

## MCFC-Electricity Generation from Biogas to Syngas Renewable Process via a Membrane Reactor

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### ABSTRACT

A new biogas based catalytic reforming-processing system for the conversion of gaseous hydrocarbons such as methane (coming from manure type anaerobic digesters) into hydrogen and carbon oxide mixtures is described and analyzed. The exit synthesis gas (syn-gas) is used to power effectively high temperature fuel cells such as MCFC types for combined efficient electricity generation.

Our paper also focuses on the description and design aspects of permreactors (permeable reformers and catalytic carriers) carrying the same type of renewable-biogas reforming reactions. Objectives of this research include turnkey process and systems development for the biogas based power/electricity generation and fuel cell industries. Also, the efficient utilization of biogas and waste type resources (coming from manure based anaerobic digesters) for green-type/renewable power generation with increased processing capacity and efficiency via fuel cells (e.g., MCFCs). Simultaneously, pollution reduction is under additional design consideration in the described catalytic processors-fuel cell systems.

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## 1 Introduction

In our previous IASTED and ACS communications (PGRES/Power and Energy Systems '02, Marina Del Ray, CA; Modeling and Simulation, '03, Palm Springs, CA; ACS-Fuel Chemistry '02, Boston, MA) we described and analyzed new findings and results on catalytic reactors for the steam reforming of methane, natural gas, and biogas reactions, for use in various types of fuel cell systems such as SOFCs (solid oxide fuel cells) [9,13,14].

Our recent communication continues this research by giving emphasis in the so-called “Biogas power” and “Bio-Energy” systems. We analyze the use of biogas mixtures (manure based generated feedstocks) as sources for electricity and heat generation using fuel cells of the MCFC type (molten carbonate fuel cells).

Use of manure based gases rich in methane and carbon dioxide, coming from anaerobic digesters, for the production of intermediate synthesis gas is an attractive route in “green power” and “biogas/manure energy” based systems [9,16,17].

There is a recent emphasis on the development and commercialization of such systems for electricity and heat generation applications. Such installations begin to exist currently mainly in US, Europe, Japan, China and other developing countries. Figure 1 below, shows the itemized distribution, utilization of biogas energy-applications coming from various renewable sources [15].

Such energy systems require the development and use of an effective catalytic reformer utilizing active metals such as Ni, Rh, Ru, Cr, or bimetallic combinations of those. Earth metal enrichment in the catalyst such as with Ca, Mg, La and K promotes the catalyst stability on stream and minimizes deactivation from carbon deposition, especially in the reactor inlet (deactivation propensity is especially high at the inlet due to the lack of hydrogen gas which is generated during the course of the reaction) [3-7,11,12,18-20]. Current plants which convert biogas to heat and electricity utilize a turbine or an engine for this purpose. Biogas coming from anaerobic digesters is converted directly into electricity and heat without the use of a reformer. However, the process has a low efficiency and the waste heat rate is high. Also CO<sub>2</sub> is not utilized within the process.

The reformer utilized in the new process can be a fixed bed catalytic reactor or a permreactor using membrane type materials as reactor walls. Use of a permreactor creates a two outlet reaction system which carries the synthesis gas product at different compositions in the MCFC. The permeate stream is richer in hydrogen and less rich in carbon oxides by the use of hydrogen selective membranes such as microporous inorganics (e.g., alumina, titania based) or metal alloys (e.g., Pd/Ag, Pd/Cu, Pd/Ni, Pd/Ta). One or both of the outlet gas streams can be used as feed in the accompanied fuel cell/MCFC. Use of the permreactor increases the conversion of the reactant biogases in the reactor due to the separation of products. This increased shift in conversion yields the required quantity of synthesis gas product for the fuel cell at a lower operation temperature than the counterpart fixed bed (impermeable) reactor [12,18-20]. Process operation at a lower temperature is beneficial for increasing the reactor and catalyst life times and for reducing the endothermic heating load (Btu/hr) of the endothermic reformer. As an example, we can see below that the membrane reactor can increase the conversion of biogas and the yield to synthesis gas by a significant percentage at the same operating temperature. At 590°C the conversion of biogas into syngas can reach at about 90-95%. The accompanied water gas shift can reach about 75% yield into H<sub>2</sub> and CO<sub>2</sub>. Thus, rich fuel gas/syn gas can be produced in the anode of the MCFC. Moreover, CO<sub>2</sub> coming either directly from biogas or as product of the reforming reaction can be used as co-oxidant (together with O<sub>2</sub>) in the MCFC cathode.

