A short-cut model for predicting biomethane availability after biogas upgrading

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Abstract

Biomethane figures with increasing importance in the bioenergy sector. As a renewable energy source that promotes waste recovery and GHG (greenhouse gases) reduction, biomethane use aligns with cleaner production principles. However, many of the final uses to biomethane require an upgrading and cleaning process, to remove contaminants such as H_2S and CO_2. Facing the great amount of technological options to promote biogas upgrading and cleaning up to this date, it might be a rather challenging task to have a first estimate of biomethane availability required for a conceptual project level. Thus, the main objective of this paper is to propose a short-cut, mass balance-based model to predict biomethane availability after promoting a biogas cleaning and upgrading process regardless of the source of organic feedstock or the choice of the cleaning technology. The model development results into interesting dimensionless parameters, such as the gas contamination factors. Relevant parameters regarding biomethane use, such as its LHV and Wobbe index are also adapted to this model. The correlation with data from literature shows that the model has a satisfactory prediction when methane losses in the upgrading process are less than 3%.

Keywords: Biogas, Biomethane, Upgrading, Availability, Shortcut

1. Introduction

The production of biofuels from biomass and waste instead of using these sources "as is" for energetic proposes is one interesting approach to achieve cleaner production principles, such as reduction of overall greenhouse gases (GHG) emissions, waste recovery and reduction of environmental impacts (Kurnia et al., 2016, Leme and Seabra, 2017). In that sense, the use of biogas produced from wastewater anaerobic digestion and landfills is aligned with cleaner production goals.

There are different options for the insertion of biogas into the energy sector. One is use of raw biogas, which enables generation of electricity and heat in a combined heat and power unit (CHP) with a simple prior cleaning step to remove contaminants that could damage the equipment, such as H_2S and siloxanes. This option is suitable for distributed generation and the conversion technologies can be, for example, Otto engines and gas micro turbines (Santos et al., 2016). The other (and much more interesting) alternative is the upgrading of biogas to biomethane for use as a transport fuel. This option is promising because renewable fuel options for vehicular use are limited, while in power generation there is great competition with other energy sources (Larsson et al., 2016). Biomethane as a vehicle fuel is already a reality, especially in Sweden, where it could potentially reduce GHG emissions in transport by 25% (Olsson and Fallde, 2015).

Biomethane is also equivalent to natural gas, which enables the possibility of injecting biomethane in the same infrastructure for natural gas transportation, enabling natural gas substitution. Since natural gas was responsible for 21.2% of the world's primary energy supply in 2014 (IEA, 2016), its substitution is desirable to achieve reduction in CO_2 emissions. In this sense, it is crucial to be able to estimate biomethane availability after biogas upgrading for energetic and environmental reasons. Biogas composition depends greatly on the organic feedstock source, but it is mainly composed of CH_4 and CO_2, with traces of other contaminant such as H_2S, NH_3 and water vapor. Thus, the separation of gases that are not methane is the focus of the cleaning process (removal of contaminants) and the upgrading and cleaning process (removal of CO_2 for adjustment of the calorific value of the gas).

There are many technologies developed for H_2S and CO2 removal. The removal of H_2S and other contaminants is critical because, despite having lower concentration, they may have corrosive or clustering power combined. Therefore, the removal of these components is a relevant stage in the production of biomethane, both in terms of guaranteeing the system's performance and the costs involved. Hence, the removal of H_2S, as well as other damaging contaminants, such as water and siloxanes, if present, should occur in an upstream stage in the process (Patterson et al., 2011).

It is possible to classify the desulfurization step by the technique's capacity of rough or fine decontamination (Deublein and Steinheuser, 2008). While, at some cases, the requirements may be met using only one decontamination technique, it is common to combine two methods in processes that present high H_2S concentrations and/or require low values of H_2S, such as in biomethane production (Leme and Seabra, 2017). In this case, technologies that require air intake should be avoided, so that biogas is not contaminated with N_2 and O_2, gases that are difficult to remove. ***Table 1*** presents the main technologies for H_2S removal and its characteristics.

