

Use of Alum and Ferric Sulphate for Treating Landfill Leachate via Coagulation Process: A Comparative Study

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ABSTRACT

Landfill leachate is a complex wastewater which may give potential problems to the environment. Besides the new treatment processes that has been implemented, coagulation and flocculation treatment are considered as the most widely used method in treating landfill leachate to reach requirements specification for the discharge of leachate. Regularly, aluminium sulphate (alum) is used as a chemical coagulant to enhance the removal contaminants in this treatment process. Besides alum, ferric sulphate also shows their ability in leachate treatment. Hence, this study has been carried out to examine the comparison between both chemical coagulants in treating leachate form Padang Siding Landfill Site. The optimum pH and coagulant dosage was evaluated by a series of Jar Test experiment. In addition, the Sludge Volume Index (SVI) test also determined for the treated leachate under optimized conditions. Ferric sulphate with optimum pH 3.0 and coagulant dosage of 6000 mg/L was successfully removed of 58.9% of COD, 72.9% of turbidity and 46.1% of suspended solid. Meanwhile, alum coagulant was able to removed 69.4% of COD, 94.5% of turbidity and 84.3% of suspended solid under optimum pH 5.0 and 8000 mg/L of optimum coagulant dosage. Besides that, SVI of alum was 46.8 mL/g and ferric sulphate gave the value of 32.5 mL/g for SVI. Hence, ferric sulphate is recommended to replace alum as a coagulant in landfill leachate treatment process since the efficiency is almost the same as alum.

Keywords: Landfill Leachate, Alum, Ferric Sulphate, Coagulation-Flocculation, Water and Wastewater.

1. INTRODUCTION

Landfill leachate can be generated by the precipitation, surface run-off, and penetration or intrusion of groundwater permeating through a landfill (Wang *et al.*, 2012). The surface water will pass over the waste in the landfill and it potentially mix with the groundwater around the site area if the landfill is not properly lined and leachate systems is not well managed. Landfill leachates are made out of inorganic and organic substances. In organic substances, the metabolic and materials of bacteria can be produced by the decomposition of living organisms. Then, the ammonium content, heavy metal and phosphorous can be found in inorganic pollutants. Not only that, landfill leachate also contains of high toxicity and various harmful contaminants that can be a potential impact to the surroundings. Hence, it is important to implement the appropriate treatment systems or design the leachate treatment operation in order to reducing or eliminating the pollutants.

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Most of the leachate treatment was adopted from water and wastewater treatment and technologies such as aerobic and anaerobic, flotation, coagulation-flocculation, chemical precipitation, adsorption, ammonium stripping, chemical oxidation, ion exchange, electrochemical treatment, microfiltration, ultrafiltration, nanofiltration and reverse osmosis (Abbas *et al.*, 2009).

In this study, coagulation and flocculation process will be focused as a method to treat landfill leachate. Coagulation is a chemical reaction which occurs when a chemical or coagulant with charges opposite to the suspended solids are added to the water to neutralize the negative charge on non-settable solids like colour-producing organic substances (Prakash *et al.*, 2014). Flocculation is gentle stirring or agitation to encourage the particles, thus formed to agglomerate into masses large enough to settle or be filtered from the solution. The effectiveness of this process will be influenced by some factors such as temperature, pH, effluent quality, dosage and coagulant type (Nnaji 2012; Jin 2005; Ma *et al.*, 2001).

This study investigates the comparative suitability of alum and ferric sulphate as coagulants for leachate treatment. Removal of chemical oxygen demand (COD), turbidity, suspended solid was monitored throughout the experiments. Thus, the effectiveness of alum and ferric sulphate as coagulants, in the determination of optimum pH and dosage were based on two factors (coagulant dosage and pH) and three responses (COD, turbidity, suspended solid).

2. MATERIAL AND METHODS

Raw landfill leachate was collected from the leachate collection pond at Padang Siding Landfill. Before jar testing process, the characterization of the leachate sample collected was analyzed. The optimum coagulant dosage and pH for both coagulants were determined through the jar test experimental works with appropriate operating parameters such as chemical oxygen demand (COD, turbidity and suspended solid. The removal efficiency of these parameters has been determined by calculation of percentage removal. Lastly, determination of the Sludge Volume Index (SVI) of the treated samples was conducted under optimum conditions.

