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EVALUATION OF BIOMASS AND COAL CO-GASIFICATION OF BRAZILIAN FEEDSTOCK USING A CHEMICAL EQUILIBRIUM MODEL

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Abstract - Coal and biomass are energy sources with great potential for use in Brazil. Coal-biomass co-gasification enables the combination of the positive characteristics of each fuel, besides leading to a cleaner use of coal. The present study evaluates the potential of co-gasification of binary coal-biomass blends using sources widely available in Brazil. This analysis employs computational simulations using a reliable thermodynamic equilibrium model. Favorable operational conditions at high temperatures are determined in order to obtain gaseous products suitable for energy cogeneration and chemical synthesis. This study shows that blends with biomass ratios of 5% and equivalence ratios ≤ 0.3 lead to high cold gas efficiencies. Suitable gaseous products for chemical synthesis were identified at biomass ratios $\leq 35\%$ and moisture contents $\geq 40\%$. Formation of undesirable nitrogen and sulfur compounds was also analyzed.

Keywords: Coal and biomass energy; Co-gasification; Equilibrium modeling.

INTRODUCTION

Biomass has been used as an energy source since ancient times. It is a renewable resource, available in many forms throughout the world. According to Parikka (2004), the worldwide potential of energy from biomass is of about 103.8 EJ (EJ = 10^{18} J) per year, equivalent to about 5.9 trillion tons of biomass generated annually. In Brazil, the potential of this resource in the national energy matrix recently reached 0.125 EJ, according to a technical report of the Energy Research Company (EPE, 2013), which is equivalent to about 7.1 billion tons of biomass. Coal, a fossil and non-renewable resource, is one of the oldest energy sources used by mankind, and is still widely used worldwide. Global coal reserves are estimated to be 861 billion tons (EIA, 2008), and account for more than 50% of the total energy matrix for some nations, such as China and India (WCA, 2012). In Brazil, coal is a fossil resource with great potential for energy cogeneration, as pointed out by the National Agency of Electrical Energy (ANEEL, 2008).

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Brazil has coal reserves of about 4.6 billion tons (EIA, 2008), corresponding to 1.4% of the national energy matrix (EPE, 2012).

The coal reserves in Brazil are concentrated predominantly in the Southern region of the country, 89.25% of them being found in the state of Rio Grande do Sul (ANEEL, 2008). This coal is predominantly sub-bituminous and has high ash content. Biomass sources are found in all regions of Brazil; however, their use in cogeneration brings technical difficulties related to the seasonal availability of certain types of biomass, their typical high moisture content and low heating value. A technical report of the National Agency of Electrical Energy (ANEEL, 2012) listed eight biomass sources suitable for thermoelectric generation: sugarcane bagasse, black liquor (from cellulose industries), wood residues, biogas, elephant grass, charcoal, rice husk and palm kernel oil. A study by the Brazilian Reference Center on Biomass (Coelho *et al*., 2012) also evaluated the potential of biomass as an energy source in Brazil. This study took into account aspects of geographical distribution and suggested the use of the following sources: sugarcane bagasse, forest residues, agricultural residues, palm oil and biogas from various renewable sources.

Gasification is a technology that allows an efficient utilization of coal and biomass as energy sources. It consists of the thermochemical conversion of carbonaceous materials at high temperatures in the presence of a gasifying agent, usually air and/or steam. The final products are essentially a gaseous mixture with higher heating value known as “syngas” and solid byproducts. The co-processing of coal-biomass blends is called co-gasification.

Co-gasification of coal-biomass blends has been increasingly studied because it leads to a cleaner use of coal. The co-processing of coal-biomass blends can be carried out such that undesirable characteristics of one fuel can be compensated by desirable ones brought by the other. Some authors (Sjöström *et al*., 1999; Hernández *et al*., 2010; Xu *et al*., 2011; Song *et al*., 2013) have observed synergies using some combinations of these fuels. However, other authors (Collot *et al*., 1999; Zhu *et al*., 2008) reported the lack of interactions among fuels. Zhou (2014) presented a review of synergy effects during co-gasification in several experimental studies.

Brar *et al*. (2012) and Emami-Taba *et al*. (2013) presented a comprehensive review of recent progress in coal-biomass co-gasification technologies. Different types of chemical reactors are used in co-gasification such as entrained-flow, fixed-bed and fluidized-bed gasifiers. According to Emami-Taba *et al*. (2013), fluidized-bed gasifiers are the most used ones. Brar *et al*. (2012) pointed out that the operating temperature of gasifiers ranges from 850 to 1000 ºC, with a maximum temperature limit established to avoid ash sintering. The authors also observed that gasifiers operate from atmospheric pressure up to 20 bar. Emami-Taba *et al*. (2013) found that the increase of biomass ratio in the blends favors the formation of CO_2, CO, and hydrocarbons due to the increase in the carbon conversion, resulting in higher biomass reactivity. Concomitantly, H_2 production is disfavored by the increase of the biomass ratio in the blends, due to the greater oxygen content present in biomass. The authors also concluded that the increase of biomass ratio increases the cold gas efficiency.

The investigation of favorable combinations of coal and biomass for co-gasification (from both technical and economic points of view) is the first step towards the design of efficient energy cogeneration and chemical synthesis processes. Experimental analysis would be costly and time-demanding, because of the great number of possible coal-biomass combinations and corresponding relative proportions. The use of theoretical analysis, employing reliable computational simulations, is an alternative and attractive approach for a preliminary screening of the best options.

In the present study, we carried out a theoretical analysis to investigate which combinations of coal and biomass sources widely available in Brazil are the most advantageous for co-gasification, as well as the optimal relative fractions of each fuel. For this purpose, we employed a thermodynamic equilibrium model, a tool widely used to study how particular fuel characteristics affect the composition of generated gaseous products. The following solid biomasses are considered in this study: (1) rice husk (*Oryza* sp.), (2) coconut residues (*Cocos nucifera* L.), (3) elephant grass (*Pennisetum purpureum* Schum.), (4) sawmill wood wastes (*Pinus elliottii* Engelm.), (5) charcoal, (6) sugarcane bagasse and (7) sugarcane straw (*Saccharum* sp.). The co-gasification potential in cogeneration and chemical synthesis is evaluated for varying operating parameters: biomass ratio in the blend (*BR*, %w/w), moisture content (*MC*, %w/w) and amount of air as gasifying agent (equivalence ratio, *ER*), keeping pressure and temperature fixed. We found out that binary coal-biomass blends with biomass ratios (*BR*) higher than 5% lead to more than 80% cold gas efficiency, and that blends with less than 35% *BR* can generate gaseous products suitable for chemical synthesis. The reduction of gaseous emissions that are undesirable from an environmental point of view was also investigated.

