TECHNICAL AND ENVIRONMENTAL ISSUES OF HYDROGEN PRODUCTION PROCESSES FROM BIOGAS AND YOUR USE IN PEMFC

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Abstract. *High energy demand required for the continuity of industrial, commercial and residential activities, and the consequential environmental pollution caused by emissions of greenhouse gases are reasons that emphasize studies of power generation from renewable resources. It is necessary to obtain greater levels of reliability in the availability of generated energy, because of the intermittent characteristics of renewable energy generation. One option to solve this problem, which is proved to be very effective, is the energy storage. In this context, fuel cell receives particular attention. Fuel cells are electrochemical devices that convert chemical energy of certain fuels to electrical energy without combustion, with higher efficiency and lower emission of greenhouse gases. However, to make fuel cell viable, it is necessary to develop effective and sustainable technologies for hydrogen production. It is possible to implement Proton Exchange Membrane Fuel Cell (PEMFC) in distributed generation, which avoids losses in power transmission. Therefore, biogas is one of the options to diversify energy sources, allowing to a planned use together with conventional sources, contributing to increase the electricity generation. This paper aims to study the technical and environmental issues of two hydrogen production processes from biogas and furthermore study the electricity generation of both processes through a PEMFC. The first process is the use of biogas in an internal combustion engine coupled with an electric generator to provide electricity to an electrolyzer, which will produce hydrogen from water. The second process is the hydrogen production by steam reforming of biogas. The studies will be based on different types of available and commercialized devices, analyzing the energy required in all processes. A pollution indicator will be used as a parameter to carry out the calculated environmental viability, with the aim of determining the ecological efficiency of all the studied processes.*

Keywords*: Hydrogen Production, Biogas, Fuel Cell, Technical Analysis, Environmental Analysis.*

1. NOMENCLATURE

AC	Alternating current “(-)”
CH_4	Methane “(-)”
CO	Carbon oxide “(-)”
CO_2	Carbon dioxide “(-)”
$(CO_2)_e$	Equivalent carbon dioxide ($kg_{emissions}/kg_{fuel}$)
$E_{cons.}$	AC power consumption of electrolyzer (kW)
F	Coulomb factor (C)
H_2	Hydrogen “(-)”
KOH	Potassium hydroxide “(-)”
LHV_{biogas}	Lower heat value of biogas (kJ/kg)
$LHV_{biogas..boiler}$	Lower heat value of biogas of steam boiler (kJ/kg)
$LHV_{biogas..reform}$	Lower heat value of biogas of reform (kJ/kg)
LHV_{H_2}	Lower heat value of hydrogen (kJ/kg)
M_{CO_2}	Carbon dioxide emission (kg_{CO2}/kg_{fuel})
M_{NO_x}	Nitrogen oxide emission (kg_{NO2}/kg_{fuel})
M_{PM}	Particulate matter emission (kg_{PM}/kg_{fuel})
M_{SO_2}	Sulfur dioxide emission (kg_{SO2}/kg_{fuel})
$\dot{m}_{biogas}$	Biogas flow (kg/s)

$\dot{m}_{biogas.boiler}$	Biogas of steam boiler flow (kg/s)
$\dot{m}_{biogas..reform}$	Biogas of reform flow (kg/s)
$\dot{m}_{H_2}$	Hydrogen flow (kg/s)
$\dot{m}_{steam}$	Steam flow (kg/s)
NO_x	Nitrogen oxide "(-)"
PEMFC	Proton exchange membrane fuel cell "(-)"
PM	Particulate matter "(-)"
SO_2	Sulfur dioxide "(-)"
V_c	Cell voltage (V)
$\dot{W}_{FC}$	Installed power of fuel cell (kW)
$\dot{W}_{gas\ genset}$	Electric power of gas genset (kW)
Greek letters	
Δh_{steam}	Enthalpy change of steam (kJ/kmol)
ε	Ecological efficiency (%)
η_{FC}	Efficiency of fuel cell (%)
$\eta_{electrolyzer}$	Efficiency of electrolyser (%)
$\eta_{gas\,genset}$	Efficiency of gas genset (%)
$\eta_{electrolyzer.gas\,genset}$	Efficiency of electrolytic process (%)
$\eta_{Rbiogas}$	Efficiency of steam reform of biogas (%)
$\eta_{steam\,boiler}$	Efficiency of steam boiler (%)
η_{system}	Global efficiency of the system (%)
$\prod_g$	Pollutant indicator (kg/MJ).

