



AENSI Journals

Advances in Environmental Biology

ISSN-1995-0756 EISSN-1998-1066

Journal home page: <http://www.aensiweb.com/AEB/>

The Study of Flocculant Characteristics for Landfill Leachate Treatment Using Starch Based Flocculant from Durio Zibethinus Seed

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ARTICLE INFO

Article history:

Received 25 July 2014

Received in revised form

8 August 2014

Accepted 15 September 2014

Available online 25 October 2014

Keywords:

Leachate Treatment, Flocculation, Natural Polymer, Starch, Durio Zibethinus Seed.

ABSTRACT

Background: Flocculation has been extensively used as a semi-aerobic landfill leachate treatment, prior to other treatment methods. Although inorganic and synthetic polymers are prominent in landfill leachate treatment, its application may introduce potentially toxic residual. As alternative to potential risk, starch based flocculant had been produced from durian seed waste. Plus, no attempt has been made to avail abundant waste of Durian Seed Starch (DSS) as a natural flocculant for wastewater and leachate treatment. In this paper, an attempt has been made to study the characterization performance of DSS in the flocculation process. Jar test results had ascertained that optimum pH and dosages values for DSS flocculant were pH 6 and 4000mg/L with removal of true colour and turbidity were 34% and 36.9% respectively. The scanning electron microscopy (SEM) images show the presence of contaminated particle layer on top of the DSS after the treatment process. The Fourier transform infrared spectroscopy (FTIR) analysis showed DSS was the major constituent of the floc. FTIR results verified that the agglomeration consist of leachate contaminants. Therefore, DSS could be a feasible selective flocculant in reducing the usage of inorganic polymer for semi-aerobic landfill leachate treatment.

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To Cite This Article: Mohd Faiz Muaz A.Z., Mohd Suffian Y., Hamidi A.A., The Study of Flocculant Characteristics for Landfill Leachate Treatment Using Starch Based Flocculant from Durio Zibethinus Seed. *Adv. Environ. Biol.*, 8(15), 129-135, 2014

INTRODUCTION

Flocculation is a process of propagation and agglomeration of destabilised particles to gather into larger flocs [1]. During flocculation, the microscopic coagulated particles aggregate with each other to form larger flocs. These bulky flocs then are able to aggregate with suspended polluting matter [2]. Throughout the decades, flocculation has become one of the most widely used techniques for the water, wastewater treatment and landfill leachate treatment [3,4]. It can be used in subsequent solid removal processes such as sedimentation, rapid or membrane filtration, and dissolved air flotation (DAF) [5,6]. Recently, flocculants as a main part in flocculation process has been given full attention by many researchers in improving the sustainability to the environment [7,8,9]. Even though the established flocculants originate from inorganic and synthetic organic polymer show good performance and efficiency, some of them were found promotes to secondary pollution and environmental issue [10]. For that reason, the study of natural based flocculant in reducing the dependent on inorganic flocculants is desired. Plus, the biodegradable nature of organic based flocculants is necessary for environment sustainability.

There are various natural based flocculant has been explored in water and wastewater treatment applications. Guar gum, protein, cellulose and starch were among the studied natural flocculant [11,12]. Owing to the omnipresence, inexpensiveness, biorenewability, biodegradability and susceptibility to modification starch has been the most extensively given attention by researchers [12]. Starch is a carbohydrate polymer which represent in general formula as $(C_6H_{10}O_5)_n$. It is derived from various sources of food plant structures such as corn, wheat, oat, rice tubers, potato, sweet potato, tapioca, nuts, mung bean, green pea and sago [13]. Nevertheless, the shapes, sizes and morphologies of each plant are dependent on their botanical origins [14]. The natural starch consists of two major macromolecular polysaccharides which are amylose and amylopectin. Amylose is a slightly branched linear macromolecule of α -(1 \rightarrow 4)-D-glucopyranosyl units with a degree of polymerization (DP) of 1000-6,000, while amylopectin is a highly branched macromolecule consist of α -(1 \rightarrow 4)-D-glucopyranose units with a DP of 300,000-3,000,000 [13,14]. The ratio of amylose: amylopectin are varies

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for each starch origin. It is reported the ratio of amylose and amylopectin typically is 1:3 respectively [13,14]. Plus, there were also contain traces of lipid (0.01-0.80 w/w %), protein/enzyme (0.10-0.40 w/w %) and phosphate monoester (0.09-0.63 w/w %) [15,16].

