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Comparative removal of suspended solids from landfill leachate by *Hibiscus rosa-sinensis* leaf extract and alum

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ABSTRACT

Hibiscus rosa-sinensis is a biodegradable material that has not yet been tested for its flocculating properties. The objective of this study was to examine the efficiency of coagulation–flocculation processes for the removal of suspended solids (SS) from landfill leachate using aluminium sulphate (alum) and *Hibiscus rosa-sinensis* leaves extract at different pH and dosages. The experiments confirmed the positive coagulation properties of the investigated natural coagulant. The extracts from *Hibiscus rosa-sinensis* leaves had an isoelectric point at pH 4 and acted as a bridging mechanism in removing 74% of SS (8,000 mg/L dosage at pH 6) which was comparable with 78% SS removal using 7,000 mg/L alum (at pH 6).

Keywords: Alum; Coagulation and flocculation; *Hibiscus rosa-sinensis*; Leachate treatment

1. Introduction

Commonly used metal coagulants are generally based on aluminium and iron and have been widely used in water treatments since the early twentieth century. However, the cost for importing aluminium sulphate (alum) and other required chemicals for conventional treatment is generally high and at times prohibitive, especially in developing countries [1]. The use of alum can affect the biogeochemical cycling of other elements on the planet through various physicochemical reactions such as hydrolysis, polymerization, flocculation, precipitation, adsorption and, in particular, the complex reactions of aquatic Al^{3+} taking place on the interfaces of minerals/water or in waters [2,3]. This leads to a possibility of several diseases such as Alzheimer's, Parkinson's and amyotrophic lateral sclerosis [4]. Natural coagulants are becoming the central focus of many researchers because of their abundant sources, low price, multiple functions and biodegradable properties [5]. In addition, natural coagulants have a wider optimum dosage range of flocculation for various colloidal suspensions [6]. Natural coagulants also fit well in the definition of sustainability and appropriateness for the developing countries [7].

Coagulation–flocculation is a relatively simple physicochemical technique commonly used in water and wastewater treatment [7,8]. The removal mechanism involved in coagulation–flocculation process primarily consists of charge neutralization of negatively charged colloids by cationic hydrolysis products followed by incorporation of impurities in an amorphous hydroxide precipitate through flocculation [8]. Currently, researchers tend to extract the natural coagulants in powder form after drying in oven before grinding it to the desired size. Afterwards, natural coagulants are kept in air tight container before making their solutions for further use.

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Most of these extracts are derived from the seeds, leaves, pieces of bark or sap and roots and fruits of trees and plants [9]. The use of leaf extracts as a coagulant has not been well reported in literature unlike its use as an adsorbent. Table 1 presents some of the natural coagulant extraction techniques as reported in literature. A method proposed by Šćiban et al. [10] has been widely used for the extraction purpose. The differences in the particular method used by each researcher are in terms of powder sizes, solvent, stirring, drying and filtration. The solvent can be an organic solvent (acetone, petroleum ether, chloroform or hexane) or solution of inorganic salts (KCl, NaCl, $CaCl_2$ or NH_4Cl).

A number of natural materials have been systematically evaluated for their coagulation properties. The species of *Plantago psyllium, Tamarindus indica, Trigonella foenum-graecum, Moringa oleifera* and *Hibiscus esculentus* have shown promising results with respect to wastewater treatment [11]. Sanghi et al. [6] reported that plants of genus *Cassia* possess medicinal values and are a good source of mucilage, flavonoids, anthraquinones and polysaccharides. Recent studies have shown interest in extracting mucilage from seeds or pads of plants in order to explore their coagulation efficiency. *Cactaceousopuntia* [5] and *Cassia javahikai* seed gum [6] are some examples of mucilage sources that have been tested alone and in conjunction with a synthetic coagulant to assess their coagulation efficiency in wastewater treatment. *Hibiscus rosa-sinensis* has been evaluated for its coagulation properties in this study for the first time in literature. *Hibiscus rosa-sinensis* belongs to the *Malvacea* family and is a glabrous shrub widely cultivated in the tropics as an ornamental plant since it is present in several forms with varying flower colours. In particular, another species of *Hibiscus* genus, namely, *Hibiscus sabdariffa* has been historically used and documented in literature for water purification [9].

To date, no reference in the literature has produced experimental data involving the use of *Hibiscus rosa-sinensis* as a coagulant for landfill leachate treatment. Starch or polysaccharide and protein contained in the mucilage are the closest explanation for the coagulation properties of the *Hibiscus rosa-sinensis* leaf extract. There is a possibility that other active components in the mucilage also possess coagulation properties. Thus, the objective of this study was to compare the efficiency of *Hibiscus rosa-sinensis* leaf extracts with alum for the removal of suspended solids (SS) from land fill leachate at different dosages and pH values. The effects of the isoelectric point (IEP) are also discussed.

