

# Performance of *Azadirachta Indica* as Bio-Flocculant in Reducing Turbidity Concentration from Landfill Leachate

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## Abstract

It is widely established that the amount of municipal solid debris produced daily continues to rise. As a result, municipal solid waste (MSW) landfills struggle with leachate formation, which puts groundwater and surface water at serious risk. Leachate is a liquid that has extracted dissolved and suspended materials from waste and travels through landfills. Dumping landfill leachate directly into a body of water or the environment puts the ecology and public health in danger since it contains high COD, pH, ammonia nitrogen, turbidity, and heavy metals. Therefore, finding and offering an efficient landfill leachate treatment is undoubtedly required. This study aims to evaluate the performance of the bio-flocculant *Azadirachta indica* (bf-Ai) as a flocculant in the coagulation-flocculation process for the treatment of landfill leachate under various experimental circumstances. Additionally, the performance of conventional coagulant alone (alum) was compared with a combination of alum (as coagulant) and *Azadirachta indica* (Ai) as bio-flocculant (alum + bf-Ai). Based on this study, the combination of alum + bf-Ai recorded higher removal of turbidity, which is 64% compared to alum alone (62%) at raw pH of leachate and the dosage of alum + bf-Ai as 0.8g and 5g, respectively. Additionally, a reduction in the usage of alum dosage can also be seen in this combination, which decreases from 1.4g to 0.8g only. This reduction is a positive sign since alum alone has produced hazardous sludge (secondary pollutants), damaging the environment and human health. Thus, further research should be conducted on the potential of Ai as a bio-flocculant aid.

## 1. Introduction

Malaysia's current daily trash production is approximately 1.1 kg. Every day, more than 26,500 tons of solid waste are almost entirely disposed of in the nation's 166 active landfills [1]. Landfilling is the predominant method for disposing of municipal solid waste (MSW) in developing countries like Malaysia, despite the availability of other

disposal options. This can be mainly attributed to its inherent strength in terms of cost savings and simplified operational mechanisms, but there is a drawback. One of the common problems with the landfilling procedure is the environmental contamination brought on by the landfill leachate [2]. Leachate from landfills contains considerable organic waste and toxins, which can harm the ecosystem and human life. It also has a high concentration of colour, ammoniacal nitrogen ( $\text{NH}_3\text{N}$ ), chemical oxygen demand (COD), biological oxygen demand ( $\text{BOD}_5$ ) and turbidity [3]. Turbid water becomes cloudy or dense, potentially interfering with downstream treatment processes and negatively affecting customer acceptance; effective drinking water treatment entails minimising it by prematurely clogging the filter. However, turbidity can make filtering difficult [4]. Therefore, this study identifies the potential of bio-flocculants in removing COD and turbidity from the landfill leachate.

The coagulation flocculation process is a well-established method used in wastewater treatment. Chemical coagulation is a process that involves adding chemicals known as coagulants to reduce the concentration of pollutants in a solution. The consequences of using chemical coagulants, such as high chemical residues, toxic sludge, and health issues from prolonged exposure, have been thoroughly examined [5]. Other than that, the disposal cost may also increase due to the amount and toxicity of the sludge and the difficulty of dewatering it. Researchers are currently extensively studying the potential of natural coagulants and flocculants to replace or reduce the usage of chemical coagulants [6]. In terms of performance and environmental sustainability, natural flocculants are competitive. Another advantage of bio-flocculants is that they are biodegradable, environmentally friendly and more affordable than chemical flocculants. The Meliaceae family includes neem (*Azadirachta indica*), commonly known as neem tree or Indian lilac, which is a natural herb with many different traditional uses, especially for hair, dental, cosmetics, health care, and pesticide products. Moreover, *Azadirachta indica* can also be found easily in most Asian countries, including Malaysia. Several research studies have explored the potential of *Azadirachta indica* as an absorbent and purifying water agent. Patel [7] recorded heavy metal removal from water up to 98.2% using this absorbent. Rubini et al. [8] studied the potential of *Azadirachta indica* as a flocculant in kitchen wastewater treatment. The results showed that the turbidity of wastewater was effectively removed up to 96%, and the use of alum dosage as the main coagulant was reduced to 50% in this treatment.

