

Exergy Analysis of Syngas Production via Biomass Thermal Gasification

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Abstract

Biomass has great potential as a clean, renewable feedstock for producing modern energy carriers. This paper focuses on the process of biomass wastes gasification, where the synthesis gas may subsequently be used for the production of electricity, fuels and chemicals. The gasification process is one of the least-efficient operations in the whole biomass-to-energy technology chain and an analysis of the efficiency of the gasifier alone can substantially contribute to the efficiency improvement of this chain. In this work, biomass wastes from canneries (peach pits) and wine industry (marcs and stalks) used for the syngas production are investigated. Thermodynamic indicators of process performance based on the second law (exergy analysis) was utilized in order to evaluate the effect of different operational parameters (temperature, supply air/stoichiometric air, supply steam/carbon ratio and moisture feed). The exergetic efficiency of the gasification process decreases when the all considered operational parameters increase. The gasification of peach pits shows the maximum values of the exergetic efficiency in all conditions.

Keywords: Gasification; biomass wastes; exergetic efficiency.

1. Introduction

The use of renewable energy sources and a more efficient use of energy have been increasing in the world some years ago, mainly motivated for a growing environmental awareness in order to reduce green-house gases emissions and the increase of fuel prices that drives up the prices of energy. Many current energy policies promote research to enhance the utilization of renewable energy sources, in large part to help mitigate environmental problems and improve the national energy security of countries dependent on the use of imported fossil fuels. Among renewable energy sources, biomass (e.g. paper, agriculture and forestry wastes, straw, wood wastes, sawdust, paddy husk) is currently one of the most popular options [1].

Agro-industrial wastes in Cuyo Region, Argentina, such as olive kernels from olive oil production units, grape wastes from wineries, or fruit stones from fruit processing industries, are inefficiently utilized or even in some cases totally wasted. The waste-to-energy technology route can lead to the displacement of fossil energy sources and therefore contribute significantly to the mitigation of greenhouse gas (GHG) emissions. The conversion of biomass wastes to useful energy forms encompasses a wide range of conversion options, such as biochemical and thermochemical processes. Overall efficiencies for biomass-based power generation tend to be low, typically ranging from 15% for small plants to 30% for larger plants, compared to those for the most efficient energy conversion plants, e.g., natural gas combined cycles [2, 3]. Due to a large variation of biomass feedstocks, conversion

technologies, and biofuels, the future bioenergy systems can be designed almost from scratch.

Considering the thermochemical processes, the biomass gasification has been proven in the past as an efficient mean to transform solid fuels into energy carriers. Gasification of biomass consists of partial oxidation at high temperature (800 – 1000°C) and subsequent reduction reactions. The process produces a gas mixture, called syngas, which contains hydrogen, carbon monoxide, carbon dioxide, water, methane and higher hydrocarbons (tar). The gasification of biomass can be done using air, oxygen steam or a mixture of them as the gasifying agents. While air based gasification results in adulteration of the syngas with high percentage of nitrogen, steam gasification of biomass requires an additional source of energy for maintaining the reactor temperature [4].

Waste biomass has special chemical properties such as carbon reactivity, high volatility and low percentage of ash and sulphur that turn it to an appropriate fuel for gasification. The process, in comparison with coal, is able to be performed at lower-level temperature, with a less amount of time and fewer difficulties concerning emissions as well as the reactor walls corrosions. Nevertheless, biomass also has significant disadvantages including its high content of moisture and low density of energy [5].

Gasification of biomass fuels results in a gas mixture mainly containing CO, H₂, N₂, CO₂, and some hydrocarbons (CH₄, C₂H₄, and C₂H₆). Very small quantities of NH₃, H₂S, and tars can also be produced [5]. During gasification small amounts of unwanted materials are also produced. They include tar, char, ash, etc. The gasification of biomass results in the production of syngas having a

wide range of heating values as a direct result of both the gasifier design used and the reactants chosen. Syngas has less energy density (kJ/m^3) than natural gas. The main characteristics of the syngas fuel are the lower heating value, the H_2/CO ratio, and the fraction (up to 50%) of non-combustible such as steam, carbon dioxide, and nitrogen. In order to select the operational conditions the gasification efficiency must be maximized, this aspect implies a correct use of thermodynamics [6].

