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Influence of Dosage, pH, and Settling Time on the Performance of Pre-Hydrolysed Polyaluminum Chloride (PAC) in Treating Ageing Matured Leachate

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Abstract

The treatment of mature leachate remains lacking due to its unique composition and characteristics, which change as the landfill ages over time. This study investigates the efficiency of the coagulation-flocculation process using Polyaluminium Chloride (PAC) to remove colour and ammoniacal nitrogen (AN) from raw matured leachate. Also, it compares the optimum removal efficiency with previous studies conducted on the same landfill leachate using PAC but in a different year. Several key parameters were evaluated, including PAC dosage (500–3000 mg/L), pH (4–9), and settling time (10–60 minutes). At the optimised designed condition of PAC dosage of 2500 mg/L, pH 6, and a 30-minute settling time, colour and AN were removed remarkably at 82% and 18% AN, respectively. These findings demonstrate significant changes in colour removal compared to previous studies, which only achieved 50–70%. While AN removal was slightly lower at 18%, the superior colour removal performance highlights the potential and relevance of this approach for treating the raw matured leachate as landfills continue to age. Thus, this study emphasised the relevance of this method for continued use in mature leachate treatment.

1. Introduction

Landfilling is one of the common methods of MSW disposal in Malaysia. However, an unavoidable result of landfill operations is the production of leachate [1]. Leachate is a highly concentrated liquid pollutant created by the percolation of rainwater and the decomposition of organic waste within the landfill [2]. Treating landfill leachate is particularly challenging as the leachate matures over time, increasing the composition of contaminants such as heavy metals, chemical oxygen demand (COD), colour, odour and AN [1]. Matured leachate is characterised by its low biochemical oxygen demand (BOD_5) and high chemical oxygen demand (COD), resulting in a BOD_5/COD ratio < 0.1, which differs from the young leachate [3]. This low BOD_5/COD ratio indicates low biodegradability. Such mature leachate requires advanced physical-chemical treatments like the coagulation-flocculation process.

Coagulation-flocculation eliminates non-biodegradable pollutants in mature leachate as it effectively removes refractory compounds, depending on the coagulant used [4]. Coagulation-flocculation uses coagulants to destabilise and aggregate dispersed particles into bigger flocs, making it easier to remove the contaminants [5]. Assisted flocculation and coagulation through optimal coagulant dosage, pH and settling time is crucial for effective floc formation and stability, enhancing the removal of contaminants from landfill leachate [6]. The performance and effectiveness of the coagulation-flocculation process in leachate are influenced by several factors, such as the flocculant dose, coagulant dose, pH, stirring time and settling time [7], [8]. This study focuses on optimising the coagulant dosage, pH adjustment, and settling time, as these factors significantly influence the coagulation-flocculation outcomes to evaluate the current relevance of this method in treating the keeps ageing mature leachate. Certain parameters are carefully controlled throughout the process to ensure consistent and effective outcomes. The control and fixed conditions included are the temperature, speed of rapid mixing, duration of rapid mixing, speed of slow mixing, and duration of slow mixing. The combination of these parameters considerably impacts the efficiency of coagulants in removing pollutants from leachate, resulting in optimal treatment outcomes [9].

Alum, ferric chloride, and polyaluminum chloride (PAC) are commonly used coagulants in this process, with each working by destabilising particles, causing them to aggregate and settle out of the solution during the flocculation process [4], [10]. Alum and ferric chloride are effective, although they frequently require greater dosages and produce large amounts of sludge, complicating the disposal process [11]. PAC, on the other hand, provides advantages in leachate treatment due to its increased efficiency at lower dosages and capacity to act across a broader pH range [12]. Moreover, the positively charged Al(OH) precipitate in the coagulant significantly enhances the humic substances and increases the flocculation kinetics [11]. Thus, to ensure the long-term effectiveness of the method, selecting the best coagulant is crucial, guided by the study of the leachate composition and optimisation of the coagulants.