Table 1: H_2S removal technologies (adapted from Deublein and Steinheuser, 2008)

H_2S removal technology	**Level of decontamination**	**Air intake into biogas required**
Internal biological desulfurization	Very rough	Yes
Percolating filter plant	Rough	Yes
Bioscrubber plant	Rough	No
Sulfide precipitation	Rough	No
Ferric chelate	Rough	Yes
$Fe(OH)_3$ – bog iron ore	Fine	Both options
Fe_2O_3	Fine	Both options
Activated carbon – KI, K_2CO_3, $KMnO_4$	Fine	No
Zinc oxide	Fine	No
Surfactant	Fine	No
Absorption at glycol and ethanolamine	Fine	No
Algae	Fine	Yes
Direct oxidation	Fine	Yes

Adding to CO_2 and H_2S removal, biomethane production may require other cleaning steps. As raw biogas has a saturated content of water vapor, which condensates causing corrosion and clogging in pipes, dehumidification occurs at an early stage. Depending on the feedstock used for biogas production, other contaminants may be present, such as siloxanes, commonly present in wastewater treatment plants and landfill gas. Siloxanes form SiO_2 after combustion and microcrystalline quartz that deposit on engine or turbines surfaces, causing severe damages (Yang et al., 2014). Siloxanes, as well as ammonia, halogen compounds, N_2 and O_2 can make the process cleaning process even more complex.

Carbon dioxide removal, on the other hand, usually takes only on one technology that can meet the standards by itself, which allows an easier comparison of their performances. Many studies have reviewed biogas upgrading technologies, analyzing parameters such as energy consumption, methane losses and methane purity achieved in the biomethane (Sun et al., 2015). ***Table 2*** shows the most reported upgrading technologies.

Table 2: Biogas upgrading technologies (adapted from Probiogas (2010); Yang et al. (2014); Sun et al. (2015); Leme and Seabra (2017)).

Upgrading technology	CH_4 losses (typical)	CH_4 (vol%) in biomethane (possible)	Observations
Water scrubbing	Low (1-3%)	> 98%	High water demand; removes NH_3
Physical absorption	Low-Medium (2-4%)	> 96%	Different organic solvents available; removes H_2S, but may difficult regeneration of the solvent
Chemical absorption (amine scrubbing)	Very low (<0.1%)	> 99%	High heat demand; Low electricity consumption; removes H_2S; may produce high quality CO_2
Pressure swing adsorption (PSA)	Medium-High (4-9%)	> 97%	N_2/O_2 removal possible; requires fine H_2S pre-removal to protect adsorbent material
Membrane separation	Low-High (1-12%)	> 96%	Compact; low-medium energy requirements; membrane can be expensive

Literature often mentions that the choice of the upgrading technology depends on the conditions and characteristics of the plant. As Sun et al. (2015) describes, the choice "must be site-specific and case sensitive". To minimize the cost per unit of biomethane, one must analyze investment and operational costs, including the affordability and availability of the equipment, raw material, labor, electricity and heat. However, it is important that the process meets the composition requirements and its safety and environmental risks are adequate to the plant's site installations and workforce preparedness (Leme and Seabra, 2017).

Therefore, the several combinations of technologies may form different biogas upgrading processes alternatives. In a very competitive market, which includes competition with natural gas and other fossil fuels, it is necessary to be able to readily estimate important data for a plant, such as the production rate of biomethane and its composition and energy content to reach a given standard. However, to do so, one may need to dominate fundamentals of cleaning and upgrading technologies, which usually are complex thermodynamic and transport phenomena principles. This may be a rather challenging goal in a conceptual level project design or an emerging biomethane market, as is seen on Brazil. Thus, this paper addresses this issue by proposing a short-cut model to estimate biomethane availability after biogas cleaning and upgrading using only simple and common variables, such as gas composition and flowrates.

2. Methods

On the proposed model, the upgrading and cleaning system model is a black box, i.e.: the model will only concern the inputs and outputs of the control volume, regardless of the process inside it, thermodynamics or transport kinetics. ***Figure 1*** shows the proposed black box model. Table 3 presents all the variables used on the model. The conditions for volumetric variables are the normal conditions (273.15 k and 101,325 Pa) and the concentrations are in dry base.

Another important aspect of the model is that, even though its variables are mass based, specifications for biomethane commonly express the gas composition in terms of its volumetric fractions. Thus, a conversion for mass fraction to volumetric fraction is necessary. Equation (1) expresses the conversion of volumetric fraction into mass fraction:

$$x_i = \frac{\rho_i . y_i}{\rho_T} \quad (1)$$

In equation (1) x_i is the mass fraction, y_i is the volumetric fraction, ρ_i is the density of a component i and ρ_T is the total density of the gas mixture.