THERMODYNAMIC EQUILIBRIUM MODELING

There are two types of phenomenological models for gasification systems, based on either a kinetic approach or an equilibrium approach (Li *et al*., 2001). Kinetic models are able to predict the behavior of gasification systems in more details; however, they require the knowledge of kinetic parameters (ranging from a few to thousands), which are typically obtained experimentally. On the other hand, equilibrium models require a reduced amount of information; however, they are only valid under chemical equilibrium conditions. Equilibrium models are valuable because they can predict the thermodynamic limits of a gasification system (Prins *et al*., 2007). This approach is independent of the gasifier type, being suitable to study the influence of operating parameters on the gaseous product composition of the process (Puig-Arnavat *et al*., 2010). According to Puig-Arnavat *et al*. (2010), the largest discrepancies between predictions of equilibrium models and the corresponding values from actual gasification systems are found under low gasification temperatures. As a result, CO and H_2 contents are typically overestimated and CO_2, CH_4, tar and char contents are underestimated.

There are few references in the literature for kinetic models of co-gasification processes (Usón *et al*., 2004; Chen and Hung, 2013; Mtui, 2013; Xu, 2013). According to Villanueva *et al*. (2008), the use of an equilibrium model is a good approach for entrained-flow gasifiers; for downdraft fixed-bed gasifiers this approach is only valid for high temperatures and large residence times in the reduction zone. However, the authors recommend the use of adapted equilibrium models for updraft fixed-bed and fluidized-bed gasifiers. For a preliminary evaluation of the potential application of different biomass sources in co-gasification, as proposed in this work, an equilibrium approach is appropriate.

The model employed uses an equilibrium approach with a non-stoichiometric formulation (Baratieri *et al*., 2008; Rodrigues *et al*., 2010). This formulation calculates the product composition by performing the total Gibbs free energy minimization of an ideal multi-phase mixture. This equilibrium model allows calculating an equilibrium state with a large number of phases and chemical species. A list of expected species in the product must be established *a priori*. Thus, a large solution space must be used, comprising even rarely detected species (Baratieri *et al*., 2008).

The main assumptions of the model are the following:

1. The feed stream (fuel and gasifying agent) consists of a combination of C, H, O, N, S, Cl, Ar, Si, Ca, Al, Fe, Na, K, Mg, P, and Ti atoms;
2. A multi-phase formulation consists of a two-phase mixture: gas- and solid-phase;
3. The fraction of non-converted solid carbon (char) is represented by graphitic carbon;
4. The process takes place in a perfectly mixed reactor in the steady-state at fixed pressure and temperature.
5. The gasification reaction rates are fast enough and residence time is sufficiently long to reach the equilibrium state.

The first and second assumptions are taken into account by choosing a list of expected chemical species from the NASA thermodynamic database (Gordon and McBride, 1971; Gordon *et al*., 1993). As a result, the solid-phase consists of 163 species and the gas-phase consists of 205 species. However, if only the products with concentrations higher than 1 ppm in the equilibrium state are considered relevant, the model is simplified to a solid-phase consisting of solid carbon and 47 chemical species and a gas-phase consisting of 38 chemical species. The chemical species considered in the model are listed in Table 1. The model was implemented using the Cantera package (Goodwin *et al*., 2013). Cantera is an open-source library of object-oriented functions for solving problems in different areas such as chemical kinetics, thermodynamics and transport processes. The Cantera library has been successfully used in the simulation of coal and biomass thermochemical conversion problems (Baratieri *et al*., 2008; Baggio *et al*., 2009; Caton *et al*., 2010; Messig *et al*., 2010).

The model was validated using experimental data for co-gasification processes from the literature and was compared with predicted values of an equilibrium model from the literature before carrying out the proposed study. A set of recent data for three experimental systems was chosen for model validation since they present detailed information regarding feedstock and product composition under typical operating conditions, and distinct gasifier configurations. More specifically, these systems involve coal-biomass co-gasification in entrained-flow (Hernández *et al*., 2010) and fluidized-bed gasifiers (Li *et al*., 2010; Song *et al*., 2013) in the temperature ranges of 1000 to 1150 ºC, at atmospheric pressure, and using air/steam as gasifying agent.

Table 1: List of chemical species considered in the model.

Phase	Group	Chemical species
Gas	Carbon compounds	C(g), CH_4, CO, CO_2
	Oxygen compounds	O, O_2
	Hydrogen compounds	H, H_2, OH, H_2O
	Nitrogen compounds	N, N_2, NH_3, NO, HCN
	Sulfur compounds	S, S_2, SH, SO, SO_2, SO_3, H_2S, COS, CS_2
	Chlorine compounds	Cl, Cl_2, HCl, ClO, SCl_2
	Phosphorus compounds	P_2, PH_3, PO, PO_2, P_2O_5, P_3O_6, P_4O_{10}, PCl_3
	Other compound	Ar
Solid	Carbon	C(s)
	Aluminium compounds	Al_2O_3, AlN, $AlCl_3$, Al_2SiO_5, $Al_6Si_2O_{13}$
	Calcium compounds	Ca, CaO, CaO_2H_2, CaS, $CaSO_4$, $CaCl_2$
	Iron compounds	Fe, FeO, Fe_2O_3, Fe_3O_4, FeS, $FeCl_2$
	Potassium compounds	K, K_2O, KCN, K_2CO_3, K_2S, K_2SO_4, KCl
	Magnesium compounds	MgO, MgS, $MgSO_4$,$MgCl_2$, $MgAl_2O_4$, $MgSiO_3$, Mg_2SiO_4
	Sodium compounds	Na_2O, Na_2CO_3, NaCN, Na_2SO_4, NaCl, $NaAlO_2$
	Silicon compounds	Si, SiC, SiO_2, Si_3N_4, Si_2N_2O
	Titanium compounds	TiC, TiO_2, Ti_2O_3, TiN, $TiCl_3$

RESULTS AND DISCUSSION

Model Validation

Figure 1 shows the deviation between estimated and experimental values from different sources (Hernández *et al*., 2010; Li *et al*., 2010; Song *et al*., 2013) for temperatures from 1000 to 1150 ºC and atmospheric pressure. This parameter is defined as:

$$\text{deviation (\%)} = \frac{1}{N} \times \sum_{i=1}^{N} \left(\frac{x_{i,exp} - x_{i,est}}{x_{i,exp}} \right) \times 100\% \qquad (1)$$

where N is the number of chemical species and $x_{i,est}$ and $x_{i,exp}$ are the estimated and measured volumetric fractions of species i respectively. The total composition of fuel and gasifying agent are expressed in terms of oxygen/carbon (O/C) and hydrogen/carbon (H/C) ratios. The model was considered satisfactory when the deviation between model predictions and experimental values as calculated by equation 1 was smaller than 35%; Esmaili *et al*. (2013) consider deviations of up to 40% appropriate for predictions using an adapted equilibrium model. According to this criterion, our calculations showed that the proposed model was considered satisfactorily accurate for temperatures from 1000 to 1150 ºC and a wide range of O/C and H/C values, as seen in Figure 1. The larger deviations (> 40%) from chemical equilibrium were observed for the data of Hernández *et al*. (2010) due to the short residence time (~1.4 s) in