The study of leachate composition and characteristics helps understand the specific contaminants present, such as ammonia, heavy metals, and organic matter, allowing for optimising coagulant dosage, pH, and settling duration to achieve maximum removal efficiency. This research chose the sample from the Simpang Renggam Landfill site (SLRS) at Johor, where several studies have been conducted in the previous year using the same PAC coagulant on this landfill leachate. However, despite using the same coagulant and sample, optimisation studies conducted over different years have yielded varying results. For example, Azizan et al. [13] managed to achieve 32% ammonia removal for the SLRS leachate sample by using PAC, while Mayangsari et al. [14] found that the same method and coagulant achieved 41% removal of ammonia. This inconsistency shows that the leachate composition has changed over time, potentially due to the ageing of landfills, highlighting the urge to reassess whether the same treatment method and coagulant remain effective under the current leachate conditions. While previous studies have explored the effectiveness of PAC as a coagulant, the effect of the ageing landfill has not been thoroughly addressed. Thus, this study was conducted to bridge the gap by comprehensively evaluating the performance of the PAC coagulant in the coagulation-flocculation method specifically applied to treating the matured leachate on its current composition. This research focuses on optimising critical operational parameters, such as the PAC dosage, pH, and settling time, to identify the best conditions for maximising pollutant removal and improving the leachate quality.

2. Materials and Methodology

2.1 Leachate Sampling

Samples were obtained from the Simpang Renggam Landfill Site (SLRS), located at the latitude of 10 53'41.64 "22'34.68 North and 1030" East in Kluang district, Johor. The site is located two kilometres from Simpang Renggam town and has been operated for over ten years. SRLS used to receive around 400 to 500 tonnes of solid waste daily from areas including Simpang Renggam, Batu Pahat, Kluang, and other surrounding regions, from permanently closed in 2019 [11]. Fig. 1 below is the map location of the SLRS, while Fig. 2 is the location of the sampling. The sampling and storage were conducted using the APHA standard method [15]. The manually collected samples by grab sampling were stored immediately in the cold room (4°C) at the Micropollutant Research Centre, Faculty of Civil Engineering and Built Environment (FKAAB), Universiti Tun Hussien Onn Malaysia. The collected sample was characterised on the same day by evaluating eight parameters: pH, suspended solids (SS), BOD, COD, AN, turbidity, and colour. The pH and turbidity were tested on-site, while the rest of the parameters were tested in the laboratory.

Fig. 1 *Map view of the SLRS*

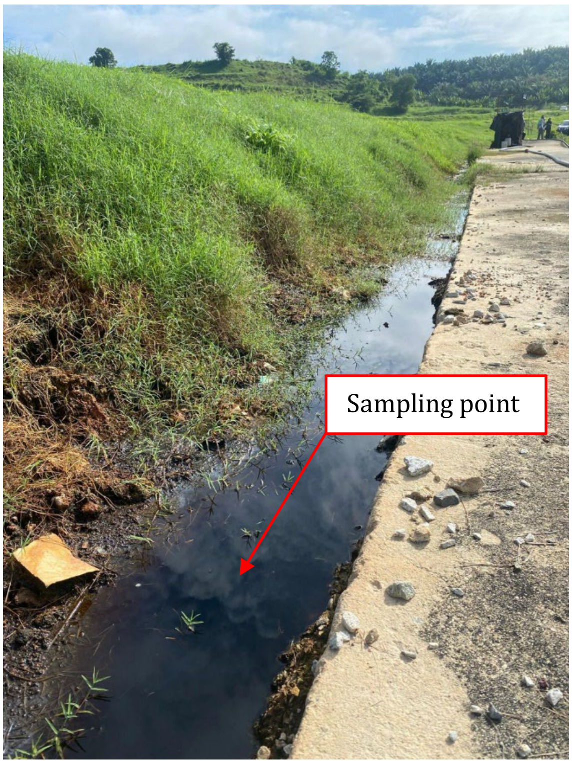

Fig. 2 *Sampling location*

2.2 Preparation of PAC coagulant

The QreC brand PAC, with a chemical composition of $Al_2Cl(OH)_5$, was purchased from a local supplier in a 500-gram bottle. Following Azizan [13], to prepare 10% of the stock solution, 10g of PAC was weighed and diluted in 100 ml of distilled water. The stock solution was prepared on the same day the optimisation was conducted. The range of dosages was calculated by using Eq. (1):

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

where M_1 = Stock solution concentration, V_2 = Volume of stock solution, M_2 = Dosage of coagulant, and V_2 = Volume of leachate sample

2.3 Jar Test

Table 1 lists the operational design parameters used in this test [16]. The experiment was conducted to find the optimal PAC dosage, pH level, and settling time by evaluating the removal percentage of colour and AN.

