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STUDY ON AMMONIA STRIPPING PROCESS OF LEACHATE FROM THE PACKED TOWERS

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Abstract: About 245 thousand tones of municipal solid waste are collected daily in Brazil. Nearly 32 thousand tones of the collected amount are treated in sanitary landfill, which generates biogas and leachate as byproduct. The leachate resulting from sanitary landfill contains high concentration of carbonaceous and nitrogenized material. The crucial question is that the biodegradation of the carbonaceous material is difficult as long as the nitrogenized material is present in the form of ammoniacal nitrogen (NH_4^+), which compromises performance of biological treatment process. Therefore, a physical and chemical treatment of the leachate should be done before its biological treatment, especially for reduction of ammoniacal nitrogen concentration and for propitiating the realization of application of biological treatment. The treatment of leachate requires specific consideration, which is not needed for other types of waste. In the specific case in this study, where ammoniacal nitrogen concentration was about 2,200 mgN L^{-1} and the BOD_5/COD ratio was 0.3, the study of ammonia stripping process was performed. Ammonia stripping process was studied in packed towers of 35 L capacity each and the parameters investigated were pH, ratio of contact area/leach volume and the aeration time. One of the parameters that influenced most in efficiency of ammonia stripping process was pH of the leachate since it contributes in conversion of ammoniacal nitrogen from NH_4^+ to NH_3.

Keywords: leachate, ammonia, pH, packed tower, stripping.

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INTRODUCTION

About 240 thousand tons of municipal solid waste is collected in Brazil daily. Major part of the collected amount is still deposited in dumping grounds and only 13% of the total collected, which is equivalent to 31.2 thousand tons, is deposited in sanitary landfill. The sanitary landfill should be regarded as a final disposal process of municipal solid waste, which still deserves better dimensioning because of local, regional and national specificity, besides its physical and chemical composition and the rate of per capita production of the municipal solid waste. In the present situation, in which all the types of municipal solid wastes are deposited in sanitary landfills and that the most representative fraction of this waste is fermentable organic material, the biodecomposition process that is initially aerobic and then passes to be anaerobic, generates by-products that require to be quantified, characterized and submitted to due treatments. Among the by-products resulting from the biostabilization process of fermentable organic material, the biogas and leach deserve special attention regarding the impact that could be created on the environment. In case of the leach, the amount produced in sanitary landfill basically depends on climatic and hydrogeological conditions of the region, on characteristics of the waste and on operational conditions of landfilling. As regards to physicochemical composition, the leachate presents elevated concentration of COD, of volatile fatty acids and of ammoniacal nitrogen (Onay *et al.,* 1989). Principal chemical characteristics of leachate from organic solid waste treated in anaerobic batch reactors are presented in **Table 1**.

Analyzing the data presented in **Table 1**, it can be verified that the leachate generated by anaerobic biostabilization process of organic solid waste presents acid characteristic and high concentration of carbonaceous material. This tendency can be explained by the characteristics of the organic solid waste, which is different from municipal solid waste, as it does not contain a significant percentage of constituents of inorganic nature. Taking in to consideration that the organic solid waste used in this work is basically constituted of vegetable waste and that at 330 days of functioning, the leachate produced still presented acid characteristic with pH equal to 4.6. The ratio of volatile acids concentration/total alkalinity during the first 30 days of functioning remained in the range of 4.8, and then gradually reduced, attaining to the level of 2, which means that the increase in total alkalinity concentration was not proportional to reduction of volatile fatty acid concentration. As regards to the carbonaceous material, a reduction of 5.5% of COD and 63% of TVS and a significant increase in concentration of total Kjeldahl nitrogen was verified.

The quantitative of leachate liquid produced in sanitary landfill, depends upon a series of factors, among them which can be pointed out are the meteorological conditions, geology and geomorphology, the operational conditions of the landfill and physical characteristics of the waste. The estimate of leachate liquid produced in sanitary landfill can be obtained by Swiss method or by *hydric* balance method. Swiss method uses empirical coefficients; but does not present a good accuracy level. It is basically in function of average annual precipitation occurred in the landfill area, of the area of landfill and of compactness of the waste. The hydric balance method produces more precise results and is function of infiltration of landfill originated only of incident precipitation, of characteristics of the waste, of the covering material used, of evaporation and of surface flow.

The leachate treatment can be realized by using differrent technological alternatives which are generally classified in three major groups: "*in situ*" treatment, which consists of equipments and internal units limiting up to the boundary of the landfill itself; conjugated treatment of leach with sanitary sewage; physicochemical treatment followed by biological treatment, or a combination of different technological alternatives (Roberts *et al.*, 1985).

One of the major problems faced by the leach treatment is related to high concentrations of ammoniacal nitrogen. An alternative, which can be used for resolving this problem, is the application of the process of ammonia stripping (Sletten *et al.*, 1997). According to MetCalf & Eddy (2003) the sweeping of ammonia with air is a modification of the aeration process used for elimination of hydrosulphuric gases in water.