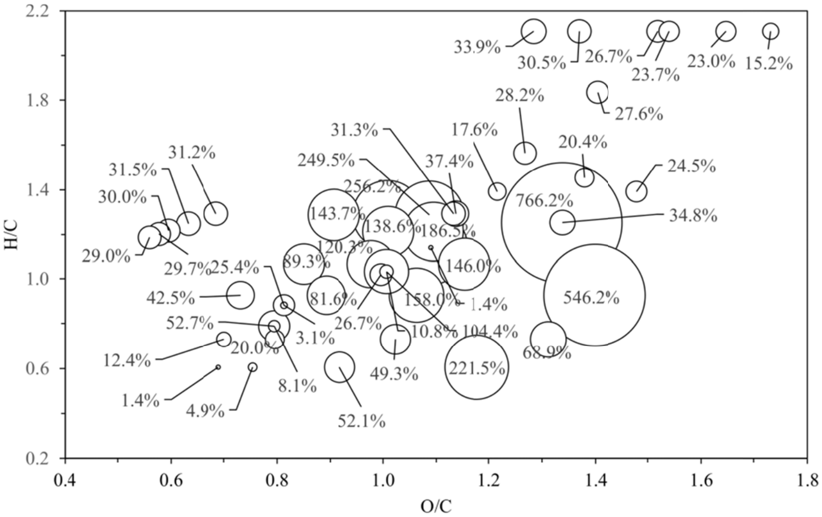

Figure 1: Deviations between experimental data (Hernández *et al*., 2010; Li *et al*., 2010; Song *et al*., 2013) and values predicted by the model for temperatures from 1000 to 1150 ºC. Feed stream compositions are represented by H/C and O/C ratios.

the entrained-flow gasifier compared to the fluidized-bed gasifiers in the Li *et al*. (2010) and Song *et al*. (2013) studies. For same set of Hernández' data it was observed that higher *BR* blends produced gaseous products closer to chemical equilibrium due to the larger volatile matter content of the biomass (four times larger than coal) that increases the feedstock reactivity. The deviations for the remaining studies (Li *et al*., 2010; Song *et al*., 2013) were smaller than 35%. It should be emphasized that, although experiments at temperatures higher than 1200 ºC are not considered here, the proposed equilibrium model is also expected to be valid for temperatures higher than 1150 ºC (as expected for any equilibrium model).

The results of the proposed model were also compared to predictions of the multi-phase equilibrium model of Li *et al*. (2004). Figure 2 shows the equilibrium composition for a proposed model of 7 major species at 1.013 bar and at two distinct temperatures (1000 K and 1100 K), in the range $0 < ER < 1$. The results represent the gasification of a representative lignocellulosic biomass of 50.9% C, 6.6% H, 40.5% O, 0.51% N, 0.34% S (%wt, daf), 1.1% ash (%wt, db), 15% moisture (%wt, wb), and 20.6 MJ/kg of *HHV*. Figure 2 can be compared to Figure 13 obtained by Li *et al*. (2004).

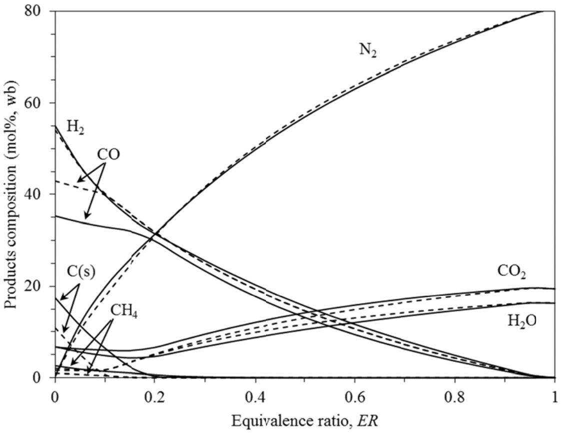

Figure 2: Composition of major gaseous and non-converted carbon for representative biomass (Li *et al*., 2004) at 1.013 bar in the range of $0 \leq ER \leq 1$. Solid lines (—) represent products at 1000 K and dashed lines (---) represent products at 1100 K.

The overall trend of the product composition predicted by the proposed model is similar to that presented in Li *et al*. (2004). The highest deviations of the results are obtained for non-converted solid carbon C(s) contents. Provided that the disappearance of non-converted carbon C(s) leads to the appearance of gaseous species, some deviations of carbon-based gaseous species are also observed. The reason of the deviations of C(s) predictions may be attributed to the fact that Li *et al*. (2004) assumed the ash content to be inert, since the authors studied only fuels with low ash content. The present model assumes the ash content to be part of system, so the ash species are allowed to interact with carbon-based species.

Sensitivity Analysis of the Model

Before starting the analysis of coal-biomass blends, simulations for coal gasification were carried out in order to evaluate the sensitivity of the syngas composition and cold gas efficiency (*CGE*) predicted by the model with respect to temperature, equivalence ratio (*ER*) and moisture content (*MC*). *CGE* and *ER* are defined as:

$$CGE = \frac{LHV_{syngas} \cdot Y}{(1 - MC) \cdot LHV_{fuel}} \times 100\% \qquad (2)$$

$$ER = \frac{n_{O_2}}{n_{O_2,stoic}} \qquad (3)$$

where n_{O_2} is the number of moles of O_2 injected into reactor, $n_{O_2,stoic}$ is the number of moles of stoichiometric O_2, *MC* is the moisture content, *LHV* is the lower heating value, *Y* is the syngas yield and 1 kg of fuel (coal or coal-biomass blends) is assumed as the basis.

The composition of gaseous products and non-converted carbon resulting from coal gasification at 1000 ºC, 1 atm, 10% *MC* and $0 \leq ER \leq 1$ is shown in Figure 3. These results show that the increase of the equivalence ratio increases N_2, CO_2 and H_2O production. This effect is observed because a larger amount of O atoms in the system displaces the chemical equilibria in the oxidation of C(s), CH_4, CO and H_2 towards CO_2 and H_2O. As a result, the variation of cold gas efficiency (*CGE*) reaches a maximum value of approximately 87.5% at $ER = 0.09$ at the point where the C(s), CO, H_2 and CH_4 amounts are decreased due to the increase of CO_2 and H_2O amounts. In those conditions the composition of the ash content is 69.6% SiO_2, 8.3% FeS, 7% Al_2O_3, 6.7% Al_2SiO_5, 1.9% $CaCl_2$, 1.9% KCl, 1.3% $MgSiO_3$, 0.9% CaS and remaining species in minor concentrations.