2. INTRODUCTION

Energy alternatives to replace or complement the sources used currently are a constant challenge for researchers. In this context, hydrogen has been researched, because of its large energy storage capacity and enables the reduction in emissions of greenhouse gases. However, hydrogen does not exist alone in nature in its pure state, being associated with other elements such as water, fossil fuels and all living beings (Silveira et al., 2008).

There are several technologies for the production of hydrogen. Some process makes use of fossil fuels, such as the steam reform of natural gas, the technology most used currently (Silveira et al., 2009). On the other hand, others makes use of renewable energy sources, among them it can highlight the electrolysis from wind energy, sun and water, such as wind turbines, solar panels and hydroelectric plant, respectively.

Costa (2006) points out that although fossil fuels exercise a fundamental role as energy source in human activities, the renewable energies, including biomass, have been increasingly studied in Brazil and abroad, as a result of growing awareness of society about the its positive aspects, mainly looking for new energy sources with the objective of not dependence on a single energy source.

Another point made by ANEEL (2015) says that biogas is one of the options to diversify energy matrix, allowing a planning of their use in combination with existing conventional sources, and thus it can implement distributed generation, that consists of micro and mini generation distributed of electricity, innovations that can combine financial savings, environmental awareness and sustainability.

Therefore, the "Fig. 1" shows the schematic representation of developed project, that is the hydrogen production processes and your use in PEMFC.

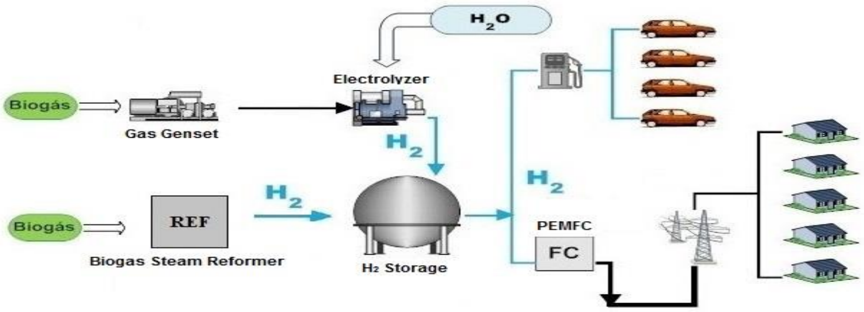

Figure 1. Schematic representation of developed project.

3. TECHNICAL ANALYSIS OF WATER ELECTROLYSIS PROCESS

3.1 Technical analysis of electrolyzer

To realize the technical analysis of electrolyzer, it was chosen the pressurized water equipment Hydrogenics HySTAT®-10-10 (Hydrogenics, 2012), where the basic technical characteristics are shown in "Tab. 1".

Table 1. Characteristics of electrolyzer Hydrogenics HySTAT®-10-10 (Hydrogenics, 2012).

Nominal hydrogen flow	10 Nm^3/h
Estimated AC power consumption	49 kW (4,9 kWh/Nm^3 at full load)
Frequency	60 Hz ± 3%
Demineralized water consumption	< 1 liter/Nm^3 H_2
Electrolyte	H_2O + 30% wt. KOH

In this cell stack, water (mixed with 30% KOH) is broken down into its basic elements, hydrogen and oxygen, by means of a DC current. The cell stack consists of a series of interconnected, circular electrolysis cells, each containing two electrodes located on either side of an advanced patented inorganic ion-exchange membrane, which is manufactured in-house. The purpose of the membrane is 2-fold: to allow ion transfer with the minimum of resistance and to prevent recombination of the produced hydrogen and oxygen (Hydrogenics, 2012).

