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Coagulation-Flocculation and Air Stripping as a Pretreatment of Young Landfill Leachate

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Abstract: Leachate from sanitary landfills is a strong wastewater in terms of organic matter and ammonia. Both biological and chemical processes can be used for organic matter but ammonia reduction by nitrification-denitrification often poses problems due to inhibition. In this study, leachate from solid waste landfill in Konya Municipal area was used and organic matter and color removal were examined by using different chemical matters (Alum, $FeCl_3$, $FeSO_4$) in the pH adjustment with lime, NaOH and H_2SO_4. For ammonium removal, air stripping and its removal efficiency were investigated. For organic matter removal, the highest efficiencies were achieved as 44% by using 9 g/L alum at pH 11 adjusted with lime and as 45% by using 15 g/L $FeCl_3$ concentration at pH 3. Color removal studies in coagulation experiments indicated that the highest color removal efficiencies could be obtained when pH was adjusted with lime. The lowest coagulant concentration 1 g/L yielded the similar removal efficiencies as the concentration increased. In ammonium removal with air stripping, the optimum flow rate was 1 L/min and the optimum aeration time was 8 hours.

Keywords: Solid waste, landfill leachate, pre-treatment, coagulation-flocculation, air stripping.

INTRODUCTION

The degradation of the organic fraction of the municipal solid waste in landfill in combination with the percolation of rainwater produces a liquid called leachate [1-3]. One of the main problems with the solid waste landfill sites is leachate depending on large amounts of organic matter, ammonia-nitrogen, heavy metals, clorinated organic and inorganic salts.

Leachate is classified as young and old according to the age of the landfill site. Young leachate is characterized by chemical oxygen demand (COD) concentration higher than 5 g/L and by low nitrogen concentration (<400 mg/L of N). Old leachate is characterized by a high nitrogen concentration (>400 mg/L of N), high content of recalcitrant compounds and low biodegradable organic fraction (BOD_5/COD $\leq$ 0.1) [4, 5].

Usually a combination of physical, chemical and biological methods are used for the treatment of landfill leachate since it is difficult to have efficient treatment by one of these methods alone [6, 7]. Aerobic, anaerobic and anoxic processes are the biological methods for leachate treatment and are usually used in combination [8-10]. Air stripping and adsorption are major physical methods whereas coagulation-flocculation, chemical oxidation are chemical treatment methods [7, 11, 12].

In water and wastewater treatment, coagulation and flocculation are used for removing high concentration organic pollutants, heavy metals and some anions [13-15]. Alumunium and iron coagulants are effective for the removal of humic substances [16-19].

Coagulation-flocculation is a relatively simple technique that may be employed successfully for the treatment of older landfill leachate [17]. However, this method may result in only moderate removals of COD (or TOC) content, apart from presenting a number of drawbacks: excessive sludge may be produced, and in certain cases, when the conventional chemical coagulants are used, increased aluminium and iron concentrations may be encountered in the resulting effluent [12]. Coagulation-flocculation has thus been proposed mainly as a pretreatment method for young leachate or as a post-treatment technique for the partially stabilized leachate [20]. Aluminium sulphate (alum), ferrous sulphate, ferric chloride and ferric chlorosulphate are commonly used as coagulants. Iron salts seem more efficient than aluminium ones [17].

Removal of nitrogen from wastewater can be accomplished through a variety of physicochemical and biological processes. Biological nitrification and denitrification are considered to be the most practical methods of treating high nitrogen in leachate. But high concentrations of organic matter, eventhough have been removed in anaerobic stages at high efficiencies, may hinder high degree of nitrification. High concentration of ammonia, inhibitors such as heavy metals, sulfides etc. which are of common existance in leachate also decrease nitrification efficiency. In such cases, physico-chemical treatment processes are more suitable [21]. One of those processes is air stripping. Collivignarelli *et al.* [22] studied ammonia removal from landfill leachate having 2100 mg/L ammonia. Experiments were carried out at 70 ^{0}C and at pH 11 and the removal efficiency was 90%. Cheung *et al.* [23] studied ammonia removal for two different flow rates (1 L/min, 5 L/min). at 20 ^{0}C and at pH 11. Results revealed that after 24 hour-aeration, 81% and 90% removal efficiencies could be obtained for flow rates of 1 L/min and 5 L/min, respectively.

