Dosage and pH optimization on stabilized landfill leachate via coagulation-flocculation process

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Abstract. Treatment on the generated landfill leachate is crucial as it can cause serious toxicological effects and environmental hazards, particularly when the unfavorable contaminants are left accumulated for a long period of time. The purpose of this study was to determine the optimum coagulant dosage of polyaluminium chloride (PAC) in selected dosage ranges (2250-4500 mg/L) and to analyse the ideal pH of leachate sample (pH 3-10). PAC was tested on stabilized leachate taken from Simpang Renggam Landfill Site (SRLS), by investigating the percentage removals of five significant parameters, which were suspended solids, chemical oxygen demand (COD), ammonia, and heavy metals (iron (Fe) and chromium (Cr)). The removal efficiency was determined by a series of experiments using jar test. From the obtained results, it was found that 3750 mg/L and pH 7 were the optimum conditions for PAC dosage and sample pH, respectively. The conventional optimization test showed satisfactory results for suspended solids, COD, Fe, and Cr at 95%, 53%, 97%, and 79% respectively, but had low removal on ammonia at 18%. It can be concluded that the coagulation-flocculation process has the potential to be applied as a primary treatment for stabilized landfill leachate in Malaysia.

1 Introduction

The age of landfill and long accumulation of contaminants will promote the degradation of organic matter present, thus influence the characteristics of leachate produced [1, 2]. As the landfill get older, the activity of anaerobic decomposition in the site will reduce the biodegradable fraction of organic pollutants, thus cause stabilized leachate become higher polluted wastewater compare to the young ones [2]. The high accumulation of inorganic toxic compounds such as heavy metals and ammonia over a long period time are recognized as toxicants to the living organisms, which can cause further damaging consequences [3]. Typically, old leachate is harder to be treated. Stabilized leachates can be acknowledged through its BOD5/COD ratio that less than <0.1 [4]. This address that it is

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important to carry out leachate characterization first to have a successful treatment strategy [5, 6].

### Table 1. Summary of BOD$_5$/COD corresponding to landfill age [4].

| BOD$_5$/COD | Age of landfill | COD     |
|-------------|-----------------|---------|
| ≥ 0.5       | Young (<5 year) | >10,000 |
| 0.1-0.5     | Medium (5-10 year) | 500-10,000 |
| <0.1        | Old (>10 year)  | <500    |

Physical-chemical treatments like coagulation-flocculation method is suitable for stabilized leachate to remove the refractory substances, with high efficiencies depending on the type of coagulants [7]. Coagulation-flocculation is one of the applications that work efficiently on stabilized leachate, alongside the utilization of chemical coagulants. These parameters need to be controlled to come out with the satisfying and optimized impurities/contaminant reduction. Coagulation needs flocculation process to work efficiently. Coagulation by itself does not help much when the addition of coagulant may increase the insoluble compounds in treated sample, thus flocculation step is needed by allowing slow mixing key to obtain optimum performance [8]. In this study, the coagulant dosage and pH are selected to be the manipulated variables due to its dominance effect, while the temperature, jar test mixing speed, and settling time are controlled on their respective working conditions.

PAC has good structure of high charged density that gather much attention due to its non-limited optimum conditions [9, 10]. Besides, low dosage applications showed excellent coagulant activity in previous reported studies [11–13]. PAC coagulants are claimed to be effective than other aluminium preparations particularly at low temperature or acidic pH for the coagulation of high turbidity waters [13]. The positively charged of Al (OH) precipitates help in adsorbing humic substances and improving flocculation kinetics [13]. As yet, there is no physicochemical method specialize in optimizing of coagulants doses by coagulation-flocculation, except using jar test. In particular, the combined coagulation–flocculation process is adopted to eliminate suspended particles and insoluble substances, which is being a challenge for the removal of toxic heavy metals from leachates due to the mixed substances present [14].

## 2 Study Area

The study site is at Simpang Renggam landfill site (SRLS), located in Simpang Renggam, Kluang, Johor (1°53'41"N 103°22'35"E). The location of SRLS is about two kilometres away from Simpang Renggam town. The landfill has about six hectares in total size. The landfill receives 400-500 tonnes/day of wet solid wastes from three covered areas, which are Simpang Renggam, Kluang, and Batu Pahat.

