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STUDY OF A HYBRID MOLTEN CARBONATE FUEL CELL AND GAS TURBINE CYCLE

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Abstract.

The continuous increase in energy demand coupled with greater knowledge about the environmental impacts of the most used processes for the energy generation has driven the development of technologies to promote efficiency in these processes. Hybrid systems for thermal machines that include the use of fuel cells have been proposed and studied with promising results. Molten carbonate fuel cells (MCFC) present numerous advantages compatible with the current demands for the energy production in a sustainable and commercially competitive way, regarding to other processes such as thermal plants. However, MCFC susceptibility to the deterioration of its components implies a reduced useful lifetime, being an obstacle to the potential of its economic attractiveness. In recent years, there have been advances in the choice of materials used in the fuel cell constitution to circumvent this problem. In addition, in-depth studies of the operation variables considered to be determinant in their efficiency are intended to compensate investments by increasing the cost-benefit ratio of the fuel cell, given its intrinsic potential. This paper presents an analysis of the main variables on the efficiency and power generation of a hybrid system comprised of a molten carbonate fuel cell and a gas turbine through the determination of fuel cell design points and subsequent hybrid system modeling. Results indicate that increase in temperature and operating pressure of the fuel cell as well as the fuel reform rate improves the performance of the hybrid system as a whole. Variation in the utilization rate, however, did not determine an expressive increase in the efficiency of the system.

Keywords: molten carbonate fuel cell, gas turbine, hybrid system, energy analysis.

1. NOMENCLATURE

A	- Area [m ² or cm ²]	δ	- Equivalent thickness of the diffusion layer [m]
E	- Operational cell voltage [V]	η	- Efficiency
F_n	- Faraday constant [96487.309 C/mol]	η_{GT}	- Isentropic efficiency of the gas turbine [-]
j	- Current density [A/m ²]	ρ_m	- Resistivity [$\Omega \cdot m$]
K_0	- Reaction equilibrium constant [-]	Subscripts	
\dot{m}	- Mass flow rate [kg/s]	0	- Standard state (1 atm and 298.15K)
P	- Pressure [kPa or MPa]	ac	- Alternative current
p_i	- Partial pressure of the component i [kPa]	an	- Anode
\bar{R}_g	- Universal gas constant [8.315 kJ/kmol.K]	cat	- Cathode
R_t	- Total resistance of the cell components [Ω]	∞	- Environment
T	- Temperature [K]	C	- Compressor
T_0	- MCFC reference temperature [K]	Comb	- Combustor
U	- Utilization [-]	F	- fuel
W_{FC}	- Power density [kW/m ²]	FC	- Fuel cell
\dot{W}	- Power [kW]	GT	- Gas Turbine
ΔG_r	- Gibbs free energy of the reaction [kJ/kmol]	ox	- oxidant

2. INTRODUCTION

The World's energy consumption, only between 2012 and 2015, rose from 22800 TWh to almost 24367 TWh (6.87%), according to the International Energy Agency (IEA, 2015), although the carbon dioxide emission has been maintained practically stable between those years (31004 Mt to 31465 Mt of CO₂ – increase of about 1.5%), according to the Global Energy Statistical Yearbook (Enerdata, 2017). The replacement of the coal use by natural gas use coupled

with a decrease in economic growth and the development of energy generation processes in countries with high energy demand, such as China and the United States, have been influencing to obtain these results. The renewable energy is still a challenge due to its installation cost. The optimization of the processes of energy generation through the fuels combustion justifies an immediate option for the pollutants emission reduction.

Hybrid systems, according to Hauschild (2006), are systems formed by two or more sources of energy production working together to meet the demand of a common consumer and have been observed as an important alternative for the optimization of energy generation processes. The main aspect studied in the development of these systems is the combination of technologies that use different mechanisms of energy generation. Starting from the same energy source (primary source), the energy not used in the energy generation by one technology is used by the other, network efficiency. In this scenario, hybrid cycles using fuel cells and thermal machines have been proposed since 1989 (Ide *et al.*, 1989; McLarty *et al.*, 2014). In order to reduce the pollutants emission and increase the energy generation efficiency, the hybrid systems can achieve efficiencies of approximately 80% PCI base, when using natural gas (McLarty *et al.*, 2014).