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Table 1. Characterization of Landfill Leachate

Parameters	Concentrations	
	Rainy Season	Dry Season
COD (mg/L)	7676-8320	39580-55450
BOD_5 (mg/L)	4050	36625
NH_4-N (mg/L)	896-1122	828-952.5
PO_4-P (mg/L)	35.18-38.8	9.52-11.5
SS (mg/L)	1400-2690	550-1500
Color (Pt-Co)	4280-5160	2961-14340
Ph	7.00-7.62	6.81-7.17
Pb (mg/L)	0.204	
Cd (mg/L)	0.118	
Zn (mg/L)	0.177	
Ca (mg/L)	139.5	
Mg (mg/L)	698	
Fe (mg/L)	7.27	
Ni (mg/L)	0.385	
Cr (mg/L)	0.661	
BOD_5/COD (mg/L)	0.44-0.8	

The aim of this study is to examine the efficiency of the coagulation-flocculation process as a pre-treatment for organic matter and color removal in young landfill leachate. To achieve this, Alum, $FeCl_3$, $Fe_2(SO_4)$ were used as coagulants. All coagulants were applied both at leachate's own pH value and at different pH values according to the selected coagulant. Also air stripping was used for ammonium removal at various air flow rates. Optimum aeration time and air flow rate for ammonium removal were determined.

MATERIAL AND METHODS

Landfill Leachate

Leachate was collected from landfill site of Konya city (Turkey) which is located between 36.5-39.5^o north latitude and 31.5-34.5^o east longitude. It is the largest province of Turkey at the altitude of about 1030 m. The population of the city is approximately 950000. The study area is a semi-arid region and receives little annual rainfall. For instance, the annual average rainfall is about 324 mm and was measured as 283 mm in the year of 2006. The average temperature of the area is about 1.2 ^oC in winter and 22 ^oC in summer months. Landfill occupies 24 ha area with an average height of 8 m in the total area of 350 ha at the present time. The amount of solid waste is about 800-850 tons/day in summer and 950-1000 tons/day in winter months yielding the average leachate flow rates of about 100 m^3/day.

Leachate samples collected from the landfill site were filled in plastic container, transported to the laboratory and stored at 4 ^0C. Before testing, samples were removed from the refrigerator and were placed for about 2 hours at room temperature for conditioning.

The main characteristics of the leachate are chemical oxygen demand in the range of 7676-55420 mg/L, biological oxygen demand after 5 days of 4050-36625 mg/L. The characterization of leachate is given in Table **1**. It is observed that presenting a relatively high value of COD and BOD_5 and the rate of COD/BOD_5 values indicates that the leachate can be defined as young. There has beeen low COD and BOD_5 concentrations according to the samples taken between January and December in 2006 depending on the monthly rainfall. It is observed that the reason of low values in COD and BOD_5 concentration is the dilution via rainfall. Changes of COD concentrations corresponding to the total amount of monthly rainfall in landfill site according to data from the meteorological services are given in Fig. (**1**).

