The use of Polyaluminum Chloride for the treatment of Landfill Leachate via Coagulation and Flocculation processes

Noor Ainee Zainol, Hamidi Abdul Aziz*, Mohd Suffian Yusoff and Muhammad Umar
School of Civil Engineering, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, MALAYSIA

Available online at: www.isca.in
(Received 26th April 2011, revised 3rd May 2011, accepted 24th May 2011)

Abstract

Leachate is a highly complex and polluted wastewater that is produced by the interaction of percolating water through the body of a landfill. Treatment of landfill leachate is essential as it could threaten the surrounding ecosystem. The aim of this research was to determine the efficiency of polyaluminum chloride (PACl) in removing chemical oxygen demand (COD), ammoniacal nitrogen (NH_3-N) and color from two different landfill sites in Malaysia. The optimum dosage in removal of these parameters was determined and found to be lower for Kulim Landfill Site (KLS) than Ampang Jajar Landfill Site (AJLS). At optimum dosage of PACl as coagulant, the COD removal for KLS was higher than that obtained for AJLS. The removal of color for both sites was almost similar. The removal NH_3-N was rather moderate for both the landfill sites with KLS exhibiting relatively higher removal rates.

Keywords: Leachate, landfill, coagulation and flocculation, polyaluminium chloride.

Introduction

Landfilling is one of the most popular methods of municipal solid waste (MSW) disposal due to its relative simplicity in terms of disposal procedures, low cost and landscape-restoring effect on the holes from mineral workings[1]. However, there is a significant concern associated to this disposal method which the generation of highly polluted wastewater called leachate. Leachate is produced when the rainwater or water inherently present within the waste cells leaches out from the body of a landfill[2,3]. Leachate may contain large amounts of organic matter (biodegradable and non-biodegradable), ammoniacal-nitrogen, suspended solids (SS), inorganic salts and significant concentration of heavy metals[4,5]. Thus, the discharge of landfill leachate without proper treatment could cause harmful effects to the surface and groundwater as it could percolate through soils and subsoils[6].

Chemical composition of landfill leachate depends on several factors including waste composition, site hydrogeology, specific climatic conditions, moisture routing through the landfill, landfill age as well as design and operation of the landfill[7,8]. In general, young leachate tends to be acidic due to the presence of fatty acids[9]. On the other hand, stabilized or old leachate manly consists of high molecular weight organics such as humic and fulvic acids and relatively low chemical oxygen demand (COD) in the range of 500-5000 ppm[10]. Therefore, biological processes are considerably effective for young leachate but less efficient for older or stabilized leachate[5].

Coagulation-flocculation is a relatively simple and frequently applied technique in water and wastewater treatment. This technique could be successfully employed for the treatment of stabilized landfill leachates[11]. The removal mechanism of this process mainly consists of charge neutralization of negatively charged colloids by cationic hydrolysis products, followed by incorporation of impurities in an amorphous hydroxide precipitate through flocculation[12].

A coagulant is defined as the substance or chemical that is added into water to begin and accomplish the coagulation-flocculation process[13]. Studies on the coagulation-flocculation process for the treatment of landfill leachates have been reported by several researchers[9,11,14]. Aluminum sulfate (alum), ferric chloride and ferric sulfate were commonly used as coagulants. However, in recent years, much attention has been paid to hydrolyzing metal salt coagulants namely polyaluminum chloride (PACl) due to its higher coagulant efficiency and relative low cost compared to the conventional coagulants[15-17]. Besides, PACI poses a good structure and higher charge density which leads to decrease in dosage requirements and hence lesser sludge production[16]. The application of PACl as a coagulant for the removal of color, COD and ammonia from water and wastewater has been investigated by several researchers[4,9,18-20] with a conclusion that PACl is capable of removing color, COD and ammonia. However, the use of PACl as a coagulant is still uncommon in leachate treatment.

