used in the reformer but for an exothermal reaction. The reactor inlet is supposed to be cooled down to the reactor temperature before entering the reactor. This defines a hot stream that will be considered in the energy integration calculation.

2.1.3. Preferential oxidation reactor

The preferential oxidation reactor is used to eliminate the CO that has not been converted in the WGS reactor. This reactor is required to reach very low level of CO content in the fuel stream at the fuel cell inlet in order to avoid poisoning of the membrane. The preferential oxidation is one of the possible technology for this task. It is using pure oxygen to proceed with the following reaction: $\text{CO} + \text{O}_2 \rightleftharpoons \text{CO}_2$. Unfortunately, the selectivity of the catalyst will not avoid the combustion of some hydrogen in the gas stream with the following reaction: $\text{H}_2 + (1/2)\text{O}_2 \rightleftharpoons \text{H}_2\text{O}$. In the model, the CO content at the reactor outlet is fixed at 10 ppm. The pure oxygen flow rate (stream #10) is computed as a function of the CO flow rate assuming one mole of O_2 per mole of CO according to the results published by [7]. An isothermal operation at a temperature of 200 °C has been considered for the PROX reactor, if needed, heat will be removed from

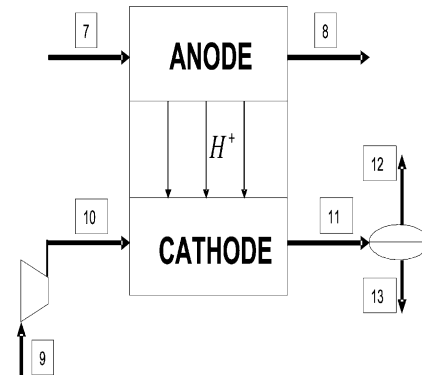


Fig. 2. PEMFC subsystem process flow diagram.

the reactor by heat exchange (see the process integration chapter for comments concerning the approach).

2.2. Proton exchange membrane fuel cell model

The PEMFC subsystem has been built considering separately the cathode and anode sides. A schematic of the PEMFC submodel is given in Fig. 2.

The operating pressure of the fuel cell is 3 bar. In the integrated system, the air compression work will be produced by expanding the post combustion gases in an expander. The compressor isentropic efficiency (η_c^{is}) is 86%. The inlet air condition are 1 bar, 25 °C and a relative humidity of 60%.

The equations of the PEMFC are the following:

- Nernst potential (V):

$$U_{\text{Nernst}} = U_{\text{Nernst}}^0(T_{\text{cell}}) + \frac{RT_{\text{cell}}}{2F} \ln \left(\frac{Y_{\text{anode}}^{\text{H}_2} (Y_{\text{anode}}^{\text{O}_2})^{1/2}}{Y_{\text{anode}}^{\text{H}_2\text{O}}} \right) + \frac{RT_{\text{cell}}}{4F} \ln(P_{\text{cell}}).$$

- Cell voltage (V): $V_{\text{cell}} = U_{\text{Nernst}} - R_{\text{cell}} i_{\text{cell}}$.
- Hydrogen flow rate through the membrane: $n_{i,\text{cathode}}^{\text{H}_2} = \frac{I_{\text{cell}}}{2F}$.
- Cell current (A): $I_{\text{cell}} = i_{\text{cell}} A_{\text{cell}}$.
- Cell power (W): $P_{\text{cell}} = V_{\text{cell}} I_{\text{cell}}$.
- Fuel cell power(W): $P_{\text{FC}} = P_{\text{cell}} N$
- Hydrogen oxidation at the cathode: $\text{H}_2 + (1/2)\text{O}_2 \rightarrow \text{H}_2\text{O}$.
- Energy balance:

$$\sum_{i=1}^{\text{inlets}} f_i h_i(T_{\text{cell}}, P_{\text{cell}}) - \sum_{o=1}^{\text{outlets}} f_o h_o(T_{\text{cell}}, P_{\text{cell}}) = E_{\text{FC}} + Q_{\text{FC}}^-$$

- Fuel utilization: $\eta_{\text{comb}} = (n_{i,\text{anode}}^{\text{H}_2} - n_{o,\text{anode}}^{\text{H}_2}) / n_{i,\text{anode}}^{\text{H}_2}$. The fuel utilization in the cell (η_{comb}) has been set at 95% according to [8]. This means that 5% of the hydrogen at the inlet of the cell is not used in the fuel cell and will be used as depleted fuel to balance the heat requirement of the fuel processing section. The sensitivity of this parameter will be described later on.

The PEMFC subsystem is supposed to be isothermal and isobar. The cell temperature T_{cell} will be considered as a decision parameter for the optimization. Heat exchange is realized to maintain the cell temperature. We assumed that the hydrogen combustion is complete on the cathode side and that only the hydrogen protons are crossing the membrane [8]. Water migration through the membrane has been ignored since it might go in both direction. Water management strategies have not been considered in the model. The sensitivity analysis procedure will be used to evaluate the possible water management strategies and their impact on the system efficiency.

The stream at the cathode outlet is cooled down and the water is separated to be partly recycled in the steam methane reformer.

2.3. Post-combustion subsystem model

The depleted fuel of the PEMFC subsystem is burned in the post combustion subsystem (Fig. 3). The generated heat will be used to balance the energy requirement of the fuel processing section. Supplementary firing of natural gas will be considered if the energy content of the depleted fuel is not sufficient to close the balance. The supplementary fuel requirement will be computed using the process integration techniques as described in the next section.

The inlet streams #8 and #12 are, respectively, the cathode and anode outlets. The assumptions are the following:

- The combustion is complete.
- Stoichiometric combustion has been assumed. This is not the case in reality, this assumption is made to compute the real potential in terms of energy integration. In reality, the air excess is related to the water management strategy: an increase of the air flow rate allows to reduce the liquid water production on the cathode side, but in the same time it reduces the post-combustion temperature decreasing by the way the available heat for the reforming. When designing the heat exchange system, the sensitivity of the assumption will be tested with respect to the system efficiency and might be removed.
- The depleted fuel and air preheating temperatures will be considered as an optimization parameter for the process integration.
- The flue gases will exchange heat with the other subsystems before being expanded in a turbine to produce the

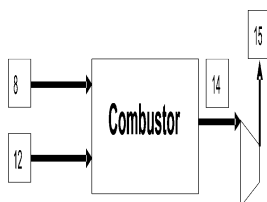


Fig. 3. Post-combustion subsystem process flow diagram.

air compression work. The inlet temperature in the turbine will be computed to balance the air compression work requirement.

- After expansion, a new heat exchange is performed to cool down the streams to a stack temperature of 120 °C and a pressure of 1 bar.