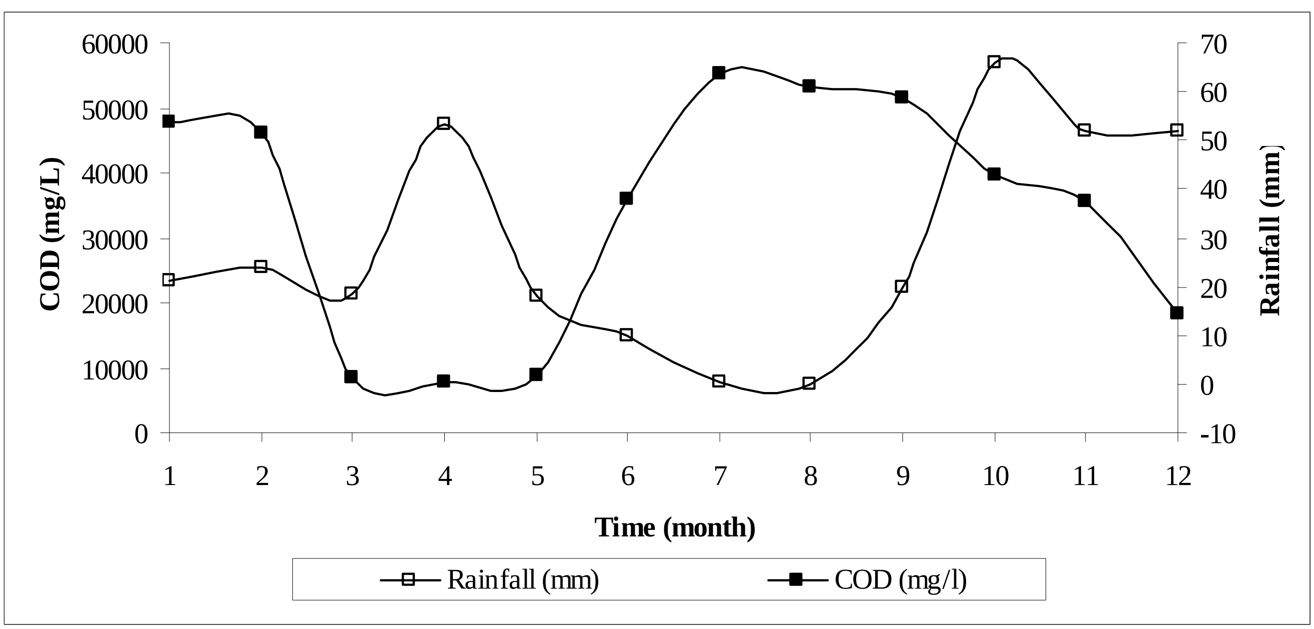

Fig. (1). Changes of COD concentrations corresponding to the total amount of monthly rainfall.

Experimental System

Studies were carried out in two steps. The first one was coagulation-flocculation for organic matter and color removal and the other was air stripping for ammonium removal. Coagulation-flocculation and precipitation studies were performed in a conventional jar test apparatus equipped with 4 beakers of 250 mL. The initial rapid mixing stage took place for 5 minutes at 126 rpm, following the slow mixing stage for 25 minute at 38 rpm and settling for 1 hour. All experiments were both performed at leachate's own pH values and appropriate pH values for the coagulants. Adjustment of the leachate's pH to the desired values was provided by addition of adequate amounts of $Ca(OH)_2$, NaOH and H_2SO_4. Alum, $FeCl_3$ and $FeSO_4$ were used as coagulants. After the settling period, the supernatant was withdrawn from the beaker and was used for analysis. In this study, $Ca(OH)_2$ is used only for pH adjustment although it can be used as a coagulant. The use of lime as an alternative coagulant presented several drawbacks such as increase of hardness, low COD removal efficiency resulting to the production of excessive sludge quantities.

Air stripping studies consisted of beakers of 500 mL. Aeration was provided by air pump via diffusers at three flow rates (1 L/min, 2 L/min and 5 L/min) throughout 24 hours and the pH was adjusted to 11 by adding $Ca(OH)_2$. Leachate was sampled after 0, 2, 4, 6, 8, 12 and 24 hours aeration time and analyzed for ammonia.

Analytical Methods

Leachate characterization studies were carried out between January and December in 2006 with two samples in a month. In the characterization studies, BOD_5, COD, NH_4-N, PO_4-P, pH, temperature and heavy metals were analyzed. Temperature and pH were measured by using a WTW Multiparameter instrument's probe. COD was analyzed by the closed reflux titrimetric methods dictated by Standart Methods [24]. Standard kits (Dr.Lange) and spectrophotometric methods were used for NH_4-N (Kit no: LCK 302) and PO_4-P (Kit no: LCK 350) analysis. Heavy metals were analyzed by ICP-EOS analyzer (Perkinelmer Optima, 2200DV). Color measurements were reported as true color (filtered by using 0.45 μm filter paper) assayed at 455 nm using DR 2000 HACH spectrophotometer according to the method given by Standart Methods [24], Method no 2120C reported in platinium-cobalt (Pt-Co), the unit of color being produced by 1 mg platinium/l in the form of the chloroplatinate ion. The performance of coagulation-flocculation was determined by the means of COD and color removal whereas air stripping was determined by means of NH_4-N removal.

RESULTS AND DISCUSSION

COD Removal

The results of alum coagulation are presented in Fig. (**2a**). COD removal as a function of alum dosage is shown at different pH values (at pH 11 and pH 5.5) adjusted with $Ca(OH)_2$, NaOH and H_2SO_4. In pH adjustment to 11 with lime ($Ca(OH)_2$), the highest value of COD removal was 39% and was obtained using alum dosage of 7 g/L whereas the highest value of COD removal is 44% at 9 g/L alum dosage in pH adjustment to 11 with NaOH. When pH was adjusted to 5.5 with H_2SO_4, COD removal efficiency was 42% and optimum alum dosage was 7 g/L.