2.1 Sampling of Landfill Leachate

According to Kamarudzaman *et al.*, (2011), the leachate samples were stored in a high density polyethylene (HDPE) bottle right after the leachate collection from the pond. Leachate sampling will be conducted once a month over three months, from November 2016 to January 2017. The samples were brought to the laboratory immediately and preserved in the refrigerator at 4°C to reduce the changes in the sample constituents. The acquisition and storage of leachate samples were followed the Standard Methods for the Examination of Water and Wastewater (APHA, 2005). Before conducting the jar test, the samples were taken out from refrigerator and left under ambient temperature for approximately 2 hours. The samples were characterized in terms of pH, COD, turbidity and suspended solid.

2.2 Preparation of Coagulant

Stock solution of both chemical coagulants was prepared prior to jar testing for determination of optimum pH and coagulant dosage. The coagulant dosage was calculated using equation 1.

$$M_1V_1 = M_2V_2 \tag{1}$$

M_1 = Concentration of coagulant (mg/L)

V_1 = Volume of coagulant (ℓ)

M_2 = Concentration of wastewater sample (mg/L)

V_2 = Volume of wastewater sample (ℓ)

2.3 Jar Test

Coagulation-flocculation studies are accomplished by standard jar test equipment. Jar test is a common laboratory procedure that can be used to determine the optimum operating conditions for wastewater treatment. Standard jar test apparatus (JLT6 VELP Scientifica) was used in this treatment process. The pH was adjusted using 1.0M NaOH and 1.0M H₂SO₄ prior to jar test process. Next, the rapid mixing speed will be fixed at 100 rpm for 3 minutes, whereas the slow mixing will be set at 30 rpm for 10 minutes. Then, 60 minutes of settling time will be given for the settlement of suspended particles at the bottom of the beaker. After that, the supernatant samples will be withdrawn using plastic syringe and filter the sample for insoluble substance removal.

In order to obtain the optimum pH for coagulation-flocculation process, the pH is varied and fixed the coagulant dosage at 5000 mg/L initially. The pH values selected for the run are pH 1.0, 2.0, 3.0, 5.0, 7.0 and 9.0. On the other hand, the various ranges of dosage are within 1000 mg/L to 10000 mg/L for both coagulants. The optimum dosage determined by varying dosage used where the other factors, pH is fixed.

After completing the jar test, parameters of the treated leachate were measured and compared with the raw leachate. The parameter that was measured are pH, turbidity, chemical oxygen demand (COD) and suspended solid. After completing the parameter analysis, the removal efficiency was calculated based on the following formula.

$$\text{Removal Efficiency} = \frac{C_o - C_i}{C_o} \times 100 \quad (2)$$

C_o is referring to initial concentrations, meanwhile C_i is final concentrations. Sludge Volume Index (SVI) was measured after determining the optimum pH and coagulant dosage for jar test process, using the sludge produced under optimum conditions.

3. RESULTS AND DISCUSSION

3.1 Raw Leachate Characteristics

Detailed characteristics of landfill leachate that was collected from November 2016 to January 2017 are given in Table 1. From the raw leachate characterization, the recorded pH value was ranging between 7.9 to 8.3. This pH value shows that the leachate is already matured and stabilized because the value higher than 7.5 (Umar *et al.*, 2010). This stabilized landfill leachate contains of extreme value of COD and ammonium concentrations, low concentration of BOD₅/COD and have dark-coloured in appearance (Kamaruddin *et al.*, 2013).

The mean value of COD measured was 3009 mg/L. Studies find out that stabilized landfill leachate characterized by moderately high strengths of COD and produce low biodegradability. Turbidity was reported as 87.7 NTU and the average suspended solid is 204 mg/L. Turbidity has closely related to the suspended solid that may indicate the amount of colloidal particles in water bodies. The presence of colloidal particles and suspended solid in water bodies reducing the transparency in turbidity.

Table 1 Characteristics of Raw Leachate Collected from Padang Siding Landfill (From November 2016 to January 2017)

Parameters	Reading	Average Reading	^a Effluent Discharge Standard
pH	7.9 - 8.3	8.1	6.0 - 9.0
COD (mg/L)	2795 - 3278	3009	400
Turbidity (NTU)	85.4 - 89.9	87.7	-
Suspended Solid (mg/L)	147 - 237	204	50

(^aEnvironmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation 2009 (ILBS,2014))

3.2 Effect of pH on Coagulation-Flocculation Process

pH is a significant factor influencing the performance of removal efficiency in coagulation and flocculation treatment process. Figure1 indicates the removal efficiency of COD using ferric sulphate as coagulant is gradually increasing and suddenly drop until pH 9. An increasing trend was observed for turbidity up to pH 3 and continues to slightly decrease until reach minimum percentage of removal efficiency at pH 9. From this result, the maximum removal of COD and turbidity is 42.9% and 74.1% respectively that obtained at pH 3. For suspended solid, this treatment allows the elimination up to 46.1% at pH 2.