Figure 4 shows the gaseous products and non-converted solid carbon composition on a dry basis obtained for $0 \leq MC \leq 50\%$ and $ER = 0.1$, keeping constant the same values for the other operating parameters used in the previous analysis. In this case, it is possible to observe that an increase of the moisture content leads to H_2, CO and CH_4 production; however, C(s) and CH_4 is consumed due to the reforming reactions (R1–R4 in Table 2), vanishing after *MC* ≈ 15%. A progressive CO consumption is also observed due to the occurrence of the water-gas shift reaction (R3 in Table 2) after *MC* ≈ 10%, which favors CO_2 and H_2 production.

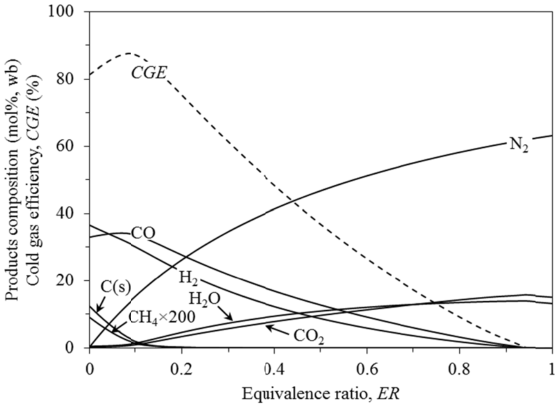

Figure 3: Composition of gaseous products and non-converted carbon and cold gas efficiency for coal gasification at 10% *MC* and 1000 ºC in the range 0 ≤ *ER* ≤ 1.

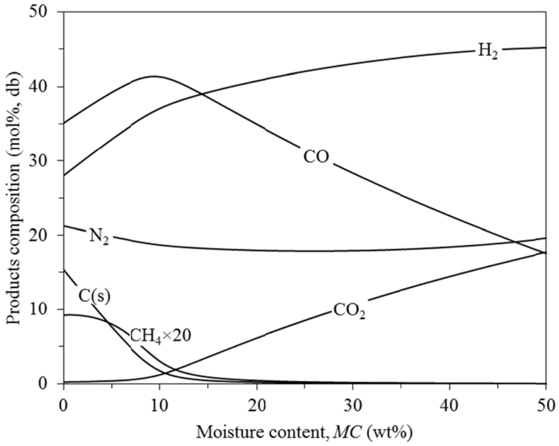

Figure 4: Composition of gaseous products and non-converted carbon for coal gasification at *ER* = 0.1 and 1000 °C in the range 0 ≤ *MC* ≤ 50%.

Table 2: List of chemical equilibrium reactions considered for sensitivity analysis of the model.

Equilibrium reaction	ΔH_r (MJ/kmol)	Equation
$CH_4 + H_2O \leftrightarrow CO + 3H_2$	+ 206.0	(R1)
$CH_4 + CO_2 \leftrightarrow 2CO + 2H_2$	+ 247.4	(R2)
$CO + H_2O \leftrightarrow CO_2 + H_2$	– 41.0	(R3)
$C(s) + H_2O \leftrightarrow CO + H_2$	+ 131.4	(R4)
$C(s) + 2H_2O \leftrightarrow CO_2 + 2H_2$	+ 90.1	(R5)

The compositions of gaseous products and non-converted carbon achieved at *ER* = 0.1 and temperatures from 700 to 1200 ºC, keeping the same values for the other operating parameters used in the previous analyses, are shown in Figure 5. The increase of temperature promotes endothermic reactions: water-gas reactions (R4–R5 in Table 2) and reforming reactions (R1–R2 in Table 2), which favor C(s), CH_4, H_2O and CO_2 conversion to CO and H_2.

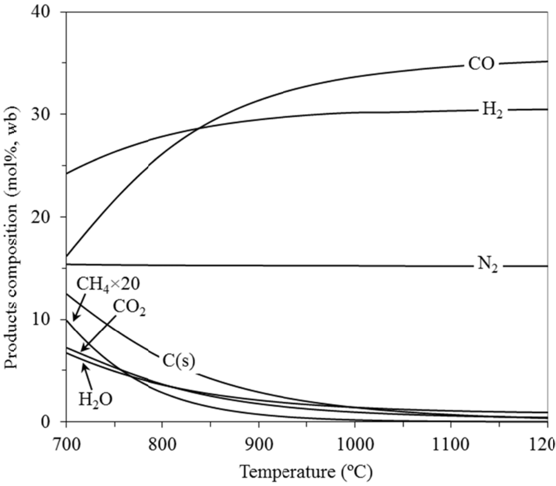

Figure 5: Composition of gaseous products and non-converted carbon for coal gasification at 10% *MC* and *ER* = 0.1 in the range 700 ≤ *T* ≤ 1200 ºC.

The results presented in this section are in agreement with results presented in the literature for similar gasification processes (Esmaili *et al.*, 2013; Shabbar and Janajreh, 2013).

Analysis of Coal-Biomass Co-Gasification

The analysis of the co-gasification of coal-biomass blends employing seven types of solid biomasses (rice husk, coconut residues, elephant grass, sawmill wood wastes, charcoal, sugarcane bagasse and sugarcane straw) was carried out within the validity range of the proposed model established above. The proximate and ultimate analyses for coal and biomass sources available in Brazil are presented in Table 3. The ash composition is given in Table 4.

The co-processing of coal-biomass blends can be advantageous when undesirable characteristics of coal gasification are eliminated or minimized by desirable characteristics of biomass gasification, by using an appropriate biomass ratio (*BR*) in the blend. The undesirable characteristics of the coal considered in this study are the high ash (42.71%) and sulfur (2.15%) contents, and low carbon content (33.39%). The use

Table 3: Characterization of the fuels considered in this study.

	Coal[1]	Rice husk[2]	Coconut residue[3]	Sawmill wood waste[4]	Charcoal[5]	Sugarcane bagasse[6]	Sugarcane straw[6]	Elephant grass[7]
Proximate analysis (%wt, db)								
Moisture (wb	11.06	12.00	83.74	11.78	7.14	50.20	2940	9.97
Volatile matter	20.29	67.80	10.20	86.44	N/A	79.90	83.30	68.93
Fixed carbon	25.09	13.60	79.70	13.22	N/A	18.00	12.80	7.95
Ash	42.71	18.60	10.30	0.35	2.43	2.20	3.90	12.84
Ultimate analysis (%wt, daf)								
C	33.39	38.30	48.23	51.46	65.19	44.60	46.20	46.52
H	3.47	4.00	5.23	6.10	5.55	5.80	6.20	5.87
N	0.61	0.50	2.98	0.26	0.83	0.60	0.50	1.47
O	16.68	38.60	33.19	41.85	25.65	44.50	43.00	46.04
S	2.15	0.08*	0.12	0.01*	0.37	0.10	0.10	0.10
Cl	0.013	0.12*	N/A	0.01*	N/A	0.02	0.10	0.21*
HHV (MJ/kg, b)	13.39	15.49	22.81	20.25	25.28	18.10	17.40	16.94

wt = weight, db = dry basis, wb = wet basis, daf = dry ash free basis, N/A = not available.

