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Application of a pre-hydrolyzed iron coagulant on partially stabilized leachate

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ABSTRACT

Leachate is a liquid produced from the biodegradation of solid waste in landfill and is normally referred as highly polluted wastewater. Various treatment methods are available and it is highly depended on the characteristics of the leachate. One of the common and simplest methods is using coagulation process. The application of pre-hydrolyzed coagulants in coagulation process is well known for water and wastewater treatment. However, information on the application of pre-hydrolyzed coagulants, especially pre-hydrolyzed iron (PHI), in partially stabilized leachate treatments is not well documented. This study examined the application of PHI on the partially stabilized leachate by determining the optimum basicity ratio, pH, and dose through the removals percentage of suspended solid (SS), color, and chemical oxygen demand (COD). Laboratory jar tests revealed that the optimum PHI basicity ratio, pH, and dosage were 0.1, 5, and 0.2 g/L Fe, respectively. The optimum removals of SS, color, and COD were 99, 95, and 58% respectively. The results indicated that PHI could be potentially useful coagulant for removing SS, color, and COD from partially stabilized landfill leachate.

Keywords: Coagulant; Pre-hydrolyzed iron; Leachate; Basicity ratio

1. Introduction

The most widely used method of solid waste disposal is by landfilling due to its simplicity and economical operation. In landfill operation, solid waste will be disposed on the upper layer and finally covered by soil. The drawback from landfill operation is the production of leachate. Leachate can be defined as heavily contaminated liquid as it contains high organic, inorganic, heavy metal, and other hazardous substances. The climate, age, precipitation, volume, and type of waste affect the amount and type of leachate produced [1]. Direct discharge of leachate into water bodies without treatment could lead into serious environmental problem. Leachate quality will be varied over a period of time and it can be categorized as fresh or unmatured leachate, stabilized or matured leachate, and partially stabilized or partially matured

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leachate [2–4]. The type of leachate will influence on the treatment method.

In general, the treatments for landfill leachates often involved a combination of various techniques (physical, chemical, and biological) depending on the age of landfill [2,5,6]. The age of landfill reflects the biodegradability of its leachates. Usually, the BOD/chemical oxygen demand (COD) ratio is used to indicate the biodegradability of leachates. The BOD/COD ratio for stabilize leachates is less than 0.1. According to Aziz et al. [7], a low BOD/COD ratio indicates that biological methods are not suitable, and that the best option is to use a physical–chemical method such as coagulation, adsorption, and filtration.

Coagulation has been applied in various types of water and wastewater treatment. Coagulation is the process of adding a coagulant or coagulants to a liquid to destabilize suspended matter, colloids, and other substances and transform them into larger particles so that it can be easily removed. The whole process reduces turbidity and dissolves the chemical species in the liquid [8]. Qian and Graham [9] stated that, in coagulation the following mechanisms will occur: (1) Formation of coagulant; (2) Destabilization of colloid/particle; and (3) Aggregation of particle. However, the performance of coagulation depends on the type of coagulant used and the characteristics of the liquid that needs to be treated.

Coagulants are divided into natural (i.e.: chitosan, hibiscus leaf, and moringaoleifera) and chemical (i.e.: alum, ferric chloride, aluminum sulfate, ferric sulfate, and PAC) [10–14]. Different coagulants yield different degrees of destabilization. The higher the valence of the counter-ion, the more the destabilizing effect and the less dosage required. In the early usage of chemical coagulants, monomeric coagulants (alum and iron salts) were normally used for water and wastewater treatment. Compared to alum, iron salt does not possess serious health effects on humans and living organisms. The growing interest on the use of iron coagulants is due to less hazardous residual compared to aluminum [15,16]. Furthermore, iron salt is more water soluble and has a wider pH range compared to alum [17].

