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EXERGY ANALYSIS OF BIOGAS PRODUCTION IN ETHANOL DISTILLERIES

Rafael Nogueira Nakashima
Escola Politécnica da Universidade de São Paulo
rafaelnnakashima@usp.br, rafaelnnakashima@gmail.com

Silvio de Oliveira Junior
Escola Politécnica da Universidade de São Paulo
soj@usp.br

Abstract. Vinasse is the main liquid effluent of ethanol distillation and, when incorrectly disposed, can cause damages in water and soil resources. Since this effluent has a high concentration of organic compounds, anaerobic digestion is a possible solution to reduce vinasse chemical organic demand (COD) while producing biogas, an alternative fuel. However, this technology has some challenges related to biogas economical devaluation and high investments costs, which demands models to analyze this systems performance. Thus, the focus of this study was to develop a biogas production model, combining Matlab® and Aspen Plus®, to evaluate the anaerobic treatment efficiency by using exergy analysis. The results show that a significant quantity of exergy can be partially retrieved (44-52%) from vinasse with biogas production. In general, increases in hydraulic retention times (HRT) and COD concentrations tend to slightly increment anaerobic digestion efficiency, while high sulfates concentrations have the opposite effect. However, the biogas plant exergy efficiency tends to increase at lower HRT due to higher biogas production and lower portion of untreated vinasse. On average, the main sources of irreversibilities are the cooling system (45%) and the anaerobic reactor (48%). Moreover, the biogas plant performance throughout the year is considerably affected by ethanol production seasonality, start-up times and number of reactors.

Keywords: biogas; exergy; vinasse; anaerobic digestion, ethanol distillery.

1. INTRODUCTION

Vinasse is the main liquid effluent of ethanol distilleries and consists of water with a high concentration of organic substances and ions. On average, 12 liters of this waste are produced per liter of ethanol (Elia Neto and Shintaku, 2009). Therefore, this wastewater represents an environmental danger if not disposed correctly. In Brazil, vinasse is used during irrigation of sugar cane crops as an alternative fertilizer, a process called fertirrigation. However, the possibility of methane emissions and contamination of water and soil resources raises concerns about the sustainability of fertirrigation (Moraes, et al., 2013) (Fuess and Garcia, 2014). In this context, anaerobic digestion is a possible solution to reduce vinasse chemical organic demand and, consequently, its pollution potential. The main advantage of this process is the production of biogas, which is an alternative fuel that can be purified to supply power and/or heat demands.

This technology has been proposed since the 1980s in Brazil, when the firsts systems were designed and installed, but nowadays few distilleries have biogas plants in operation (Poveda, 2014). Biogas produced from vinasse was first planned as a fossil fuel substitute for transportation in ethanol distilleries, due to the high prices of oil during the 1970s energy crisis (Braunbeck and Cortez, 2002). However, the stabilization of oil prices associated with technological difficulties stagnate further investigations in Brazil for some years (Pinto, 1999). Today, biogas installations are studied to improve ethanol distilleries productivity by offering a by-product from vinasse, which can be used to produce heat and power or be sold as an alternative natural gas.

In general, studies of biogas production and use in ethanol distilleries suggest a reduction energy consumption and enhancement in overall system efficiency. For example, Palacios-Berenche et al. (2013) investigated the exergetic performance of integrated first and second-generation ethanol and determined better results for systems including biogas as an additional product. Fuess and Garcia (2015) studied the energy balance for ethanol production and found that biogas could supply 5-15% of the consumed energy. Barrera et al. (2016) examined different biogas plants

configurations and showed that these systems could be 39% more exergetic efficient compared with the traditional treatment.

Nevertheless, there is a lack of studies focusing on analyzing biogas production including the biological reactions of anaerobic digestion. Thus, this paper attempts to develop a methodology for analyzing biogas production using the exergy method.