Fuel cell produces electrical energy through oxy-fuel reduction reactions; thus presenting a non-limitation to the Carnot Cycle and its efficiency. High temperature fuel cells also have the advantage of promoting the capture and use of CO₂ (Beér, 2007) and the flexibility of fuel utilization through the possibility of internal reforming and gasification of energy sources such as coal and biomass (Naidja *et al.*, 2003; Freni, 2001; Richards *et al.* 2001; Aravind, 2012). However, in this type of fuel cell almost half of the energy from the fuel is lost in the form of heat. The use of thermal engines within the hybrid cycle is aimed at harnessing the heat generated.

Hybrid systems with solid oxide (Roberts; Brouwer, 2006) and molten carbonate fuel cells (FuelCell Energy Inc., 2006) were successfully demonstrated in the sub-MW size class and reached efficiencies of 53% and 56%, PCI base, respectively. Another system involving a molten carbonate fuel cell associated to a gas turbine achieved 74.4% efficiency, PCI base. A unified hybrid fuel cell and thermal engine model was demonstrated by Zhang *et al.* (2014) reproducing important results of the literature. Some limiting aspects such as production cost and durability, especially in fuel cells, however, still represent an important challenge for the diffusion of these systems (Energy Nexus Group, 2008; Shipley, 2004).

The aim of this paper is to evaluate thermodynamically a hybrid system comprised of a molten carbonate fuel cell and a gas turbine. This evaluation seeks the analysis of the main variables on the efficiency and power generation of the hybrid system through the determination of fuel cell design points and subsequent hybrid system modeling. First, an evaluation of the operation parameters of the molten carbonate fuel cell at the design point was carried out, identifying those that most influence in the energy production of the device. Afterwards, an analysis of the fuel cell - gas turbine hybrid cycle was accomplished, using Cycle-Tempo software from Delft University of Technology. The validation process was performed with the comparison between the results of the fuel cell / hybrid system model and those available in the literature. The results indicate that the variation in the operational temperature and pressure of the fuel cell, and the fuel reform rate improve the performance of the hybrid system as a whole. The variation of the utilization rate did not determine an expressive increase in the efficiency of the system.

3. DESCRIPTION AND MATHEMATICAL MODELING OF THE HYBRID SYSTEM

Hybrid fuel cell / gas turbine systems show a potential for high efficiency, low pollutant emissions, fuel flexibility and dynamic responsiveness (Roberts; Brouwer, 2006; McLarty *et al.*, 2012). Both the experimental and theoretical analyzes of such hybrid systems confirm this potential. The integration displayed in hybrid cycles shows synergies that are not present in the separated cycles, where the fuel efficiency are greater than the fuel cell or the gas turbine alone. The advancement of high temperature fuel cell technology in the last decade has been significant and the expectation is that it will become commercially feasible in the coming decades.

3.1 Molten Carbonate Fuel Cell (MCFC)

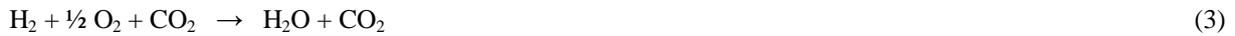
Among the types of fuel cells currently available, MCFC is especially applicable in stationary processes. This fact is due to the operating temperature of the fuel cell around 923.15 K, which is sufficient for the electrochemical conversion processes without the need for noble metals catalysts. The high temperature also favors the reforming reaction of conventional fuels that can be realized internally in the own cell, simplifying the system. Since part of the energy used for the reforming processes comes from the heat generated in the cell itself, the efficiency of the system is increased. Additionally, CO and CO₂ are welcome constituents in the electrochemical reactions that occur in the MCFC and, together with the possibility of fuel reforming, make possible the use of a wide range of fuels (Sundmacher *et al.*, 2007).

The half-cell reactions of the MCFC are:





Combining the half-cell reactions of the anode and the cathode gives the overall reaction of the fuel cell:



The performance of the fuel cell can be described by knowing the equilibrium potential (E_{eq}) of the cell and the irreversible losses (α_{cat} and α_{an}) of the process (Chan *et al.*, 2001):

$$E = E_{eq} + j(R_t + \alpha_{an} + \alpha_{cat}) \quad (4)$$

$$E_{eq} = E^0 + \frac{R_g T}{2 F_n} \ln \left(\frac{p_{\text{H}_2} p_{\text{O}_2}^{0.5} p_{\text{CO}_2, \text{cat}}}{p_{\text{H}_2\text{O}} p_{\text{CO}_2, \text{an}}} \right) \quad (5)$$

$$E^0 = \frac{R_g T}{2 F_n} \ln(K_0) \quad (6)$$

$$K_0 = \exp \left(\frac{-\Delta G_r}{R_g T} \right) \quad (7)$$

$$R_t = \delta \left\{ a \cdot \exp \left[b \left(\frac{1}{T} - \frac{1}{T_0} \right) \right] \right\} \quad (8)$$