The model uses some simplifying assumptions, which are listed below:

- Inert gases (N_2 and O_2) are not captured by the cleaning and upgrading system;
- A loss stream exists, but it is composed solely of methane;
- Biogas enters the cleaning and upgrading systems at a dry state;
- The thermodynamic behavior of the gas is assumed to be ideal, following the ideal gas model;
- The thermodynamic behavior of the gas mixture is assumed to be ideal. Thus, Amagat's law of additive volume can be implied;
- The system shall be as economic as possible. Thus, if possible, the output concentrations shall not be less than those determined by biomethane standards.

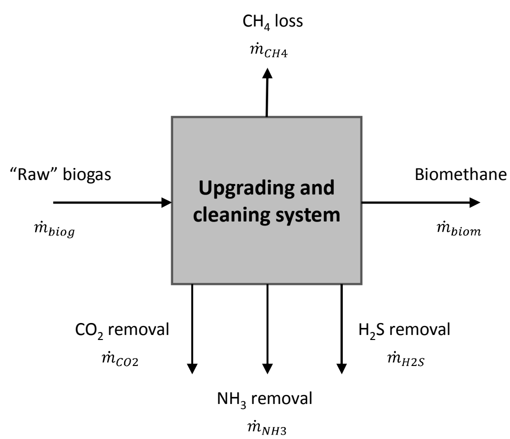

Figure 1: Black-box model for biogas cleaning and upgrading.

Table 3 shows expressions using both mass and volumetric fractions. Even though analytic methods and standards usually presents results using volumetric factions, the model requires mass fractions for the mass balance. Browsing through the presented expressions, it becomes clear that, to determine the value of the mass fraction of a component, the total density of the gas mixture, which also depends on the mass fraction of this same component, must be also determined. Hence, the problem becomes iterative and rather difficult to solve. It might be more practical to simply calculate $x_i = \frac{\rho_i . y_i}{\sum \rho_i . y_i}$ to first determine the mass fraction of each component and later calculate the total mixture density. Estimative for the individual density of each component comes from the ideal gas model behavior. Should the biogas or biomethane density be an easily accessible data (found on literature or be a project data), the first expression yields faster results.

The global and component mass balances yields the following equations:

Global mass balance: $\dot{m}_{biog} = \dot{m}_{CH4} + \dot{m}_{CO2} + \dot{m}_{H2S} + \dot{m}_{NH3} + \dot{m}_{Si} + \dot{m}_{biom}$ (2)

Methane mass balance: $x'_m . \dot{m}_{biom} + \dot{m}_{CH4} = x_m . \dot{m}_{biog}$ (3)

CO_2 balance: $\dot{m}_{CO2} = x_c . \dot{m}_{biog} - x'_c . \dot{m}_{biom}$ (4)

H_2S balance: $\dot{m}_{H2S} = x_s.\dot{m}_{biog} - x'_s.\dot{m}_{biom}$ (5)

NH_3 balance: $\dot{m}_{NH3} = x_n.\dot{m}_{biog} - x'_n.\dot{m}_{biom}$ (6)

Inerts balance: $(1 - x'_m - x'_c - x'_s - x'_n - x'_{Si}).\dot{m}_{biom} = (1 - x_m - x_c - x_s - x_n - x_{Si}).\dot{m}_{biog}$ (7)

Table 3: Model variables.