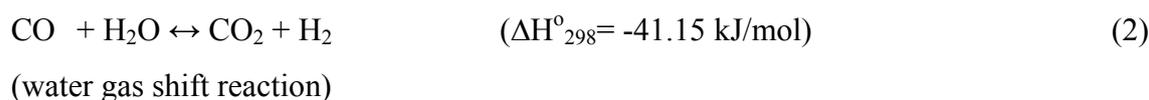
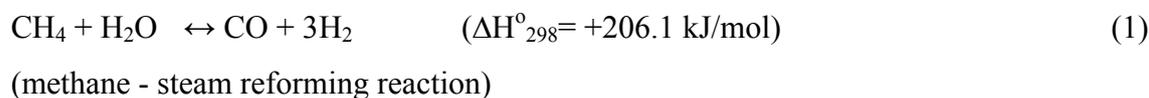
Below, we give design emphasis in both reformer configurations for the generation and delivery of hydrogen rich synthesis gas into the accompanied molten carbonate fuel cell.

## 2 Process-fuel cell analysis and modeling

The process of reforming methane or higher hydrocarbons with steam is a key catalysis route for producing high quality hydrogen or synthesis gas in an economical way [3-14,16,19]. Synthesis gas contains hydrogen mixed with carbon monoxide and possibly carbon dioxide as well. The reforming processes are endothermic and use similar catalyst metals as these described above.

Use of biogas based feedstocks as the reactant gases constitute for a methane (CH<sub>4</sub>) rich feed in the reformer which is converted with steam into a H<sub>2</sub> and CO rich mixture. The exit

hydrogen-rich gas is used as fuel in the anode of the molten carbonate fuel cell. The following reactions take place in the reformer by adding steam in the feedstock as the oxidant, as shown below:



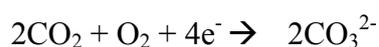
Here we assume that the biogas has been purified before entering into the reformer from the various impurities to avoid the deactivation of the catalyst [16-19].

The interconnected molten carbonate fuel cell (MCFC) produces electricity by the following dual electrochemical reaction:

In the MCFC anode:



In the MCFC cathode:



With the overall reaction to be:



Part of the hot gas exiting from the MCFC according to (3) can be diverted in the shellside of the membrane reactor or the catalytic fixed bed reactor to provide the necessary endothermic heat for running the reformers.

Mathematical modeling of the CH<sub>4</sub>-H<sub>2</sub>O reformer for a steady state fixed-bed catalytic reactor includes the species reaction terms in the mass balance equations and is as follows:

$$dX^A/dz = (\pi d_T^2 / 4n_{A0}^T) \rho_B R_A \quad (4)$$

Species A can be any of the reactants and products of the reactions (1) and (2) above.

$$\begin{aligned} \text{With: } R_{\text{CH}_4} &= -R_1 \\ R_{\text{CO}_2} &= R_2, \quad R_{\text{CO}} = R_1 - R_2, \\ R_{\text{H}_2} &= 3R_1 + R_2, \quad R_{\text{H}_2\text{O}} = -R_1 - R_2, \end{aligned}$$

Where  $R_1$  and  $R_2$  are the heterogeneous reaction rates of the reactions (1) and (2) given above.

The thermal balance in a non-isothermal reformer with T and S sides, is given as follows:

$$dT_T/dz = (\pi d_T^2/4) (1/m'c_p) \{ \rho_B [(-\Delta H_r^1) R_1 + (-\Delta H_r^2) R_2] - 4 (U/d_T) (T_T - T_S) \} \quad (5)$$

The reformer pressure balance which describes the pressure drop along the fixed bed of catalyst is given as follows:

$$-dP_T/dz = (2 f \rho_g u_s^2) / (g_c d_p) \quad (6)$$

The above equations are complemented by initial conditions as shown below:

$$\text{at } z=0 \text{ (reactor inlet), } X^A=0, \quad T_T=T_0, \quad P_T = P_{T_0}$$

A detailed analysis of the model, its parameters and their variation is given in earlier communications [2],[15],[16].

