HAMADAN LANDFILL LEACHATE TREATMENT BY COAGULATION-FLOCCULATION PROCESS


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Received 3 October 2009; revised 27 May 2010; accepted 20 June 2010

ABSTRACT
In most countries, sanitary landfilling is the common way to dispose municipal solid wastes. In the operations, leachate treatment is a difficult and expensive process. Although, leachate can be treated by biological processes, COD removal efficiency is usually low due to high ammonium ion content and the presence of toxic compounds such as metal ions. This experimental study was conducted to investigate the effect of coagulation–flocculation process on the Hamadan landfill leachate treatment in the city of Hamadan. Also the effects of different coagulants with various dosages and pH values in the removal of chemical oxygen demand (COD) and total suspended solids were studied. Results showed that the efficiency for COD removal by Poly Aluminum Chloride at pH=12 and 2500 mg/L of coagulant, by alum at pH=12 and 1000 mg/L of coagulant dose and by ferrous sulfate at pH=12 and 1500 mg/L of ferrous sulfate dose were 60%, 62.33% and 70.66%, respectively. Also results showed that, the efficiency for Total Suspended Solids removal by Poly Aluminum Chloride that was obtained at pH=12 and 2500mg/L concentration of Poly Aluminum Chloride, by alum at pH=2 and 1500 mg/L concentration of alum and by ferrous sulfate at pH=7 and 2500mg/L of ferrous sulfate, were 39.14%, 58.37% and 35.58%, respectively. Based on results of this study, the best coagulant for COD removal was ferrous sulfate and the physico-chemical process may be used as an effective pretreatment process, especially for young leachate, prior to post-treatment (polishing) for partially stabilized leachate.

Key word: Leachate; Treatment; Coagulation-flocculation; Landfill

INTRODUCTION
Increasing the quality of life and industrial and commercial growth in many countries around the world in the past decades have been accompanied by rapid increases in both municipal and industrial solid waste production. Municipal solid waste (MSW) generation continues to grow both in per capita and overall terms (Melike et al., 2007; Ajit Singh and Vidyarthi, 2008). The sanitary landfill method for the ultimate disposal of solid waste material continues to be widely accepted and used due to its economic advantages (Javaheri et al., 2006). Leachate from municipal landfills are defined as the aqueous effluent generated as a consequence of rainwater percolation through wastes, chemical biological processes in waste’s cells and the inherent water content of wastes themselves. The combination of the previous factors results in a dark effluent whose properties highly depend on the age of the landfill (Javier et al., 2008). Leachate from municipal solid waste landfill sites are often defined as hazardous and heavily polluted wastewaters. Leachate may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist an important group,
as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts (Wang et al., 2002).

The appropriate treatment methods are mainly based on specific characteristics leachate under examination (Chian and DeWalle, 1976). Alternative methods of leachate management have been also suggested, including the recirculation of produced leachate through he disposed wastes. The principal benefits of this approach are simplicity of operation and low capital and operation costs. In such a method, recirculated leachate are amenable to biological treatment, due to microorganisms inhabiting the bulk of landfill (Forgie, 1988; Renoua et al., 2008).

Coagulation-flocculation is an essential process in water and industrial wastewater treatment. Several studies have been reported on the examination of coagulation–flocculation for the treatment of landfill leachate, aiming at performance optimization, i.e. selection of the most appropriate coagulant, determination of experimental conditions, assessment of pH effect and investigation of flocculant addition. Aluminum sulfate (alum), ferrous sulfate, ferric chloride and ferric chloro-sulfate were commonly used as coagulants (Amokrane and Comel, 1997; Vaezi et al., 2005).

Iron salts were proved to be more efficient than aluminum ones, resulting in sufficient chemical oxygen demand (COD) reductions (up to 56%), whereas the corresponding values in case of alum or lime addition were lower (39 or 18%), respectively (Diamadopoulos, 1994). Furthermore, the addition of flocculants together with coagulants may enhance the floc-settling rat (Amokrane and Comel, 1997; Zong ping et al., 2002).

