Modelling of a HTPEM-based micro-combined heat and power fuel cell system with methanol

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Abstract

A fuel cell-based combined heat and power system using a high temperature proton exchange membrane fuel cell has been modelled. The fuel cell is fed with the outlet hydrogen stream from a methanol steam reforming reactor. In order to provide the necessary heat to this reactor, it was considered the use of a catalytic combustor fed with methanol. The modelling aims to fit the hydrogen production to the demand of the fuel cell to provide 1 kW_e, maintaining a CO concentration always lower than 30,000 ppm. A system with 65 cells (45.16 cm^2 cell area) stack operating at 150 ºC and hydrogen utilization factor=0.9 (with O_2/methanol ratio=2 at combustor; H_2O/methanol ratio=2 and temperature=300 ºC at reformer) needed a total methanol flow of 23.8 mol h^{-1} (0.96 L h^{-1}) to reach 1 kW_e, with a system power efficiency (LHV basis) ca. 24 % and a CHP efficiency over 87 %. The ability to recycle the non-converted hydrogen from the fuel cell anode to the combustor and to use the heat produced at the fuel cell for obtaining hot water increased the global energy efficiency.

Keywords: fuel cell system, micro-CHP, HTPEMFC, modelling, methanol steam reforming.

1. Introduction

Decentralized power generation combined with heat supply, named as combined heat and power (CHP) is a feasible technology developed to increase the energetic efficiency and the safety of energy supply and to reduce costs in order to produce energy in a more environmentally friendly way. For these reasons, the European Union and the governments of many countries promote the use of this kind of systems in order to fulfil the pollutants emission agreements assumed in the Kyoto protocol, both in the global framework and in every individual country [1].

One of the most interesting technologies to be used in CHP at low scale (μ-CHP) to generate both electricity and heat (hot water) are the fuel cells. Their use in a μ-CHP provides a significant reduction in CO_2 emissions in comparison with the conventional condensing boilers in residential applications. Among fuel cells, proton exchange membrane fuel cells (PEMFC) are considered as the most efficient for working at low temperatures (<100 ºC). However, the possibility of increasing the operation temperature of the fuel cell at values around (or higher than) 150 ºC should be more suitable to reach better energy efficiency [2].

This option has motivated the development of high temperature proton exchange membrane fuel cells (HTPEMFC). But there are more compelling reasons to use this kind of fuel cells [3]: i) electrochemical reaction kinetics in both electrodes are much better; ii) water management at cathode is easier because only gas steam phase is present; iii) fuel cell cooling system is simpler due to a higher temperature gradient between fuel cell and refrigerant; iv) waste heat recovered can be used for example for producing hot water, v) CO concentration limits in the fuel can be increased because the poisoning of Pt catalysts decreases with temperature.

Moreover, fuel cells systems that work at temperatures lower than 80 ºC exhibit a poor capacity to extract the generated heat and it requires exchangers with great surface areas, which implies an increase in system weight, volume and cost. The use of HTPEMFC with temperatures higher than 150 ºC makes easier the heat transfer. As a result, the cooling system is simpler and the global efficiency becomes higher when temperature increases.

Power generation efficiency of a low temperature PEMFC with a reformer system is a drawback during partial load operation [4]. While the PEMFC usually increases its power efficiency at partial loads (that means lower current density but higher voltage), other parts of the system are not at optimal operation and their efficiencies decay. Some authors have proposed different ways to overcome this effect, like using the excess power at full load (when partial loads are required) to electrolize water and produce hydrogen or using smaller and several reformers and fuel cells (at partial load at least one of them is working at full load) [5]. In this way, the HTPEMFCs (as water is already in vapour phase) require less number of auxiliary equipments like the humidifier, compressor and radiator. This is another advantage of HTPEMFC systems because they diminish the decay in the overall efficiency due to the partial load operation.

Among candidate fuels to use in fuel cell systems are natural gas, methanol, ethanol and dimethyl ether. Methanol does not require special storage conditions, has high hydrogen to carbon ratio (4:1) and does not present C-C bonds, which minimizes the coke formation [6]. Moreover, methanol can be obtained from biomass (bio-methanol) [7].