2.4. Heat exchange subsystem model using process integration techniques

Intentionally, the heat exchangers have not been considered in the simulation model described above because the heat exchange system has been considered as being unknown apriori. Our goal is to define the heat exchange network structure for optimized operating conditions. We have therefore used the hot and cold composite curves concept of the pinch technology to model the integrated heat exchange system without having to impose a heat exchange network structure. In this approach, the results of the simulation model will characterize the hot and cold streams of the system.

The process integration method (or pinch technology) (e.g. [4]) has been developed for analyzing the energy integration in industrial processes to help identify energy savings opportunities. This method is typically applied in two major steps. The first is a targeting phase that aims at identifying the minimum energy requirement of the system by computing the possible energy recovery from the hot streams to heat up the cold streams of the system. The second step is the synthesis of the heat exchange network that will implement the targeted energy recovery. This method has been widely applied in the process industry for the both grassroots and retrofitting problems. It has also been widely used as a tool for process synthesis. Process integration studies start with the definition of a list of hot and cold streams. The hot and cold streams define, respectively, heat sources and heat requirements of the system that are characterized by a heat-temperature diagram usually defined by a heat load, an inlet and a target temperature. The heat sources are then composed to compute the hot composite curve that represents the heat availability in the system as a function of the temperature. The same procedure is applied for the cold streams to draw the cold composite curve. The composite curves can be considered as being one single hot and cold streams that will exchange heat in a virtual counter-current heat exchanger that models the heat exchanger network of the system under study. Considering that the heat exchange will be technically feasible if the temperature difference between the hot and cold streams will always be superior to a predefined ΔT_{min} , the maximum heat recovery by heat exchange between the hot and the cold streams will be obtained when the ΔT_{min} constraint will be activated. This point is called the system pinch point. By heat balance, one may then compute the minimum energy requirement of the system and the minimum heat losses to be evacuated from the system. When some of the streams have unknown flow rate or

Table 1
The Fuel processing subsystem streams, syngas composition: CO, H₂O, H₂, CO₂, CH₄

#	Stream	Description	T _{in}	T _{target}	Flow type
1	Cold	Water preheating for SMR reaction (including liquid water preheating, the vaporization and the steam superheating)	20 °C	T _{i,SMR}	H ₂ O
2	Cold	Methane preheating for SMR reaction	25 °C	T _{i,SMR}	CH ₄
3	Hot	The stream between the SMR and WGS reactions	T _{o,SMR}	T _{i,WGS}	syngas
4	Hot	The stream between the WGS and PROX reactions	T _{o,WGS}	T _{i,PROX}	syngas
5	Hot	The generated heat by the WGS reaction	T _{o,WGS}	T _{o,WGS}	Heat
6	Hot	The generated heat by the PROX reaction	T _{o,PROX}	T _{o,PROX}	Heat
7	Cold	The required heat by the SMR reaction	T _{o,SMR}	T _{o,SMR}	Heat

Table 2
The PEMFC subsystem streams, flue gas composition: H₂O, CO₂, O₂, N₂

#	Stream	Description	T _{in}	T _{target}	Flow type
8	Hot	The stream between the fuel processing subsystem outlet and PEMFC inlet	T _{o,PROX}	T _{cell}	Syngas
9	Hot	The air compressor outlet cool down to the cathode inlet	T _{o,comp}	T _{i,cell}	Air
10	Hot	The stream between the cathode outlet and the water separator	T _{o,cell}	35 °C	Flue gas
11	Hot	The generated heat by the PEMFC	T _{cell}	T _{cell}	Heat

Table 3
The post-combustion subsystem streams

#	Stream	Description	T _{in}	T _{target}	Flow type
12	Cold	The air preheating for the combustion	35 °C	500 °C	Flue gas
13	Cold	The depleted fuel preheating for the combustion	T _{cell}	500 °C	Syngas
14	Hot	The generated heat by the combustion	T _{o,comb}	T _{i,exp}	Flue gas
15	Hot	The combustion gases stream after the expander	T _{o,exp}	120 °C	Flue gas

temperature, optimization methods are used to compute the optimal values that give the best integration (e.g. [5]).

Using the composite curve calculation together with a simulation model, we define the simulation of the heat exchanges network without knowing about its configuration and we will be able to compute the influence of the major decision variables to later on define the best system configuration. The difficulty here is that the heat exchange model has to be solved as an optimization problem.

The efficiency of the application of the pinch technology is highly related to the appropriate definition of the hot and cold streams, i.e. the definition of the heat load and the temperatures in the system. These have to be defined in such a way that heat recovery options will not be missed or hidden. This explains the choice of isothermal reactions and some of the preheating operations that have been introduced in the model. Once the results will be obtained, the usefulness of the heat exchange requirements will be analyzed and only the useful exchanges will be considered in the heat exchanger network design.

Tables 1–3 show the definition of the hot and cold streams of the system.

The heat sources and sinks are used to compute the hot and cold composite curves (Fig. 4). Assuming a ΔT_{min} of 10 °C and 25 °C at the SMR, we have computed the heat

recovery between the heat sources (hot streams) and the heat requirements (cold streams).

The ΔT_{min} constraint is used to impose the technical feasibility of the heat exchanges. The pinch point is the temperature at which the constraint is activated. If the available heat in the hot streams above the pinch point is lower than the heat required by the cold streams, a supplement of heat

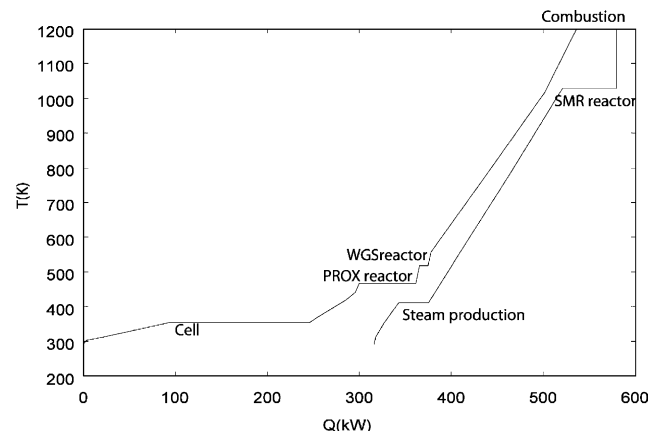


Fig. 4. The composite curves of the TPPEMFC system with a steam to carbon ratio of 1.4 (SMR and PEMFC temperatures fixed at 750 and 85 °C and a fuel utilization of 95%) and a stoichiometric combustion.

will have to be supplied to the system. In our case, the heat supplement will be supplied by a supplementary firing with natural gas. The combustion will define a hot stream (flue gas from the adiabatic temperature of combustion to the stack temperature). It should be noted that if the pinch point temperature is the reforming temperature (high temperature), the supplementary firing should take into account the possible air preheating below the pinch point that will reduce the fuel consumption. In the model developed, we used an optimization model [5] that allows the calculation of the optimal fuel flowrate and the corresponding optimal air preheating temperature.