Based on the physical observation and interviews, the available treatments in the landfill were still insufficient. Based on the preliminary data of leachate characterization, the landfill site alleged to generate stabilized leachate [15] which did not fit for biological treatment adoption. Thus, coagulation-flocculation was applied.
3 Methodology

3.1 Leachate sampling

The sampling point was located at the entrance of leachate pond. The sampling and storage of leachate samples were carried out according to APHA standard method. Samples of leachate influent was collected manually by grab sampling and immediately transferred into a cool room of 4°C at Waste Water Laboratory, Faculty of Civil and Environmental Engineering, Universiti Tun Hussien Onn Malaysia (UTHM). In the laboratory, the samples were tested as soon as possible for chemical oxygen demand (COD), biochemical oxygen demand (BOD), ammonia, suspended solids, and heavy metals (Fe and Cr). The characteristics of selected parameters are able to indicate the type and suitability of leachate for this study.

Table 2. Characteristic of leachate in January, 2018 from this study.

| Parameter         | Average Value |
|-------------------|----------------|
| pH                | 8.76           |
| COD (mg/L)        | 2954           |
| BOD$_5$ (mg/L)    | 166.89         |
| Ammonia (mg/L)    | 920.4          |
| Suspended solid (mg/L) | 275        |
| Fe (mg/L)         | 7.61           |
| Cr (mg/L)         | 0.56           |
| BOD$_5$/COD       | 0.058          |
3.2 Preparation of coagulant

PAC has a good structure of having higher charged density, been gather much attention due to its non-limited optimum conditions and low dosage utilization. It has been committed excellent coagulant activity in previous reported studies [9], [11]–[13], [16]. PAC coagulant is claimed to be effective than other aluminium preparations particularly at low temperature or acidic pH for the coagulation of high turbidity waters [13]. The positively charged of Al (OH) precipitates help in adsorbing humic substances and improving flocculation kinetics [13]. According to the studies made by [12], the optimum pH for PAC is favourably higher than alum and showed efficient results in the view of physical-chemical treatment when tested on partially stabilized leachate, which also a highly polluted wastewater. Figure 2 shows the scanning electron microscopy (SEM) image of PAC in 10% stock solution at 600x magnification. The SEM analysis of PAC was done at the Environmental Analysis Laboratory, Faculty of Civil and Environmental Engineering, UTHM.

Commercial PAC was purchased from local laboratory supplier. The preparation of PAC stock solution was prepared according to [16], [19]. 10 grams of PAC was weighed and diluted in 100 ml of distilled water, to produce 10% concentration of stock solution coagulant. The stock solution was prepared on the same day as the experiment jar test experiment.

Fig. 2. SEM image of PAC in 10% stock solution at 600x magnification.

3.3 Working conditions (Jar test, coagulant dosage, pH)

All working conditions in this study were done by following the methodology of [17], as in Table 3 that shows the ranges of operating parameters for coagulation-flocculation test. The working conditions of jar test were applied to obtain the respective optimum pH of leachate sample and PAC dosage. The performance of coagulant in this study was compared with other utilizations of PAC in past studies in order to collate its efficiency. The obtained results were analysed using Excel to obtain the optimum value using graph; this procedure is also known as the conventional optimization method.

A coagulation process consists of three distinct steps. Firstly, the PAC coagulant was added to the sample with a determined pH value of leachate, before rapid mixings was initiated. The aim was to obtain a complete mixing of the coagulant with the leachate sample in order to maximize the effectiveness of the destabilization of colloidal particles.
Next, the suspension was slowly stirred to increase contact between the coagulation particles and to initiate the development of large floc in the flocculation process. Third, the mixing was stopped and the flocs was allowed to settle. After that, supernatant was collected three centimetre (cm) from the surface by using a plastic syringe for analytical measurement. The collected leachate sample was analysed to obtain the optimum conditions based on COD, suspended solids, ammonia, Fe, and Cr removals (standard method listed in Table 4). The removal efficiency of COD, suspended solids, ammonia, Fe, and Cr were calculated based on the equation below;

\[
\text{Percentage removal (\%) = } \left(\frac{\text{Initial concentration} - \text{Final concentration}}{\text{Initial concentration}}\right) \times 100
\]

\[\text{(1)}\]