The system of equations (4), (5) and (6), is integrated numerically as an initial value problem to provide the reactant conversions, product yields, reactor temperature and pressure along the axial length and to obtain the axial profiles of these variables and their values at the reactor exit.

By employing an inorganic permreactor as the main catalytic processing unit to convert biogas feedstocks into fuel cell gas, the above design equations are modified accordingly to

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include the permeation effects via the membrane of the different components. The following term has to be added at the right hand side of equation (4) to account for the permeation effects within the mass balance equations:

$$- (2\pi/n_{A0}^T) P_{A,e} [ (p_A^T - p_A^S) / \ln (r_1/r_2) ] \quad (7)$$

wherein  $P_{A,e}$  (gmol/s.cm.atm) is the effective permeability coefficient of species A via the catalytic or non-catalytic (blank) membrane. In our earlier experimental reaction studies we utilized mesoporous aluminum oxide membranes having a thin permselective layer (3-5 $\mu$ m thickness, 50% porosity) with 40-50 $\text{\AA}$  pore diameter [8,12,18,19]. In the case of a permreactor the corresponding mass, temperature, and pressure variation equations are written as well for the gas which permeates via the membrane wall material and flows in the permeate side (S) of the membrane reactor. We assume that there are no reactions occurring in the permeate membrane side. The detailed model for the permreactor has been described as well in earlier communications [11,12,18,19].

By using the above equations (4),(5),(6), and (7) within the modeling procedure a detailed reactor analysis is obtained for the two different reformer configurations. Solution of the equations is obtained numerically by using an initial value integration technique for ordinary differential equations with variable stepsize to ensure higher accuracy (implicit Adams-Moulton method) [18,19].

In our previous communications we have described and analyzed the reaction, separation (i.e., permeation), and process (conversion, yield, selectivity) characteristics of permreactors (membrane based catalytic reactors) and related processes for methane-steam reforming, water gas shift, and methane-carbon dioxide reforming reactions including catalysis and membrane materials characteristics. The main types of reactors described were membrane reformers which were utilized as single permreactor [19]; permreactor-separator or reactor-separator sequence and permreactor-permreactor sequence, [8,11,18-20].

These effective and versatile catalytic systems were applied for pure hydrogen ( $H_2$ ),  $H_2$  and  $CO_2$ , or  $H_2$  and  $CO$  (syn-gas) generation to be used as fuel gas for power generation or as

synthesis gas for production of specialty chemicals (such as methanol and higher hydrocarbons) [19]. More than one type of membrane reformers were examined based on the location of the fixed catalytic bed and the inert or catalytic nature of the inorganic (alumina based) membrane tube. Computational modeling of the described perm-reformers was performed which shows performance measures (reactant conversion, product yield, product selectivity) versus variation of intrinsic model parameters (reactor space time, reaction temperature and pressure, feed composition, sweep gas to feed gas ratio). The models also analyze and show performance measures under new operating conditions which are of interest to new energy and chemical process applications [8,12,18,19].

The interconnected or integrated molten carbonate fuel cell is fed directly by the fuel gas generated by the described reformers. The focus of our studies includes solutions in a number of problems associated with the installation, operation, and mass, energy conservation of the entire MCFC and membrane reformer-processing unit.

One possible effective process and flow configuration is the separation of CO<sub>2</sub> before the reformer. CO<sub>2</sub> can be separated from methane using a number of methods such as polymer membrane separation technology, or cryogenic separation, or pressure swing adsorption [18,19]. With this way pure CH<sub>4</sub> (methane) flows into the reformer and reacts according to reactions (1) and (2). On the other hand, the separated CO<sub>2</sub> stream is directed in the cathode of the MCFC to act as the additional oxidant (i.e., equation (3)). It is possible also that some CO<sub>2</sub> flows into the reformer as a mixture with methane. CO<sub>2</sub> will react within the reformer and catalyst with the reverse of the water gas shift reaction (reverse of reaction (2)) and produce some CO. Also, it may react with methane via the direct CO<sub>2</sub>-methane reforming reaction (dry reforming) to produce more CO and hydrogen according to the following reaction stoichiometry ( CO<sub>2</sub> + CH<sub>4</sub> = 2CO + 2H<sub>2</sub> ) [8,12,19].