The total resistance (R_t) is related to the cell building material, δ is the equivalent thickness of the diffusion layer and a and b are constants. Calculation of the equilibrium potential predicts that increasing the operating pressure of the fuel cell results in increased partial pressures of the reactants, the solubility and mass transport rates. However, an excessive increase in the operating pressure will favor the carbon deposition (Boudouard reaction) and suppress the reforming of methane to form H_2 by the Le Chatelier principle, as Hirschenhofer (1998) emphasizes, reducing the gases flow and the fuel availability for the reaction, respectively.

Regarding the irreversible losses (α_{cat} and α_{an}) of the process, Selman and Lin (1993) demonstrate that the polarization in the electrodes can be obtained by:

$$\alpha_{an} = 2.27 \cdot 10^{-9} (p_{\text{H}_2})^{-0.42} (p_{\text{CO}_2})^{-0.17} (p_{\text{H}_2\text{O}})^{-1.00} \exp \left[\frac{53500}{R_g} \left(\frac{1}{T_0} - \frac{1}{T} \right) \right] \quad (9)$$

$$\alpha_{cat} = 7.51 \cdot 10^{-10} (p_{\text{O}_2})^{-0.43} (p_{\text{CO}_2})^{-0.09} \exp \left[\frac{53500}{R_g} \left(\frac{1}{T_0} - \frac{1}{T} \right) \right] \quad (10)$$

Ohmic polarization occurs due to the resistance of both the carbonate ions flow in the electrolyte and the electron in the electrodes and interconnectors. Activation polarization occurs due to an energy barrier that the reagents must overcome in order for the reaction to occur. On the other hand, the concentration polarization results from the difference between the rate of diffusion of the reactant gases by the electrodes and the reaction rate (Zhang, 2014). Therefore, it is evident the influence of the operating pressure and temperature, the composition of the gases used and their use in the performance of the cell. It is necessary to observe the influence of these parameters in the hybrid system to analyze the best relationship among them.

The values of the gas concentrations required in the calculation of the anode and cathode resistances were obtained through the results of chemical equilibrium, fuel reforming and combustion reactions, respectively, in the CEA program (Chemical Equilibrium with Applications, 2018). The values of the equilibrium constants at different fuel cell temperature and pressure conditions were also calculated using CEA.

Finally, the electric energy density produced by the fuel cell can be calculated as (Chan *et al.*, 2002):

$$W_{FC} = E \cdot j \quad (11)$$

Where: W_{FC} is the fuel cell power density produced [$\text{kW} \cdot \text{m}^{-2}$], E is the operational cell voltage [V] and j is the current density [$\text{A} \cdot \text{m}^{-2}$].

3.2 Hybrid system simulation

Pure methane gas (CH_4) was used as the fuel for hydrogen production, assuming two steam/carbon ratio (S/C of 2.0 and 3.0) plus a temperature range of 500°C to 973.15 K and pressure range from 1 to 6 atm, to observe the influence of

each variable in the system. Furthermore, the partial pressure was obtained based on the average molar concentrations of the fluid components at the inlet and outlet of each electrode, so that the fuel utilization rate ($U_{f_{an}}$) in the anode was also a parameter to be considered and was analyzed in this study with a variation of 25% to 100%.

The physical properties of the MCFC was obtained from the literature (Hirschenhofer *et al.*, 1994; Zhang *et al.*, 2014) and the input parameters of each component of the hybrid system are listed in Tables 1 and 2, respectively. The process was considered adiabatic, without losses, in the electrical generator connected to the fuel cell output.

Table 1 – Characteristics of the MCFC components (Hirschenhofer *et al.*, 1994; Zhang *et al.*, 2014).

Flowing length in electrolyte	δ (m)	0.0017
	α (m)	0.0294
	A (m ²)	0.00005
	b (K)	3016
Oxidant utilization	U_{ox} (-)	0.5
MCFC reference temperature	T_o (K)	923.15

Table 2 – Equipment entry parameters.