**Table 3.** Ranges of operating parameters for coagulation-flocculation test.

| Parameter                     | Working condition          |
|-------------------------------|----------------------------|
| Speed of rapid mixing (rpm)   | 200                        |
| Duration of rapid mixing (min)| 4                          |
| Speed of slow mixing (rpm)    | 30                         |
| Duration of slow mixing (min) | 15                         |
| Settling time (min)           | 30                         |
| Temperature (°C)              | Room temperature (20-25°C) |
| pH                            | 3-10                       |
| Dosage                        | 2250-4500 mg/L             |

**Table 4.** List of used standard methods in this study according to Standard Methods for Water and Wastewater (APHA).

| Parameter                  | Methods                          |
|----------------------------|----------------------------------|
| Chemical oxygen demand (COD)| APHA Method: 5220 C              |
|                            | HACH Method: 8000                |
| Suspended solids           | APHA Method: 2540 D              |
|                            | HACH Method: 630                 |
| Ammonia                    | HACH Method: 8038                |
| Fe and Cr                  | APHA Method: 3120                |

**4 Result and Discussion**

**4.0 Optimization of coagulation-flocculation**

Determination of optimum condition for coagulation-flocculation using selected variables was taken place by using conventional method of ‘change one factor at a time’, which involved the trial and error practices [17]. Specifically, the coagulant dosage and pH were the considered factors in this study. By using this method, pH factor was fixed at 7 while coagulant dosage range was between 2250 mg/L, 2500 mg/L, 2750 mg/L, 3000 mg/L, 3250 mg/L, 3500 mg/L, 3750 mg/L, 4000 mg/L, 4250 mg/L, and 4500 mg/L, respectively. The optimum coagulant dosage was determined by the highest removal percentages on chemical oxygen demand (COD), suspended solids, ammonia, iron (Fe), and Chromium (Cr). Next, by using the optimum and constant coagulant dosage, the pH range was varied from 3 to 10 to decide the ideal conditions for leachate sample. The pH of leachate sample then was altered to pH 7 by using 1N hydrochloric acid.
Based on the observation on the Figure 3 (a) the removal percentages on suspended solids and Fe were continuously increasing from dosage 2250 to 4500 mg/L. However for Cr, the removal percentages decreased at dosage of 4500 mg/L. While the removal percentages for other parameters were bit fluctuating at the initial and final dosage ranges. From the outcome results, the removal percentages for COD and ammonia were the lowest compared to others. The removals were lower might due to its chemical characteristics that did not counterpart much with coagulation-flocculation process. Nonetheless, the removal percentages for suspended solids, Fe, and Cr were quite satisfying. Through this first step of optimization process, it was noticed that the optimum coagulation-flocculation occurred at 3750 mg/L with 96%, 44%, 52%, 96%, and 66% for suspended solids, ammonia, COD, Fe, and Cr respectively. In order to confirm the optimum coagulation-flocculation process of single PAC dosage at 3750 mg/L, the second jar test was carried out with the same conditions.

Based on the second jar test in optimizing the single PAC dosage (Figure 3 (b)), the results showed better removal percentages for parameter COD and Cr compared to the previous one. Overall, through Figure 3 (b), the graph showed the removal percentages were increasing gradually without much fluctuating like in experiment 1, with all the applied PAC dosages. Nevertheless, after the application of 3750 mg/L PAC dosage, the removal percentages of most parameters were decreasing, such as SS, AN, and COD. While the after treatment affect decreased at 4500 mg/L for the removal heavy metal Cr. The removal of Fe after treatment indicated continuous removal even though at dosage 4500 mg/L, which showed the same trend in the first jar test (Figure 3 (a)). As a whole, the second carried out jar test showed the optimum single PAC dosage occurred at 3750 mg/L as well, with removal percentages of 94, 17, 70.32, 96.94, and 83.12% for SS, AN, COD, Fe, and Cr respectively. Since the optimum coagulant dosage of single PAC was achieved, the conventional optimization was continued with varied pH of leachate samples. The results for third jar test were presented as in Figure 3 (c).
The pH of leachate samples were altered using 1N of hydrochloric acid and sodium hydroxide, from pH 3 to pH 10. Based on Figure 3 (c), the removal percentages of most...
parameters showed increasing trends from pH 3 to 6, but decrease at pH 7 to 10. However, for certain parameters, the significant removal percentages were recorded at different pH values. Individually, the highest removal for each parameter was 98% for suspended solids at pH 6, 19% for ammonia at pH 5, 54% for COD at pH 6, 97% for Fe at pH 7, and 79% for Cr at pH 7. In the third-repeated jar test, the removal percentages of 3750 mg/L dosage of PAC at both pH 6 and 7 had almost approximate outcomes. At pH 6, the gained results was 98%, 18% 54%, 95%, and 76%, meanwhile its 95%, 18%, 53%, 97%, and 79% at pH 7, consecutively resembling for suspended solids, ammonia, COD, Fe, and Cr parameters. Therefore it showed that based on the removal percentages at both pH values, pH 6 and 7 had the same dominance, with the difference was not more than 5% each. Since the average pH value of raw leachate in this study was 8.76, pH 7 was chosen as the optimum pH condition in this study of single PAC coagulant. It was considered at pH 7 due to the less used volume of hydrochloric acid in altering the pH of leachate sample. Nevertheless, it was also considered due to the required ‘real-life’ application on site, which neutral pH is prioritized [19]. The result in this study was compared with the application of PAC coagulant from previous literature reviews. Table 5 shows the utilization comparison of PAC coagulant on leachate sample with previous studies.

