

Effects of using *Tamarindus indica* Seeds as a natural coagulant aid in landfill leachate treatment

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Abstract

Uncontrolled landfill leachate generation portends danger to the environment and aquatic ecosystem, especially without prior treatment before discharge. The application of Al-based coagulants, such as polyaluminum chloride (PACl), has the potential of introducing Al residuals into water bodies. Therefore, an alternative natural coagulant was recommended to reduce the usage of Al-based coagulants. In this study, a coagulation–flocculation process using the combination of PACl as coagulant and *Tamarindus indica* seed (TiS) as coagulant aid was used in treating the landfill leachate from the Alor Pongsu Landfill Site in Malaysia. Some of the optimum operational conditions determined were the pH and dosage of the coagulant aid, and their effect was considered on parameters, such as suspended solids (SS), color, and COD, using standard jar test procedures. The combination of TiS flocculant reduced the dosage of PACl coagulant from 5,000 mg/L to 2,750 mg/L with removal efficiencies of 99.3%, 97.3%, and 67.4% for SS, color, and COD, respectively.

Keywords: *Tamarindus indica*; PAC1; landfill leachate, coagulant, coagulant aid

1. Introduction

Landfill leachate is generally referred to as a complex liquid that may contain a large amount of organic matters, ammonia–nitrogen, suspended solids (SS), chlorinated organic and inorganic salts, and heavy metals (Foo and Hemeed, 2009; Bashir *et al.*, 2010). The disposal of landfill leachate can be a source of severe pollution, especially for soil, water, and groundwater contamination (El-Salam and Abu Zaid, 2015). Similarly, public health hazards may also be caused by the discharge of untreated landfill leachate, especially into water bodies (Akinbile *et al.*, 2012; Al Hamadani *et al.*, 2011; Aziz *et al.*, 2011). In an attempt to prevent an occurrence of this scenario, related government regulatory agencies are becoming more stringent and restrictive in the enforcement of environmental rules and regulations concerning the monitoring of contaminants

from leachate waste streams (Maizatun, 2011). Adequate and appropriate landfill leachate treatments have been widely proposed to alleviate challenges associated with untreated leachate (Akinbile *et al.*, 2016; Amuda *et al.*, 2006; Aziz and Mojiri, 2014). Many well-documented treatment techniques are available, including biological, physical, or physicochemical methods (Akinbile *et al.*, 2016; Aziz *et al.*, 2011; Alias *et al.*, 2010). Several types of physicochemical treatments, such as chemical precipitation, coagulation–flocculation, adsorption, ion exchange, and ammonium stripping, are available (Kamaruddin *et al.*, 2013; Turovsky and Mathai, 2006). The coagulation–flocculation process is a comparatively simple and widely applied technique used in water and wastewater treatment (Aziz and Mojiri, 2014; Aziz *et al.*, 2007; Amuda *et al.*, 2006) and has also been proven as a cost-effective treatment approach in landfill leachate treatment (Rajaram *et al.*, 2011). The mechanism involves charge neutralization by adding coagulants to destabilize negatively charged colloids for the agglomeration of fine particles and colloids into large particles, thereby reducing turbidity, natural organic matter, and other soluble organic and inorganic pollutants in wastewater (Teh *et al.*, 2016). The commonly used commercial coagulants are aluminum sulfate (alum), polyaluminum chloride (PACl), ferrous sulfate, ferric chlorosulfate, and ferric chloride (Lee, 2013). Although inorganic coagulants are generally effective, some drawbacks are observed related to the high amount of metal ions in sludge (Mishra, 2016). On the contrary, natural coagulants are found to produce a relatively low sludge volume and are safe to humans compared with inorganic coagulants (Madhukar and Yogesh, 2013). Although natural coagulants have been widely applied in wastewater treatment (Santos *et al.*, 2016; Kos, 2016), their application in landfill leachate treatment is limited despite their abundance, relatively low cost, and being environmentally friendly (Meraz *et al.*, 2016).