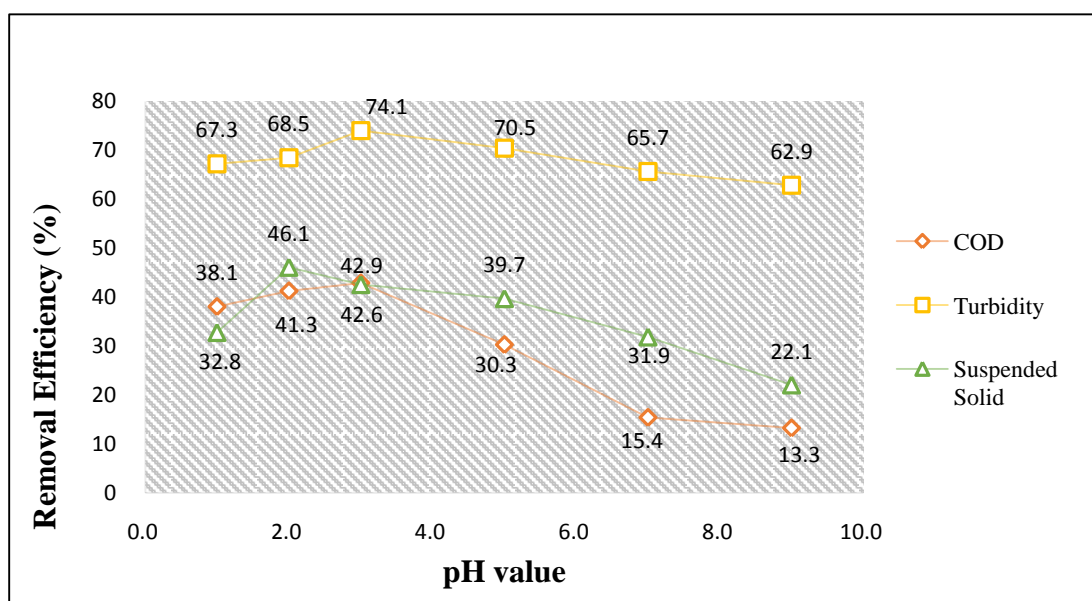


Figure1. Percentage of COD, turbidity and suspended solid removal against pH at 5000 mg/L (ferric sulphate)

The removal efficiency of suspended solid dropped as the pH increase after the optimum level. Under the same dosage of coagulant and due to acidic conditions of ferric sulphate coagulant, this treatment showed better result at lower pH which is the overall optimum pH is pH 3. According to Cheng *et al.* (2002), as the pH increase in coagulation-flocculation treatment process, the ferric ions may still react with humic acid but unable to neutralize the surface charges completely.

In Figure 2, the highest percentage removal efficiency for COD using alum coagulant was recorded at pH 3. The highest reduction for COD reaches up to 50.5%. Meanwhile turbidity and

suspended solid achieved their maximum removal at pH 5 with the value of 85.9% and 80.4% respectively. After COD, turbidity and suspended solid reached their maximum percentage removal, then they slightly decrease with increasing pH. This optimum point indicated that the performance of removal efficiency in leachate treatment was enhanced in acidic condition (Wang *et al.*, 2002). Under acidic condition, all organic compounds were fully oxidized to carbon dioxide. Even though anaerobic organisms are not an oxidizing agent, degradation of the organic content can occur through dissolved microorganisms in the leachate sample (Kamaruddin *et al.*, 2013). As a conclusion for this study, leachate treatment using alum coagulant reached the optimum efficiency removal at pH 5.0. This result supported by Kang and Hwang, (2000) which indicated that the optimum pH using alum in coagulation is within range of pH 3.0 to 6.0.