*Azadirachta indica* has the potential to be a highly effective bio-flocculant in green technology due to its high positive zeta potential of +34.6 mV [9]. Based on this characteristic, a high positive charge in *Azadirachta indica* can neutralise the suspended particle in leachate, which has -22.94 mV of zeta potential [9],[10], and allow destabilisation of a particle as well as enhance the agglomeration of the suspended particle during the treatment. Additionally, the surface characteristics of Ai, which are porous and have an irregular surface structure, can potentially become an active surface area that favours the adsorption of pollutants. Therefore, those factors will increase the effectiveness of the treatment. However, the removal mechanism of b-Ai and the factors influencing landfill leachate treatment performance are not well established. Furthermore, *Azadirachta indica* is a local plant, non-toxic, green material, cheap and easy to obtain in our country compared to a chemical coagulant. This underscores the significance of investigating the possibility of this bio-flocculant, as its health-promoting properties are associated with its high levels of antioxidants. Besides, *Azadirachta indica* naturally contains antibacterial qualities that can help remove harmful bacteria and lower the bacterial load in water or wastewater when employed as a coagulant [11]. This is due to its ability to hold significant levels of microbiological pollutants, making it beneficial in treating landfill leachate. *Azadirachta indica* was utilised as a natural flocculant to treat anaerobic stabilised landfill leachate in this case study. This study investigates the ability of the green local flocculant *Azadirachta indica* (bf-Ai) to remediate landfill leachate.

## 2. Methodology

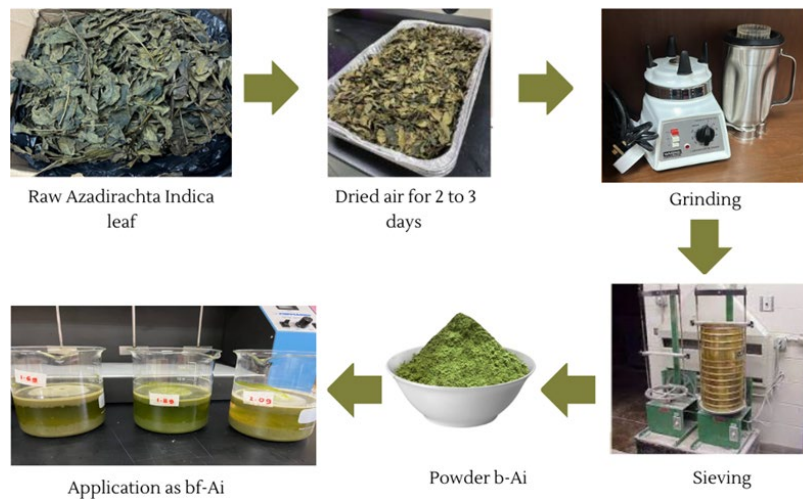
### 2.1 Sampling

Sampling of landfill leachate was conducted at one of the municipal landfill sites in Kota Kinabalu, Sabah and then characterised in the Environmental Laboratory, Faculty of Engineering, Universiti Malaysia Sabah (UMS). The sample was promptly taken to the UMS environmental laboratory for characterisation and testing. The sampling, preservation, and sample analysis procedures followed Standard Methods for the Examination of Water and Wastewater [12].

### 2.2 Preparation of Coagulation-Flocculation Material of *Azadirachta Indica* and Alum

*Azadirachta indica* (Ai) was collected from local suppliers and air dried for 2 to 3 days before grinding into fine particles. After that, the dried *Azadirachta indica* was ground into powder form through a typical 90-micron sieve. Subsequently, the sieved powders were washed with distilled water, filtered and dried again in an oven at a temperature of 105°C for 24 hours before being used as bio-flocculant *Azadirachta indica* (bf-Ai) [8], as illustrated in Fig. 1. The prepared bf-Ai is stored in a glass container at room temperature before being used as a coagulant. In this study, conventional coagulant Aluminium Sulphate  $\text{Al}_2(\text{SO}_4)_3$  or alum, supplied by a local supplier, was used

in powder form. Additionally, the surface morphology and the chemical compositions of b-Ai were observed using Field Emission Scanning Electron Microscopic (FESEM-EDX).