The gasification efficiency depend on operating parameters such as gasification temperature (T), equivalence ratio (ER, supply air/stoichiometric air) and supply steam/carbon ratio (SBR) [7]. Thermodynamic indicators of process performance based on the second law (exergy analysis) are nowadays commonly accepted as the most natural way to measure the performance of different processes, ranging from energy technology, chemical engineering, transportation, agriculture, etc. The identification of the inefficiencies and their sources allows effective management and optimization of gasification process [8].

Reasonably, two fundamental aspects related to biomass use as fuel are: (1) to extend and improve the basic knowledge on composition and properties; and (2) to apply this knowledge for the most advanced and environmentally safe utilization. Taking into account this aspect, the objective of this study is to investigate the thermodynamic performance by making application of the mathematical modeling technique to establish conditions leading to the production of maximum combustible syngas composition with maximum efficiencies. Three biomass feedstock are studied using the exergy analysis, marcs, stalks and peach pits from canneries and wine industry.

2. Materials

The raw material used in this work were peach pits, from canneries, marc and stalks from wine industry. Both industries are located in the province of San Juan, Cuyo Region of Argentina. The weight loss at 105°C , ash and organic matter content were determined according to ASTM standards (ASTM D3173-87, ASTM D3172-89 (02)). Ultimate analyses of the samples were performed using EuroEA3000 model elemental analyzer.

The values of lignin, cellulose and holocellulose contents, expressed in percentage, were determined by ASTM standard (ASTM D1106-56, ASTM D1103-60 and ASTM D1103-60, respectively). The hemicellulose contents was determined by difference between holocellulose and cellulose contents. These analysis were carried out by the laboratory of the Chemistry Section of Agro Products (EEAOC-Estación Experimental Agroindustrial Obispo Colombres) -Tucumán / Argentina. The analysis results are expressed in dry basis. The results are shown in Table 1, 2 and 3.

Table 1. Results of Proximate Analysis (Dry Basis, Weight Percentage).

Biomass type	Proximate analysis (%), dry basis			
	Moisture	Volatile matter	Fixed carbon	Ash
Peach pit	35.57	79.12	19.85	1.03
Grape stalk	73.22	60.58	27.37	12.05
Grape marc	55.05	68.54	21.98	9.48

The higher heating value was estimated using the correlation proposed by Channiwala and Parikh [9]:

Table 2. Results of Ultimate Analysis (Dry Basis, Weight Percentage).

Biomass type	Ultimate analysis (%), dry basis				
	C	H	O*	N	S
Peach pit	53.01	5.90	36.89	2.32	1.88
Grape stalk	46.14	5.74	37.54	6.37	4.21
Grape marc	52.91	5.93	30.41	5.41	5.34

*By difference

Table 3. Contents of lignin, cellulose and hemicellulose determined by ASTM standard methods.

	Peach pit	Marc	Stalk
Hemicellulose (%)	21.02	4.96	5.78
Cellulose (%)	31.58	15.31	16.02
Lignin (%)	27.53	37.97	30.79

$$HHV(\text{MJ/kg}) = 0.3491C + 1.1783H + 0.1005S - 0.1034O - 0.0151N - 0.0211A \quad (1)$$

where C, H, S, O, N and A are the contents of carbon, hydrogen, sulfur, oxygen, nitrogen and ash in the peach pits, mark and stalk respectively. The calculated high calorific values are 20.65, 18.33 and 20.41MJ/kg for peach pit, grape stalk and marc, respectively.

The concentration of 28 elements samples peach pits, marc and stalks were determined using a plasma mass spectrometer with Shimadzu ICPE 9000. The results are shown in Table 4. Fe, Cr, Pb, Sn, Mo, Ni, Ag, Ti, V, Mn, S, Cd and B are undetectable.

Table 4. Metals contents in agro-industrial wastes (dry basis).