The choice of the electrolyzer was based on hydrogen production capacity in order to supply the demand required by the PEMFC, aiming its use in automotive vehicle.

3.2 Technical analysis of gas genset

To accomplish the technical analysis of the gas genset, which is the internal combustion engine associated to an electric generator, it was chosen the equipment Stemac® MWM/International G6.12T (Stemac, 2012) where the technical characteristics of the selected model are shown in "Tab. 2". This system produce electricity to run the electrolyzer and for other requirements in the local of hydrogen production.

Table 2. Characteristics of gas genset Stemac® MWM/International G6.12T (Stemac, 2012).

Estimated AC power consumption	70 kW
Installed power	87 kVA
Fuel	Biogas
Frequency	60 Hz

The mentioned equipment can be used in three different situations; each one has different electric power. For technical analysis, it was used the continuous mode (base), that means without interruption of electricity supply. However, the gas genset can operate in emergency mode (Standby) (120 kVA/96 kW), acting as a generator in a power failure, or Prime mode (104 kVA / 83 kW) for operation peak hours, in the range between 17h and 22h.

It is important to point out that the choice of equipment was based on the electric power of electrolyzer, where the generator must provide equal or greater electric power to supply electricity to electrolytic process.

3.3. Thermodynamic Efficiency

The calculation of the theoretical efficiency of the electrolytic process is based on the maximum electrical efficiency of equipment. For the electrolyzer, its efficiency was calculated using a commercially available electrolyser. On the other hand, the efficiency of the genset was collected from the literature.

Considering the electrolyser Hydrogenics HySTAT®-10-10, it can calculate thermodynamic efficiency using "Eq. (1)", the hydrogen flow equal to 10 Nm³/h (247,22.10^{-6} kg/s), LHV of the same gas is 119.950 kJ/ kg and device power equal to 49 kW. (Braga, 2014 apud Krona, 2012).

$$\eta_{electrolyzer} = \frac{\dot{m}_{H_2} . LHV_{H_2}}{E_{cons.}} \tag{1}$$

The thermodynamic efficiency of the electrolytic process is described by "Eq. (2)" considering the electrical efficiency medium of gas engines (Otto cycle) is 27.5% (Silva, 2015).

$$\eta_{electrolyzer.gas\,genset} = \eta_{electrolyzer} * \eta_{gas\,genset} \tag{2}$$

3.4 Energy Efficiency

The objective of energy analysis in the electrolytic process is determining the biogas flow required by gas genset to provide electricity to electrolyzer. It was used the “Eq. (3)” to calculate the biogas flow based on its LHV.

$$\dot{m}_{biogas} = \frac{\dot{W}_{gas\ genset}}{\eta_{gas\ genset} * LHV_{biogas}} \tag{3}$$

4. TECHNICAL ANALYSIS OF STEAM REFORMING OF BIOGAS

The objective of thermodynamic analysis of steam reforming of biogas is determining the energy efficiency. The steam reforming process consists of a steam generator and two reformers, being of reform and shift reactor. Thus, the overall process product is hydrogen and, as reagents, there are the fuel used in the steam generator and reform.

In the biogas steam reforming process, biogas was also considered as a fuel, not only burned in the steam boiler, but also consumed by the steam reforming reaction.

The "Figure 2" illustrates the hydrogen production by steam reforming of biogas

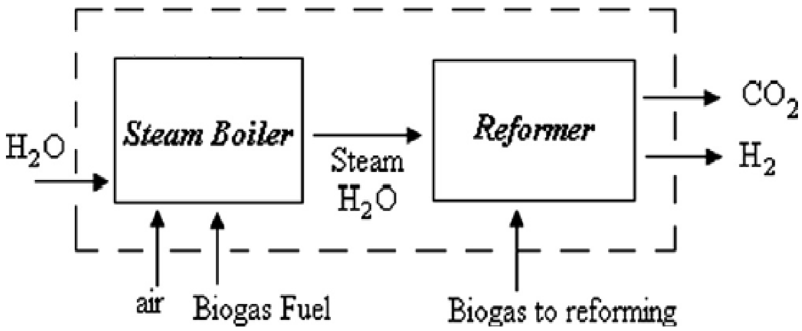

Figure 2. Systems of the reformer and the steam boiler (Braga et al., 2013).

