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Potential Use of Polyaluminium Chloride and Tobacco Leaf as Coagulant and Coagulant Aid in Post-Treatment of Landfill Leachate

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Abstract

A study was conducted to treat stabilized leachate by applying polyaluminium chloride (PAC) and tobacco leaf extract as a coagulant and coagulant aid. Experimental results indicated that the tobacco leaves were positively charged. The removal rate of the chemical oxygen demand, using 1500 mg/L PAC as a sole coagulant, was approximately 63% and increased to 91% when 1000 mg/L PAC was mixed with 1000 mg/L tobacco leaf. Additionally, 1500 mg/L PAC with 250 - 1000 mg/L tobacco leaf and 54% ammoniacal nitrogen was removed, compared with only 46% reduction using 1500 mg/L with only 46% reduction.

Keywords: Polyaluminium Chloride (PAC), Tobacco Leaf, Landfill Leachate, Coagulation-Flocculation

1. Introduction

The rapid worldwide industrial and commercial growth of the past decades has led to an increasing amount of municipal solid-waste production. Toward the end of the 1990s, the annual waste production calculated ranged from 300 to 800 kg per person in developed countries, while other countries generated less than 200 kg per person. These large amounts of solid waste mainly are disposed in landfill sites due to the economic advantages. However, a sanitary landfill will produce wastewater known as leachate, which has become of significant interest, as it threatens surface and groundwaters (1). Sanitary landfills are developed to cater to long-term disposal of refuse. Over time, the waste characteristics contained in the leachate change depending on the age of the sanitary landfill. A stabilized landfill leachate (which exists more than 10 years) is defined by high-strength wastewater, constituting a major fraction of high molecular-weight organics, about 30% humic acids, ammonia, heavy metals, and color (2, 3). It also contains toxic compounds, such as adsorptive organic halogen (AOX) compounds, chloride compounds, and ammoniacal nitrogen (NH3-N), which are identified as the major toxicants to living organisms (e.g., Salmo gairdnieri and Onchorhynchus nerka). Among the common characteristics of a stabilized landfill leachate is high-strength NH3-N (3000 - 5000 mg/L), moderately high-strength chemical oxygen demand (COD) (5000 - 20000 mg/L), and a low ratio of biochemical oxygen demand (BOD) over COD, which is less than 0.1 (4, 5).

Untreated leachate potentially can permeate the groundwater or mix with surface waters, causing severe chemical contamination in dissolved or suspended forms and lead to a long-term impact on the environment (3, 4). Due to the significant pollution and environmental concerns, numerous studies have been conducted on the treatment of stabilized leachate. Among them are reverse osmosis (5), Fenton processes (3, 6), and physicochemical techniques, such as coagulation-flocculation, chemical precipitation, ammonia stripping, membrane filtration, and adsorption (4). Depending on many factors, including operation conditions, the efficiency of each treatment may differ (1). According to Ntampou et al. (5), physicochemical treatment are favorable for stabilized leachate. Coagulation-flocculation has been a common technique practiced using a variety of conventional coagulants such as alum, polyaluminium chloride (PAC), and polyaluminium silicate chloride (7-12). It has been reported that conventional coagulants are efficient in removing 10% - 25% of COD from young leachates and 50% - 65% of COD from old leachates (13). However, this may lead to moderate removals of COD (or total organic compound [TOC]) content and create drawbacks, for example, generating excessive sludge and producing an increase of aluminium or iron concentrations in the final effluent.

In recent years, natural coagulants have been researched as a more environmentally friendly approach to substitute for the commercial coagulants in coagulation-flocculation treatment. Plant-based coagulants (14, 15), such as cactus (16), nuts such as chestnuts and acorns (17), starch (18, 19), and Moringa oleifera (20-24) have been investigated on other types of wastewater as either a primary coagulant or as a coagulant aid for a chemical coagulant. For leachate treatment, very limited information is available pertaining to applications of natural coagulants (25, 26). Besides being biodegradable, natural coagulants also are presumed safe for human health and unhazardous toward the environment.

In support of the research mentioned above, this study aims (1) to develop a new natural coagulant made from tobacco resources in order to treat stabilized leachate; (2) to extract a coagulant from the leaves of tobacco, and examine its characteristics based on the zeta potential and element composition; and (3) to evaluate the efficiency of coagulation-flocculation processes for the removal of turbidity, SS, color, COD, and NH3-N from landfill leachate using 1500 mg/L PAC with tobacco leaf extract as a coagulant aid.

2. Materials and Methods

2.1. Leachate Sampling and Characterization

Samples of leachate were collected from the Alor Pongsu landfill in Bagan Serai, Perak. The Alor Pongsu landfill started operations in 2000 and receives an average of 660,000 metrics tons of solid waste per year. As this landfill has operated for more than 10 years, it falls under the category of an old and stabilized landfill. The characteristics of the leachate taken from the detention pond at Alor Pongsu are listed in Table 1.