However, according to Jiang and Wang [18] and Lei et al. [19], the disadvantage of monomeric (alum and iron) coagulants is their inability to control the nature of the coagulating species that are formed by rapid hydrolysis and precipitation, especially in the pH range of 6–8. During rapid hydrolysis, only some of the hydrolysis products are believed to be neutralized by the colloid charge, the rest chemically interact with dissolved components in the raw water [20]. Furthermore, changes in the raw water characteristics such as temperature and pH affect the performance of alum and iron coagulants [19].

Pre-polymerized or pre-hydrolyzed coagulants are made by partially neutralizing the monomeric coagulants to a different basicity ratio to control the formation of coagulating species. This process produces larger and highly positive coagulant species, thereby allowing pre-hydrolyzed coagulants to perform better than conventional coagulants.

Pre-hydrolyzed iron (PHI) is better than the monomeric coagulant [21]. Based on a study by Jiang and Wang [18], the precipitates of poly iron carry higher cationic charge and molecular weight than the precipitates of iron salt. Apart from that, PHI as a coagulant has lower sensitivity to raw water temperature and creates less Fe residues compared with the monomeric forms of iron [21]. The other advantages of PHI as a coagulant include lower dosage, less sludge production, able to perform in colder temperature, effectively precipitating organic substances, and producing only minimal iron residues in the treated water.

Basicity (B) ratio is considered as the key feature of PHI coagulants [22]. B ratio is defined as the molar ratio of the hydroxide ions bound per mole of iron coagulant [18,23,24]. B ratio is related to the concentration of the base added during the hydrolysis process. A high B ratio means more bases are added. When more bases are added, the alkalinity of the coagulant increases, thereby reducing the effect of pH reduction by the coagulant.

Different liquid samples have different optimum B ratios. A study done by Gao et al. [25], showed that the optimum value of B ratio was 1.0 for PHI when used in textile applications. For synthetic wastewater made from kaolin clay, the best B ratio was recorded at 2.0 by Liu and Chin [26], whereas 0.5–1.0 was recorded by Hong-Xio and Stumm [20]. A study by Jiang and Wang [18] reported that $B = 0.3$ showed minimal residues of DOC and color in a sample mixture containing humic acid and kaolin clay particles treated by PHI. For eutrophicated raw wastewater, the basicity ratio of 0.2 displayed the highest percentage removal of algae [19]. Based on a study by Lei et al. [19], the type of sample does affect the optimum value of the PHI B ratio. Thus, it is important to find the suitable B ratio as the effect of B ratio is unique in every type of sample.

Different types of coagulants (alum, ferrous sulfate, ferric chlorosulfate, ferric chloride, and natural coagulants) have been used by several researchers for leachate treatment [12,27–29]. However, to date, limited information exists on the application of PHI coagulants on partially stabilized leachates by coagulation. Therefore, this study aimed to optimize the coagulation process of PHI by varying three factors (pH, PHI basicity, and PHI dose) involved in evaluating the removal of suspended solids (SS), color, and COD in partially stabilized leachates.

2. Materials and methods

2.1. Preparation of PHI

The PHI used in this study was prepared based on the method of Lei et al. [21]. About 0.5 mol/L of NaOH was gradually added into a solution of 0.1 mol/L $FeCl_3$ at the flow rate 0.0038 mL/s with the stirring speed of 500 rpm at room temperature. The volume of NaOH added to $FeCl_3$ solution was determined based on the target basicity (0.1–1.0). Then, the PHI mixture was left to aged at room temperature for 24 h before it is able to be used. The characteristics of the synthesized PHI are shown in Table 1. The intensity of the PHI color increased as the basicity increased. The color changed from yellow to orange and then to brown. Increase in the intensity of color was also recorded during the PHI aging process.

2.2. Leachate sampling and characterization

Leachate samples were collected from the Matang landfill site (MLS) located in the northern region of Malaysia. MLS is situated at 4˚49´20.08´´N and 100˚40´44.08´´E near Taiping town in Perak, Malaysia. MLS is equipped with a leachate collection pond. The collection pond acts as a detention pond. The total landfill area is 12 ha. It is classified as an improved anaerobic landfill. MLS is more than 14 years old. The landfill receives about 300 tons of solid waste daily. Recycling is practiced at the site mainly by scavengers. The remaining solid waste is then dumped on site and covered with local soil.