Tamarind is widely spread throughout Asia and Southeast Asia, especially in India and Thailand where the species is planted in large plantation scales and is economically important. In comparison with these two countries,

tamarind is not commonly found or seen throughout Malaysia, except in the northern region where the ecological condition is better suited than that in the other regions. No actual data are available on the production and area of tamarind plantation in Malaysia. Tamarind is mostly grown in small-scale plantations. It is mainly cultivated in Penang and Kedah. Obtaining precise data on the production and acreage of this species is relatively difficult due to its small production. Furthermore, most of the fruits are either collected from the wild by rural people or harvested in isolated areas. Some of the tamarinds are also imported and repacked locally. A number of food factories process and sell tamarinds in the form of seedless paste, in which the seed is disposed untreated. The actual number of factories is not available. As tamarind is one of the most common ingredient added in many dishes, its wide usage is expected for over 28 million of the current population. Furthermore, tamarind is a common ingredient in Malay, Indian, and Chinese cuisines in Malaysia and a “must-have” ingredient in almost all dishes.

Several studies have been conducted on the application of *Tamarindus indica* seed (TiS) in the coagulation–flocculation method, as well as adsorption. Mostly, the application of TiS in the coagulation–flocculation process involves the use of low concentrated wastewater. For example, Phani Madhavi and Rajkumar (2013) used TiS powder for the treatment of low turbid wastewater. They achieved 78% removal turbidity at optimum pH of 8 and optimum dosage of 150 mg/L. Murugan and Subramanian

(2006) applied tamarind seeds as an absorbent in water treatment method. Total defluoridation and subsequent regeneration of adsorbent were performed with a household water filter and fixed bed column for domestic and industrial applications, respectively, and maximum defluoridation was achieved at pH 7.

The focus of previous studies when using TiS as coagulants in the coagulation–flocculation process has been mostly for the treatment of turbid or wastewater, whereas any utilization for leachate treatment, either as a primary coagulant or as a flocculant, is hardly available. Therefore, the main motive of this study is on the effectiveness of utilizing TiS as a coagulant for landfill leachate with the combination of PACl in removing SS, color, and COD. This study also aims to determine the optimum dosage of TiS and PACl as coagulants for landfill leachate treatment by conducting performance evaluation on the parameters of SS, color, and COD.

2. Materials and methods

2.1 Leachate Sampling and Characterization

Leachate samples were collected from the Alor Pongsu Landfill Site (APLS) in Bagan Serai, Perak, Malaysia from January to April, 2016. Sampling was conducted using the grab sampling method, whereas preservation was performed in accordance with the Standard Methods for the Examination of Water and Wastewater (APHA, 2005). Table 1 presents the initial characteristics of the six leachate samples obtained.

Table 1. APLS Raw Leachate Characteristics

Parameters	Unit	Min	Max	Average
Temperature	°C	28.19	38.85	31.34
pH	-	7.78	8.12	7.99
DO	mg/L	0.05	0.27	0.17
COD	mg/L	3610	4113	3925
BOD ₅	mg/L	107	176	131
BOD ₅ /COD	-	0.03	0.05	0.03
NH ₃ -N	mg/L	1010	1480	1296
TSS	mg/L	356	478	397
TDS	g/L	9.13	9.61	9.35
Turbidity	NTU	33.4	49.9	40.1
Colour	Pt Co	10317	18733	13787
Conductivity	μS/cm	15134	18413	16128
Zeta Potential	mV	-24.8	-21.6	-23.2
Chromium (Cr)	mg/L	0.03	0.15	0.09
Iron (Fe)	mg/L	8.66	10.91	9.96
Manganese (Mn)	mg/L	0.15	0.38	0.26
Zinc (Zn)	mg/L	0.15	1.04	0.46