Durian (*Durio zibethinus*) is a seasonal fruit tree grown in South East Asia and belongs to family of Bombacaceae [17]. Durian is normally being eaten fresh. Previous studies had shown that durian was very nutritious and had high fiber content [18]. However, only one-third of durian is eatable, whereas the seeds and the shell are usually thrown away. Unlike most of the existing botanical flocculants, the use of Durian Seed Strach (DSS) as a natural flocculant for landfill leachate treatment has not yet been investigated. As leachate are mostly toxic and portends detrimental psycho-physiological effects on human physical and mental health [19], there should be an effort to improve its treatment process. Therefore, this preliminary study reports the flocculant characteristics for landfill leachate treatment using starch natural based flocculant from *Durio Zibethinus* Seed.

MATERIALS & METHODS

Leachate characteristic:

A sample of leachate was obtained from Matang Landfill, situated in Kuala Sepetang, Taiping Perak. Leachate samples were collected manually by composite sampling method. All samples were kept in HDPE (High Density Polyethylene) containers with sealed caps. Samples were transported to the laboratory within 1 h and stored in a cold room at 4°C to minimise biological and chemical reactions prior to any treatability study. Before experiments, leachate samples were conditioned by putting them at room temperature for 2-3 h and homogenised by manual agitation. During the study, the leachate samples were characterised before and after each treatment. All the analytical procedures were performed according to the Standard Method of Water and Wastewater APHA (2005).

Starch extraction method:

Starch was obtained by using dry milling method. The durian seed were peeled to separate from its yellow colour aril that can be eaten and then the seed is washed with tap water as mention by Tongdang [20]. The durian seeds were cut into small cubes and dried for 24 hours in the open air. The durian seed were then grounded using heavy duty blender and then it is sieved until it passing the 75µm size. The sieve process using the 75µm sieve and become powder to use in the flocculation process.

Jar Test:

A conventional jar test apparatus (VELP-Scientifica, Model J LT6, Italy) with impellers equipped with 2.5 cm×7.5 cm rectangular blades was used for coagulation-flocculation processes. Leachate samples were removed from the cold room, conditioned and agitated before transferred into corresponding jar test beakers in appropriate volume. The pH values of samples were adjusted to the desired values by addition of appropriate amounts of NaOH (3M) and H₂SO₄ (3M) solutions. The jar test consisted of three subsequent stages: (1) rapid mixing stage, (2) slow mixing stage and (3) final settling. During rapid mixing, the DSS were added into the beakers while the impellers maintained at the fast speed. After a certain rapid mixing period, the stirrers were set to a slower speed for another period of time. After that, the stirrers were stopped and the samples were left for final settling. Then, the samples were withdrawn using plastic syringe from 10 cm below the surface for the analytical determinations. Analyses were undertaken in triplicates. The removal analysis of the leachate was done by analyzing colour and turbidity removal using DR 2800 Spectrophotometer. Removal efficiency of leachate colour and turbidity was obtained using the following equation:

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

Where C_i and C_f are the initial and final concentrations of leachate, respectively.

Characterization of the DSS flocculant:

The surface morphology of DSS flocculant was characterized using Field emission scanning electron microscope (FESEM) ZEISS SUPRA 35VP. Samples were attached to an aluminium mount with carbon double-sided tape and sputter coated with Au/Pd by using Palaron SC 515 sputter coater to eliminate the electron charging and poor image resolution effects; It was operated at working distances down to 1 mm and extended accelerating voltage range of 30 kV-100 V. However, the sample of DSS-leachate flocculant was previously filtered by micro glass fibre filter (Grade 259) with pore size of 1.6 µm. While the functional group and composition analysis of DSS was done by using Fourier Transform Infrared (FTIR) Spectrometer Perkin-Elmer System 2000. The FTIR spectra were recorded in wavelength range of 550-4000 cm⁻¹ at 4 cm⁻¹ resolution using KBr pellets method.

RESULTS & DISCUSSIONS

Raw Leachate Characterisation:

Table 1: Matang Landfill Site raw leachate characteristics.

Parameter	Average	Standard*
pH	8.28	6.0-9.0
Residual conductivity ($\mu\text{S}/\text{cm}$)	880.94	-
Zeta potential (mV)(at original pH)	-28.77	-
Particle size, d (μm)	61.15	-
Turbidity (NTU)	220	-
Ammoniacal-nitrogen (mg/L)	840.00	5.0
Colour (PtCo)	4080	100 ADMI
Appearance (colour)	Brownish-black	-

*Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulations 2009

The leachate collected could be classified as alkaline since the pH was around 8.28. High pH value occurred due to the conversion of intermediate organic acids into methane (CH_4) and CO_2 by methanogenic bacteria during methane fermentation phase or so-called methanogenic stage of the landfill decomposition phase [21]. Ammoniacal-nitrogen concentration was around 840 mg/L. Ammoniacal-nitrogen was leached from the biodeterioration of protein and other nitrogen-containing organic matters. In nature, diminishing $\text{NH}_3\text{-N}$ concentration to safe level could take decades [22].