3. Results and discussions

The integrated process model has been used to determine the optimal operating conditions to be considered in the system.

The influence of the following parameters has been studied:

- The steam to carbon ratio (steam/carbon).
- The SMR temperature ($T_{o,SMR}$).
- The PEMFC temperature (T_{cell}).
- The fuel utilization (η_{comb}).

Analyzing the composite curves of the system allows then to study the process modifications that will further improve the overall efficiency of the system.

3.1. The self-sufficient limit

One of the interests of the combined use of process simulation and process integration techniques is the determination of the self-sufficient limit of the TPMEFC system that is defined as being the limit at which the depleted fuel at the outlet of the fuel cell allows satisfying the energy requirement of the system. This limit is obviously related to the decision parameters that have been used in this study. Figs. 4 and 5 show the composite curves obtained for two different values of the steam to carbon ratio at fixed $T_{o,SMR} = 750^\circ\text{C}$, $T_{cell} = 85^\circ\text{C}$ and $\eta_{comb} = 95\%$. In Fig. 4, the depleted fuel satisfies the energy requirement of the system. When increasing the steam to carbon ratio, a pinch point is created by the steam production (at the vaporization temperature), because the heat requirement has been increased and because the excess of water leads to a higher water content in the depleted fuel and also because the air flowrate reduces the adiabatic temperature of combustion. In Fig. 5, we should also observe an increase of the WGS reactor performance that corresponds to a decrease of the PROX reactor, revealing that the CO conversion has been higher in the WGS reactor due to the presence of an excess of water in the syngas produced by the reformer. We should also remark that by the non selectivity of the PROX catalyst, part of the oxygen entering the reactor will burn

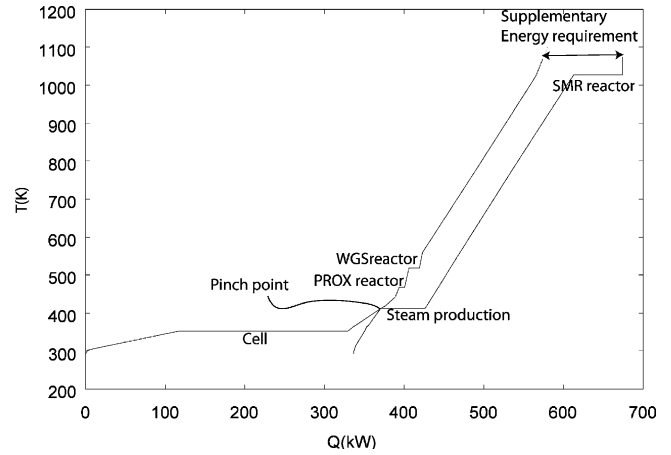


Fig. 5. The composite curves of the TPMEFC with a steam to carbon ratio of 2.4 (SMR and PEMFC temperatures fixed at 750 and 85 °C and a fuel utilization of 95%) and a stoichiometric combustion.

part of the hydrogen produced reducing the production of electricity in the cell. The results of Fig. 4 are obtained for a stoichiometric combustion of the depleted fuel. If air excess is used to control the water balance of the cathode, we obtain the composite curves of Fig. 6. The combustion temperature is reduced and the pinch point is activated at the reformer temperature. In the case of Fig. 6, the air excess correspond to a near saturation condition at the cathode outlet.

When the system is not self-sufficient, additional natural gas is used to balance the system requirement. In this case, the overall efficiency of the system is computed by Eq. (1).

$$\eta = \frac{P_{elec}}{\text{LHV}_{fuel}(f_{fuel}^{FC} + f_{fuel}^{add})} \quad (1)$$

where P_{elec} is a generated electric power (kW), LHV_{fuel} the lower heating value of fuel (kJ/kg), f_{fuel}^{FC} the fuel flow rate processed in the fuel processing subsystem (kg/s) and f_{fuel}^{add} is the additional flow rate required in the post-combustion subsystem (kg/s).

Fig. 7 shows the influence of the steam to carbon ratio on the system efficiency. The optimum steam to carbon ratio (1.8) leads to a maximum efficiency of 46%. In Fig. 7, we have located the self-sufficient limit. Here, the interest of the additional firing is demonstrated: without supplementary firing, the maximum efficiency would be of 42%. It becomes 46% with supplementary firing. The maximum performance for this operating conditions corresponds to an additional firing of 4% based on the total fuel inlet.

3.2. The SMR reactant preheating train

From the analysis of the composite curves of the system, several improvements might be suggested. In order to reduce the plateau at the steam vaporization level, it might be envisaged to install a special preheating device that will

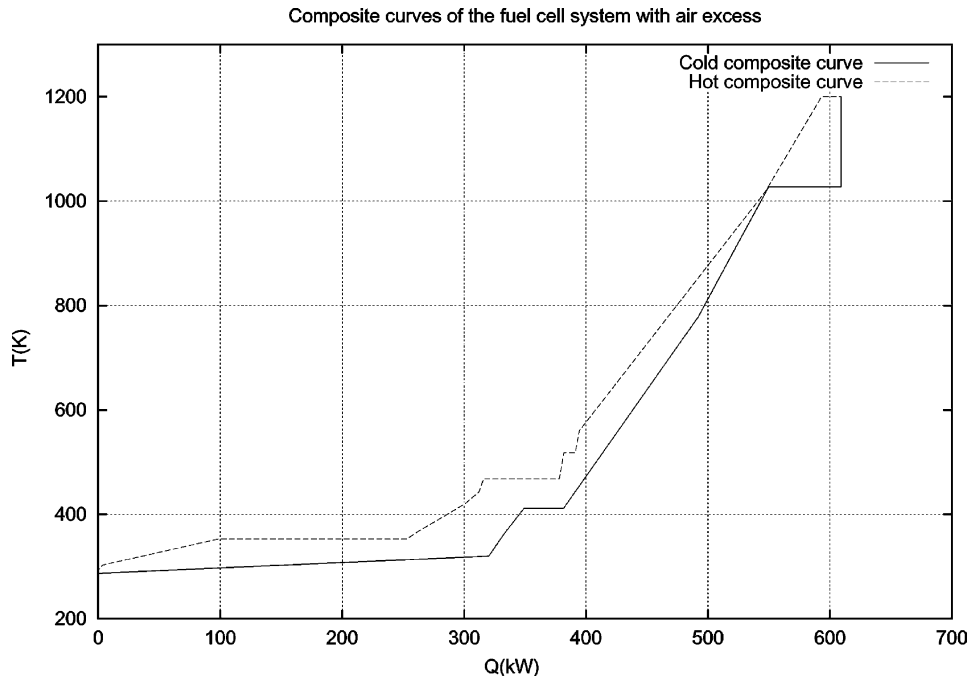


Fig. 6. The composite curves of the TPFC system with a steam to carbon ratio of 1.4 (SMR and PEMFC temperatures fixed at 750 and 85 °C) and an oxygen content in the flue gases of 6%.