2.2 Preparation of TiS as Coagulant

Fresh *T. indica* fruits at 500 ± 0.5 g were obtained from the local market and prepared using distilled water. The seeds were then oven-dried for 1 h at 105 °C and, once weakened, the seeds were crushed using a mortar and pestle to separate the seed husks and kernels. The crushed kernels were oven-dried for 1 h to ensure that the seeds

were completely dried prior to milling to produce powder by using a ring mill. TiS at 400 g was weighed and blended with distilled water using a domestic blender for 2 min to extract the active ingredients. The suspension was filtered through a filter paper into a beaker. The TiS powder (50%) in the solution produced a concentration of 200,000 mg/L, which was ready to be utilized as a natural

coagulant/flocculant for the jar test experiments (Alias *et al.*, 2010).

2.3 Characterization of Leachate and TiS as a Function of pH

In determining the effect of pH on the zeta potential and particle size of the leachate and TiS, the leachate samples and TiS solutions were separately titrated using 0.5 M HCl and 0.5 M NaOH within a pH range of 2–12 under continuous agitation. Zetasizer Nano ZS was used in measuring the zeta potential and particle size in triplicate runs. A sample at 1 mL was administered into the capillary cell by using a pipette and placed into the Malvern Zetasizer Nano ZS. During measurement, samples were assayed by a 633 nm He–Ne laser beam using dynamic light scattering method with a scattering angle of 173° (Omar, 2015). The instrument software program was run, and the zeta potential value was generated using Smoluchowski equation. For particle size measurement, a transparent, disposable cuvette was used in place of the capillary cell. Different standard operating procedures were used for the measurement of zeta potential and particle size. The graphs of two y-axes consisting of zeta potential and particle size versus pH value were generated separately for leachate and TiS. From the results obtained, the values of IEP and PZC for the leachate and TiS were identified.

2.4 Jar Test

Coagulation test was performed by using a jar test equipment (SW6 Stuart Bibby Scientific Limited, UK). NaOH and HCl at 3 M each were used to adjust the pH sample. The test involved rapid mixing, slow mixing, and sedimentation in a batch process. For jar tests using 18% PACl as the main coagulant, the rapid mixing was set at 200 rpm for 3 min and slow mixing at 40 rpm for 30 min; the settlement was 30 min (Zainol *et al.*, 2013). Meanwhile, the operational conditions of the jar tests using TiS as the main coagulant were rapid mixing at 100 rpm for 8 min and slow mixing at 30 rpm for 20 min, and the settlement was 30 min (Muyibi *et al.*, 2002). Leachate samples at 500 mL were filled into six beakers and agitated simultaneously while varying the rotational speed and allowing the simulation of different mixing intensities and resulting flocculation process (Mokhtar *et al.*, 2011).

3. Results and discussion

3.1 Characteristics of Leachate Zeta Potential and Particle Size as a Function of pH

A general but gentle decline was observed in the trend of the zeta potential of leachate when compared with the observed increase in pH. This trend suggested that the zeta potential was closely dependent on the pH value of the leachate (Fig. 1). The zeta potential decreased with the increase in pH value. Lin *et al.*, (2013) remarked that a strong surface charge in the particle could result in a significant mutual repulsion. Similarly, an irregular sinusoidal trend was observed in the behavior of the particle size distribution of the leachate with respect to the increasing pH ranging from 2 to 12, as shown in Fig. 1. At

an approximately natural pH value of 8 for the leachate, the zeta potential was found to be at -23.43 ± 1.46 mV, which indicated that the surface charge of the leachate was negatively charged. For the coagulant to be highly effective, the zeta potential is required to be higher than +20 mV, such that it can neutralize and attract the particles to form flocs, which agrees with the findings of Al-Hamadani *et al.*, (2011). As soon as the pH was adjusted to the acidic region, the surface charge of the leachate became less negative and approached -5 mV (Fig. 1), which caused the particle size of the leachate to increase. In view of the zeta potential between pH values of 4 and 7, stabilization in the particle size of the leachate was observed, whereas the zeta potential remained the same at an approximate value of -20 mV. Within the alkaline region of pH values of 8 and 11, the particle size of the leachate remained unchanged at an approximate value of 320 d.nm, whereas the zeta potential of the leachate was constantly decreasing and becoming highly negatively charged. Electrostatic repulsion of the particles occurred, thereby increasing the size of particles. However, when the pH value approached 12, the particle size of the leachate decreased to 246 d.nm and tended to reduce further under high alkaline pH value of 14, which agrees with the findings of Kamaruddin *et al.*, (2015).