2. Materials and methods

2.1. Preparation of leaf extracts

Rather than using methods listed in Table 1, this study provides a new extraction method that is much simpler and cost-effective. The collected fresh leaves were repeatedly washed with water to remove dust and soluble impurities. The X_1 g of the green leaves was then placed in a beaker containing Y_1 L of distilled water. The mixture was blended using a domestic blender operated at a high speed to extract the active ingredient of the *Hibiscus rosa-sinensis*. The suspension was then filtered using Whatman Grade No. 1 filter paper into a beaker. The leaf dregs were dried in an oven for 48 h at 105°C; meanwhile, the filtrate stock solutions were stirred using a magnetic stirrer for 30 min at room temperature. The dry leaf dregs were weighed and the amount was defined as X_2. The pure concentration of the dissolved filtrate stock solution, HBaqs, M_1 was calculated as follows:

$$M_1 = (X_1 \times \mathrm{DM}/100 - X_2)/Y_1 \quad (1)$$

where DM refers to dry matter of green leaves in percentage (%).

HBaqs filtered stock solution was used for the coagulation experiments either immediately or after storing at 4°C.

2.2. Sampling of the leachate influent

Leachate samples were collected from Pulau Burung Landfill Site (PBLS) which is located near shoreline about 50 m from straits of Malacca within Byram Forest Reserve at 5°24 N and 00°24 E in Penang, Malaysia [12]. The site receives approximately 1,800 tons of solid waste daily from Penang Island [13]. The PBLS was developed into a semi-aerobic system employing a controlled tipping technique and leachate recirculation system in March 2001 [14]. The total operational area of the landfill is 33 ha and it is equipped with a leachate collection pond. This site has a natural marine clay liner and a semi-aerobic system, making it one of the only three such sites in Malaysia. The samples were kept in a container and placed in a cool room at 4°C to restrain the micro-organism activities. However, the samples were left for 2 h at ambient temperature before each experiment. This landfill produces a dark black-green coloured leachate that can be classified as stable due to high concentrations of chemical oxygen demand (COD) and ammoniacal nitrogen and a low BOD_5/COD ratio [15,16]. The samplings of the leachate influ-

Table 1
Extraction techniques of natural coagulants

Source of natural coagulant	Extraction process	Findings	References
Tamarindusindica	Seeds were thoroughly washed and soaked in distilled water overnight. The mucilaginous extract was filtered through muslin cloth. It was precipitated by adding three parts of iso-propanol to one part of the aqueous extract. The residue was then washed with acetone two or three times to remove impurities and finally dried by keeping it in an oven at 40˚C for 24 h	At neutral pH, the maximum removal was 60% for golden yellow after 2 h and 25% for direct fast scarlet after 1 h. The optimum mucilage dosage was 10 and 15 mg/L, respectively	[26]
Opuntiaficus-indica	Pods were cut and external layer was removed manually. Internal fraction was milled in a domestic blender. About 200 g of the resultant juice was put into a beaker and it was filled up to 1 L with distilled water. Then it was kept at 60˚C for 24 h. Later, the mixture was filtered and concentrated by vacuum evaporation to one-third of the initial volume. Then, it was precipitated with ethanol twice, in order to achieve clean impurities-free mucilage. The resultant mix of ethanol and mucilage was dried in a heater at 60˚C for 12 h. Final product presents a green, crystal aspect	Approximately, 5% of sodium lauryl sulphate was removed by 100 mg/L *Opuntiaficus-indica mucilage* at neutral pH	[7]
Chestnut (*Aesculushyppocastanum* and *Castaneasative*) Acorn (*Quercusrobur*, *Quercuscerris* and *Quercus rubra*)	The seeds were ground to fine powder using laboratory mil (separately for each chestnut and acorn). All grounded materials were sieved through 0.4 mm sieve and the fraction with particle size less than 0.4 mm was used. About 50 g of prepared powder was suspended in 1 L of distilled water or NaCl solution and the suspension was stirred using a magnetic stirrer for 10 min to extract the coagulation active components. The suspension was then filtered through a rugged filter paper and kept in refrigerator for further used	Coagulation capabilities to remove turbidities from synthetic water depend on pH. Highest CA derived from *Ae. Hyppocastanum* and *Q. robur* were 70 and 80%, respectively, at low dosage, 0.5 ml/L	[10]
Mallow (*Malvasylvestris*) Okra (*Hibiscus esculentus*)	The seedpods of the plant were initially washed thoroughly with water to remove any impurities, dried at 110˚C for 6 h and then ground with a microhammer cutter mill and sieved to a 32 mesh (500 μm) particle size (the solvent used to prepare stock solution was not mentioned)	With the help of 0.025 Mm Al^{3+} addition (no pH controls), 62.5 mg/L mallow and 2.5 mg/L okra successfully removes 63 and 72% turbidity, respectively in the biologically treated effluents	[11]

ent were taken from the active detention pond of the PBLS, 14 times, from 1 October 2008 to 30 November 2009. All experiments were undertaken in triplicate according to the procedures detailed in the Standard Method for Examination of Water and Wastewater [17]. The pH was measured using a portable pH meter (Hach, sens ion 1, USA). SS were determined by using APHA Method No. 2540D. The NH_3–N was determined using nesslerization method (4500-NH_3). COD was determined using colorimetric method (5220-D). Turbidity was measured using 2020 turbidimeter (LaMotte). Colour measurements were reported as true colour (filtered using 0.45 μm filter paper) assayed at 455 nm using DR 2000 HACH spectrophotometer. Method No. 2120C reports colour in Platinum–cobalt (PtCo), the unit of colour being produced by 1 mg platinum/L in the form of the chloroplatinate ion. The effect of filtration on colour removal was corrected by means of a control sample. The characteristics of the leachate are given in Table 2.

