

Treatment of landfill leachate by using lateritic soil as a natural coagulant

Syafalni^{a,*}, Han Khim Lim^a, Norli Ismail^b, Ismail Abustan^a, Mohamad Fared Murshed^a, Anees Ahmad^c

^a School of Civil Engineering, Engineering Campus, Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia

^b School of Industrial Technology, Universiti Sains Malaysia, 11800 USM Penang, Malaysia

^c Analytical and Environmental Chemistry Division, Department of Chemistry, Aligarh Muslim University, Aligarh, India

ARTICLE INFO

Article history:

Received 15 June 2010

Received in revised form

5 July 2012

Accepted 1 August 2012

Available online 8 September 2012

Keywords:

Coagulation–flocculation

Landfill leachate

Lateritic soil

Natural coagulant

ABSTRACT

In this research, the capability of lateritic soil used as coagulant for the treatment of stabilized leachate from the Penang–Malaysia Landfill Site was investigated. The evaluation of lateritic soil coagulant in comparison with commercialized chemical coagulants, such as alum, was performed using conventional jar test experiments. The optimum pH and coagulant dosage were identified for the lateritic soil coagulant and the comparative alum coagulant. It was found that the application of lateritic soil coagulant was quite efficient in the removal of COD, color and ammoniacal-nitrogen content from the landfill leachate. The optimal pH value was 2.0, while 14 g/L of lateritic soil coagulant was sufficient in removing 65.7% COD, 81.8% color and 41.2% ammoniacal-nitrogen. Conversely, the optimal pH and coagulant dosage for the alum were pH 4.8 and 10 g/L respectively, where 85.4% COD, 96.4% color and 47.6% ammoniacal-nitrogen were removed from the same leachate sample. Additionally, the Sludge Volume Index (SVI) ratio of alum and lateritic soil coagulant was 53:1, which indicated that less sludge was produced and was an environmentally friendly product. Therefore, lateritic soil coagulant can be considered a viable alternative in the treatment of landfill leachate.

© 2012 Elsevier Ltd. All rights reserved.

1. Introduction

Landfilling is one of the most widely accepted methods for the disposal of municipal solid waste (MSW) in several countries (Aziz et al., 2007b) because of its lower maintenance and operation costs. The degradation of the organic fraction of the wastes in combination with percolating rainwater leads to the generation of leachate (Kurniawan et al., 2006). Landfill leachate is considered a high-strength wastewater, which is characterized by extreme pH, biochemical oxygen demand (BOD), chemical oxygen demand (COD), inorganic salts and toxicity (Keenan et al., 1984). The leachate composition depends on many factors, such as waste composition, the availability of moisture and oxygen, the design and operation of the landfill, site hydro-geological factors and landfill age (Aziz et al., 2004). Moreover, landfill leachates have also been identified as a potential source of surface and groundwater contamination (Aziz et al., 2007a). This phenomenon is due to leachate migration through soils and sub-soils, which cause pollution of streams, water wells and creeks if they are not properly collected, treated and disposed (Tatsi et al., 2003). There are several types of treatment technologies drawn from wastewater and

drinking water technology that are applicable for leachate treatment, including physical, chemical, biological or a combination of treatment alternatives as precautionary approaches. Biological treatment alternatives are effective for freshly produced leachate but are ineffective for stabilized ones (more than 10 years). Alternatively, physical–chemical treatment methods are not favored for the treatment of young leachates but are advised for the treatment of old leachate (Ghafari et al., 2009).

Coagulation–flocculation processes have been widely used as treatment alternatives to remove pollutants such as BOD, COD, total suspended solids (TSS), heavy metals, color, and nitrogen compounds in landfill leachate or industrial wastewater prior to other treatment methods (Chu, 1999; Burke et al., 2000; Fan et al., 2003; Aziz et al., 2004, 2007a, 2007b; Ghafari et al., 2009). However, the coagulation–flocculation treatment methods involve chemicals as the treating agents, which could constitute a potential risk to the quality of the environment itself and cause excess sludge generation during the coagulation–flocculation treatment process (Tatsi et al., 2003). Thus, it is important to apply the appropriate coagulant in the coagulation–flocculation process to overcome these potential disadvantages and increase the efficient application of an alternative treatment.

Lateritic soil, also known as red earth, is found in the tropics and subtropics (Maji et al., 2008). It is a residual product of a wide variety of intensive chemical weathering processes that affect rocks

* Corresponding author. Tel.: +60 45996291; fax: +60 45941009.
E-mail address: cesyafalni@eng.usm.my (Syafalni).

under strong oxidizing and leaching conditions (Ko et al., 2006). In addition, lateritic soil is enriched with aluminum silicates, aluminum hydrosilicates, iron oxides and iron hydroxides because the water leaches out the bases and the silic acid (Maji et al., 2008). Such phenomenon can be proved by the iron compound, which leads to the typical red color of the soil. Furthermore, lateritic soil is a kind of soil with abundant clay minerals that show a high affinity for immobilizing cationic or organic contaminants due to their large specific surface area and negatively charged surface (Wang et al., 2008). Additionally, developments in using lateritic soil as an adsorbent for gas cleaning and wastewater treatment have also been widely reported in recent years (Maji et al., 2008; Ko et al., 2006; Wang et al., 2008; Yu et al., 2008; Mohapatra et al., 2009). In addition, lateritic soil is known to contain high concentrations of aluminum ions (Al^{3+}) and ferric ions (Fe^{3+}), where both of these ions are the primary functional compounds in widely use chemical coagulants.