saturate the natural gas with a liquid water injection while preheating the mixture. This device will profit from the partial pressure effect to vaporize part of the water, allowing to transform the constant temperature plateau into a temperature profile as indicated in Fig. 8. In this way, the cold and hot composites are closer meaning that both the required

energy and energy losses are minimized. Fig. 9 points out the comparison between the efficiencies with and without vaporization. The maximum efficiency of the system is increased of about 3% and corresponds to a higher steam to carbon ratio. The efficiency seems to stabilize after the maximum is reached for higher steam to carbon ratio.

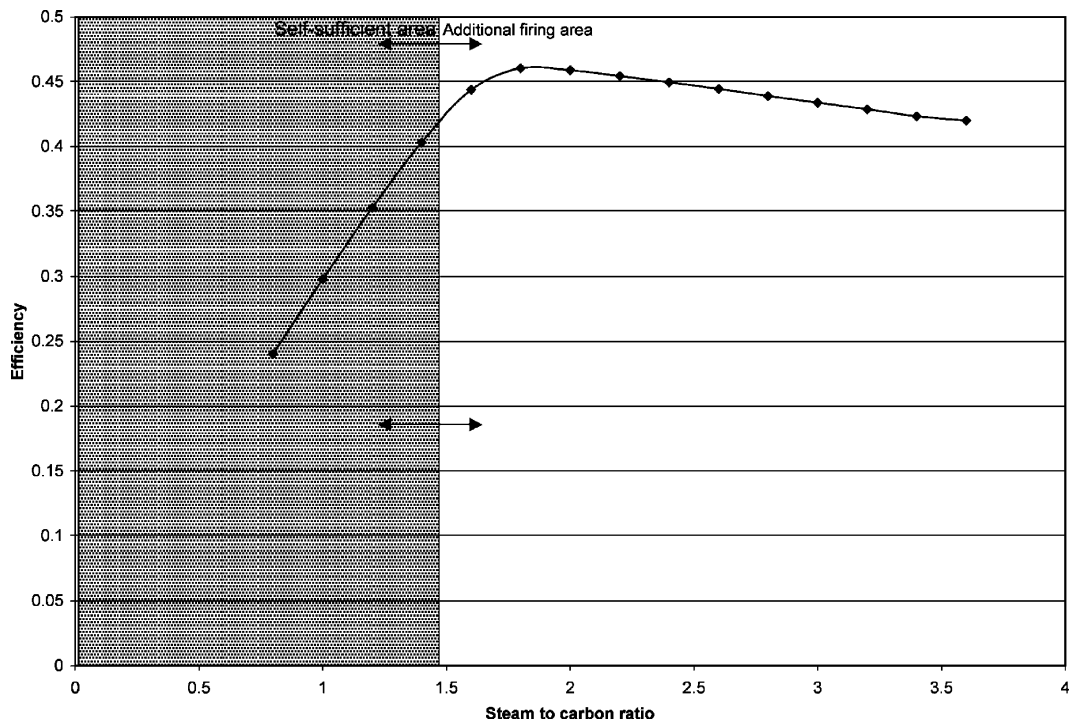


Fig. 7. The efficiency as a function of steam to carbon ratio (SMR and PEMFC temperatures fixed at 750 and 85 °C and a fuel utilization of 95%).

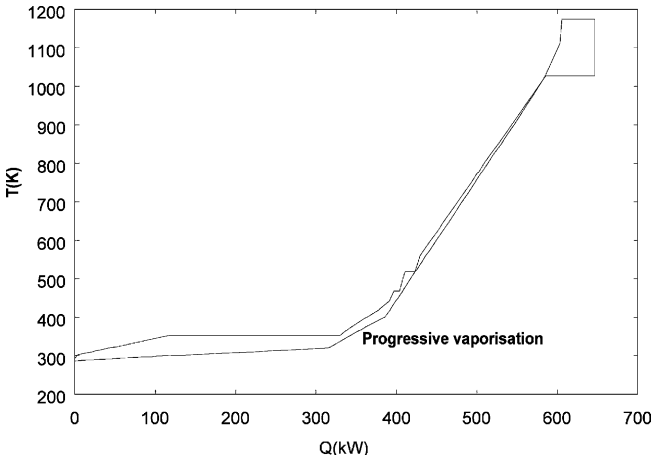


Fig. 8. The composite curves of the TPMEFC system without vaporization for a steam to carbon ration of 2.4 (SMR and PEMFC temperatures fixed at 750 and 85 °C and a fuel utilization of 95%).

The limit of the self-sufficient process is pushed to the right but additional firing is still needed to reach the optimum. The self-sufficient system has a higher efficiency when the saturation device is used.

3.3. The air combustion and depleted fuel preheating

Air combustion and depleted fuel preheating (combustion preheating) recovers heat from below the reforming temperature to make it available above the reforming temperature. This reduces the additional fuel required and increases the

self sufficient limit. The results shown in Fig. 10 demonstrate that the optimum efficiency is reached for smaller steam to carbon ratio (2.4 instead of 4). The systems with preheating will therefore be more compact and less expensive. The maximum efficiency is of 49% with preheating while it is of 47% without preheating. As it will be shown in Fig. 15, the difference in efficiency is even higher for lower fuel utilization. Working with combustion preheating will also help in water management issues: increasing the air excess to reduce the water content at the cathode will have a smaller effect on the overall performance of the system (Fig. 11).

3.4. The influence of the steam methane reformer temperature ($T_{o,SMR}$)

The developed model gives information about the optimal reforming temperature. In Fig. 12, we present the efficiency as a function of the steam to carbon ratio for different values of $T_{o,SMR}$.

Similar optimal system efficiencies may be reached but for different steam to carbon ratio. For similar efficiencies, the curve corresponding to the temperature of 750 °C corresponds to the smallest steam to carbon ratio and to the highest efficiency. $T_{o,SMR} = 750$ °C seems to be the optimal temperature. We should observe that the curves corresponding to $T_{o,SMR} = 700$ and 800 °C are practically superposed meaning that a range of $T_{o,SMR}$ will provide similar efficiencies. In Fig. 12, we also compare the hydrogen molar fraction at the fuel processing subsystem outlet and the system efficiency.