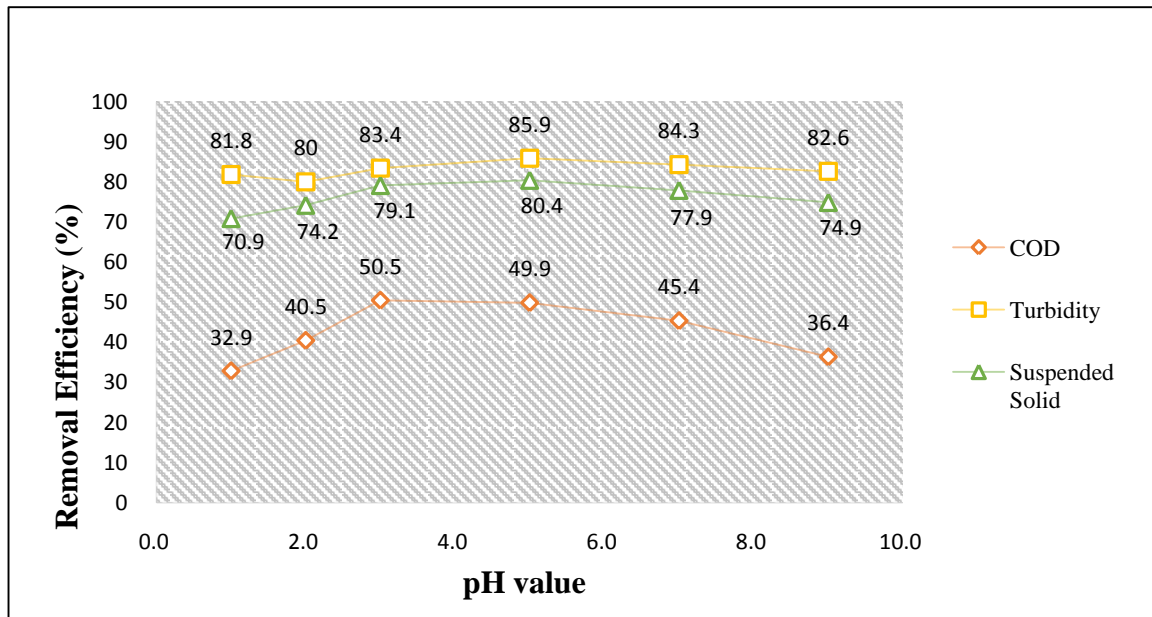


Figure 2. Percentage of COD, turbidity and suspended solid removal against pH at 5000 mg/L (alum).

Generally, the pH of medium changes during the process. The changes of pH in landfill leachate after coagulation and flocculation process by jar test presented in Table 2. Changes of pH occurred due to chemical reaction of acidic coagulant used in the treatment process when mix with landfill leachate. The pH drops also affected from solubility and charge neutralization at a certain pH.

Table 2 pH value measured after treatment

Initial pH	pH measured after treatment	
	Ferric Sulphate	Alum
1.0	1.0	1.1
2.0	1.8	1.9
3.0	3.2	3.3
5.0	5.1	4.8
7.0	7.2	7.2
9.0	8.7	9.1

3.3 Effect of Dosage on Coagulation-Flocculation Process

Coagulant dosage is one of important role in removing contaminants in coagulation and flocculation process. According to Aguilar *et al.*, (2005), optimum dosage of coagulant is defined as a maximum value which there is no significant increase in removal efficiency with further addition of the coagulant. The dosage used for both ferric sulphate and alum ranged from 1000 mg/L to 10000 mg/L. The optimum pH for ferric sulphate and alum was pH 3 and pH 5 respectively. The effect of ferric sulphate as coagulant in identifying the optimal coagulant dosage was shown in Figure 3. Turbidity removal shows the increasing of percentage consistently until reached highest removal at dosage 8000 mg/L with 75.3% efficiency. But after achieve that optimum coagulant dosage, the removal efficiency decrease as the coagulant dosage increase. For COD, the trends of the result were smoothly increased at first before the fluctuations of value occur. COD and suspended solid managed to remove the pollutants as much as 58.9% and 46.1% respectively. Both parameters were reached the optimal dose at 6000 mg/L of ferric sulphate dosage.