2.3. Characterization of HBaqs

2.3.1. Isoelectric point

The surface charge of HBaqs was evaluated by determining the electrophoretic mobility of suspended particles with Malvern Zetasizer Nano ZS. Measurements were taken at 25°C with distilled water as dispersal medium. IEP for HBaqs was determined by the titration of 10 mL HBaqs with 0.25 M HCl or 0.25 M NaOH from pH 3 to 12 or from pH 12 to 3, respectively depending on the initial zeta potential of the titration.

2.3.2. Fourier transform infrared spectra

Fourier transform infrared (FTIR) Perkin Elmer was used to investigate the structure of HBaqs and to analyze their functional groups. The leaves of *Hibiscus rosa-sinensis* in powder form were used because the solution would only result in water spectra.

2.4. Jar test

A 1,000 mL beaker was filled with 500 mL of the leachate for each test run. The pH value of each jar test was adjusted to the desired value by using NaOH and H_2SO_4 within the range of 2–12. HBaqs dosages varied from 500 to 10,000 mg/L. The leachate sample was agitated at 160 rpm for 2 min (rapid mixing), after which the speed of mixing was reduced to 30 rpm and kept constant for 20 min. After the settling period of 30 min, the supernatant was withdrawn from the beaker using a plastic syringe from the point approximately 2 cm below the liquid level for analysis [18,19]. The entire experiment was repeated under the same conditions using the alum solution as a coagulant.

The removal efficiency of SS was calculated by using Eq. (2) as follows:

$$\text{Removal } (\%) = [(C_i - C_f)/C_i] \times 100 \tag{2}$$

where C_i and C_f are the initial and final concentrations of the sample, respectively.

The coagulation activity (CA) of the coagulants was analyzed using Eq. (3) to evaluate whether the removal of parameter tested was solely caused by the coagulant or by chemical precipitation, especially at high pH.

$$\text{Coagulation activity} = (RT_B - RT_S)/RT_B \tag{3}$$

where RT_S refers to the residual parameter measured after settling and RT_B is the residual parameter measured after settling with no coagulant (as a blank) [10].

Table 2
Characteristics of raw leachate from PBLS

Parameter	Units	Minimum	Maximum	Average[a]	Standard[b]
pH	–	8.24	8.82	8.65	6.0–9.0
SS	mg/L	165	211	189	50
NH_3-N	mg/L	1,840	2,465	2,125	5
COD	mg/L	2,873	3,360	3,170	400
Turbidity	FAU	275	325	303	–
Colour	PtCo	4,780	5,120	4,950	100

[a] Average of 14 samples taken from October 2008 till November 2009.

[b] The standard requirements of the Environmental Quality Act 1974 and the limits set down by the Environmental Quality (Control of Pollution from Solid Waste Transfer Station and Landfill) Regulation 2009, Malaysia.

3. Results and discussion

3.1. Characteristics of HBaqs

Coagulant characterization was carried out to provide basic understanding of the chemistry of the coagulant and its effectiveness in removing both particulate and organic materials. Thus, determining the following important individual properties of HBaqs was essential.

3.2. Isoelectric point

The zeta potential of HBaqs at various pH values is illustrated in Fig. 1. The surface charge of HBaqs was absolutely neutral at pH 4 while it was positively charged at pH below 4 and negatively charged at pH above 4. Hence, HBaqs was an anionic coagulant when coagulation process was run at pH above 4 and a cationic coagulant at pH below 4. It is believed that the aggregation of the constituents was mainly governed by the bridging mechanism which is in agreement with the study of Zhang et al. [20].

3.3. Functional group analysis of HBaqs using FTIR

FTIR spectra analysis is an important step in polymer characterization. FTIR has been used extensively for the characterization of polymer materials in polymer chemistry. To determine the main functional groups of HBaqs, FTIR spectra analysis was carried out based on the previous studies [21–23]. The results of the analysis are presented in Fig. 2 and Table 3.

Since the FTIR spectra of HBaqs were not determined quantitatively, the transmittance intensity could not be used to quantify the measured functional groups. However, Li et al. [22] used a Gaussian curve fit to calculate the area ratios of each peak to all peaks of the same curve, with the assumption that such ratios represent the relative quantities of the peaks. Zhang et al. [20] claimed that the FTIR spectrum of TJ-F1 contained carboxyl, hydroxyl and amino or amide ($-NH_2$) groups as well as hydrogen bonds which are the preferred groups for the flocculation process. The chemical structure of the polymer might possess two common features: (1) the structure consists of a hydrocarbon backbone forming a long chain; and (2) the long chain carries active groups that could be anions, cations or non-ionic entities [24].