To reform of biogas, it was considered two reactions occurring simultaneously in the first reactor, the steam reform and dry reform. The "Table 3" shows the stoichiometric reactions in reactor 1 (reform), the reactor 2 (shift reactor) and subsequent, the global reaction of the process.

Table 3. Stoichiometric reactions for the biogas reform.

Steam reform + dry reform (reactor 1)	$CH_4 + 0,669\ H_2O + 0,331\ CO_2 \longrightarrow 1,331\ CO + 2,669\ H_2$
Shift reform (reactor 2)	$1,331\ CO+1,331\ H_2O \longrightarrow 1,331\ CO_2+1,331\ H_2$
Global steam reform (reactor 1 + reactor 2)	$CH_4+2\ H_2O \longrightarrow CO_2+4\ H_2$

The flows of reagents and products of the reform process were calculated by stoichiometry for the production of 10 Nm^3/h ($2,472.10^{-4}$ kg/s) of hydrogen, based on the biogas and steam flows in the reformer.

For calculating the biogas flow used in the steam boiler, it was used “Eq. (4)” considering the boiler efficiency operating with biogas equal to 90% (Lora, E; Nascimento, M, 2004). Thus, using steam at a temperature of 700 °C, the efficiency process is calculated using the “Eq. (5)”

$$\eta_{steam\ boiler} = \frac{\dot{m}_{steam}.\Delta h_{steam}}{\dot{m}_{biogas}.LHV_{biogas}} \tag{4}$$

$$\eta_{Rbiogas} = \frac{\dot{m}_{H_2}.LHV_{H_2}}{\dot{m}_{biogas.boiler}.LHV_{biogas.boiler} + \dot{m}_{biogas.reform}.LHV_{biogas.reform}} \tag{5}$$

5. ENRIRONMENTAL ANALYSIS OF HYDROGEN PRODUCTION PROCESSES

The environmental analysis was performed to evaluate how pollutant is a biogas reforming system. This analysis was based on concepts of equivalent carbon dioxide, pollutant indicator, and ecological efficiency.

5.1 Equivalent carbon dioxide

The calculation of equivalent carbon dioxide $(CO2)_e$ is made considering the concentrations of each component separately (CO_2, SO_2, NOx, PM) for burning 1 kg of fuel, which, in this work is biogas. "Equation (6)" shows how it can be calculated carbon dioxide equivalent (Silveira et al., 2012):

$$(CO_2)_e = M_{CO_2} + 80*M_{SO_2} + 50*M_{NO_x} + 67*M_{PM} \tag{6}$$

5.2 Pollutant indicator

The pollutant indicator is used to quantify the environmental impact of burning fuel in relation to its energy potential, evaluated by the division between the amount of $(CO2)_e$ emitted by LHV of the fuel, that is biogas (Silva, 2010), as defined by the "Eq. (7)" (Coronado et al., 2010).

$$\Pi_g = \frac{(CO_2)_e}{LHV_{biogas}} \tag{7}$$

5.3 Ecological efficiency

The ecological efficiency considers emissions by fuel and electricity used in a system, allowing quantifies its level of pollution. The ecological efficiency has a range from zero up to one: a value equal to zero means 100% of environmental impact, determining a high pollution system and a value equal to one, it means 0% environmental impact, in others words, the system is not polluter (Coronado et al., 2010).