Hydrogen production can be carried out by methanol steam reforming reaction (Eq. 1) [8].

$$CH_3OH(g) + H_2O(g) \rightarrow CO_2\,(g) + 3H_2\,(g) \qquad (1)$$

This reaction produces an outlet stream rich in hydrogen (~75%) but the endothermic character of the reaction (ΔH^o_{298} = 49.5 kJ mol^{-1}) forces to supply heat from an external source (i.e. a combustor). Some methanol reforming-based HTPEMFC systems have shown that operation with high CO contents is possible with reasonable voltage outputs in a lab-made system [9] and a commercial device [10], including a design with coupled reformer and fuel cell for heat integration purpose [11]. However, no data is available about fuel cell and specially overall system theoretical efficiencies.

In the present work, a μ-CHP system that provides 1 kW_e and heat for residential applications is proposed using methanol as a fuel to feed a steam reforming reactor for obtaining the necessary hydrogen flow rate to generate heat and electricity in a HTPEMFC. For this propose, a model with AspenOne Hysys® is designed. In order to calculate the produced heat and electricity, a semi-empirical model is used for the fuel cell.

2. Model development

The system consisted of three main parts: a combustor, a methanol steam reforming reactor and a HTPEMFC (Figure 1). Several pumps, centrifugal fans and heat exchangers must be incorporated to the system in order to move the fluid streams and to transfer heat among different devices and streams. The characteristics of the main equipments should be designed in detail to obtain the most efficiency of the system maintaining a compact arrangement.

2.1 Catalytic combustor

The catalytic combustor must both start up the system and provide enough heat to keep the methanol steam reforming reaction at the desired temperature using methanol as fuel. Moreover, the HTPEMFC anode outlet stream is recycled to this equipment in order to make the most of the hydrogen and methanol that remains unreacted from the fuel cell. There are catalysts able to accelerate the combustion of both methanol and hydrogen with air and the resultant mixes of both fuels. The use of $Pt/\gamma\text{-}Al_2O_3$ and $Pt\text{-}Mn/\gamma\text{-}Al_2O_3$ pellets in a fixed bed reactor were reported, the first one applied to 1/3 of the bed as primary ignition catalyst and the other as a secondary combustion catalyst [12]. The option “Gibbs reactor” was chosen to simulate the combustion reaction because this assumes minimization of energy in products and it is a good reference value for real situations.

The combustor was designed with internal heat exchangers to preheat combustor and reformer inlet streams and to heat the reformer. The energy required in each heat exchanger was adjusted (AspenOne Hysys®) by burning the necessary fuel (methanol) amounts at different zones of the combustor. Thus, both operating and gas outlet temperatures were kept conveniently low. Residual heat from the combustor exhaust was taken to produce hot water (60 ºC).

2.2 Methanol steam reforming reactor

In order to carry out the methanol steam reforming reaction is necessary to transfer the heat generated in the combustor to the reactor. This is achieved by leading the combustor outlet stream through the reactor to exchange the heat. One strategy to significantly improve this process is the use of a fluidized bed reactor [13]. This favours the catalyst circulation along the bed avoiding the formation of temperature profiles. Several catalysts have been proposed in literature but it is assumed that copper-based ones (especially $CuO-CeO_2$) exhibit high activity and excellent selectivity to both H_2 and CO_2 [14]. It has been proven that $CuO-CeO_2$ catalyst is significantly affected by copper loading and different authors have studied the optimum composition of $CuO-CeO_2$ prepared by co-precipitation. So, Liu et alt. [15] obtained better results for a 26.9 wt.% Cu loading (50 at.% Cu) whereas Oguchia et alt. [16] found the best performance with 80 wt.% CuO loading (90 at.% Cu). Pan et al. [9] worked with a Cu/ZnO catalyst in a small reactor (2.4 cm in diameter and 19 cm long) and reached conversions close to the equilibrium ones (almost 100 %) at temperatures above 200 ºC, showing the feasibility of this catalyst use.

In the present work the reformer was simulated in AspenOne Hysys® by an equilibrium reactor, which approximates the reaction outlet to the equilibrium. HTPEMFCs can deal with high CO content. That implies high reforming temperatures and the thermodynamic equilibrium conversion is enhanced. The high water/methanol feed ratio (2 times the stoichiometric one) favours the almost total methanol conversion and avoids significant methanol presence at fuel cell anode.

The outlet reformer stream temperature must be reduced to get the fuel cell requirement, so excess heat can be recovered by preheating to some extent the water to the reformer.

2.3 HTPEMFC

The design of the suitable HTPEMFC can be adapted according to the material used in the different parts [17]: gas diffusion layers [18], electrocatalyst [19, 20], electrodes [21] and, specially, the membrane [22]. The bipolar flow field configuration have also influence on the operating conditions of the stack [23]. Moreover, the CO level in the fuel plays a key role in the performance of the fuel cell due to its strong adsorption on the surface of the Pt supported on the electrocatalyst, occupying hydrogen oxidation reaction sites [24].