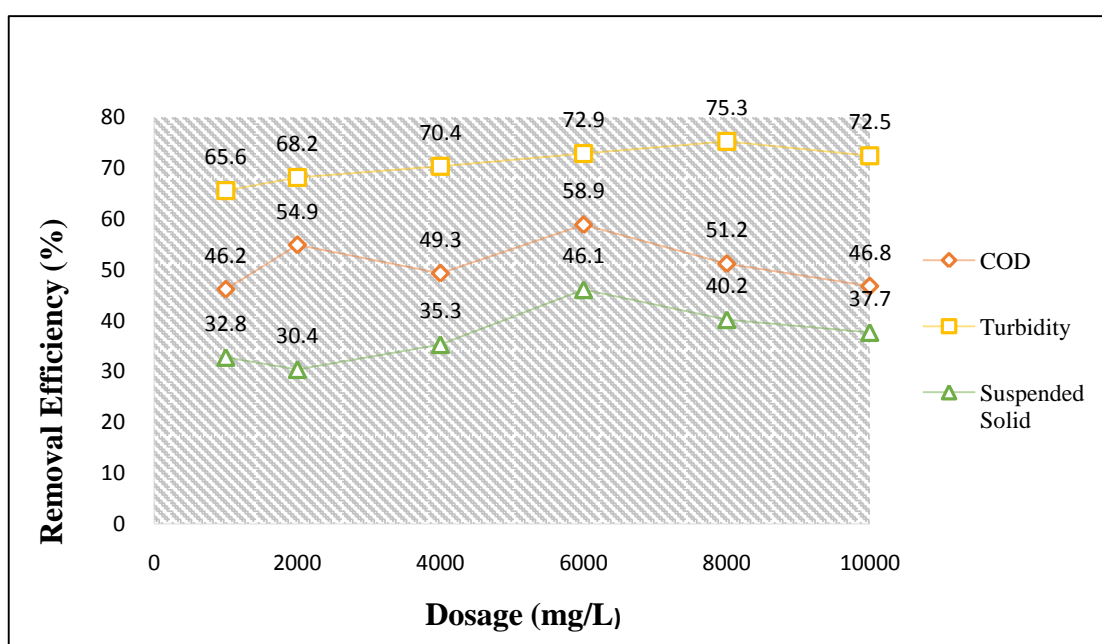


Figure 3. Percentage of COD, turbidity and suspended solid removal against dosage at pH 3 (ferric sulphate).

The removal pattern of COD and turbidity treated with alum almost similar as shown in Figure 4. Initially, the removal efficiency gradually increased up to dosage 4000 mg/L and it slightly drops at dosage 6000 mg/L. An increasing trend was observed after dosage 6000 mg/L and followed by a decrease when the dosage was increased to 10000 mg/L. In this experiment, the optimal coagulant dosage that was determined is 8000 mg/L induced a 94.5% for turbidity reduction and 69.4% of COD removal. For suspended solid, the maximum removal occurred at a dose of 6000 mg/L with the value of 85.6%. The pattern for suspended solid was increased from the started point until reached the optimum coagulant dosage then it reduced as the coagulant dosage increase.