This research study investigated the suitability of lateritic soil as a coagulant for landfill leachate treatment and is compared it with a conventional coagulant, aluminum sulfate (alum). The appropriate implementation of this method depends upon how precisely the pH and coagulant dosage of the lateritic soil are chosen. Central composite design (CCD) of response surface methodology was used to build model and determine the optimum conditions. COD, color, ammoniacal-nitrogen removal and SVI determination were monitored throughout the experiment. Hence, the statistical design was based on two factors (pH and coagulant dosage) and two responses (percentage of COD and color removal).

2. Materials and methods

2.1. Landfill leachate sampling and characterization

Leachate samples were collected from a landfill site situated within the Byram Forest Reserve Area, Penang, Malaysia. The site is a semi-aerobic landfill with an area of 23.7 ha and receives 1500 tons of solid waste daily (Ghafari et al., 2009). Moreover, the site is equipped with a leachate collection pond. Leachate samples were obtained from eight sampling points (composite sampling procedure) from one of the detention ponds, three times, and at two-week intervals in February and March 2009. The leachate samples collected were stored in 25-L plastic carboys and packed in cooler boxes containing ice cubes for the journey back to the laboratory from the site. In the laboratory, the leachate samples were kept in the refrigerator at 4 °C and taken for use when needed so that the potential for sample volatilization or biodegradation of the samples was minimized. The collection and preservation of the samples were done according to the Standard Methods for the Examination of Water and Wastewater (APHA, AWWA and WEF, 2005).

Landfill leachate samples were characterized immediately right after the samples arrived in the laboratory. The pH, COD, color, total solids (TS), TSS and turbidity were analyzed according to the Standard Methods for the Examination of Water and Wastewater (APHA, AWWA and WEF, 2005). The results obtained from the characterization studies are presented in Table 1 (MDC, 2006).

2.2. Lateritic soil sampling and characterization

Lateritic soil was collected at Kuari Beseri, Perlis and ground into powder using a ball mill grinder. The soil used was the outermost layer (topsoil). It is important to determine the characteristics of lateritic soil must be determined prior to use as a coagulant for the treatment of landfill leachate. The parameters characterizing the lateritic soil were pH, total organic carbon and the chemical

Table 1

Characteristics of the raw landfill leachate from a landfill site from Penang–Malaysia.

Parameter	Range	Mean	Effluent (standard B) ^a	S.D. ^b
pH	8.30–8.45	8.38	5.5–9.0	0.11
Temperature (°C)	25–31	28	40	4.24
COD (mg/L)	2950–4050	3500	100	777.82
Color (PtCo)	4650–4850	4750	–	141.42
Total solids (mg/L)	9790–9950	9870	–	113.14
Total suspended solids (mg/L)	86–298	192	100	149.91
Turbidity (NTU)	58–74	66	–	11.31
Ammoniacal-nitrogen (mg/L)	1820–2260	2040	–	311.13

^a Environmental Quality (Sewage and Industrial) regulations, 1979 (MDC, 2006).

^b Standard Deviation.

composition of the powdered lateritic soil determined by energy dispersive x-ray spectroscopy (EDX). The chemical elements found in the soil as a percentage of concentration (%) using EDX are reported in Table 2.

2.3. Coagulation–flocculation

Coagulation–flocculation studies were conducted in a conventional jar test apparatus (VELP-Scientifica, Model: JLT6, Italy) with impellers equipped with 2.5 cm × 7.5 cm rectangular blades (Ghafari et al., 2009) and six 500 mL beakers. First, the landfill leachate samples were taken out of the refrigerator and were allowed to reach ambient temperature (approximately 2 h). Next, the sample bottle was thoroughly agitated for re-suspension of possibly settled solids. Then, 200 mL of the leachate sample was transferred into each of the corresponding jar test beakers and the coagulation–flocculation experiment was initiated using alum and lateritic soil. The alum used in this study was in powder form with the formula $Al_2(SO_4)_3 \cdot 16H_2O$ ($M = 630.39$ g/mol) and supplied by SYSTERM, Malaysia. The operating parameters used were based on literature reviewed. The speed of rapid mixing was set at 100 rpm for 3 min, while the speed of slow mixing was set at 30 rpm for 10 min. A settling time (30 min) was allocated for the settlement of suspended particles after the jar test process.

2.4. Experimental design and data analysis

The 2^k factorial design was used in this research to determine the joint effects of several factors on a response. This design provides the smallest number of treatment combinations with which k factors can be studied in a complete factorial arrangement. Important interactions among factors may not be detected without the use of this design and may lead to misleading conclusions. The pH and coagulant dosage were chosen as independent factors, while the percentage of COD and color removal were the dependent responses variables investigated throughout the study.

Central composite design (CCD) of the response surface methodology (RSM) was also used to optimize two important operating factors (pH and coagulant dosage) of the lateritic soil being used as

Table 2

Chemical elements found in lateritic soil using EDX.

Element	Weight%	Atomic%
Carbon	5.94	9.86
Oxygen	46.91	58.43
Aluminum	18.01	13.31
Silicon	22.70	16.11
Iron	6.44	2.30

a coagulant for the treatment of landfill leachate. The experiment began with a preliminary study to narrow the range of pH and coagulant dosage prior to designing of the experimental runs (Ghafari et al., 2009). In addition, a wide range of pH (1–12) was examined, and numerous coagulant dosages, starting from 2 g/L, were incrementally investigated until appreciable reductions were observed in the process responses (COD and color) (Ghafari et al., 2009). Consequently, pH between 1.0–5.0 and coagulant dosages of 8–16 g/L were chosen as the study ranges for the lateritic soil. The coded values for pH (A) and coagulant dosage (B) were set at five levels: –1.5 (minimum), –1.0, 0 (central), 1.0 and 1.5 (maximum). To obtain optimum pH and coagulant dosage, two dependent parameters were analyzed (COD removal and color removal). Experimental results are shown as the percent removal of COD and color. When a process is relatively close to the optimum, the second-order model that incorporates curvature is required to approximate the response (Montgomery, 2006) as expressed in Eq. (1):

$$Y = \beta_0 + \sum_{i=1}^k \beta_i x_i + \sum_{i=1}^k \beta_{ii} x_i^2 + \sum_{i < j} \beta_{ij} x_i x_j + \epsilon \quad (1)$$

where Y is the predicted response; i is the linear coefficient; j is the quadratic coefficient; β is the regression coefficient; k is the number of factors studied and optimized in the experiment; and ϵ is the random error.

