

Computer Simulation of Stationary Energy Production from Biomass by Molten Carbonate Fuel Cells

Maurizio Fermeglia and Gennaro Longo

ICS-UNIDO, AREA Science Park, Padriciano 99, 34012 Trieste, Italy
DICAMP, University of Trieste, Piazzale Europa 1, 34127 Trieste, Italy

maurizio.fermeglia@dicamp.units.it gennaro.longo@ics.trieste.it

ABSTRACT

This paper presents an integrated approach to the steady state and dynamic simulation of fuel cells and power generation processes. Attention is devoted to Molten Carbonate Fuel Cells (MCFC) due to the relative low cost, simple construction and flexibility of use of the fuel. Two models are used in this work, both recently presented in the literature: (i) a steady state model simulating a global (MCFC) power plant and (ii) a dynamic model for the same cell (MCFC). The models are based on real plant data and the simulations are selected according to real operating conditions. The computer simulation is finally applied to a feasibility study of a MCFC coupled with a biomass gasification process.

This paper reports the results of computer simulation for a realistic configuration of a power system generated by MCFC, focusing on macroscopic quantities of interest such as stack efficiency, global process electrical efficiency, co-generative efficiency, integrated with a consistent dynamic model of the cell able to consider microscopic and distributed variables behaviour such as temperature and concentration in the cells.

The software developed allows the *in silico* study of the effect of variations in the process conditions as well as modification of the input fuel, thus providing a useful tool for supporting technical decisions and feasibility studies on the use of fuel cells in developing countries.

Keywords: molten carbonate fuel cells, computer simulation, biomass gasification.

1. INTRODUCTION

Deregulation of the electric power industry and more stringent emission controls are stimulating investments in fuel-cell systems, which are virtually free from noxious emissions of nitrogen oxides normally associated with burning fossil fuels. They can convert chemical energy directly into electricity with greater efficiency than most other devices, thus conserving fuel resources and reducing CO₂ emissions. Although hydrogen is considered the ideal fuel for many energy-conversion systems, its widespread use is dependent on technological breakthroughs in its cost and storage. So, it is best to assume that in the immediate future, except for a few niche markets, fuel cells will have to use hydrocarbons (such as methane) or alcohols (such as methanol) as fuel.

The application of fuel cells is particularly attractive in the renewable energy arena: biomass could be used as fuel for hydrogen production, thus addressing the problems of generation of carbon dioxide at the same time. This topic is also of interest to developing countries for off-grid power generation with low impact on the environment. It is therefore important to be able to carry out feasibility studies and decide if a given process is suitable for a task.

Among the different fuel cells developed so far, molten carbonate fuel cells (MCFC), also referred to as 'second generation' cells (Kraaij et al., 1998), have been thoroughly investigated over the last decade for several advantages with respect to other technologies. MCFC's simulation may be helpful in their further development before commercialization (Bishoff et al., 2002, Joon, 1996).

In fuel cells - MCFC in particular - high global efficiency, with respect to other electrical energy production systems, can be reached in merit to many contributory causes (Hart et al., 1967, Kordesh et al., 1996):

- working temperatures are sufficiently high to enhance chemical and physical processes, but not so high as to lower the free energy;
- construction materials for the cells are not very expensive and no costly catalysts are needed;
- discharged heat can be used either inside the cell or transferred to down stream systems;
- carbon monoxide tolerance, a pollutant in other technologies, can be burned in MCFC;
- low emissions are reached since many pollutants are not present and exhaust gases are processed and partly recycled;
- power scalability can be achieved by simple addition of compact modules, transportable by rail or road;
- low pressures are involved and voltage can be improved by increased partial pressures of reactants, keeping in mind that mechanical resistance of materials requires less than 3.5 bars operating pressure.

A molten carbonate fuel cell plant integrates a stack with a pre-treatment of fuel conditioning and post-treatments of residual energy recover. The former is essentially desulphurization and reforming, the latter makes use of steam and gas micro-turbines. Thermal control is obtained by varying both the exhaust gas recycle and the inlet temperature. The stack is made up by a number of piled up cells, and soft joints, to comply with mechanical tolerances. Figure 1 shows the general process for energy production from natural gas.