In $FeCl_3$ coagulation, pH 3, pH 11 and leachate's own pH values were studied. The results of the effects for different dosages of $FeCl_3$ as coagulant on the removal of COD from the landfill leachate were presented in Fig. (**2b**). The highest COD removal was 55% and was reached at 15 g/L $FeCl_3$ dosage at pH 11 adjusted with lime but system didn't reach the steady state condition. At leachate's own pH value, the highest COD removal was 33% which was the lowest value of the set and was reached at 15 g/L $FeCl_3$ dosage. At pH 3, COD removal was higher according to leachate's own pH and was 45%. And at pH 11 adjusted with NaOH, COD removal was 36% at 11 g/L $FeCl_3$ dosage. This result shows similarity with the study of Maranon *et al.* [28] that they found pH 3.8 as the optimal pH for $FeCl_3$ on the color, COD and turbidity removal.

In $FeSO_4$ coagulation, pH 3, pH 11 and leachate's initial pH values were studied. Results are presented in Fig. (**2c**). Of all the pH adjustment alternatives, the highest COD removal was obtained at 3 g/L $FeSO_4$ dosage in pH adjustment with lime and the efficiency was 43%. In pH adjustment with NaOH, the highest COD removal was 32% at 7 g/L $FeSO_4$ dosage whereas the highest values of COD removal was 35% at 9 g/L $FeSO_4$ dosage when pH was adjusted to 3 with H_2SO_4.

Coagulation-flocculation is more effective in old landfill leachate rather than in young landfill leachate, so number of studies was applied to old landfill leachate [15, 25-27], but studies for young landfill were limited leachate [20, 28]. So when compared to the studies on old and young landfill leachate there are some distinction in COD removal efficiencies. Tatsi *et al.*, [20] studied both young and old landfill leachate without and with pH adjustment and the results on young landfill leachate almost were similar to this study. The maxium COD removal efficiency for alum was 38% at the dosage of 3 g/L at pH 10 adjusted with lime. In this study the COD removal efficiency for alum was 33% at the dosage of 3 g/L at pH 11 and increased with coagulant concentration and was maxium at 7 g/L alum (39%).

Color Removal

In pH adjustment to 11 with lime and NaOH, color removal efficiencies were 99% and 98% at 3 g/L and 5 g/L alum concentrations, respectively. No color removal was obtained in pH adjustment to 5.5. During coagulation it was realized that there was not any distinctive difference in removal efficiency at higher coagulant concentrations so the minimum coagulant concentration was capable of obtaining color removal as much as higher concentrations could do. The results are shown in Fig. (**3a**).

In leachate's own pH value, color removal was 88% at 15 g/L $FeCl_3$ concentration wheras color removal was 63% at pH 3 adjusted with H_2SO_4. This value was the lowest color removal efficiency of the set. And at pH 11 adjusted with NaOH, color removal was 93% at 11 g/L $FeCl_3$ dosage. The highest color removal efficiency was achieved at 1 g/L $FeCl_3$ concentration at pH 11 adjusted with lime and was 98%. Color removals in $FeCl_3$ coagulation are presented in Fig. (**3b**).