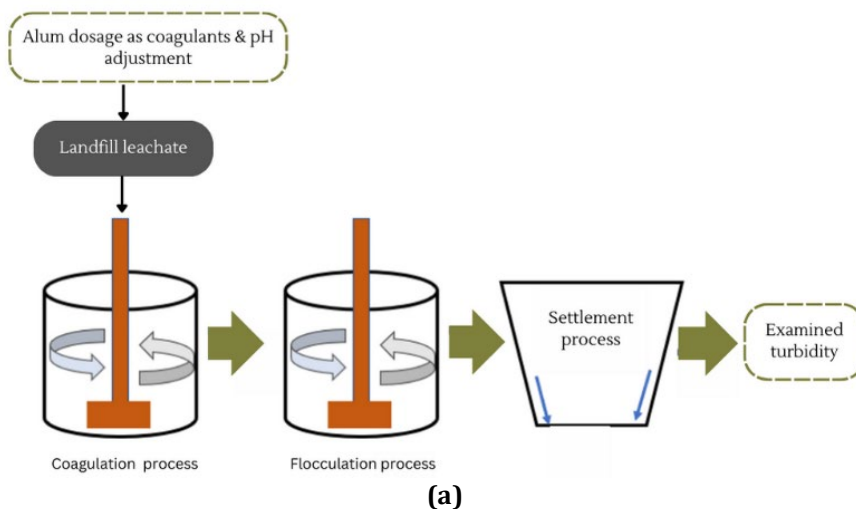
**Fig. 1** Preparation of bio-flocculant *Azadirachta indica* (bf-Ai)