The Minitab Release 14 Statistical Software was used for the statistical design and data analysis to estimate the response of the dependent variables and obtain the effects, coefficients, standard deviation of coefficients and other statistical parameters of the model. The optimum conditions were obtained graphically from the contour plot and by solving the polynomial regression equation. Goodness of fit was expressed by the coefficient of determination R^2 . Statistical significance was analyzed through Analysis of Variance (ANOVA), where the level of significance at a 5% probability level was given as the P value with a 95% confidence level.

3. Results and discussion

3.1. Screening of independent factors for percentage COD and color removal

In this study, the independent factors screened consisted of pH and coagulant dosage, while the influence of these factors was determined by measuring the percentage of COD and color removal. Subsequently, the effects of these factors and their interactions were studied by performing the experiment described in Table 3, in triplicate using 2^2 full factorial design. Factor levels were coded as – (low level) and + (high level). The pH and coagulant dosage (lateritic soil), along with the experimental ranges and levels for the 2^2 full factorial design, are presented in Table 4. The Analysis of Variance (ANOVA) presented in Table 5 shows that pH and coagulant dosage were significant in COD reduction and color removal of the landfill leachate. Therefore, both of the parameters must be considered during the optimization of experimental work. Moreover, the interaction between pH and coagulant dosage were significant in the experimental work mentioned.

3.2. Optimization of percent COD and color removal

Central Composite Design (CCD) was used to obtaining the relationship between the independent variables and two important process responses (COD and color removal efficiency) for the coagulation–flocculation process. Significant model terms are needed to get a good fit in a particular model (Ghafari et al., 2009).

Table 3

The input factors of pH/coagulant dosage (lateritic soil) and the experimental ranges and level for the 2^2 full factorial design.

Experiment	Block	Experimental factors	
		A (pH)	B (mg/L)
1	2	1.5	16,000
2	2	2.0	14,000
3	2	1.5	12,000
4	2	1.5	12,000
5	2	2.5	16,000
6	2	2.0	14,000
7	1	2.5	12,000
8	1	1.5	16,000
9	1	2.0	14,000
10	1	1.5	12,000
11	1	2.0	14,000
12	1	2.5	16,000
13	3	1.5	16,000
14	3	2.5	12,000
15	3	1.5	12,000
16	3	2.0	14,000
17	3	2.5	16,000
18	3	2.0	14,000

Factors	Levels	
	– (Low level)	+ (High level)
A: (pH)	1.5	2.5
B: (Coagulant dosage, mg/L)	12,000	16,000

Table 4

CCD results for the study of two experimental variables for lateritic soil used as a coagulant.

Run no.	Lateritic soil			
	Experimental design		Results (removal %)	
	A: pH	B: mg/L	COD	Color
1	2.00 (0)	14,000 (0)	65.71	81.75
2	2.00 (0)	10,000 (–1.5)	57.86	72.22
3	2.00 (0)	14,000 (0)	66.43	83.33
4	2.00 (0)	18,000 (1.5)	62.14	77.78
5	1.00 (–1.5)	14,000 (0)	63.57	78.57
6	3.00 (1.5)	14,000 (0)	58.57	65.87
7	2.00 (0)	14,000 (0)	65.00	80.16
8	1.50 (–1)	12,000 (–1)	62.85	75.40
9	2.00 (0)	14,000 (0)	67.14	81.75
10	1.50 (–1)	16,000 (1)	62.14	76.26
11	2.50 (1)	12,000 (–1)	57.85	65.07
12	2.50 (1)	16,000 (1)	60.71	67.46
13	2.00 (0)	10,000 (–1.5)	59.29	75.32
14	2.00 (0)	18,000 (1.5)	62.86	78.57
15	3.00 (1.5)	14,000 (0)	60.00	65.07
16	2.00 (0)	14,000 (0)	65.71	82.53
17	2.00 (0)	14,000 (0)	68.57	84.13
18	1.00 (–1.5)	14,000 (0)	65.00	76.19
19	2.50 (1)	12,000 (–1)	56.43	64.29
20	1.50 (–1)	12,000 (–1)	64.29	78.57
21	2.50 (1)	16,000 (1)	59.29	67.46
22	2.00 (0)	14,000 (0)	65.71	81.75
23	2.00 (0)	14,000 (0)	66.43	83.33
24	1.50 (–1)	16,000 (1)	63.57	76.98
25	2.00 (0)	14,000 (0)	65.00	80.16
26	1.50 (–1)	12,000 (–1)	61.43	74.60
27	2.50 (1)	16,000 (1)	60.00	68.25
28	2.50 (1)	12,000 (–1)	57.14	63.49
29	2.00 (0)	14,000 (0)	67.14	81.75
30	1.50 (–1)	16,000 (1)	62.86	76.19
31	2.00 (0)	14,000 (0)	65.71	82.53
32	2.00 (0)	10,000 (–1.5)	61.43	76.26
33	3.00 (1.5)	14,000 (0)	57.85	65.87
34	2.00 (0)	14,000 (0)	68.57	84.13
35	1.00 (–1.5)	14,000 (0)	66.43	77.78
36	2.00 (0)	18,000 (1.5)	62.14	76.98