2. MODEL DESCRIPTION

2.1 Vinasse composition

Since usually effluents are not well-defined mixtures, their properties are difficult to evaluate. Therefore, a representative composition for vinasse was assumed in order to model anaerobic digestion and calculate physical and chemical exergy. This composition was based on substances modeled on ADM1 (Anaerobic Digestion Model Nº1) (Batstone, et al., 2002) and concentrations reported by Barrera et al. (2015). It was assumed that each organic compound concentration (C_i) was proportional to the effluent chemical organic demand (COD_{op}) and its representative mass fraction (m_i) in kgO_2, according to Eq. (1) and Tab. 1.

$$C_i = COD_{op} \cdot m_i \tag{1}$$

Table 1. Mass fraction of organic substances

Substance	m_i (kgO_2/kgO_2)[1]	Modeled as
Sugar	0.518	Dextrose ($C_6H_{12}O_6$)
Amino acids	0.089	Average amino acid ($C_{4.82}H_{8.95}O_{2.88}N_{1.13}S_{0.02}$)
Long-chain fatty acids (LCFA)	0.001	Linoleic acid ($C_{18}H_{32}O_2$)
Acetic acid	0.021	Acetic acid ($C_2H_4O_2$)
Carbohydrates	0.106	Sucrose ($C_{12}H_{22}O_{11}$)
Proteins	0.001	Average amino acid ($C_{4.82}H_{8.95}O_{2.88}N_{1.13}S_{0.02}$)
Lipids	0.002	Linoleic acid ($C_{18}H_{32}O_2$)
Inert	0.260	Phenol (C_6H_6O)
Biomass / Composite	0	Ethyl cyanoacetate ($C_5H_7NO_2$)

[1]calculated based on values reported by Barrera et al. (2015)

The mean concentrations of inorganic substances reported by Elia Neto and Shitaku (2009), presented in Tab. 2, were assumed constant for different COD concentrations and used to characterize Brazilian sugarcane vinasse. A related chemical substance was used to evaluate the physical properties of generic organic components, as it can be seen in Tab. 1. In the case of amino acids, mean properties were calculated based on weight distribution of 14 amino acids present in vinasse (Waliszewski, et al, 1997). Moreover, proteins and lipids properties were assumed equivalent to amino acids and LCFA respectively, while ionic substances were modeled as shown in Tab. 2.

Table 2. Mass fraction of organic substances (Elia Neto and Shintaku, 2009)

Substance	Concentration (mg/L)	Modeled as
Nitrogen	37	NH_3
Phosphorus	34	H_3PO_4
Potassium	2206	KOH
Calcium	832	$Ca(OH)_2$
Magnesium	262	$Mg(OH)_2$
Sulfate	1149	H_2SO_4

2.2 Biogas plant model

The biogas plant model, shown in Fig. 2, is based on the anaerobic digestion unit of São Martinho plant (Pradópolis, São Paulo, Brazil) as described by Poveda (2014) and Souza et al. (1992). Vinasse produced by wine distillation is distributed for anaerobic digestion in a UASB (Upflow Anaerobic Sludge Blanket) reactor, which temperature and pH are maintained at 35ºC (T_{op}) and 7.5, respectively. Thus, vinasse preparation consists of a cooling unit and the addition of a caustic solution (100 kg/m³ of NaOH), which is determinated based on the bicarbonate concentration on the digested vinasse (Metcalf & Eddy, 2003). After anaerobic digestion, biogas is separated to cleaning, storage and use, while digested effluent is disposed in sugarcane fields with the untreated vinasse.
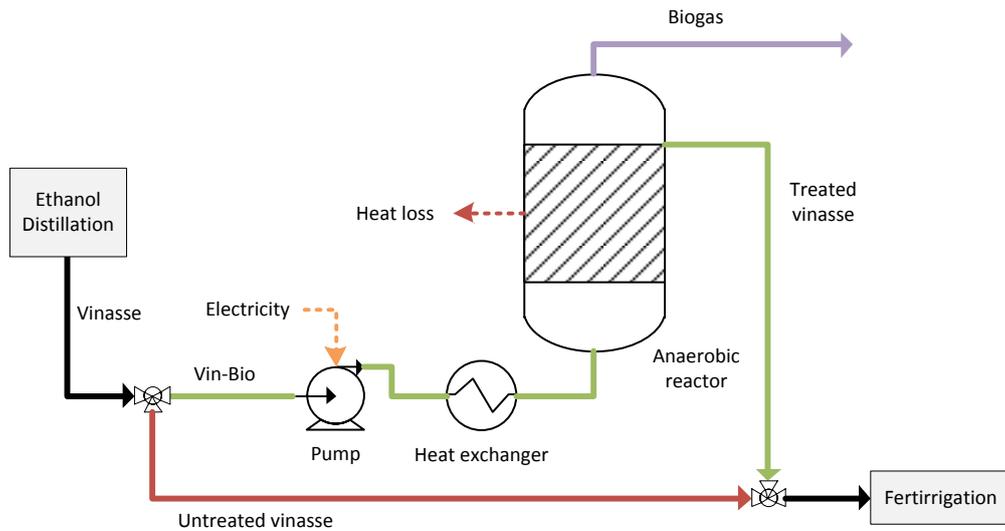
Figure 1. Biogas plant model in Aspen Plus®