The stripping of ammonia is a physical process of removal of gaseous phase from the liquid, principally due to elevation of total contact surface of liquid phase with the surrounding (atmospheric) medium, so that the effects of dragging and molecular diffusion promote its passage for the last (Ozturk *et al.*, 1999). The removal process of free ammonia from the liquid medium causes dislocation of equilibrium in the direction of its formation. Ammonia, in aqueous phase, is found in equilibrium of two forms, which are the ionic (NH_4^+) and the gaseous molecular (NH_3). The equilibrium of conversion of ammonium ion for ammonia is shown by **Eq. (1)**.

$$NH_4^+(aq) + OH^- \leftrightarrow NH_3(g) + H_2O(aq) \quad (1)$$

Table 1. Chemical parameters of leachate originated from the organic solid waste in function of biostabilization time

Time (days)	pH	TA (mg $CaCO_3$ L^{-1})	VFA (mg HAc L^{-1})	COD (mg L^{-1})	KTN (mg L^{-1})	VS (mg L^{-1})
30	4.2	2875.0	13 500.0	36 169.0	829.0	34 892.0
90	4.2	2000.0	16 500.0	37 382.0	941.0	41 134.0
150	4.0	2350.0	14 040.0	47 995.0	1155.0	32 730.0
210	4.3	2408.0	17 570.0	47 692.0	2038.0	22 152.0
270	4.7	5500.0	10 650.0	31 621.0	1994.0	30 108.0
330	4.6	3625.0	7 138.0	34 090.0	1557.0	12 892.0

Where: TA is Total Alkalinity; VFA is Volatile Fat Acids; COD is Chemical Oxygen Demand; KTN is Kjeldhal Total Nitrogen; VS is Volatile Solids.
Source: Lopes (2000).

The equilibrium of the conversion process depends on pH, and for pH around 7.2 the tendency is that the equilibrium is shifted to the left. With an increase in pH, there is a shift in equilibrium for the right and consequently a higher elevation of the gaseous fraction (Muniz *et al*., 1989).

As the equilibrium of concentration of ammonium ion and of ammonia gas depends on pH, the percentage distribution of ammonia and ammonium ion can be determined by using **Eq. (2)**.

$$\%NH_3 = \left(\frac{[NH_3]}{[NH_3]+[NH_4^+]}\right)100 \qquad (2)$$

The quantity of air required for extracting ammonia from residual water is determined by mass balance equation at stable equilibrium state for a sweeping tower (Kyosai *et al*., 1991), as described by **Eq. (3)**.

$$G(y_2 - y_1) = L(x_2 - x_1) \qquad (3)$$

where G is moles of affluent gas per unit of temperature (K); L is moles of affluent liquid per unit of time (s); y_1 is solute concentration in gas in lower part of the tower (moles of solute/mole of gas); y_2 is solute concentration in gas in upper part of the tower (moles of solute/mole of gas); x_1 is solute concentration in liquid in lower part of the tower (moles of solute/mole of liquid); x_2 is solute concentration in liquid in upper part of the tower (moles of solute/mole of liquid).

MATERIALS AND METHOD

Experimental work was performed at Environmental Sanitation Laboratory of Department of Chemistry, CCT/UEPB. An experimental system basically composed of the following components was constructed for realization of the experimental part: (1) 4 stripping towers of 35.3 L capacity each; (2)1 reservoir for stocking the *in natura* leachate; (3) 1 device for pH correction of the leachate; (4) 1 leachate feeding system; (5) 1 air feeding system.

Schematic diagram of packed towers used for realization of ammonia stripping process is demonstrated in **Fig. 1**. The work was performed in triplicate for the four treatments. Operational parameters applied to the packed towers are presented in **Table 2**.

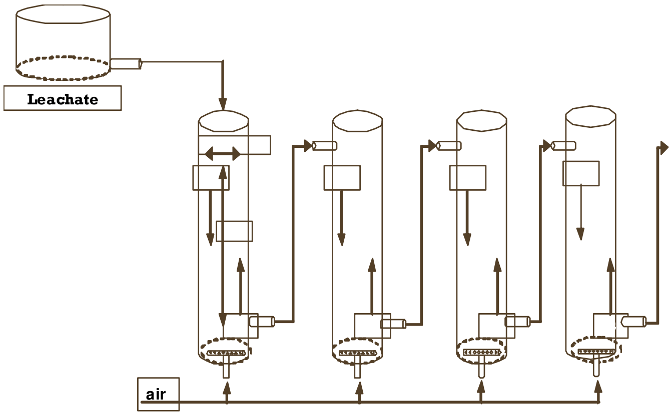

Fig. 1. Schematic diagram of packed towers.