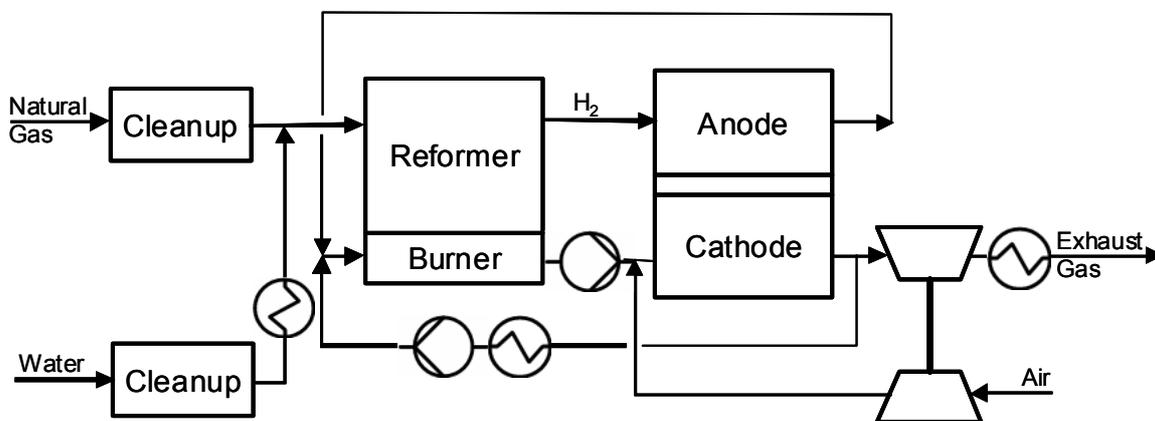


Figure 1:

Many are the intervening parameters for defining the optimum process condition and its stability with respect to possible external perturbations. It is therefore important to simulate the process for different steady state conditions to determine the best process parameters and study the effect of process modifications with respect to efficiency and energy production. It is also very motivating to study the dynamic behaviour of the process for determining the time dependency of key process parameters for system changes.

Mathematical modelling is used as an investigation technique to improve knowledge of limiting factors (Kang et al., 2001). In the past few years many authors have developed steady state models

based on macro balances (Miyake et al., 1995) applied to both single cell and stack, or differential equations (Yoshida et al., 1998, Bosio et al., 1999, Koh et al., 2000, Arato et al., 2001, Kim et al., 2002, Park et al., 2002, Mangold et al., 2003) applied to both mean and distributed variables. An interesting review of steady state simulation of fuel cells is reported in (Koh et al., 2001). Many problems were solved in assumption of hypothesis, in formulation of equations and in approximation of accessory devices (De Simon et al., 2003), but many others are still to be solved, such as integration of the cell model in the plant and the control strategies.

Models for electrodes were developed assuming various structures for micro pores and internal meniscus (Fontes et al., 1995 and 1997, Fehribach et al., 2000) but such models are not always easily integrated into the macrostructure to be simulated.

It is interesting to analyse the models available in the literature according to the number of dimensions considered. There are only a few papers containing a multidimensional approach. Three dimensional approaches (Yoshida et al., 1998, He et al., 1995) have been applied to the stack rather than the cell, thus giving far fewer details in the distribution of the variables in the cell. Two dimensional approaches (Bosio et al., 1999, Kim et al., 2002, Park et al., 2002) are available but consider only steady state conditions. Other models are available the mono-dimensional approach (Lukas et al., 2001, Heidebrecht et al., 2003).

MCFC dynamic simulations are carried out assuming separated modules (He et al., 1995, Lukas et al., 2001, Heidebrecht et al., 2003, Shinoki et al., 1995, He, 1997 and 1998) exchanging information among them and obtaining the solution by means of a sequential modular approach: each module is solved and the results are sent to the next module until convergence is achieved. Specific modules are used to calculate electrical, physical and chemical properties. Recently, another interesting approach (Heidebrecht et al., 2003) considered the dynamic simulation of a MCFC with internal reforming and one dimension counter flow with isobaric conditions in the duct. The model yielded interesting results for the simplified case of negligible heat conduction and lumped solid phases. The model was written in terms of dimensionless variables and tested for step input of some variables.