Table 5
Analysis of Variance for 2² full factorial design.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
COD R-Sq = 0.9483 R-Sq (adj) = 0.9200						
Blocks	2	4.260	4.260	2.130	2.12	0.166
Main effects	2	61.261	61.261	30.631	30.54	0.000
2-Way interactions	1	6.135	6.135	6.135	6.12	0.031
Curvature	1	130.569	130.569	130.569	130.17	0.000
Residual error	11	11.034	11.034	1.003		
Lack of fit	8	9.500	9.500	1.188	2.32	0.263
Pure error	3	1.534	1.534	0.511		
Total	17	213.259				
Color R-Sq = 0.9766 R-Sq (adj) = 0.9638						
Blocks	2	6.447	6.447	3.223	1.86	0.202
Main effects	2	305.378	305.378	152.689	87.98	0.000
2-Way interactions	1	13.293	13.293	13.293	7.66	0.018
Curvature	1	470.239	470.239	470.239	270.96	0.000
Residual error	11	19.090	19.090	1.735		
Lack of fit	8	12.201	12.201	1.525	0.66	0.714
Pure error	3	6.889	6.889	2.296		
Total	17	814.447				

Table 6
Analysis of Variance of Central Composite Design (CCD).

Source	DF	Seq SS	Adj SS	Adj MS	F	P-value
COD R-Sq = 0.9280 R-Sq (adj) = 0.8990						
Regression	5	368.142	368.142	73.628	62.16	0.000
Linear	2	130.616	130.616	65.308	55.14	0.000
Square	2	231.392	231.392	115.696	97.68	0.000
Interaction	1	6.135	6.135	6.135	5.18	0.032
Residual error	25	29.612	29.612	1.184		
Lack of fit	19	16.334	16.334	0.860	0.39	0.946
Pure error	6	13.278	13.278	2.213		
Total	30	397.754				
Color R-Sq = 0.9670 R-Sq (adj) = 0.9540						
Regression	5	1374.45	1374.448	274.890	141.91	0.000
Linear	2	549.28	549.277	274.638	141.78	0.000
Square	2	817.71	817.714	408.857	211.07	0.000
Interaction	1	7.46	7.458	7.458	3.85	0.061
Residual error	25	48.43	48.427	1.937		
Lack of fit	19	40.84	40.843	2.150	1.70	0.264
Pure error	6	7.58	7.585	1.264		
Total	30	1422.88				

The CCD, as presented in Table 4, allowed the development of mathematical equations, where predicted results (Y) were analyzed as a function of pH (A) and coagulant dosage (B) and calculated as the sum of a constant, two first-order effects (A term and B term), one interaction effect (AB) and two second-order effects (A² and B²) according to Eq. (1). Hence, range and levels of factors were based on the earlier screening results indicating the presence of a curvature indicating that the experimental ranges were relatively close to the optimum. The quadratic regression models for the percentage COD and color removal, in terms of coded factors regardless of their significance, are given in the following equations:

$$Y_{(COD)} = 66.4143 - 2.1016 A + 0.8400 B - 2.3006 A^2 - 2.7228 B^2 + 0.7150 AB; \tag{2}$$

$$Y_{(color)} = 82.2307 - 4.5324 A + 0.9990 B - 5.5878 A^2 - 3.5300 B^2 + 0.7883 AB, \tag{3}$$

where Y is the predicted response; and A and B are in coded form. The results of the ANOVA show significant effect of the selected parameters in the optimization work of landfill leachate treatment (Table 6).

The R² coefficient presents the proportion of the total variation in the response predicted by the model, indicating the ratio of the sum of squares due to regression to the total sum of squares (Ghafari et al., 2009). In addition, an R² value close to 1 with desirable and reasonable agreement with the adjusted R² was necessary; high R² coefficient ensured a satisfactory adjustment of the quadratic model to the experimental data. Therefore, the values of R² 0.9280 (COD) and 0.9670 (color) (Table 6) showed a satisfactory prediction of the experimental data.

The effects on COD and color removal efficiencies are shown in 3D diagrams (Figs. 1 and 2) for lateritic soil used as a coagulant. Thus, Fig. 1 shows the maximum COD removal of 65% is achieved at a pH level (A) between -1 and 0 (pH between pH 1.5 and pH 2.0) and level of dosage (B) between -1 and 0 (dosage between 12 and 14 g/L). This phenomenon indicates that the most favorable conditions for COD removal are at lower range of pH and coagulant dosage. On the other hand, Fig. 2 shows the maximum color reduction of 80% is achieved at level of pH (A) between -1 and 0 (pH between pH 1.5 and pH 2.0) and level of coagulant dosage (B) between 0 and 1 (dosage between 14 and 16 g/L). This phenomenon

shows that the most favorable conditions for color removal were at lower pH and higher coagulant dosage but were not to exceed 16 g/L because the 3D diagram remained within the range level -1 and 1.