Metal	Stalk	Peach Pits	Marc
	ug/g	ug/g	ug/g
Cu	17,31	15,32	14,86
Al	28,86	27,19	26,97
Si	4,22	1,66	4,042
Na	149,1	103,1	105,9
K	191,9	118,5	160,7
Ca	276,6	53,4	165,4
Mg	63,72	26,85	48,67
Zn	24,26	22,42	22,36
P	63,64	15,03	72,27
Ba	5,109	12,71	5,362
As	3,647	3,472	2,119
Co	3,571	3,616	3,544
In	8,139	7,77	8,275
Li	5,985	5,73	5,759
Tl	10,38	11,14	10,55
Sr	1,755	0,49	0,803

3. Methodology

In this work, the nonstoichiometric equilibrium model was applied in order to obtain the composition of syngas, providing valuable information about the optimal parameters of the gasification process, with the aim of

obtain a gaseous fuel with higher calorific power. This model is based on minimizing Gibbs free energy in the system without specification of the possible reactions taking place [10]. In the absence (or substoichiometric presence) of oxygen, several reduction reactions occur in the 800–1000°C temperature range. These reactions are mostly endothermic. The important chemical reactions that occur during biomass gasification are based on the following assumptions: 1) biomass is represented by the general formula CH_aO_b . Due to the biomass contains negligible amount of N and S in comparison to the previous elements, they are not considered 2) the reactions are at thermodynamic equilibrium (at atmospheric pressure = 1 atm), 3) the reactions proceed adiabatically (heat losses neglected), 4) ash of biomass wastes are not considered, 5) the reactions of heat losses are neglected (Adiabatic process), 6) no chars living with the exit of the gasifier products.

The main reactions in this category are as follows:

Steam gasification:



Boudouard reaction:



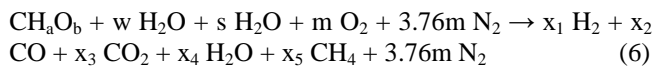
Water-gas-shift reaction:



Methane reaction:



The syngas composition (the H_2 , CO , CH_4 , CO_2 , H_2O and N_2 content) depends on the gasification agent (air/steam ratio) and the others process parameters. The important reactions that occur during biomass conversion based on the above assumptions can be described using the following global gasification equation:



where w is the water mol / biomass mol (moisture content), s is steam mol / biomass mol, m , the oxygen mol / biomass mol, x_1 , x_2 , x_3 , x_4 and x_5 , are unknown fractions of exit gas composition. If the gasification is carried out only with air, $s = 0$.

3.1. Exergy Efficiency of Gasification Process

The mass and energy balances form the basis for exergy analysis [10]. Exergy of all process streams (mass streams including biomass, and heat and work) is calculated according to the method described by Szargut [11] and recently updated by Rivero [12]. The exergy analysis technique estimates the efficiency of the process and determines the energy quality and usefulness [13]. This analysis makes us able to specify the maximum performance of a system and the sources of the irreversibilities [14], and it had been used to evaluate the performance of different systems and to improve their efficiencies [15–16]. The relationship between the exergy analysis and the sustainability shows that diminution the exergy loss leads to increment of the sustainability of energy use [17].

During the gasification the chemical energy of the biomass changes into the thermal and chemical energy of the syngas. Gasification achieves a higher conversion rate in comparison with pyrolysis and combustion [18]. The energy efficiency of biomass conversion depends on the

operational conditions of the gasification, its performance is evaluated using the second thermodynamic law, applying the exergy analysis concept to the model of energy exchange process.

This analysis requires identification of the possible products, which are emitted after the gasification process takes place. In this analysis, these products are comprised of a mixture of H_2O , N_2 , H_2 , CO_2 , CH_4 and CO . The following assumptions are made in the analysis and simulation:

- The entire flow processes occur at steady state conditions, and in order to avoid the kinetic effects, residence time is not considered.
- Catalysis is not considered in the simulation or thermodynamic assessments.
- Ambient air is considered on a volume basis as 79% nitrogen and 21% oxygen.
- The reference-environment temperature is 25°C and pressure is 1 bar, and these values apply to ambient air.