Table 2. Operational data applied to the packed towers

Parameter\ Treatment	V_T (L)	V_B (L)	V_L (L)	Q_G (m^3/h)	t (hours)	pH
1	35.3	29.0	10.0	6.3	5.5	8.0
2	35.3	29.0	10.0	6.3	5.0	9.0
3	35.3	29.0	10.0	6.3	4.0	10.0
4	35.3	29.0	10.0	6.3	3.0	11.0

V_T: volume of tower; V_B: volume of grit; V_L: volume of leachate; t: time; Q_{ar}: air flow rate

82.1% of the total volume of the towers was filled with grit nº 4 leaving 49% of the space empty, resulting thus an available volume of 20.5 L per tower. On an average, only half of the useful volume available of the tower was occupied with leachate. The aeration process causes a significant formation of foam and consequently dragging a part of the leachate. The functioning and monitoring of the experimental system consisted in ascendant feeding of air and descendent of the leachate, with sample collecting of the final effluent at frequency of 30 min. for determination of monitored analytical parameters: pH, ammoniacal nitrogen, volatile fatty acids and total alkalinity.

Leachate used for realization of the experimental work was collected from the metropolitan sanitary landfill of João Pessoa city, capital of Paraíba state in northeast region of Brazil. The leach was collected *"in natura"* and transported in tank to Environmental Sanitation Laboratory of Department of Chemistry, CCT, UEPB, which is at a distance of 130 km from the sanitary landfill. The leach was stocked in closed tanks of hard PVC and distributed daily for equalization and pH correction tanks and for feeding towers. The pH correction was done by using the chemical species sodium hydroxide and calcium hydroxide, with the objective of estimation of cost of processing of ammonia stripping. The analytical determinations were done taking in to consideration the methods given by APHA (1998).

RESULTS AND DISCUSSION

The data resulting from chemical characterization of leachate used for realization of tests of ammonia stripping process in four different treatments are presented in **Table 3**.

Table 3. Data of chemical characterization of the leachate

Parameter	Magnitude	Unit
pH	8.0	-
Total alkalinity	14 291.0	$mgCaCO_3\,L^{-1}$
Volatile fatty acids	5 907.0	$mgH_{AC}\,L^{-1}$
COD	25 478.0	$mg\,L^{-1}$
BOD_5	3 760.0	$mg\,L^{-1}$
KTN	4 881.0	$mg\,L^{-1}$
Ammoniacal nitrogen	2 738.0	$mg\,L^{-1}$
TS	25 490.0	$mg\,L^{-1}$
VS	11 884.0	$mg\,L^{-1}$
SS	644.0	$mg\,L^{-1}$
VSS	490.0	$mg\,L^{-1}$
DS	24 846.0	$mg\,L^{-1}$
DSV	11 394.0	$mg\,L^{-1}$
DSF	13 452.0	$mg\,L^{-1}$

Analyzing the data demonstrates in **Table 2** it can be verified that the leachate collected in sanitary landfill of João Pessoa City presented elevated concentration of ammoniacal nitrogen and low BOD_5/COD ratio, which contributes negatively for application of any alternative of biological treatment. It can also be verified that the leachate is basically composed of 2.5% total solids and 97.5% water, the value which is 25 times higher than that of municipal sewage. One other particularity that deserves to be pointed out is the dissolved solids that represent 72.1% of fraction of total solids, the value sufficiently higher than that of the municipal sewage. As regards the nitrogenated material, it is verified that the concentration of ammoniacal nitrogen in the leach was on an average 54 times higher than in municipal sewage, which justifies the necessity of application of the process of ammonia stripping for reduction of this concentration to a suitable for feasibility of biological treatment.

Profile of variation of the parameters pH, ammoniacal nitrogen, volatile fatty acids and total alkalinity in relation to treatment 1, in which the leachate was adjusted pH to 8.0, is demonstrated in **Figs 2** and **3**.

Analyzing the behavior of variations, it is observed that the average concentration of ammoniacal nitrogen of the leachate used in treatment 1 was initially 2050.0 mg N L^{-1} and was reduced for values lower than 100 mg L^{-1} on 7 hours of aeration. Along with the decrease in concentration of ammoniacal nitrogen, an increase in pH values was verified. The stripping process drags some chemical species as well from the leachate. In the ammonia stripping process the ammonium ion converts in ammonia, thus consuming alkalinity, which justifies the reduction of the initial concentration of 15 100.0 mg $CaCO_3$ L^{-1} to 7900.0 mg C_aCO_3 L^{-1}. As regards the volatile acids, the initial concentrations varied in the range 1128.0 to 2715.0 mg N L^{-1} and after the functioning period the residual concentrations varied in the range of 96.0 to 1086.0 mg N L^{-1}. The reduction of concentration of volatile fatty acids is favored by dragging of determined chemical species present in the liquid medium that are detainers of slightly acid characteristic. In the specific case of this treatment, in which the stripping process of ammonia was realized in the leachate at pH 8 and at this pH, only about 5.3% of ammoniacal nitrogen is found in unionized form, which requires a greater time interval for converting a more representative parcel of ammoniacal nitrogen in the unionized form and consequently taken out of the liquid medium. The according Metcalf & Eddy (2003), a theoretical relation established between the concentrations of nitrogen species in function of pH can be determined by **Eq. (4)**.

$$pH = 9.25 - \log f + \log\left(\frac{[NH_3]}{[NH_4^+]}\right) \quad (4)$$

where f is ionic coeficiente actividade.

