

The Effects of Mixing Speed and Reaction Time on the Removal of Colour and Turbidity from Alor Pongsu Landfill Using Tin Tetrachloride



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Abstract The release of landfill leachate to the environment without appropriate treatment creates problems to the surroundings as it might affect the groundwater and surface water sources. The use of divalent and trivalent coagulants, for example, alum and ferric chloride are common in the coagulation and flocculation process of leachate. Still, the application of tetravalent chemicals is relatively scarce. In this study, experiments were carried out to assess the use of tin tetrachloride (SnCl_4) as a potential coagulant in removing the colour and turbidity from the leachate taken from Alor Pongsu landfill site, Kerian, Perak. The effects of mixing speed and reaction time for various coagulant dosages and pH ranges were evaluated using a standard jar test. It was found that 99.5% of turbidity and 98.3% of colour were removed at 1 g/L dosage and an agitation speed of 220 rpm. Also, the mixing time of 3 min was chosen as the best agitation time as removal of 99.3% of turbidity and 98.2% of colour were achieved for the same dosage. The study inferred that the agitation speed and time are two important factors among the different mixing conditions that need to be determined in optimizing the coagulation/flocculation process.

Keywords Tin tetrachloride (SnCl_4) · Leachate · Coagulation · Flocculation · Mixing speed · Time

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1 Introduction

The most regular practices of municipal solid waste (MSW) disposal is landfilling or open dumping [60]. However, landfilling method leads to the production of landfill leachate. Landfill leachate is a liquid produced by the penetration of the precipitate across the landfill or the cap of the completed site. Normally, it comprises of a huge amount of harmful and polluting components as well as a multi-mixture of organic and inorganic constituents, which can be categorised as inorganic macro compounds, heavy metals, dissolved organic matter and xenobiotic organic complexes [3, 28, 29, 52]. Due to the existence of these constituents, the leachate is typically recognized as recalcitrant wastewater. Improper collection, management, and discharge of raw landfill leachate will pollute the groundwater, the surface water [14], and this will eventually cause harm to the environment. Therefore, appropriate leachate treatment and supervision are necessary to inhibit or to reduce the hazardous impact on the environment.

An appropriate leachate treatment and supervision are necessary to inhibit or to reduce the impact on the environment. There are about 200 harmful substances and other toxic chemicals found in landfill leachate. Compounds such as phenols, pesticides, halogenated compounds, aromatics compound, heavy metals and ammonium may pose a damaging and hazardous impact on the aquatic life and ecosystem which lead to many serious health problems, for instance, carcinogenic effects, severe toxicity as well as genotoxicity. Also, heavy metals content in leachate might enter the food chain, as heavy metals are not biodegradable and can infiltrate into the water and groundwater sources. Also, the ammonia produced from the leachate will gradually nitrify to nitrite and nitrate, which will lead to the eutrophication problem. Eutrophication happens when a high concentration of ammonia and phosphate mix with the surface water. This phenomenon gives adverse impacts to the aquatic life as it limits the penetration of light into the lake or river. Landfill leachate also causes aesthetic problems as landfills produce awful odour, noise, dust, insects, rodents, snakes and scavenger birds [5, 17, 36, 57].

There are diverse treatment practices that can be applied for leachate treatment, for example, physical treatment, chemical treatment, physicochemical treatment or biological treatment [46, 47]. The most practical physicochemical process employed for leachate treatment is coagulation and flocculation. Coagulation is described as the destabilization of colloids in a solution by neutralizing the repulsive electrical charges among the particles which allow them to get closer and stick together. Meanwhile, the flocculation process is a process wherein the destabilized colloids are gathered and agglomerated into bigger flocs [15]. In the process, the common coagulants such as the positively charged inorganic metals salts are tested first to the wastewater then, continued by the addition of long chains of non-ionic or negatively charged polymers, usually called as flocculants (Zahrim et al. 2019). Trivalent coagulants such as ferric chloride, polyaluminium chloride (PAC), alum and aluminium sulphate are some of the frequently used coagulants [39]. The positive electric charges provided by the cationic coagulants help in reducing the

