Environmental Engineering and Management Journal

October 2014, Vol.13, No. 10, 2687-2695 http://omicron.ch.tuiasi.ro/EEMJ/



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LANDFILL LEACHATE RECIRCULATION. PART I: SOLID WASTE DEGRADATION AND BIOGAS PRODUCTION

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Abstract

Anaerobic degradation (AD) of municipal solid waste (MSW) depends on the moisture and nutrient distribution within the bioreactor; these factors are affected by leachate recirculation. In this work, the effects of leachate recirculation of 15 to 120 by volume percentage (%V) on AD were studied. The ojectives of this study are as follows: a) to determine which recirculation rate provides the best conditions for accelerated anaerobic degradation (AD) and b) to determine the optimal range of recirculation rates for methane (CH₄) generation. In the first group of experiments, denoted as the exploratory range, laboratory-scale bioreactors (LSBs) were operated at leachate recirculation rates of 15, 30, 60 and 120% V. In a second group of experiments (denoted as narrowed range), a group of seven LSBs were operated at rates of 40, 60 and 80% V, and seven were employed as control, without recirculation. In this stage, LSBs were periodically dismantled to allow testing of the digested MSW. The AD rate was monitored for 201 days along with other variables, including the total volatile solids, holocellulose, lignin, organic carbon, total nitrogen and pH of the MSW matrix, the characteristics of the produced and recirculated leachates and CH₄ production rates. The results indicated that methane production during the methanogenic fermentation stage is directly correlated with the recycling rates. The 120%V recirculation rate was observed to cause washout in the waste matrix. The suggested range on the basis of CH₄ generation per liter of recirculated leachate was 30 to 40% V.

Key words: biomethanation, leachate recirculation, municipal solid waste

Received: December, 2012; Revised final: June, 2014; Accepted: June, 2014

1. Introduction

Mexico produced 112,500 tons of municipal solid waste (MSW) per day in 2011, and up to 23% of that waste was not disposed of properly (INEGI, 2013). Currently, Mexican disposal sites that do comply with environmental norms (LGPGIR, 2013) do not always have systems for biogas collection or

appropriate systems for leachate treatment. According to the National Emissions Inventory of Greenhouse Gases in 2010, MSW in Mexico produced emissions of 22148 Gg CO_2e , which corresponded to 2.96% of the total national emissions and were three times the emissions due to MSW in 1990 (SEMARNAT-INECC, 2013).

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One option for reducing these emissions is to operate sanitary landfills (SL) as bioreactors and use methane (CH₄) as an alternative energy source (Pacey et al., 2000; Reinhart et al., 2002). The goal of accelerating the anaerobic degradation (AD) of organic matter (OM) in MSW through water addition and/or leachate recirculation was proposed over 30 years ago and is currently studied in various parts of the world (Abichou et al., 2013; Benson et al., 2007; Chugh et al., 1998; Francois et al., 2007; Pohland, 1975; Schiappacasse et al., 2010; Sponza and Agdag, 2004). Leachate recirculation promotes the uniform distribution of nutrients and microbial extracellular enzymes involved in AD through the matrix, buffers pH, dilute inhibitory compounds and spreads methanogenic bacteria through the reactor (Barlaz et al., 1990; Pohland and Kim, 1999; Sang et al., 2012). However, leachate recycling can also cause problems, such as the accumulation of volatile organic acids (VOA) and ammonia as nitrogen (NH3-N), flooding, washing and/or excessive compaction of the MSW matrix and landfill slope failure (Ellis et al., 2005; Reinhart et al., 2002; Reddy et al., 2009; Sponza and Agdag, 2004). Therefore, it is important to determine the appropriate recirculation rate and conditions to avoid potential adverse effects on methanogenesis and to reduce the MSW stabilization time (Francois et al., 2006; Pohland, 1975; Sri Shalini et al., 2010; Valencia et al., 2009).

Several reports have partially responded to the aforementioned challenges using a variety of approaches. There have been studies that investigated leachate recirculation using simple step systems (Pohland et al., 1992; Sponza and Agdag, 2004; Swati et al., 2005) or using a control system (El-Fadel, 1999; Mehta et al., 2002), corroborating the effectiveness of recirculation for enhancing AD. Recirculation frequency has also been the subject of several studies that varied the periodicity of liquid addition within different AD stages (Trankler et al., 2005; Sanphoti et al., 2006). Several other reports indicated the recirculation rate used but did not necessarily seek to compare different recirculation rates. Moreover, it is difficult to make an accurate comparison between recirculation rates because there are at least two ways to report the amount of fluid that is recirculated. It can be reported as a volume percentage (% V), which is defined as the leachate volume x 100 divided by the volume of the loaded residues, or as the wet basis moisture content percentage (% MC), which refers to the wet basis moisture content of the studied residues.