3.3. Effects of pH

The highest COD and color removal efficiency values were attained by applying alum as the coagulant were achieved with a pH 4.80 as presented in Fig. 3. The percentage of COD removal at pH 4.80 was 68.9%, while the percentage of color removal was 89% at the same pH level. This phenomenon is in agreement with previous experiments that showed the extent of pH not only depends on the type and concentration of the coagulant but also on the characteristics of the wastewater itself (Aziz et al., 2007b). Under acidic conditions, the amount of cationic ions (Al³⁺) increased, resulting in high coagulation activity because the charge neutralization effect between negatively charged suspended or colloidal particles was maximized. Alternatively, the best pH value for the lateritic soil used as a coagulant is at a pH of 2.00 and is displayed in Fig. 4, where the percentages of COD and color removal were 40% and 61.9%, respectively. The treatment of leachate using lower pH values shows better results Wang et al. (2002). The soils generally act as ion exchangers because they are aluminosilicates. In water solution, exchange may take place releasing the hydrogen ions which is responsible for the acidic pH. Then, the pH was

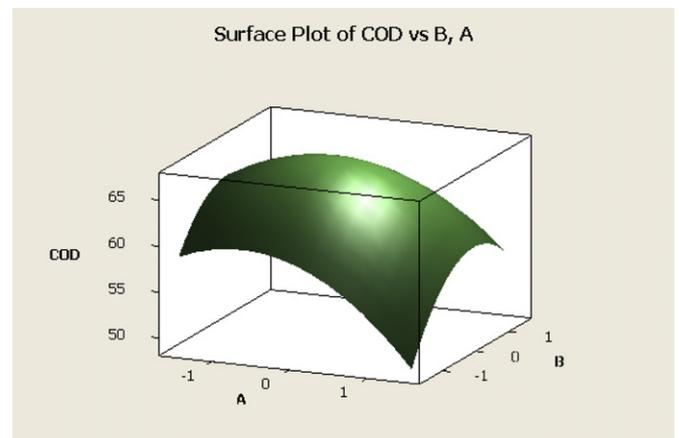


Fig. 1. 3D plots of percentage COD removal.

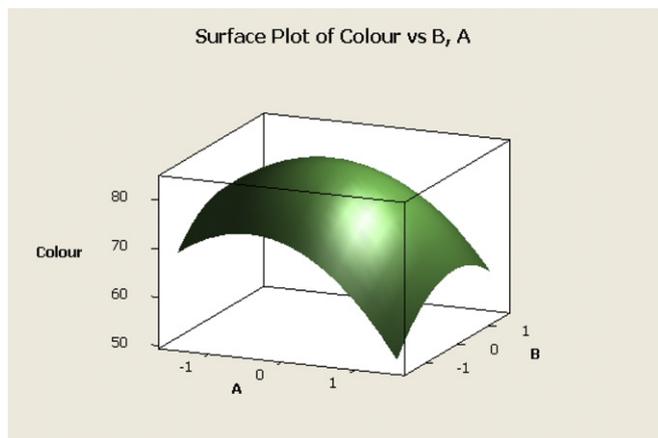


Fig. 2. 3D plots of percentage color removal.

adjusted to pH 2.00 which gives the best results. At this pH, hydrogen ion compete with extra aluminum ions and get exchange in the soil releasing thereby maximum aluminum ions which is responsible for the coagulation process. Second factor might be partial dissolution of the soil releasing aluminum ions for the coagulation process. Additionally, the trend depicted in Fig. 4 a decrement of percentage of COD and color removal when the pH of the solution exceeds pH 2.00, and an increase in the COD and color when compared the raw leachate sample. The dissolution of metal ions increases at lower pH range and becomes deficient when the pH falls in the alkaline range, thus competition between the electrolyte cations and metals adsorbed to the colloidal particles may occur due to the increment of ionic strength of soil solution (lower pH) (Lombnæs et al., 2008). Therefore, the effectiveness of the dissolution of metal ions from soil enhances the treatment efficiency of the landfill leachate. The COD and color removal of the leachate by the lateritic soil increases up to pH 2.00 and then decreases with the increasing of pH as shown in Fig. 4. After pH 4.00, the trend of COD and color removal become similar and show negative values. This may be accounted for the change in surface charge which is zero at pH 4.00 (Chairidchai and Ritchie, 1990) and become positive after that. In acidic medium especially at pH 2.00, the humic substances are insoluble in water (Thurman and Malcolm, 1981) and do remain associated with the soil imparting negative charge to the soil and contributing in the coagulation process. With the increase in pH, the solubility of humic substances increases resulting thereby decrease in surface charge of the soil which is responsible for coagulation. Though lateritic soil might release the humic substances and color which can increase the TOC

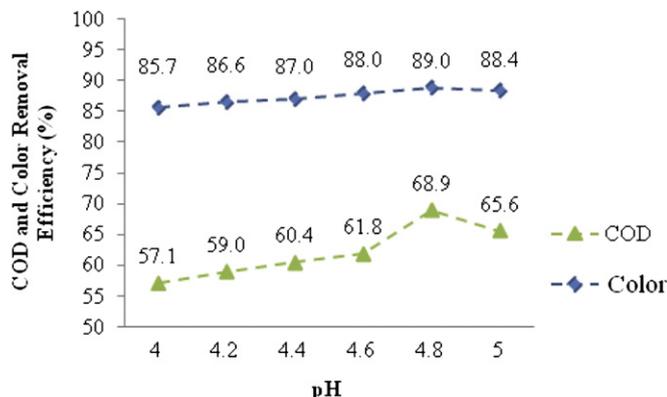


Fig. 3. Percentage of COD and color removal versus pH (alum).