negative charge of the colloids. This process initiates the collision between the colloids to form bigger flocs and allows settlement. To properly disperse the coagulant throughout the liquid, rapid mixing is essential [15]. Coagulation and flocculation are one of the broadly used methods for leachate treatment. Generally, this process is favoured over others due to its simple operation as well as the effectiveness of removal of the suspended particles. The coagulation efficiency is controlled by many aspects, for instance, pH, dosage and coagulant/flocculant type, agitation speed and duration, temperature as well as settling time [13]. Of all the factors, the mixing time and speed play an essential part in the primary stages of the treatment. This is because high mixing strength and longer stirring time will disrupt the formed aggregated flocs. Therefore, the determination and optimization of these factors are crucial as they can influence the extent of removal of pollutants from leachate [46, 59].

Physico-chemical treatment focuses on the separation of colloidal particles in the wastewater. Eventually, the physical condition of colloid will change by the addition of chemicals, which will lead them to be in an unstable state and finally form a particle with settling properties [2]. Initially, the physicochemical treatment aim was to improve the effectiveness of a biological treatment system. Therefore, it was implemented as a biological pre-treatment. It can either be a single stage treatment or a combination with other treatment processes during the pre-treatment. The elimination of pollutant in water or wastewater through physicochemical treatment was achieved by modifying the chemical or physical structure of the elements [31, 42]. Application of this method in wastewater treatment can contribute in reducing the COD, BOD, metals, colour, ammonia, turbidity, and suspended solids content in the wastewater [12, 33]. The selection on which treatment method is suitable for leachate treatment generally depends on the characteristics of the leachate. Different leachate requires different treatment method as their chemical compositions. Usually, young or freshly produced leachate can effectively be treated using biological treatment processes. The old leachate with low biodegradability index can be treated using physico-chemical treatment. Since the leachate used in this research is considered as old leachate, therefore, the recommended treatment method is by physico-chemical treatment. The typical physico-chemical treatment includes adsorption, chemical precipitation, air-stripping, membrane filtration, chemical precipitation and coagulation/flocculation process [1, 2, 35, 53]. Physico-chemical treatments are usually chosen as one of the leachate treatment methods because of the effectiveness, quick, compact, economically feasible and excellent for colour removal [37].

The coagulation/flocculation performance is also influenced by the type of metal coagulant salts used. Generally, coagulants can be divided into chemical based coagulants and natural coagulants. The most applied chemical-based coagulants in the coagulation and flocculation process are an alum, ferric chloride and polyaluminium chloride (PAC) [26]. The introduction of higher charged cations into the water helps in destabilizing the colloidal particles, and large aggregates will be formed. Theoretically, according to Hardy Schulze law, the coagulation of particles depends on the valency of the ion with a charge contrary to that of the colloidal

particles. In simple words, the higher the valency of the opposite ion of electrolyte added into the solution, the quicker the coagulation process [38]. The role of coagulant is influenced by several factors such as the amount and reactivity of complexing ligands, hydrolysis products, and the degree of mass transfer among those constituents. Hence, the type of coagulant chosen has a major impact on coagulation or flocculation efficacy [65]. Many researchers had studied the efficiencies of trivalent coagulants in removing contaminants from landfill leachate. Fe (III) and Al (III) coagulants are widely used because of the economical factor and sufficient availability [25, 51]. Aziz et al. [8] studied the effectiveness of three types of coagulants namely, ferric chloride (FeCl_3), ferrous sulphate (FeSO_4) and aluminium sulphate (alum), on the removal of colour. Alum removed 60.4% of colour at pH 4 with coagulant dosage of 600 mg/L. Meanwhile, 2500 mg/L of FeCl_3 dosage removed 96.5% of colour at pH 6, and only 63% of colour was removed using 600 mg/L of FeSO_4 at pH 12. Thaldiri et al. [55] investigated the removal of colour using Polyaluminium chloride (PAC) and found that 96.4% of colour was removed at pH 7 using 1000 mg/L of PAC. Studies by Zainol et al. [63] using PAC in landfill leachate treatment, found that 7200 mg/L of PAC was required to remove 70–73% of the colour from the leachate. Yusoff et al. [61] required 8000 mg/L of PAC at pH 7 to remove 90.7% of turbidity and 99.4% of colour.