Table 1 *Experimental design parameter*

Parameters	Experimental design
Temperature (°C)	Room temperature
Speed of rapid mixing (rpm)	200
Duration of rapid mixing (min)	4
Speed of slow mixing (rpm	30
Duration of slow mixing (min)	15
Coagulant dosage (mg/L)	500-3000
pH	4-9
Settling time (min)	10-60

The removal percentage was calculated using Eq. (2):

$$\text{Removal}\ (\%) = \frac{C_0 - C_f}{C_0} \times 100\% \tag{2}$$

where C_0 = Initial concentration of the sample (mg/L), and C_f = Final concentration of the sample (mg/L). The results were further analysed using conventional Microsoft Excel, and the graphs were generated using Origin 2024. The removal of AN and colour was measured using the standard methods for water and wastewater (APHA).

2.4 Coagulation-Flocculation Optimisation

The optimum conditions for coagulation-flocculation were determined using the conventional "one factor at a time" method, focusing on selected variables [17]. This study focused on the PAC dosage, pH, and settling time by following the method by Azizan [16]. The optimum dosage was obtained by pouring a 500 mL volume of leachate into a 1000 mL beaker, and PAC coagulant was added in the range of 500 mg/L to 3000 mg/L while the pH and settling time were fixed at pH 5 and 30 minutes. The fixed operating condition is referred to in Table 1. The optimum dosage was determined by evaluating the colour and AN removal. Subsequently, with the optimum coagulant dosage kept constant, the pH range was varied from 4 to 10 to identify the ideal pH condition for the leachate sample. pH was adjusted by using 0.1 M sodium hydroxide and 0.1 M hydrochloric acid. Lastly, the optimum settling time was determined by varying the settling duration from 10 to 30 minutes, with the optimum dosage and pH maintained throughout the experiment.

3. Results and Discussion

3.1 Leachate Characterisation

Table 2 demonstrates the sample characteristics. Samples were in alkali pH (8-9), BOD_5/COD 0.064 < 0.1, AN concentration of 1626 mg/L >400 mg/L and COD of 2187 mg/L <4000, indicating that the leachate sample was a mature leachate. Compared to previous studies that used the same method and sample, it was proven that the leachate composition keeps changing over time, with the current concentrations of pollutants such as turbidity, SS, colour and AN being remarkably increased [13], [18]. As mentioned in Table 2, the current pH of the SLRS matured leachate was also found to become more alkaline as the year went by, compared to previous studies [19]. These results prove that, when evaluating the same method, the coagulation-flocculation process using PAC on the same samples was needed to examine its relevance to the current mature leachate composition condition.

3.2 Dosage Optimisation

The coagulation process is influenced by several critical factors, with coagulant dosage being one of the significant factors. Based on the result in Fig. 3, when the PAC dosage was altered from 500 mg/L to 2500 mg/L, the colour and AN were removed gradually, slightly fluctuating before reaching their highest removal at 86% and 19%, respectively, when PAC dosage was used at 2500 mg/L. Meanwhile, after exceeding 2500 mg/L dosage, the removal for both started to decline to 67% and 10%, indicating the overdosing of coagulant, which is caused by the repelling of the positively charged particles of the excessive PAC, causing flocs deflocculation [20]. Moreover, a study by Mustereț et al. [21] proved that the coagulant efficiency decreases when it exceeds the optimal value due to the colloid restabilisation. This suggests that exceeding the dosage of 2500 mg/L does not significantly improve the removal performance of PAC in terms of colour and AN on the current leachate composition. Thus, 2500mg/L was chosen as the optimum PAC dosage with 86% removal of colour and 19% removal of AN. Compared to a study by Azizan et al. [13], the higher removal efficiency of colour (94%) and AN (32%) were achieved under similar 2500 mg/L PAC dosage conditions, while Mohd-Salleh et al. [18] recorded 98% colour removal and 28% AN removal at a higher PAC dosage of 3750 mg/L. Although this study achieved a lower removal with the exact dosage used, the initial concentrations were significantly higher, with 9333 ADMI for colour and 1626 mg/L for AN, compared to Azizan et al. [13], who reported 4700–6100 ADMI for colour and 692–1272 mg/L for AN.