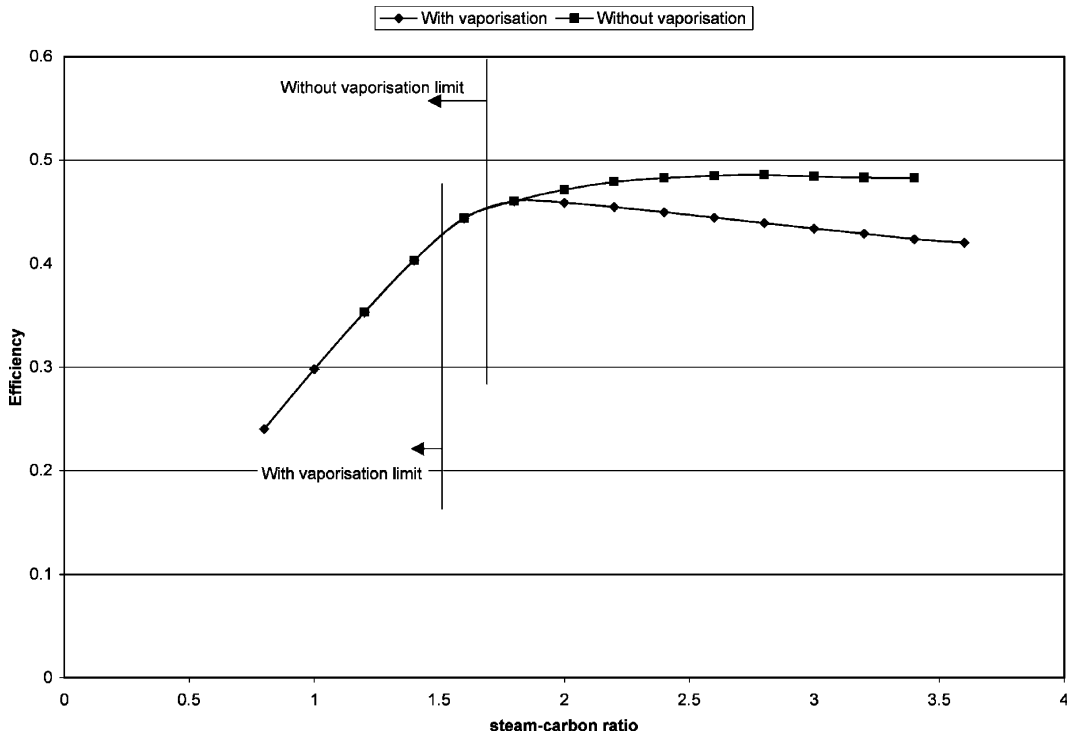


Fig. 9. The efficiency with and without vaporization as a function of steam to carbon ratio (SMR and PEMFC temperatures fixed at 750 and 85 °C and fuel utilization 95%).

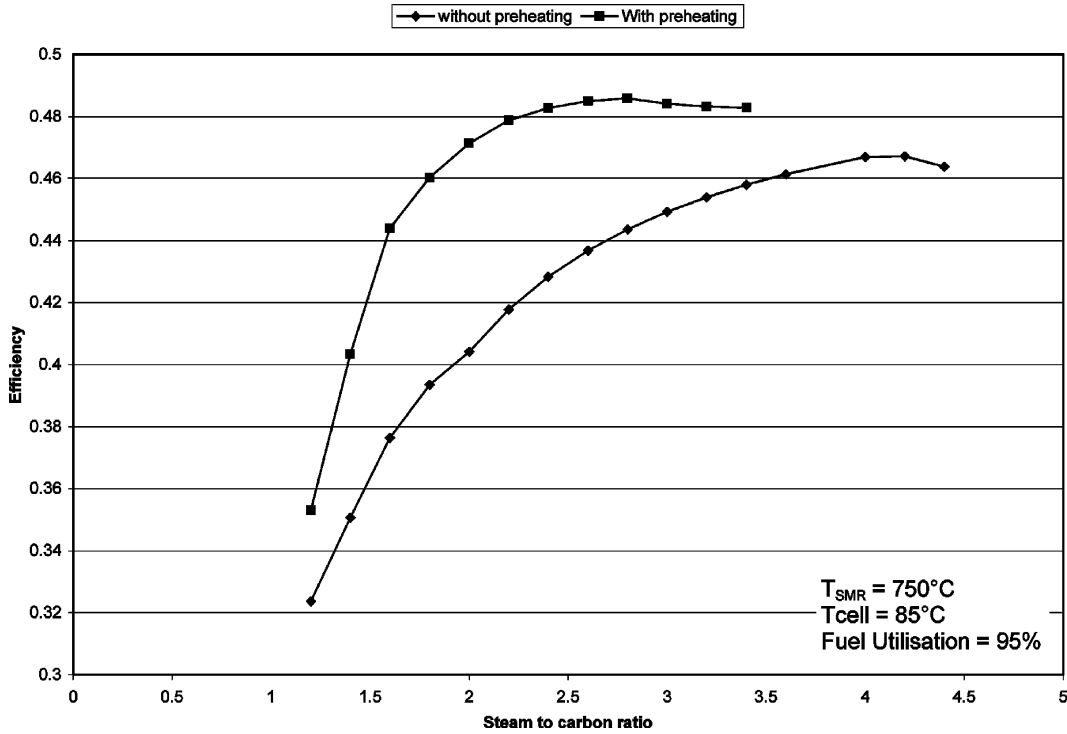


Fig. 10. The efficiency with and without combustion preheating as a function of steam to carbon ratio (SMR and PEMFC temperatures fixed at 750 and 85 °C and a fuel utilization of 95%).

The optimum steam to carbon ratio for the fuel processing and for the global efficiency do not coincide indicating the importance of adopting an integrated approach rather than separated optimizations.

3.5. The influence of the cell temperature T_{cell}

Fig. 13 shows the relative humidity at anode and cathode outlets and inlets for two different temperature of the cell.