Table 5. Utilization comparison of polyaluminium chloride (PAC) coagulant on leachate sample with previous studies.

| Optimum dosage (mg/L) | Optimum pH | Removal parameter (%) | Reference |
|-----------------------|------------|-----------------------|-----------|
| 3750                  | pH 7       | Suspended solids (95%) | From this study |
|                       |            | Ammonia (18%)         |           |
|                       |            | COD (53%)             |           |
|                       |            | Fe (97%)              |           |
|                       |            | Cr (79%)              |           |
| 2500                  | pH 7       | Suspended solids (92%) | [19]      |
|                       |            | Turbidity (77%)       |           |
|                       |            | Colour (94%)          |           |
|                       |            | COD (37%)             |           |
|                       |            | Ammonia (32%)         |           |
| 2000                  | pH 7       | COD (49%)             | [20]      |
|                       |            | Ammonia (29%)         |           |
| 7200                  | pH 6       | COD (55%)             | [21]      |
|                       |            | Colour (80%)          |           |
|                       |            | Suspended solids (95%)|           |
| 1900                  | pH 7       | COD (57%)             | [12]      |
|                       |            | Colour (97%)          |           |
|                       |            | Suspended solids (97%)|           |
|                       |            | Turbidity (99%)       |           |

In the coagulation-flocculation process using single PAC coagulant, optimization at pH 7 seems general, according to [12], [19-21]. In this study, it was found that 3750 mg/L dosage of PAC works well with pH 7, which proven through the first and third jar tests. At certain parameter, the removals were found to be higher at the first jar test, such as suspended solid and ammonia at 96% rather than 95%, and 44% rather than 18%, consecutively. Meanwhile, for parameter Cr, the removal at third jar test was higher, which
at 79% rather than 66%. Based on the observations, the obvious difference values could be seen in parameter of ammonia and chromium. Generally, through comparison with the previous studies, the removal of ammonia was usually low, especially when coagulation-flocculation process was used [20]. The ammonia concentration in the leachate sample used in the first jar test might be lesser than the leachate sample used in the third jar test, which cause higher removal than the last one. As eloquently stated by [6], the ammonia nitrogen content loads in the leachates are toxicants and may become the eradicate factor to physical-chemical treatment method. Thus, the pre-treatment to get rid ammonia pollutants first is such a crucial step, for instance, by integrating biological practices [22]. Moreover, the leachate in SRLS nowadays has become more high-strength, due to the dosage comparison from previous study [19], that used the same source of leachate sample taken from SRLS. Logically, if higher pollutants are present, higher dosage is required as well.

5 Conclusion

Hence, in a conclusion, the optimization of single PAC dosage occurs at 3750 mg/L dosage and pH 7 with the results of removal percentages at satisfying outcomes, at 95%, 18%, 53%, 97%, and 79% for suspended solids, ammonia, COD, Fe, and Cr respectively. it is recommended to reduce the dosage of PAC coagulant or any other chemical coagulants that are frequently used in the wastewater treatment for future research and studies. This is due to, despite the superiority of of these chemical coagulants, they also have negative sides by referring to the past literature [23]–[28][23]–[27], [29], such as hazards on living organisms and human health [26], [27], [30], [31]. Sustainability treatments are preferable when talking about forthcoming-next green remediation. Therefore, by studying on new sustainable coagulants, these drawbacks at least can be tackled if not entirely, just to lessen the flaw is already a pleased advancement.

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