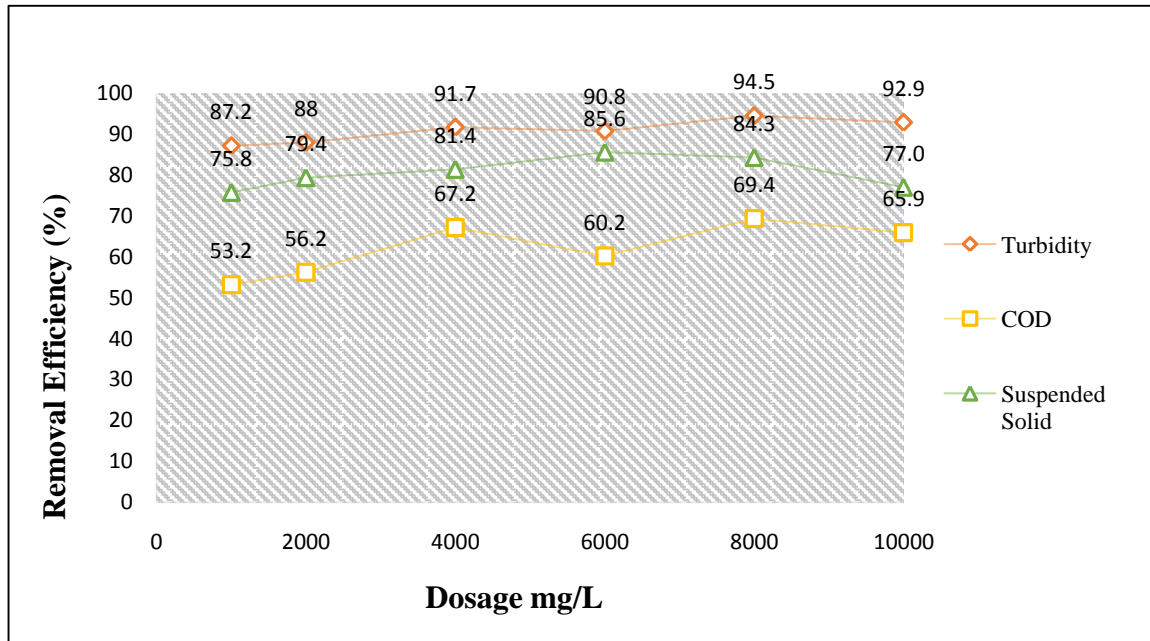


Figure 4. Percentage of COD, turbidity and suspended solid removal against dosage at pH 5 (alum).

3.4 Sludge Volume Index (SVI) for Alum and Ferric Sulphate under Optimum Conditions

Generally, the determination of sludge volume index (SVI) depends on the coagulant or flocculant used and operating conditions that has been considered during coagulation and flocculation process. SVI was identified in this study for both ferric sulphate and alum under optimum conditions and set 30 minutes of settling time. Figure 5 demonstrates the SVI treated for both coagulants. Ferric sulphate produces 32.5 mL/g of SVI whereas 46.8 mL/g of SVI generated from alum coagulant. Thus, alum has higher SVI compared to ferric sulphate. SVI result with lower value indicates a good settling sludge, which has a bigger floc size and need a shorter time for the sedimentation process (Bhatia *et al.*, 2007). The capability in producing sludge is almost the same for both coagulants. Nevertheless, ferric sulphate is more capable to produce compact sludge in lower settling time than alum. Based on this result, ferric sulphate will give more advantage in sludge operation which is the treatment plant will produce less sludge at final discharge point. It will be easier for the sludge conductor to handle the sludge disposal.

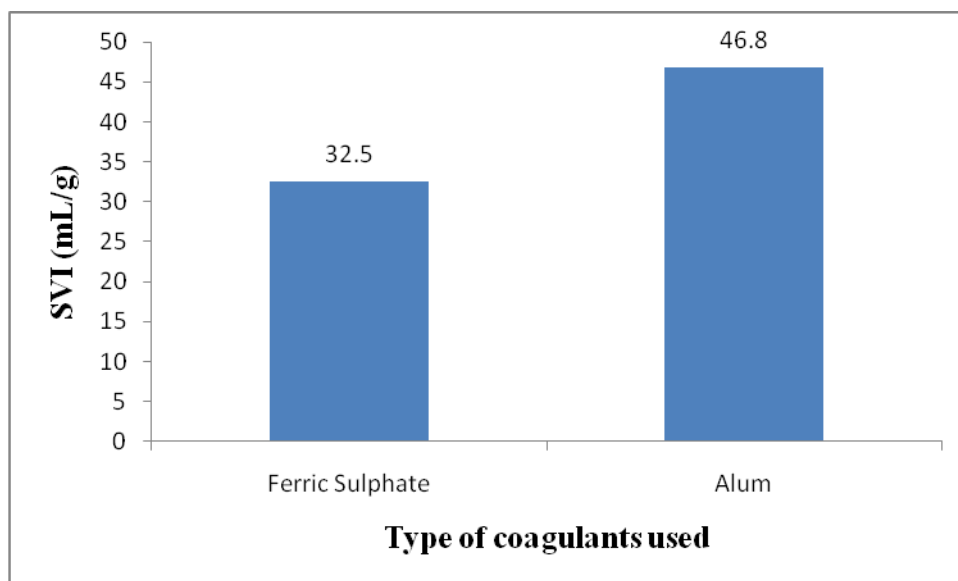


Figure 5. Sludge Volume Index (SVI) against type of coagulants used.