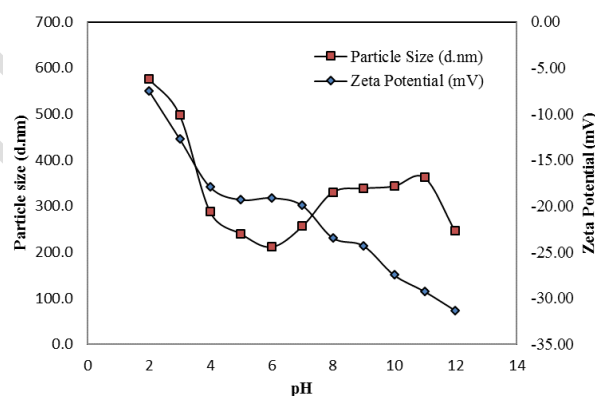


Figure 1. Characteristics of Leachate as a function of pH

3.2 Characteristics of TiS Zeta Potential and Particle Size as a Function of pH

Fig. 2 shows the characteristics of the zeta potential and particle size for TiS under varying pH values of 2–12. A slightly distinctive decline in the zeta potential of TiS was observed from the pH value of 2–11. Meanwhile, an extremely sharp decline was recorded for pH values of 11–12 with the zeta potential values decreasing from -0.3 mV to -36.83 mV (Fig. 2). Moreover, a sharp contrast was observed in the particle size when a mild undulating trend was noticed for the pH values from 2 to 8. The value range of the particle size was from 17 μ m to 38 μ m. From pH values of 8–12, a sudden increase was observed as the particle size increased from 20 m to 169.874 m (Fig. 2). Overall, the behavioral trend of the particle size of TiS was the opposite of the zeta potential trend, where the particle size of TiS increased with the pH value. At pH 2, the zeta potential was positive at 1.87 ± 0.49 mV, whereas the

particle size recorded was 31.879 m. Thereafter, the zeta potential tended toward zero charge as the pH value increased. Within the alkaline region, the zeta potential fluctuated from the range of -2.50 mV to -4.00 mV, whereas the particle size increased rapidly with an increasing pH value. All these observations agree with the findings of Aziz *et al.*, (2011).