The sampling procedures were conducted according to the method for collection and preservation of the samples [30]. All the collected samples were immediately transported to the laboratory and stored in a cold room at 4˚C to minimize biological and chemical reaction. Prior to analysis, the samples were placed under room temperature for 2 h. All parameters were measured according to the APHA [30] standard method. The characteristics of the raw MLS leachates are shown in Table 2. The value of the BOD/COD ratio for MLS is 0.12–0.16. This value indicates that MLS is a mature landfill with partially stabilized leachates (BOD/COD > 0.1) [20].

Table 1
Characteristics of PHI with different basicity value

No.	*B*	pH	Turbidity (NTU)
1	0.1	1.61	20
2	0.2	1.65	7
3	0.3	1.64	9
4	0.4	1.67	11
5	0.5	1.73	12
6	0.6	1.74	20
7	0.7	1.82	23
8	0.8	1.78	25
9	0.9	1.80	28
10	1.0	1.82	28

2.3. Jar test

The jar test apparatus (SW6 Stuart, Bibby Scientific Limited, UK) was used for the coagulation experiments. The tests were carried out at room temperature. A 1 L beaker was used as the reactor for 500 ml of leachate sample. NaOH and HCl were used for pH adjustment. A coagulant was added into the reactor and immediately mixed with the leachate. The mixture was slowly stirred for 50 min and was allowed to settle before the supernatant samples were collected with a syringe 3 cm below the surface of the leachate. The supernatant samples were collected and analyzed for SS, color, and COD. Analyses were conducted in duplicates and the mean values are presented in the result and discussion sections. Through the jar test, the optimum conditions of basicity, pH, and dose of PHI were determined. The other coagulation factors (Table 3) were determined through preliminary experiments.

3. Results and discussion

3.1. Optimum PHI basicity

The range of the pH basicity tested in this experiment was from 0.1 to 1.0 at pH 8 and dosage 0.4 g/L Fe. Based on Fig. 1, it can be concluded that the basicity value influenced the removal of the SS, color, and COD. Generally based on Fig. 1, PHI performance is reduced as basicity is increased, which is probably due to the reduction of charge and polymerization fraction [31]. As the basicity increased, the percentage of iron precipitate species is also increased. The sudden decrease in COD removal from 0.6 to 0.7, might be caused by the amount of positive charge iron species formed at this basicity was not enough to neutralize the iron precipitate. This iron precipitate is inorganic and this will increase the COD value in the treated leachate. Treatment of MLS leachate by $B = 0.1$

Table 2
Characteristics of raw leachate at MLS

No.	Parameters	Value		MEQA (1974)[b]
		Range	Average[a]	
1	Temperature (°C)	28–31	29	40
2	pH	7.96–8.17	8.1	6.0–9.0
3	BOD5 (mg/L)	60–184	109	20
4	COD (mg/L)	470–1,261	770	400
5	SS (mg/L)	222–303	271	50
6	Ammonia-N (mg/L NH_3-N)	311–693	500	5
7	Total phosphorus (mg/L PO_4^{3}-TNT)	22–54	42	–
8	Iron (mg/L)	2.3–3.1	2.7	5
9	Turbidity (NTU)	15–41	28	–
10	BOD/COD	0.12–0.16	0.14	–

[a] Average of 12 samples taken from December 2011–April 2012.

[b] Malaysian environmental quality (control of pollution from solid waste transfer station landfill) regulations 2009, under the laws of Malaysia environmental quality Act (MEQA) 1974.