The exergy balance of biomass gasification can be expressed:

$$\sum \varepsilon_{in} = \sum \varepsilon_{out} + I \quad (7)$$

where ε_{in} and ε_{out} are the input and output exergy, I is the produced irreversibility during the conversion process. The available exergy in a stream of material can be calculated as the sum of its chemical and physical exergy.

$$\varepsilon = \varepsilon_{ch} + \varepsilon_{ph} \quad (8)$$

The kinetic exergy is negligible. The physical exergy for the gaseous material is calculated with the following expression:

$$\varepsilon_{ph} = \sum_i y_i \cdot \left[\left(\int_{T^\circ}^T C_{p_i} \cdot dt \right) - T^\circ \left(\int_{T^\circ}^T \frac{C_{p_i}}{T} \cdot dT \right) \right] \quad (9)$$

where i is the gaseous component, y_i and C_{p_i} are the molar fraction and heat capacity at constant pressure (kJ/mol) of component i , respectively. The physical exergy represents the enthalpy change of a particular species from its standard state to a determinate temperature and pressure and also counts the entropy change in the process.

The mix chemical exergy is the standard chemical exergy of all mix component (first term of Eq. (10)) and the entropic change due to the mixing of different gaseous species (second term of Eq. (10)). This exergy can be calculated with the following equation:

$$\varepsilon_{ch} = \sum_i y_i \cdot \varepsilon_{ch,i} + RT^\circ \sum_i y_i \ln y_i \quad (10)$$

where $\varepsilon_{ch,i}$ is the standard chemical exergy of gaseous component i , R is the universal gas constant 8.314 kJ/mol K, y_i are the molar fraction of the species i .

The values of the molar enthalpy, entropy and standard chemical exergy of the gaseous component are shown in Table 5, the chemical exergy of char is 410260 kJ/kmol [19].

Due to the biomass is fed to gasification reactor at environmental temperature, the biomass physical exergy is negligible. The biomass chemical exergy is calculated by:

$$\varepsilon_{ch,biomass} = m_{biomass} \cdot \beta \cdot LHV_{biomass} \quad (11)$$

Table 5. Molar enthalpy, standard entropy and chemical exergy for gaseous compounds [20].

Component	H° (kJ/kmol)	S° (kJ/kmol K)	$\varepsilon_{ch,i}$ (kJ/kmol)
N ₂	0	191.61	668
O ₂	0	205.033	3970
H ₂ O(g)	-228.583	188.720	9500
CO	-137.150	197.543	275100
CO ₂	-394.374	213.685	19870
H ₂	0	130.574	236100
CH ₄	-74.850	186.16	831650

where $LHV_{biomass}$ is the lower heating value of biomass, and β is a factor dependent upon mass fraction of oxygen, carbon, hydrogen, and nitrogen in the biomass and is expressed as [21]:

$$\beta = \frac{1.044 + 0.0160 \cdot \left(\frac{H}{C}\right) - 0.3493 \cdot \left(\frac{O}{C}\right) \cdot \left[1 + 0.0531 \cdot \left(\frac{H}{C}\right)\right] + 0.0493 \cdot \left(\frac{N}{C}\right)}{1 - 0.4124 \cdot \left(\frac{O}{C}\right)} \quad (12)$$

where H/C, O/C y N/C are the atomic ratios of different elements present in the biomass.

The biomass low heating value can be calculated by:

$$LHV_{biomass} = HHV_{biomass} - 9 \cdot m_H \cdot \Delta H_{steam} \quad (13)$$

where m_H is the hydrogen mass fraction in the biomass, ΔH_{steam} is the latent heat of water vaporization at 298K and 1 atm expressed in MJ/kg, and $HHV_{biomass}$ is the biomass higher heating value reported previously.

The exergy efficiency is a concept applied to stationary open system and it links the effective exergy leaving the system with the introduced exergy into the system. The exergy efficiency of gasification reactor is defined by:

$$\eta_{exergy} = \frac{\varepsilon_{product}}{\varepsilon_{feed}} = \frac{\varepsilon_{ch,gas} + \varepsilon_{ph,gas}}{\varepsilon_{ch,bio\ mass} + \varepsilon_{ph,agent}} \quad (14)$$

where $\varepsilon_{ch,gas}$ is the chemical exergy of gaseous product, $\varepsilon_{ch,biomass}$ is the chemical exergy of biomass, $\varepsilon_{ph,gas}$ is the physical exergy of gaseous product and $\varepsilon_{ph,agent}$ is the exergy of gasification agent.

