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Characterization and Chemical Treatment of Taman Beringin Landfill Leachate

Nor Asikir M. and **Agamuthu, P.***

Institute of Biological Sciences, Faculty of Science, University of Malaya, 50603 Kuala Lumpur, Malaysia
nor_asikir@perdana.um.edu.my
*agamuthu@um.edu.my (corresponding author)
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ABSTRACT Characterization of landfill leachate from Taman Beringin landfill, Kuala Lumpur, Malaysia was carried out immediately after sample collection. Subsequently, it was treated using alum, ferric chloride ($FeCl_3$), Fenton's reagent and polyaluminium chloride (PAC) individually. Raw leachate contained COD (5400 mg/L), BOD (1900 mg/L), NH_3-N (3500 mg/L), sulphide (68 mg/L), chloride (260 mg/L) and heavy metals such as Pb (3.38 mg/L), Cu (0.48 mg/L), Zn (4.5 mg/L) and Fe (35.8 mg/L). All these parameters were above the EQA 1974 (Standard A and B) limit. At optimal ratio of 1:6 (Fe to H_2O_2), with rapid and slow mixing at 100 - 120 rpm and 30 - 40 rpm, respectively Fenton's reagent was able to reduce about 62% of COD, 58% of BOD, 65% of NH_3-N, 100% of Pb, Cu and Zn, 95% of Al and 82% of Fe. PAC, also at optimal concentration of 0.4g/500 ml, and at pH of 7 with rapid and slow mixing at 100 - 120 rpm and 30 - 40 rpm, was capable of reducing COD and BOD by 59% and 55% respectively. The reduction of NH_3-N, Pb, Al, Fe, Cu and Zn ranged from 51% to 100%. At the optimum concentration of 5g/500ml, alum was capable of reducing about 53% of COD, 49% of BOD, 63% of NH_3-N, 99% of Pb, 100% of Cu, 91% of Al, 100% of Zn and 81% of Fe. Finally, $FeCl_3$, at optimum concentration, pH and mixing speed, reduced COD by 38% while BOD was reduced by 42%. The reduction of NH_3-N, Pb, Al, Zn and Fe ranged from 41% to 85%. Fenton's reagent was most effective in reducing most of the above parameters compared to PAC, alum and $FeCl_3$. Some parameters such as COD, BOD, NH_3-N, Al and Fe still exceeded the EQA 1974 (Standard A and B), although Fenton's reagent was able to reduce Pb, Cu and Zn to below the EQA Standard.

ABSTRAK Pencirian air larut resapan dilakukan sebaik sahaja sampel diambil dari tapak perlupusan sampah Taman Beringin, Kuala Lumpur. Kemudian, ia akan dirawat menggunakan alum, ferik klorida, reagen Fenton dan polialuminium klorida. Air larut resapan mempunyai kandungan COD 5400 mg/L, BOD (1900 mg/L), NH_3-N (3500 mg/L), sulfida (68 mg/L), klorida (260 mg/L) dan logam berat seperti Pb (3.38 mg/L), Cu (0.48 mg/L), Zn (4.5 mg/L) dan Fe (35.8 mg/L). Kesemua parameter ini melebihi had EQA 1974 (Standard A dan B). Pada nisbah optimal, 1:6 (nisbah Fe kepada H_2O_2), dengan pengacauan perlahan dan laju pada 100 - 120 rpm dan 30 - 40 rpm setiap satunya, reagen Fenton berupaya mengurangkan 62% COD, 58% BOD, 65% NH_3-N, 100% Pb, Cu dan Zn, 95% Al dan 82% Fe. PAC, pada kepekatan 0.4g/500 ml, dan pH 7 dengan pengacauan perlahan dan laju pada 100 - 120 rpm dan 30 - 40 rpm, berupaya mengurangkan COD dan BOD sebanyak 59% dan 55%. Pengurangan NH_3-N, Pb, Al, Fe, Cu dan Zn adalah dalam julat 51% ke 100%. Pada kepekatan 5g/500ml, alum berupaya mengurangkan COD kepada 53%, 49% BOD, 63% NH_3-N, 99% Pb, 100% Cu, 91% Al, 100% Zn dan 81% Fe. Sementara itu, $FeCl_3$, pada kepekatan, pH dan kadar pengacauan optimal,berupaya mengurangkan COD sebanyak 38%, BOD 42%, NH_3-N, Pb, Al, Zn dan Fe dari 41% ke 85%. Melalui kejian ini, didapati, reagen Fenton adalah reagen yang paling berkesan untuk menurunkan bacaan parameter-parameter berbanding PAC, alum and $FeCl_3$. Setengah parameters seperti COD, BOD, NH_3-N, Al dan Fe masih melebihi EQA 1974 (Standard A dan B), walaupun reagen Fenton berupaya mengurangkan nilai-nilai Pb, Cu dan Zn di bawah had yang ditetapkan.