Using the Nernst equation for the generated electrical potential of a MCFC we have [19, 20]:

$$E = E^{\circ} + RT \ln [P_{H_2} P_{O_2}^{1/2} / P_{H_2O}] / (2F) + RT \ln [P_{CO_2c} / P_{CO_2a}] / (2F)$$

Where the subscripts **a** and **c** indicate the anode and cathode gas sides respectively.

Also,  $E$  is the EMF (in volts)

$E^\circ$  is the EMF at standard pressure (in volts);

$P_j$  are the partial pressures of the participated gases (in kPa).

One more possible process flow configuration will be the addition of both oxygen ( $O_2$ ) and carbon dioxide ( $CO_2$ ) into the reformer feed inlet. With this way a four component reactive system is maintained ( $CH_4-CO_2-H_2O-O_2$ ) which is mostly exothermic and therefore no heat input in the reactor is necessary. This reaction system is capable to produce syngas as well ( $H_2$  and  $CO$ ) to feed the MCFC. However, the issues of increased carbon formation/deposition on the catalyst and the formation of unwanted byproducts have to be addressed.

The economic feasibility of the overall fuel cell installation is correlated with higher efficiencies (e.g., 42%-65% for advanced units) and high current density output ( $A/cm^2$ ), increased system reliability for continuous dispersed power generation, and reduced plant installation, operation and maintenance cost. These targets combined with virtual elimination of pollution by use of fuel cells in stationary (e.g., central and remote power stations) and mobile/transportation (e.g., automobile) sources make this technology highly applicable and attractive. Clean fuel cell power minimizes  $NO_x$ ,  $CO$ , and hydrocarbon species in the emissions [19, 20].

## 3 Results and discussion

### 3.1 Study Details

Use of biogases coming from manure-type anaerobic digesters, within the reformer, constitutes an innovative approach in previous attempts for direct use of those feedstocks for power generation [2]. There are significant renewable resources of biogas feedstocks today generated from the large herds of farm animals grown in local and remote farms.

Gas coming out from the proper treatment of manure of farm animals in anaerobic digesters is rich in methane and constitutes the proper mixture for direct conversion into the described reformer/MCFC system. Catalytic conversion of these gases in the reformer yields a hydrogen rich mixture for properly powering the interconnected MCFC. As the flowrate of the biogases increase (for larger sites and treatment systems) a larger capacity reformer and

fuel cell are required to handle the conversion; consecutively, the final MCFC power output (kW/cm<sup>2</sup>) increases as well.

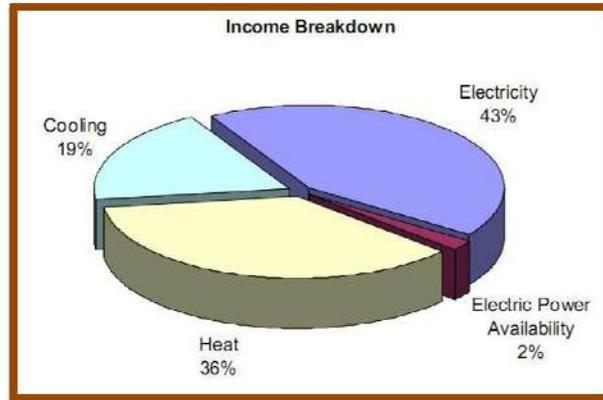


Figure 1: Percentage of income from the direct utilization of biogas coming from agricultural sources and farm animal wastes [15].

Figure 1 shows the percentage of income from the direct utilization of biogases coming from agricultural and farm animal waste sources. The figure shows the different energy utilization of used biogas in terms of percentage [15].