[1]Kalkreuth *et al*. (2006), [2]CIENTEC (1986) *apud* Hoffmann (1999), [3]Andrade *et al*. (2004) and Nogueira *et al*. (2000) *apud* Andrade *et al*. (2004), [4]Wander *et al*. (2004), [5]Fuwape (1993), [6]Linero and Lamônica (2005) *apud* Seabra *et al*. (2010), [7]Broetto *et al*. (2012)

*Mean value from Vassilev *et al*. (2010).

Table 4: Ash composition of the fuels considered in this study.

	Ash composition (%wt, db)									
Fuel	**SiO_2**	**CaO**	**Al_2O_3**	**Fe_2O_3**	**Na_2O**	**K_2O**	**MgO**	**P_2O_5**	**TiO_2**	**SO_3**
Coal[1]	29.04	1.02	9.02	4.21	0.16	0.70	0.41	0.03	0.36	2.28
Rice husk[2]	94.48	0.97	0.21	0.22	0.16	2.29	0.19	0.54	0.02	0.92
Coconut residue[3]	29.14	25.99	4.49	5.60	2.54	19.40	5.60	5.92	0.24	3.27
Sawmill wood waste[4]	22.22	43.03	5.09	3.44	2.85	10.75	6.07	3.48	0.29	2.78
Charcoal[4]	22.22	43.03	5.09	3.44	2.85	10.75	6.07	3.48	0.29	2.78
Sugar cane bagasse[5]	46.79	4.91	14.60	11.12	1.61	6.95	4.56	3.87	2.02	3.57
Sugar cane straw[6]	43.94	14.13	2.71	1.42	1.35	24.49	4.66	4.13	0.16	3.01
Elephant grass[7]	46.18	11.23	1.39	0.98	1.25	24.59	4.02	6.62	0.08	3.66

wt = weight, db = dry basis.

[1]Kalkreuth *et al*. (2006), [2]Mean value from five sources (Vassilev *et al*., 2010), [3]Mean value for natural biomass (Vassilev *et al*., 2010), [4]Mean value for woody biomass (Vassilev *et al*., 2010), [5]Mean value from two sources (Vassilev *et al*., 2010), [6]Mean value for straws (Vassilev *et al*., 2010), [7]Mean value for grasses (Vassilev *et al*., 2010).

of a coal-biomass blend can properly raise or reduce these contents; also, the high oxygen content in biomass (25.65 to 46.04%) can favor gasification reactions without injecting additional oxygen into the gasifier. Some biomasses also have lower nitrogen content (0.26 to 0.5%) compared to coal (0.61%), which decreases the production of nitrogen-containing compounds (NH_3), minimizing the environmental impact caused by the emissions.

Gas Composition and Carbon Conversion

The composition of the gaseous products from gasification of pure coal and different types of biomasses (each one in pure form) were computed for 1000 ºC, 0% *MC*, $ER = 0.4$ and $0 \leq BR \leq 100\%$. The results are summarized in Table 5. Then, the product composition resulting from coal-biomass co-gasification under different biomass ratios was calculated using the same set of parameters (1000 ºC, 0% *MC* and $ER = 0.4$). Results for wood waste are shown in Figure 6. We observed that the product composition estimated for gasification of a coal-biomass blend is practically identical to the mass average of the values estimated for gasification of each individual fuel; deviations were smaller than 1.5%, which can be attributed to numerical errors in calculations. In other words, the product composition varied linearly with *BR*. We observed an analogous behavior for the other computed properties, such as gas yield, heating value of gaseous products and cold gas efficiency. Consequently, the gaseous product composition and the other aforementioned parameters from coal-biomass co-gasification can be estimated from mass-averaged values of the corresponding parameters obtained for gasification of pure coal and biomass. In the following paragraphs, we discuss the influence of the biomass ratio on the operational parameters of interest directly from the values obtained for the gasification of pure fuels, given in Table 5.

Table 5: Results for dry coal and biomass gasification at 1000 ºC, 1 atm and *ER*=0.4.

	Gas composition (%vol, wb)							Gas yield (Nm^3/kg, db)	*HHV* (MJ/kg, db)	*CC* (%)	*CGE* (%)
	H_2	CO	CH_4 (ppm)	CO_2	H_2O	H_2S (ppm)	NH_3 (ppm)				
High ash coal	10.6	21.5	0.77	5.51	4.50	1690	3.53	0.66	4.31	99.8	44.0
Rice husk	12.2	23.1	0.67	8.64	7.55	86.8	4.07	0.96	6.26	99.9	53.4
Coconut residue	13.8	24.1	1.26	6.03	5.70	31.2	5.00	1.34	9.12	100	55.9
Wood waste	14.6	23.6	1.18	6.77	6.91	0.0002	5.31	1.44	9.37	100	66.1
Charcoal	13.3	28.1	2.87	3.29	2.58	187	4.85	1.76	13.7	100	75.9
Cane bagasse	14.5	22.0	0.84	8.15	8.88	52.2	5.17	1.27	7.79	100	60.7
Cane straw	14.9	21.8	0.96	7.54	8.55	37	5.45	1.32	8.03	100	66.8
Elephant grass	13.7	21.5	0.73	8.35	8.79	7.06	4.80	1.12	6.84	100	56.5

vol = volume, wb = wet basis, db = dry basis, *HHV* = higher heating value, *CC* = carbon conversion, *CGE* = cold gas efficiency.

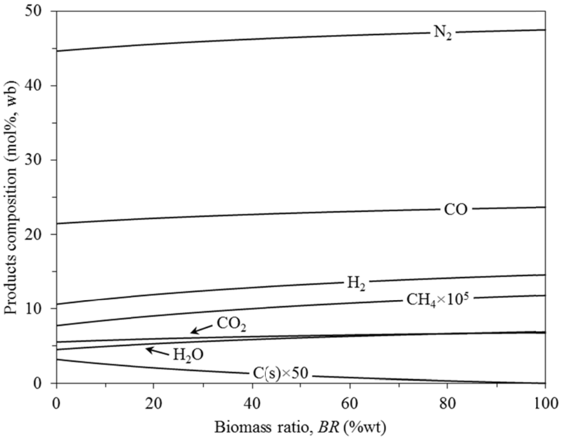

Figure 6: Composition of gaseous products and non-converted carbon for dry coal-wood co-gasification at *ER* = 0.4 and 1000 ºC in the range 0 ≤ *BR* ≤ 100%.