Reference	Name	Parameters
A	Fuel source	CH ₄ : 100%. Pressure = 1.013 bar. $\dot{m} = 0.05$ kg/s
B	Air source	N ₂ : 77.29%, O ₂ : 20.75%, H ₂ O: 1.01%, CO ₂ : 0.03% and Ar: 0.92% Pressure = 1 atm and temperature = 298.15 K.
T	Turbine	$\eta_{GT} = 0.80$ $\eta_{GTM} = 1$
G	Combustor	$\Delta_p = 0$ atm
S	Stack	$P_{in} = 1$ atm
C ₁	Compressor	$\eta_C = 1$ $\eta_{CMI} = 1$

The current density function of the MCFC was plotted in the Microsoft Excel® program as well as the anode and cathode resistance based on different pressure inputs, reforming rate, fuel utilization and fuel cell operating temperature. The data were compiled for the respective component in the Cycle-Time™ program for analysis of the hybrid system.

4. RESULTS AND DISCUSSION

Figure 1 presents the results for the ohmic polarization of the MCFC for the anode (a) and cathode (b) as a function of the operating pressure, steam/carbon ratio varying from 2 to 3 and fuel utilization in the anode (U_{fan}) ranging from 0.25 to 1.00. The results considered the temperature of 923.15 K.

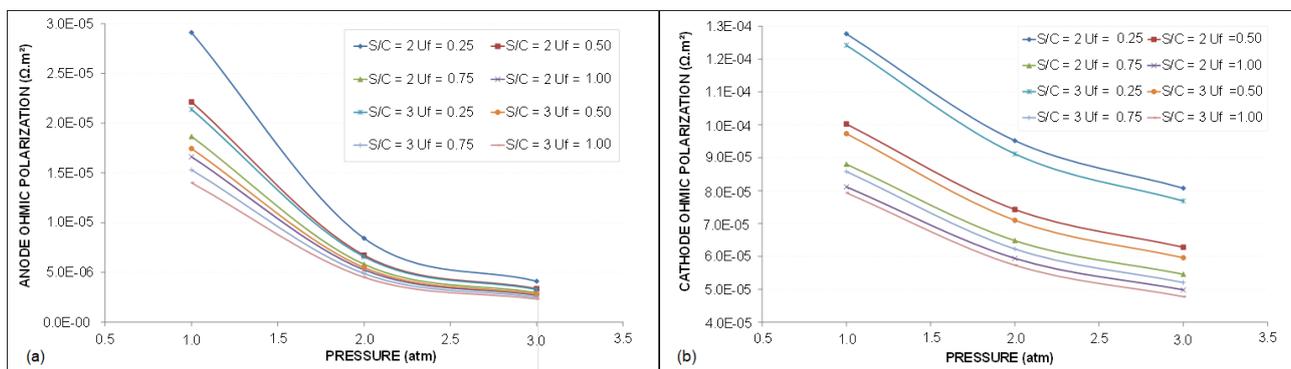


Figure 1. Results for ohmic polarization in the anode (a) and cathode (b) as a function of the pressure and steam to carbon ratio (S/C) for the operating temperature of 923.15 K.

The results show that the resistance in the electrodes decreases exponentially with increasing pressure and the steam/carbon (S/C) ratio, followed by the fuel utilization in the anode. This was expected based on: (i) the higher the S/C ratio, the greater the conversion of the fuel to H₂, favoring an increase in the availability in the anode and reducing the concentration resistance; (ii) a higher rate of fuel utilization also implies higher activity on this electrode; (iii) the

increase of the pressure favors the availability of H_2 at the catalyst sites, besides promoting the diffusibility of the ions by the electrolyte after the reaction at the electrode, releasing the sites so that the reaction occurs at higher rates. The reduction in resistance in the electrodes will imply in a smaller reduction in the operating voltage of the cell and, in turn, in a higher power density. These results confirm with that described by US DEO (2004), which emphasizes that the increase in operating pressure results in increased fuel cell voltage due to the increased solubility of gases and increased mass transport due to the increase in the partial pressures of the gases.

Regarding the influence of the fuel cell operating temperature on the resistance, already demonstrated by Leal *et al.* (1999), the results obtained are expressed in Fig. 2, for which the total resistance of the cell was given as a function of the temperature (ranging from 773.15 K to 973.15 K) at different pressures. It is possible to observe that the temperature has a greater influence on the resistance of the fuel cell than the operating pressure. With these results, it was possible to determine the points of greatest energy production by the fuel cell, shown in Fig. 3. These results was used in the calculation of the power produced by the hybrid system in the Cycle-Tempo™ software. The parameters obtained and used for the fuel cell model was its operating temperature and pressure, current density, output voltage, fuel and oxidant utilization, and the internal reforming rate.