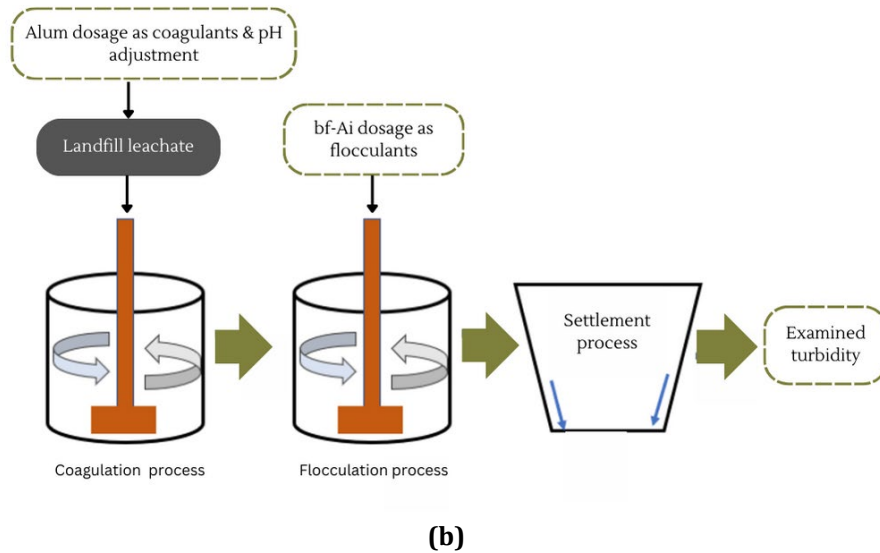
### 2.3 Batch Study

The jar test aimed to evaluate the performance of alum as a coagulant and *Azadirachta indica* as a bio-flocculant in the coagulation/flocculation process. A six-beaker jar test apparatus was used to simulate the coagulation/flocculation process. In this study, the performances of alum alone and alum with bf-Ai were determined by varying two influential factors: the dosage and pH of the solution. Therefore, two sets of experiments were conducted. To evaluate alum's performance alone, the alum dosage varied from 0.2 g to 2.0 g while the pH was from 5 to 10, respectively, at 500 mL of leachate (Fig. 2(a)). The pH of leachate was adjusted using hydrochloric acid (5 M) and sodium hydroxide (5 M). Next, the performance of bf-Ai was tested by varying the dosage of bf-Ai and pH of the solution, from 0.2g to 10 g and 5 to 10, respectively (Fig. 2(b)). Concerning the practical condition, reaction time and the rotating speed of the paddle were constant. It is set to be 200 rpm for the initial 3 minutes (rapid mixing), 40 rpm for the subsequent 10 minutes (slow mixing), and 0 rpm for the final 30 minutes (settling) of all experiments [13]. Turbidity was selected as a measurement parameter for this study, and its concentration was measured using a turbidity meter. The turbidity removal efficiency was obtained using the following Eq. (1).

$$\text{Percentage Removal (\%)} = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

where  $C_i$  and  $C_f$  refer to the initial and final turbidity concentrations, respectively.



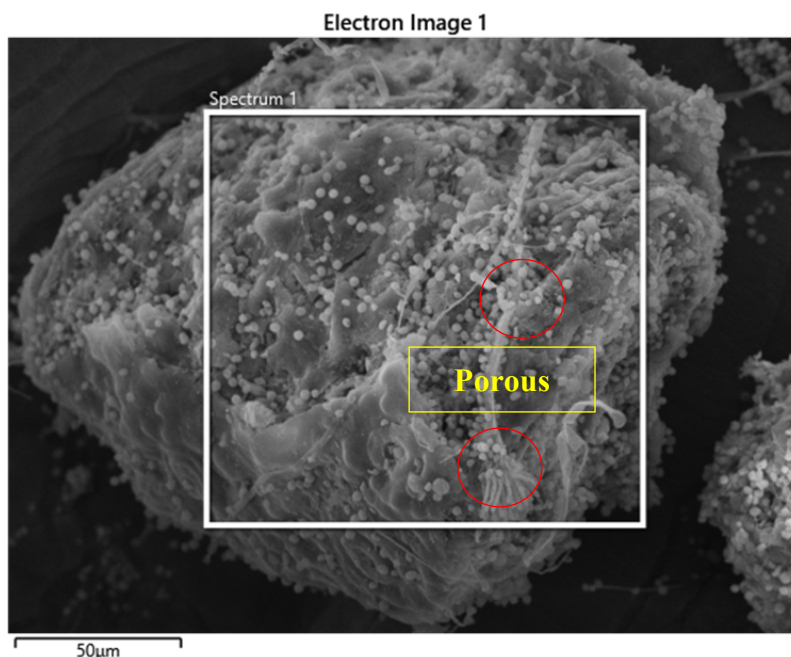


**Fig. 2** Schematic diagram for jar test process: (a) alum alone; and (b) alum + bf-Ai

### 3. Results and Discussion

#### 3.1 Characteristics of *Azadirachta Indica*

The surface morphology of b-Ai at a magnification of 1600x is presented in Fig. 3. The surface of bf-Ai is irregular in shape, porous, and contains small rounded white particles. Nandyanto [14] mentioned that the porous surface enhances the absorption of the particles. The elemental EDX analysis, as depicted in Fig. 4, highlights the existence of carbon (C), Oxygen (O), Calcium (Ca), Potassium (K), Silica (Si), Copper (Cu), Phosphorus (P) and several other elements which are either presented in small amount or negligible. This data shows carbon and oxygen elements are predominant in the composition, recorded at 65.8% and 31.6%, respectively. According to Zainal et al. [10], the carbon in bf-Ai might contribute to the presence of polysaccharides, consisting of cellulose, hemicellulose, and lignin. Polysaccharides are bio-polymers capable of neutralising particle charges and aggregating flocs through bridging mechanisms [10]. Thus, b-Ai has the potential to function effectively as a natural flocculant.