Moreover, this suggests that although the removal percentages in this study were slightly lower, the treatment was applied to more concentrated leachate. The results demonstrate the effectiveness of PAC even under more challenging conditions. These differences between the study findings can be attributed to the changes in the leachate composition over time. Compared to the previous study, it can be concluded that the current leachate samples used in this study contained a higher concentration of dissolved organic matter and recalcitrant compounds, reducing PAC's overall effectiveness. This aligns with the study by Zheng et al. [22], who highlights the effect of complex organic matter on coagulation performance.

Table 2 *Characteristic of SLRS leachate sample from 2017-2024*

Parameter	(January – April 2017)	(January – December 2018)	(October – December 2021)	(March-September 2024)	Environmental Quality Act (EQA 1974) Standard
pH	7.6 – 8.3	8.11 - 7.76	8.1	8.68	6.0-9.0
Turbidity (NTU)	84-244	63.5 - 160	23.04	347	-
SS (mg/L)	78-268	69 - 291	82.92	634	50
Colour (ADMI)	4700-6100	1902 - 7101	2069	9333	100
AN (mg/L)	692- 1272	131.21 - 963.24	1218	1626	5
COD (mg/L)	1836-2150	996 - 2954	706.30	2187	400
BOD_5 (mg/L)	-	54.38 - 253.88	-	138.90	20
BOD_5/COD	0.085	0.02 - 0.211	-	0.064	-
No. of samples	12	24	9	8	-
Reference	[13]	[18]	[19]		-

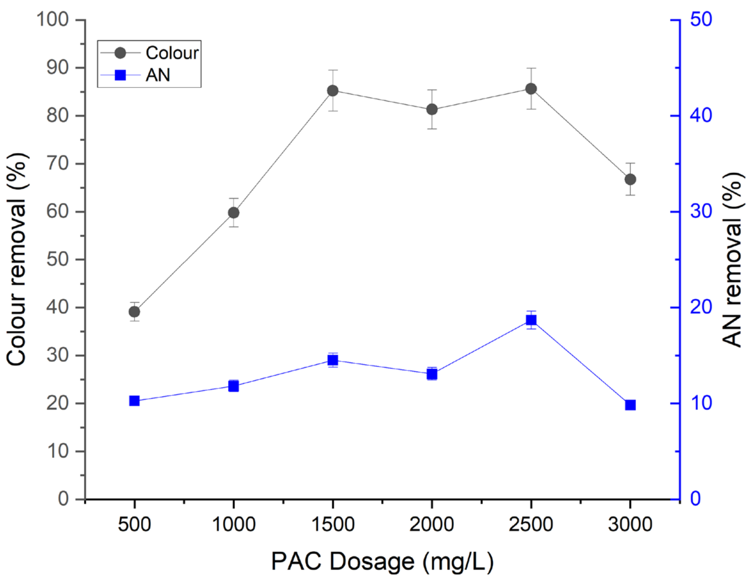

Fig. 3 *Optimisation of PAC dosage at pH 7 and settling time 30 minutes (rapid mixing 200 rpm for 4 minutes and slow mixing 30 rpm for 15 minutes)*