In addition, the sizes of leachate colloids appeared to be distributed with volume-weighted mean gyration diameter of 61.15 μm . The smallest size was 0.45 μm , while the biggest one could go up to 94.66 μm . Organic colloidal particles also accounted for the high values of turbidity (220 NTU), and colour (4080 PtCo). The leachate samples appeared to be brownish-black in colour with high colour value of 4080 PtCo due to the humid substances formed from organic material biodegradation [23]. Recalcitrant humid substances like humic acids and fulvic acids are soluble at higher pH and difficult to be treated. These substances could inhibit organic pollutant decomposition in acidic condition form carcinogens like trihalomethanes and halogenic acids when disinfected with chlorine [24].

Colour and Turbidity Removal:

For this preliminary study, DSS were only applied as primary flocculant in of leachate treatment. The effects of pH and flocculant dosage on leachate clarifying performances were evaluated based on the removal turbidity and colour. Figure 1 shows the respond of colour removal by DSS-leachate different pH and dosage values after 1 hour of settling time. The results demonstrated that the DSS-leachate flocculation performance was highly dependent on pH and dosage. At the lowest dosage of DSS (1000 mg/L) the removal was dominated by the condition of pH values. As the acidity of leachate increase the colour removal were increased. At this early stage, the pH condition was primary influence the condition of the treatment. However as the dosage of DSS flocculant increased the colour removal was gradually increased without mainly depending on acidity of the leachate condition. The results demonstrated that DSS flocculation performance was highly dependent on pH, but the responses tended to fluctuate within the pH. This abnormality was attributed to the intrinsic characteristics of DSS as natural polysaccharide, which might have rather inconsistent quality and relatively low uniformity compared to chemical flocculants. In this colour removal the optimum pH value is pH 6 with 34% of removal at 4000 mg/L flocculant dosage.

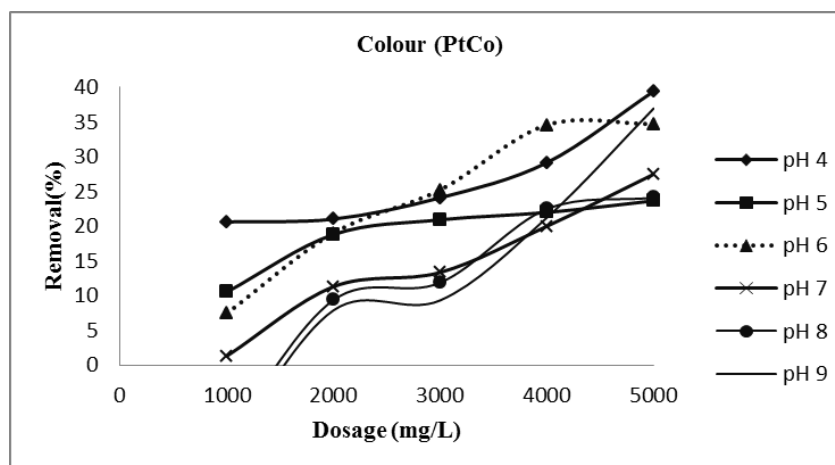


Fig. 1: The colour removal by DSS-leachate different pH and dosage.

Figure 2 shows the respond of turbidity removal by DSS-leachate at different pH and dosage values after 1 hour of settling time. The results demonstrated that the DSS-leachate flocculation performance was also highly dependent on pH and dosage. There was no removal recorded for all pH values at low dosage, the turbidity was increased as DSS introduced to the system. As the dosage increased to 4000 mg/L the turbidity removal was firstly occurred at pH 6 with the removal of 28%. While remaining pH values were only shows removal as the dosage increased to 5000 mg/L. The removal for pH 4,5,6,7 and 9 were 35.7%, 18.8%, 36.9%, 17.7% and 8.4% respectively. This turbidity removal pH 6 shows highest removal performance with 36.9% at 5000 mg/L flocculant dosage. These observations might contributed by the floc formation through the flocculation process. At the low dosage amount the floc have low strength to attract each other. However as the dosage was increased the floc has become stronger to agglomerate with the polluted particles and become larger to settle to the bottom.