The reactor was considered directly heated by the entering waste water, whose temperature was estimated by the energy balance of the anaerobic digester, described in Eq. (2), where $\dot{H}$ and $\dot{Q}_{loss}$ are the enthalpy flowrate and reactor heat rate loss. Moreover, the subscripts 'i', 'o' and 'g' refer to input vinasse, digested vinasse and biogas produced, respectively. The heat rate loss was calculated using Eq. (3), assuming heat transfer coefficients (U) based on common materials for reactors (Metcalf & Eddy, 2003), which are shown on Tab. 3. Furthermore, the ambient temperature (T_0) was considered 25ºC and the reactor surface area (A) was estimated assumimg a tubular form with a diameter/height ratio of 1:3. Other types of losses, such as friction and heat convection, were not included in this model.

$$\dot{H}_i = \dot{H}_g + \dot{H}_0 - \dot{Q}_{loss} \tag{2}$$

$$\dot{Q}_{loss} = U \cdot A \cdot (T_0 - T_{op}) \tag{3}$$

Table 3. Heat losses parameters

Surface	Area (m²)	Heat transfer coefficient	Value (W/m²)
Walls	1556	Plain concrete walls; 300 mm; not isolated	5.1
Floor	130	Plain concrete floors; 300 mm; moist earth	2.85
Cover	130	Fixed steel covers 6 mm thick	5.4

The anaerobic digestion reactions were modeled using ADM1 (Batstone, et al., 2002), which consists in a system of ordinary differential equations that was solved by using Matlab numerical solver *ode15s*. Moreover, this model includes the sulfate reduction reactions and calibrated constants for vinasse anaerobic degradation reported by Barrera, et al. (2015). The initial conditions were determined by iteration using vinasse with an equal distribution of bacteria concentration. Furthermore, the mass balance for steady state results were calculated using long time simulations (>200 days) results.

A software interface between Matlab® and Aspen Plus® was developed to analyze the biogas production. This link was made by connecting Aspen Plus® *USER2* tool with an Excel® spreadsheet, which uses an Add-In to call a Matlab® function to solve ADM1. In other words, Matlab® computes the anaerobic reactor mass balance, using ADM1, and Aspen Plus® calculates the energy and entropy balances, using the NRTL (Non-random two-liquid model) method. The connection between these softwares was described in detail by Nguyen (2014) and Fontalvo Alzate (2014).

Table 4 shows the main parameters considered in this analysis, the base case values assumed were 45 kgO$_2$/m³, 12 kgO$_2$/m³-day and 0.048 gSO$_4^{2-}$/gO$_2$ for chemical organic demand concentration (COD), organic loading ratio (OLR) and sulfate relative concentration (SO$_4^{2-}$/COD), respectively. The vinasse flow rates (Q) and hydraulic retention times (HRT) were calculated using Eq. (4), assuming a reactor volume (V$_{reactor}$) of 5000 m³, for the interval of calibrated organic loading ratios for the model (Barrera, et al, 2015). The COD and SO$_4^{2-}$/COD concentrations are based on average values of Brazilian ethanol distilleries reported by Elia Neto and Shitaku (2009).

Table 4. Main parameters used in the biogas plant model

UASB reactor		Vinasse	
Parameter	Value	Parameter	Value
Volume	5000 m³	Initial temperature	90 °C
Temperature	35 °C	Pressure	1 atm
pH	7.5	Flow	480 m³/h
OLR	12-7 kgO$_2$/m³-day	COD	65; 45 e 33 kgO$_2$/m³
		SO$_4^{2-}$/COD	0.02 ; 0.03 e 0.048 g/gO$_2$