The effect of different operational parameters on the gasification process efficiency was evaluated. Thus, equilibrium composition of the product gas is obtained by simultaneously solving mass and energy balance equations and equilibrium constant expression of chemical reaction using Mathcad software.

3.2. Model Validation

With the purpose of evaluate the proposed model; the predicted values were compared with experimental data obtained from literature [22]. The statistical parameter, RMSE (root mean square error) was used. This parameters is defined by [23]:

$$RMS = \sqrt{\frac{\sum(X_e - X_p)^2}{N}} \quad (15)$$

where X_e , X_p and N are experimental data, predicted values and observation numbers, respectively. The conditions of the experimental data, obtained of bamboo gasification, were: T = 850°C, ER = 0.3, moisture content = 20%. The bamboo are similar composition and higher heating values than the peach pits, marcs and stalks. The obtained results are shown in Table 6. The achieved RMSE average value was 2.78 when the experimental results are compared with their corresponding theoretical predictions. The predicted values are similar to the experimental data, particularly, the exergetic efficiency values, for all studied different biomass wastes.

Table 6. Syngas composition (dry basis) obtained of bamboo gasification (experimental data) and the predicted values obtained applying the model.

	Peach pit	Grape stalk	Grape marc	Experimental (Dutta et al., 2013)
H ₂ (%)	21.26	22.37	20.34	21.43
CO (%)	22.53	21.60	22.07	24.28
CO ₂ (%)	8.92	13.21	12.78	8.19
CH ₄ (%)	0.41	0.42	0.34	0.78
N ₂ (%)	37.99	42.39	44.47	44.90
RMS	3.21	2.81	2.34	---
η_{exergy}	26.12	30.08	30.30	27.60

4. Results and Discussion

The analyses based on the biomass gasification model are used to perform a parametric study, by varying a number of parameters while holding others fixed values.

4.1. Effect of Gasification Temperature on the Exergetic Efficiency of Process

The gasification process was studied between 450-900°C. The temperature influence was evaluated taking into account the molar composition of syngas (H₂, CO, CO₂, CH₄, H₂O and N₂) in the temperature range mentioned. The exergy content was measured as the sum of physical and chemical exergies. For all wastes, the produced CH₄ concentration decreases in all temperature range but, the produced H₂ concentration increases around 650°C, and then it diminishes, whereas produced CO content augment since 3% to 13% approximately. The highest exergy efficiency was predicted for the peach pits in all analyzed temperature range. Figure 1 shows the variation of exergetic efficiency with the temperature for the three biomass wastes. In this case, the gasification agent was air (SBR = 0), the moisture content of biomass wastes was equal to 40%. The model provides de required air quantity for each temperature. ER varies between 0.03-0.38 when the temperature increases since 450°C to 900°C.

4.2. Effect of Supplied Air Quantity (ER) on the Exergetic Efficiency of Process

The produced syngas composition varies with the amount of supplied air to the process. The exergetic efficiency decrease with ER in all studied temperature range. These relations are illustrated in Figure 2. The simulation was carries out for a temperature equal to 800°C, moisture content of biomass wastes equal to 40% and air like gasification agent. All the gas constituents except CO₂ decrease with ER. This is due to shifting of the

process more towards combustion at higher ER values. Even though the number of moles of CO₂ increases, there is a decrease in CO₂ mole fraction with ER. This is attributed to the higher rate of increase of total number of moles of gas constituents, due to the addition of N₂, compared to the increase in number of moles of CO₂. With the increase in ER, the temperature of the product gases increases, however, the amount of syngas in the product gases is less than other gases like CO₂ and H₂O.