The objectives of this study were the examination of coagulation–precipitation process efficiency for the treatment of both fresh (raw), as well as partially stabilized (by re-circulation) leachate, especially in terms of organic matter and total solids removal. More specifically, the aim was the determination of most appropriate coagulant type and dose, studying of pH effect on removal efficiency, the investigation of combined action of coagulants–flocculants and the identification of optimum experimental conditions for the efficient application of this process. This experimental study was conducted to investigate the efficiency of coagulation–flocculation process on Hamadan city landfilling site leachate treatment. Also the effects of various dosages of coagulant and different pH values on the coagulation processes were compared.

**MATERIALS AND METHODS**

**Leachate characteristics**

The leachate was collected from Hamadan city landfilling site. Characteristics of leachate samples are given in Table 1. Leachate was stirred 7 min after the addition of FeCl₃·6H₂O, HCl and NaOH that were used to adjust the pH to the desired value.

**Analytical methods**

The samples were transported to the laboratory in sealed plastic barrels and then stored at 4 °C before being used and analyzed.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>COD</td>
<td>mg/L</td>
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</tr>
<tr>
<td>BOD</td>
<td>mg/L</td>
<td>2500</td>
</tr>
<tr>
<td>pH</td>
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</tr>
<tr>
<td>Salinity</td>
<td>%</td>
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<tr>
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<td>EC</td>
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<tr>
<td>TSS</td>
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<td>31625</td>
</tr>
<tr>
<td>TDS</td>
<td>mg/L</td>
<td>9540</td>
</tr>
</tbody>
</table>

The pH was determined by a pH meter (model HACH sension1), the COD was determined by titration(APHA, 1998) and the TSS was determined by gravimetric standard method in 103°C to 105°C. TDS and EC were determined by EC meter (model HACH sension5).

**Chemical coagulation**

By using Jar Test equipment in the physicochemical treatment experiments were investigated the effect of Al₂(SO₄)₃·18H₂O (supplied from Merck), poly aluminum chloride (PACl) and ferrous sulfate at various dosages on removing the COD and TSS content of leachate.
The jar test was carried out: first, pH of samples were readjusted to desired pH (2, 7, 12) and then the varying coagulant concentrations (500, 1500, 2000 and 2500 mg/L) at room temperature (20°C) were dosed into 1 L of a leachate sample. The fast mixing for 3 minute (150 rpm), slow mixing for 20 min. (70 rpm) and 1 hour settling time were applied sequentially in chemical precipitation. After that, the supernatant was analyzed for COD and TSS concentration according to standard methods. The removal efficiency of total suspended solids (TSS) and COD was obtained by using equation (1):

\[
\text{RE} (%) = \frac{(C_0 - C_t)}{C_0} \times 100
\]  

(1)

Where, \( C_0 \) and \( C_t \) are the initial and final concentrations of suspended solids and COD.

**RESULTS**

**Alum efficiency**

The effect of various doses of alum and different pH values on the COD removal efficiency is shown in Fig. 1. The removal of COD and TSS increased with increasing concentration of added alum. It observed that, when the dose of alum was greater than 1500 mg/L, the removal of COD increased slowly. The high efficiency for COD removal by alum was obtained at pH=12 and 1000 mg/L concentration of alum.

The effect of various doses of alum and different pH values on the removal of TSS is shown in Fig. 2. The high efficiency for TSS removal by alum was obtained at pH=2 and 1500 mg/L concentration of alum.

**PACl efficiency**

The effect of different doses of PACI and different pH values for COD and TSS removal is shown in Fig. 3 and Fig. 4. The removal of COD and TSS increased with increasing concentration of PACI. The high efficiency for COD removal by PACI was obtained at pH=12 and 2500mg/L concentration of PACI.