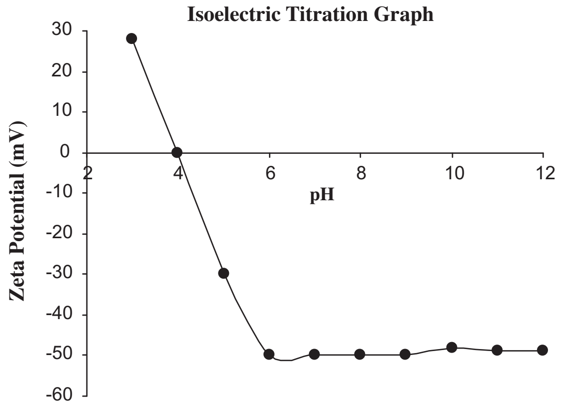

Fig. 1. Effect of pH on the zeta potential values of HBaqs.

As mentioned earlier, the main mechanism that governed the CA by HBaqs was bridging. Local coulombic forces between the divalent metal cations (Ca^{2+}, Mg^{2+}, etc.) at the particle surface of the leachate and the dissociated carboxyl ($-C{=}O^-$) groups on the HBaqs chain induced the CA of HBaqs. The carboxyl groups could serve as ion bridges or binding sites for the divalent cations [22–24]. Additionally, –H bonding between monovalent cations (K^+ and Na^+), the amide ($-NH_2$) and the hydroxyl (–OH) groups also contributed to the removal of SS. Indeed, –H bonding is mostly seen when anionic polymers are used in the coagulation–flocculation system. However, this type of bonding has been suspected to occur when using a long polymer chain with a high molecular weight [25,26].

3.4. Optimum pH

The pH is the most significant variable in the coagulation–flocculation process for water and wastewater treatments [10,27,28]. Controlling the pH is critical because coagulation occurs within a specific range for each coagulant [29]. The optimum pH values found could be related to the IEP of the mixture of coagulant and wastewater and not the coagulant alone.

Currently, the measurement of the zeta potential is used extensively to predict coagulant requirement and optimum pH level. A couple of studies [20,30] examined the effects of the zeta potential at varying pH (IEP) and dosages in a heterogeneous suspension. Their studies were mainly focused on a heterogeneous suspension that consisted of coagulants and colloidal particles (water sample) with or without different electrolyte backgrounds after the coagulation process.

3.4.1. Determination of the optimum pH for alum

Fig. 3 demonstrates the percentage reduction of SS at various pH values. According to Fig. 3, an optimum pH for alum at 4,000 mg/L dosage was 6. The results revealed that a variation in pH noticeably affected the SS deduction with a removal of 77% at an