This study focused on two different types leachate generated from Kulim Landfill Site (KLS) and Ampang Jajar Landfill Site (AJLS). KLS is located in Kulim, Kedah, Malaysia. It is an open dumping site that started operations in 1996 and was upgraded to sanitary landfill in 2006. AJLS is located in Seberang Perai, Penang, Malaysia. This site is administrated by Majlis Perbandaran Seberang Perai (MPSP) and is closed for more than 10 years. The purpose of this study was to examine the efficiency of coagulation-flocculation processes for the removal of COD, NH_3-N and color from KLS and AJLS leachate using PACI. Thus, the results would be useful in establishing the removal pattern of each parameter in landfill leachate.

Material and Methods

Leachate: Leachate samples were collected from KLS and AJLS between June and December 2010. Samples were collected in 25-L plastic containers, transported to the laboratory and stored at 4 °C. Leachate samples were removed from the refrigerator and left under ambient temperature for about 2 hrs before the jar test was performed. Then, samples were thoroughly agitated for re-suspension of settled solids before any tests were conducted[9]. The samples were characterized in terms of pH, NH_4-N, COD, turbidity and color. Table 1 and 2 show the general characteristics of samples determined in accordance with the Standard Methods for the Examination of Water and Wastewater[21].

Coagulation-Flocculation: A hydrolyzed solution of PACl with the formula Al(OH)xCly (where x is in the range 1.35–1.65, and y=3−x) and pH 2.3–2.9 (due to the presence of hydrochloric acid) was supplied by Hasrat Bestari Sdn Bhd, Penang, Malaysia. An 18% solution of PACI was used as a stock solution throughout the experiments. Coagulation-flocculation experiments were performed in a conventional jar-test apparatus (VELP-Scientifica, Model:J LT6,Italy) comprising of six paddle rotors (2.5cmx7.5cm), equipped with 6 beakers of 1L each. The time and speed for rapid and slow mixing were set with an automatic controller. For each experiment, each beaker used for testing was filled with 500 ml of sample. Before rapid mixing, different dosages of PACl were added to each beaker together with a pH adjustment using sulphuric acid and sodium hydroxide. For this research, the operating parameters were adopted as rapid mixing speed 80 rpm, slow mixing speed 30 rpm, rapid mixing time 2 min, slow mixing time 30 min and settling time 30 min.

Analyses: After accomplishment of the jar-tests, supernatant samples were withdrawn from the beaker by using a plastic syringe from the point located about 2 cm below the liquid level for the analysis. pH value was measured by pH meter (CyberScan 20) while turbidity was measured using 2100N HACH laboratory turbidimeter. COD concentration was determined using closed reflux and Colorimetric Method (5220D). NH_3-N was determined using Nesslerization method (4500-NH_3). COD was determined using Colorimetric Method (5220-D). Color measurements were reported as true color,filtered using GC-50 filter papers (Advantec Toyo Kaisha Ltd., Japan) with 0.45 µm pore size, assayed at 455 nm using DR2800 HACH spectrophotometer. Method No.2120C reports color in Platinum-cobalt (PtCo), the unit of color being produced by 1 mg platinum/L in the form of the chloroplatinate ion. The effect of filtration on color removal was corrected by means of a control sample. Removal efficiency for parameters was obtained using the following formula.

$$\text{Removal } (\%) = [(C_i - C_f)/C_i] * 100 \qquad (1)$$

where C_i and C_f are the initial and final concentrations of leachate sample in mg/L, respectively.

Results and discussion

Coagulant dosage plays an important role in the removal of target pollutants in coagulation-flocculation studies. The optimum dosage of coagulant is defined as a value above which there is no significant increase in removal efficiency with further addition of the coagulant[22].