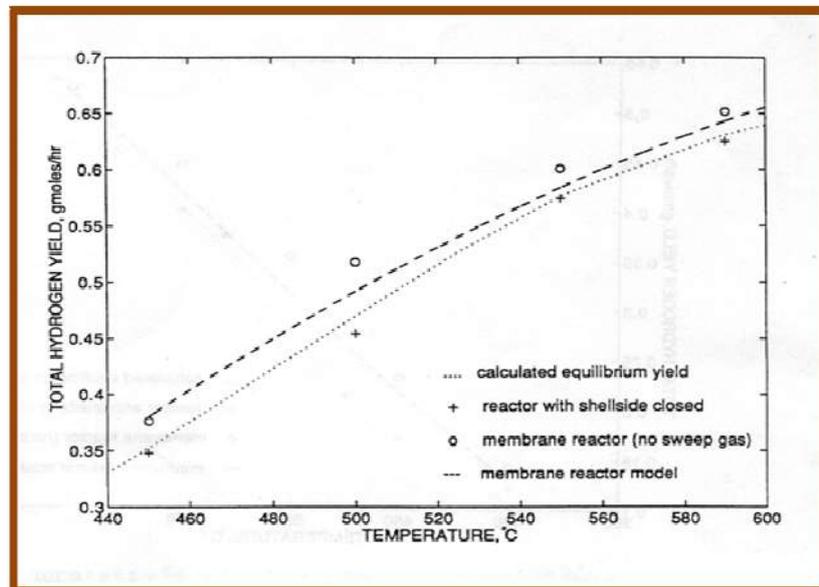


Figure 2: Experimental and modeling results of two biogas-steam reformers for syngas production and MCFC continuous operation.

Total hydrogen yield data; ( $P_{T_0}=1.17$  atm, Space time = 54.0 g<sub>cat</sub>.hr/gmole<sub>CH<sub>4</sub></sub>)

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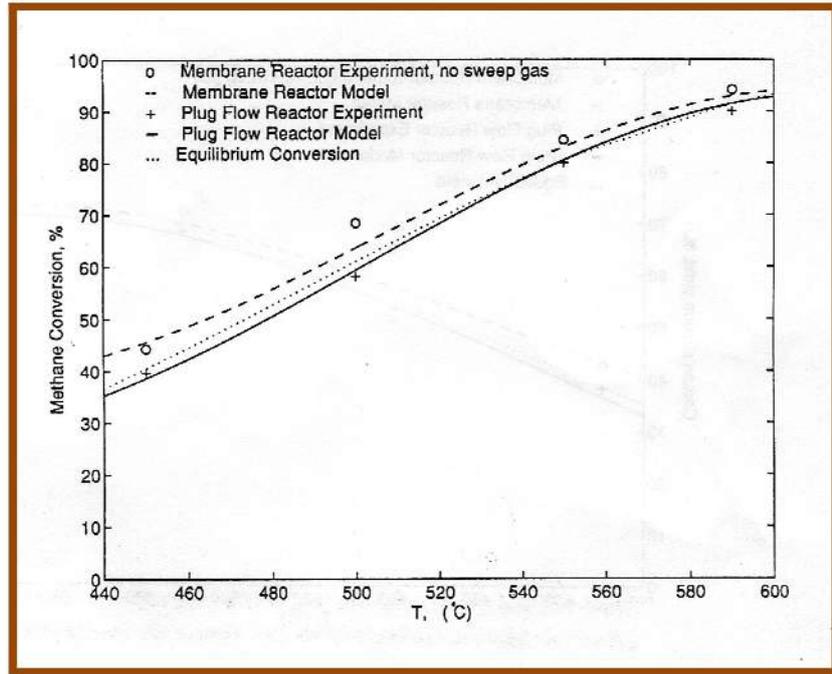


Figure 3: Experimental and modeling results of two biogas-steam reformers for syngas production and MCFC continuous operation.

Total methane conversion data; ( $P_{T_0}=1.17$  atm, Space time =  $54.0 \text{ g}_{\text{cat}}\cdot\text{hr}/\text{gmole}_{\text{CH}_4}$ )