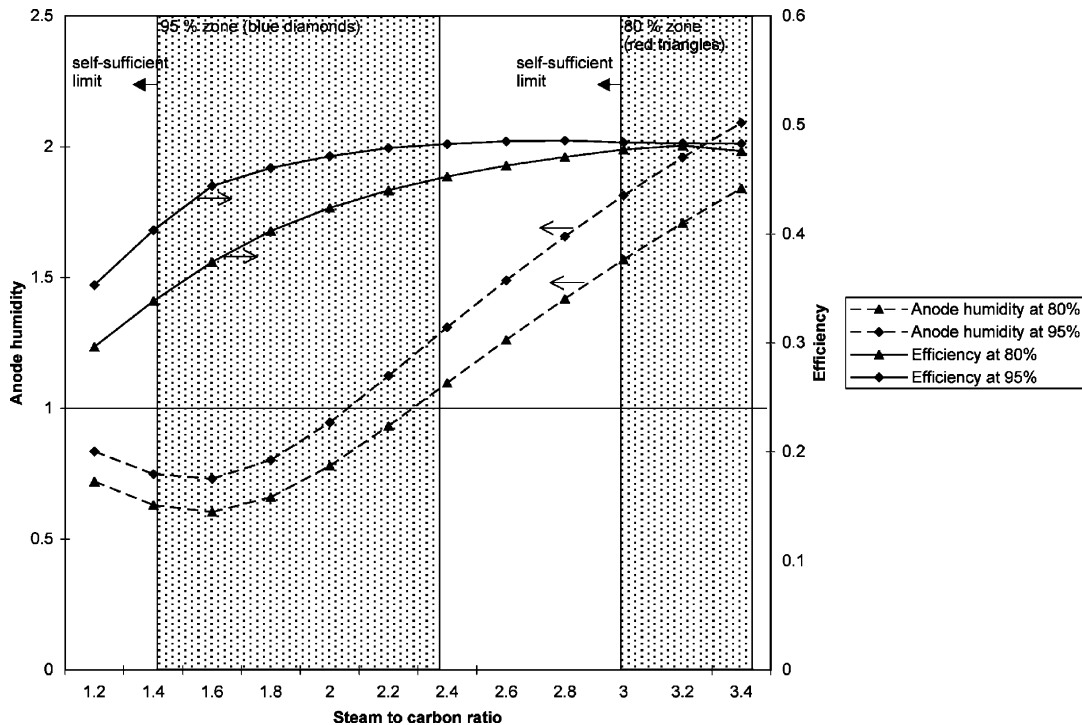


Fig. 11. The influence of the fuel utilization on the self-sufficient limit.

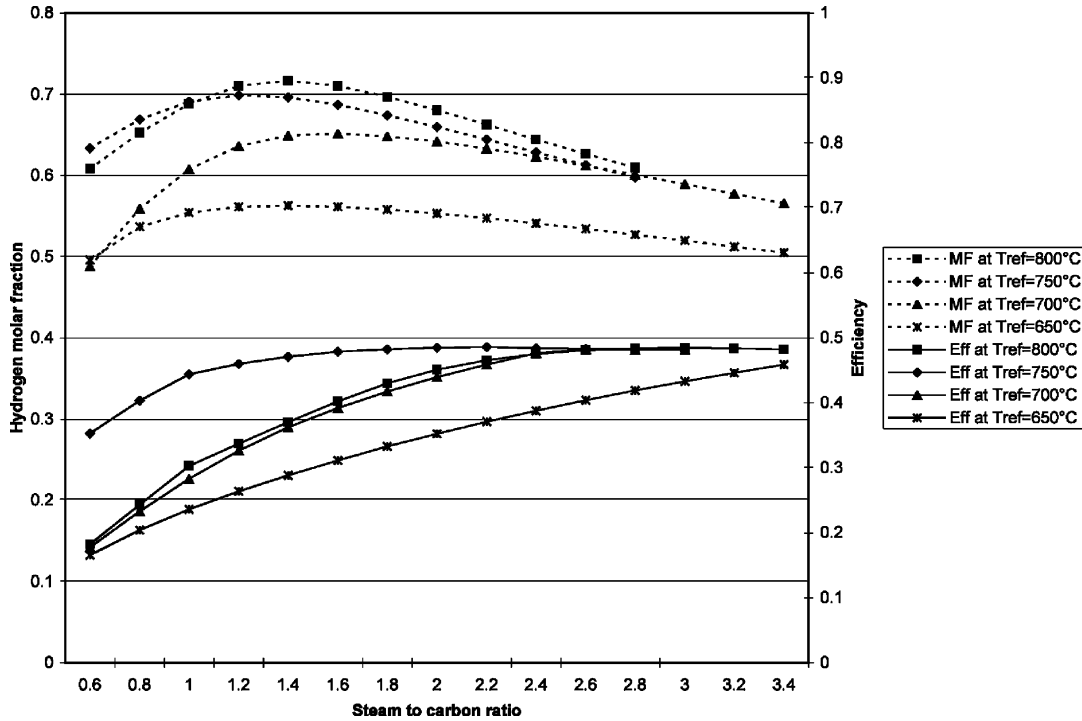


Fig. 12. The efficiency and the H₂ molar fraction for different reforming temperatures, the cell temperature is fixed at 85 °C and a fuel utilization of 95%.

The steam to carbon ratio and the reforming temperature have a big influence on the humidity and therefore on the water management strategy. As all results have been carried out with the mass flow rate of water fixed at zero through the polymer membrane, a careful analysis should be performed to study these phenomena and introduce the conclusions as

constraints in the simulation and integration model. The modeling strategy adopted here will help in computing the impact at the system level.

The cell temperature does not really affect the overall system efficiency. The impact will be rather on the technology. The humidity decreases with an increase of the cell