The simulation of this equipment has been carried out taking into account a semi-empirical model proposed by the Aalborg University group [25-28]. The overall cell potential V_{cell} is calculated from the open circuit voltage V_0, taking into account the activation (Tafel equation), ohmic and diffusion-concentration polarizations (η_{act}, η_{ohmic} and η_{diff}).

$$V_{cell} = V_0 - \eta_{act} - \eta_{ohmic} - \eta_{diff} \tag{2}$$

$$V_{cell} = V_0 - RT/(n\alpha F)\cdot \ln((i+i_0)/i_0) - R_{ohmic}\cdot i - R_{conc}\cdot i/(\lambda_{air}-1) \tag{3}$$

where n is the number of electrons participating in the reaction, α is the charge transfer coefficient, i is the current density, i_0 is calculated by a specific equation, R_{ohmic} and R_{conc} are calculated from linear temperature dependent equations and λ_{air} is the cathode air stoichiometric ratio [26].

The heat loss at fuel cell can be obtained from the thermodynamic energy released from reaction [29].

$$q = (\Delta H_r/(nF) - V_{cell})\cdot i \cdot \text{cell area} \cdot \text{number of cells} \tag{4}$$

The expressions and the parameters values used were based on an extensive set of measurements performed by the authors [25-28] at temperatures from 120 ºC to 200 ºC, cathode stoichiometric ratios from 2 to 5 and CO concentrations from 0.1 to 10 %. However, they detected that for tests performed at 10 % CO permanent degradation seemed to occur at temperatures above 180 ºC. From data provided by Serenergy, Serenus 90-cell stack, the first commercially available, low pressure, air-cooled HTPEMFC system in the world, is able to work at CO levels up to 30,000 ppm [30]. So, in the present work, this value was chosen as an operation limit. In order to compare results, the same cell area (45.16 cm^2) [28] was used. Using this dimension and the parameters provided by the Aalborg group it was noticed that 65 cells were enough to reach the required electrical power, with a current density of 0.75 A cm^{-2}. In order to obtain a compact CHP system, this number was considered suitable. A heat recovery from the fuel cell, for producing hot water (60 ºC), has been considered in the system, in order to improve the global energy efficiency (CHP efficiency).

2.4 System simulation

The complete arrangement of equipments was done using the AspenOne Hysys® program. As well as the three main equipments mentioned above, several heat exchangers were added in order to make the most of the heat of every hot stream. As it was previously mentioned, HTPEMFCs have waste streams with high enough temperatures to extract heat from them and increase the energy efficiency.

First at all, the fuel cell calculations were done using the above equations. These were included in the AspenOne Hysys® program file to simultaneously interact with the other parts of the system. The fuel cell was taken as a “conversion reactor”, with a conversion equal to the hydrogen utilization factor (reacted/fed hydrogen ratio at fuel cell).

The methanol flow rate to feed the reformer was calculated from the hydrogen necessary to reach the 1 kW_e at the fuel cell, taking into account the hydrogen utilization factor whose values ranged from 0.7 to 1.

The combustor was incorporated to the system to maintain the reforming temperature. A proper quantity of methanol should be added for this purpose and also to preheat the combustor and reformer feed streams.

Simple heat integration through heat exchangers was done, as streams temperature differences were easily managed. Streams at higher temperatures than 300 ºC were used to heat the combustor and reformer feed flows. Streams with lower temperatures than 300 ºC were employed to preheat in some extent also the feed flows and to produce hot water (60 ºC), which should greatly increase the overall (CHP) system efficiency.

3. Results and discussion

According to the calculations with the semi-empirical model, the hydrogen flow rate to produce 1 kW_e when the fuel cell works at 150 ºC (current density of 0.75 A cm^{-2}) with a hydrogen utilization factor of 1 should be 15.33 NL min^{-1} (41.07 mol h^{-1}). This value is the fuel quantity that the reformer needs to continuously generate that power. From the results of the system simulation, it was observed that the air flow rate injected to the combustor was an important operation variable. This parameter is expressed as the oxygen flow rate over the stoichiometric one for the methanol combustion (λ). In Figure 2 it can be observed that a higher air/methanol (O_2/methanol) ratio makes the methanol consumption higher due to the increase in the energy requirement to preheat the air inlet. Another consequence is that the maximum combustion operation temperature ($T_{combustor}$) decreases when air flow rate is higher. This happens specially when $\lambda>2$, but does not significantly fall (521 ºC) when $\lambda=4$ and it implies, obviously, a lower power efficiency. The need to reduce this combustor maximum temperature using a catalytic combustor gave λ=2 as the more interesting value for later simulations. For this value, the maximum combustor temperature was around 600 ºC.