For every coal-biomass blend tested for the parameters specified in the previous paragraph, we observed that the increase of *BR* leads to larger H_2 production due to the higher H content in biomass for a given *ER* (i.e., total O content remains constant). An increase of *BR* increases CO production for all the blends. CH_4 production usually increases with the increase of *BR*, except for rice husk and elephant grass blends. Charcoal blend exhibited an interesting feature: the increase of *BR* reduces CO_2 and H_2O production; consequently, it is possible to obtain gaseous products with higher heating value from these blends compared to others. The temperature and *ER* conditions used in these calculations led to a 100% carbon conversion (*CC*) for most of blends, corresponding to a full conversion of solid carbon to gaseous products.

The main results for coal and wood gasification at 1000 ºC, 1 atm pressure, and values of *ER* = 0.1, 0.2, 0.4 and 0.6, and *MC* = 0, 10, 20 and 40% are shown in Table 6. The increase of *ER* leads to a higher conversion of H_2, CO and CH_4 to H_2O and CO_2 as expected. The moisture content has little influence on H_2 production for *MC* < 40%. However, the increase of *MC* reduces CH_4 production. At 40% *MC*, the H_2/CO ratio is larger than 1.0, which indicates higher H_2 over CO content in gaseous products. At low equivalence ratio (*ER* = 0.1), H_2 contents higher than 31.4% are achieved for 15% *MC* and 5% *BR* except for the charcoal blend that requires higher moisture contents. CO contents higher than 34.2% are obtained for 10% *MC* and 5% *BR* for all blends. The H_2/CO ratio equal to 2.0 (recommended value for chemical synthesis) is obtained for values of 5 ≤ *BR* ≤ 25%, 45 ≤ *MC* ≤ 50%, and 0.1 ≤ *ER* ≤ 0.6 for most of blends. CH_4 contents higher than 0.03% can be achieved using *ER* = 0.1, 0% *MC* and 5% *BR* for all blends.

Gas Yield, Heating Value and Cold Gas Efficiency

The gas yield (*Y*) is directly related to the conversion of solid carbon to gaseous products. The blends investigated here showed that the increase of *BR* favors gas yield, as observed experimentally (Emami-Taba *et al*., 2013). The gas yield can exceed 0.66 Nm^3 N_2-free gas per kg of coal, d.b., up to 1.76 Nm^3/kg for charcoal blends as shown in Table 5. In general, the gas yield increases with the increase of *BR* due to the higher content of H and O in biomass, which increases the carbon conversion. In addition, the increase of *BR* decreases the content of ash in the blend, which increases the amount of solid available for gas production.

The highest heating values of gaseous products were observed for charcoal, 13.7 MJ/kg, and wood, 9.37 MJ/kg as shown in Table 5. The heating value of the gaseous product is directly related to its composition. However, the higher content of ash led to lower heating values. This effect can be seen for cane bagasse (7.79 MJ/kg) and elephant grass (6.84 MJ/kg), for example. These biomass sources led to

Table 6: Results for coal and wood gasification at 1000 ºC and 1 atm.

	MC (%wt)	*ER*	Gas composition (%vol, wb)							*CC* (%)	Gas yield (Nm^3/kg, db)	*HHV* (MJ/kg, db)	*CGE* (%)
			H_2	CO	CH_4 (ppm)	CO_2	H_2O	H_2S (ppm)	NH_3 (ppm)				
High ash coal	0	0.1	22.4	28.1	296	0.17	0.23	2920	8.33	69.6	0.59	3.76	50.2
		0.2	19.0	31.1	85.6	0.39	0.39	2340	7.34	90.4	0.68	4.85	58.4
		0.6	4.92	11.6	0.02	10.5	7.39	1320	1.21	100.0	0.59	2.85	24.9
	10	0.2	22.8	27.7	10.1	3.39	4.63	2070	8.71	99.3	0.79	4.33	75.3
		0.4	12.3	16.9	0.45	7.83	9.43	1570	4.21	99.9	0.70	3.34	48.4
		0.6	5.89	9.16	0.02	11.3	12.1	1260	1.53	100.0	0.62	2.19	26.5
	20	0.2	23.4	21.1	3.11	6.47	11.9	1830	8.46	99.8	0.82	3.32	82.9
		0.4	12.8	12.8	0.23	9.42	15.6	1430	4.27	100.0	0.71	2.51	51.5
		0.6	6.13	6.81	0.01	11.9	17.7	1170	1.56	100.0	0.61	1.59	26.7
	40	0.2	20.5	10.8	0.41	9.41	29.5	1370	6.17	100.0	0.78	1.74	95.4
		0.4	10.9	6.26	0.03	10.6	30.7	1130	3.07	100.0	0.65	1.21	52.7
		0.6	4.42	2.74	0.001	11.7	31.2	95	0.88	100.0	0.53	0.62	20.8
Wood waste	0	0.1	33.2	38.7	1130	0.12	0.17	1.62×10^{-4}	12.6	81.7	1.46	9.82	90.5
		0.2	27.6	39.4	13.8	0.58	0.67	1.42×10^{-4}	11.2	100.0	1.63	11.9	100.0
		0.6	7.23	13.3	0.06	11.2	10.1	1.89×10^{-4}	2.04	100.0	1.27	6.54	38.8
	10	0.2	28.2	34.8	22.6	2.88	3.86	1.94×10^{-4}	11.3	100.0	1.67	10.0	104.1
		0.4	15.2	21.0	0.86	7.97	9.51	2.17×10^{-4}	5.53	100.0	1.47	7.87	68.2
		0.6	7.59	11.8	0.05	11.7	12.4	2.26×10^{-4}	2.16	100.0	1.28	5.45	39.4
	20	0.2	28.1	29.9	9.39	5.09	7.93	2.56×10^{-4}	11.0	100.0	1.67	8.24	105.4
		0.4	15.2	18.0	0.55	9.18	12.8	2.68×10^{-4}	5.46	100.0	1.45	6.39	67.5
		0.6	7.51	9.93	0.03	12.3	15.4	2.79×10^{-4}	2.10	100.0	1.26	4.31	37.6
	40	0.2	25.7	19.7	1.98	8.77	19.0	4.12×10^{-4}	9.30	100.0	1.54	5.01	97.5
		0.4	13.4	11.3	0.14	11.2	21.9	3.98×10^{-4}	4.37	100.0	1.30	3.63	56.5
		0.6	5.73	5.35	0.005	13.2	23.4	3.84×10^{-4}	1.36	100.0	1.08	2.09	25.6

wt = weight, vol = volume, wb = wet basis, db = dry basis, *HHV* = higher heating value, *CGE* = cold gas efficiency.

similar gaseous products composition, but elephant grass has about 6 times higher ash content than cane bagasse. The cold gas efficiency (*CGE*) relates the heating value of the gaseous products with the heating value of solid fuel. The coal gasification reached a *CGE* of 44.0% for the operating conditions studied here. The co-gasification of coal-biomass blends could attain a *CGE* of 75.9% by using charcoal blends.