Profile of variation with passage of time of the parameters pH, ammoniacal nitrogen, volatile fatty acids and total alkalinity, for the three different tests and the arithmetical mean of the tests in relation to treatment, is demonstrated in **Figs 4** and **5**. In this treatment, the leachate pH was corrected to 9 and the monitoring time was stipulated in 5.5 hours.

In treatment 2, average concentration of ammoniacal nitrogen was reduced from 1582.0 to 113.0 mg N L^{-1} in a period of 5 hours, propitiating the reduction efficiency to 93%. Analyzing the profile of ammoniacal nitrogen concentration presented in **Fig. 3**, it is verified that in the first four hours of functioning, in all the tests realized, the leachate already presented low concentration of ammoniacal nitrogen. As regards the pH, it is verified that there was a striking increase, passing the level from 9.2 to 10.4. The average concentration of volatile fatty acids was reduced from 1128.0 to 192.0 mg $H_{AC}\,L^{-1}$, achieving the reduction efficiency of 82.9%. The total alkalinity concentration was reduced from 19 800.0 to 5200.0 mg $CaCO_3\,L^{-1}$ and the reduction efficiency, which was 73.7% is directly associated to the process of ammonia stripping. The profile of variations of parameters monitored in treatment 3 is presented in **Figs 6** and **7**.

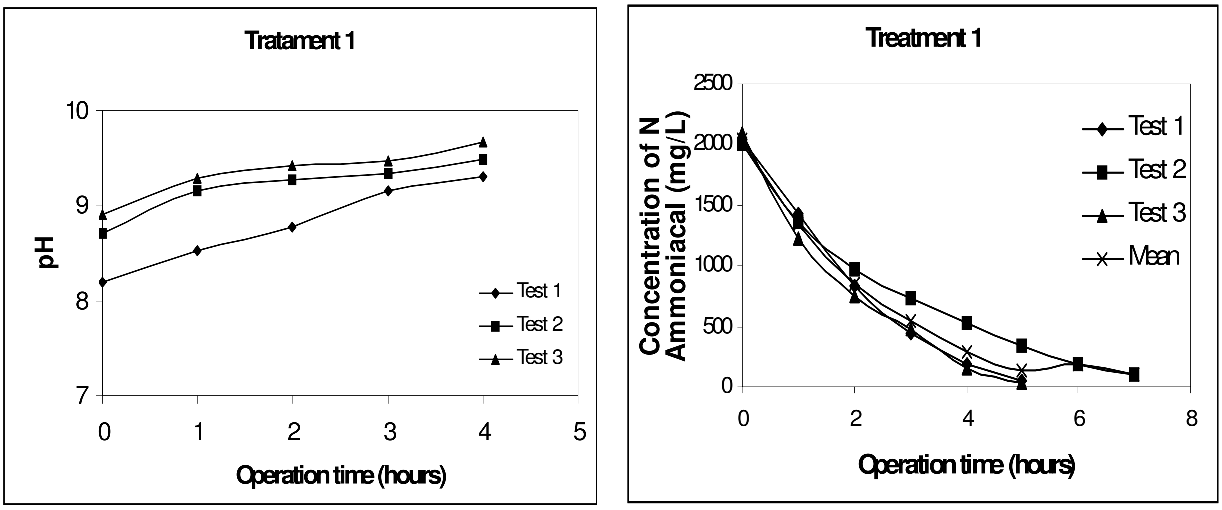

Fig. 2. Profile of the pH in and concentration of nitrogen ammoniacal in treatment 1.