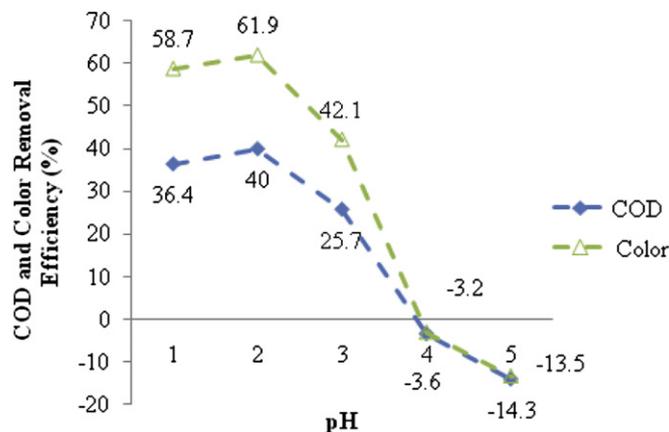


Fig. 4. Percentage of COD and color removal versus pH (lateritic soil).

of the equilibrating solution but the observation showed that the net reduction in the COD and color of the leachate by the soil for the sake of economic point of view and obtained the positive result. Additionally, the removal of COD of the leachate decreases when the pH of the solution exceeds pH 2.00 might be due to deprotonation of the hydroxyl groups at mineral surfaces in soil decreases the positive net surface charge plus organic molecules turn into more electronegative. The organic materials and mineral surfaces repel each other and limit the surface complex formation, releasing the organic materials into the soil solution and thus increase the COD of the soil solution (Grybos et al., 2009).

3.4. Effects of coagulant dosage

The percent removal of COD increased as the dosages of alum and lateritic soil increased. The observed removal efficiency of COD using alum as a coagulant increased linearly up to 85.4% at a dosage of 10,000 mg/L but decreased as the alum dosage reached 11,000 mg/L, with a COD removal efficiency of 84.9%, as shown in Fig. 5. This phenomenon could be attributed to the restabilization of colloidal particulates when the coagulants were used at dosages in excess of the optimum value (Aziz et al., 2007a). However, this reduction is not very significant and is within the experimental error. Additionally, the lateritic soil coagulant showed the same

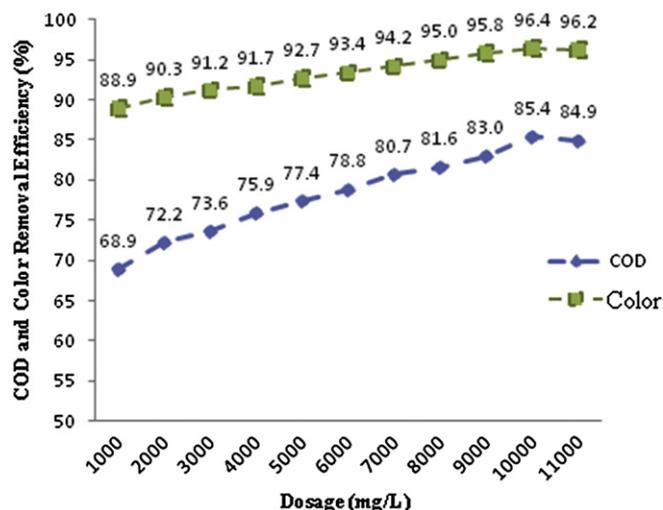


Fig. 5. Percentage of COD and color removal versus dosage (mg/L) by using alum.

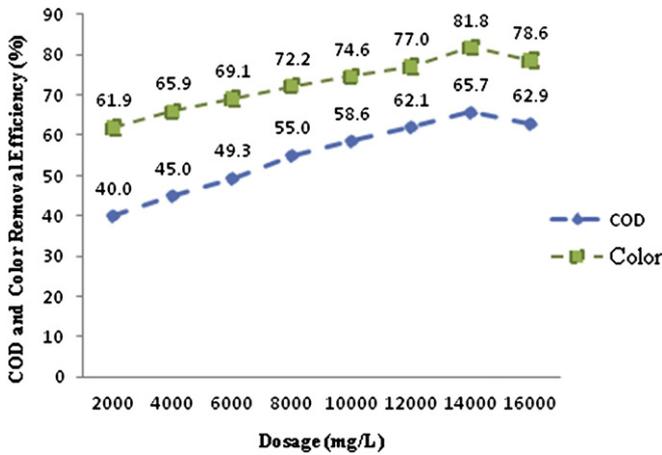


Fig. 6. Percentage of COD and color removal versus dosage (mg/L) by using lateritic soil.

effect as the alum coagulant. The trend in Fig. 6 shows that the COD removal efficiency increased as the lateritic soil dosage increased. Hence, the maximum point of the COD removal of leachate by using lateritic soil as a coagulant is at a dosage level 14,000 mg/L, with a removal efficiency of 65.7%. In addition, the alum coagulant is better than the lateritic soil coagulant with regards to COD removal efficiency because the alum dosage was less than the dosage of lateritic soil but still effectively treated the landfill leachate (COD). Based on the color removal efficiency, the alum coagulant performed better than the lateritic soil coagulant, as shown in Figs. 5 and 6. This phenomenon resulted from the fact that the lateritic soil was red, clay rich soil and hence, it may have contributed to the color of the landfill leachate (Maji et al., 2008). The color removal efficiency using alum was 96.4%, whereas it was 81.8% when using lateritic soil as the coagulant. The difference between the color removal efficiencies for the landfill leachate was 14.6%.