The increase of *ER* led to the decrease of gas yield, heating value and cold gas efficiency, as shown in Table 6. Actually, higher gas yield could be observed at lower *ER* due to non-converted solid carbon. At higher *ER*, the remaining air amount decreases the yield of useful gas (H_2O and N_2-free basis) as soon as solid carbon is converted to gaseous products. The same trend was observed with the increase of *MC*; however, the highest values of those parameters were observed at $10 \leq MC \leq 20\%$. These trends were also observed for the other blends. The coal-biomass co-gasification allows obtaining values of *CGE* higher than 80% with at least 5% *BR*, $MC \geq 5\%$ and $0.1 \leq ER \leq 0.4$ for most of cases.

Nitrogen and Sulfur Compounds

The evaluation of nitrogen and sulfur compound production (NH_3 and H_2S) is important when the gaseous products are applied in chemical synthesis, fuel cells, and energy cogeneration. There are tolerance limits for NH_3 and H_2S contents according to the application. According to Emami-Taba *et al.* (2013), NH_3 and H_2S contents must be smaller than 1 ppm (10^{-4} %vol) in chemical synthesis, and sulfur release must be close to zero in fuel cells. H_2S content should be smaller than 47 ppm and NH_3 smaller than 65 ppm for use in energy cogeneration.

Table 5 shows that coal gasification generates gaseous products that meet the limits for NH_3 content (3.53 ppm) but not for H_2S content (1690 ppm) in energy cogeneration. The appropriate H_2S content can be achieved using blends of coconut residue (31.2 ppm), wood waste (0.2 ppb), sugarcane straw (37.0 ppm) or elephant grass (7.06 ppm). This happens due to the lower content of S in such biomasses compared to coal. However, some types of biomasses with lower content of S have greater H_2S production because of the type of ash composition that may support H_2S release.

The content of H_2S in the products of dry coal-wood blends co-gasification at 1 atm is shown in Figures 7 and 8. Figure 7 shows the resulting content of H_2S at $ER = 0.4$ for $75 \leq BR \leq 100\%$ and temperatures of 900, 1000, 1100 and 1200 ºC. Moreover, Figure 8 shows the variation of H_2S at 1000 °C for $0 \leq ER \leq 1$, and 75, 80, 85, 90, and 95% *BR*. The increase of *BR* led to the decrease of H_2S content in the gaseous product. Lower temperature and larger *ER* also contributed to the decrease of H_2S formation. These trends have been observed experimentally in the literature (Emami-Taba *et al*., 2013). At a value of *ER* approximately equal to 0.2, H_2S consumption surpasses its production, which can be explained by the conversion of solid-S (mainly FeS and CaS) in ash contents to gaseous-S in gaseous products. The entire consumption of H_2S takes place at *ER* = 1.0 due to the oxidation of H_2S to SO_2. The variation of *MC* revealed a minor influence on H_2S formation.

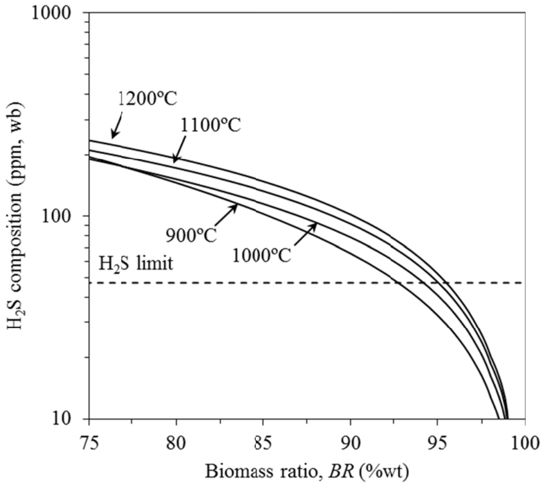

Figure 7: H_2S content for dry coal-wood co-gasification in the range of $75 \leq BR \leq 100\%$ and $900 \leq T \leq 1200$ °C at *ER* = 0.4. Dashed line (---) indicates the H_2S limit for cogeneration.

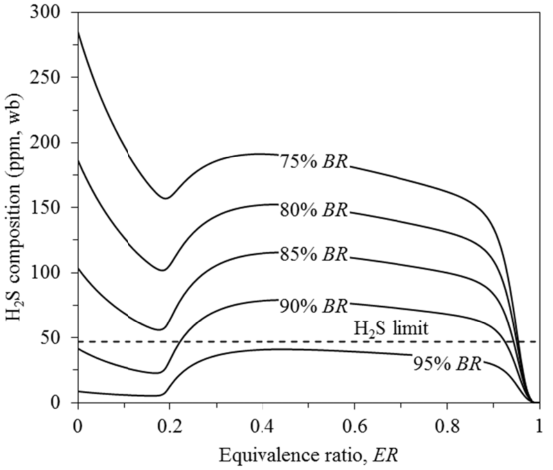

Figure 8: H_2S contents for dry coal-wood co-gasification in the ranges $0 \leq ER \leq 1$ and $75 \leq BR \leq 95\%$ at 1000 ºC. Dashed line (---) indicates the H_2S limit for cogeneration.

The appropriate H_2S contents could be achieved with at least 95% *BR* for co-gasification of wood blends at *ER* = 0.4 and 1000 ºC. In the case of coconut blends, these conditions were achieved with at least 90% *BR*, 99% *BR* of sugarcane straw and 69% *BR* of elephant grass also achieved this conditions. Higher values of *ER* decrease both H_2S and NH_3 production. There is an increase of H_2S and a decrease of NH_3 formation at higher values of *MC* as shown in Table 6. The recommended H_2S and NH_3 limiting values for chemical synthesis were not observed for most of the studied cases.

Summary of Results by Evaluation of Scenarios

This section summarizes the results of the analysis of coal-biomass co-gasification for energy cogeneration and chemical synthesis purposes. Table 7 presents a summary of guidelines to help in the selection of binary blends of coal and biomass. For each coal-biomass blend, there are two scenarios corresponding to the utilization of gaseous products for cogeneration and chemical synthesis purposes. Scenario (I) considers the recommended biomass ratio in blends to reach a cold gas efficiency higher than 80% for cogeneration, and H_2/CO ratio equal to 2.0 ± 5% for chemical synthesis. Scenario (II) considers the same conditions of scenario (I) and appropriate H_2S and NH_3 contents, i.e., scenario (II) shows an environmental friendly scenario. Both scenarios are also shown for pure coal, to demonstrate the benefits of using coal-biomass blends instead of pure coal.