$$OLR = \frac{COD}{HRT} = \frac{Q \cdot COD}{V_{reactor}} \tag{4}$$

The anaerobic digestion performance throughout the year was divided in three periods: start-up, steady state and offseason. The first one refers to the acclimatization and reproduction of bacteria to achieve a constant organic loading ratio and biogas production. This process was modeled by a linear increase in vinasse flow rate (45 gO$_2$/L and 0.048 gSO$_4^{2-}$/gO$_2$) entering the anaerobic digester during a certain period of time. Furthermore, fluctuations in biogas composition during the start-up were not considered in this model. The second period is the stable state of vinasse treatment, characterized by the maximum OLR possible (12 kgO$_2$/m³-day). The last period is the inactive phase, when ethanol and vinasse are not produced, usually for 4 months (120 days), because there is no sugarcane available. The efficiency and biogas production were considered as null in this phase. Moreover, only the active days (start-up and steady state regime) were used to calculate the mean efficiency and methane production. The vinasse rate of production throughout the year was estimated using the data of ethanol production for 2015/2016 season reported by UNICA (2016).

2.3 Exergy analysis

The thermodynamic properties of vinasse were evaluated by its representative composition properties. The specific physical exergy (b_p) was calculated using Eq. (5), where h and s are the specific enthalpy and entropy in a given state, while the subscript '0' refers to the environmental state (25 °C and 1 atm).

$$b_p = h - h_0 - T_0(s - s_0) \tag{5}$$

All streams were considered ideal mixtures and their chemical exergy ($b_{c,mix}$) was evaluated using Eq. (6), in which x_j and R are the component 'j' molar fraction and universal gas constant. The chemical specific exergy (b_c) values reported by Szargut (2005) were used for well-defined substances. Moreover, the representative composition of vinasse was used to estimate the effluent molar fraction distribution of organic compounds.

$$b_{c,mix} = \sum_j x_j b_{c,j} + RT_0 \sum_j x_j \ln x_j \tag{6}$$

The chemical exergy of the organic compounds in vinasse was evaluated by a correlation with its theoretical COD (ThOD), as done by previous studies on effluent chemical exergy (Tai, et al., 1986)(Khosravi, et al., 2013), but modified to correspond with environmental conditions and assumptions proposed by Szargut (2005). The ThOD represents the mass of oxygen needed to oxidize an organic compound in a reference reaction, which was assumed as described in Reaction (R1) and Eq. (7). In this reaction, nitrogen atoms in organic compounds are assumed to be converted in N$_2$ in order to improve the correlation with chemical exergy.

$$C_aH_bO_cN_dS_e + (a + \frac{b}{4} - \frac{c}{2} + 1.5e)O_2 \rightarrow aCO_2 + (\frac{b}{2} - e)H_2O + dN_2 + eH_2SO_4 \tag{R1}$$
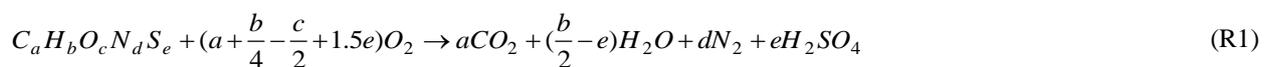
$$ThOD(\frac{gO_2}{mol}) = 32(a + \frac{b}{4} - \frac{c}{2} + 1.5e) \tag{7}$$

This methodology was used to minimize uncertainties in the evaluation of vinasse chemical exergy, since COD concentrations are independent from assumptions in the representative composition. The final correlation, Eq. (8), was

determined by a linear correspondence between ThOD and b_c for 32 organic substances, including different types of sugars, alcohols, organic acids, amino acids, fats and others that possibly constitute vinasse, as seen in Fig. 2.

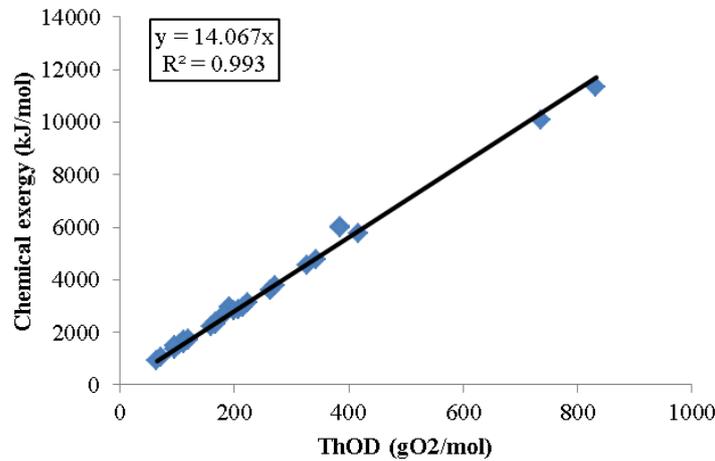$$b_{ch}(\frac{kJ}{mol}) = 14.1 \cdot ThOD(\frac{g}{mol}) \tag{8}$$