**Fig. 3** Surface morphology of b-Ai at 1600x magnification

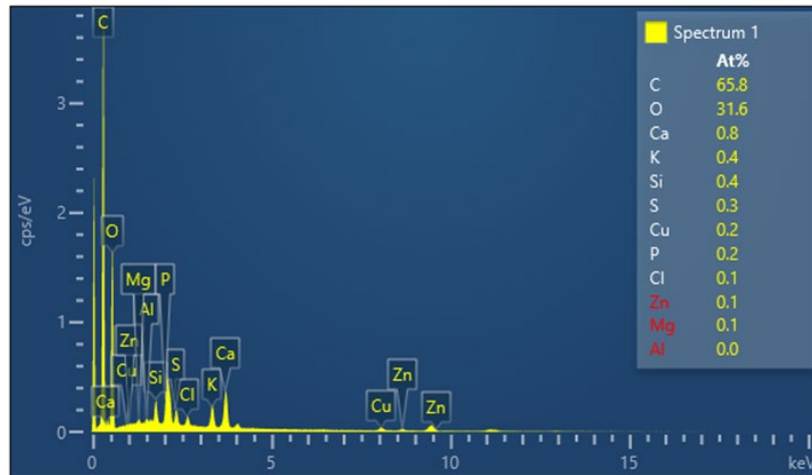


Fig. 4 EDX elementary results for bf-Ai

### 3.2 Influence of Dosage Toward Alum Alone, Ai Alone and Al + bf-Ai in Removing Turbidity

Varying amounts of alum powder were used to assess the impact of alum dosage, Ai alone, and Al + bf-Ai on turbidity removal. The dosage of coagulants ranged from 0.2g, 0.4g, 0.6g, 0.8g, 1.0g, 1.2g, 1.4g to 1.6g, 1.8g, 2.0g, 5.0 g, 10 g and 20g. Despite the raw leachate's pH being 8, the test's pH value was kept constant. The removal percentage for turbidity is depicted in Fig. 5.

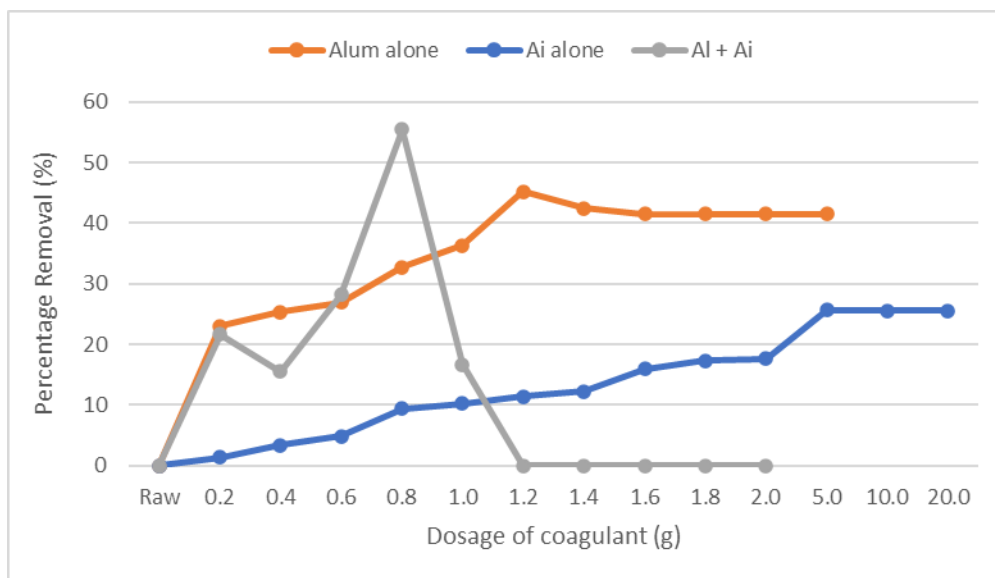


Fig. 5 Effect of coagulant dosage on turbidity removal in landfill leachate (experiment conditions: volume sample 500 mL, raw pH and reaction times of rapid mixing 200 rpm (3 minutes), slow mixing 40 rpm (10 minutes) and settlement 30 minutes)