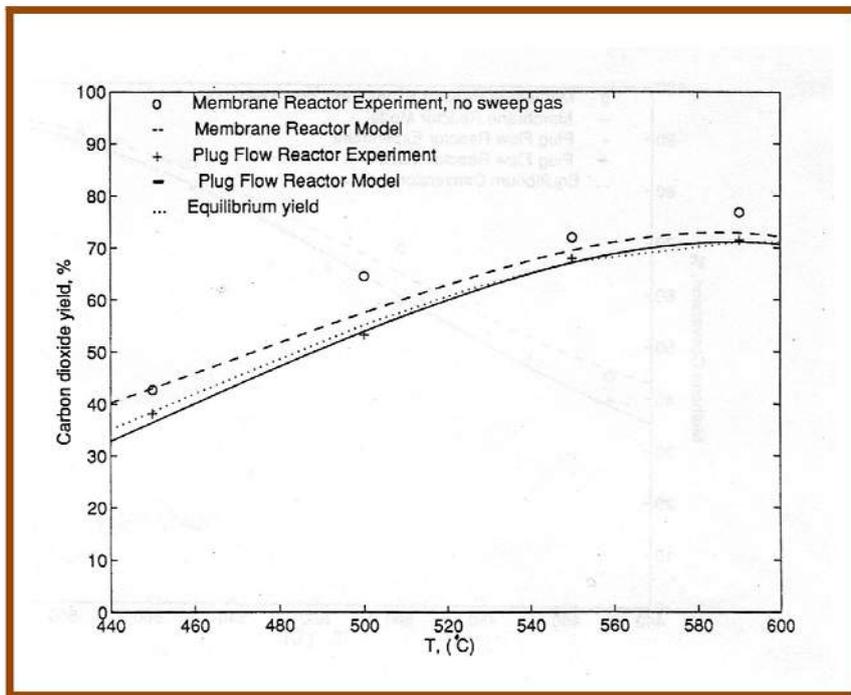


Figure 4: Experimental and modeling results of two biogas-steam reformers for syngas production and MCFC continuous operation.

Total carbon dioxide yield data; ( $P_{T_0}=1.17$  atm, Space time =  $54.0 \text{ g}_{\text{cat}}\cdot\text{hr}/\text{gmole}_{\text{CH}_4}$ )

Moreover, we report below the performance of two types of reformers for the specific conversion reactions (1) and (2). In our experiments and models we used methane as the simulated biogas feed. Thus, Figure 2 shows the total hydrogen yield produced from these conversion reactions within the reformer and specifically at the reformer exit as function of the reaction temperature. We report results from a membrane type reformer and from a conventional (non-membrane) plug flow type reformer. The membrane reformer exceeds in total hydrogen yield the non-membrane reformer and this is also shown by the accompanied modeling results which simulate well the experimental membrane reformer data. Moreover, the plug flow type reformer produces results that are very close to the calculated equilibrium hydrogen yields which are calculated at the tubeside (T) reaction conditions. Hydrogen and CO produced under these conditions are directed in the MCFC anode to drive the electrochemical reactions (3) shown above. The feed composition in the tubeside of the membrane reformer was maintained at  $\text{CH}_4 : \text{H}_2\text{O} : \text{Ar} : \text{H}_2 = 1 : 7 : 1 : 0.75$ . Ar gas was added in the feed as a diluent to examine the effect of diluting the methane/biogas feed. The catalyst used in the reformers was a 15% NiO catalyst on calcium aluminate and magnesium aluminate supports. 8.02g of catalyst was loaded in the reformers in the form of particles with 0.92mm average diameter. The space time of the reactors tubeside was maintained at  $54.0 \text{ gr}_{\text{cat}} \cdot \text{hr} / \text{gmole}_{\text{CH}_4}$ . The reaction range examined in the two reactors was varied from 450-590-C. The pressure in the tubeside of the reformers where the catalyst lies was maintained at about 2-3 psig (1.17atm) during the course of these experiments.

Further, we report below methane conversion and  $\text{CO}_2$  yield data at various reaction temperatures (450-590-C). This data is indicative of the course of the two catalytic reactions (1) and (2) within the reformers. The reaction conditions remained the same as with the above plot (hydrogen yield data). Thus, Figure 3 reports methane (simulated biogas) conversion versus reaction temperature data for the temperature range reported above.

Methane conversion data is reported for the two types of reformers. The data is accompanied by simulation fittings by the numerical model developed and described above.

Figure 3 shows that the membrane reformer conversions are higher than the conventional (plug flow) reformer values. The membrane reformer methane conversions exceed the equilibrium conversions calculated at the tubeside reactions conditions. The plug flow reformer conversions are very close to the equilibrium calculated conversions. Moreover, both types of conversions are simulated well by their respective modeling lines. The models use data from both reformers and from the reaction conditions which occur within the catalyst. The kinetic data and the permeation/permeability data used within the models were obtained from separate/independent experiments which took place earlier [18,19]. The membrane reformer conversions are higher because of the equilibrium shift of reaction (1) due to the effect of the membrane in the reformer. The membrane assists in the continuous hydrogen permeation and separation which leads into the above beneficial increases of the methane conversion and the CO<sub>2</sub> yield as also shown in the Figure 4.

It is characteristic that the methane (simulated biogas) conversion exceeds 90% at the 590·C especially for the membrane reformer case.