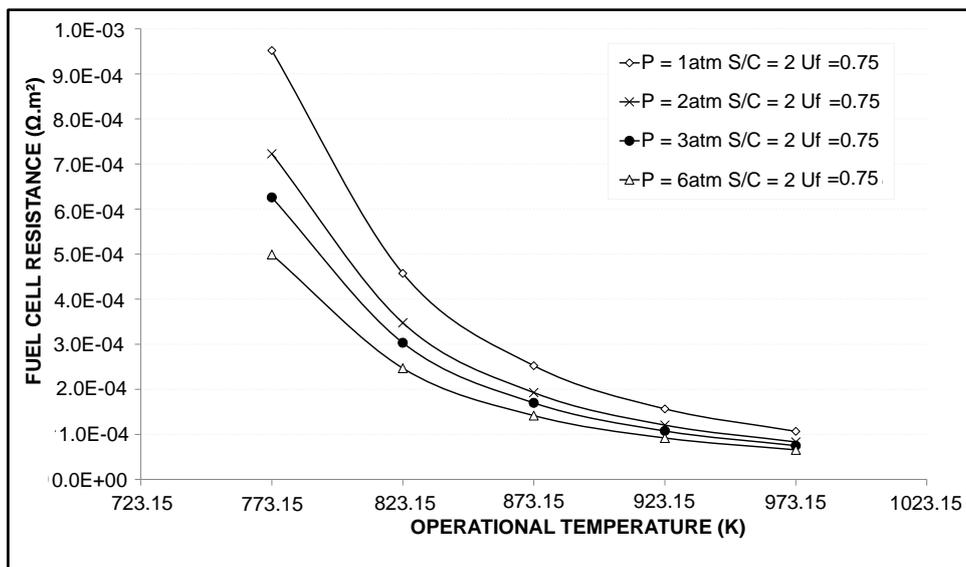


Figure 2. Results of the fuel cell resistance as a function of the operating temperature ($773.15 \text{ K} \leq T \leq 973.15 \text{ K}$) and pressure (from 1 atm to 6 atm) setting the steam to carbon ratio of 2.0 and fuel utilization of 0.75.

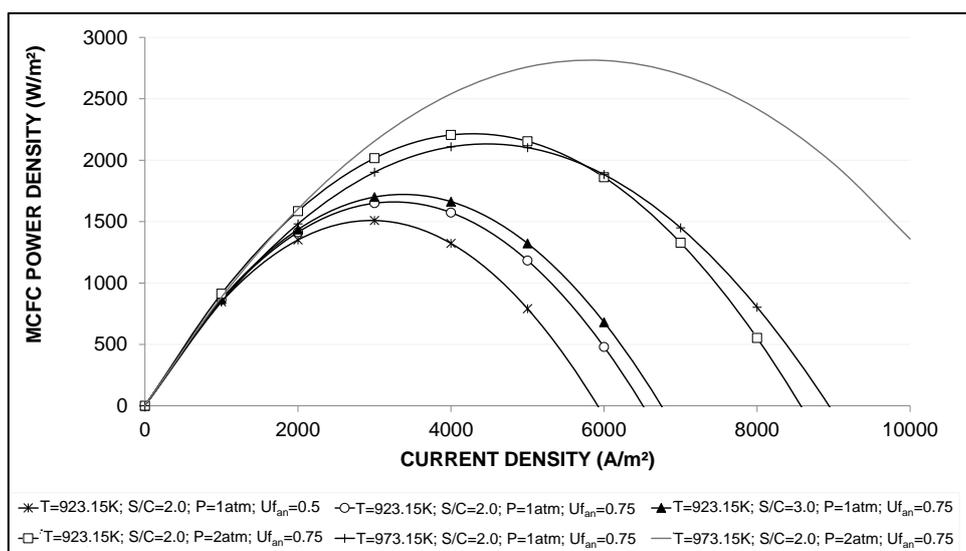


Figure 3. Results of the fuel cell power density as a function of the current density ($0 \text{ A/m}^2 \leq j \leq 10000 \text{ A/m}^2$), operating pressure (1 atm and 2 atm), fuel utilization of 0.50 and 0.75, and steam to carbon ratio of 2.0 and 3.0.