The methanol steam reforming reactor operational conditions to generate hydrogen must satisfy the CO level limit to operate without degradation (30,000 ppm). In Figure 3a, the influence of reformer temperature ($T_{reformer}$) in the simulation model is presented. It can be noticed that CO level continuously increases when temperature is higher. The CO concentration shows values from 11,600 ppm (290 ºC) to 18,500 ppm (330 ºC). The temperature selected was 300 ºC because it combines high methanol conversion, low heat needs and acceptable CO downstream concentration. This behaviour was also observed by other authors [12, 31]. More influence had the H_2O/methanol ratio λ' in the CO level (Figure 3b). For the studied range ($1< \lambda'<3$), the CO level falls from 58,200 to 6,000 ppm. The same trend was noticed in previous works [32]. However, an excess of water implies higher energy cost because the need to heat water (fresh or recycled from the hot water system). A value of $\lambda'=2$ (13,200 ppm CO) was taken.

Both voltage and power curves obtained in the fuel cell are presented in Figure 4 for several operating temperatures. It can be observed that the higher temperature the fuel cell operates the better fuel cell performance. However, as already said above, it is not convenient to work at temperatures higher than 180 ºC in order to avoid material degradation. For this temperature value, the cell works with a current density of 0.529 A cm^{-2} to get 1 kW_e. Anyway, it was reported in a study over a HTPEMFC operating at temperatures between 150 and 190 ºC that a higher fuel cell temperature results in a higher voltage but also in a shorter cell life [33].

Due to not all of hydrogen reacts at the fuel cell, the unconverted hydrogen can be recycled to the combustor to produce heat. The fraction of converted hydrogen in the fuel cell (hydrogen utilization) depends on the purity of the anode feed hydrogen stream [34]. In Table 1 the influence of this variable is showed. It can be noticed that when this factor decreases, more methanol and air are needed in the system (both in the reformer and the combustor). The consequence is a decrease of power efficiency, as could be expected. However, a higher hot water flow rate is obtained. The increase of fuel cell temperature ($T_{fuel\ cell}$) (Table 2) provides higher power efficiencies with lower methanol, air and water flow rates due to the fuel cell performance is improved. The CHP efficiency scarcely changes. Therefore, a lower hot water flow rate is produced.

4. Conclusions

A model for a new system for micro-combined heat and power (μ-CHP) generation using a combustor, a methanol steam reforming reactor and a high temperature proton exchange membrane fuel cell has been designed. The influence of several operating variables in the power and global CHP efficiency has been studied. Due to the performance of this kind of fuel cells, a CO level up to 30,000 ppm and a temperature not higher than 180 ºC are feasible for a real application. The influence of increasing the O_2/methanol ratio implies that more methanol to combustor is needed in order to provide the suitable heat flow to keep the reactor temperature. It decreases the power efficiency but, as a positive aspect, the maximum combustor temperature also decreases. The reforming reactor temperature has influence in the CO level but this, at the temperature range studied, is lower than the maximum limit for fuel cell degradation in all cases. Higher CO levels than 30,000 ppm were obtained for H_2O/methanol ratios lower than 1.5. The fuel cell temperature also determines its performance. So, for obtaining 1 kW_e at 150 ºC, the current density should be 0.75 A cm^{-2} and at 180 ºC it should be necessary to work at 0.529 A cm^{-2} in a stack with 65 cells with a cell area of 45.16 cm^2. Finally, in order to simulate a more realistic system, the H_2 utilization factor was studied. When this factor decreases, more methanol and air flow rates are necessary to incorporate to the system and the power efficiency also decreases. However, more hot water is obtained, with a roughly similar global CHP efficiency.

The increase of fuel cell temperature makes the power efficiency better with less needs of methanol, air and water to the system. Of course this causes a higher degradation of PEMs and a lower life-time of the fuel cell. A compromise between both aspects should be considered depending on the number and duration of specific high demanding situations and an affordable system life.

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Figure Captions

Figure 1: Diagram of the μ-CHP proposed in this work.

Figure 2: Effect of O_2/methanol ratio (λ) in the combustor: a) methanol and air flow rates, b) combustor maximum temperature and power efficiency.