Table 1. Characteristics of Raw Leachate From Alor Pongsu

Parameter	Unit	Leachate, Average	Standard[a], (EQA 1974)
pH	-	8.22	6.0 - 9.0
Temperature	°C	28.85	40
Turbidity	NTU	173	-
SS	mg/L	513	50
Color	PtCo	15469	100
COD	mg/L	3863	400
NH3-N	mg/L	1519	5

[a]The standard is according to the environmental quality regulation 2009 (control of pollution from solid waste transfer station and landfill).

2.2. Preparation of Tobacco Leaf Base Coagulant Aid

Dried tobacco leaves were obtained from a local tobacco farm in Kota Bharu, Kelantan. The tobacco leaves first were washed repeatedly with water to remove dust and insoluble impurities. Next, the leaves were weighed, and the reading was recorded at a fixed weight of 120 g. They then were blended at a high speed in a solution of 550 mL of distilled water by using a domestic blender. The mixture was poured through muslin cloth and squeezed dry to separate the solids from the solution. The leaf dregs were dried in an oven at 105°C for 48 hours while the suspension was filtered into a beaker. The leaf dregs were weighed again. Then, the filtrate stock solution was stirred using a magnetic stirrer for 30 minutes at room temperature. The final concentration of the tobacco leaf extract was calculated at 93.5 mg/L. This stock solution was stored in the cold room at 4°C for future use.

2.3. Characterization of Tobacco Leaf

The morphology of the tobacco leaf was analyzed using scanning electron microscopy (SEM). The major elements were detected using the Fourier transmission infrared-red technique. The zeta potential was analyzed using Nano Zetasizer ZS (Malvern, UK).

2.4. Jar Test Experiments

In this research, PAC was purchased in liquid form (18% [w/v]) from water purifying materials Co. Ltd. It was utilized as the main coagulant, followed by the tobacco leaf natural coagulant aid, to conduct the jar test experiments. The conventional jar test with a six-unit multiple stirrer system was used. Six beakers with a 1000 mL volume were filled with 500 mL leachate. A dosage of 1500 mg/L PAC was added into each beaker, and the pH of the suspension was fixed at 6. The leachate was stirred at a rapid speed of 250 rpm for 5 minutes. Tobacco leaf extract then was added into each beaker at different dosages, for example, 500, 750, 1000, 3000, 5000, and 10000 mg/L. One beaker remained as the control without any addition of tobacco leaf. This was followed by slow stirring at a speed of 60 rpm for 30 minutes. The suspension then was allowed a settling time of 45 minutes.

The supernatant samples were withdrawn from 2 cm below the surface of the suspension. The parameters of turbidity, suspended solids, color, COD, and NH3-N were tested in accordance with the APHA standard method.

2.5. Field Emission Scanning Electron Microscope and Energy Dispersive X-ray

An image of flocs produced from the coagulation of leachate was captured using SEM to determine the morphology and the surface composition at the optimum condition. An appropriate amount of the sludge sample was withdrawn from the bottom of the cylindrical beaker at the end of the settling period. Then, the sludge was filtered by using filter paper, and finally, the sludge was allowed to dry at room temperature. The dried sludge was ground using a mortar and pestle into a fine powder form. A small amount of this dried powder was attached to double-sided carbon tape. SEM images were taken at various magnifications (50, 100, and 500 X). Then, measurements of energy dispersive X-rays (EDAX) were taken to detect that the significant element exists in the dried sludge. The first sample was sludge from the coagulation process using PAC with optimum pH 6 at a dosage of 1500 mg/L. The second sample was sludge from the coagulation process using PAC and tobacco leaf at a dosage of 1500 mg/L and 1000 mg/L, respectively, with optimum pH 6.

3. Results and Discussion

3.1. Characteristics of Tobacco Leaf

Figure 1 shows the morphology of the tobacco leaf analyzed using SEM. Three major elements were detected using the Fourier transmission infrared-red technique: calcium, oxygen, and carbon, with a weight percentage of 53.99%, 26.10%, and 6.60%, respectively. Other minor elements were detected: sulphur (S), aluminium (Al), phosphorus (P), potassium (K), chlorine (Cl), and silicon (Si). Based on the finding, the composition of the tobacco leaf is generally nontoxic and biodegradable. According to Masson (27), tobacco is known to be a plant with high tolerance and can accumulate high concentrations of toxic elements in its leaves without suffering any impairment of growth. The zeta potential obtained for tobacco is -3.57 mV. It can be classified as an anionic coagulant with a negative surface charge. If a pre-polymerized coagulant is combined with a natural coagulant with a lower zeta potential (e.g., anionic polyelectrolyte), the aggregating ability would be improved (25).