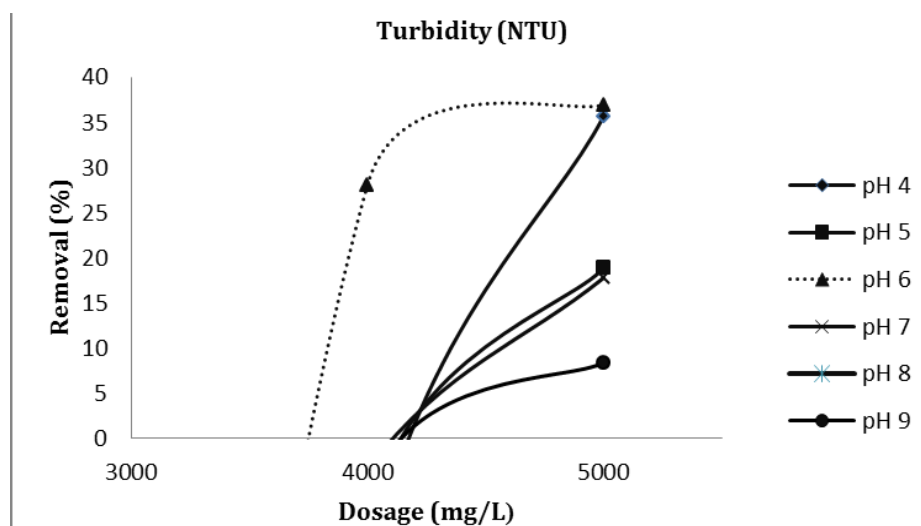


Fig. 2: The turbidity removal by DSS-leachate different pH and dosage.

Structural Morphology:

Figure 3 shows the morphology of extracted DSS flocculant before the treatment process. It is observed the shape and surface features of DSS were almost similar with the previous research. Its irregular oval and truncated ellipsoidal-granules was parallel with the previous research observations [25,26]. The truncated ends observed could be formed due to the starch compaction in starch producing organelle i.e. amyloplast over the maturation years [27].

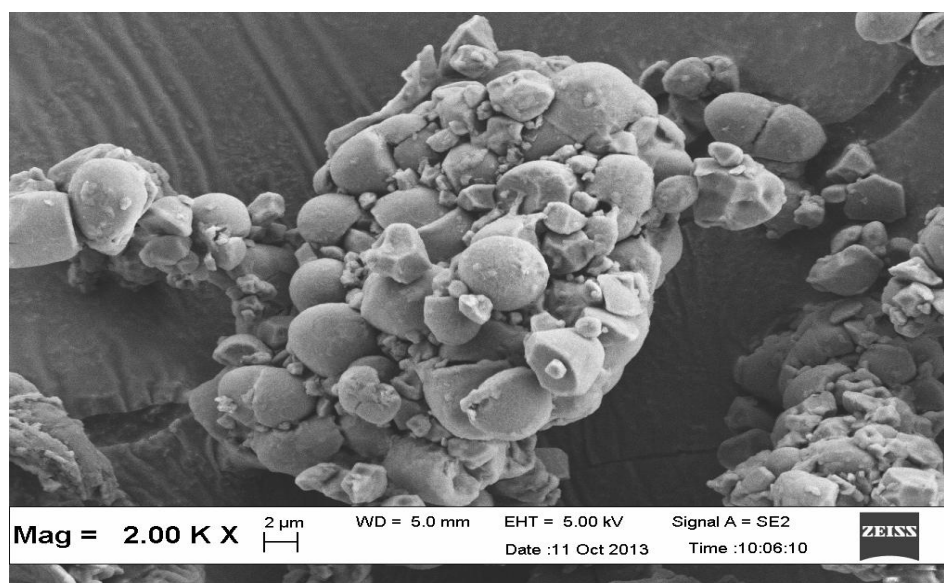


Fig. 3: The morphology of extracted DSS flocculant before the treatment process.

Figure 4 shows the surface features of DSS flocculant after the treatment process. Unlike to Figure 3, this DSS- leachate flocculant granule has been covered by different layer of particles on top of the starch surface. This wave-like layer was creases together with the starch and forming different granules which is not smooth and ends with radial pattern. Plus, a long thin particle which attached on top of the granules has strengthened the opinion that the DSS flocculant has been contaminated. Further analysis of the particles contamination was shown by the composition analysis in Figure 5.