Figure 2. Correlation between chemical exergy and ThOD for organic substances

Exergy efficiency can be defined as the ratio between exergy of useful products and feeding exergy of a certain process. Since biogas is considered the only product, the anaerobic digestion (AD) and biogas plant (BP) efficiency (η_{AD} and η_{BP}) are defined as presented in Eq. (9) and (10), respectively, in which $\dot{B}$ is the exergy flowrate for streams shown in Fig. (1). The difference between these parameters is that the former accounts only for the vinasse sent for anaerobic digestion (*Vin-Bio*), while the latter considers all vinasse produced from distillation (*Vinasse*).

$$\eta_{AD} = \frac{\dot{B}_{biogas}}{\dot{B}_{vin\text{-}bio} + \dot{B}_{eletricity} + \dot{B}_{NaOH}} \tag{9}$$

$$\eta_{BP} = \frac{\dot{B}_{biogas}}{\dot{B}_{vinasse} + \dot{B}_{eletricity} + \dot{B}_{NaOH}} \tag{10}$$

In the efficiency analysis throughout the season, it was assumed that $\dot{B}_{eletricity}$ and $\dot{B}_{NaOH}$ are much smaller than the vinasse exergy ($\dot{B}_{vinasse}$), thus Eq. (9) and (10) can be combined as described in Eq. (11), where $\dot{v}$ is the volumetric flow rate. The anaerobic digestion efficiency was calculated as function of HRT using results from the parametric analysis, as shown in Eq. (12) and (13).

$$\eta_{BP} = \frac{\dot{B}_{vin-bio}}{\dot{B}_{vinasse}} \cdot \eta_{AD} = \frac{\dot{v}_{vin-bio}}{\dot{v}_{total\,vinasse}} \cdot \eta_{AD} \tag{11}$$

$$\text{If } HRT > HRT_{max}, \text{ then}: \quad \eta_{AD} = \eta(HRT_{max}) \tag{12}$$

$$\text{If } HRT_{min} \leq HRT \leq HRT_{max}, \text{ then}: \quad \eta_{AD} = \eta(HRT) \tag{13}$$

3. RESULTS AND DISCUSSION

As it can be seen in Fig. 3, for the base case, most of the input exergy is available as organic matter easily degradable (66%) and inert organic substances (23%). Thus, due to high chemical exergy of organic compounds present in this effluent, vinasse is the main exergy flow rate (99.9%) in anaerobic digestion. Electricity and NaOH exergies represent a little percentage (0.1%) in comparison with the others, thus they have low influence on the efficiencies. Still,

most of the output exergy is available as methane gas (44%), followed by inert (23%) and bacteria (11%) in the effluent.

For this base case, the anaerobic digestion exergy efficiency is 46%, with 7% of entering exergy being destroyed in the system, while the biogas plant efficiency was 5% for only one reactor. Although the biogas production has a good performance, one anaerobic digester can only treat 11% of the total vinasse produced, therefore, limiting the impact of the treatment for the ethanol distillery.

(a)   (b)
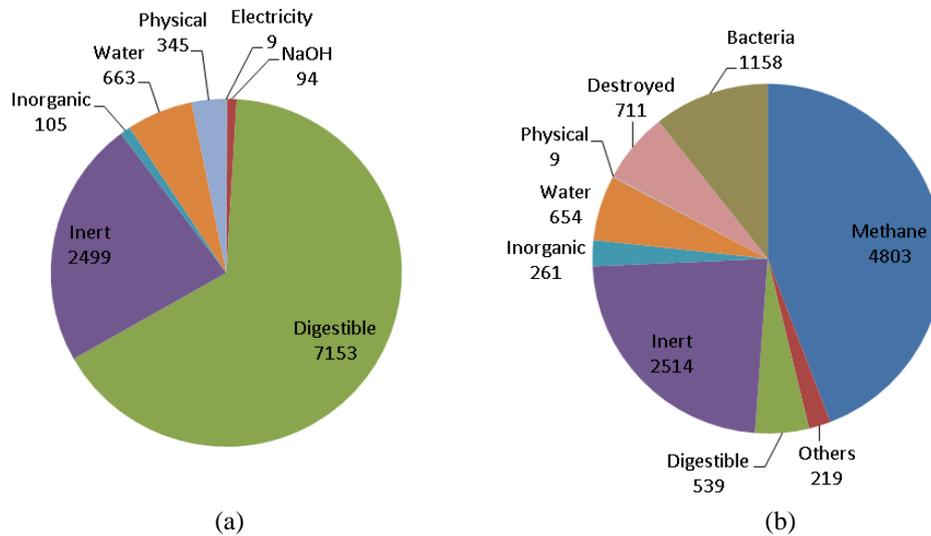
Figure 3. Anaerobic digestion exergy flow rates (in kW) (a) input, (b) output