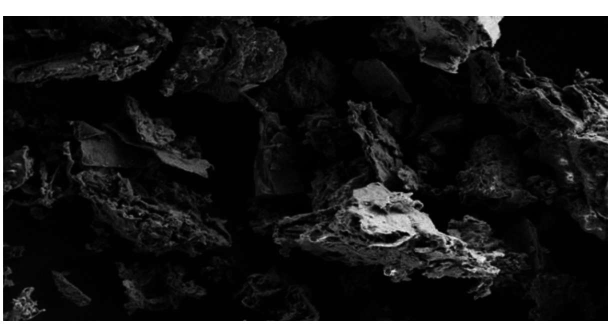

Figure 1. SEM Image of a Tobacco Leaf

3.2. Removal Rate

A shown in Figure 2, a high correlation between turbidity and SS was observed because the sampling was made at a single particular site. As the sole coagulant, 1500 mg/L PAC successfully removed 99% of turbidity and 95% of SS. The combination of 1000 mg/L tobacco leaf extract and 1500 mg/L PAC removed approximately 21% of turbidity and 48% of SS. In particular, the removal of the turbidity and SS proportionately decreased as the tobacco leaf extract dose was increased to over 10000 mg/L.

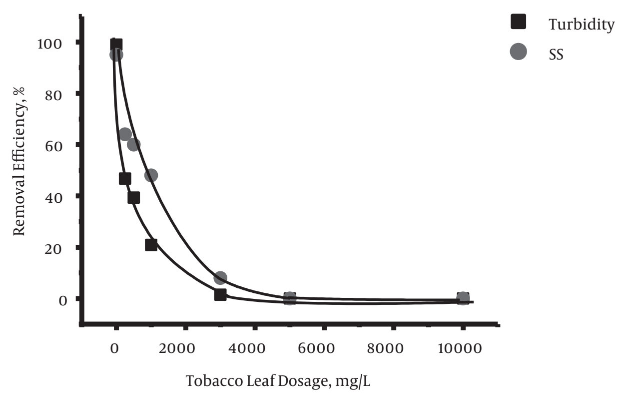

Figure 2. Removal Efficiency of Turbidity and Suspended Solids by Applying PAC and Tobacco Leaf Coagulant Aid at Various Dosages

Increasing the tobacco leaf extract dose significantly affected the color removal. In fact, 1500 mg/L PAC combined with 1000 mg/L tobacco leaf extract only managed to remove 86% as compare to 96% with 1500 mg/L PAC alone, as shown in Figure 2. The removal rate for COD with 1500 mg/L PAC was 62%, 1% below the removal rate using 1000 mg/L tobacco leaf as the sole coagulant at pH 4. However, a combination of PAC at 1500 mg/L and tobacco leaf at 1000 mg/L effectively could remove COD up to 91%, as shown in Figure 3.

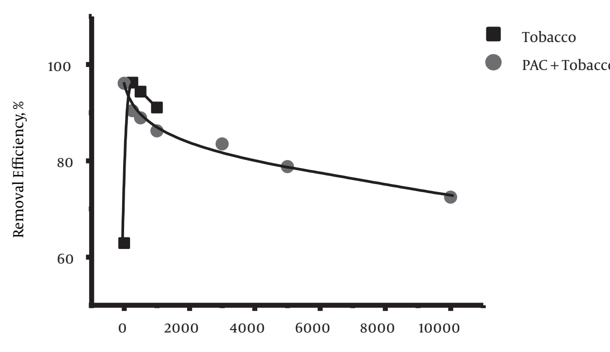

Figure 3. Removal Efficiency of COD by Applying PAC and Tobacco Leaf Coagulant at Various Dosages

The removal rate for NH3-N with 1500 mg/L of PAC was

46%, 7% above the removal rate using a combination of PAC and tobacco leaf at a dosage of 1000 mg/L, as shown in Figure 4. However, further experiments showed that tobacco leaf extract effectively could remove NH3-N in leachate up to 75% using 1500 mg/L of PAC combined with a tobacco leaf extract dosage up to 10000 mg/L, as shown in Figure 4.