Despite their wide applications, the trivalent metal coagulant salts also presented a few drawbacks. For example, the use of Al in purified water increase the turbidity level which leads to low turbidity removal efficiency, decrease the disinfection level and a loss in hydraulic capacity [51]. Also, the use of these metal coagulants hindered the effectiveness of the wastewater treatment as they produced huge amounts of sludge [40]. To improve treatment efficiency, another metal-based coagulant with a higher valency (tetravalent coagulant) is considered. The examples of currently used tetravalent metal-base coagulants for water and wastewater treatment are titanium and zirconium salts [25]. These two tetravalent coagulants were reported not only raise the performance of coagulation/flocculation process but also reduced the amount of sludge formed with an increase in the sludge settling time [40]. Jarvis et al. [27] reported that the floc formed from coagulation using Zirconium (Zr) was larger (930 mm) than flocs produced by Ferric (Fe) salt (710 mm). Zr was also able to decrease 46–150% of dissolved organic carbon (DOC) compared to the Fe salt [51, 65] used zirconium-glycine complex to remove 93.8% of turbidity compared to 87.9 and 60% removal of turbidity using PAC and $\text{Al}_2(\text{SO}_4)_3$, respectively. Aziz et al. [11] employed TiCl_4 on landfill leachate treatment and reported that, at pH 6, 600 mg/L TiCl_4 was able to reduce 81.4% of colour and 86.7% of suspended solids. Pushpalatha and Lokeshappa [43] also reported on the coagulation performance using TiCl_4 to treat stabilized landfill leachate and concluded that TiCl_4 showed a better removal of turbidity, colour and COD compared to FeCl_3 and alum. TiCl_4 removed 78.3% of COD, 72.6% of turbidity and 75.4%, of colour at optimum dosage of 0.6 g/L. Meanwhile, 0.4 g/L FeCl_3 removed 42.3, 52.1, 59.8% of COD, turbidity and colour, respectively. On the other hand, alum reduced 26.2% of COD, 35.3% of turbidity, and 45.3% of colour at optimum dosage of 0.8 g/L.

The elements are expected to form trivalent or tetravalent coagulant by losing their valence electrons. Elements which have 3 or 4 electrons in their outermost shell are expected to form trivalent or tetravalent cations by losing all their valence electrons. For example, the Sn atom, which contains 4 valency electrons can transform into Sn^{4+} or Sn^{2+} by losing all their four outermost electrons or losing only 2 valence electrons [48]. The efficiency of coagulation performance is affected by the oppositely charged ions from the electrolyte. Meanwhile, the power of an ion affecting the coagulation depends on its valency. The effect of colloidal coagulation is greater when the valency of the ion is higher. For instance, trivalent cations such as Al^{3+} is more efficient in the coagulation of negatively charged particles, compared to the bivalent cation (Ba^{2+}) or monovalent cation like Na^+ . Generally, the order of the effect of charge ions will be: trivalent cations > divalent cation > monovalent cation [32].

Thus, this research investigates the coagulation and flocculation performance of tin tetrachloride (SnCl_4) as an alternative tetravalent coagulant in treating the landfill leachate. By applying the tetravalent cations (Sn^{4+}), it is expected that the effect of coagulation will be much higher than the lower cations. The influence of operational conditions like rapid mixing and time were evaluated with regard to the colour and turbidity as target parameters. The focus of the paper is to evaluate the effects of mixing speed and reaction time on the coagulation and flocculation process of the landfill leachate. For this, the initial finding on the two most common parameters of leachate was selected, i.e., colour and turbidity. This has been widely practised by other researchers, such as [10, 39, 41, 62]. These effects on the other leachate parameters such as COD and $\text{NH}_3\text{-N}$ are part of the on-going investigations.