2. THEORY AND MODEL DEVELOPMENT

In comparison to other devices, fuel cells are complicated by the electrochemistry of the system. A fully distributed bi-dimensional open loop model was implemented in a software application, to understand physical and chemical phenomena ruling MCFC. A steady state model of MCFC was developed in a previous work (De Simon et al., 2003) and has been extended recently to dynamic simulation of the single cell (Fermeglia et al., 2005). In both models, the fundamental chemical-physical equations are organized to give an 'easy-to-solve' system which deeply integrates fluid dynamical, pressure drop, and reaction equations, thus avoiding the 'off line' approach in the solution of the set of equations, and solving the system in an equation oriented approach. Equations are grouped together logically, but not analytically, in modules (duplicated variables are eliminated during the solution) thus allowing simple and fast modifications of the system for screening many different configurations. Chemical reactions are coupled with homogeneous and heterogeneous heat and mass transfer thus generating a system of non-linear differential equations. The models account for heat transfer by convection and conduction and each solid phase is considered separately: each with their own physical properties and characteristics. Furthermore the anode and cathode are modelled independently.

The input variables are velocities, temperatures and densities and the water gas shift reaction is considered at equilibrium and constrained by the continuity equation. The model is written in terms of dimensional variables and tested for a wide range of input variations: steps, linear ramps and sinusoidal ramps.

2.1 The energy production process

The molten carbonate fuel cell plant described in the introduction, and reported in Figure 1, is the reference flow sheet for the development of the steady state simulation of the process. Figure 2 shows the essential geometry of the single cell as modelled in this paper. The main components of the MCFC are: (i) two metal current collectors, also active in distributing the gas streams across the electrodes surfaces; (ii) a NiO-based porous cathode, where the reduction reaction occurs, resulting in the production of CO_3^{2-} ion, that must travel across (iii) an electrolyte tile (made up of molten K, Na and Li carbonates in a solid porous and inert matrix), and reach (iv) a Ni-based porous anode, where oxidation occurs.

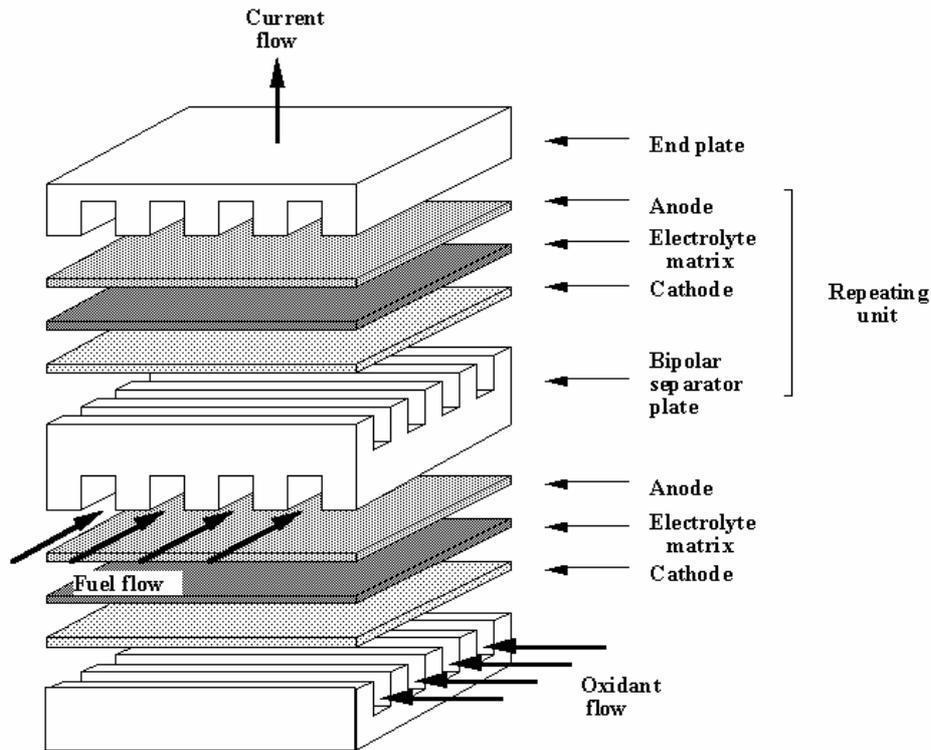


Figure 2:

The cathode reaction is:



The anode reaction is:



The total reaction is essentially the hydrogen combustion reaction, combined with a CO_2 transfer from cathode to anode:



The stack is made up of a number of piled up cells separated by cooling plates, used to maintain a constant temperature, and soft joints to comply with mechanical tolerances.