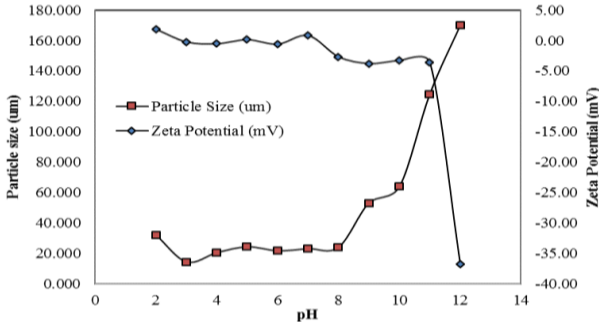


Figure 2. Characteristics of *Tamarindus indica* Seeds (TiS) as a function of pH

3.3 Optimum Dosage for TiS as a Flocculant in the Removal of SS, Color, and COD

The optimum dosage of TiS as a flocculant was determined by using jar test procedures with rapid mixing at 200 rpm for 3 min, slow mixing at 40 rpm for 30 min, and 30 min settlement (Zainol *et al.*, 2013). The tests were conducted at 31.3 °C; the initial concentrations of SS, color, and COD were 397 mg/L, 13,787 Pt Co, and 3,925 mg/L, respectively. Fig. 3 presents the removal efficiency of SS with various dosages of PACl as coagulant and TiS as flocculant. High efficiency in SS removal was observed when specific quantities ranging between 3,000 mg/L and 4,000 mg/L of PACl were introduced with varied dosages of TiS into the leachate. From the figure, the introduction of a low PACl dosage of 2,750 mg/L with 2,000 mg/L of TiS was found to be highly efficient at 99.3% in removing SS from the leachate. Identical performance was achieved using 5,000 mg/L of PACl as the sole coagulant, which provided a 99.5% removal efficiency. Ebeling *et al.*, (2005) confirmed this high efficiency in a similar study conducted.

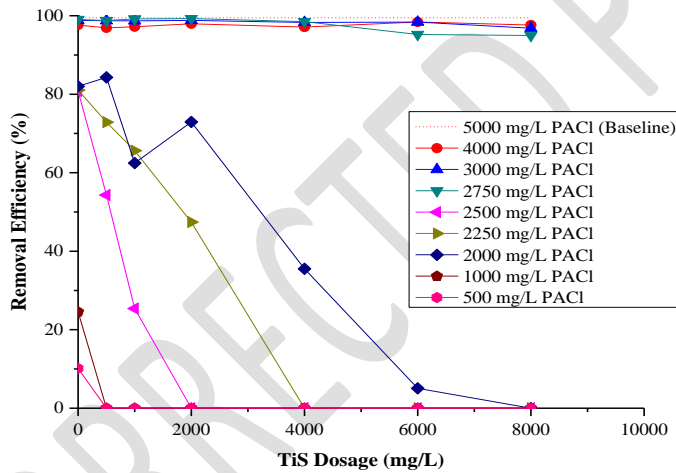


Figure 3. SS removal efficiency with varied dosage of PACl as coagulant and TiS as flocculant

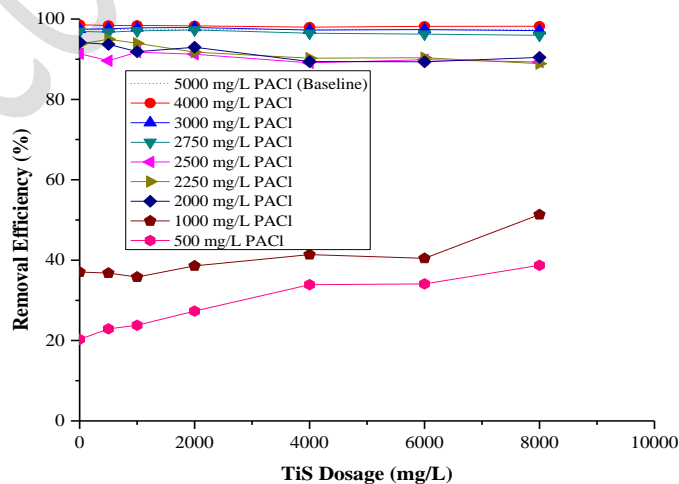


Figure 4. Colour removal efficiency with varied dosage of PACl as coagulant and TiS as flocculant

In the color removal (Fig. 4), using 2,000 mg/L of PACl with 2,000 mg/L of TiS as flocculants achieved 93.0% removal

efficiency, whereas 97.4% removal efficiency was obtained when the initial TiS values were used with 5,000 mg/L of

PACl as the sole coagulant. However, when the PACl dosage was lowered to 1,000 mg/L and below, the efficiency of color removal declined considerably and therefore confirmed the assertion of Aziz *et al.*, (2007), who suggested that color in landfill leachate is mostly presented by organic matters with some unsolvable forms that reveal turbidity and SS.