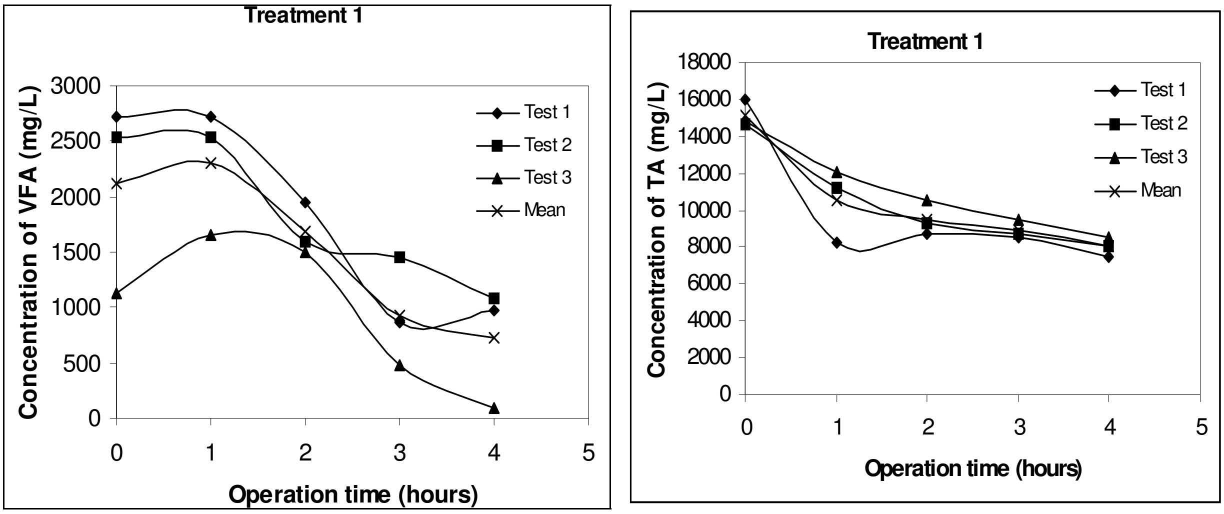

Fig. 3. Profile of the VFA in and TA in treatment 1.

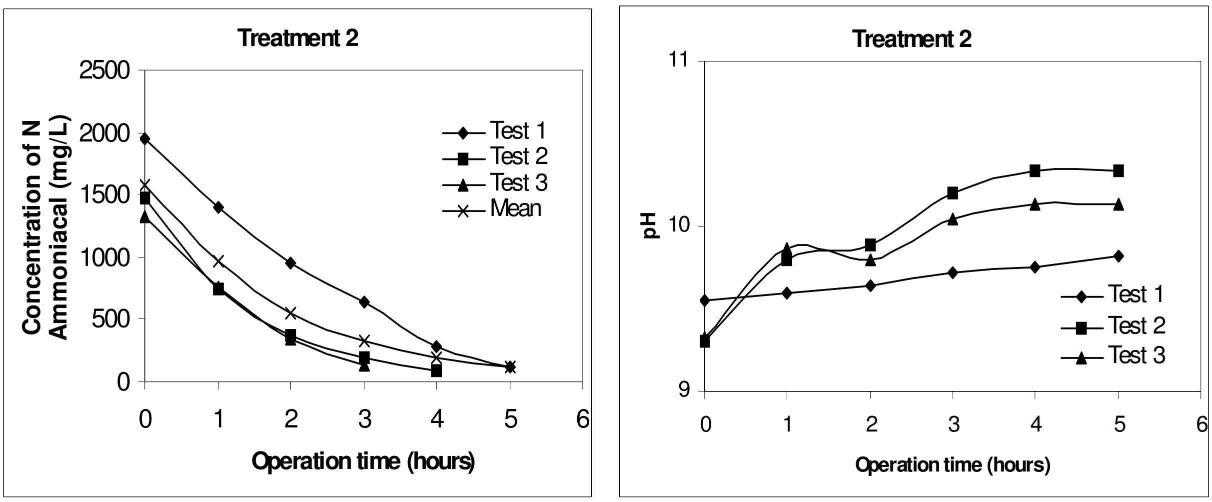

Fig. 4. Profile of the pH in and concentration of nitrogen ammoniacal in treatment 2.