The fuel, after a primary treatment, is mixed with steam, pre-heated in a regenerative heat exchanger, and fed to the reformer, where CO and H_2 are produced. Exit gas passes again through

the regenerative exchanger, and is set ready to be processed at the anodic side of the fuel cells stack. After electrochemical and gas shift conversion, residual concentration of fuel is still present, principally because CH_4 not converted in the reformer is inert in the stack. Moreover, to avoid a diffusion-controlled electrochemical process, not all H_2 and CO is consumed. Consequently, fuel containing effluent is mixed with part of the cathodic exhaust, and fed to a catalytic burner where the heat necessary for endothermic reactions taking place in the reforming process is produced and directly exchanged. CO_2 rich and hot gas is mixed with fresh air coming from the compressor, and fed to the cathode where O_2 and CO_2 are consumed by reaction (1). Part of the cathode exhaust gas is sent to a turbine for recovering residual energy, which is used to drive an air compressor and to produce further electrical energy. The hot effluent eventually releases the heat necessary to produce reforming steam, and for eventual cogeneration.

2.2 The single cell dynamic simulation model

The dynamic simulation is characterized by the simultaneous integration of all the process equations. The simulation of a molten carbonate fuel cell (MCFC) system in dynamic condition has been implemented by some of us (Fermeglia et al., 2005) with a bi-dimensional cross flow fuel cell numerical model: in this paper we refer to that model. The model developments and the peculiarities of the dynamic simulation of fuel cell systems is thoroughly discussed and presented in (Fermeglia et al., 2005). All the equations describing the dynamic behaviour of a MCFC have been derived and presented in detail.

The dynamic simulation is characterized by the simultaneous integration of all the process equations and is implemented using a commercial equation solver (Aspen Custom Modeler™), thus taking advantage of the available data banks and physical property calculation methods. All the equations are solved in an equation oriented approach, thus coupling fluid dynamic, pressure drop, reaction, and thermo physical property equations to obtain the behaviour of the key parameters in the time domain.

Steady state simulations are run with the dynamic model to obtain the base case, and sensitivity analysis is performed. The model dynamic behaviour is compared with results obtained by a different model and software operating in steady state conditions, showing good consistency. The dynamic model developed is based on real plant data and the simulation conditions are selected according to real operating conditions.

The dynamic simulation reported with the description of the model (Fermeglia et al., 2005) showed the quantitative response of the system to several disturbances and proved to be a valid tool for determining the temperature, the current and the voltage profiles as a function of time within the bi-dimensional cross flow fuel cells.

2.3 The steady state fuel cells stack simulation model

The steady state simulation model is based on the approach of Arato et al., (2001) as implemented by some of us in a previous paper (De Simon et al., 2003). The dynamic model of the fuel cell derived in the previous section was simplified, and a local steady state model was derived and interfaced with the process simulator. The local behaviour is described as a simple electrical circuit, series of an ideal voltage, determined by the Nernst equation, and an internal resistance, made up by the sum of the three contributions: (i) contact resistance (R_c) between electrode and current collector, and electrolyte tile, (ii) Ohmic resistance (R_e), and (iii) polarization contribution (R_p). The first term is found to be constant, the second term is an exponential function of temperature, and the last one depends on both temperature and partial pressures of the species involved in the electrochemical reaction. All coefficients of this model are obtained by experimental data fittings (De Simon et al., 2003 and references therein).

The main assumptions made in the model development are (i) steady state conditions; (ii) the anodic CO electrochemical reaction neglected due to its very low rate; (iii) non-limiting diffusion in macro-pores of the electrode and in gas stream; (iv) current collector as an ideal conductor (constant voltage on whole surface); (v) adiabatic conditions; (vi) water gas shift reaction considered at the equilibrium due its high rate; (vii) ideal gas. It is important to note that diffusion phenomena occur at high reactants utilization, and heavily reduce the device efficiency. For this reason, they must be avoided by defining the cell working conditions in terms of low gas utilization. So, assumption (iii) is justified.