As shown in the results of the total resistance (Fig. 2), the increase in pressure and in the operating temperature of the fuel cell results in significant increases in the power production (Fig. 3), being the association between the increase of these two parameters. It can also be verified, according to Fig. 3, a parabolic behavior of the relationship between power and current density, and for higher temperatures, the higher the power limit will be. For the temperature of 973.15 K, at a pressure of 1 atm; U_{fan} of 0.75 and S/C ratio of 2, the power reached a maximum value of 2108 W/m² at 4000 A/m², while for the temperature of 923.15 K a power output reached 1509 W/m² at 3000 A/m². This means an increase of 39.7% in power density and 33.3% in the peak current density for a 7.7% change in temperature. In addition, for the temperature of 923.15 K and current density of 4000 A/m², the variation in the fuel utilization resulted in the increase of 19.0% (at 1 atm pressure and S/C ratio of 2) and 15.1% (at 1 atm pressure and S/C ratio of 3). The pressure increase from 1 to 2 atm resulted in increases of up to 40.2% (U_{fan} of 0.75 and S/C ratio of 2), while the increase in the S/C ratio increased by 9.2%, at pressure of 1 atm and U_{fan} of 0.5, 5.7%, at a pressure of 1 atm and U_{fan} of 0.75.

The hybrid system was built in Cycle-Tempo® and the points of greatest energy production by the cell were inserted in the configuration to evaluate the influence of these parameters on the complete hybrid system. Figure 4 shows the schematic diagram of the hybrid system with the results obtained at a temperature of 973.15 K, operating pressure of the fuel cell at 6 atm, fuel utilization at the anode of 0.75 and S/C ratio of 3.0.