Figure 3: Effect of operating conditions of the methanol steam reforming reactor in CO level of the outlet stream: a) temperature, b) H_2O/methanol ratio (λ’).

Figure 4: Influence of temperature in fuel cell polarization curves: a) I-V curve, b) I-P curve.

Table1

Table 1: Effect of H_2 utilization in flow rates, efficiencies and hot water production.
Conditions: λ=2; $T_{reformer}$= 300 ºC, λ’=2; $T_{fuel\ cell}$= 150 ºC, Dcc= 0.75 A cm^{-2}.

H_2 utilization factor	1	0.9	0.8	0.7
H_2 flow rate (mol h^{-1})	41.07	45.63	51.34	58.67
Methanol flow rate fed at reforming reactor (mol h^{-1})	14.00	15.55	17.50	20.00
Methanol flow rate fed at combustor (mol h^{-1})	7.42	8.27	9.31	10.63
Air flow rate at combustor (mol h^{-1})	105.0	118.2	133.0	151.9
System power efficiency (%)	26.41	23.74	21.1	18.46
System CHP efficiency (%)	87.48	87.52	87.76	89.03
Hot water production (L h^{-1})	51.23	61.5	74.46	80.9

Table 2: Effect of fuel cell temperature in flow rates, efficiencies and hot water production. Conditions: λ=2; $T_{reformer}$= 300 ºC, λ'=2; H_2 utilization factor = 0.9.

$T_{fuel\ cell}$ (ºC)	150	160	170	180
Dcc (A cm^{-2})	0.75	0.631	0.569	0.528
H_2 flow rate (mol min^{-1})	45.63	38.39	34.62	32.16
Methanol flow rate fed at reforming reactor (mol min^{-1})	15.55	13.09	11.80	10.96
Methanol flow rate fed at combustor (mol min^{-1})	8.27	6.97	6.29	5.88
Air flow rate at combustor (mol min^{-1})	118.20	99.60	89.81	87.93
System power efficiency (%)	23.74	28.19	31.27	33.60
System CHP efficiency (%)	87.52	87.49	87.49	87.36
Hot water production (L h^{-1})	61.50	48.50	41.70	37.31