3.3 pH Optimisation

The pH is one of the vital factors that influence the effectiveness of the coagulation-flocculation process, as it relates to the zeta potential and surface charge of colloidal particles, as well as the hydrolytic response of the coagulant [4]. In this optimisation process, the pH was varied from 4 to 9, with the optimum pre-determined dosage of PAC (2500 mg/L). Fig. 4 demonstrates the result of the pH optimisation. Based on the result, an increase in pH starting from pH 4 showed a significant climb in the removal of colour and AN, with the highest removal of 67%-80% and 19%-27%, respectively, occurring at pH 6. However, the removal of both parameters started to drop gradually when exceeding pH 6, with less than 75% for colour and less than 24% for AN. Thus, pH 6 was the optimum pH, which could produce 80% colour and 27% of AN removal. This result aligns with the study by Chen et al. [4] that mentioned too high pH could be ineffective due to the strong alkaline condition, causing a decrease in the positive charge carried by the coagulant, thus producing a weaker attraction between the anionic organic compounds and the positive charge of coagulant However, previous studies reported higher removal efficiencies at similar and slightly higher pH levels.

Azizan et al. [13] found that pH 7 led to 94% colour and 32% AN removal, while Mohd-Salleh et al. [18] and Mayangsari et al. [14] reported comparable results of 98% colour with 28% AN removal, and 92% colour with 41% AN removal, both at pH 6. The same optimum pH obtained in this research and previous studies resulted in different removal percentages due to the variations in the initial concentration of the colour (9333 ADMI) and AN (1626 mg/L), influenced by the ageing of the landfill. The slight differences in the optimum pH value and contaminant removal percentage compared to previous studies can be attributed to the differences in the leachate composition over time [3]. Nevertheless, an alkaline to neutral pH range is proven to remain the optimum pH condition for mature leachate treatment [11].

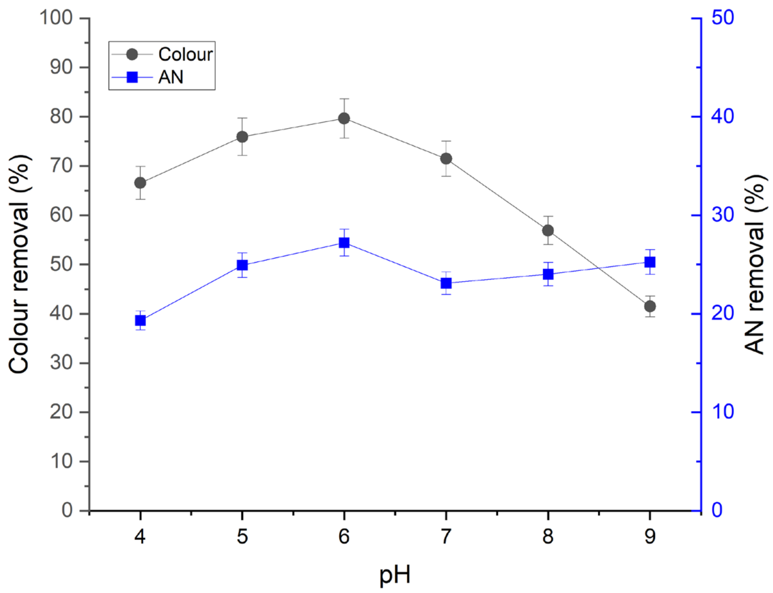

Fig. 4 *Optimisation of PAC dosage at pH 7 and settling time 30 minutes (rapid mixing 200 rpm for 4 minutes and slow mixing 30 rpm for 15 minutes)*

3.4 Settling Time Optimisation

The settling time was optimised at different times of 10 to 60 minutes, emphasising the necessity of achieving equilibrium for optimal results. This process used the pre-determined optimum dosage and pH at 2500 mg/L of PAC and pH 6. Fig. 5 reveals that the coagulation process's efficiency diminished after a certain settling period, indicating the existence of an optimal settling duration. Based on Fig. 5, when the settling time increased from 10 minutes to 30 minutes, removal of colour occurred from 53% to 82% and 12% to 18% for AN. However, when the settling was conducted further, the result showed a gradual drop in the removal of colour and AN, decreasing to 73% and 13%. The result showed that the optimum settling time for this treatment was 30 minutes, with 82% removal of colour and 18% removal of AN, respectively. This showed that less than 30 minutes is insufficient for all the substances to settle completely. In contrast, a more extended time above the optimum time leads to the resuspension of flocs, causing the contaminants to re-enter the samples after treatment [23]. These results aligned with the previous study by Azizan et al. [13], Salleh [17] and Mayangsari et al. [14], which concluded that 30 minutes is the optimum settling time for the matured leachate treatment. This result also confirmed that the differences in the initial leachate composition may not affect the settling time after treatment.