Table 3
Optimum coagulation factors determined from preliminary experiment

Coagulation factors	Unit	Value
Rapid mixing speed	rpm	250
Rapid mixing time	minutes	4
Slow mixing speed	rpm	50
Slow mixing time	minutes	25
Settling time	minutes	60

yielded an optimum SS, color, and COD of 41, 46, and 6%, respectively. Therefore, basicity at 0.1 was selected as the optimum value for PHI for the determination of optimum pH.

3.2. Optimum pH

Treatment of MLS leachate by $B = 0.1$ (dosage = 0.4 g/L Fe) was examined at various pH conditions (2–11). The increase of the removal percentage was recorded for SS, color, and COD as the pH increased from 2 to 5 (Fig. 2). Decrease in the removal percentage was recorded at pH 6–10. A slight increase in the removal percentage was recorded at pH 11. The highest removal percentage was recorded at pH 5, with 96, 84, and 44% removal of SS, color, and COD, respectively.

SS, color, and COD are part of the natural organic matters NOM in leachate. The removals of organic matters are related to the pH. pH plays a vital role in the hydrolysis process of PHI [27]. Based on Fig. 2,

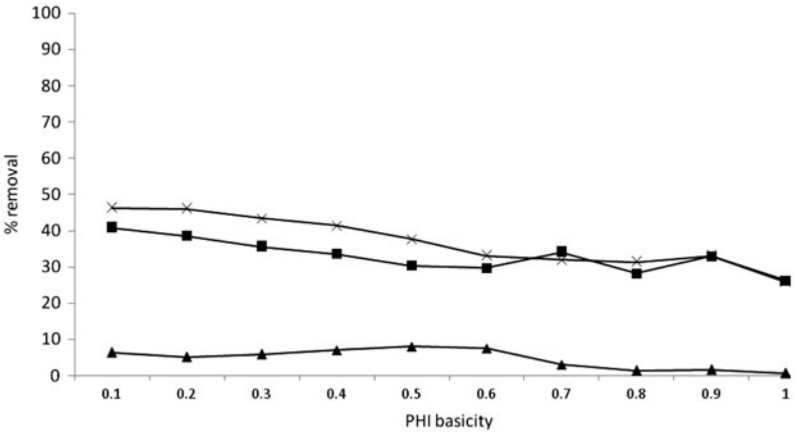

Fig. 1. Treatment of MLS leachate by varying basicity of PHI (Experimental condition: coagulant dose: 0.4 g/L Fe, rapid mixing duration: 4 min, rapid mixing speed: 250 rpm, slow mixing speed: 60 rpm, slow mixing duration: 25 min, settling time: 60 min, and pH:8) (—■— SS, —×— color, —▲— COD).

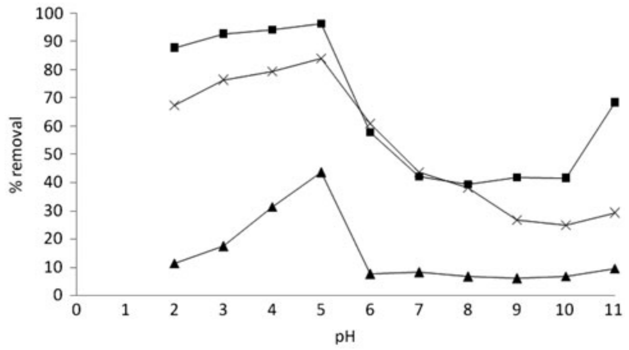

Fig. 2. Treatment of MLS leachate by varying pH of leachate (Experimental condition: PHI basicity: 0.2, coagulant dose: 0.4 g/L Fe, rapid mixing duration: 4 min, rapid mixing speed: 250 rpm, slow mixing speed: 50 rpm, slow mixing duration: 20 min, and settling time: 30 min) (—■— SS, —×— color, —▲— COD).