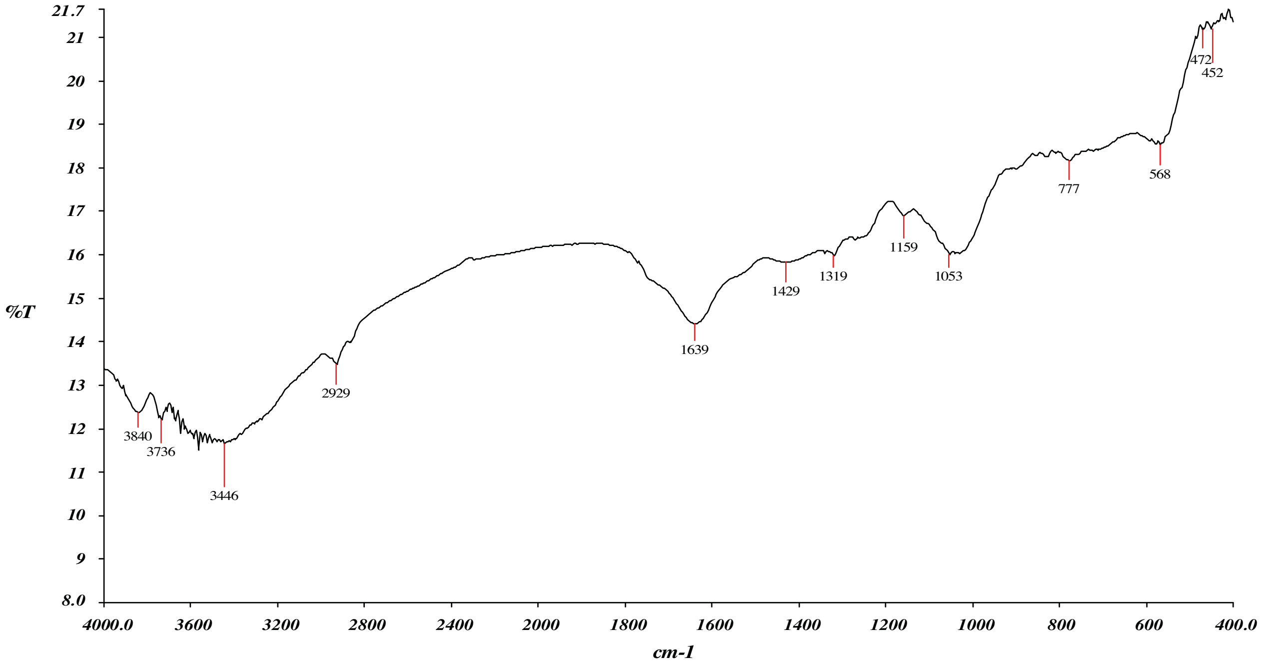

Fig. 2. The FTIR spectra of HBaqs.

Table 3
Main functional groups of HBaqs from FTIR spectrum analysis

Vibration type	Wavenumber (cm^{-1})
Hydroxyl and amines (O–H and N–H stretching)	3,446
Alkanes (C–H stretching)	2,929
Carboxyl (C=O stretching)	1,429–1,639
Methoxyl (C–O stretching of carboxylic acids)	1,319
Methoxyl (C–O stretching of alcoholic groups)	1,053–1,159

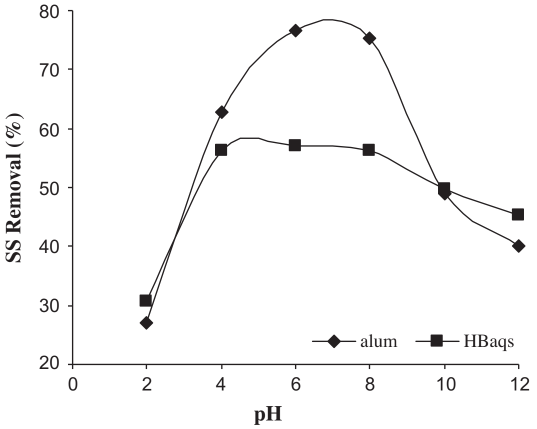

Fig. 3. Percentage removal of SS by 4,000 mg/L alum and 4,000 mg/L HBaqs at different pH values (Initial SS concentration = 189 mg/L).

optimum pH. The optimal pH value obtained is not in accordance with the results obtained by several authors [19,31,32], where optimal pH was 7. Nevertheless, several other researchers [28–30] claimed that the optimum pH for alum (for various wastewater types) was approximately from 5 to 6.

This narrow range occurred due to the variance in wastewater composition, the types of parameters used for measurement and the initial alum dosage/concentration (Table 4). An experiment on the effect of dosage was performed by Zouboulis et al. [30] for the humic acid removal from biologically pre-treated landfill leachate. The report indicated that the optimal pH between 5.5 and 6 was more evident for low dosages of alum (20 mg/L). The effect of pH was not so noticeable when the coagulant dosage was increased up to 50 mg/L. Thus, an efficient removal was obtained for a greater pH range. In addition, Huang and Shiu [33] studied the various alum dosages and pH values for the removal of humic acids by coagulation. Their study showed that an 80% removal of humic acid could be achieved in two regions. The regions consisted of a low dosage of alum at pH 5 and a high dosage of alum at pH above 6. Hence, 4,000 mg/L alum with an optimum pH at 6 was reasonable and acceptable when compared with the results obtained by others [31,32].

Table 4
Comparison of optimal dosage and pH of alum in a variety of wastewaters

Water sample	Parameter tested	Optimum pH	Optimum dose (mg/L)	Findings	Reference
Leachate	SS	6	7,000	Achieved 78% SS removal	This Study
Leachate	COD, turbidity, colour and TSS	7	9,400	COD, turbidity, colour and TSS removal were 61, 93, 88 and 92%, respectively	[32]
Leachate	COD and colour	7	9,000	93 and 46% for colour and COD removal, respectively	[31]
Cork processing industry wastewater (three water samples with different organic matter concentration was labelled as I, II, and III)	COD, TP and A	5	66 for Water I, 100 for Water II, and166 for Water III	Yielded similar values of the removal of COD (approximately 55%), TP (approximately 80%) and A (approximately 85%) in the three types of water	[28]
Slaughterhouse wastewater	COD, BOD_5 and TSS	5	600	COD, BOD_5 and TSS removal were 85, 88 and 95%, respectively	[29]
Humic acids	Humic acids	5.5−6	50	95% humic acids removal	[30]
Leachate	COD	7	500	COD removal capacities up to 56% for the partially stabilized leachate	[19]

TSS: total suspended solids; TP: polyphenols; A: aromatic compound.

Notably, the removal efficiency of SS using alum increased from pH 4 to 6 (Fig. 3). These results are vastly similar with a study regarding the influence of pH on coagulation using alum by conventional and electrochemical dosing [27]. Only ionic species has to be considered in explaining the coagulation results outside the range of pH 5 to 6, where the influence of aluminium hydroxide is present. This work is supported by Aguilar et al. [29], who claimed that aluminium hydroxide species grow best at pH values between 4 and 6 (where the positively charged species are predominant), whereas the range 5.5–7.5 is best for the $Al(OH)_3$ precipitate.