In this study, recirculation rates are reported as %V. Recirculation rates reported in the literature range from 2 to 43% V of loaded residues, and previous studies have used a variety of influents: leachate only (diluted, concentrated, young or mature), leachate mixed with sludge, neutralized leachate, water, etc. (Benbelkacem et al., 2010; Calabro et al., 2010; Chugh et al., 1998; Francois et al., 2007; Reddy et al., 2011; Yong-Jun et al., 2008; Zhou et al., 2012). Providing landfill operators with information regarding leachate recycling is vital, and data obtained with local conditions in countries where landfills are starting to be operated as bioreactors is needed. The scarcity of data can be addressed, as a first approach, by the use of laboratory scale studies. Although laboratory-scale research does not allow results to be implemented in the field (Fellner et al., 2008), it offers a variety of relevant advantages, as it permits (i) a systematic comparison among a wide range of leachate recycling rates, (ii) control of MSW characterization and variable follow-up, (iii) the inclusion of replicates in the study and (iv) a manageable timescale, and it is more economical than field-scale experimentation.

This study is the first part of a study aiming to test the performance of leachate recirculation rates from 15 to 120%V in laboratory scale bioreactors (LSB) to determine the best recirculation rate for MSW AD and CH₄ generation.

2. Materials and methods

2.1. Preparation of solid waste samples

A sample of approximately 50 kg of fresh refuse was collected from the landfill in the Pátzcuaro Municipality, Michoacán, (México). The sample was hand-sorted and separated into 15 waste categories, then reduced to a particle size ≤ 1 cm to prepare the mixture shown in Table 1.

2.2. Assembly of laboratory scale bioreactors (LSB)

Cylindrical LSBs with the dimensions 18 cm x 4.5 cm ID were constructed using schedule 40 PVC pipes. Each LSB was packed with 215.19 g of the MSW mixture described above (Table 1). A layer of 32.15 g of soil was added as cover material, and both were compacted to 600 kg/m^3 . The LSBs were sealed hermetically and flushed with oxygen-free N₂ to displace the air in the system. Each LSB was then connected to both the leachate recirculation and the biogas measuring systems (Fig. 1).

2.3. Operation of laboratory scale bioreactors

2.3.1. Leachate recirculation: exploratory range

Eight LSBs were operated at four different leachate recirculation rates (15, 30, 60 and 120 %V), as shown in Table 2, and two were used as controls without recirculation. Leachate recirculation began with the addition of tap water to each LSB in the amounts indicated in Table 2 to increase the waste moisture content. The first produced leachate was quantified and poured into a reservoir with enough water (Fig. 1) to make up 10 times the LSB volume (this was performed only once) to obtain enough leachate to operate at each recycling rate. Independent reservoirs were used for each recirculation rate. Table 2 shows the volume of leachate recirculated per MSW mass. Fig. 2 shows the operation protocol for leachate recirculation that was implemented twice per week. All LSBs were operated at room temperature ($23 \pm 2^{\circ}$ C), and all were dismantled on day 201, three days after the last recirculation.



Fig. 1. Leachate recirculation LSB: a) biogas measurement system b) biogas sampling point, c) leachate recirculation entry port, d) sieve dish distributor e) LSB body, f) produced leachate sampling point, g) peristaltic pump, h) water/diluted leachate reservoir

2.3.2. Leachate recirculation: narrowed range

After completing the exploratory range, a narrowed range (around 60%V) was selected. To study and compare leachate recirculation volumes, 28 LSBs (four groups of seven reactors) were operated in the narrowed range stage. During this stage, one LSB from each group was dismantled on days 15, 45, 75, 105, 135, 165 and 201. The leachate recirculation rates studied in this stage were 40, 60 and 80 %V, with recirculation volumes of 165, 248 and 330 ml, respectively. The fourth reactor set was operated under no recirculation and was used as a control group. Fig. 3 shows all recirculation rates tested during the exploratory and narrowed ranges in this study.