PHI performed better at low pH. This was due to the weak hydrolysis of PHI that occurred at a low pH, and more iron species were formed at this range. This resulted a sufficient charge neutralization action that occurred during coagulation–flocculation, and resulted in higher removal of SS, color, and COD. At pH 5, PHI produced the most effective range of iron species, thus making it the best removals recorded at this point. Lower than pH 5, PHI will produce more iron species or more positive charge of PHI. However, this will result in destabilization of flocs and increase the repulsion force between flocs. Therefore, lower pH does not always produce higher removal of pollutant for PHI. As the pH increased, the portion of positive charged iron species decreased, thus reduced the removal of SS, color, and COD (refer Fig. 2). Moreover, alkaline condition is not a favorable condition for the removal of negative charged leachate.

At optimum pH, the coagulant will produce the most effective hydrolysis species which will be responsible for the removal of pollutants. Based on previous findings, different coagulants will perform only at a certain range of pH (Table 4). The optimum pH of PAC in leachate samples was recorded between 7.5 and 7 by Ghafari et al. [32]. A study by Aziz et al. [7] found pH 6 as optimum condition for for $FeCl_3$. Both researchers tested the coagulant in leachate samples from Pulau Burung Landfill, yet the optimum pH was different as the coagulant used also differed. The optimum pH of PHI in this study was 5.

The percentage removals without pH adjustment were 40, 35, and 7% for SS, color, and COD, respectively. By adjusting the pH of MLS to 5, a huge increase in the percentage removal was recorded. Therefore, pH is an important factor during the destabilization process. This finding confirms Bratby's [23] claim and is consistent with the results of previous studies on the coagulation treatment of leachates [7,32,33].

3.3. Optimum dose

The amount of coagulant added into the sample is one of the key factors in determining the efficiency of the coagulation process. The dosage of the coagulant should be kept sufficient enough to allow the coagulant to perform at the required efficiency. Overdosing should be avoided because it would cause the restabilization of particles.

The range of doses tested for MLS were between 0.05 and 1 g/L Fe at pH 5. Results showed almost similar trend for SS, color, and COD removals. At lower dose (0.05–0.1 g/L Fe) color showed higher removal compared to SS and COD. Flocs formed at lower dose but the size and amount were small. However for color, the sample was filtered and the filtering process has helped in removing the flocs and SS from the leachate. A rapid increment in the slope of the trend line was recorded as the PHI dose was increased from 0.05 to 0.2 g/L Fe (Fig. 3). After the dose of 0.2 g/L Fe, the percentages of the removal of SS, color, and COD decreased slowly. Overdosing or in excess of optimal dosage occurred after 0.2 g/L Fe that caused restabilization of colloidal particulates due to increment of positive charged flocs.

Generally, as iron coagulants are added into the wastewater, it will hydrolyze and polymerize rapidly and produced various monomers and polymers with positive charges. These positively charged iron hydrolysis species were attracted to negatively charged colloid particles in wastewater. Then, it will result in charged neutralization action of the colloidal particle and causing the aggregation and finally formation of flocs [34]. At lower dose (<0.2 g/L Fe), the amount of positive charged monomer and polymer is not enough to fully neutralize the negative charged colloid. While at excess of optimal dose (>0.2 g/L Fe), the steady colloid was fully neutralized, while the redundant positive charge would be attracted to the particles and causing repulsion and restabilization, thus causing the colloid to be in steady state again. Therefore, the optimum dosage must be determined to ensure better performance of the coagulant relative to the cost. At the dosage of 0.2 g/L Fe, the removal percentages of 99, 95, and 58% were recorded for SS, color, and COD, respectively. Thus, 0.2 g/L Fe was selected as the optimum PHI dose.

Table 4
Optimum pH and dose of coagulant for leachate treatment

Coagulant	Sample	pH	Dose	SS[a]	Color[a]	COD[a]	Reference
$FeCl_3$	Leachate**	6	0.53 g/L Fe	99	97	45	[7]
Alum	Leachate**	6	2.5 g/L	70	55	27	[7]
PAC	Leachate***	7–7.5	2 g/L	92	91	43	[32]
$FeCl_3$ + DAF*	Leachate***	4.76	0.13 g/L Fe	–	93	75	[33]
PHI	Leachate***	5	0.2 g/L Fe	99	95	58	This study

[a] In percentage removal.