The model considers local temperature, pressure and composition changes along the gas path in the ducts, because of reactions, heat transfer and pressure drop. To find overall cell behaviour it is necessary to solve four sets of equations simultaneously: mass balance for each gas component, momentum balance for cathode and anode gas streams, energy balance, and local kinetics. The boundary conditions are input gas streams temperature, pressure and composition, and the assumption of adiabatic conditions. Physical and chemical properties are calculated from internal models available in the process simulator, or estimated from an empirical equation (De Simon et al., 2003). Finite difference and relaxation methods have been employed to solve numerically the system of equations in an in-house Fortran90 developed module.

2.4 The energy production simulation model

In the steady state simulation of the process, the in-house developed described above for the fuel cells stack has been introduced in a commercially available process simulator to take advantage of the available physical property calculations methods and data banks. The electricity production process around the MCFC has been modelled by using standard simulation modules. All the necessary pre-treatments, independent of process conditions, have not been considered, since they can be included in the input conditions.

The gas is heated again and sent to the compact reformer for converting part of the methane and water. The reformer and the catalytic burner - modular integrated reformer (MIR) - not available as an Aspenplus™ standard module, were modelled using other available models. It consists of plates and frames heat exchanger with one side filled with reformer catalyst. The heat required by the reaction is supplied by the gas flowing in the other side of the exchanger.

The compressor, turbine and blower are assumed to work isentropically at a fixed mechanical efficiency, and the flow rate and/or pressure are fixed at the outlet conditions. The heat exchangers, including the cogeneration heat exchanger, have fixed outlet temperatures. Heat losses and pressure drop (except those in the fuel cell) have been neglected.

2.5 The biomass gasification simulation model

The model developed for the simulation of the biomass gasification had never been presented before. It is based on experimental data of gasification behaviour of specific biomasses. The model was implemented in Aspenplus™ by using two standard chemical reactor unit operation models: RYield e RStoic.

The first reactor (RYield) basically splits the biomass into its fundamental elements (H_2O , N_2 , O_2 , H_2 , S, C, ASH and Cl_2) based on the experimental ultimate analysis of the biomass. The necessary heat for the reaction is supplied by the next module (RStoic).

In the second reactor (Rstoic) all the product formation reactions (CH_4 , H_2 , CO , CO_2 ...) that are present in the real mixture of gas after gasification were implemented. The flow rate of each component was calculated from the experimental information through the conversion of each species. The heat generated in this unit is partly transferred to the previous one (RYield), partly lost in heat losses and the rest is used to heat up the exit stream that leaves the second reactor at high

temperature (around 870 °C). Input to this unit is of course the output of the previous unit plus steam and oxygen.

The two couplet blocks allow calculating the material and energy balances, the composition of the gas at outlet, which matches the experimental data, if the ultimate analysis of the biomass is known.

The gas produced by the reaction subsystem is then treated in the clean-up unit, which is made up by (i) a tar-cracker, (ii) an H₂S removal system and (iii) a filter for the removal of the fine particulate. The clean up section is needed because the gas outlet of the gasification reaction contains tars, H₂S (which is a poison for the reforming catalyst and for the anode fuel cell), alkali and particulate. The clean up section must work at a lower temperature in order to be efficient. For this reason the temperature is reduced to 400 - 450°C by means of a heat exchange with water in the catalytic cracking reactor (Lubachyov and Richter, 1998). The H₂S is separated at 400°C by means of adsorbents (ZnO) (vv. Authors, 2001). Finally a ceramic filter for the removal of particulate is used at a temperature of around 500°C. At the end of the treatment, the clean gas is sent to the reforming unit at about 400°C. All the reactors in the clean up section have been modelled by Aspen+ RGibbs units.

Figure 3 shows the general schema of the integrated process in which one can easily distinguish the gasification section, the clean-up section, the reforming-burner and the fuel cell stacks.