The removal efficiency of COD by using various dosages of PACl as coagulant and TiS as flocculant was unsatisfactory,

as shown on Fig. 5. Further increase in TiS dosages ironically further reduced the efficiency of COD removal. However, when comparing the removal efficiencies of using 2,750 mg/L of PACl with 2,000 mg/L of TiS with using 2,750 mg/L of PACl only as the primary coagulant, the COD removal efficiency increased to 67.4% for the former and 73.6% for the latter. Further reduction in PACl dosage to 2,500 mg/L was observed to contribute to a 60% removal efficiency (Fig. 5).

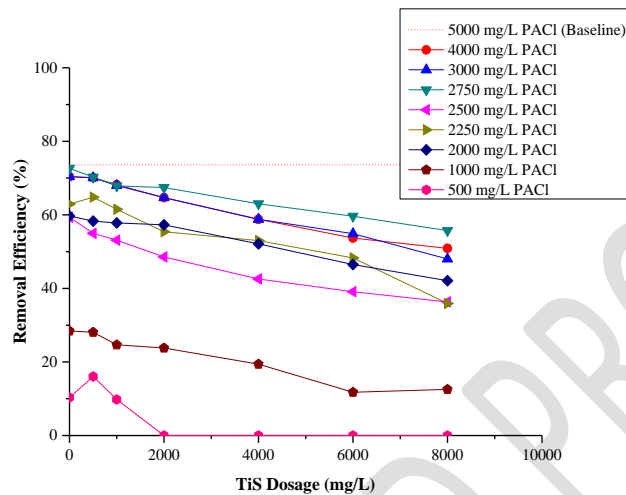


Figure 5. COD removal efficiency with varied dosage of PACl as coagulant and TiS as flocculant

3.4 Efficiency of TiS as Flocculant

A comparative analysis of the best combination of PACl and TiS obtained in Figs. 3, 4, and 5 was performed and reported in Fig. 6. From this figure, identical results were obtained for SS and color but a slightly different outcome was observed for COD. The removal efficiencies for SS and color removal differed by 0.02% and 0.01%, respectively, which indicated non-significant differences. Lowering the

amount of PACl by introducing TiS reduced the COD removal by 6.2%. This slight difference remained within the acceptable range. In comparison of the overall performance of all the three removal parameters, with 2,000 mg/L of TiS, the amount of PACl usage for leachate treatment could be reduced from 5,000 mg/L to 2,750 mg/L, which would result in a 45% reduction in PACl dosage.

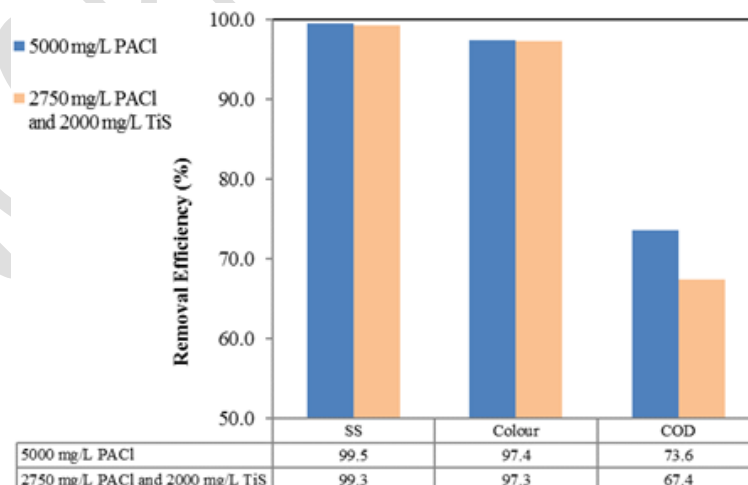


Figure 6. Comparison of pollutants removals at optimum conditions of PACl with and without TiS

The efficiency of TiS as the flocculant with PACl as the primary coagulant could be further proven through the performance of floc obtained from the coagulation–flocculation process. Flocs obtained from the jar test with 5,000 mg/L of PACl and the combination of 2,750 mg/L of