Figure 4 shows the anaerobic digestion and biogas plant efficiency for the hydraulic retention times and COD concentrations indicated in Tab. 4. As it can be seen in Fig. 4 (a), the anaerobic digestion exergy efficiency varies from 43.5% to 51.7% for stable regimes and drops to 10% for low retention times. This is partially due to the imbalance between the anaerobic digestion reactions rates and to the perfectly mixed assumption, which does not include solid retention. In general, the system is more efficient for higher HRT and COD concentrations, because there is more time and organic compounds for bacteria development and biogas production. Furthermore, HRT has a low influence on efficiency for constant chemical organic concentrations, around 2.1-7.6% variations, and COD does not seem to have a direct influence on biogas plant efficiency.

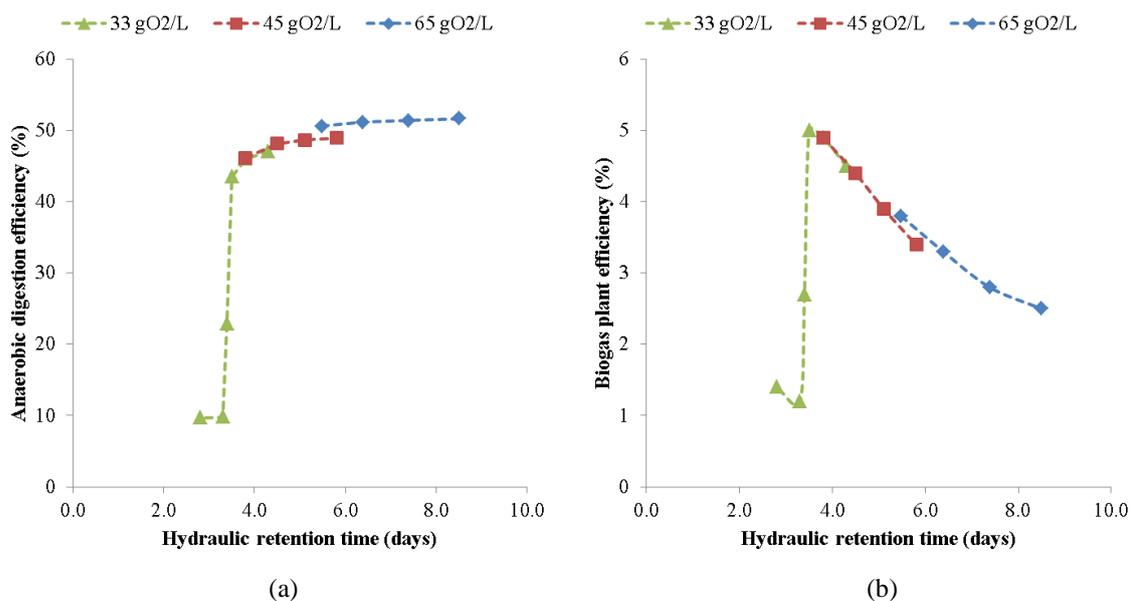(a)   (b)

Figure 4. Exergy efficiencies per HRT and COD concentrations (a) Anaerobic digestion, (b) Biogas plant