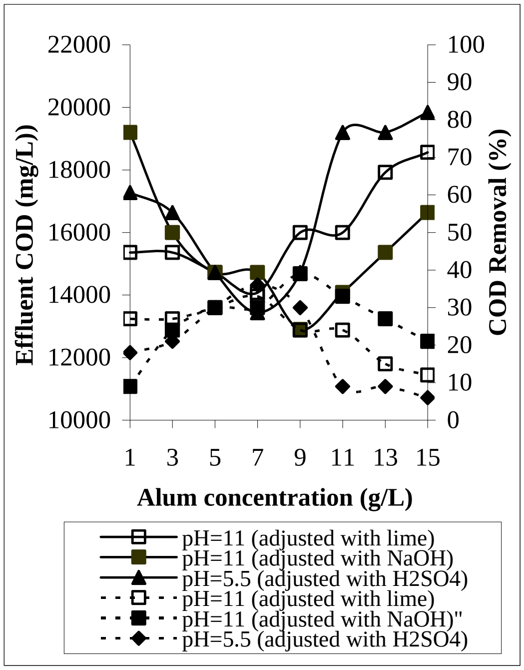

Figure 2a. Coagulation with Alum

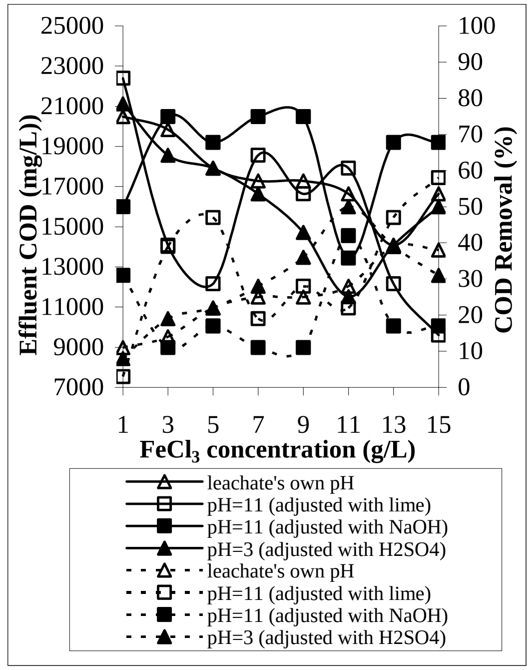

Figure 2b. Coagulation with $FeCl_3$

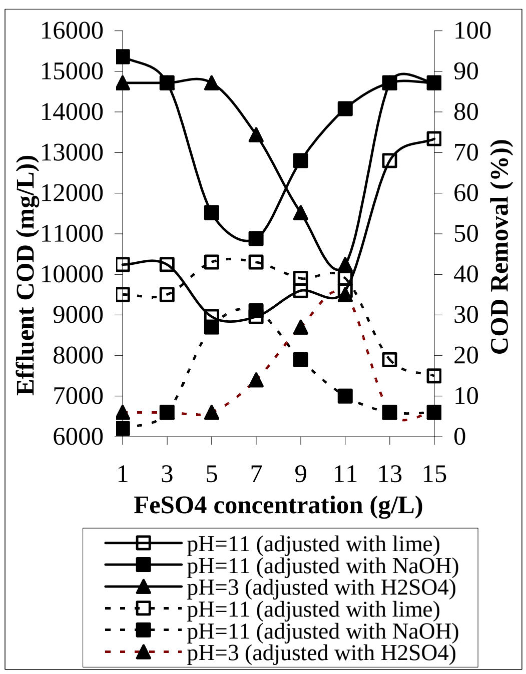

Figure 2c. Coagulation with $FeSO_4$

Fig. (2). Effluent COD concentration and COD removal with chemical matters (data shown with straight line represent effluent COD whereas dash line represent COD removal).

In $FeSO_4$ coagulation, the highest color removal was 99% and was obtained at 1 g/L $FeSO_4$ concentration in pH adjustment with lime. In adjustment with NaOH, color removal efficiency was 87% whereas it was 67% in pH adjustment to 3 with H_2SO_4, at 7 g/L and 9 g/L $FeSO_4$ dosages, respectively. Color removals in $FeSO_4$ coagulation are presented in Fig. (**3c**).