3.4.2. Determination of optimum pH for HBaqs

The removal of SS as a function of pH by HBaqs is also shown in Fig. 3. The overall maximum percentage removal for HBaqs in the leachate sample was observed at pH 4 to 8 (56–57%), although a considerable percentage removal was also seen at pH 10 (50%). The effect of pH on the removal of SS by the application of HBaqs was significant throughout the experiments and different SS removal rates were achieved at different pH levels. These results are not completely in accordance with the results obtained by Sanghi et al. [6], who stated that natural polysaccharides or polymers are usually inert to pH changes. Therefore, the CA caused by pH variation is believed to be due to the effect of pH on the constituents of the leachate sample. At neutral pH, the formation of hydrogen bonding between neighbouring hydroxyls and surface adsorbed water results in disruption of surface hydroxyls which increases the percentage removal [34]. On the other hand, at acidic pH (4), the metallic ions present in the effluent get oxidized which results in the aggregation of particles [35].

Thus, to prove that the removal of SS was solely due to the natural precipitation by HBaqs, jar tests at varying dosages and pH of 6 and 10 were carried out. Blank samples (without addition of coagulant) were used to define the CA of pH alone, whether by HCl at low pH or by NaOH at high pH. A pH of 10 was chosen because this pH was in agreement with the previous findings that reported higher pH values as being optimal for other natural coagulants, such as from chestnut and acorn [10] and *Moringaoleifera* [36].

3.5. Optimum dosage

The coagulant dosage is an important factor in the coagulation process. The CA highly depends on the optimum relationship between the coagulant dosage and bivalent cations in water [36]. The optimum dosage of a coagulant is defined as the value above which there is no significant increase in the removal efficiency with the further addition of a coagulant [29].

3.5.1. *Determination of the optimum dosage for alum*

Fig. 4 illustrates the CA and the removal of SS using alum and HBaqs. The optimum dosage for alum was approximately 7,000 mg/L with 78% removal. The results generally depict that the overall removal efficiency of the SS increased with an increase in alum dosage until reaching an optimal value. This could be attributed to the restabilization of colloidal particulates when coagulants are used at dosages in excess of the optimum value [18]. However, coagulant overdosing leads to the restabilization of the suspension and therefore to the release of heavy metals into the treated water.

The predominant removal mechanisms at a low dose of alum (below 7,000 mg/L) were adsorption and charge neutralization. The charge neutralization occurred when the zeta potential of particles was close to zero (at 7,000 mg/L) and the sweep coagulation took place at a sufficiently high alum dosage (10,000 mg/L with 8% SS removal), causing the precipitation of the amorphous metal hydroxide.

3.5.2. *Determination of optimum dosage for HBaqs*

There was a distinctive difference between coagulation by HBaqs at pH 6 and 10 (Fig. 4). A higher removal of SS (74% removal with 8,000 mg/L HBaqs at pH 6 and 56% removal with 2,000 mg/L HBaqs at pH 10) and a broadened dosage range (2000 to 10,000 mg/L at pH 6 and 2,000 to 4,000 mg/L at pH 10) was achieved at pH 6 (Fig. 4).

The overdosing of HBaqs at pH 10 led to a sudden decrease in SS removal, whereas the removal of SS reached positive increased CA at pH 6. This overdosing resulted in the saturation of the polymer bridges and caused restabilization of the destabilized particles because of an insufficient number of free particles to form more interparticle bridges. The adsorbed layers might also have caused steric repulsion [11,25].

The obtained result indicated that the removal of SS by HBaqs was comparable with alum at dosage of approximately 4,000–6,000 mg/L at pH 6, whereby the removal of 70–77% for alum and 66–69% for HBaqs was observed. Moreover, the CA for alum was 0.66 at 7,000 mg/L dosage, whereas it was 0.54 (pH 6) and −1.36 (pH 10) for HBaqs (Fig. 4).

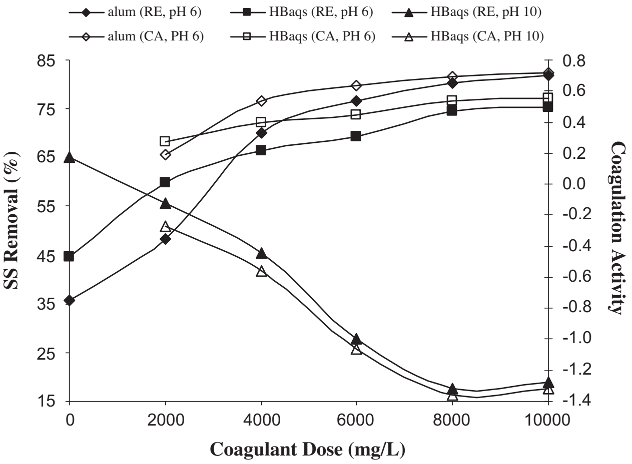

Fig. 4. Plot of coagulant dose for alum and HBaqs, where RE stands for percentage removal of SS, while CA indicates coagulation activity by coagulants without being influenced by pH factor (Initial SS concentration = 189 mg/L).