The coagulant dosages of PACl for both leachates ranged from 0 mg/L to 9000 mg/L at varying increments. The effect of different dosages of PACI on AJLS leachate is shown in Fig. 1 while fig. 2 shows the effect of various PACl dosages on KLS leachate. The removal of COD increased initially but a decreasing trend was observed at PACl dosage of 2700 mg/L followed by an increase when the dosage was increased to 3600 mg/L (fig. 1). However, beyond this dosage, a consistent increase in COD removal was observed and it reached maximum at PACl dosage of 7200 mg/L (fig. 1). Interestingly, the removal pattern for NH_3-N was similar to COD removal initially; however, for dosages beyond 3600 mg/L, the removal pattern varied greatly and reached maximum at 5400 mg/L PACl followed by a decrease with further increase in PACl dosage (Fig. 1). As shown in Fig. 1, the removal of color was more consistent with increasing PACl dosage and it reached maximum when PACl dosage increased to 8100 mg/L.

According to fig. 2, for KLS leachate, the removal trend of COD was different to that of AJLS. A smooth decrease in COD was observed with increasing PACl dosage and it peaked at a dosage of 4500 mg/L followed by a consistent decrease with further increase in the dosage of PACl (fig. 2). Again, NH_3-N showed a peculiar trend and its removal was not consistent with the increasing dosages of PACl as shown in Fig. 2. Although the removal of color remained almost same for PACl dosages of 900 and 1800 mg/L, it was found to decrease drastically when PACl dosage was increased to 2700 mg/L. However, it remained almost linear for dosages higher than 2700 mg/L (fig. 2).

Based on the above results, the optimum dosages of PACI for AJLS and KLS were chosen at 7200 mg/L and 4500 mg/L, respectively. At neutral pH, these optimum dosages gave COD removals of 50% and 67% for AJLS and KLS, respectively. It is well known that COD intensity is due to humic substances in the leachate. Therefore, with the addition of hydrolyzing coagulants such as PACI, humic substances were successfully removed from the leachate. The present results agree with those described elsewhere[23]. The difference in removal rate could be attributed to the age of the landfill sites. It should be noted that KLS is currently operational and is producing highly polluted leachate in comparison to AJLS which has been closed for more than 10 years, hence produces mature/stabilized leachate with relatively higher percent of recalcitrant compounds which are more difficult to remove.

The removal of color at optimum dosages was rather similar for both leachate types using the optimum PACl dosage and it ranged between 70-73% (Figs. 1 and 2). However, the percentage removal of NH_3-N was lower with merely 26% and 45% for AJLS and KLS, respectively. This is in agreement with the previous findings[4] whereby the authors used PACl as a coagulant for the treatment of semi-aerobic landfill leachate. Overall, the percent removal of color was higher than that of COD and NH_3-N for both sites regardless of the PACl dosages (figs. 1 and 2).

In general, it could be noticed that the percentage removal of parameters increased with an increase in coagulant dosage until reaching an optimum value. This could be attributed to the notion that an excessive amount of coagulant caused restabilization of colloids and re-dispersion of the colloidal particulates[9]. Hence, the process should be optimized to reduce both the operational costs and improve the removal efficiency. The use of local natural polymers as coagulant aid with PACl could also be considered to further reduce the overall coagulant dosage as reported in earlier works[24,25].

Conclusion

As a comparison, different optimum dosages were obtained for PACI at both the locations. Optimum dosage for PACI at KLS was lower than AJLS indicating lower coagulant requirements and possibly less sludge production. It could be concluded that treatment of landfill leachate from KLS and AJLS by coagulation-flocculation process in the presence of PACI as a coagulant was effective in the removal of color and COD but poor in case of NH_3-N removal. Hence, PACl could be used as an effective pretreatment step for further biological treatment.

Acknowledgement

The authors would like to acknowledge Universiti Sain Malaysia (USM) for providing grant to conduct this research. The authors also wish to acknowledge cooperation extended by the Majlis Pebandaran Seberang Perai, Majlis Perbandaran Kulim and the contractor Idaman Bersih Sdn. Bhd. Penang during this study.