PACl as coagulant and 2,000 mg/L of TiS as flocculant were characterized and compared. No significant floc formed while using TiS as the sole coagulant in the jar test. The floc was characterized in terms of particle size. Figs. 7 and 8 show the floc sizes formed with different PACl dosages and

a constant PACl dosage of 2,750 mg/L with different TiS dosages, respectively. From Fig. 7, the largest floc size was obtained by using a PACl dosage of 5,000 mg/L as the sole coagulant compared with other dosages with a floc size of 107.815 μ m. Then, a gradual decrease in floc size with increasing applied dosages was observed. Fig. 8 presents the result of floc size formed when combining 2,750 mg/L of PACl as coagulant with 2,000 mg/L of TiS as flocculant. The largest floc size of 153.496 μ m was observed at 2,750 mg/L of PACl, which is consistent with the findings of Lin *et al.* (2013).

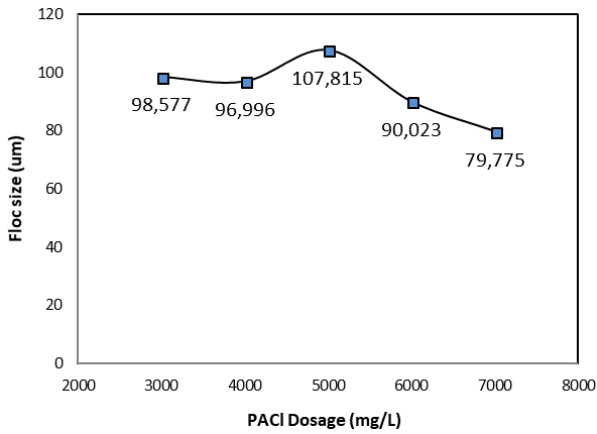


Figure 7. Size of floc formed by using various dosages of PACl as a sole coagulant

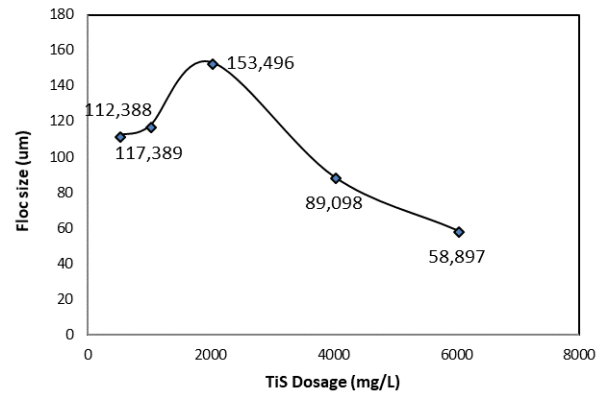


Figure 8. Size of floc formed by using 2,750 mg/L PACl as a coagulant and various dosages of TiS as a flocculant

Fig. 9 presents the comparison of the particle sizes between two treatments and the raw leachate. The sizes of floc formed when combining PACl and TiS were considerably large compared with the particle size of the raw leachate, which indicated an effective floc formation. Effective floc formation would enhance floc performance, thereby further enhancing the efficient removal of pollutants. The floc size formed by applying the combination of PACl and TiS was larger than that when PACl was used alone. Thus, TiS aided in the aggregation of colloids and improved the performance of sludge settling. This result was due to the TiS having a high molecular weight, which was therefore able to provide long bridges during the mechanism of bridging in the flocculation process, as well as enhance the floc size (Amuda *et al.*, 2006).