Figure 1

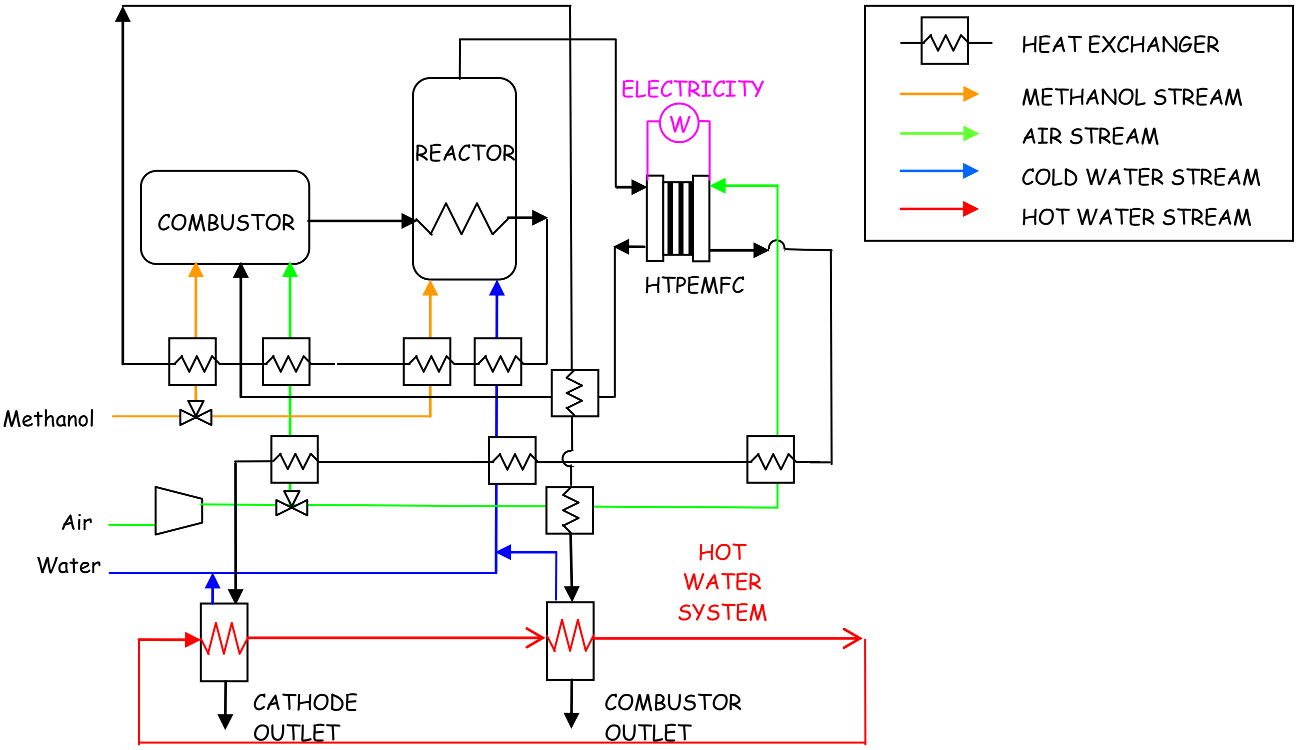

Figure 1

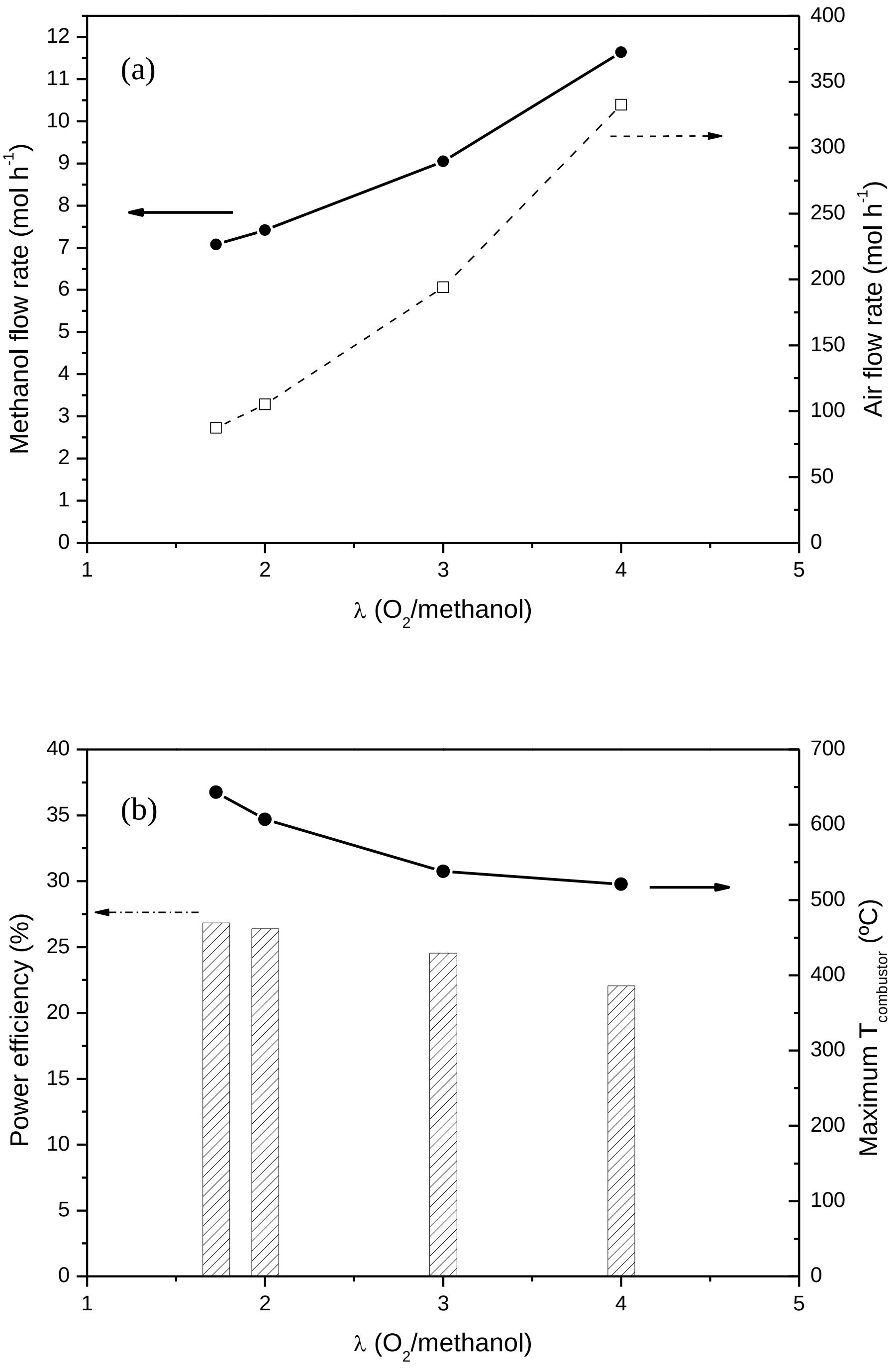
(a)
Methanol flow rate (mol h^{-1})
Air flow rate (mol h^{-1})
λ (O$_2$/methanol)
0
1
2
3
4
5
6
7
8
9
10
11
12
0
50
100
150
200
250
300
350
400
1
2
3
4
5
(b)
Power efficiency (%)
Maximum T$_{combustor}$ (ºC)
λ (O$_2$/methanol)
0
5
10
15
20
25
30
35
40
0
100
200
300
400
500
600
700
1
2
3
4
5