2.4. Analytical and statistical determinations

2.4.1. Solid waste analyses

The parameters shown in Table 3 were determined for MSW and soil.

2.4.2. Biogas analysis

Biogas generated in each LSB was measured by displacement of brine (pH=4) (Fig. 1a), and CH₄ content was analyzed twice per week using a gas chromatograph (Varian 3800) fitted with a stainless steel column (2 m x 2 mm ID) packed with HAYESEP Q 80-100 MESH. The temperatures for the oven, injector and detector were 90, 200 and 210 °C, respectively.

Table 1	. Waste	categories and	l quantities	for i	initial	MSW	mixture
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Waste estacom	М	SW
waste category	$\begin{tabular}{ c c c c c c } \hline & & & & & & & & & & & & & & & & & & $	g
Food residues	34.25	73.71
Yard trimmings	31.86	68.56
Paper	5.97	12.84
Plastic film	4.74	10.20
High density polyethylene (HDPE)	3.81	8.21
Glass	3.48	7.49
Rags	3.31	7.12
PETE (Polyethylene terephthalate)	2.67	5.75
Construction material	1.91	4.10
Disposable diapers	1.89	4.06
Ferrous material	1.76	3.78
Non-ferrous material	1.31	2.83
Waxed cardboard	1.21	2.60
Cardboard	1.07	2.30
Styrofoam	0.76	1.64
Total	100.00	215.19

%: w/w, weight/wet mass percentage

Recirculation rates (% V)	Water/leachate volume (ml)	Leachate recirculated per MSW mass (ml/kg)
15	62	288
30	124	576
60	248	1152
120	496	2305

Table 3. Mexican standard norms and methods used to characterize municipal solid waste

Parameter	Mexican standards norms and methods
pH	NMX-AA-25-1984 ^a
Moisture content	NMX-AA-016-1984 ^a
Total nitrogen	NMX-AA-24-1984 ^a
Organic carbon	Dichromate method ^b
Holocellulose	Chlorite method ^c
Lignin	ASTM D1106-56 modified method ^d
Total volatile solids	2540G method ^e

^a SEMARNAT, 2012 ^bWalkley and Black, 1982; ^c Wise et al., 1946; ^d Colín-Urieta et al., 2007; ^e APHA, 2005



Fig. 2. Operation protocol for leachate recirculation



Fig. 3. Diluted leachate recirculation rates in exploratory and narrowed ranges

Biogas samples (30 μ l) were collected manually from each bioreactor sample port (Fig. 1b) using a 50 μ l Hamilton® syringe. Methane and biogas data were analyzed statistically using ANOVA and Tukey's HDS test (95% significance) through the STATGRAPHICS Plus 5.0 software, considering the leachate recirculation rate as a variable control.

3. Results and discussion

3.1. Municipal solid waste characterization

The characterization of refuse and soil is shown in Table 4. For the fresh refuse used in this study, the total volatile solids (TVS), holocellulose (HOL), lignin (LIG) and organic carbon values were higher than those reported by other authors. This discrepancy is observed because OM (food residues, yard trimmings, paper, rags and cardboard) accounted for 76.46% of the MSW studied here (Table 1), which is common in rural Mexico (Buenrostro and Israde, 2003). The C/N ratio of the prepared MSW was close to 30 (Table 4), suggesting a suitable nitrogen (N) concentration.

The soil used as cover material neutralized the pH and reduced the OM content (expressed as TVS, HOL, LIG and C) of the loaded MSW in each LSB although it accounted for only 15% of the weight of the initial residues.

3.1.1. Waste anaerobic degradation in the exploratory range

The moisture content was progressively incremented until the field capacity was reached (approximately 65 %MC). The LSBs operated at 15, 30, 60 and 120 %V produced their first leachate at 231.7, 64.3, 2.82 and 2.10 h, respectively. It was found that, independent of the leachate recirculation rate, the final pH of the unloaded MSW was close to neutrality for all recirculating LSBs. For the control LSB, however, the final pH of the MSW was acidic, and other parameters showed that the AD of OM was carried out at lower rate (Table 5). The TVS removal data, as well as the HOL/LIG and C/N indexes of solid waste, reveal that the highest AD rate occurred at a 60 %V recirculation rate (Table 5). The solid waste HOL/LIG and C/N results at 120 %V showed lower AD levels, which can be

attributed to the washout of microorganisms and nutrients as a result of high recirculation volume (Sponza and Agdag, 2004). The leaching of nutrients could also explain the lower N concentration in the solid waste matrix at this recirculation rate.