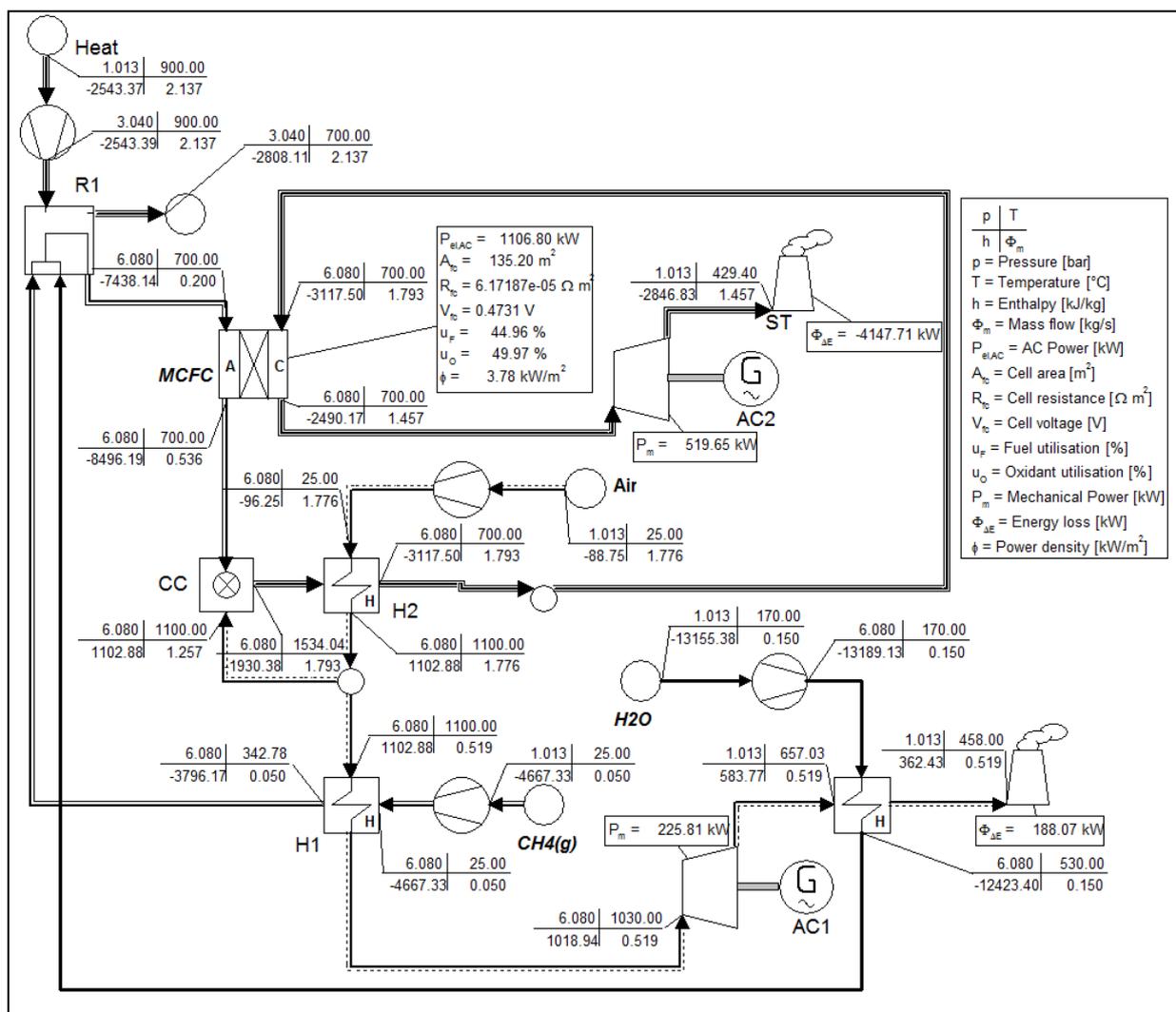


Figure 4. Schematic diagram of the hybrid system with the results obtained at a temperature of 973.15 K, operating pressure at 6 atm, fuel utilization of 0.75 and S/C ratio of 3.0.

The results obtained in Cycle-Tempo software and Excel software for the power density showed a good agreement in all temperature range simulation. However, the total fuel cell resistance showed a difference of 20% in the temperature of 923.15 K. Figure 5 shows the results of Cycle-Tempo software for power delivered and net delivered power efficiency as a function of operating pressure from 3 atm to 6 atm, temperature of 923.15 K and 973.15 K, fuel utilization of 0.75, and steam to carbon of 2.

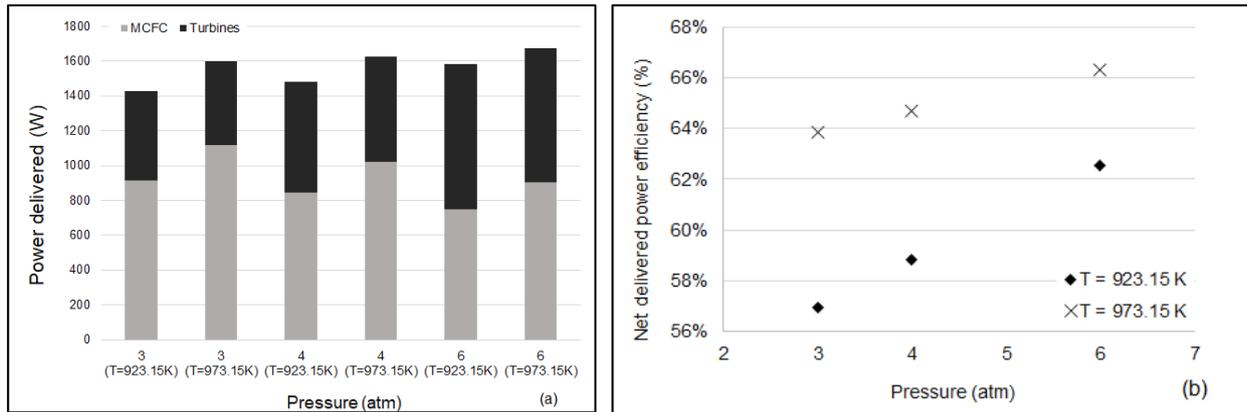


Figure 5 – Results of power delivered (a) and net delivered power efficiency (b) as a function of pressure (3 atm ≤ P ≤ 6 atm), temperature of 923.15 K and 973.15 K for fuel utilization of 0.75 and steam to carbon of 2.

Figure 5 relates the delivered power to the pressure variation for temperatures of 923.15 K and 973.15 K. For the same fuel mass flow rate in the system, it is possible to observe that the variation in the operating temperature of the fuel cell resulted in a greater increase in the total power of the hybrid system, when compared to the results of the pressure increase. The increase in temperature from 923.15 K to 973.15 K resulted in a maximum increase of 12.0% in delivered power and corresponding to a 6.95% system efficiency increase, demonstrated in Fig. 5(b). This effect was a consequence of an increase in the delivered power of the fuel cell, already expected and described with the individual modeling of the device.

Increase in pressure from 3 to 4 atm resulted in a maximum increase of 3.86% in delivered power, corresponding to an increase of 2.22% system efficiency (T = 923.15 K). This is because, although an increase in the cell's power density was observed for the same mass flow rate in the system, the pressure negatively influenced the value of the total delivered power of the fuel cell. For this pressure variation, there was a reduction of 7.79% in the delivered power of the MCFC, although the delivered power of the gas turbine increased by 24.7%, also showing a higher effect of the pressure on the turbine operation regarding to the working fluid temperature. Results of turbines and MCFC power delivered for a variation in steam to carbon ratio, operating temperature, fuel utilization variation, are shown in Fig. 6.