Figure 2

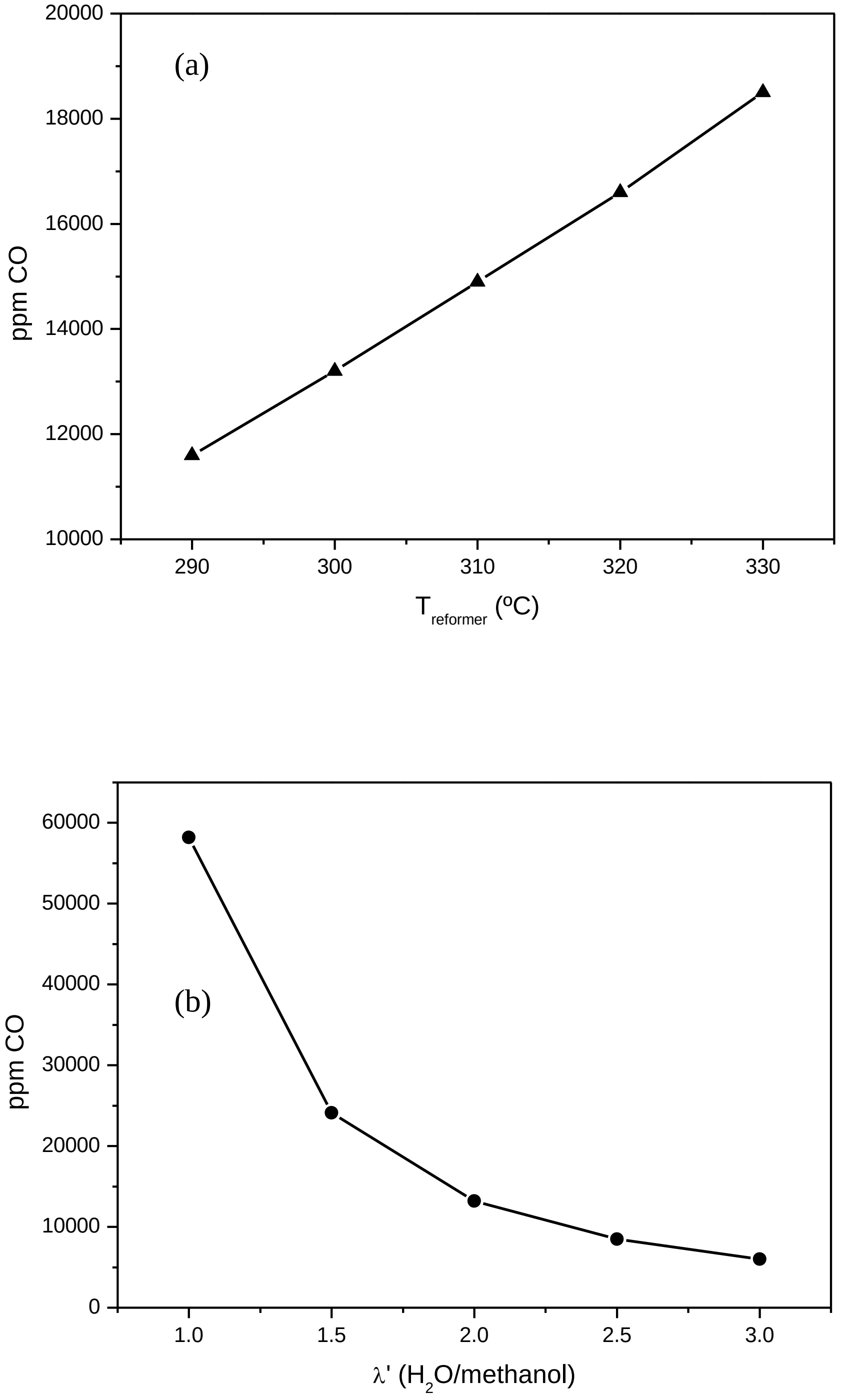
(a)
ppm CO
20000
18000
16000
14000
12000
10000
290
300
310
320
330
T reformer (ºC)
(b)
ppm CO
60000
50000
40000
30000
20000
10000
0
1.0
1.5
2.0
2.5
3.0
λ' (H2O/methanol)