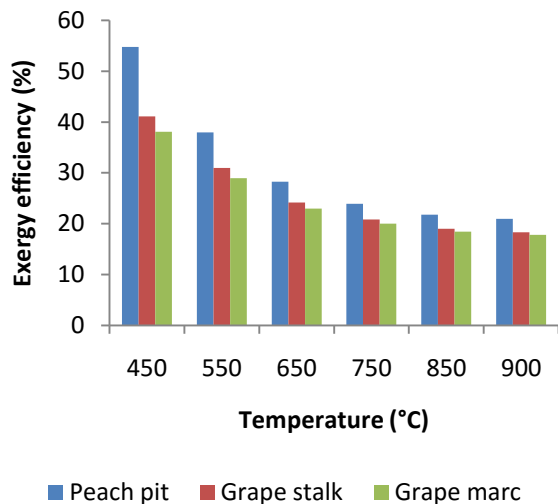


Figure 1. Effect of temperature on the exergetic efficiency of gasification process.

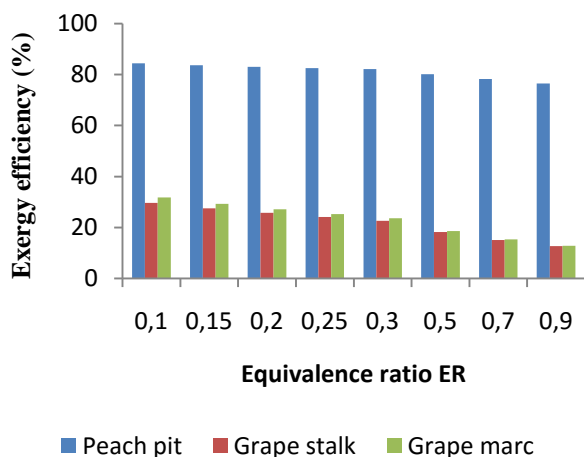


Figure 2. Effect of ER on the exergetic efficiency of gasification process.

4.3. Effect of Moisture Content of the Biomass Wastes on the Exergetic Efficiency of Process

Biomass consists of a significant amount of moisture and high moisture content affects the chemical reactions in the process. To understand how the gasification responds to variations in biomass moisture content, the process is assessed for a range of biomass moisture fractions from the biomass dryer and the results are also shown in Figure 3. In this case, the gasification agent was air (SBR = 0), the temperature was equal to 800°C and ER value equal to 0.3. The effect of moisture content was studied between 0 and 40%. The exergetic efficiency decreases with the moisture content increase. With the increase in moisture content, the rate of CO₂ production increases, which itself is an exothermic process. In steam-biomass gasification, the

liberated heat is consumed by water evaporation rather by the endothermic CO and H₂ reactions. Thus, the CO concentration decrease significantly and the H₂ concentration increase slightly to 30% of moisture approximately, and then it decrease, explaining the exergetic efficiency decreased with biomass wastes moisture. On the other hand, moisture content has a negative effect on the chemical and physical exergy.

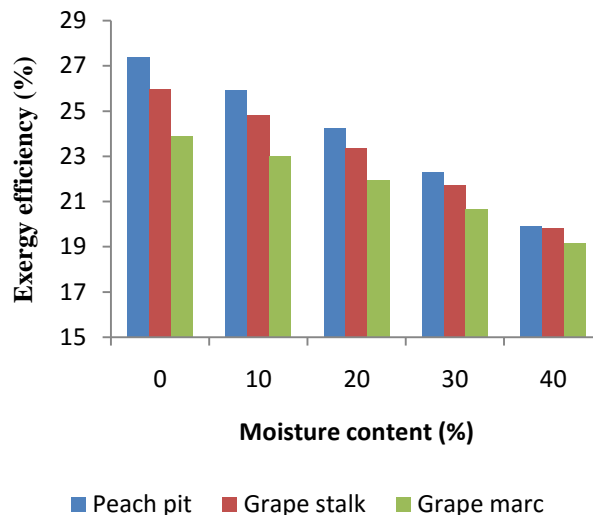


Figure 3. Effect of moisture content of the biomass wastes on the exergetic efficiency of gasification process.

4.4. Effect of the Steam/biomass Waste Ratio (SBR) on the Exergetic Efficiency of Process

The influence of steam addition on product gas composition is depicted through Figure 4.