Notably, a higher removal of SS (65%) was achieved at pH 10, even without the addition of coagulants. This suggests that the removal rate at pH 10 was higher even without the presence of coagulants which was a result of the influence of NaOH. This also indicates that NaOH (alkaline) alone could have acted as a coagulant simultaneously with other factors such as the IEP and the charged particles carried by a natural organic matter in the leachate sample. In this case, caustic soda composed mainly of NaOH was used as an alternative in order to produce the hydroxyl ions needed to raise the pH to levels at which precipitation of divalent metal ions (Ca^{2+} and Mg^{2+}) would be favourable in indulging a CA.

4. Conclusions

Apart from having a wider effective dosage range (2,000–8,000 mg/L) of flocculation, high removal rates of SS of 60–74% and positive coagulation activities indulged by HBaqs at an optimum pH 6, proved that *Hibiscus rosa-sinensis* deserves further exploration as a coagulant in landfill leachate treatment. Moreover, the CA and the removal of SS by 8,000 mg/L HBaqs (CA, 0.59; removal, 74%) were comparable with 7,000 mg/L alum (CA, 0.66 and removal, 78%).

References

[1] R. Sanghi, B. Bhatttacharya, V. Singh, Seed gum polysaccharides and their grafted co-polymers for the effective coagulation of textile dye solutions, React. Funct. Polym. 67 (2007) 495–502.

[2] E. Gauthier, I. Fortier, F. Courchesne, P. Pepin, J. Mortimer, D. Gauvreau, Aluminum forms in drinking water and risk of Alzhemier's disease, Environ. Res. Sect. A84 (2000) 234–246.

[3] W.J. Lukiw, N.G. Bazan, Neuroinflammatory signaling upregulation in Alzheimer disease, Neurochem. Res. 25 (2000) 1173–1184.

[4] D.R. Parker, P.M. Bertsch, Identification and quantification of the "Al13" tridecameric aluminum polycation using ferron, Environ. Sci. Technol. 26 (1992) 908–914.

[5] J. Zhang, F. Zhang, Y. Luo, H. Yang, A preliminary study on cactus as coagulant in water treatment, Process Biochem. 41 (2006) 730–733.

[6] R. Sanghi, B. Bhatttacharya, V. Singh, Use of Cassia javahikai seed gum and gum-g-polyacrylamide as coagulant aid for the decolorization of textile dye solution, Bioresour. Technol. 97 (2006) 1259–1264.

[7] J. Beltrán-Heredia, J. Sánchez-Martín, Removal of sodium lauryl sulphate by coagulation/flocculation with *Moringaoleifera* seed extract, J. Hazard. Mater. 164 (2009) 713–719.

[8] S. Ghafari, H.A. Aziz, M.H. Isa, A.K. Zinatizadeh, Application of response surface methodology (RSM) to optimize coagulation-flocculation treatment of leachate using poly-aluminum chloride (PAC) and alum, J. Hazard. Mater. 163 (2009) 650–656.

[9] M. Pritchard, T. Mkandawire, A. Edmondson, J.G. O'neill, G. Kululanga, Potential of using plant extracts for purification of shallow well water in Malawi, Phys. Chem. Earth 34 (2009) 799–805.

[10] M. Šćiban, M. Klašnja, M. Antov, B. Škrbić, Removal of water turbidity by natural coagulants obtained from chestnut and acorn, Bioresour. Technol. 100 (2009) 6639–6643.

[11] K. Anastasakis, D. Kalderis, E. Diamadopoulos, Flocculation behavior of mallow and okra mucilage in treating wastewater, Desalination 249 (2009) 786–791.

[12] M. Umar, H.A. Aziz, M.S. Yusoff, Variability of parameters involved in leachate pollution index and determination of LPI from four landfills in Malaysia, Int. J. Chem. Eng. (2010). doi: 10.1155/2010/747953 (Article ID 747953)

[13] M.J.K. Bashir, M.H. Isa, S.R.M. Kutty, Z.B. Awang, H.A. Aziz, S. Mohajeri, I.H. Farooqi, Landfill leachate treatment by electrochemical oxidation, Waste Manage. 29 (2009) 2534–2541.

[14] A. Omran, A.O. El-Amrouni, L.K. Suliman, A.H. Pakir, M. Ramli, H.A. Aziz, Solid waste management practices in Penang state: A review of current practices and the way forward, Environ. Eng. Manage. J. 8 (2009) 97–106.

[15] N. Aghamohammadi, H.A. Aziz, M.H. Isa, A.A. Zinatizadeh, Powdered activated carbon augmented activated sludge process for treatment of semi-aerobic landfill leachate using response surface methodology, Bioresour. Technol. 98 (2007) 3570–3578.