2 Methodology

2.1 Leachate Sampling and Characterization

Landfill leachate samples were collected from Alor Pongsu landfill site, located in Perak, Malaysia. To minimize the day to day, change of characteristics, the sample was kept at 4 °C in a cold room, and the samples were preserved according to the Standard Methods for the Examination of Water and Wastewater [7]. The leachate samples were examined for suspended solids (SS), pH, turbidity, Biochemical Oxygen Demand (BOD_5), Chemical Oxygen Demand (COD), colour and ammoniacal nitrogen. The characterization of the leachate is tabulated in Table 1.

Table 1 Raw characteristics of Alor Pongsu Landfill leachate (an average of 6 samples, taken from Nov 2018 until April 2019)

No	Parameter	Average	Discharge limit ^a
1	pH	8.18	6.0–9.0
2	BOD ₅ (mg/L)	90	20
3	COD (mg/L)	2223	400
	BOD/COD	0.1	–
4	Suspended Solids (mg/L)	435	50
5	Ammoniacal nitrogen (mg/L NH ₃ -N)	1030	5
6	Colour (Pt. Co)	13,597	–
7	Turbidity (NTU)	16.3	
8	Zeta potential	–19.8	

^aAccording to environmental quality (Control of pollution from solid waste transfer station and landfill) Regulations 2009 (PU (A) 433)

2.2 Preparation of Tin Tetrachloride (SnCl₄)

Tin tetrachloride (SnCl₄), supplied by BG Oil Chem Sdn. Bhd, Penang, Malaysia was used as a coagulant. The preparation of SnCl₄ stock solution was done by dissolving the needed volume of SnCl₄ into distilled water. To prevent degradation in molarity, the stock solution was only prepared when needed.

2.3 Jar Test

The best condition of mixing was evaluated using a standard jar test apparatus (SW6 Stuart Bibby Scientific Limited, UK) which comprises of six pedals. Leachate samples were conditioned to room temperature for around 1 h. Then, the required pH value was adjusted using 3M NaOH and 3M HCl. For the determination of best agitation speed and time, the experimental conditions of speed and time were varied from 80–260 rpm and 2–9 min, respectively. Based on similar previous research, certain guidelines were established to fix other operating parameters such as mixing speed, mixing time and settlement time at 60 rpm, 30 min, and 20 min, respectively. These operating parameters were chosen as they had been previously applied in treatment of landfill leachate by researchers such as Taoufik et al. [54], Verma and Kumar [58] and Amr et al. [6]. Also, these values were in the range of critical operating parameter obtained from the literature [16, 49, 50]. Moreover, Aziz et al. [11] had applied the same agitation, speed and settling time to treat leachate using TiCl₄. Six beakers of 500 mL leachate solutions were agitated simultaneously. After the experiments, samples were collected 2 cm below the supernatant and were tested for turbidity and colour removal using the DR 2800 Spectrophotometer.

3 Results and Discussion

3.1 Characteristics of Alor Pongsu Leachate

Table 1 shows the physicochemical properties of raw leachate obtained from Alor Pongsu landfill site. The characteristics of the sample were taken for six months from November 2018 until April 2019.

The pH value increase with the increase of landfill age. The site is considered as a mature landfill because the pH value of the leachate is greater than 7.5, which meets the criteria for more than ten years landfill [45]. The leachate colour is deep black with an average concentration of 13,596.7 Pt. Co, because of the existence of high fulvic and humic acids, and other organics in the waste. There is also a high concentration of degradable organic matters in the leachate as the COD levels are 2223 mg/L. The period phase of leachate, which explained the condition of the sample, was determined by the BOD₅/COD value ratio. Leachate is considered young, partially stabilized, and stabilize when the BOD₅/COD ratio > 0.5, 0.1–0.5 and <0.1, respectively [30]. The BOD₅/COD ratio of Alor Pongsu leachate is 0.04, which and classified as stabilized leachate with low biodegradability and considered recalcitrant. Hence, biological treatment processes alone may not be feasible.