The results show that all coal-biomass blends can be used for cogeneration purposes for any values of biomass ratio and moisture content and low values of equivalence ratio. The recommended H_2S and NH_3 limiting values were not observed for pure coal when applied to cogeneration purposes. Rice husk, wood and sugarcane bagasse blends achieved the appropriate H_2S and NH_3 limiting values for $BR \geq 95\%$. Coconut and sugarcane straw blends achieved the appropriate H_2S and NH_3 limiting values for $BR \geq 75\%$. Elephant grass blends achieved the appropriate H_2S and NH_3 limiting values for $BR \geq 55\%$. The recommended H_2S and NH_3 limiting values were not observed for charcoal blends. All coal-biomass blends can also be suitable for chemical synthesis purposes for $BR \leq 35\%$ and higher moisture contents. Equivalence ratio did not have a significant effect on the H_2/CO ratio; however, higher values of ER would lead to lower content of H_2 and CO that could be inconvenient for chemical synthesis purposes. Rice husk, coconut, wood, sugarcane bagasse, sugarcane straw and elephant grass blends achieved the recommended H_2S

Table 7: Guidelines for selection of binary coal-biomass blends.

Blend	**Scenario***	**Cogeneration purposes**			**Chemical synthesis purposes**		
		***BR* (%)**	***MC* (%)**	***ER***	***BR* (%)**	***MC* (%)**	***ER***
Coal	(I)	N/A	10–50	≤ 0.25	N/A	40–50	—
	(II)	Not found			N/A	50	≥ 0.75
Coal-rice husk	(I)	—	—	≤ 0.35	—	40–50	—
	(II)	≥ 95	≤ 5	≤ 0.1	≤ 20	50	≥ 0.75
Coal-coconut	(I)	—	—	≤ 0.4	≤ 65	40–50	—
	(II)	≥ 70	5–30	≤ 0.3	≤ 10	50	≥ 0.75
Coal-wood	(I)	—	—	≤ 0.3	≤ 60	40–50	—
	(II)	≥ 90	≤ 25	≤ 0.3	≤ 10	50	≥ 0.75
Coal-charcoal	(I)	—	—	≤ 0.5	≤ 35	40–50	—
	(II)	Not found			≤ 5	50	≥ 0.75
Coal-cane bagasse	(I)	—	—	≤ 0.4	≤ 85	40–50	—
	(II)	≥ 95	≤ 10	≤ 0.1	≤ 10	50	≥ 0.75
Coal-cane straw	(I)	—	—	≤ 0.45	≤ 90	40–50	—
	(II)	≥ 75	≤ 20	≤ 0.3	≤ 10	50	≥ 0.75
Coal-elephant grass	(I)	—	—	≤ 0.4	—	40–50	—
	(II)	≥ 55	—	≤ 0.4	≤ 15	50	≥ 0.75

N/A = not applicable, —(dash) = any value.

[*] Scenario (I) considers the recommended *BR* to reach *CGE* ≥ 80% for cogeneration purposes and $H_2/CO = 2.0 \pm 5\%$ for chemical synthesis purposes. Scenario (II) includes scenario (I) and appropriate H_2S and NH_3 contents.

and NH_3 limiting values for $BR \leq 10\%$, ~50% *MC*, and equivalence ratio values higher than 0.75. Charcoal blends achieved the recommended H_2S and NH_3 limiting values for $BR \leq 5\%$.We can point out that higher moisture contents in the analysis may be assumed as a combination of moisture and steam amounts in the feed stream as long as the simulations do not include the initial state of H_2O as liquid (moisture) or vapor (steam).

CONCLUSIONS

This study evaluated the potential of co-gasification of coal-biomass blends from sources available in Brazil for energy cogeneration and chemical synthesis purposes. A thermodynamic equilibrium model was employed, requiring a reduced amount of information. This model proved to be satisfactory, comparing its predictions to experimental and simulated data available in the literature at high temperature ($1000 \leq T \leq 1150$ ºC). The study of the co-gasification potential of these blends identified the operating conditions (*ER*, *MC* and *BR*) required to achieve the optimal gaseous products in cogeneration and chemical synthesis. Synergic effects were not observed in the predictions of the equilibrium model; the product compositions showed linear relationships with the linearly changing biomass ratio. Charcoal and wood blends led to gaseous product with higher heating values for 0% *MC* and $ER = 0.4$. Values higher than 80% *CGE* were achieved by blends with $ER \leq 0.3$. Based on technical criteria, suitable conditions for obtaining co-gasification products appropriate to use in chemical synthesis were identified for $BR \leq 35\%$ and $40 \leq MC \leq 50\%$. Another aspect evaluated was the content of undesirable nitrogen and sulfur compounds in the gaseous products. The results showed that is possible to obtain a suitable gas fuel for cogeneration within recommended H_2S limits only for $BR \geq 95\%$ by gasifying most of the coal-biomass blends. However, NH_3 release was shown to be adequate for all cases studied. The contents of H_2S and NH_3 in the products were below the values recommended in chemical synthesis for $BR \leq 5\%$ and higher values of *MC* and *ER*. This study is the first step towards the selection of optimal coal-biomass blends for co-gasification. Future studies will take into account economic criteria and other technical issues, such as regional and seasonal availability of biomass sources.

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NOMENCLATURE

ΔH_r	Heat of reaction (MJ/kmol)
N	Number of chemical species
n_{O_2}	Number of moles of O_2 injected into the reactor (kmol)
$n_{O_2,stoic}$	Stoichiometric number of moles of O_2 (kmol)
$x_{i,est}$	Estimated volume fraction of species i (m^3/m^3)
$x_{i,exp}$	Measured volume fraction of species i (m^3/m^3)
Y	Gas yield (Nm^3/kg)

Abbreviations

BR	Biomass Ratio (kg/kg)
CC	Carbon Conversion (kmol/kmol)
CGE	Cold Gas Efficiency (MJ/MJ)
daf	Dry ash-free basis
db	Dry basis (H_2O-free basis)
ER	Equivalence Ratio (kmol/kmol)
HHV	Higher Heating Value (MJ/kg)
LHV	Lower Heating Value (MJ/kg or MJ/Nm^3)
MC	Moisture Content (kg/kg)
N/A	Not Available, Not Applicable
%vol	Volume percent (m^3/m^3)
wb	Wet basis
%wt	Weight percent (kg/kg)

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