*Dissolve air floatation method.

**Unmatured.

***Matured.

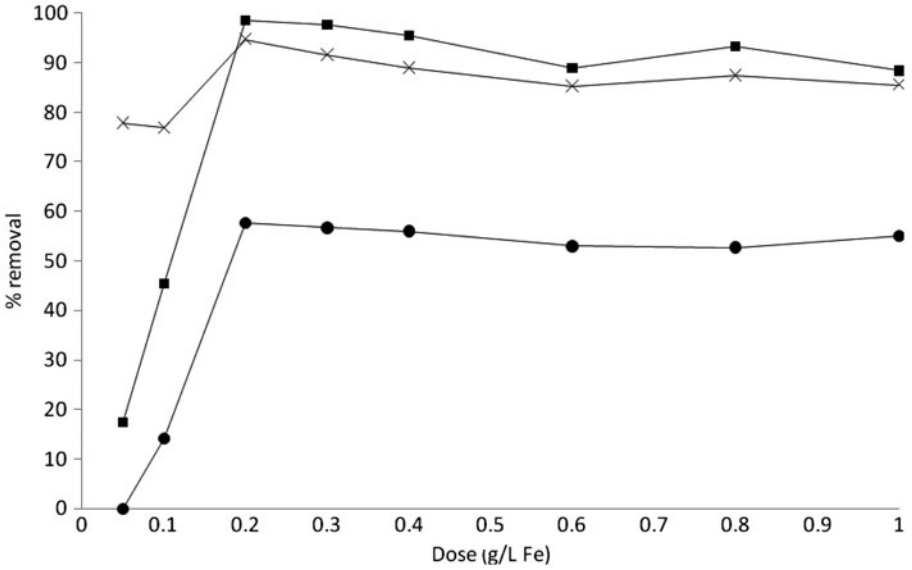

Fig. 3. Treatment of MLS leachate by varying dose of PHI (Experimental condition: PHI basicity: 0.2, rapid mixing duration: 4 min, rapid mixing speed: 250 rpm, slow mixing speed: 50 rpm, slow mixing duration: 20 min, settling time: 30 min, and pH:5) (—■— SS, —×— color, —▲— COD).

At pH 6, with 0.53 g/L Fe of $FeCl_3$, the percentages removal of 99, 96.5, and 44.7% for SS, color, and COD, respectively, were recorded by Aziz et al. [7] for leachate from Pulau Burung Landfill (Table 4). Based on this finding, PHI produced a concurrent removal percentage of SS, color, and COD except with 50% reduction of the dosage (optimum dose of PHI: 0.2 g/L Fe). Aziz et al. [7], tested alum at pH 6, dosage 2.5 g/L on Pulau Burung landfill leachate, and recorded the percentages removal of 70.4, 54.9, and 26.9% for SS, color, and COD, respectively. At the optimum dose of 2 g/L PAC, the percentages removal of 92, 91, and 43% for SS, color, and COD, respectively, were recorded by Ghafari et al. [32]. Compared with the SS, color, and COD removals by alum and PAC, PHI produced a much better result. Moreover, comparing removal of COD by coagulation of PHI with adsorption of municipal leachate with coal flyash, PHI removed 44% while coal flyash only removed 28% [35].

4. Conclusion

The optimization of the coagulation process of PHI on partially stabilized MLS leachates was investigated. The optimum values of basicity, pH, and dose were 0.1, 5, and 0.2 g/L Fe, respectively. At the optimum condition, the percentages removal of SS, color, and COD were 99, 95, and 58%, respectively. Basicity, dose, and pH optimization did significantly improve the performance of PHI. Judged against the findings of previous studies, the SS, color, and COD removals of PHI at the optimum condition in the current study resulted in almost the same efficiency as that of conventional coagulants, but utilized only half of the dosage.

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