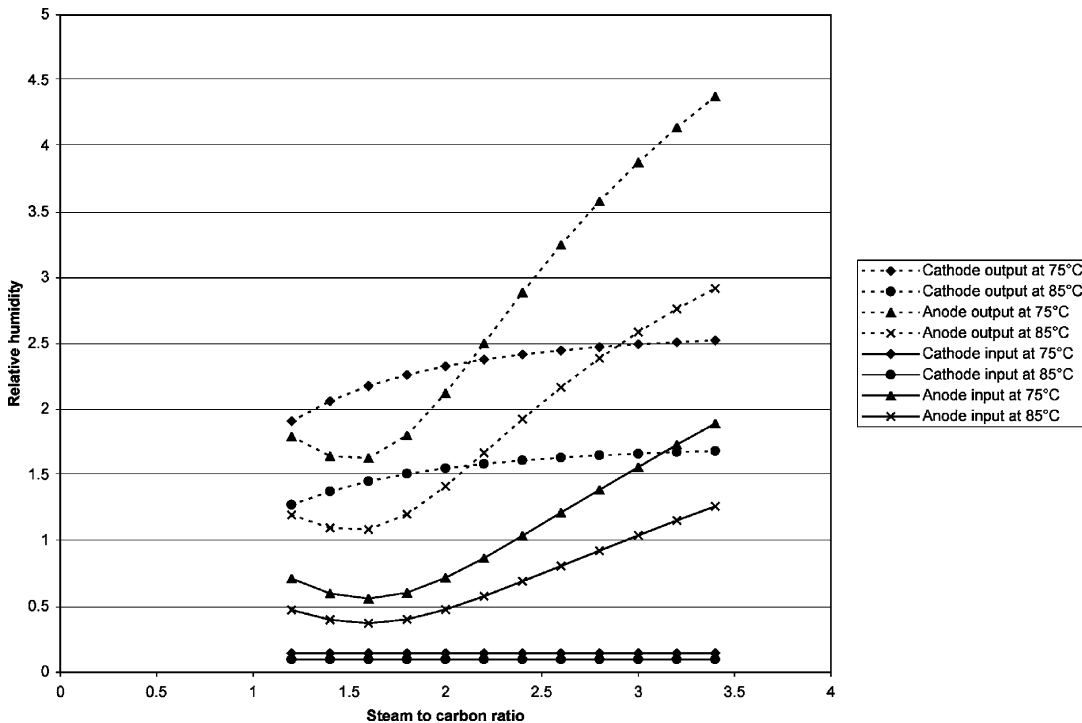


Fig. 13. Anode and cathode humidities at the inlet and outlet of the cell as a function of the steam to carbon ratio for two different cell temperatures.

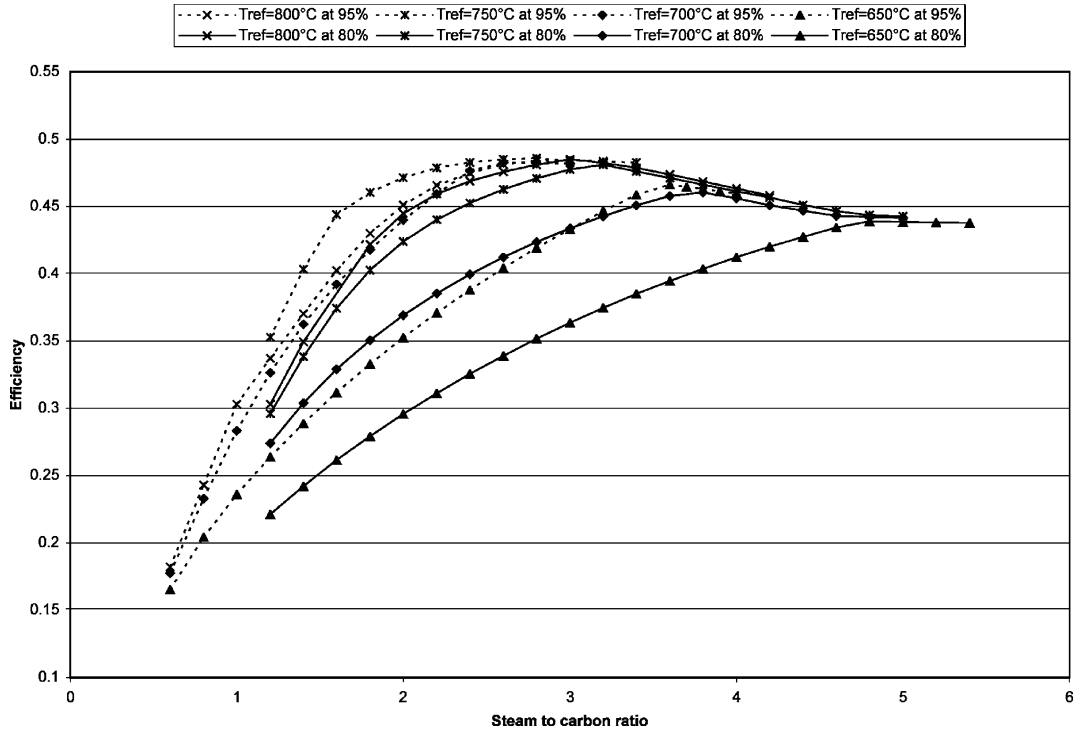


Fig. 14. The efficiency for different fuel utilization's and different reforming temperatures.

temperature, as shown in Fig. 13, reasonable efficiency can be reached within the optimal operating temperature range and with acceptable humidity. The difference between the inlet and the outlet humidity is higher for lower temperatures

(Fig. 13) indicating that working with higher temperatures will create less problems of water repartition in the cell.

According to the affirmations above, the cell temperature has to be as high as possible meaning also a higher energy

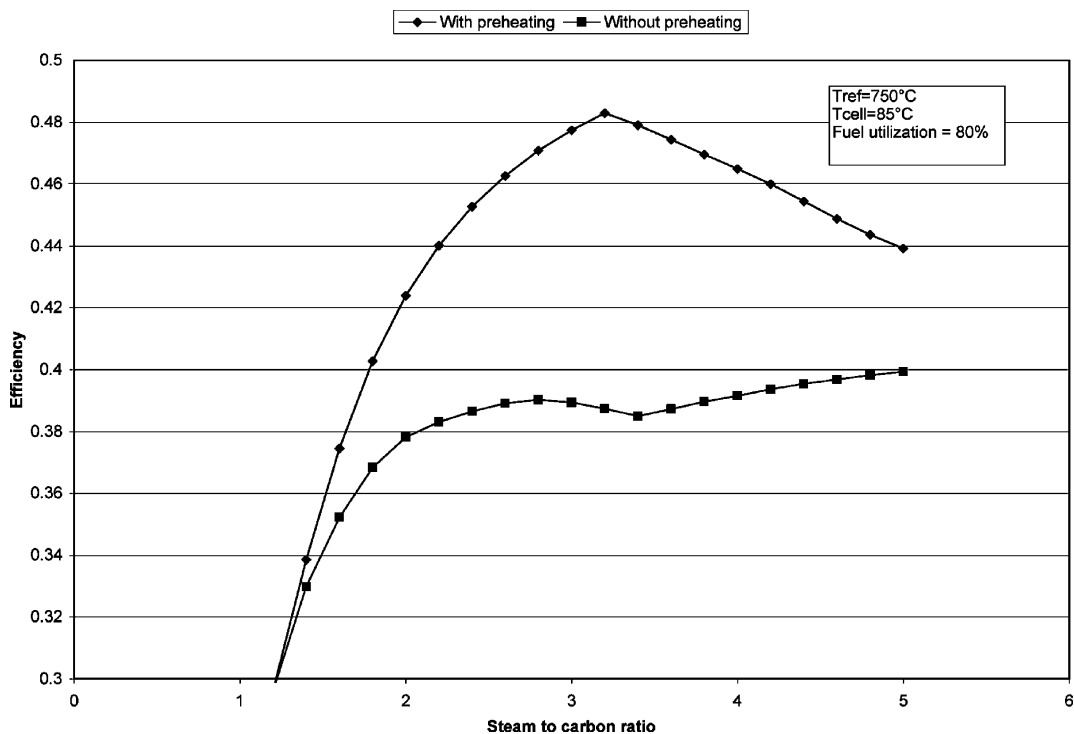


Fig. 15. The efficiency for a fuel utilization of 80% with and without preheating.

content in the available heat for combined heat and power purposes.