[16] S.Q. Aziz, H.A. Aziz, M.S. Yusoff, M.J.K. Bashir, M. Umar, Leachate characterization in semi-aerobic and aneaerobic sanitary landfills: A comparative study, J. Environ. Manage. 91 (2010) 2608–2614.

[17] APHA, Standard methods for the examination of water and wastewater, 21st ed., American Public Health Association, Washington, DC, 2005.

[18] A.H. Aziz, S. Alias, M.N. Adlan, F.A.H. Asaari, M.S. Zahari, Colour removal from landfill leachate by coagulation and flocculation processes, Bioresour. Technol. 98 (2007) 218–220.

[19] A.A. Tatsi, A.I. Zouboulis, K.A. Matis, P. Samaras, Coagulation–flocculation pretreatment of sanitary landfill leachates, Chemosphere 53 (2003) 737–744.

[20] Z. Zhang, S. Xia, J. Zhao, J. Zhang, Characterization and flocculation mechanism of high efficiency microbial flocculant TJ-F1 from *Proteus mirabilis*, Coll. Surf. B: Biointerfaces 75 (2010) 247–251.

[21] M.A.K.M. Hanafiah, W.S.W Ngah, H. Zakaria, S.C. Ibrahim, Batch study of liquid-phase adsorption of lead ions using Lalang (*Imperata cylindrical*) leaf powder, J. Biol. Sci. 7 (2007) 222–230.

[22] W.W. Li, W.Z. Zhou, Y.Z. Zhang, J. Wang, X.B. Zhu, Flocculation behavior and mechanism of an exopolysaccharide from the deep-sea psychrophilic bacterium *Pseudoalteromonas* sp. sm9913, Bioresour. Technol. 99 (2008) 6893–6899.

[23] P. Prasertsan, W. Dermlim, H. Doelle, J.F. Kennedy, Screening, characterization and flocculating property of carbohydrate polymer from newly isolated *Enterobacter cloacae* wd7, Carbohydr. Polym. 66 (2006) 289–297.

[24] B. Yarar, Evaluation of flocculation and filtration procedures applied to WSRC sludge. Report No. WSRC-TR-2001-00213, Colorado School of Mines, USA, 2001, pp. 1–34.

[25] B. Ersoy, Effect of pH and polymer charge density on settling rate and turbidity of natural stone suspensions, Int. J. Miner. Process 75 (2005) 207–216.

[26] A. Mishra, M. Bajpai, The flocculation performance of Tamarindus mucilage in relation to removal of vat and direct dyes, Bioresour. Technol. 97 (2006) 1055–1059.

[27] P. Cañizares, C. Jiménez, F. Martínez, M.A. Rodrigo, C. Sáez, The pH as a key parameter in the choice between coagulation and electrocoagulation for the treatment of wastewater, J. Hazard. Mater. 163 (2009) 158–164.

[28] J.R. Domínguez, T. González, M.H. García, F.S. Lavado, J.B. De-heredia, Aluminium sulfate as coagulant for highly polluted cork processing wastewaters: Removal of organic matter, J. Hazard. Mater. 148 (2007) 15–21.

[29] M.A. Aguilar, J. Saez, M. Llorens, A. Soler, J.F. Ortuno, V. Meseguer, A. Fuentes, Improvement of coagulation–flocculation process using anionic polyacrylamide as coagulant aid, Chemosphere 58 (2005) 47–56.

[30] A.I. Zouboulis, X.L. Chai, I.A. Katsoyiannis, The application of the bioflocculant for the removal of humic acids from stabilized landfill leachates, J. Environ. Manage. 70 (2004) 35–41.

[31] Z. Daud, H.A. Aziz, M.N. Adlan, Y.T. Hung, Application of combined filtration and coagulation for semi-aerobic leachate treatment, Int. J. Environ. Waste Manage. 4 (2009) 457–469.

[32] S. Ghafari, H.A. Aziz, M.J.K. Bashir, The use poly-aluminum chloride and alum for the treatment of partially stabilized leachate: A comparative study, Desalination 257 (2010) 110–116.

[33] C. Huang, H. Shiu, Interaction between alum and organics in coagulation, Colloids Surf. A: Physicochem. Eng. Aspects 113 (1996) 155–163.

[34] I. Ayumi, U. Teruyuki, A. Jiro, T. Toshiyoki, M. Koji, Removal of heavy metals from anaerobically digested sewage sludge by a new chemical method using ferric sulfate, Water Res. 34 (2000) 751–758.

[35] M.B. Hocking, K.A. Klichuk, S. Lowen, Polymeric flocculants and flocculation, J. Macromol. Sci. Rev. Macromol. Chem. Phys. C39 (1999) 177–203.

[36] T. Okuda, A.U. Baes, W. Nishijima, M. Okada, Isoalation and characterization of coagulant extracted from *Moringaoleifera* seed by salt solution, Water Res. 35 (2010) 405–410.