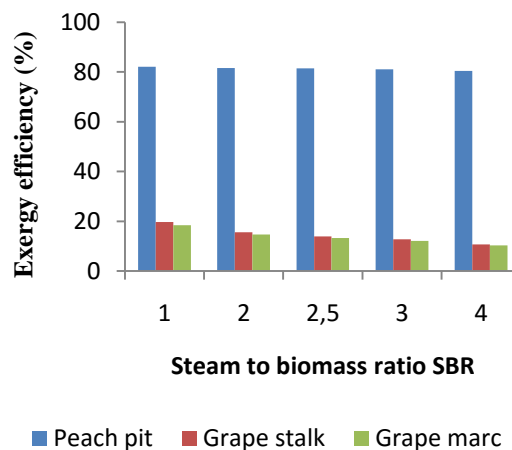


Figure 4. Effect of SBR on the exergetic efficiency of gasification process.

The steam and gasifier temperature were equal to 400°C and 800°C, respectively. Increasing SBR, the H₂ mole fraction also increases slightly (since 31% to 41%) but, CO mole fraction decreases since 40% to 7%, due to the effect of water gas shift reaction. Influence of methanation reaction on gasification process is reflected in the decrease of CH₄ mole fraction with SBR increase. On the other hand, the physical exergy of steam is considered in the denominator of the exergetic efficiency expression definition (Eq. 18). As results, the exergetic efficiency decrease slightly with the SBR increase.

4.5. Effect the Biomass Composition

The influence of the biomass waste composition on the product gas composition can be observed in Figures 1, 2, 3, and 4.

Table 1 shows that the proximate analysis results of biomass wastes. These properties differ substantially in moisture and ash content. Peach pits is a relatively dry feedstock with low ash content. The high moisture contents of grape stalk and marcs decrease the exergetic efficiency due to oxygen is added mainly to generate heat and evaporate moisture present in the fuel. If the exothermic oxidation reactions could drive endothermic gasification reactions, rather than endothermic evaporation of water, the gasification process is much more efficient.

Taking into account the ultimate analysis (Table 2), the peach pits present the lower hydrogen/carbon ratio and moreover, it contains less impurities, particularly N and, S than the marcs and stalks.

Considering the results shown in Table 3, the highest hemicellulose and cellulose contents are present in peach pit. Due to the high content of benzene rings with high stability, high molecular weight compounds are produced during the lignin decomposition, contributing to the char and tar production. Therefore, the winery solid wastes are not suitable to gasify due to the formation of these products [24]. This fact can be verified in all results of proposed model.

5. Conclusions

This process in which biomass wastes are converted into clean and combustible gas can be studied using thermodynamic equilibrium model allowing the predicting of the main product gas compositions CO, CO₂, H₂, CH₄ and N₂, and the efficiency process, which is an important step in modeling the gasification. The model assumes that the principle reactions are at thermodynamic equilibrium. The model are computed using the Mathcad software. The three types of biomass wastes from Argentine are used for prediction of equilibrium gas compositions.

These three samples are compared with results based on percentage mole concentration values that compare favorably well with results obtained from previous studies.

The wastes were peach pits, marcs and stalks, from canning industries and wineries, respectively. It was observed that the energy and exergy of process output was predominantly chemical energy of the fuel. The results show that the largest internal exergy losses (irreversibilities) take place during the studied process.

The exergetic efficiency was evaluated taking into account the operational conditions of process like temperature, wastes moisture, ER and SBR. The exergetic efficiency decreases when all these operation variables increase.

For all wastes, the H₂ concentration contained in syngas, increases around 650°C, and then it diminishes, whereas produced CO content augment since 3% to 13% approximately.

Increasing SBR, the H₂ concentration increases slightly but, CO and CH₄ mole fractions decrease due to the effect of water gas shift reaction and the influence of methanation reaction on gasification process, respectively.

It is shown that exergetic efficiency depends on feedstock composition. It was concluded that gasification of peach pits is very efficient comparing with the gasification of marcs and stalks. Peach pits is a relatively dry feedstock

with low ash content. On the other hand, this agro-industrial waste present the lower hydrogen/carbon ratio and moreover, it contains less impurities, particularly N and, S than the marcs and stalks. On the other hand, it is important to consider that higher lignin content produces greater char amount or carbonaceous residues, because of a lower thermal degradation. This fact can be verified in all results of proposed model.

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