References

1. Bashir M.J.K., Aziz H.A., Yusoff M.S. and Adlan M.N., Application of response surface methodology (RSM) for optimization of ammoniacal nitrogen removal from semi-aerobic landfill leachate using ion exchange resin, *Desalination,* **254**, 154-161 **(2010)**

2. Umar M., Aziz H.A. and Yusoff M.S., Trends in the use of Fenton, electro-Fenton and photo-Fenton for the treatment of landfill leachate, *Waste Manage,* **30**, 2113–2121 **(2010)**

3. Umar M., Aziz H.A. and Yusoff M.S., Variability of parameters involved in leachate pollution index and determination of LPI from four landfills in Malaysia, *Int. J. Chem. Eng.,* Article ID: 747953 **(2010)**

4. Aziz H.A., Daud Z., Adlan M.N. and Hung,Y.T., The use of polyaluminum chloride for removing colour, COD and ammonia from semi-aerobic leachate, *Int.J.Environ.Eng*., **1**, 20-35 **(2009)**

5. Renou S., Givaudan J.G., Paulain S., Dirassouyan F. and Moulin P., Landfill leachate treatment: review and opportunity, *J.Hazard. Mater*., **150**, 468-493 **(2008)**

6. Bashir M.J.K., Isa M.H., Kutty S.R.M. Awang Z.B., Aziz H.A., Mohajeri S. and Farooqi I.H., Landfill leachate treatment by electrochemical oxidation, *Waste Manage*., **29**, 2534-2541 **(2009)**

7. Aziz H.A., Yusoff M.S., Adlan M.N., Adnan N.H. and Alias S., Physico-chemical removal of iron from semi-aerobic landfill leachate by limestone filter, *Waste Manage*, **24**, 353-358 **(2004)**

8. Ghafari S., Aziz H.A., and Bashir M.J.K., The use of poly-aluminum chloride and alum for the treatment of partially stabilized leachate: a comparative study, *Desalination*, **257**, 110-116 **(2010)**

9. Aziz H.A., Alias S., Adlan M.N. Asaari F.A.H. and Zahari M.S., Colour removal from landfill leachate by coagulation and flocculation processes, *Bioresour. Technol*., **98**, 218–220 **(2007)**

10. Kulikowska D. and Klimiuk E., The effect of landfill age on municipal leachate composition, *Bioresour. Technol.,* **99**, 5981-5985 **(2008)**

11. Amokrane A., Comel A. and Verson, J., Landfill leachate pretreatment by coagulation-flocculation, *Water Res.,* **31,** 2775-2782 **(1997)**

12. Duan J.M. and Gregory J., Coagulation by hydrolyzing metal salts, *Adv. Colloid Interface Sci.,* **100–102**, 475–502 **(2003)**

13. Davis M.L. and Cornwell D.A., Introduction to Environmental Engineering, Fourth ed. Mc-Graw Hill, New York, **(2008)**

14. Tatsi A.A., Zouboulis A.I., Matis K.A. and Samaras P., Coagulation-flocculation pretreatment of sanitary landfill leachates, *Chemosphere*, **53**, 737-744 **(2003)**

15. Hu C., Liu H. and Qu J., Preparation and characterization of polyaluminum chloride containing high content of Al_{13} and active chlorine, *Colloids and Surf. A: Physicochem. Eng. Asp.* **260**, 109-117 **(2005)**.

16. McCurdy K., Carlson K. and Gregory D., Flocs morphology and cyclic shearing recovery: comparison of alum and polyaluminum chloride coagulants, *Water Res.,* **38**, 486–494 **(2004)**

17. Wang Y., Gao B.Y., Xu X.M., Xu W.Y. and Xu G., Characterization of floc size, strength and structure in various aluminum coagulants treatment, *J. Colloid Interf. Sci.,* **332**, 354-359 **(2009)**

18. Gao B., Hahn H.H. and Hoffmann E., Evaluation of aluminium –silicate polymer composite as a coagulant in water treatment, *Water Res*., **36**, 3573-3581 **(2002)**