Table 4. Comparison	between the measured	d characteristics of the	MSW tested her	re and previously	y reported values
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Bayamatans		Loaded components	Reported values	
Furumeters	Soil	MSW and soil	Fresh MSW	Fresh MSW
Moisture content (%MC)	33.2±0.35	38.96±0.42	40.69±0.51	43.5 ^{a;} 54.7 ^b
pH	6.66±0.03	6.56±0.07	5.65 ± 0.08	6.8 ^{b;} 6.9 ^c
TVS (%)	7.8±0.32	71.35±0.26	79.53±0.41	38.5 ^a ; 50.0 ^b ; 69.3 ^d ; 52.9 ^e
HOL (%)	1.81±0.12	56.91±0.46	61.77±0.37	52-62 ^f ; 50.38 ^g
LIG (%)	1.26±0.23	16.88±0.27	20.33±0.32	10-15 ^f ;19.96 ^g
HOL/LIG	1.44	3.37	3.04	2.52 ^g ; 3.0-4.0 ^h
C (%)	1.93±0.33	32.25±0.54	39.16±0.46	24.0 ^a ; 29.0 ^d ; 19.6 ^e ; 36.5 ⁱ
N (%)	0.10±0.03	1.15±0.14	1.31±0.16	1.63 ^a ; 1.2.0 ⁱ
C/N	19.3	28.04	29.89	71.8 ^d ; 30.42 ⁱ

Average ± standard deviation; ^a Valencia et al., 2009; ^b Elango et al., 2007; ^c Hernández-Berriel et al., 2008; ^d Francois et al., 2006; ^e Bareither et al., 2012; ^f Barlaz et al., 1989; ^g Hossain et al., 2003; ^h Barlaz et al., 1997; ⁱ Sri Shalini et al., 2010. % Related to the dry content.

D anamatans	Control	Leachate recirculation levels (%V)					
Furumeters	Control	15	30	60	120		
Moisture content (%MC)	37.34±0.39	64.92±0.49	65.37±0.45	66.06±0.44	64.91±0.46		
pH	5.47±0.04	6.98±0.03	7.14±0.04	6.92±0.04	7.98±0.04		
TVS (%)	63.51±0.35	56.97±0.47	55.97±0.44	54.88±0.30	56.71±0.47		
TVS removal (%)	10.99	20.15	21.56	23.08	20.52		
HOL (%)	54.18±0.35	41.53±0.37	33.13±0.42	29.85±0.34	36.69±0.38		
LIG (%)	16.69±0.26	27.74±0.32	29.41±0.29	31.16±0.35	28.11±0.30		
HOL / LIG	3.25	1.50	1.13	0.96	1.31		
C (%)	29.51±0.22	20.54±0.31	15.98±0.47	14.81±0.55	19.26±0.57		
N (%)	1.04±0.12	0.96±0.01	0.89±0.21	0.90±0.07	0.49±0.04		
C / N	28.38	21.39	17.96	16.46	39.41		

 \pm standard deviation

3.1.2. Waste anaerobic degradation in the narrowed recirculation range

As discussed above, the second part of the experiment used rates of 40, 60 and 80 %V, and reactors were periodically dismantled during this stage.

The recirculation of leachate maintained moisture content values between 63.5 - 67.8 % MC, which contributed to the AD of OM, and the pH surpassed neutrality after day 75 for recirculating reactors (Fig. 4). All recirculating reactors also reached TVS removal rates between 22.16 - 23.35% (Fig. 5). For the control LSB, pH values ranged between 4.88 - 5.47, and the MC decreased to 37.34 \pm 0.39 on day 201, suggesting that the AD process remained in the acidogenic phase (Mehta et al., 2002; Pohland and Kim, 1999).