3.2 Effect of Rapid Mixing Speed on Coagulation

The purpose of rapid mixing in the coagulation process is to give a thorough and uniform distribution of coagulant in the solution. Once the coagulant is fully mixed, the efficiency of particle destabilization is maximized and thus, will initiate the coagulation process [18]. To determine the best mixing speed for the reduction of turbidity and colour, standard jar test experiments were conducted. Figure 1 illustrated the result of agitation speed (ranging from 80 to 260 rpm with increments of 20 rpm) on the removal performance for SnCl₄ dosage of 1 g/L. The mixing speed was fixed at 3 min with an original pH of 8.1. The removal of turbidity shows an increase up to 220 rpm and decreased beyond 220 rpm. Meanwhile, for the removal of colour, there was no major variance in the removal at all mixing speeds. However, a noticeable variation of the colour was observed at 220 rpm. The highest percent change in colour and turbidity at this speed was 98.3 and 99.5%, respectively. The variation percentages at a lower mixing speed are due to the insufficient mixing of coagulant, termed as an incomplete state. For a good coagulation performance, conditions of high energy and sufficient mixing are required so that the coagulant is dispersed properly and thereby promoting particle collisions. The removal efficiencies at 200 rpm agitation speed and original pH of leachate (pH 8.1) were 98.1% for colour and 90.9% for turbidity. To assess the potency of the tetravalent coagulant, the results were compared with the removal efficiency of other trivalent coagulants, at 200 rpm. Nur and Omar [39] reported that, at 200 rpm

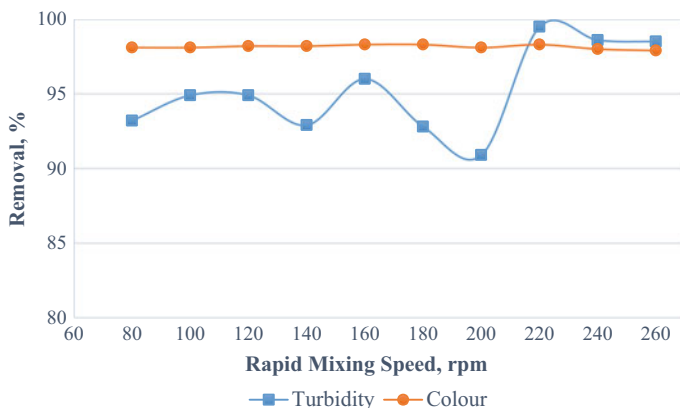


Fig. 1 Effect of rapid mixing speed on turbidity and colour removal at pH 8.1 and 1 g/L of SnCl_4

rapid mixing, only 83% of colour was removed using 2.5 g/L of alum at pH 6. Yusoff et al. [62] reported that at 900 mg/L of PAC, the reduction of colour was 93.5% while the turbidity was 90.3% removed for the same mixing speed. It is shown that, at the same mixing speed, the tetravalent salts exhibited a higher removal than the trivalent salts. However, the dosage used in Nur and Omar [39] study was higher than the study done by Yusoff et al. [62].

The optimum coagulation condition as defined by Edzwald and Haarhoff [19], originally described by Edzwald and Tobiason [20, 21] covered multiple objectives. One of the descriptions is the optimum condition where the coagulant dosage and pH maximize the removal of the particle by physico-chemical treatment. On the other hand, the United States Environmental Protection Agency [56] presented the Enhanced Coagulation term to describe the coagulation conditions designed in the treatment to improve particle removal. Considering these two terms, the poor coagulation performance in rapid mixing determination can be described as the removal of particle below the optimum value (maximum value) during the design of the coagulation condition. Using a study conducted by Ernest et al. [22] as an example, poor coagulation performance is explained as follows. Ernest et al. [22] studied the effect of mixing on the removal of turbidity from river water by varying the rapid mix at a different speed, started from 40 to 200 rpm, with 40 rpm increment each. After a series of jar test, it was found that the optimum rapid mix was at 120 rpm with 86.6% removal. At a lower mixing speed (40 rpm) and higher mixing speed (220 rpm), the removal of turbidity was 80.5% and 79.8%, respectively. Based on this situation, the mixing speed below (40 rpm) and higher (200 rpm) the optimum value (120 rpm) is not favourable in the coagulation process. This condition happens due to the insufficient or over mixing speed which leads to the inadequate or breakage of particles during the coagulation [4].