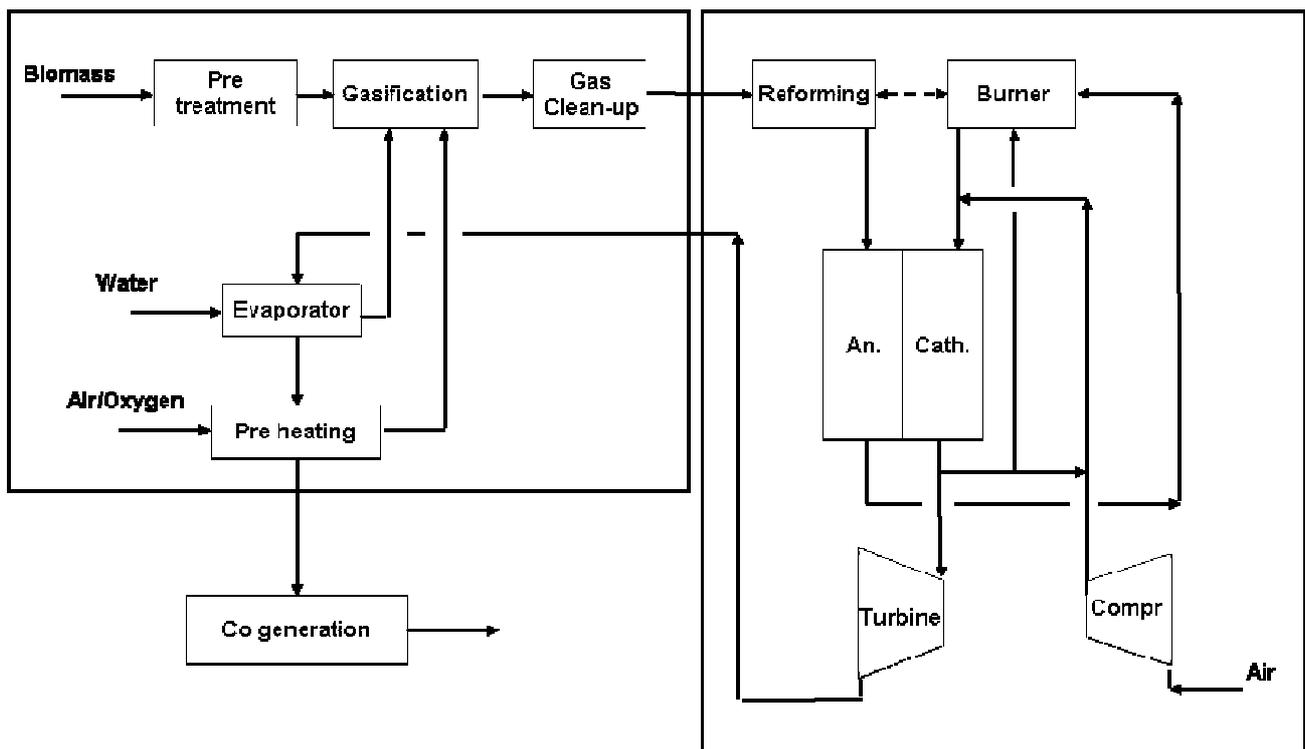


Figure 3: general schema of the process.

2. RESULTS AND DISCUSSION

Figure 4 shows the integrated simulation flow sheet as implemented in this paper. The biomass is fed to the gasification reactor (RYeld and RStoic) described above, together with oxygen, and steam preheated by exhaust gases from the turbine of the lower cycle. Stream 42 is sent to the co-generation and is used to heat stream n. 18.