4. CONCLUSIONS

Leachate from Padang Siding Landfill was treated using aluminium sulphate (alum) and ferric sulphate as coagulant. The optimum condition from both coagulants was successfully determined. Ferric sulphate with optimum pH 3.0 and coagulant dosage of 6000 mg/L was induced a 58.9% of COD, 72.9% of turbidity and 46.1% of suspended solid removal. Meanwhile, alum coagulant was able to removed 69.4% of COD, 94.5% of turbidity and 84.3% of suspended solid under optimum pH 5.0 and 8000 mg/L of optimum coagulant dosage. This study showed that the usage of alum as coagulant in leachate treatment is more effective in removing COD, turbidity and suspended solid. But ferric sulphate also obtained the removal efficiency almost similar to alum performance. Moreover, Sludge Volume Index (SVI) test showed that ferric sulphate generated less sludge compared to alum coagulant. However, the excess amount of sludge produced at the sludge final discharge in coagulation-flocculation treatment process will cause the problematic issues such as sludge disposal problems to the operator and environment. Due to less sludge produced from alum coagulant, ferric sulphate is recommended as a coagulant in landfill leachate treatment process since the efficiency is almost the same as alum performance.

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REFERENCES

- [1] Abbas, A. A., Jingsong, G., Ping, L. Z., Ya, P. Y., & Al-Rekabi, W. S. Review on Landfill Leachate Treatments. *Journal of Applied Sciences Research* **5**, 5 (2009) 534-545.
- [2] Aguilar, M. I., Saez, J., Lloréns, M., Soler, A., Ortuno, J. F., Meseguer, V., & Fuentes, A. Improvement of coagulation–flocculation process using anionic polyacrylamide as coagulant aid. *Chemosphere* **58**, 1 (2005) 47-56.
- [3] American Public Health Association, American Water Works Association, Water Pollution Control Federation, & Water Environment Federation. *Standard methods for the examination of water and wastewater (Vol. 2)*. American Public Health Association, (1915).
- [4] American Public Health Association, American Water Works Association, Water Pollution Control Federation, & Water Environment Federation. *Standard methods for the examination of water and wastewater (Vol. 2)*. American Public Health Association, (1915).
- [5] Bhatia, S., Othman, Z., & Ahmad, A. L. Pretreatment of palm oil mill effluent (POME) using *Moringa oleifera* seeds as natural coagulant. *Journal of Hazardous Materials* **145**, 1 (2007) 120-126.
- [6] International Law Book Services. *Laws of Malaysia – Environmental Quality Act 1974 (Act 127) Regulations, Rules, Orders*. Kuala Lumpur, Malaysia. (2014).
- [7] Jin, Y. Use of a high resolution photographic technique for studying coagulation/flocculation in water treatment (Doctoral dissertation, University of Saskatchewan Saskatoon). (2005).
- [8] Kamaruddin, M. A., Yusoff, M. S., Aziz, H. A., & Basri, N. K. Removal of COD, ammoniacal nitrogen and colour from stabilized landfill leachate by anaerobic organism. *Applied Water Science* **3**, 2 (2013) 359-366.
- [9] Kamarudzaman, A. N., Aziz, R. A., & Jalil, M. F. A. Removal of heavy metals from landfill leachate using horizontal and vertical subsurface flow constructed wetland planted with *Limnocharis flava*. *Int. J. Civil Environ. Eng* **11**, 5 (2011) 85-91.
- [10] Kang, Y. W., & Hwang, K. Y. Effects of reaction conditions on the oxidation efficiency in the Fenton process. *Water research* **34**, 10 (2000) 2786-2790.
- [11] Ma, J., Li, G. B., Chen, Z. L., Xu, G. R., & Cai, G. Q. Enhanced coagulation of surface waters with high organic content by permanganatepreoxidation. *Water science and technology: water supply* **1**, 1 (2001) 51-61.
- [12] Nnaji, P. C. An investigation of the performance of various coagulants/flocculants in removing the turbidity of coal washery effluents. M. Eng (Doctoral dissertation, Thesis, Federal University of Technology, Owerri). (2012).
- [13] Prakash, N. B., Sockan, V., & Jayakaran, P. Wastewater treatment by coagulation and flocculation. *Int J Eng Sci Innov Technol* **3**, 2 (2014) 479-484.
- [14] Umar, M., Aziz, H. A., & Yusoff, M. S. Variability of parameters involved in leachate pollution index and determination of LPI from four landfills in Malaysia. *International Journal of Chemical Engineering*, (2010).
- [15] Wang, Z. P., Zhang, Z., Lin, Y. J., Deng, N. S., Tao, T., & Zhuo, K. Landfill leachate treatment by a coagulation–photooxidation process. *Journal of Hazardous Materials* **95**, 1 (2002) 153-159.