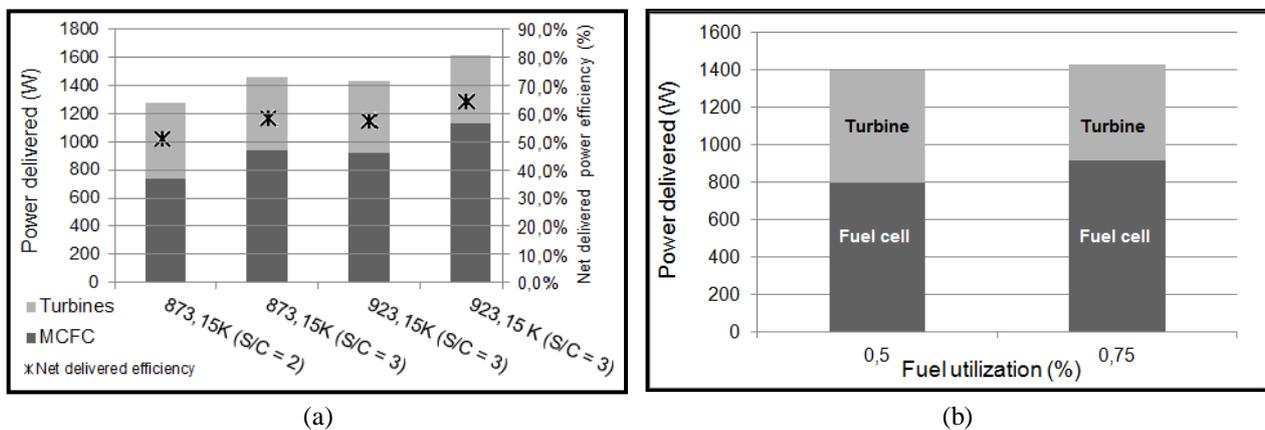


Figure 6 – Results of the power delivered and efficiency of each main component as a function of the operating temperature and steam to carbon ratio (a), and the fuel utilization with temperature of 923.15 K, both in the pressure of 3 atm.

Results presented in Fig. 6 (a) and (b) indicate that the variation in the reform rate from 2 to 3 promoted an increase in the power delivered by the MCFC for both temperatures of 873.15 K and 923.15 K compared (27.11% and 23.0%, respectively). On the other hand, this increase resulted in a reduction in power delivered by the turbine pair, with a reduction of 4.5% for the temperature of 873.15 K and 6.0% for the temperature of 923.15 K. Nevertheless, there was an increase in total delivered power and in the system's network efficiency. For 873.15 K, an increase of 13.7% in the delivered power and 7.0% in the network efficiency was observed, and at 923.15 K, 13.0% and 7.17%, respectively.

In turn, it was possible to conclude that the variation in the fuel utilization rate from 0.50 to 0.75 also promoted a 14.5% increase in the power delivered by the MCFC to the system studied at a temperature of 923.15 K and a pressure of 3atm. As for the steam / carbon ratio, this increase resulted in a reduction in the power delivered by the turbine pair by 14% in the delivered power. The result was expected, given that the increase in fuel utilization by the cell reduces

the availability of fuel to be used in generating turbine power. The final result was a slight increase in power and total delivered network efficiency (2.3% and 2.4%, respectively).

5. CONCLUSIONS

It becomes clear that, in the design point, the fuel cell is the main energy generator, while both turbines are, together, the second, but they are comparable to the pressure of 6 atm and a temperature of 973.15 K. It was also possible to conclude that, for the hybrid system, the increase in cell operating temperature and fuel reform promoted an increase in cell and system delivered power, although the increase in temperature did not significantly influence the power delivered by the pair of turbines and the reform has resulted in a decrease of this. The increase in pressure, on the other hand, resulted in an increase in the power delivered by the pair of turbines and in a decrease of the cell, maintaining the fixed mass flow of fuel, although the power density of the cell increases. Moreover, both the increase in the cell's operating temperature and in the pressure and reform rate culminated in an increase in the efficiency of the hybrid system. The variation in the rate of fuel utilization by the cell, on the other hand, did not demonstrate an expressive increase in the efficiency of the system or in the total delivered power, since the increase in energy production by the cell was affected by the reduction of energy production by the turbines.

Further research is needed to evaluate the influence of the studied parameters on carbon deposition in the MCFC and evaluation of its lifetime for economic analysis of the increase of these variables in the system.

6. ACKNOWLEDGEMENTS

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8. RESPONSIBILITY NOTICE

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