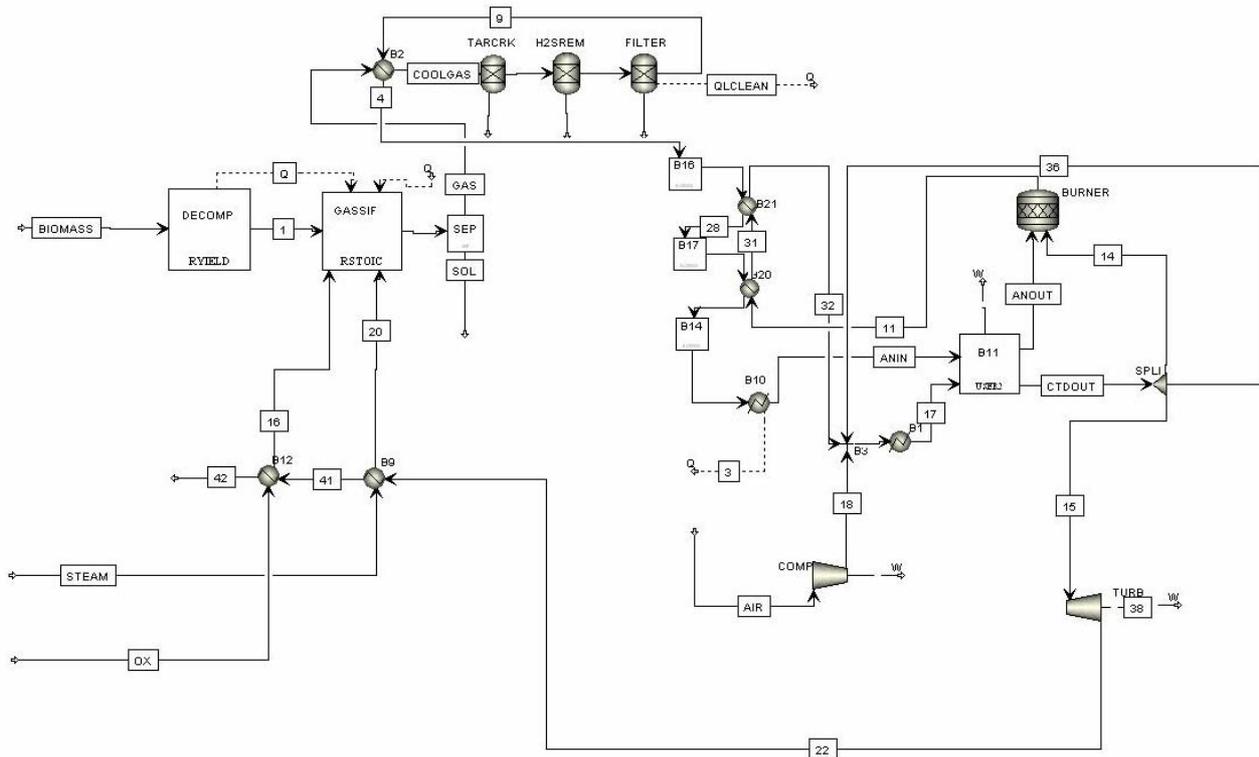


Figure 4: general schema as implemented in ASPEN+ of the coupled gasification fuel cell stack process.

The clean up section is described above. The clean gas enters the reforming unit which is modelled by 3 combined RGibbs reactors. Before entering the reactors the stream is preheated in counter current with the stream output of the burner. The gas out of the reformer is fed to the anode.

The feed to the cathode is made up of (i) the air entering the process, (ii) the stream recycled from the cathode and (iii) the output from the burner.

The cathode outlet stream is split into 3 streams. One stream is sent to the lower cycle and therefore is not recycled. A second stream is sent to the burner that requires an oxygen excess of about 30%. By doing this, the total flow rate to the burner is strongly reduced (made up of the anode output plus the part recycled from the cathode), with the direct advantage of reducing the heat dilution and improving the heat transfer for the reforming and the conversion of the fuel (the conversion is about 95%). The exit stream from the burner transfers heat to the stream entering the reformer and is recycled back. The third stream forming the cathode input is the output of the cathode that is not sent to the burner or to the lower cycle.

The model developed is used for the simulation of 3 different biomass feed conditions: (i) sugarcane bagasse (BG) which is a residue from sugar cane treatment, (ii) switchgrass (SW) and (iii) nut shells (NT) which is a mixture of 20% walnut shell, 40% hazel nut shell and 40% wood.

The results obtained are compared to experimental data of gasification of similar biomasses. The experimental and simulated data are close. Furthermore, the entire system converges to a solution that makes engineering sense and that is in line with published results, thus allowing us to apply the model to the analysis of feed variations in the system.

Table 1 reports the ultimate and proximate analysis of the three feeds, the humidity and the R ratio.