3.6. The influence of the fuel utilization in the cell (η_{comb})

The fuel utilization in the cell is an important factor that affects the balance of the plant and therefore the global efficiency of the system. A higher fuel utilization leads to a decrease of the available energy for post-combustion leading therefore to the possible increase of the additional fuel requirement. This may be compensated by an increase in the fuel cell power production. The process is less influenced by the fuel utilization with the higher reforming temperatures (Fig. 14). The difference decreases when the steam to carbon ratio increases. In fact, for values above a steam to carbon ratio of 3, the curves are superposed. For lower reforming temperatures, the situation is different and the differences are much bigger.

At a constant steam to carbon ratio of 1.5, the difference reaches up to 10% in terms of system efficiency. For higher steam to carbon ratio, the difference is attenuated. The importance of the fuel utilization rate is higher for $T_{o,\text{SMR}} = 750^\circ\text{C}$ than for 800°C . The fuel utilization has also an influence on the self-sufficient limit since the depleted fuel has a lower hydrogen content for higher fuel utilization rate. This is shown in Fig. 11. A lower fuel utilization does not need additional firing to reach the maximum efficiency of the system, but in this case the steam to carbon ratio is much higher. This might simplify the design and the control of the system. One should remind

that these calculations are made with a stoichiometric combustion. Increasing the air excess will lower the self-sufficient limit.

The influence of the air and depleted fuel preheating has been analyzed in Fig. 15 for a fuel utilization rate of 80%. This figure should be compared with Fig. 10 that refers to a fuel utilization of 95%. The efficiency gain obtained by preheating the depleted fuel and the combustion air to a temperature of 500°C is of 9 points of efficiency (from 39 to 48%) which is quite considerable. The air and depleted fuel preheating heat exchange before combustion is therefore an important device in order to maximize the overall efficiency of the system especially when lower fuel utilization are expected in the cell. The preheating will compensate the efficiency losses.

4. Conclusions

A combined simulation and process integration model has been developed to optimize the performances of a proton exchange membrane fuel cell system for stationary applications. The process integration techniques have been used to model the heat exchanger network without having to consider its configuration. This approach reveals to be very interesting in order to study the impact of the major decisions parameters (steam to carbon ratio, reforming and cell temperatures). The interest of the process integration techniques stands in the fact that it is able to identify heat exchange bottlenecks in the system (pinch points) and to propose process modifications that will increase the overall

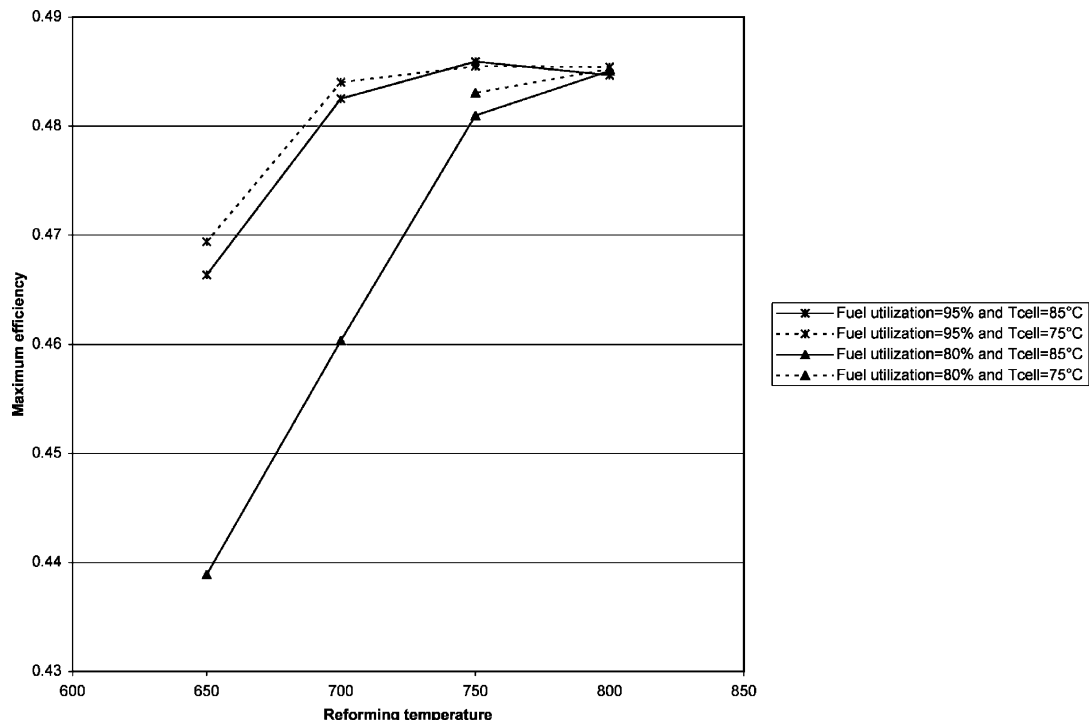


Fig. 16. Maximum system efficiency as a function of the reforming temperatures for different cell temperatures and fuel utilizations.

efficiency of the system (saturation/preheating of the natural gas feed, combustion preheating). The combined use of process modeling using commercial software and process integration techniques implemented into an optimization frame work leads to propose a new integrated system structure whose optimum is not the optimum of the separated subsystem.

The model has been used to optimize the system performances and analyze the sensitivity of the decision parameters with respect to the major unknowns of the future cell performance (e.g. fuel utilization in the cell). The results are not only optimal decision parameters values but also optimal process configurations. In Fig. 16, we present the evolution of the optimal efficiency as a function of the steam methane reformer temperature for different values of the fuel utilization and the cell temperature. Each point in the curves correspond to an optimal value of the steam to carbon ratio and to a given process configuration (i.e. an investment) that remains to be determined more accurately by designing the corresponding heat exchange system and modeling the integrated system.

The combined model has also been used to study the influence of the technological constraints to be considered for the cell (e.g. the water management issues, fuel utilization) and deduce the corresponding optimal process configurations.

The solutions obtained from the application of the method to the TPMEFC system feature total efficiency as high as 49% that are much higher than the one of the original system design.

The results of this preliminary analysis will be used to design the heat exchange network for the system and per-

form a more accurate thermo-economic optimization where the trade-off between the efficiency increase w.r.t. investment will be considered. The advantage of the adopted approach stands in the fact that by using process integration techniques the changes in the heat exchange system configuration are allowed and the structure will adapt according to constraints trade-off.

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