**Ferrous sulfate efficiency**

The effect of different doses of Ferrous Sulfate and pH values on the COD removal is shown in Fig. 5. The removal of COD and TSS increased with increasing concentration of ferrous sulfate.

The effect of various doses of Ferrous Sulfate and different pH values in the TSS removal efficiency is as shown in Fig. 6. Also the effect of pH values on COD removal efficiency by using different coagulants showed in Fig.7.
Fig. 2: The effect of alum doses and pH on the removal of TSS from landfill leachate

Fig. 3: The effect of PACl doses and pH values on the removal of COD from landfill leachate

Fig. 4: The effect of PACl doses and pH values on the removal of TSS from landfill leachate
Fig. 5: The effect of ferrous sulfate doses and pH values on the removal of COD from landfill leachate

Fig. 6: The effect of ferrous sulfate doses and pH values on the removal of TSS from landfill leachate

DISCUSSION

(Tchobanoglous et al., 1993) reported that the levels of suspended solids and COD for leachate younger than 2 years may reach 2000–30000 mg/L and 3000–60000 mg/L, respectively (Tchobanoglous, Theisen et al., 1993). According to (AWWA, 1971), pH is the most important variable in the coagulation process for wastewater treatment. The extent of pH range is affected by the types of coagulant used and by the chemical composition of wastewater as well as by the concentration of coagulant (AWWA, 1971). When coagulant doses were set at 1500 mg/L and the effects of varying pH were evaluated, it was observed that relatively higher removals of suspended solids, and COD were observed at lower and higher pH values (pH = 4 and 12). For example, alum
had removed 58% of suspended solids at pH= 2 compared with less than 12% at pH= 12. Ferrous sulfate was found to be generally superior to the other coagulants for removal COD monitored. For example, at pH= 12, over 70.66% of COD was removed by ferrous sulfate, while alum and PACI coagulants removed only from 44 and 49.33% of COD, respectively, at pH= 12. Amokrane et al., (1997) similarly found ferric chloride to be more effective than aluminium sulphate (94 and 87%, respectively) for coagulation–flocculation pretreatment turbidity removal from of landfill leachate (Amokrane and Comel 1997). Overall, alum performed better than FeSO4 and PACI in removing TSS.

At pH= 2, TSS removals for alum, FeSO4 and PACI were 58% and 32.39% and 36.88 %, respectively. At pH= 7, the removal of COD was fairly low. These results are in agreement with other reports of FeCl3, FeSO4, or alum coagulation and flocculation applied to textile and municipal wastewater (Nicolaou and Hadjivassilis 1992; Selcuk 2005). Also the results of the present study are in agreement with those in several other reports.

The greatest source of uncertainty remains with the optimum pH region for the coagulation–flocculation process to remove the contaminants tested. According to Stephenson & Duff (1996), the influence of pH on chemical coagulation–flocculation may be considered as a balance between two competitive forces: (1) H+ and metal hydrolysis products for interaction with organic legands; and (2) hydroxide ions and organic anions for interaction with metal hydrolysis products. Coagulation and flocculation studies conducted on the leachate from the Hamadan Landfill Site in Iran indicated that alum was capable of removing suspended solids, and ferrous sulfate was best capable of removing COD. The best removal efficiency was observed at pH= 2 and pH=12 than at pH= 7. However, about 2500 mg/ L of coagulants were required to give good removal efficiency at pH= 7. Flocculation processes could be applied to improve the leachate characteristics as part of an integrated treatment system for semi-aerobic landfill leachate. This physico-chemical process may be used as a useful pretreatment step, especially for fresh leachate, prior to post-treatment (polishing) step for partially stabilized leachate.

ACKNOWLEDGEMENTS

The authors are most grateful to the laboratory staff of the Department of Environmental Health Engineering, School of Public Health, Hamadan University of Medical Sciences, for financial support and their collaboration in this research.

REFERENCES