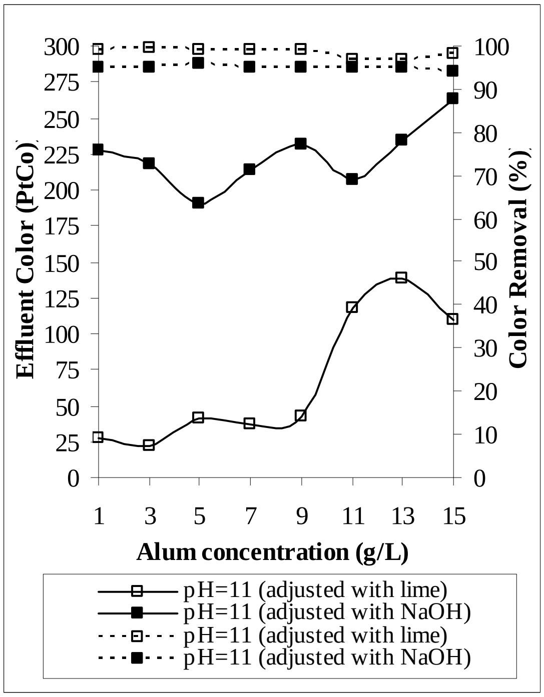

Figure 3a. Coagulation with alum

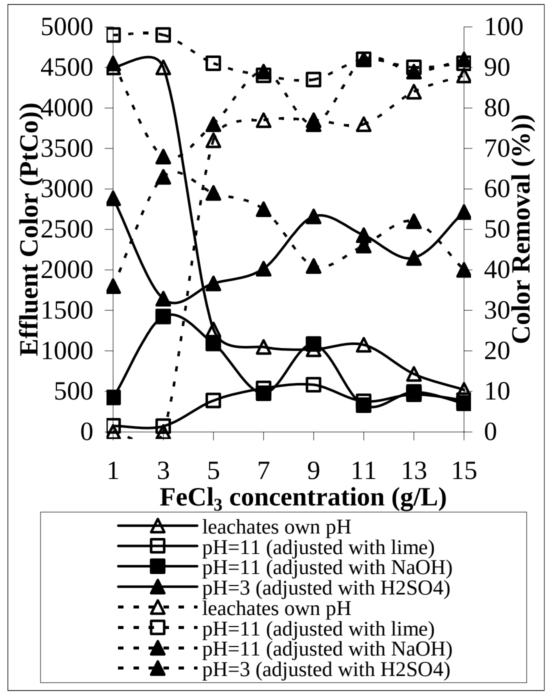

Figure 3b. Coagulation with $FeCl_3$

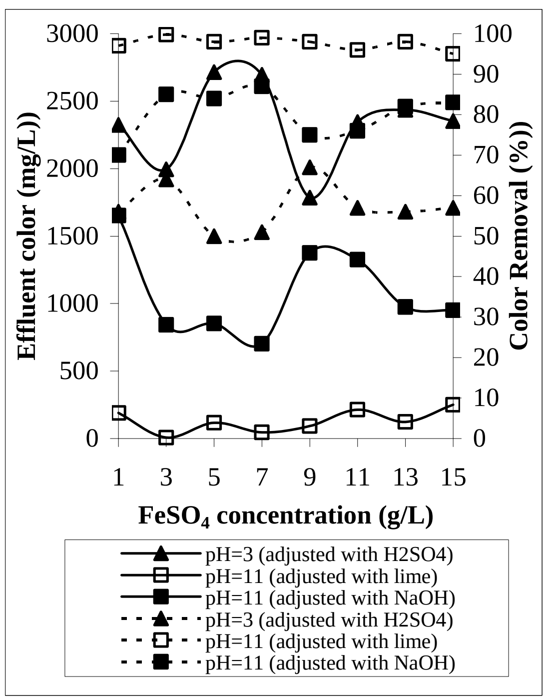

Figure 3c. Coagulation with $FeSO_4$

Fig. (3). Effluent color and color removal with chemical matters (data shown with straight line represent effluent COD whereas dash line represent COD removal).

In this study, high color removal efficiencies were obtained with high coagulant dosages. Studies with young landfill leachate were performed with lower coagulant concentrations than this study. Maranon et al (2008) [28], studied $FeCl_3$ and $Al_2(SO_4)_3$ at 0.4-0.8 g/L coagulant concentrations and obtained 28.1% and 27% COD removal, respectively. Also they studied color removal and at these concentrations, removal efficiencies were 78.4% and 84.3%. Similar to this study, highest color removal efficiency was achieved with alum coagulation.