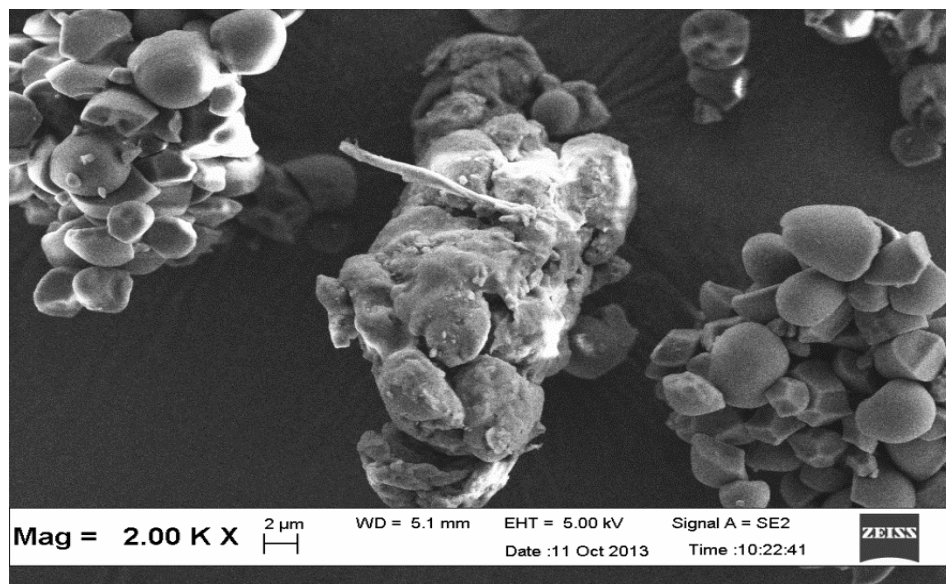


Fig. 4: The surface features of DSS flocculant after the treatment process.

Compositions Analysis:

Figure 5 shows the composition analysis of DSS flocculants using the FTIR spectrometer. The characteristics bands of DSS was slightly shifted after the treatment process .FTIR results verified that the agglomeration did consist of leachate contaminants like humic and fulvic acids (2918 and 1735 cm^{-1})[28,29,30], nitrate (1379 cm^{-1})[31,32] and P-H phosphate (1039 cm^{-1})[33] . These carboxyl, carbonyl and hydroxyl groups are weak cationic charge. Therefore, it is believe the removal performance of DSS flocculant might be promotes by adsorption of ion exchange and complexation of starch during the treatment process.

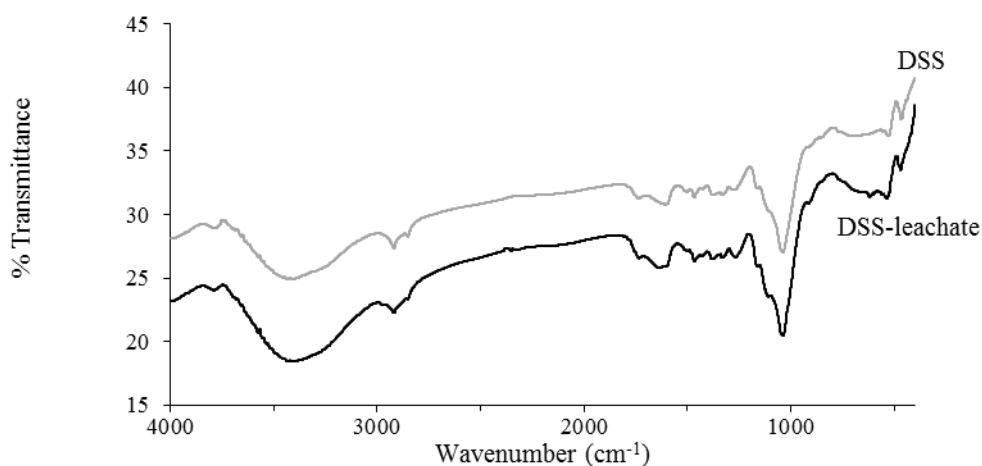


Fig. 5: The composition analysis of DSS flocculants using FTIR spectrometer.

Conclusions:

In this preliminary study DSS flocculant showed good potential in the flocculation process of landfill leachate treatment. It was noted that the appropriate pH and the flocculant dosage was needed for the removal of the leachate colour and turbidity. The optimum value of removal was recorded at pH 6 with the dosage of 4000 mg/L. The combination analysis using SEM and FTIR has revealed the potential of DSS flocculant. The SEM

images show the presence of contaminated particle layer on top of the DSS after the treatment process. While the Fourier transforms FTIR analysis confirmed the composition of the flocculants and verified that the agglomeration of leachate contaminants. Therefore, DSS flocculant could be a feasible selective flocculant in reducing the usage of inorganic polymer for semi-aerobic landfill leachate treatment. A further research should be taken in explore more interesting potentials of DSS flocculant.

ACKNOWLEDGEMENT

The financial support provided by RUI USM grant (1001/PAWAM/814166) is gratefully acknowledged.

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