Higher recycling rates were associated with greater TVS removal (up to day 165) (Fig. 5) for the narrowed range, confirming that, as reported by Benbelkacem et al., (2010), leachate perfusion is fundamental to the mass transfer between the solid and liquid phases and promotes greater nutrient availability. At the end of the experiment, however, similar TVS removal rates were detected in the 40,

60 and 80 %V LSBs. There are two potential explanations for this result. It could be attributed to the high initial LIG content in the MSW; lignin's complex structure could have interfered with the AD of HOL (Barlaz et al., 1997; Barlaz, 2006; Micales and Skog, 1997), preventing the attainment of lower C levels after 201 days. It could also be explained by the compaction of the waste matrix (data not shown) (Fei and Zekkos, 2013; Hossain et al., 2003; Reddy et al., 2011; Reinhart et al., 2002); the TVS removal rate was > 0.1% TVS removed per day until day 165 for the three recirculation rates, after this, the rate dropped to 0.07% TVS removed per day for the 80 %V reactor. This means that if the MSW includes a high fraction of yard trimmings (\geq 30%, w/w), different recirculation rates will not necessarily result in higher AD (Table 6).

3.3. Biogas and methane

3.3.1. Biogas and methane statistical analysis

Table 7 presents the results of the biogas and CH_4 Tukey's HDS test during the acidogenic and methanogenic fermentation phases, using a CH_4 concentration of 40 %v/v as a limit. In the control

LSB, no CH_4 production was detected during the experiment, most likely due to the low temperature and the lack of added moisture (Rajasekaran et al., 1986; Warith et al., 2005).

3.3.2. Methane and biogas production

In the acidogenic phase, CH₄ concentrations (%v/v) obtained from the LSBs operating at 15 and 30 %V were both similar (Fig. 6), as shown by the Tukey's HDS test (Table 7) and the estimated production rate in %v/v CH₄/d shown in Table 8. However, the establishment of CH₄ production was detected 14 days later for the LSBs operating at recirculation rates of 15 and 30 %V than for the other recirculation rates; CH₄ production was established on day 38 for the 15 and 30%V reactors and day 24 for the others. During the acidogenic phase, the cumulative biogas and CH₄ production (Figs. 7a and 7b) in the LSBs operating at 40, 60 and 80 %V were similar at day 52 (Table 7), possibly because TVS removal remained at less than 10% until day 45 for these three reactors (Fig. 5). The estimated rates of biogas and CH₄ accumulation in the LSB operating at 120 %V were higher than those in the other reactors, though this difference was not statistically significant (Tables 7 and 8).

In the methanogenic fermentation stage, lower CH₄ concentrations were detected in the LSBs operating at 15 and 120 %V than in the other reactors (Table 8). According to Tukey's HDS tests, the CH₄ concentrations at 15 and 120 %V were significantly different from those found at the other recirculation rates (Table 7). The accumulated biogas curves (Fig. 7a) for the LSBs operating at 30 to 80 %V were similar at this stage. Although the rate of accumulated CH₄ per day in the LSB operating at 30 %V was lower than in the ones operating at 40 to 80 %V (Table 8), the Tukey's HDS test indicates that this difference is not statistically significant (Table 7). With the exception of the LSB operating at 120 %V, the results for the cumulative production of biogas (Fig. 7a) and CH₄ (Fig. 7b) in the methanogenic fermentation stage showed a direct



Fig. 4. Solid waste pH dynamics for the narrowed recirculation range

correlation between biomethanation and recirculation rate (Table 8).

This relationship occurs because recirculation better distributes nutrients through the waste matrix, stimulating microbial activity (Chugh et al., 1998; Ravishankar et al., 2007; Warith et al., 2005). The decrease in biogas and CH₄ production observed in the LSB operating at 120 %V between days 130 and 140 suggests that this recirculation volume resulted in the dilution and wash-out of nutrients and microorganisms, producing free spaces in the waste matrix (Ellis et al., 2005, Hernández-Berriel et al., 2010; Sponza and Agdag, 2004).

The Tukey's HDS test did not identify any significant differences between the recirculation rates from 30 to 80 %V with respect to the cumulative production of biogas and CH₄ (Table 7). The production rates reported here are lower than that reported by Sanphoti et al., (2006) for leachate recirculation with the addition of supplemental water (54.87 l/kg MSW dry) and higher than that reported for leachate recirculation alone (17.04 l/kg MSW dry). This indicates that recirculation of diluted leachates is more effective because it avoids the presence of various compounds at toxic concentrations (Gerardi, 2003).