The percentage of turbidity that can be removed by alum effectively is 45.2% at a dosage of 1.2g. As seen in Fig. 5, removal of turbidity increases with alum dosage. This pattern might be due to the positively charged ions that are produced during the hydrolysis process of alum in water. These flocs serve as coagulants and aid in destabilising the water's suspended particles and colloids. More positively charged flocs are produced as the alum dosage rises, enhancing coagulation and boosting removal effectiveness [15]. The graph shows constant readings from 1.4 g to 2.0 g dosage of alum due to large flocs forming; thus, the coagulant may become saturated, meaning that all of the active sites on the floc surfaces would be taken up. Further increases in the alum dosage will, therefore, have an insignificant effect on eliminating new pollutants, resulting in a steady reading. The subsequent ion neutralisation and destabilisation process between the negative and positive charges of alum can be used to explain the elimination of turbidity [16]. They neutralise the negative charges on suspended particles and colloidal matter in the leachate, allowing them to come together and form larger flocs. This process is called coagulation

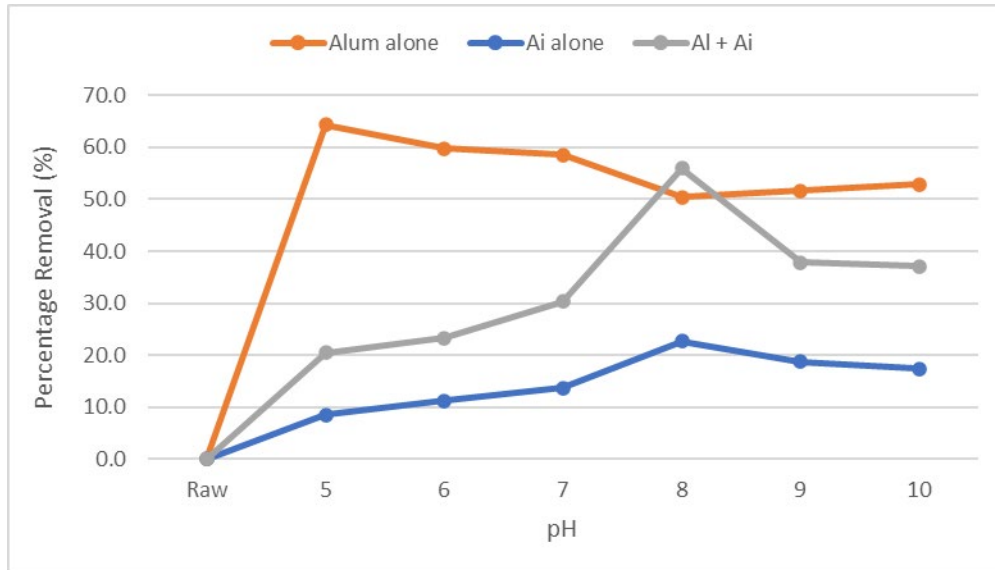
and flocculation. It is important to note that the specific reactions and mechanisms may vary depending on the composition of the leachate and the pH conditions.

Next, the performance of Ai as a coagulant was also studied. Based on the graph in Fig. 5, the highest percentage removal of turbidity is 25.6% at a bf-Ai dosage of 5.0g. This value is low compared with the performance of alum prior. Nevertheless, the data indicate that bf-Ai can function as a coagulant, facilitating optimal interaction with pollutants and promoting the formation of stable flocs. Besides, an observation during the test shows that larger flocs are formed by the coagulant during the coagulation process, aiding in the trapping and aggregation of smaller particles. A previous study by Khan et al [17] on *Azadirachta indica* found that the optimal dosage for water treatment to remove 86% of turbidity (considered a low to medium strength pollutant) was 3 g/L. The high strength of pollutants in landfill leachate and different ranges might be explained by the lower removal of turbidity recorded in this study compared to previous research data. Therefore, a combination of Ai with other coagulants might increase the performance of this treatment method.