19. Tapas N., Vyras R.D., Shastry S. and Kaul S.N., Optimization of coagulants for pre-treatment of printing ink wastewater, *Environ. Eng. Sci,*. **19**, 1-7 **(2002)**

20. Wang D., Sun W., Xu Y., Tang H. and John G., Speciation stability of inorganic polymer flocculant-PACI, *Colloid Surf. Physicochem. Eng. Asp.,* **243**, 1-10 **(2004)**

21. APHA, Standard Methods for the Examination of Water and Wastewater, Twenty first ed., American Public Health Association, Washington DC **(2005)**

22. Aguilar M.I., Sáez J., Lloréns M., Soler A., Ortuño J.F., Meseguer V. and Fuentes A., Improvement of coagulation-flocculation process using anionic polyacrylamide as coagulant aid, *Chemosphere*, **58**, 47-56 **(2005)**

23. Ghafari S., Aziz H.A. and Zinatizadeh A.A., Application of response surface methodology (RSM) to optimize coagulation-flocculation treatment of leachate using poly-aluminum chloride (PAC) and alum, *Hazard. Mater.,* **163**, 650-656 **(2009)**

24. Aziz H.A., Adlan M.N., Mohamed A.M.D., Raghavan S., Koflly M., Isa M. and Abdullah M.H., Study on the anionic natural coagulant aid for heavy metals and turbidity removal in water at pH 7.5 and alum concentration 25 mg/L-laboratory scale, *Ind. J. Eng. Mater. Sci.,* **7**, 195-199 **(2000)**

25. Al-Hamadani Y.A.J., Yusoff M.S., Umar M., Bashir M.J.K. and Adlan M.N., Application of psyllium husk as coagulant and coagulant aid in semi-aerobic landfill leachate treatment, *J. Hazard. Mater.,* doi:10.1016/j.jhazmat.2011.03.087 **(2011)**.

Table-1: General characteristics of raw leachate from Kulim Landfill Site (KLS)

Parameter	Unit	Range
pH		7.85-8.28
NH_3-N	(mg/L)	600-720
COD	(mg/L)	110-300
Turbidity	(NTU)	11.65-24.9
Color (PtCo)	(mg/L)	279-494

Table-2: General characteristics of raw leachate from Ampang Jajar Landfill Site (AJLS)

Parameter	Unit	Range
pH		7.13-8.25
NH_3-N	(mg/L)	258-570
COD	(mg/L)	117-260
Turbidity	(NTU)	15-20.3
Color (PtCo)	(mg/L)	134-287