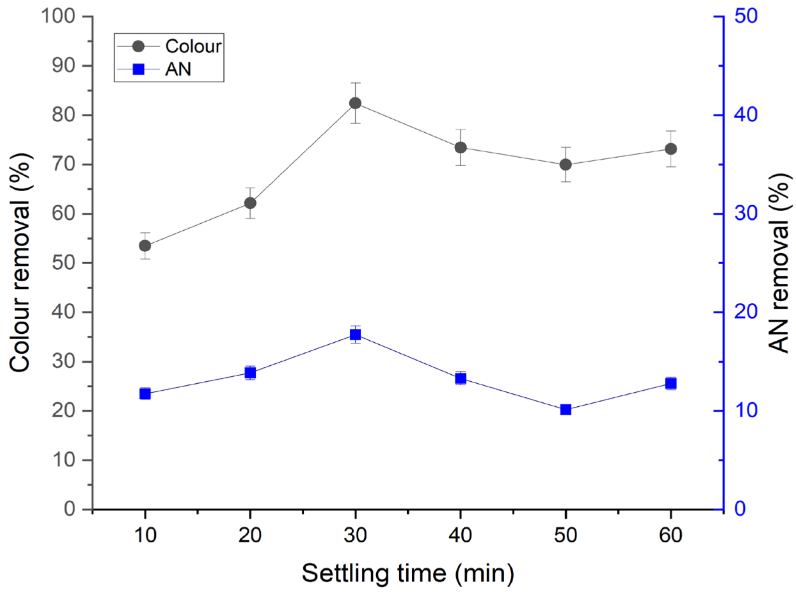

Fig. 5 *Optimisation of PAC dosage at pH 7 and settling time 30 minutes (rapid mixing 200 rpm for 4 minutes and slow mixing 30 rpm for 15 minutes)*

4. Conclusion

In summary, the result obtained from the optimisation study confirmed the removal ability of PAC in the coagulation-flocculation process, and the PAC dosage, pH, and settling time have significantly influenced the removal efficiency. This study managed to remove 82% and 18% for colour and AN, demonstrating the potential of PAC for treating leachate under optimum conditions. However, while this method was proven to remove the colour effectively, it showed limited efficiency in removing AN. This limitation may be due to the complicated nature of AN in leachate, which necessitates additional treatment to obtain better removal rates. Furthermore, differences in the removal performance compared to previous studies on the same landfill leachate indicate that changes in the leachate composition over time influence treatment outcomes. Variations in the initial contaminant concentration and organic content may contribute to the variances, emphasising the necessity for continuous reassessment of the treatment effectiveness. Overall, the findings emphasise the significance of optimising treatment conditions for effective leachate management, as well as the applicability of PAC for leachate treatment.

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Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of the paper.

Author Contribution

The authors confirm their contribution to the paper as follows: ***Study conception and design:*** *Nur Ain Natasha Abdul Rasid, Nur Shaylinda Mohd Zin, Laila Wahida Mohamad Zailani, Wan Afnizan Bin Wan Mohamed @ Wan Abd Ghani;* ***Data collection:*** *Nur Ain Natasha Abdul Rasid, Laila Wahida Mohamad Zailani, Mohd Fadzly Rashim;* ***Analysis and interpretation of results:*** *Nur Ain Natasha Abdul Rasid, Nur Shaylinda Mohd Zin, Laila Wahida Mohamad Zailani, Wan Afnizan Bin Wan Mohamed @ Wan Abd Ghani;* ***Draft manuscript preparation:*** *Nur Ain Natasha Abdul Rasid, Nur Shaylinda Mohd Zin, Laila Wahida Mohamad Zailani, Wan Afnizan Bin Wan Mohamed @ Wan Abd Ghani, Siti Nor Aishah Mohd Salleh. All authors reviewed the results and approved the final version of the manuscript.*

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