In comparisons with this study, Heng et al. [24] who used FeCl_3 could only remove 80% of suspended solids and 59% of turbidity at 300 rpm which is higher

mixing speed than this study. A similar result was also obtained by Zand and Hoveidi [64] who used aluminium sulfate (alum) and poly-aluminium chloride (PAC) to remove turbidity. At 350 rpm of mixing speed and 1 min of mixing, the turbidity removal was 82% and 93%, respectively. Aziz et al. [9] also applied alum and PAC in the removal of colour from leachate. The percentage removal of colour using alum and PAC were 83% and 85%, respectively at 300 rpm of mixing speed. These three studies required higher mixing speed but obtained a lower removal compared to the mixing condition of SnCl_4 but with higher removal (99.3% of turbidity removal and 98.2% for colour).

3.3 Effect of Rapid Mixing Time on Coagulation

Another factor that influences the efficacy of the coagulation process is the mixing time. The impact of rapid mixing time ranging from 2–9 min with an increment of 1 min, were examined in this experiment. Figure 2 displayed the percentage removal of turbidity and colour when 1 g/L of SnCl_4 was used at the original pH 8.1 and 220 rpm of rapid mixing speed. Based on the graph, the removal pattern of colour is stable as there is not much difference in the removal percentage with an increase in the time. Throughout the time, the removal of colour exhibited a saturated 98.2–98.3%. Compared to the behaviour of turbidity, the removal of colour was not much affected by the mixing time. The turbidity removal maintained within 96–99% for all the mixing times with maximum removal of 99.3% occurring at after 3 min. Raghav et al. [44] reported that at 3 min of rapid mixing, for a 90 mg/L of Alum dosage 82.5% of the turbidity was removed while Verma and Kumar [58]

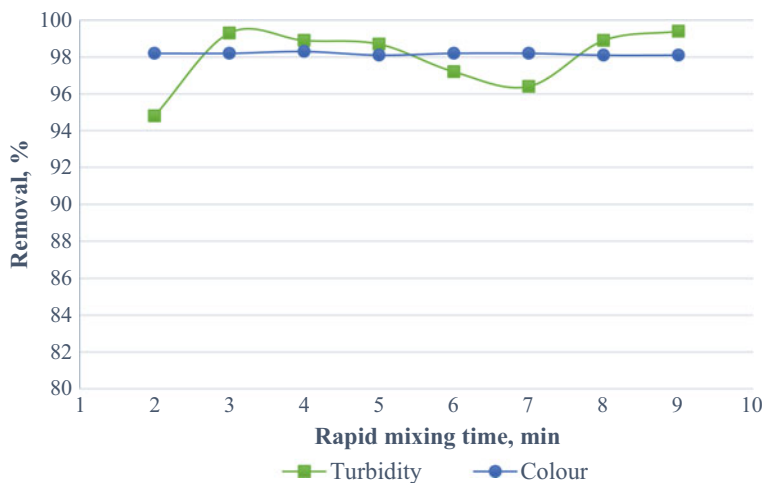


Fig. 2 Effect of rapid mixing time on turbidity and colour removal at pH 8.1 and 1 g/L of SnCl_4

stated that the removal efficiency of colour at 970 mg/L of FeCl_3 and pH 5 was only 60% for 3 min of rapid mixing.

It can be inferred that mixing time contributes significantly to the performance of the coagulation process [34]. However, longer mixing times can also disturb the performance of coagulation because the aggregated particles tend to break and drop the size of the flocs formed [23]. Hence, the 3 min reaction evaluated in the current study is considered optimal to allow enough collisions between the coagulant and the particles for a good settlement.

4 Conclusion

Rapid mixing speed and time are the two of the essential factors that need to be determined for a good coagulation and flocculation process. In this study, it was found that the best mixing speed to remove turbidity and colour was 220 rpm, which exhibited 99.5 and 98.3% reductions of colour and turbidity, respectively. The mixing time that achieved 99.3% of turbidity removal and 98.2% for colour was 3 min. Overall, the studies concluded that inadequate mixing speed and time could lead to a poor coagulation performance.

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