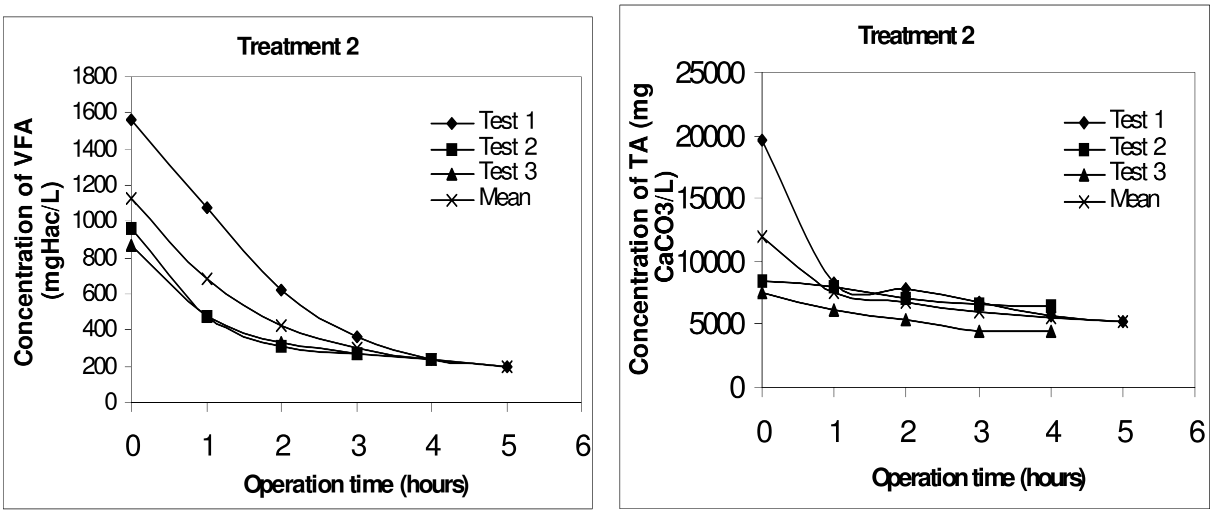

Fig. 5. Profile of the VFA in and TA in treatment 2.

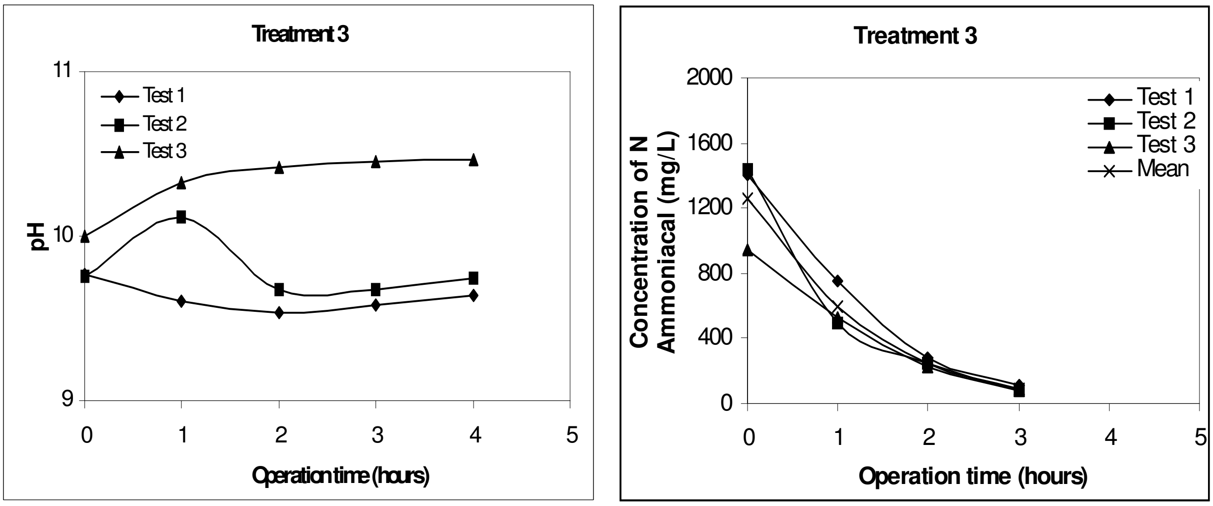

Fig. 6. Profile of the pH in and concentration of nitrogen ammoniacal in treatment 3.

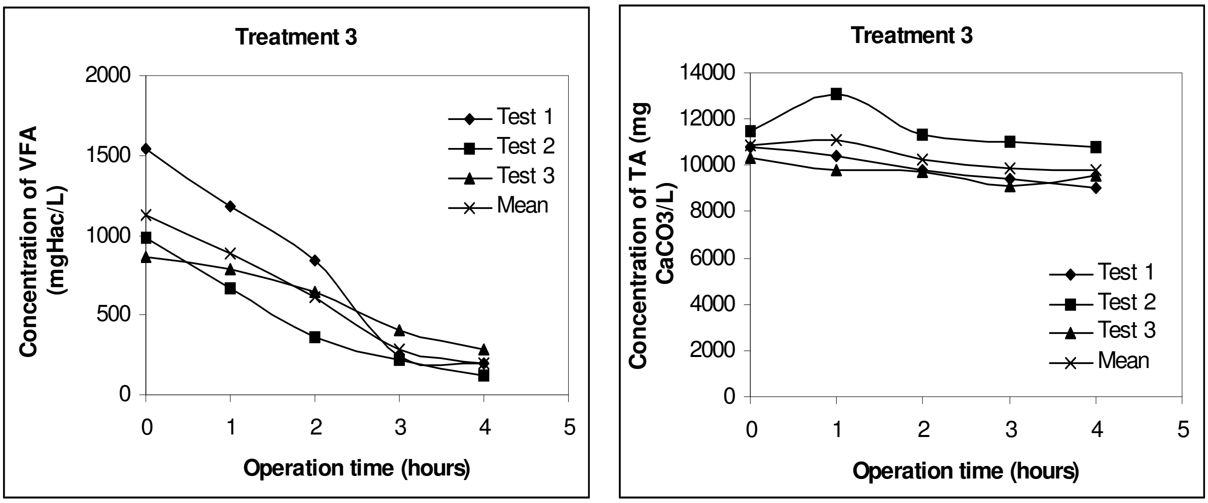

Fig. 7. Profile of the VFA in and TA in treatment 3.