3.5. Removal of ammoniacal-nitrogen under optimized conditions

The ammoniacal nitrogen concentration of the raw leachate is 2040 mg/L (mean value), as presented in Table 1. Ammoniacal nitrogen in leachate may last for several decades and is a result of slow leaching and the release of soluble nitrogen from solid waste in landfills (Aziz et al., 2004). From Fig. 7, the removal efficiency of ammoniacal nitrogen using the alum coagulant was 47.6%, while 41.2% was removed using the lateritic soil as a coagulant under

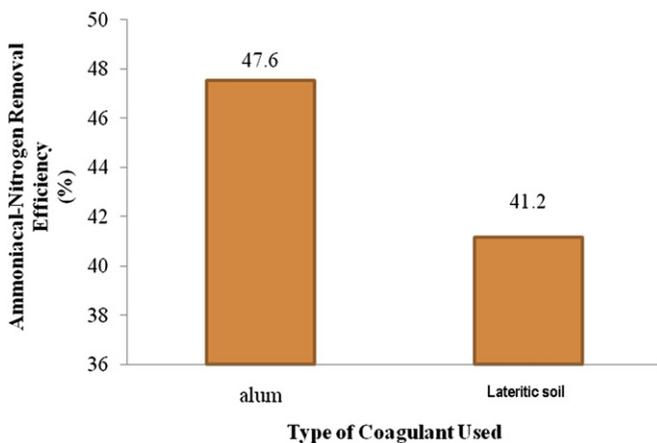


Fig. 7. Percentage of ammoniacal-nitrogen removal against type of coagulants used.

optimized conditions (alum: pH 4.80, dosage 10,000 mg/L; lateritic soil: pH 2.0, dosage 14,000 mg/L). Although the removal percentage was higher using alum, the lateritic soil also provided an ideal removal percentage for ammoniacal nitrogen.

The possible mechanism for the removal of ammoniacal nitrogen might be the adsorption of ammonium ion (NH₄⁺) on the coagulated material (lateritic soil) that contains high iron clay (Umapriya and Shrihari, 2010) and presents of potential of ion exchange for charged pollutant (Vimonses et al., 2009), especially for material which have negative charges on the surface. The ammonium ion being positively charged species are probably acting as complementary species to extend the effect of Al³⁺ and Fe³⁺ for the coagulation of suspended particles which can be presented as follows:

- (1) Al³⁺–*SP–NH₄⁺–SP–Al³⁺
 - (2) Fe³⁺–SP–NH₄⁺–SP–Fe³⁺
 - (3) Al³⁺–SP–NH₄⁺–SP–Fe³⁺
- (*SP = Suspended Particles)

Thus, the ammonium ions are acting as bridging element in the coagulation process and for their adsorption cum co-precipitation with sludge. The whole model can be visualized as 3D matrix of aluminum (or iron) and suspended particles (negatively charged) where NH₄⁺ and other counter ions (negatively charged) are embedded alternatively to the oppositely charged particles in the void spaces of the matrix. The overall percentage of removal of the ammoniacal nitrogen with the laterite soil is lower than the alum. These can also be explained in terms the proposed mechanism. With the presence of laterite soil, the total surface area in the sludge matrix is relatively lower due to the occupancy of soil particles in the sludge and hence relatively less NH₄⁺ ions are adsorbed and removed. The proposed mechanism can further be supported with the chemical compositions of the lateritic soil itself which contains of aluminum, silica and iron (Table 2). These essential compounds exhibit similar adsorption behavior as reported by Theriault and Mcnamee (1936) that aluminosilicate complexes in the activated sludge manage to remove ammonia and organic matters from wastewater based on the criterion of zeolitic behavior.

3.6. Sludge Volume Index (SVI) for alum and lateritic soil under optimized conditions

Fig. 8 shows that the Sludge Volume Index (SVI) generated by the alum (chemical coagulant) was higher than for the lateritic soil

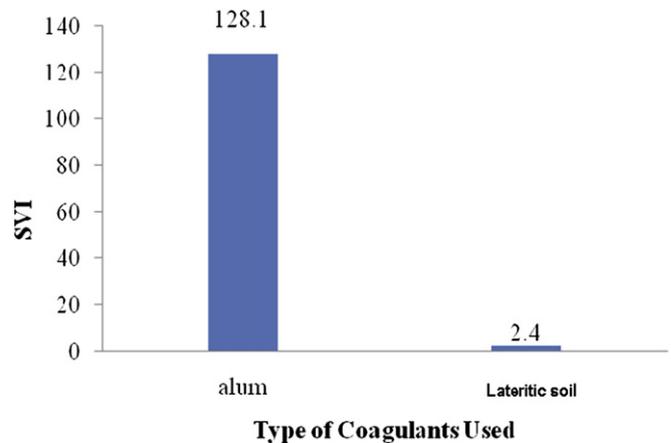


Fig. 8. Sludge Volume Index (SVI) against type of coagulants used.

(natural coagulant). Alum resulted in a 128.1 SVI, while the lateritic soil SVI was approximately 2.4. The ratio for the volume of sludge produced in the case of the alum and lateritic soil coagulants was 53:1. Furthermore, the lateritic soil produced a relatively compact sludge as compared to the alum because lateritic soil is an organic product that is environmentally friendly.

4. Conclusions

This study has successfully examined the treatment of landfill leachate via coagulation–flocculation purification methodology using lateritic soil as a coagulant and alum as a comparative coagulant. The lateritic soil coagulant was enriched with aluminum ions and iron ions (cationic ions) that destabilize the colloidal and suspended particles in the liquid media such as landfill leachate, via a coagulation mechanisms relying on adsorption and charge neutralization plus inter-particle bridging. The optimum pH for using the lateritic soil as a natural coagulant was 2.0, with a coagulant dosage of 14 g/L. The lateritic soil coagulant was also able to remove 65.7% COD and 81.8% color from the landfill site leachate under an optimum pH and coagulant dosage. Alternatively, the alum coagulant removed 85.4% COD and 96.4% color from the same leachate sample under optimized conditions (pH 4.80 and alum dosage of 10 g/L). Additionally, alum as a chemical coagulant reduced the concentration of ammoniacal nitrogen by 47.6%, while lateritic soil reduced it by 41.2%. In addition, the ratio of alum to lateritic soil was determined as 53:1 throughout this research project, where the SVI of alum coagulant was 128.1, while it was 2.4 for the lateritic soil under optimized conditions.