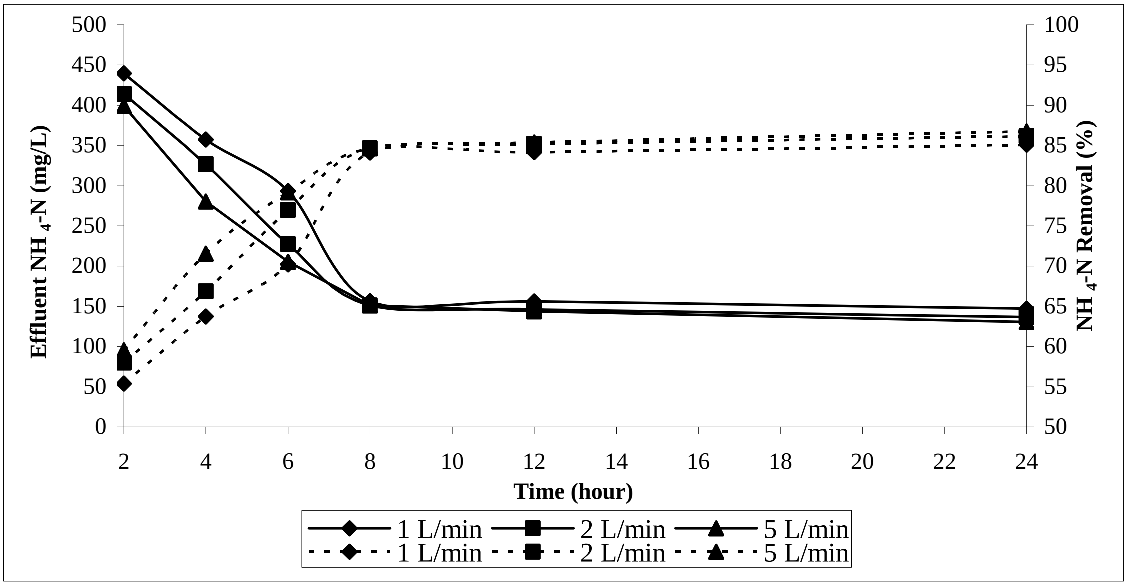

Fig. (4). Effluent ammonium concentrations at various flow rates and aeration times (data shown with straight line represent effluent NH_4-N whereas dash line represent NH_4-N removal).

Air Stripping

The effluent ammonium concentrations at various flow rates and aeration times are presented in Fig. (**4**). As it can be seen in the figure, the initial ammonium concentration was 985.5 mg/L at the beginning of the air stripping studies. After 2 hour-aeration, ammonium removal rates were 55%, 58% and 60% at the flow rates of 1 L/min, 2 L/min and 5 L/min, respectively. With increasing aeration time, ammonium removal was also increased and after 24 hours aeration, the highest removal rates were obtained. The ammonium removal rates were 85%, 86% and 87% at the flow rates of 1 L/min, 2 L/min and 5 L/min, respectively. But after 8 hours aeration, the ammonium removal efficiencies were 84%, 84% and 85% at the flow rates of 1 L/min, 2 L/min and 5 L/min, respectively. There were not many changes for the removal efficiencies after 8 hour-aeration and the increase in the removal efficiencies was not meaningful beyond that. For this reason and economical aspects, the optimum ammonium removal efficiencies were obtained at 8 hour-aeration and 1 L/min flow rate. Similar results were seen in the study of Cheung *et al.* [23]. Their ammonium removal efficiency was increased proportional to increased air flow rate and aeration time and after 24 hour-aeration, 81% and 90% ammonia removals were achieved at flow rates of 1 L/min and 5 L/min, respectively. In this study, higher removal efficiencies at pH 11 were obtained when compared with the study of Ozturk *et al.* and Kabdasli *et al.* [21, 29].

CONCLUSIONS

The coagulation-flocculation experiments indicated that the highest COD removals were achieved at 9 g/L alum and 5 g/L $FeSO_4$ concentrations at pH 11 adjusted with the use of lime. The COD removal efficiencies were 44% and 43%, respectively. In $FeCl_3$ coagulation, 55% COD removal efficiency at pH 11 adjusted with lime was achieved but the system could not reach a steady state condition for the $FeCl_3$ coagulation. In addition to this, it was seen that the optimum concentration was 11 g/L at pH 3 adjusted with H_2SO_4 and COD removal efficiency was 45%.

Color removal studies in coagulation experiments indicated that the highest color removal efficiencies could be reached when pH was adjusted with lime. The similar removal efficiencies were obtained with the lowest coagulant concentration, 1 g/L, as the concentration increased. So it is determined that the optimum concentration in coagulation with alum, $FeSO_4$ and $FeCl_3$ was 1 g/L. In air stripping studies, the optimum flow rate was 1 L/min and the optimum aeration time was 8 hours. With increasing flow rates and aeration times, there was not any considerable increase in NH_4-N removal.

As a result of this study, it is determined that high chemical concentrations were required when coagulation-flocculation method was used as a pre-treatment method. It can be concluded that this method could be used as a post treatment method for young landfill leachate. Therefore, this provides both low chemical use and low sludge production.

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