The leachate pH was corrected to 10 in treatment 3 and the concentrations of ammoniacal nitrogen of the leachate used in the tests varied from 945.0 to 1399.0 mg L^{-1}. This variation of ammoniacal nitrogen concentration should be due to the time for which the leachate passed at the corrected pH 10, where a part of the ammoniacal nitrogen was converted to ammonia and liberated from liquid medium before being loaded to the towers. It is found that with three hours of functioning, the average ammoniacal nitrogen concentration was reduced from 1424.0 to 67.0 mg L^{-1}, with reduction efficiency of 95.4%, the value which is slightly higher that presented by treatment 2. For pH around 10, not any significant increase in pH is observed, considering the initial pH level of 9.7 finally reached to 10.4. The concentration of volatile acids was reduced significantly, passing from 1536.0 to 192.0 mg H_{Ac} L^{-1} with removal efficiency of 87.5% in the first test. The removal efficiencies for the other tests were 75 and 86%. As regards the total alkalinity, low reduction efficiency can be noted for all the realized tests. The profile of variations of the parameters and their respective average values for treatment 4 is presented in **Figs 8** and **9**.

For the leachate with pH around 11, the initial concentration of ammoniacal nitrogen varied from 1229.0 to 1607.0 mg N L^{-1} and after the monitoring period of three hours, the residual concentration of ammoniacal nitrogen varied from 56.0 to 70.0 mg N L^{-1}, with an average reduction efficiency of 95.5%. In relation to the pH, a small decrease is observed in all the tests, stabilizing around 10 pH, which did not occurred in the previous treatments. The reduction efficiency of volatile acids concentration was about 68.6%, where as the total alkalinity concentration for this level of pH presented small oscillations in all the tests. Estimation of cost of ammonia stripping process from leachate in filling towers is presented in **Table 4**. For realization of cost estimation, the price of the chemical species used for pH correction (sodium hydroxide and lime) and the cost of residential electric energy charged by the local electricity company ENERGISA are taken in to consideration.

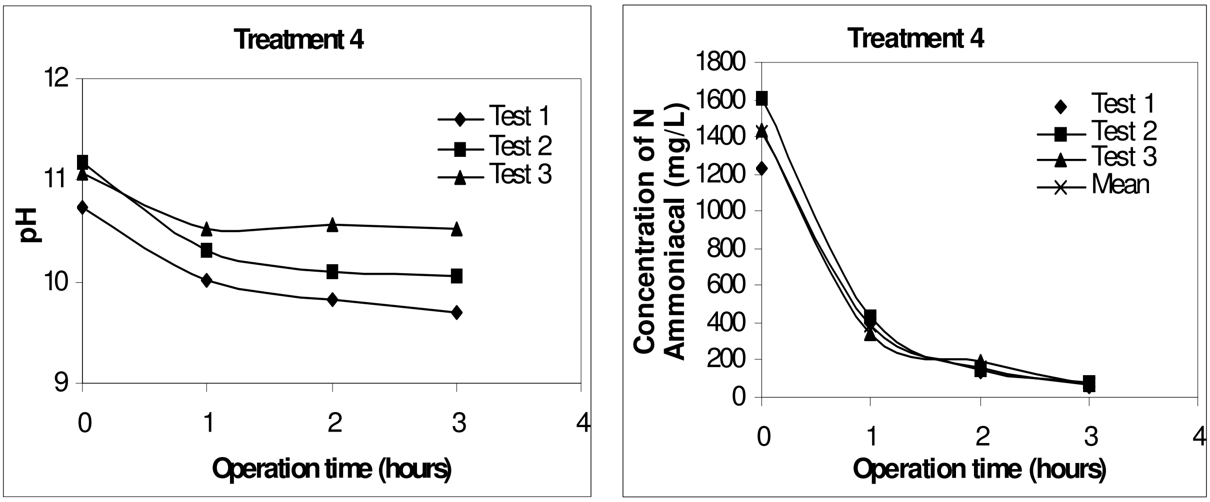

Fig. 8. Profile of the pH in and concentration of nitrogen ammoniacal in treatment 4.