To calculate the ecological efficiency of the system, it is used two fuels as a reference, representing the pollution extremes. Hydrogen is the least polluting fuel, presenting pollutant indicator equal to zero ($\Pi g = 0$ kg/MJ). Nevertheless, sulfur is admitted as a more polluting fuel ($\Pi g = 134$ kg/MJ). Based on these conditions, the ecological efficiency of any system that uses hydrogen is equal to one ($\varepsilon = 1$) and any system that uses sulfur as fuel will have ($\varepsilon = 0$), independent of the energy efficiency system values. "Equation (8)" shows how the ecological efficiency is calculated (Silveira et al., 2012):

$$\varepsilon = \left[\frac{0,204 \,.\, \eta_{system} .\, ln\left(135 - \Pi_g\right)}{\eta_{system} + \Pi_g}\right]^{0,5} \tag{8}$$

6. HYDROGEN USE IN FUEL CELL FOR ELECTRICITY GENERATION

This topic approaches the use of hydrogen produced by both processes with application in an electric automotive vehicle operating with a fuel cell, with the aim to stimulate technological advances in transport systems.

For the application of hydrogen in the fuel cell, it was selected Proton Exchange Membrane Fuel Cell (PEMFC) as technology, in view of its application in transportation and electric utility systems.

The technical characteristics of the selected model, Honda FCX Clarity can be seen in the "Tab. 5".

Table 5. Characteristics of vehicle Honda FCX Clarity (Honda, 2016).

Fuel cell type	Proton Exchange Membrane Fuel Cell (PEMFC)
Required fuel	Hydrogen gas compressed
Installed power	100 kW

6.1 Technical analysis of PEMFC

6.1.1 Hydrogen consumption

The hydrogen input flow required for a fuel cell is proportional to the current or operation of the fuel cell power, i.e, the gas is consumed in accordance with the variation of the current (Dicks, A; Larminie, J., 2003).

Thus, the chemical reaction that occurs in the cell produces for every 2 moles of hydrogen together with 1 mol of oxygen, 2 moles of water, where hydrogen molar mass is $2{,}02.10^{-3}$ kg/kgmol (Dicks, A; Larminie, J., 2003). Therefore, "Eq. (9)" was used to estimate the consumption of H_2 during the operation of the fuel cell, considering cell voltage (V_c) equal to 0,65 V and Coulomb factor (F) equal to 96485 C.

$$\dot{m}_{H_2}=\frac{2,02.10^{-3}.\dot{W}_{FC}}{2.V_c.F} \tag{9}$$

6.1.2 Thermodynamic efficiency of PEMFC

The First Law of thermodynamic efficiency is represented by the division of the products by the energy source used. Thus, electricity generation efficiency of the First Law for the system is the power output of fuel cell divided by energy provided by hydrogen, according to "Eq. (10)" (Colombaroli et al., 2015).

$$\eta_{FC}=\frac{\dot{W}_{FC}}{\dot{m}_{H_2}.LHV_{H_2}} \tag{10}$$

6.1.3 Global thermodynamic efficiencies of the systems

The global thermodynamic efficiencies of the systems that involve the electrolysis of water (Gas Genset/Electrolyser/PEMFC) and steam reforming of biogas (Reformer/PEMFC) are based on the "Eq. (11)" and "Eq. (12)".

$$\eta_{global.electrolysis}=\eta_{electrolyzer}*\eta_{gas\,genset}*\eta_{FC} \tag{11}$$

$$\eta_{global.reform}=\eta_{reform}*\eta_{FC} \tag{12}$$

7. RESULTS AND DISCUSSION

From the calculation methodology related to efficiency of hydrogen production processes, it can compare the performance of each procedure and evaluate which is more advantageous thermodynamically and ecologically, as well as evaluate the global thermodynamic efficiency of each process with the use of PEMFC.

With the aim to check the influence of the biogas composition in the processes, it was made a literature review to obtain compositions of biogas for different types of biomass, in order to calculate the LHV of each mixture based on the calculation method available in the literature. It is important to point out that the final composition of biogas depends on the type of substrate to be digested, influencing the LHV and proportions of CO_2 and CH_4 presents in the constitution of the gas. The "Table 6" describes the different compositions of biogas for each type of biomass and the LHV it was calculated according "Eq. (13)", considering the molar mass of CH_4 and CO_2 equal to 16 kg/kmol and 44 kg/kmol, respectively (Campani et al.,2009).

$$LHV_{biogas} = 50,052\ MJ/kg\ x\ (mass\ percentage\ of\ CH_4\ in\ the\ biogas) \tag{13}$$