Acknowledgment

This research described in this paper was supported by USM and partially supported by MOSTI Science Fund (Project No. 03-01-05-SF0430).

References

- APHA, AWWA, WFE, 2005. In: Standard Methods for the Examination of Water and Wastewater, 21st ed. American Public Health Association (APHA), Washington DC.
- Aziz, H.A., Shahrir, M.N., Zahari, M., Alias, S., 2004. Removal of ammoniacal nitrogen (N-NH₃) from municipal solid waste leachate by using activated carbon and limestone. *Waste Manage. Res.* 22, 371–375.
- Aziz, H.A., Alias, S., Adlan, M.N., Asaari, F.A.H., Zahari, M.S., 2007a. Colour removal from landfill leachate by coagulation and flocculation processes. *Bioresour. Technol.* 98, 218–220.
- Aziz, H.A., Alias, S., Assari, F., Adlan, M.N., 2007b. The use of alum, ferric chloride and ferrous sulphate as coagulants in removing suspended solids, colour and COD from semi-aerobic landfill leachate at controlled pH. *Waste Manage. Res.* 25, 556–565.
- Burke, G.H., Singh, B.R., Theodore, L., 2000. *Handbook of Environmental Management and Technology*, second ed. Wiley-Interscience, New York, pp. 217–235.
- Chairidchai, P., Ritchie, G.S.P., 1990. Zinc adsorption by a lateritic soil in the presence of organic ligands. *Soil Sci. Soc. Am. J.* 54, 1242–1248.
- Chu, W., 1999. Lead metal removal by recycled alum sludge. *Water Res.* 33, 3019–3025.
- Fan, M., Brown, R.C., Sung, S.W., Huang, C.P., Ong, S.K., Leeuwen, J.H., 2003. Comparisons of polymeric and conventional coagulants in arsenic (V) removal. *Wat. Environ. Res.* 75, 308–313.
- Ghafari, S., Aziz, H.A., Isa, M.H., Zinatizadeh, A.A., 2009. Application of response surface methodology (RSM) to optimize coagulation–flocculation treatment of leachate using poly-aluminum chloride (PAC) and alum. *J. Hazard. Mater.* 163, 650–656.
- Grybos, M., Davranche, M., Gruau, G., Petitjean, P., Pédrot, M., 2009. Increasing pH drives organic matter solubilisation from wetland soil under reducing conditions. *Geoderma* 154, 13–19.
- Keenan, J.D., Steiner, R.L., Fungaroli, A.A., 1984. Landfill leachate treatment. *J. Wat. Pollut. Control Fed.* 56 (1), 27–33.
- Ko, T.H., Chu, H., Lin, H.P., Peng, C.Y., 2006. Red soil as a regenerable sorbent for high temperature removal of hydrogen sulfide from coal gas. *J. Hazard. Mater.* B136, 776–783.
- Kurniawan, T.A., Lo, W.H., Chan, G.Y.S., 2006. Physico-chemical treatments for removal of recalcitrant contaminants from landfill leachate. *J. Hazard. Mater.* B129, 80–100.
- Lombnaes, P., Chang, A.C., Singh, B.R., 2008. Organic ligand, competing cation, and pH effects on dissolution of zinc in soils. *Pedosphere* 18 (1), 92–101.
- Maji, S.K., Pal, A., Pal, T., 2008. Arsenic removal from real-life groundwater by adsorption on lateritic soil. *J. Hazard. Mater.* 151, 811–820.
- MDC Sdn. Bhd., 2006. *Laws of Malaysia—Environmental Quality Act 1974 and Regulations*, sixteenth ed. Malaysia, Kuala Lumpur.
- Mohapatra, M., Anand, S., Mishra, B.K., Giles, D.E., Singh, P., 2009. Review of fluoride removal from drinking water. *J. Environ. Manage.* 91, 67–77.
- Montgomery, D.C., 2006. In: *Design and Analysis of Experiments*, fifth ed. Wiley India (P) Ltd., New Delhi.
- Tatsi, A.A., Zouboulis, A.I., Matis, K.A., Samaras, P., 2003. Coagulation–flocculation pretreatment of sanitary landfill leachates. *Chemosphere* 53, 737–744.
- Theriault, E.J., McNamee, P.D., 1936. Adsorption by activated sludge. *Ind. Eng. Chem.* 28 (1), 79–82.
- Thurman, E.M., Malcolm, R.L., 1981. Preparative isolation of aquatic humic substances. *Environ. Sci. Technol.* 15, 463–466.
- Umapiya, R., Shrihari, S., 2010. *Pathogen Removal Under the Influence of Iron*, vol. 72. World Academic of Science, Engineering and Technology, pp. 851–855.
- Vimonses, V., Jin, B., Chow, C.W.K., Saint, C., 2009. Enhancing removal efficiency of anionic dye by combination and calcination of clay materials and calcium hydroxide. *J. Hazard. Mater.* 171 (1–3), 941–947.
- Wang, Z.P., Zhang, Z., Lin, Y.J., Deng, N.S., Tao, T., Zhuo, K., 2002. Landfill leachate treatment by a coagulation–photooxidation process. *J. Hazard. Mater.* 95 (1/2), 153–159.
- Wang, T.H., Li, M.H., Yeh, W.C., Wei, Y.Y., Teng, S.P., 2008. Removal of cesium ions from aqueous solution by adsorption onto local Taiwan lateritic. *J. Hazard. Mater.* 160, 638–642.
- Yu, X.H., Zhu, L.J., Guo, B.W., He, S.Y., 2008. Adsorption of mercury on lateritic from Guizhou Province, China. *J. Environ. Sci.* 20, 1328–1334.