Table 1: biomass properties; data from the Gas Technology Institute, 2002.

| | BG | SW | NT |
|---------------------------------------------------------------|-------|-------|--------|
| Proximate analysis | | | |
| Ash | 6.99 | 5.24 | 2.38 |
| Volatile Subst. | 80.06 | 80.09 | 76.28 |
| C residual | 12.95 | 14.67 | 21.34 |
| HHV (MJ/kg) | 17.77 | 18.62 | 19.80 |
| Ultimate analysis | | | |
| C | 46.46 | 47.73 | 48.51 |
| H | 5.4 | 5.56 | 5.65 |
| N | 0.18 | 0.67 | 0.77 |
| S | 0.06 | 0.01 | 0.01 |
| Ash | 8.5 | 5.24 | 3.07 |
| O | 39.36 | 40.68 | 41.98 |
| Cl | 0.04 | 0.11 | 0.01 |
| Other properties | | | |
| $R = \frac{(kg_{H_2} + kg_{co})^{out}}{(kg_{biomassa})^{in}}$ | 0.24 | 0.34 | 0.38 |
| Humidity | 20 % | 12 % | 12.5 % |

The three different feeds are simulated and the results obtained are shown in Table 2. For the different biomasses the total electrical efficiency, the cogeneration efficiency, the biomass flow rate, the total electrical power generate, the falsifier cold efficiency and the conversion at anode is reported. Comparing the three feedstock considered, the net electric efficiency is similar for switch grass and nutshell (about 40%), not so for bagasse that reaches only 36,5%. The gasification efficiency for bagasse is 76.5%, for switchgrass and nutshell 82.8% and 84.5% respectively. Therefore, for constant hydrogen and carbon monoxide fed into the anode, in the bagasse case a higher feed rate is necessary, producing a poorer gas. The co generative efficiency in all the cases is not very high (less than 70%), due to the thermal dilution caused by the high fresh air flow rate.

An analysis of table 3 shows clearly that, at constant conversion at anode and constant total electrical power, the electrical and cogeneration efficiency of the different biomasses are similar but a much lower flow rate is used in the case of nut shell biomass, which is directly related to the higher heating power of this biomass.

Table 2: comparison of the performance of different biomass feeds for constant conversion in the cell.

| Biomass | Electrical Efficiency (%) | Cogeneration Efficiency (%) | Biomass Flow rate (Kg/h) | Total Electrical Power (kW) | Gasifier efficiency % | Conversion at anode (%) |
|---------|---------------------------|-----------------------------|--------------------------|-----------------------------|-----------------------|-------------------------|
| BG | 36.5 | 68.4 | 1900 | 2739.5 | 76.5 | 75 |
| SW | 40.3 | 69.1 | 1550 | 2841.0 | 82.2 | 75 |
| NT | 40.2 | 69.9 | 1450 | 2802.4 | 84.5 | 75 |

3. CONCLUSIONS

This paper presented the development and the implementation of an integrated approach to the steady state and dynamic simulation of fuel cells and power generation processes coupled with the gasification of biomass. The fuel cell used in the process is a Molten Carbonate Fuel Cell and two MCFC models were used: (i) a steady state model simulating a stack of fuel cells and a power plant and (ii) a dynamic model for the single cell. Both models are based on real plant data and the simulations are selected according to real operating conditions. The paper also presents the details of a novel integrated gasification – fuel cell steady state model.

The paper reports results of computer simulation for a realistic configuration of power generation system by MCFC, focusing on macroscopic quantities of interest such as stack efficiency, global process electrical efficiency, cogenerative efficiency, integrated with a consistent dynamic model of the cell able to consider microscopic and distributed variables behaviour such as temperature and concentration in the cells.

The software developed allows to study ‘in silico’ the effect of variations of the process conditions as well as modification of the input fuel, thus providing a useful tool for supporting technical decisions and feasibility study on the use of fuel cells in developing countries. The model was applied to three different biomasses allowing us to compare the performances of the different feeds and to understand the details of the entire process. Among the three biomass considered, the bagasse permit the lowest electric efficiency because of its difficulties in being gasified, performing the worst gasification cold efficiency and then the lowest fuel content in the producer gas.

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