Table 6 - Composition of biogas from different types of biomass and its LHV (Lins, P et al., 2015; Ensinas, A. V, 2003)

Types of Biomass	Biogas Composition			LHV (kJ/kg)
	CH_4 (%)	CO_2 (%)	H_2S (ppm)	
Cassava starch	54,3	44,9	97	15.287,99
Landfill	55,13	44,16	-	15.627,61
Bovine breeding	59,6	39,1	329,3	17.849,51
Pig breeding	62	37	2.782,30	18.950,99
Mixed (Rearing poultry + Bovine breeding)	69,2	29,8	64,1	22.914,97

Thus, the efficiency of electrolysis of water process is 15,28% ($\eta_{electrolyzer}$= 55,56% ; $\eta_{gas\,genset}$= 27,5 %) , according to "Eq. (5)". The hydrogen consumption of PEMFC is $1,61.10^{-3}$ kg/s, according to "Eq. (9)". Therefore, the thermodynamic efficiency of PEMFC is 51,77 %, according to "Eq. (10)". Finally, the global thermodynamic efficiency of electrolysis of water is 7,91 %, according "Eq. (12)".

The "Figure 3" shows the comparison between the thermodynamic efficiencies of hydrogen production processes, as well as the global efficiency of the hydrogen use in PEMFC. In relation to steam reforming of biogas, it was considered the average efficiency from all types of biomass.

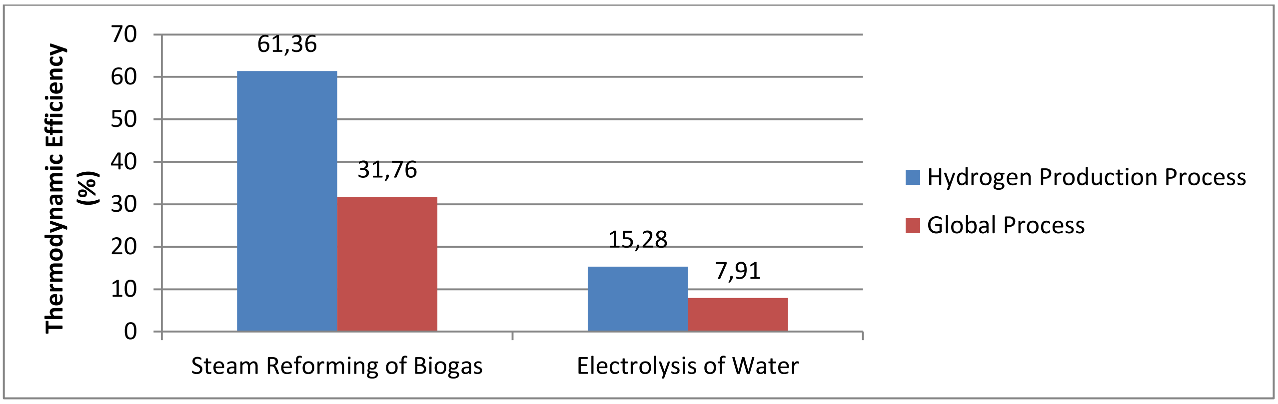

Figure 3. Comparative of thermodynamic efficiencies.

One reason for the efficiency of electrolysis is less advantageous compared to steam reforming is the inclusion of several equipment in the hydrogen production process, occurring losses during the process.

With the aim to check the influence of the biogas composition in the processes, "Fig.4" shows the thermodynamic efficiency of biogas steam reforming process and "Fig.5" illustrates the energy efficiency of gas genset, where both figures are according to concentration of CH_4 present in biogas.

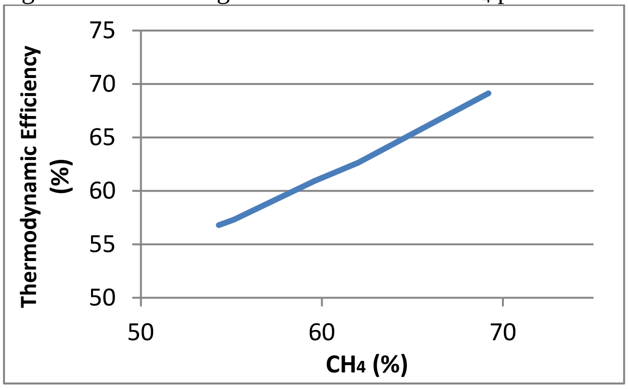

Figure 4. Thermodynamic efficiency of biogas steam reforming process.