Variable		Biogas	Biomethane
Flowrate	Mass (kg/s)	$\dot{m}_{BG}$	$\dot{m}_{BM}$
	Volumetric (Nm³/s)	$Q_{BG} = \frac{\dot{m}_{BG}}{\rho_{BG}}$	$Q_{biom} = \frac{\dot{m}_{BM}}{\rho_{BM}}$
Density	kg/Nm³	$\frac{1}{\rho_{BG}} = \sum \frac{x_i}{\rho_i}$ *or* $\rho_{BG} = \sum y_i.\rho_i$	$\frac{1}{\rho_{BM}} = \sum \frac{x'_i}{\rho_i}$ *or* $\rho_{BM} = \sum y'_i.\rho_i$
Composition (volumetric fraction)	CH_4 (% vol)	y_m	y'_m
	CO_2 (% vol)	y_c	y'_c
	H_2S (% vol)	y_s	y'_s
	NH_3 (% vol)	y_n	y'_n
	Inert (% vol)	$y_{in} = 1 - y_m - y_c - y_s - y_n$	$y'_{in} = 1 - y'_m - y'_c - y'_s - y'_n$
Composition (mass fraction)	CH_4 (% wt)	$x_m = \frac{\rho_m.y_m}{\rho_{BG}}$ *or* $\frac{\rho_m.y_m}{\sum \rho_i.y_i}$	$x'_m = \frac{\rho_m.y_{\prime m}}{\rho_{BM}}$ *or* $\frac{\rho_m.y'_m}{\sum \rho_i.y'_i}$
	CO_2 (% wt)	$x_c = \frac{\rho_c.y_c}{\rho_{BG}}$ *or* $\frac{\rho_c.y_c}{\sum \rho_i.y_i}$	$x'_c = \frac{\rho_c.y'_c}{\rho_{BM}}$ *or* $\frac{\rho_c.y'_c}{\sum \rho_i.y_{\prime i}}$
	H_2S (% wt)	$x_s = \frac{\rho_s.y_s}{\rho_{BG}}$ *or* $\frac{\rho_s.y_s}{\sum \rho_i.y_i}$	$x'_s = \frac{\rho_s.y'_s}{\rho_{BM}}$ *or* $\frac{\rho_s.y'_s}{\sum \rho_i.y'_i}$
	NH_3 (% wt)	$x_n = \frac{\rho_n.y_n}{\rho_{BG}}$ *or* $\frac{\rho_n.y_n}{\sum \rho_i.y_i}$	$x'_n = \frac{\rho_n.y_{\prime n}}{\rho_{BM}}$ *or* $\frac{\rho_n.y'_n}{\sum \rho_i.y'_i}$
	Inert (% wt)	$x_{in} = 1 - x_m - x_c - x_s - x_n$	$x'_{in} = 1 - x'_m - x'_c - x'_s - x'_n$

In order to use a recurring performance parameter of methane-concentrating technologies, the rate of methane loss will be accounted as a factor of loss related to the input rate of methane (f_{loss}):

$\dot{m}_{CH4} = x_m.\dot{m}_{biog}.f_{loss}$ (8)

The proposed model will rise from the combination and rearrangement of equations (2)-(8) and the introduction of the following dimensionless parameters:

$X_{BM/BG} = \frac{\dot{m}_{BM}}{\dot{m}_{BG}}$ - mass conversion factor of biogas into biomethane by the process; (9)

$r = \frac{1}{1 - x_c - x_s - x_n}$ - biogas mass contamination factor; (10)

$r' = \frac{1}{1 - x'_c - x'_s - x'_n}$ - biomethane mass contamination factor; (11)

$r_m = \frac{1}{x_m.f_{loss}}$ - methane mass retention factor of the upgrading and cleaning system (12)

$R_{CO2} = \frac{\dot{m}_{CO2}}{x_c . \dot{m}_{BG}}$ - CO_2 mass removal factor of the upgrading and cleaning system (13)

The objective is to present a model that consists only of dimensionless parameters to predict the biomethane availability after any given biogas cleaning and upgrading process. Once this model is proposed, other important parameters, like the biomethane lower heating value (LHV), Wobbe Index will be combined to it. In order to validate the model, this study searched for data from case studies, experimental or simulation of biogas upgrading, regardless of technology or organic matter feedstock. Data such as composition and flowrates were collected and the parameters$X_{BM/BG}$, r and r' were calculated and fitted to the model using the software Microsoft Excel®. This study shall use graphic correlations to assess the fitting of the data to the model.