Although the highest cumulative CH₄ production was obtained at 80 %V (Table 8), this rate differed from that obtained at 60 %V by only 0.24 1 CH₄/kg MSW (dry basis), and the recirculation volume used was 0.624 l/kg MSW (dry basis). A recirculation rate of 80 %V would therefore not represent an economic advantage at the field scale. On day 201, the LSBs operating at recirculation rates of 30, 40, 60 and 80 %V had produced cumulative CH₄ volumes of 24.59, 19.72, 13.33 and 10.12 L CH₄/L recirculated leachate, respectively. Because of this result and considering the practical problems posed by handling large volumes of leachate at the field scale (Benson et al., 2007; Reinhart et al., 2002; Xu et al., 2013), the recommended recirculation range for MSW showing a composition similar to that of the Pátzcuaro landfill is 30 - 40 %V.



Fig. 5. TVS removal dynamics from solid waste for the narrowed recirculation range

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Table 6. Final characterization of solid waste for LSBs operating in the narrowed leachate recirculation range

Dayam stars	Le	achate recirculation levels (%	5V)
Furumeters	40	60	80
TVS (%)	55.54±0.43	54.88±0.30	54.69±0.40
TVS removal (%)	22.16	23.08	23.35
HOL (%)	30.46±0.38	29.85±0.34	29.36±0.41
LIG (%)	30.30±0.39	31.16±0.35	31.89±0.38
HOL / LIG	1.01	0.96	0.92
C (%)	14.43±0.61	14.81±0.55	13.92±0.42
N (%)	0.89±0.13	0.90±0.07	$0.87{\pm}0.08$
C/N	16.21	16.46	15.99

Average values \pm standard deviation

Table 7. Statistical analysis of biogas and CH₄ production (%v/v) for the acidogenic and methanogenic phases

Phases	CH ₄ concentration (%v/v)	Accumulated biogas (l/kg MSW*)	Accumulated CH ₄ (l/kg MSW*)
Acidogenic phase	No ≠	No ≠	No ≠
	15-30	15-30	15-30
Methanogenic fermentation	15-40	15-40	15-40
	15-60	15-60	15-60
	15-80	15-80	15-80
	30-120		30-120
	40-120		40-120
	60-120		60-120
	80-120		80-120

No ≠: *The values were statistical similar in all recirculation rates, MSW*: MSW dry basis*

Table 8. Flow rates of methane and biogas production in LSBs

		Leachate recirculation rates (%V)						
Phase / Parameter	15	30	40	60	80	120		
Acidogenic phase (days)	38-66	38-66	24-48	24-48	24-48	24-48		
CH_4 concentration/d (%v/v / d)	1.025	1.193	0.996	1.015	1.008	1.187		
Accumulated biogas/d (l/kg MSW d)	0.089	0.163	0.074	0.077	0.093	0.185		
Accumulated CH ₄ /d (l/kg MSW d)	0.018	0.036	0.018	0.020	0.039	0.043		
Methanogenic fermentation phase (days)	69-201	69-201	52-201	52-201	52-201	52-201		
CH_4 concentration (%v/v)	49.43±3.37	58.39±3.32	58.70 ± 3.91	60.81 ± 4.18	58.80 ± 3.91	44.29 ± 9.17		
Accumulated CH ₄ / day (l/kg MSW d)	0.113	0.198	0.216	0.220	0.230	0.171		
Accumulated biogas/d (l/kg MSW d)	0.223	0.334	0.364	0.350	0.371	0.307		
Accumulated CH ₄ , day 201 (l/kg MSW ^a)	15.26	23.21	24.77	25.17	25.41	17.07		

^a: dry basis



Fig. 6. Methane concentration dynamics in biogas

4. Conclusions

The 60%V was the recirculation rate that generated the highest anaerobic digestion of the MSW. A narrowed recirculation range of (40, 60 and 80%V) was therefore selected.



Fig. 7. Cumulative production: a) Biogas, b) Methane

The recirculation range within 15 to 80%V was positively correlated with both solid waste biodegradation and methane generation. The 120%V regime impacted negatively in biodegradation and biomethanation.

Difference in methane production rates obtained in the 30, 40, 60 and 80 %V regimes suggest the 30 to 40%V as recommended recirculation rate. Future research will include pilot studies at recirculation rates of 15 to 60 %V with the goal of establishing practical field applications

Acknowledgements

The authors gratefully acknowledge the generous funding of the National Council of Science and Technology (CONACyT) in México by means of grant 62776, and project 47187. The funding of COECyT Michoacan is also acknowledged. Finally, all comments and suggestions made by the referees are fully appreciated.

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