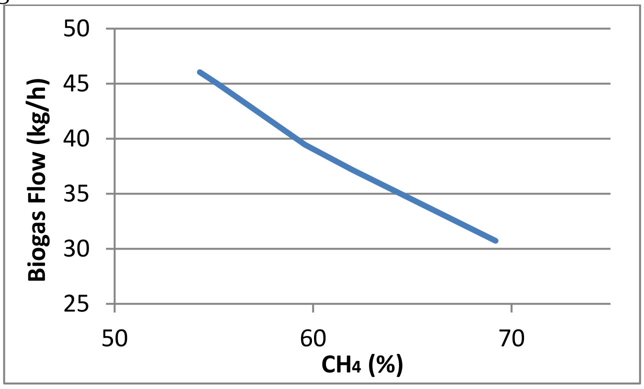

Figure 5. Energy efficiency of gas genset.

According to "Fig. 4" and "Fig. 5", the concentration of CH_4 present in the biogas influences directly the biogas flow required to meet the gas genset, as well as the thermodynamic efficiency of steam reforming, showing the superiority of CH_4 LHV in relation to LHV of CO_2. However, CH_4 is more harmful to the environment than CO_2, requiring caution in its use in order to reduce environmental impacts on energy conversion. Thus, the "Table 7" describes the ecological efficiencies of each composition of biogas from different types of biomass.

Table 7 – Ecological efficiencies of each composition of biogas from different types of biomass.

	Cassava starch	Landfill	Bovine breeding	Pig breeding	Mixed
Steam Reforming of Biogas	79,16 %	79,31 %	80,24 %	80,63 %	81,80 %
Electrolysis of Water	77,22 %	77,43 %	78,67 %	79,19 %	80,72 %

It is important to point out that the emissions of CO_2 from both processes do not contribute to global warming. It can be explained by the fact that CO_2 from biogas is from recently alive plant matter (even if it was fed to animals), it is part of a CO_2 cycle – i.e. CO_2 given off by burning biogas is absorbed by plants that will provide future biogas; besides that animal manures release methane into the atmosphere. CH_4 is a more potent greenhouse gas than CO_2, so it is a good idea to use it to produce hydrogen rather than releasing it (Braga et al., 2013).

8. CONCLUSIONS

Based on obtained results and information available in references, this paper shows that steam reforming of biogas is most advantageous in thermodynamic aspect than electrolysis of water to produce hydrogen and electricity by PEMFC.

According to the ecological analysis, the hydrogen produced by both processes, is an environmental promising technology, due to its high ecological efficiency when considering the CO_2 cycle. It is important noteworthy that these procedures are sustainable way to produce hydrogen in large scale. However, it cannot affirm ecological superiority of one process, because the results obtained are very close, and the difference each other, at most 2%, is contained in the statistical error.

Therefore, this paper shows that the fuel cell could be a promising alternative for electricity generation due to the high efficiency and the lower emission of pollutants in comparison with gas genset. From the thermodynamic point of view, the data shows that the fuel cell system can be more efficient than the gas genset. The gas genset using biogas as fuel has thermodynamic efficiency of 27,5%, and fuel cells have thermodynamic efficiency of 51,77%.

With the use of biogas to generate electricity and/or hydrogen, it will be given a noble destination for this energy source, instead of execute its burn and release CO_2 into the atmosphere.

The hydrogen, the main energy carrier to fuel cells and its production by the steam reforming of biogas, is the best way to guarantee the volume of production necessary to produce hydrogen in sustainable way. The integration or association of hydrogen production with the electrolysis of water can certainly boost the "Hydrogen Era" in the near future.

9. ACKNOWLEDGEMENTS

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