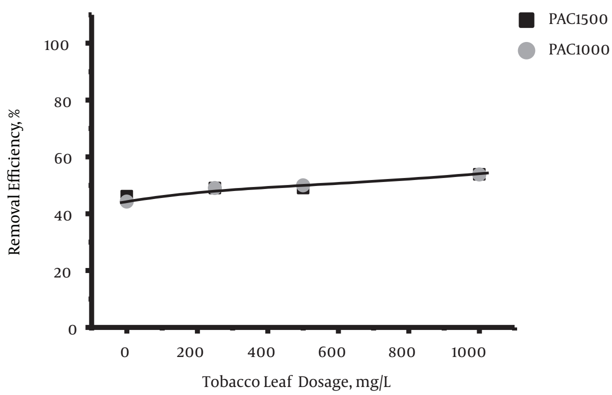

Figure 4. NH3-N Removal Efficiency by Applying PAC and Tobacco Leaf Extract

In general, results showed that the optimum dosage for PAC as the sole coagulant at pH 6 in leachate was 1500 mg/L. At this optimum dose, PAC effectively removed 99% of turbidity, 95% of SS and 96% of color, as compared with the use of 1500 mg/L PAC combined with 1000 mg/L tobacco leaf extract, which had only about 21%, 48%, and 86% removals, respectively. However, a combination of PAC and tobacco leaf extract removed 91% of COD and 54% of NH3-N, compared with the use of PAC as the sole coagulant, which only managed to remove 63% of COD and 46% of NH3-N. This indicated that tobacco leaf enhanced the performance of PAC in removing COD and NH3-N in leachate.

3.3. Sludge Analysis

The surface morphology in the experiment involving PAC as the sole coagulant and PAC combined with tobacco leaf as a coagulant aid was determined by the SEM technique, as shown in Figure 5. In this test, high-resolution SEM was applied at a magnification of 500X for both coagulants. As depicted, the image of flocs size for PAC as the sole coagulant is much smaller than using PAC with tobacco leaf extract as a coagulant aid. By comparing both images, the combination of PAC with tobacco leaf extract as a coagulant aid was proven to form bigger flocs than PAC as the sole coagulant.

3.4. Particle Size Analysis Using Mastersizer

In this study, coagulant and the adsorbents were tested and compared for their size before and after the reaction.

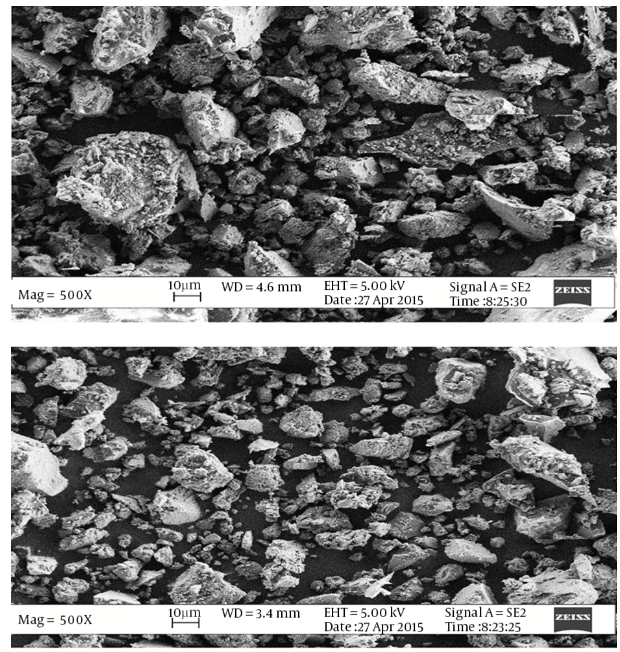

Figure 5. SEM Image After Treatment, a, Using PAC as the sole coagulant; and b, using PAC and tobacco leaf extract as a coagulant aid.

Table 2 shows the results obtained from Mastersizer analysis. As shown in the Table 2, the coagulation and flocculation product was bigger than the original coagulant. Accumulation occurred in the coagulation and flocculation processes, which formed flocs. This data also proved that the reaction that occurred during the jar test had an affinity towards the pollutant. Adsorption refers to the attraction and the binding to the surface of the adsorbent. When the sample that carried the pollutant was flowing into the adsorbent, the binding of oppositely charged elements happened on the surface of the filtration media. It increased the size of the flocs, with additional pollutant elements.

Table 2. Summary of the Mastersizer Analysis for the Coagulants Involved in This Study

Coagulant	Size of Particle, μm	
	Before Reaction	After Reaction
PAC	0.212	33.364
PAC + Tobacco leaf extract	0.212	34.511

4. Conclusions

It can be concluded that 1500 mg/L PAC combined with 1000 mg/L tobacco leaf exhibits a synergic effect on COD and NH3-N removals and is more advantageous than the use of 1500 mg/L PAC alone. However, removal of turbidity, SS and, color is more efficient when using 1500 mg/L PAC as the sole coagulant. Therefore, this study reveals that PAC is more efficient in improving physical characteristics of leachate, but a combination of PAC and tobacco leaf extract better removes COD and NH3-N.

Footnotes

Authors' Contribution All authors did a fair share in both academic writing and research work.

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