The beneficial increase in CO<sub>2</sub> yield with the use of the membrane reformer is shown in Figure 4 below, together with the other data. The CO<sub>2</sub> yield is indicative of the extent of the water gas shift reaction (reaction (2)). Reaction (2), produces more suitable fuel (H<sub>2</sub>) and oxidant (CO<sub>2</sub>) for the interconnected MCFC. The CO<sub>2</sub> yield also corresponds to the CO conversion according to reaction (2). Moreover, there is a good fit by the modeling results (simulation lines) to the experimental CO<sub>2</sub> yield data. The reaction conditions are the same with those described above. The above plots (Figs. 2, 3, 4) are shown the type of syngas (in terms of composition) which is entering into the MCFC system for electricity generation according to reactions (3), [12-14,19,20].

The described data shows that fuel gas rich in H<sub>2</sub> and CO compounds can be produced from the described reformers and especially from the membrane reformer for the continuous operation and electricity generation of the MCFC. A related representative plot is shown in Figure 5. It shows the generated power by the MCFC for various feed ratios and reforming conditions (i.e., reaction temperature). The plot assumes a 60% fuel cell efficiency at equilibrium fuel gas composition according to reactions (1) and (2). Higher power outputs (kW) can be achieved usually between 600-800·C.

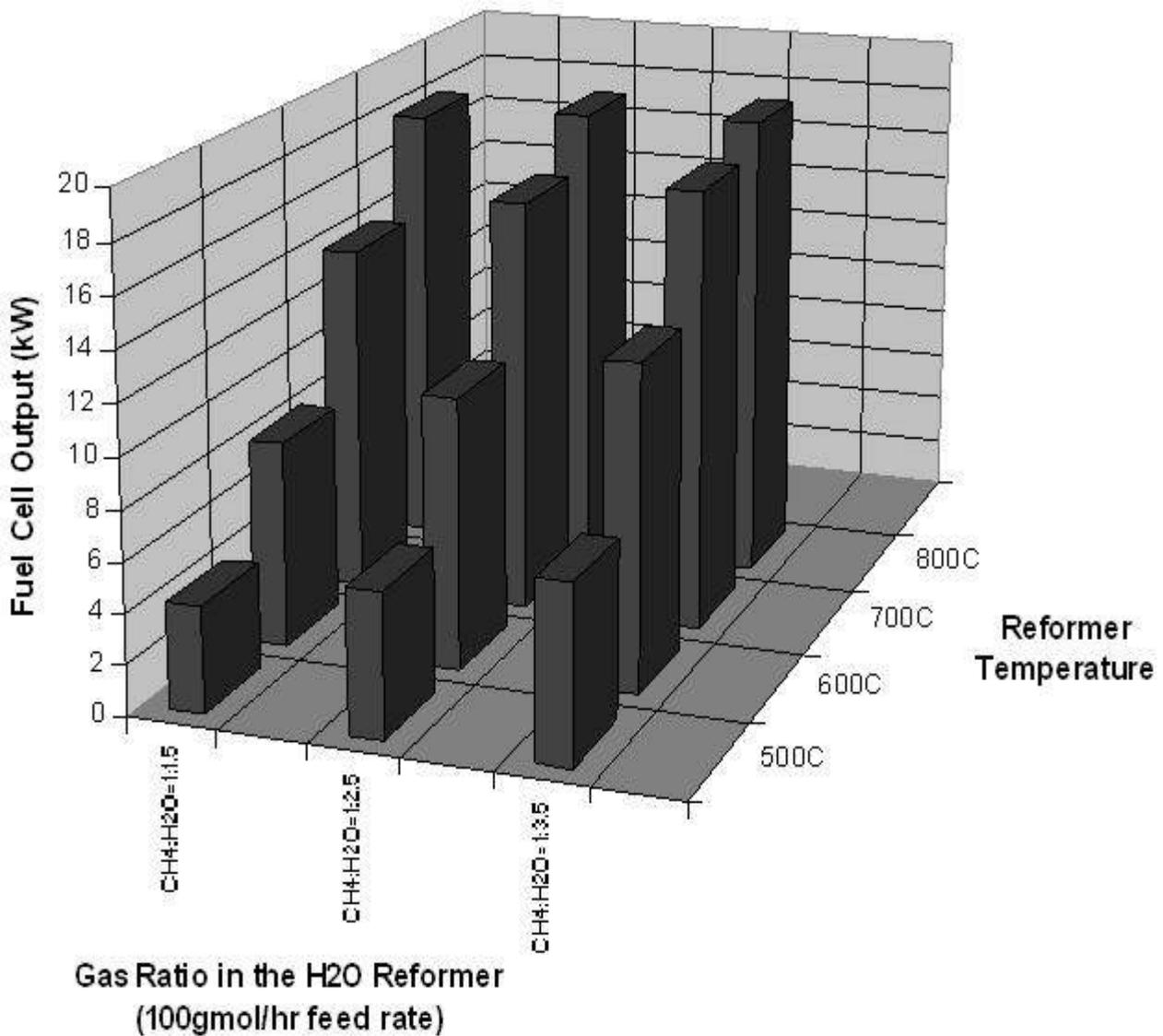


Figure 5: MCFC power (kW) versus catalytic reforming conditions; 60% MCFC efficiency at equilibrium fuel gas composition.

Moreover, Table 1 shows a summary of specifications from a biogas processing plant for energy cogeneration (biogas is coming from farm animal wastes [17]). The table provides details on the energetic distribution outcome of the entire plant. This table is given for comparison purposes, in order to be evaluated the potential of the newly described biogas to MCFC unit. We should point out that the electricity generated by the MCFC (assuming a 60% efficiency) can cover the needs of the farm and any excess electricity can be sold in the nearby

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electrical network. The useful heat from the MCFC can cover the heating needs of the farm (e.g., via a boiler) and those of the endothermic reformer. An optimum flow chart for the specific process therefore can be generated which can utilize properly the electricity and heat produced by the MCFC.

Table 1: Specifications of a representative biogas steam reforming-MCFC plant, for electricity and heat cogeneration.

| Specifications of a MCFC plant   |   |
|--|---|
| Biogas production volume:<br>(From the Manure Anaerobic Digester)                      | 3,600 m <sup>3</sup> /day                 |