As seen in Fig. 5, the combination of alum as coagulant and Ai as bio-flocculant (alum+bf-Ai) records higher removal than other methods. The most effective alum dosage is 0.8 g and bf-Ai 5g, which reduces turbidity to 55.5. Additionally, the use of alum dosage decreases by 33.3% when compared to using alum alone. Reducing alum use will directly lessen the production of hazardous sludge, lower the risk of environmental issues, and reduce operational costs in treatment plant management. Ai also contains bioactive compounds with flocculating properties, such as proteins, tannins, and polysaccharides. These substances can speed up the process of removing suspended solids through sedimentation or filtration methods and promote the clumping of those solids [18]. Insufficient dosages may lead to ineffective neutralising charges against negative charges on kaolin particles [19]. Conversely, overdosing might result in saturated polymer bridge sites. A lack of suspended particles to create more bridges between particles will cause unstable particles to stabilise. These facts explained the decremental performance of alum+bf-Ai after reaching the optimum alum dosage at 0.8g. Based on this data, Ai works effectively as a bio-flocculant compared to a coagulant.

### 3.3 Influence of pH Toward Alum Alone, Ai Alone and Al + bf-Ai in Removing Turbidity

Another studied factor is pH. The influence of pH on turbidity under those three methods was tested; the findings are presented in Fig. 6.



**Fig. 6** Effect of pH on turbidity removal in landfill leachate (experiment conditions: volume sample 500 mL, dosage coagulants and flocculant are taken from section 3.2 (at optimum condition) and reaction times of rapid mixing 200 rpm (3 minutes), slow mixing 40 rpm (10 minutes) and settlement 30 minutes)

Based on Fig. 6, the performance of alum alone, Ai alone and alum + bf-Ai is effective at pH 5 and 8, respectively. The maximum percentage removal of turbidity recorded for alum alone, Ai alone, and alum + bf-Ai is 64.4%, 22.8% and 56%, respectively. As seen, alum performs well at pH 5, which shows that acidic condition accelerates alum's performance to break down pollutants inside the leachate, while Ai and alum+bf-Ai record high removal in alkaline conditions (pH 8). According to Sahoo et al. [20], bf-Ai has a high positive charge, which attracts ions with a negative charge in the leachate, causing floc to coagulate and sink to the bottom. Theoretically, a natural coagulant's negatively charged active component might draw bivalent cations from water, form a net-like structure, and remove those cations by sweeping coagulation in an alkaline state, and vice versa [21]. Therefore,

the efficiency of turbidity reduction increases with the rising pH levels as depicted in Fig. 6. Previous researchers also supported these data as neutral or alkaline conditions of the solution showed higher removal efficiencies than acidic conditions [22]. pH 8 was the most cost-effective pH for adjusting the leachate's pH, as it fell within the optimal pH range for treatment.

#### 4. Conclusion

The application of Ai in landfill leachate treatment shows low performance compared to other coagulants. However, the combination of alum+bf-Ai improves the removal of turbidity (56%) and records a reduction in the usage of alum dosage at pH 8. The characteristics of Ai obtained from the FESEM test also highlighted that the composition of Ai is dominated by carbon elements, which might contribute to the presence of polysaccharides and consist of cellulose, hemicellulose, and lignin. These elements can influence the coagulation-flocculation mechanisms, such as neutralising particle charges and aggregating flocs through bridging mechanisms. Based on the data obtained, Ai can work as an effective bio-flocculant and reduce the usage of alum dosage. However, additional research and development are needed, including finding the optimisation condition and effectiveness when combined with existing landfill leachate treatment. The long-term effects of employing these compounds on water quality, managing leftover sludge, and human health should also be closely watched and evaluated.

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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 contribution to the paper as follows: **study conception and design:** Siti Nor Farhana Zakaria, Hamidi Abdul Aziz; **data collection:** Monika Selvam, Elfreda Peter; **analysis and interpretation of results:** Nurmin Bolong; **draft manuscript preparation:** Elfreda Peter, Maheera Mohamad. All authors reviewed the results and approved the final version of the manuscript.*

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