3. Results

3.1 Model development

By manipulating equations (2)–(8) and using the dimensionless parameters (9)–(13), one is able to obtain equation (14). This equation correlates the conversion factor with the gas quality factors before and after the upgrading and cleaning process and with the methane retention by the process. Equation (15) shows the methane concentration, obtained by combing equations (3), (5) and (14).

$$X_{BM/BG} = r' \cdot \left(\frac{1}{r} - \frac{1}{r_m}\right) \quad (14)$$

$$x'_m = \frac{x_m - 1/r_m}{X_{BM/BG}} \quad (15)$$

Though it may vary greatly according to the organic matter feedstock, biogas composition typically remains inside a quite narrow range of values. ***Table 4*** shows these typical values, as well as typical biomethane concentration and the parameter r or r' for each case. Additionally, as can be seen in ***Table 2***, most of the current upgrading technologies commonly have low methane loss factors, which leads to high values of the parameter of methane retention, r_m. Inspecting ***Table 4***, it becomes clear that, regardless of the source, the parameter r is much smaller than the parameter r_m. Therefore, the term $1/r_m$ is negligible if methane losses are low. Hence, equations (14) and (15) become equations (16) and (17), respectively:

$$X_{BM/BG} = \frac{r'}{r} \quad (16)$$

$$x'_m = \frac{x_m}{X_{BM/BG}} \quad (17)$$

Table 4: Typical dry biogas composition according to organic matter feedstock and typical dry biomethane composition (adapted from Allegue & Hinge, 2012; Ett. Et al, 2013 and). CH_4 loss is 2% ($\boldsymbol{f_{loss}}$=0.02).

Parameter	**Wastewater treatment plants**		**House-hold waste**		**Agricultural wastes**		**Biomethane**	
Classification	**Poor**	**Rich**	**Poor**	**Rich**	**Poor**	**Rich**	**Poor**	**Rich**
CH_4 (%wt)	37.70	58.30	30.00	39.10	35.80	57.00	90.00	97.00
CO_2 (%wt)	57.00	40.60	62.70	60.90	54.10	39.70	4.90	3.00
N_2+O_2 (%wt)	1.70	0.00	6.50	0.00	1.60	0.00	3.10	0.00
H_2S (%wt)	3.50	1.10	0.80	0.10	8.40	3.20	0.02	0.00
NH_3 (%wt)	0.00	0.00	0.00	0.00	0.10	0.10	0.02	0.00
r or r'	2.646	1.715	3.333	2.564	2.793	1.754	1.183	1.153
r_m	132.63	85.76	166.67	127.88	139.66	87.72	-	-

In order to determine the value of the CO_2 concentration in biomethane, the dimensionless parameter R_{CO2} comes in need. Uniting equations (4) and (13), one obtains:

$$x'_c = x_c \cdot \frac{1-R_{CO2}}{X_{BM/BG}} \quad (18)$$

Finally, in order to obtain the volumetric conversion of biogas into biomethane ($Y_{BM/BG}$), simply manipulating the ratio of volumetric rates of biogas and biomethane yields:

$$Y_{BM/BG} = X_{BM/BG} \cdot \frac{G_{BG/m}}{G_{BM/m}} \quad (19)$$

Where $G_{BG/m}$ and $G_{BM/m}$ are the specific gravities of biogas and biomethane, respectively, related to pure methane. Additionally, one can write $G_{BM/m}$ using the equations shown ***Table 3***. As a simplification, the model will disregard the H_2S and NH_3 contributions to the final density, since the concentrations of these components will be very low on biomethane, as shown on ***Table 4***. Thus, one obtains:

$$G_{BM/m} = \frac{X_{BM/BG}}{F_{mc}} \quad (20)$$

Where $F_{mc} = \left(x_m + \frac{\rho_m}{\rho_c} \cdot x_c.(1 - R_{CO2})\right)$ is dimensionless density correction factor due to the removal of CO_2 and the concentration of CH_4. $G_{BG/m}$ is easily calculated using equations on ***Table 3*** and biogas data.

Two other important parameters for biomethane use for injection on the natural gas pipeline are its energy content (LHV) and its gas interchangeability factor (Wobbe index) (Wolfgang, 2013). The biomethane LHV is the weighted average of each of its components LHV. Again, the model disregards H_2S and NH_3 contributions. Being inert to combustion, CO_2 also does not contribute to the LHV. Thus, only the methane contribution will take place, leading to equation (21). The definition of the Wobbe Index $Wb = LHV/\sqrt{G_{gas/air}}$, where $G_{gas/air}$ is the specific gravity of a gas related to air (Suwansri et al., 2014). Hence, using equations (20) and (21), equation (22) express the Wobbe Index:

$$LHV_{BM} = \frac{x_m.LHV_m}{X_{BM/BG}} \quad (21)$$

$$Wb_{BM} = \frac{x_m.LHV_m}{X_{BM/BG}^{1,5}} \cdot \left(\frac{F_{mc}}{G_{m/air}}\right)^{0,5} \quad (22)$$

3.2 Model validation

Table 5 shows the data retrieved from literature already in the form of the calculated parameters of the model. When plotting $X_{BM/BG}$ against r'/r, a linear correlation with angular coefficient equal to 1 and linear coefficient equal to zero is expected. ***Figure 2*** shows the data plot against the model. Methane losses under 1% were considered negligible.