On the other hand, as presented in Fig. 4 (b), the operation at lower retention times treats higher volumes of vinasse and, therefore, increases the amount of biogas produced per reactor. Although the anaerobic digestion efficiency is lower, the biogas plant efficiency is increased because more biogas is produced. Nevertheless, since anaerobic reactions can reach an unbalance, as shown for 33 gO_2/L concentration, the reduction of HRT is limited by the reactor type and vinasse composition. Other interesting analysis, shown in Fig. 5, is the influence of the relative concentration of sulfates (SO_4^{2-}/gO_2). As expected, the inhibitory nature of sulfates for methanogenic bacteria negatively affects the efficiency, with a maximum decrease of 5.8%, and highly rises the H_2S generation. This possibly increases the amount of resources needed to desulfurize the gas for storage and use, reducing the performance of these systems.
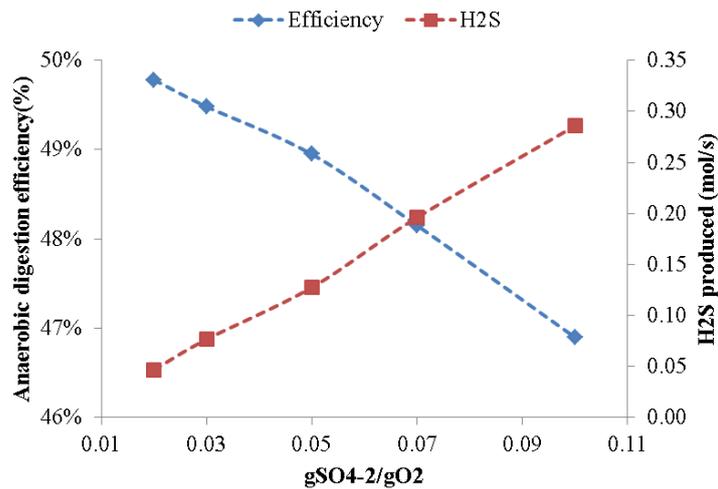

Figure 5. Anaerobic digestion efficiency for different sulfates concentrations

On average, the exergy destroyed in biogas production is mainly due to the cooling system (45%) and the anaerobic reactor (48%), as shown in Fig. 6. The impact of the cooling system is lower for vinasses with higher COD concentrations, due to the lower influence of physical exergy over the total and lower vinasse flow rates, which increases the entering vinasse temperature. Since vinasse initial temperature has to be adjusted to mesophilic regions, if thermal recover systems are not included, most of vinasse physical exergy is destroyed to the environment. Moreover, the anaerobic digester reactions and heat losses are significant sources of irreversibilities for biogas production. In perspective, the mean system exergy destruction represents 13% of the biogas produced exergy.
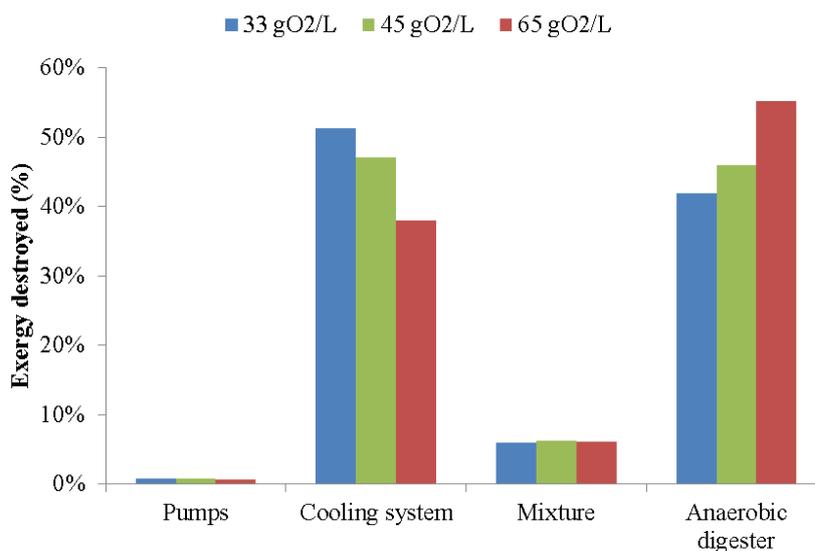

Figure 6. Exergy destroyed per process

A possible enhancement is to increase the anaerobic reactor temperature (thermophilic), which could reduce the cooling demand and increase the yield of methanogenic bacteria. However, thermophilic temperatures increase the

reactor instability and heat losses, which can increase the digester exergy destruction. Other alternative is to use vinasse to heat the wine, before distillation, or the digester, to maintain the operational temperature. Furthermore, solid retention could reduce exergy of digested vinasse and improve the rate of biogas production, enhancing the exergetic efficiency.