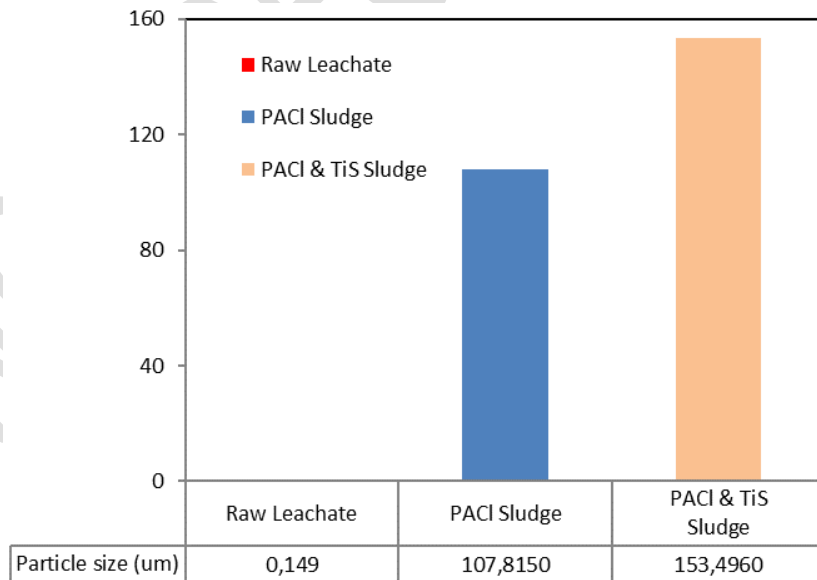


Figure 9. Comparison of particle size between raw leachate and sludge of PACl in the presence and absence of TiS

Generally, a flocculant with high molecular weight produces a relatively large floc size (Ebeling *et al.*, 2005). The floc sizes recorded from the jar test were all smaller than 0.2 mm. Hence, the organic part of the sludge decays rapidly with the increase in the quantity of finely dispersed and colloidal particles; this phenomenon will result in the

decrease in the separation of water from the sludge and poor dewaterability (Turovsky and Mathai, 2006).

3.5. Coagulant Cost Estimation

Generally, the leachate treatment cost depends on factors, such as landfill design, quantity of leachate, level or degree

of treatment needed, and final removal method for residues and effluent. Obtaining data on the cost of leachate treatment is difficult because it requires the cooperation of the company in charge. Therefore, on the basis of the chemicals used, the costs of both coagulants are estimated (Al-Shukaili and Agamuthu, 2008). Table 2

Table 2. Cost comparison

Coagulant	Price of Chemical (RM)	Optimum Concentration Used	Amount of Chemical to Treat 1m ³ of Leachate/day	Cost to Treat 1 m ³ of Leachate (RM)	Total (RM)
5000 mg/L PACI	300/L	13mL/500mL	26 L	7,800	7,800
2750 mg/L PACI	300/L	7.6mL/500mL	15.2 L	4,560	4,646
2000 mg/L TiS	4.30*/kg	10g/500mL	20 kg	86	

*1 RM = Ringgit Malaysia = 3.91 USD, (Al-Shukaili and Agamuthu, 2008; Choenkwan et al., 2014)

4. Conclusion

Applying PACI as the sole coagulant at pH 6 and concentration of 5,000 mg/L successfully removed SS (99.5%), color (97.4%), and COD (73.6%), whereas pH 4 and TiS dosage of 5,000 mg/L were identified as the optimum pH and coagulant dosage when TiS was used as the sole coagulant. This case resulted in color and SS removal of 41.9% and 5.9%, respectively, whereas SS removal was found to be insignificant. At optimum operating conditions of pH 6, when the combination of PACI and TiS was applied, such that PACI was used as the main coagulant and TiS as flocculant, PACI usage was reduced to 2,750 mg/L with the aid of 2,000 mg/L of TiS. The removal efficiencies achieved were 99.3%, 97.3%, and 67.4% for SS, color, and COD, respectively, which were comparable to the treatment performance with the usage of 5,000 mg/L of PACI. Therefore, TiS has the potential to be used as a natural coagulant/flocculant in landfill leachate treatment.

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shows the cost comparisons when PACI was used alone and when TiS was used as the flocculant. From the comparisons, a cost of RM 7,800 was determined when 5,000 mg/L of PACI was used alone, whereas the use of 2,000 mg/L of TiS as flocculant only costed RM 4,646.

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