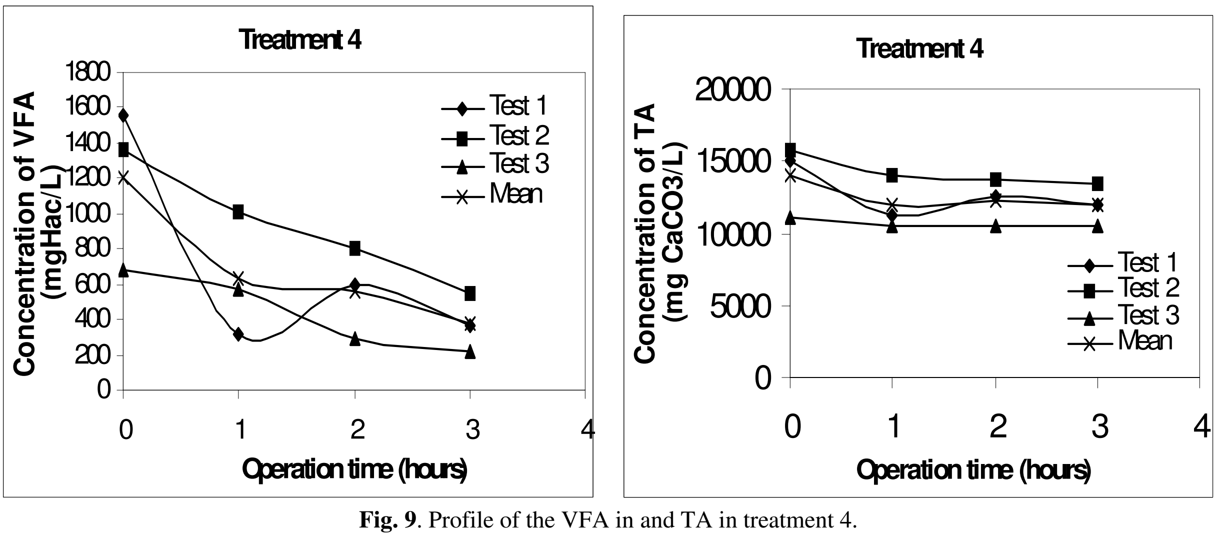

Fig. 9. Profile of the VFA in and TA in treatment 4.

Table 4. Cost estimate of ammonia stripping process of the leachate

pH	8.0		9.0		10.0	
Chemical species	NaOH (g m^{-3})	$Ca(OH)_2$ (g m^{-3})	NaOH (g m^{-3})	$Ca(OH)_2$ (g m^{-3})	NaOH (g m^{-3})	$Ca(OH)_2$ (g m^{-3})
Concentration	91.0	141.0	1.2	4.9	2.9	5.8
Cost (US$$/m^3) of chemical	0.50	0.03	6.80	1.30	15.70	1.58
Cost (US$/m^3) of electric energy	28.75	26.25	18.33			
Total cost (US$/m^3)	29.25	28.80	33.00	27.58	34.00	19.90

Analyzing the data presented in **Table 3**, it is observed that for pH 8, the costs estimated for realization of ammonia stripping process of leachate, varied proportionally to the time of aeration of the towers and on the chemical species used for pH correction. When the chemical specie used was sodium hydroxide, the total cost reached to US$ 29.25 and for the chemical specie lime the total estimated cost was US$ 28.80 per cubic meter leachate. It is interesting to point out that for this pH level, 98% of the estimated total cost, correspond to electrical energy. In general, it was verified that even if using lime as chemical specie for pH elevation, in specific case of this work, for cubic meter of leachate, 5.8 kg lime was consumed, corresponding to a total cost of US$ 19.90, which makes the ammonia stripping process technically and financially impracticable.

CONCLUSION

Referring to data analysis results of this work, one can conclude that: Ammonia stripping process occurs with higher efficiency in medium with pH higher than 10. However, the costs associated with the elevation of pH from 7.8 to 10.0 are of US$ 1.58 per cubic meter of leachate when the alkalinizing specie was lime and of US$ 15.70 per cubic meter of leachate when the alkalinizing specie was sodium hydroxide.

The removal efficiency of ammoniacal nitrogen in packing towers was higher than 90% for all the treatments realized. However, the aeration time varied from 3 to 5.5 hours so that the leachate remains with ammoniacal nitrogen concentration at about 100 mg L^{-1}. As the inherent costs of aeration of the process are directly proportional to the aeration time, therefore, higher the pH elevation, lower the cost of aeration and greater with that of the alkalinizing specie.

In the specific case of this work, the costs were estimated for three pH levels, two alkalinizing chemical species and aeration time varying from 3.5 to 5.5 hours. It was verified that for the leachate at pH 10, with ammoniacal nitrogen concentration of 1411.0 mg N L^{-1} reduced to 125 mg N L^{-1}, with aeration time of 5.5 hours, will spend about US$ 19.90 per cubic meter of leachate when lime was used as alkalinizing specie and US$ 34.00 per cubic meter when sodium hydroxide was used as alkalinizing specie.

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