Figure 3

Figure 4

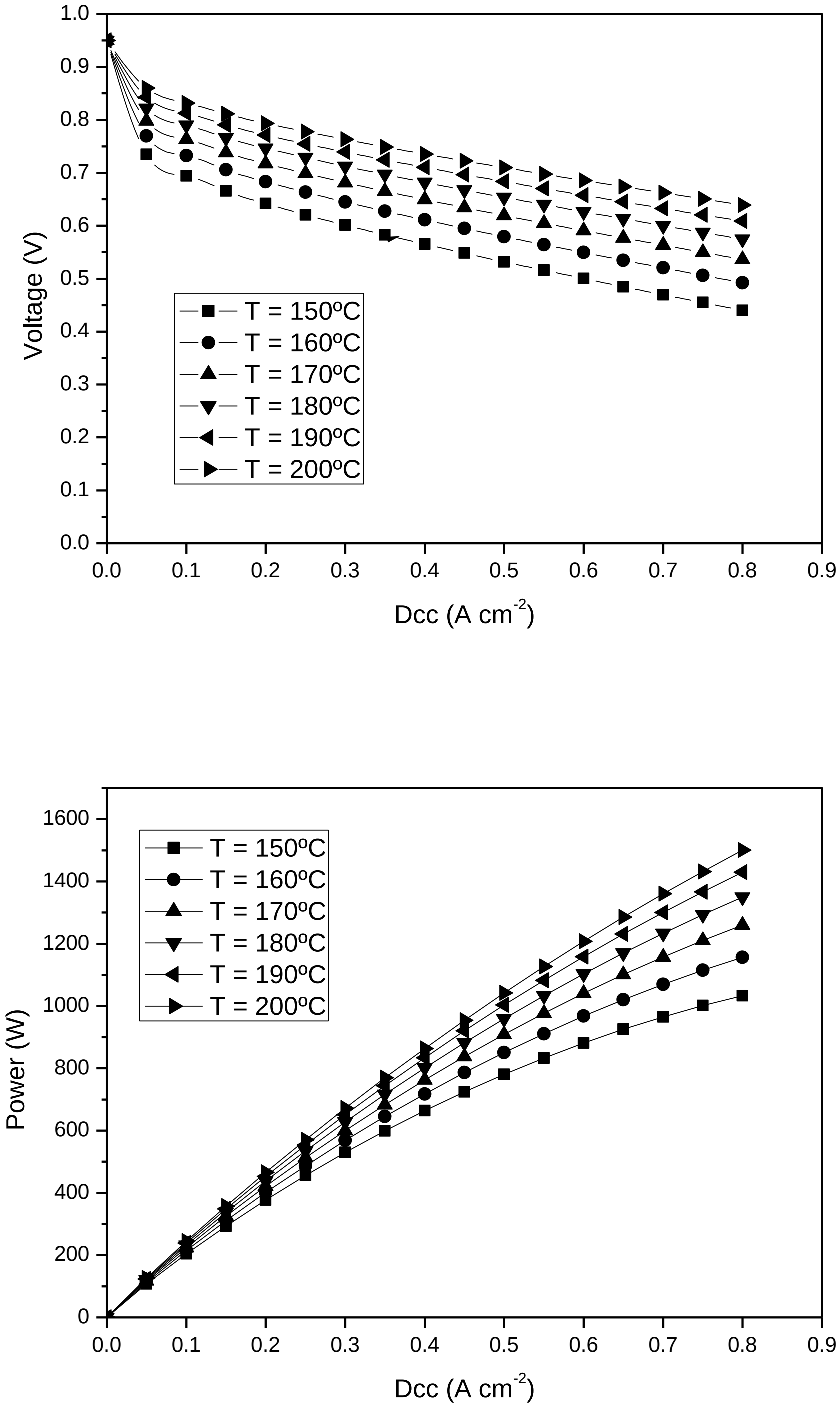

Figure 4