Figure 7 presents the biogas plant exergy efficiency for different numbers of reactors along 240 days, based on ethanol production data described in UNICA (2016). This efficiency starts with low values between 4% and 13% and steadily rises until it remains almost constant, as in the nine reactors case, or it oscillates and sharply rises towards the end of season. These fluctuations are due to variations in ethanol production, which reduces or increases the amount of effluent to be accounted for in Eq. (11). As expected, the systems perform better for cases with more reactors, because less vinasse is left untreated. However, for periods of vinasse shortages, such as between days 200-240, the difference is reduced or almost negligible.
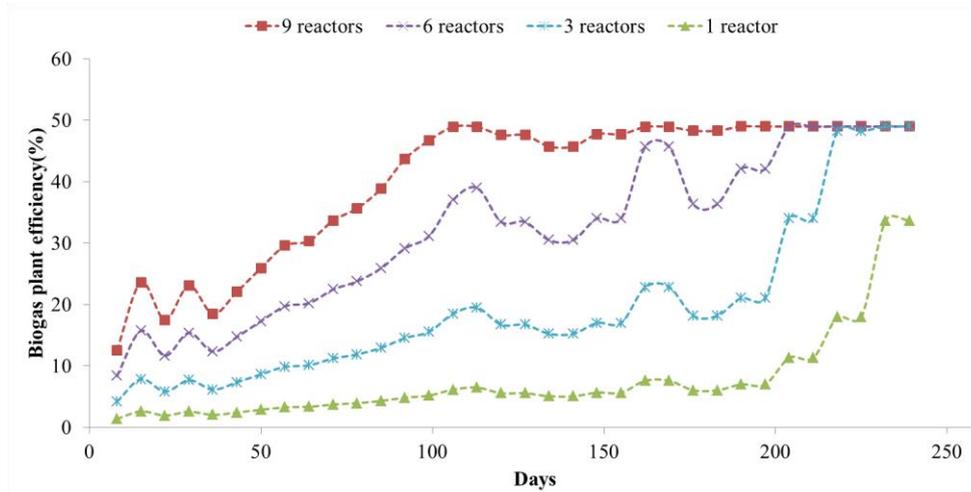

Figure 7. Biogas plant exergy efficiency in a season

The start-up process, which can take 2-4 months (Deublein and Steinhauser, 2011), has a reduced efficiency compared to more stable states because in this period the organic loading ratio is reduced and, therefore, less vinasse can be treated. The start-up can be improved by using inoculating bacteria from other waste water treatment, ideally also operating with vinasse, or by retaining bacteria.

As seen in Fig. 8, the mean biogas plant efficiency throughout the season is close to the stable steady state, except for the nine reactors case that has a 28% reduction. Although all cases have the total efficiency reduced by the start-up period, the vinasse shortages significantly increased the efficiency for systems that treated less vinasse than produced. Thus, for reactors operating with breaks during the off-season, start-up times and vinasse rates fluctuations have to be taken into account in this systems design.
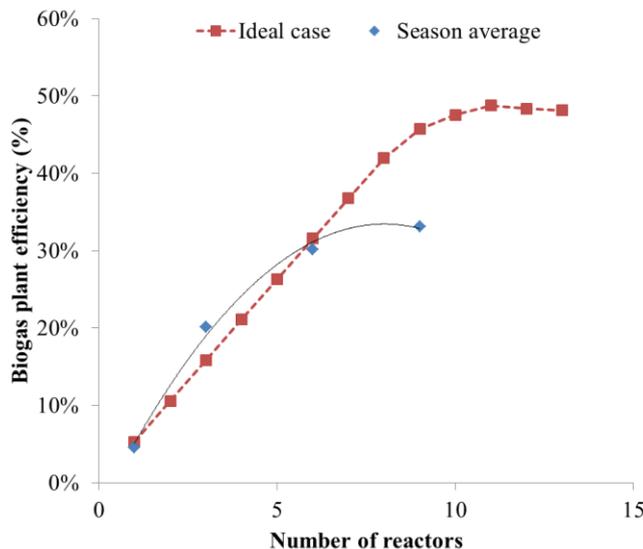

Figure 8. Comparison between ideal efficiency and mean efficiency per season

4. CONCLUSIONS

This study has shown that a significant quantity of exergy contained in vinasse can be partially retrieved (44-52%) by producing biogas. Moreover, the effects of hydraulic retention time, COD and sulfates concentrations were investigated for the anaerobic digestion and biogas plant system. The main sources of irreversibilities found were due to the cooling process and the anaerobic digester. Nevertheless, the findings suggest that the anaerobic digestion annual performance is significantly affected by the seasonality of ethanol production and start-up times.

5. ACKNOWLEDGEMENTS

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