| Total number of farm animals :<br>(swines)   | 2210                                      |
| Methane production volume:   | 2,520 m <sup>3</sup> CH <sub>4</sub> /day |
| Total energy generation:   | 35,705 kWh/day                            |
| Electricity generation/MCFC<br>(60% min):  | 21,423 kWh/day                            |
| Heat generation<br>(30% min):  | 10,712 kWh/day                            |
| Waste heat<br>(about 10%):   | 3,570 kWh/day                             |
| Annual Electricity<br>Generation:  | 7,700 MWh/year                            |
| Income about<br>(Sale price to DEH, Greek Electricity<br>Authority: 73 Euro/MWh,)      | 562,000 Euro/year                         |
| Annual Heat generation:  | 3,856 MWh/year                            |
| Equivalent Oil capacity<br>(Annual)<br>Oil sale price: about 1,100 Euro/m <sup>3</sup> | 846 m <sup>3</sup> /year                  |

## 4 Conclusion

High temperature MCFCs (molten carbonate fuel cells) can be coupled with reforming operations of biogases coming from anaerobic digesters of manure (farm animal wastes). The MCFCs can operate in series or integrated with a catalytic reformer or a membrane reformer which convert biogas into fuel syngas at various operating conditions. These feedstock biogases are rich in CH<sub>4</sub> and are converted into a H<sub>2</sub> and CO mixture suitable for the continuous operation of the MCFC unit. At the same time the unused CO<sub>2</sub> constituent of the biogas or the

CO<sub>2</sub> which is coming as product of the water gas shift reaction can be used as direct oxidants in the MCFC. Our reformers have been also simulated by computational models which account for the reaction and hydrogen separation in the permeable reformers. Several flexible operating conditions in the permeable-reformers have been tested via these models and useful operation results have been derived.

Useful distributed power generation within a wider power grid can be accomplished with this MCFC operation, which can cover the local needs of municipal and remote areas. MCFC power relates to the reformer conversion and the efficient utilization of the syngas by the fuel cell. The waste heat from the conversion of syngas to electricity can be reduced through the fuel cell operation and specifically through the increase of the efficiency of the MCFC. Fuel cell/MCFC continuous operation and power generation from manure biogas offer as well pollution minimization, higher power density and increased efficiency in comparison with conventional power systems. Clean MCFC power also minimizes NO<sub>x</sub>, CO, and hydrocarbon species from the emissions of this stationary biogas fed power unit.

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