Inspecting ***Table 5***, one can perceive that the model fits quite well to data from literature where the methane loss is lower than 3%. When the methane loss is high, the data seems to deviate from the model, which is according to the hypothesis that the model may disregard the methane loss if this parameter is low enough. To corroborate this observation, ***Figure 3*** shows the plot whit data corresponding to high methane loss removed***.*** The fitting to the model then becomes nearly perfect, with a fitting parameter R^2 of 0,994.

Figure 4 and ***Figure 5*** show the calculated LHV and Wobbe Index using the model. A threshold of the minimum and maximum LHV and Wobbe Index calculated from ***Table 2*** with 5% tolerance was chosen as the range of acceptable values. Most of the results fall within the established range, which shows, once again, that the model fits quite well to literature data. Outliers are mainly from data tied to high methane losses.

Table 5: *Data from literature.*

Data	Upgrading technology	Methane loss	Calculated $X_{BM/BG}$	Calculated r/r'	Reference
1	Water scrubbing	2.0%	0.378	0.386	Leme and Seabra, 2017
2	Organic scrubbing	3.0%	0.374	0.386	Leme and Seabra, 2017
3	Amine scrubbing	Neg.	0.365	0.365	Leme and Seabra, 2017
4	Membrane	1.0%	0.367	0.370	Leme and Seabra, 2017
5	PSA	8.0%	0.347	0.386	Leme and Seabra, 2017
6	Water scrubbing	Neg.	0.381	0.383	Rotuno et al., 2017
7	Water scrubbing	Neg.	0.381	0.383	Rotuno et al., 2017
8	Water scrubbing	Neg.	0.382	0.383	Rotuno et al., 2017
9	Water scrubbing	Neg.	0.380	0.383	Rotuno et al., 2017
10	Water scrubbing	Neg.	0.383	0.383	Rotuno et al., 2017
11	Water scrubbing	Neg.	0.381	0.383	Rotuno et al., 2017
12	Caustic scrubbing	1.0%	0.096	0.097	Leonzio, 2016
13	Membrane	11.5%	0.295	0.334	Valenti et al., 2016
14	Cryogenic separation	Neg.	0.399	0.399	Youssef et al., 2016
15	Water scrubbing	Neg.	0.383	0.373	Xu et al., 2015
16	Water scrubbing	3.5%	0.387	0.399	Morero et al, 2017
17	Amine scrubbing	Neg.	0.431	0.421	Collet et al., 2017
18	Membrane	7.5%	0.372	0.419	Collet et al., 2017
19	Water scrubbing	1%	0.424	0.429	Wylock and Budzianowski, 2017
20	Membrane	14.8%	0.463	0.547	Molino et al, 2013