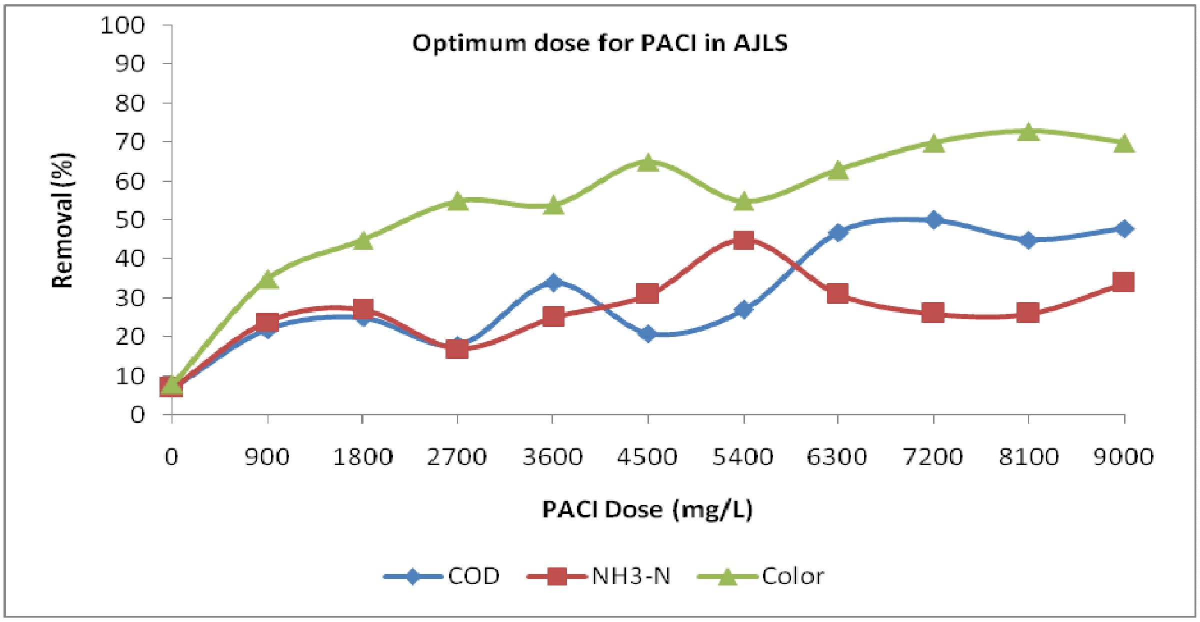

Figure-1: The effect of coagulant dosage on the removal of COD, NH_3-N and color for AJLS leachate

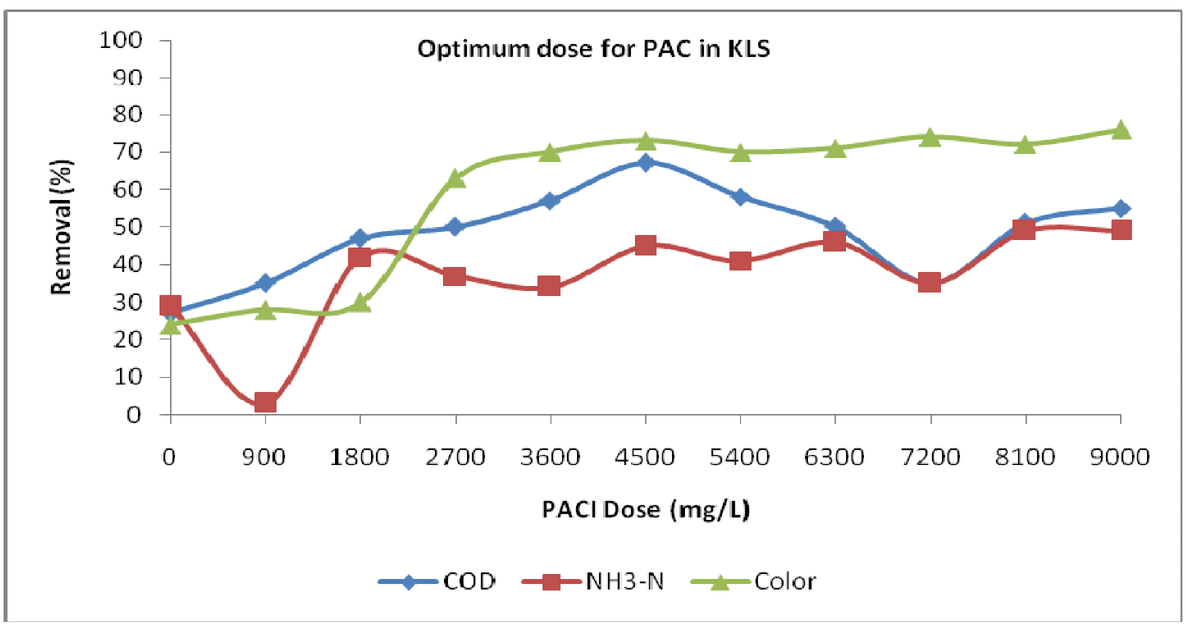

Figure-2: The effect of coagulant dosage on the removal of COD, NH_3-Nand color for KLS leachate