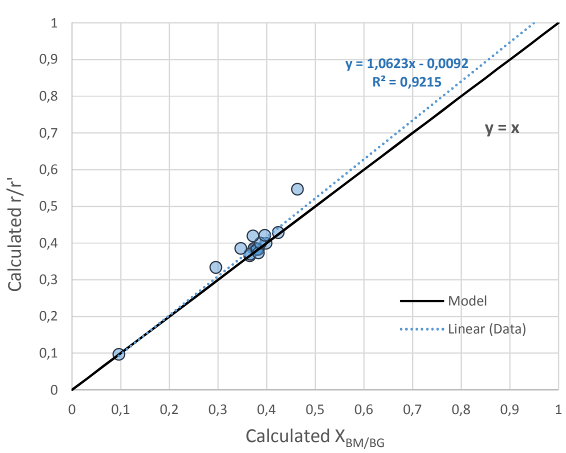

Figure 2: *Data and model correlation.*

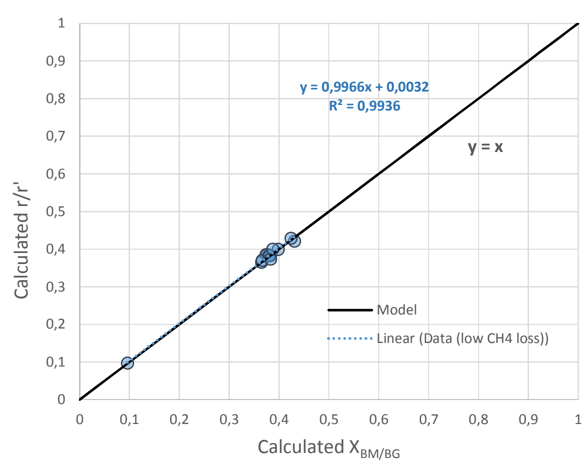

Figure 3: *Data and model plot considering only data with methane loss lower than 3%.*

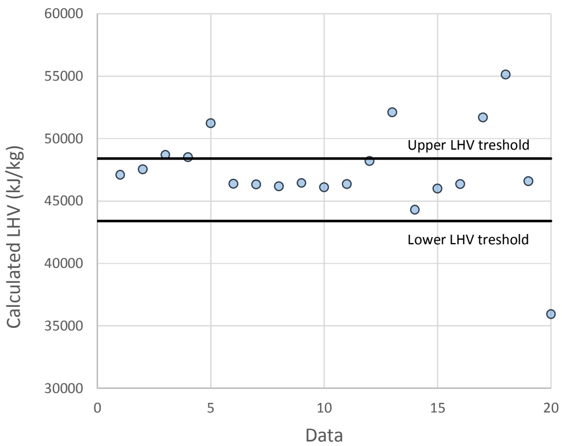

Figure 4: *Model fitting for LHV.*

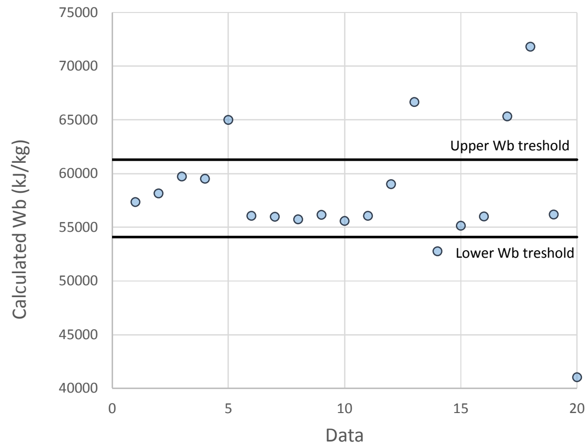

Figure 5: *Model fitting for Wobbe Index.*

4. Conclusions

The presented short-cut model allows to calculate important parameters of a biogas upgrading process, such as the conversion of biogas to biomethane, its LHV and Wobbe Index. Data from literature fitted to the model with a R^2 parameter of 0,994, and calculated LHV and Wobbe Index were inside the defined range of acceptable values, which shows a very satisfactory correlation.

It is important to highlight that the model, based solely on a mass balance, makes no differentiation on data regarding the organic feedstock for biogas or the technology used to perform the upgrading. The simplicity of the model makes it very suitable for conceptual engineering projects and estimation of CO_2 emissions reduction, and may provide strategic advantage, especially in countries like Brazil, where the interest on biomethane environmental and market potentials is increasing. However, a limitation of the model is predicting biomethane availability on upgrading process with CH_4 losses higher than 3%. Hence, it might not be a suitable choice if the upgrading technology is one that typically has a high CH_4 slip, such as PSA or membranes separation. Further studies should assess if the insertion of the parameter rm on the proposed equations would provide a better fit to literature data with high methane loss.

5. Acknowledgments

The authors gratefully acknowledge support from Shell and FAPESP through the "Reserch Centre for Gas Innovation - RCGI" (FAPESP Proc. 2014/50279-4), hosted by the University of Sao Paulo, and the strategic importance of the support given by ANP (Brazil's National Oil, Natural Gas and Biofuels Agency) through the R&D levy regulation. Special thanks to RCGI Project 27. The authors also thanks CNPq (Conselho Nacional de Desenvolvimento Científico e Tecnológico) for the financial support.

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