(Landfill leachate, characterization, coagulation, flocculation, Fenton's reagent, polyaluminium chloride [PAC], alum, $FeCl_3$)

INTRODUCTION

In Malaysia, there are 144 landfills which receive about 8 million tonnes/yr of municipal solid waste (MSW). The urban population, which is about 50% of the total population in West Malaysia, generates about 3 million tonnes of domestic refuse per year. About 60 - 75% of MSW in Kuala Lumpur is organic, putrescible and also contains high plastic components (10 - 16%). Leachate from rural areas has a COD of 2 - 3 times lower when compared to the leachate from landfills in Kuala Lumpur [1] and this lower COD is due to the lower amount of putrecible waste disposed in rural areas [2]

This study focused on leachate generated from the Taman Beringin Landfill, Kuala Lumpur which received an average of 2000 metric tons of MSW daily. Aerators were used to treat leachate before it was released into the Jinjang River. Taman Beringin landfill operated for nine years before it was closed in 2004. In 2002, transfer station was operated using post-land compaction method. The function of this station was as transit area before the compacted waste was sent to Bukit Tagar landfill. This station could receive 1300 - 1400 tonnes of MSW daily.

A wide range of treatment options have been utilized to treat leachate. Biological processes such as activated sludge and aerated lagoons are the most common leachate treatment to remove biodegradable organic fractions [2]. Ammonium ion concentrations can be reduced with microbiological treatment via nitrification and denitrification processes [3]. However, the removal of COD during these processes is fairly low. The coagulation process generally employs several steps. It involves the addition of coagulant into the effluent followed by rapid and high intensity mixing.

The objective of mixing is to obtain intimate mixing of the coagulant with the wastewater which increased the effectiveness of particle destabilization and initiates coagulation. The subsequent stage involved the occurrence of flocculation for a period of 30 minutes. In the latter stage, the suspension is stirred slowly to increase the possibility of contact between coagulating particles and to facilitate the development of large flocs. These flocs are then transferred to a clarification basin in which they settle and are removed from the bottom, while the clarified effluent overflows [4].

PAC is a polymer and its usage is becoming popular for heavy metal removal. PAC hydrolyzes with great ease as compared to alum, emitting polyhydroxides with long molecular chains and greater electrical charges in the solution, thus contributing to maximize the physical action of the flocculation. Better coagulation is obtained with PAC, as compared to alum for medium and high turbidity waters. Floc formation with PAC is quite rapid. The sludge produced by PAC is more compact than that produced by alum. PAC is an effective and useful substitute for solid alum, which is conventionally used as a coagulant in most of the water treatment plants. It can cause rapid coagulation of water at different turbidities, producing less sludge and leaves less residual aluminium [4].

Fenton's reagent is a mixture of hydrogen peroxide (H_2O_2) and ferrous iron salts that reacts to form hydroxyl radicals (HO), ferric iron (Fe^{3+}), hydroperoxyl radicals (HO_2) and superoxide radicals (O_2). The reagents for H_2O_2 cracking are easy to handle and the process is environmentally friendly since the final decay products (water and oxygen) introduce no further pollution. In recent years, studies using Fenton chemical oxidation have been successful in treating various types of wastewater that effectively reduced COD, colour [5] and several pollutants such as aromatic amines [4], chlorinated aromatics [6] and 2,4 – dichlorophenol [7]. Wanpeng *et al.* [8] also found that wastewater pre-treated with Fenton reagents had improved biodegradability and efficiency of coagulation.

The aim of this study is to investigate the effect of reagent concentration, pH and mixing speed of Fenton's reagent, PAC, alum and $FeCl_3$ that could give the maximum pollution reduction in landfill leachate.

MATERIALS AND METHODS

Leachate characterization

Raw leachate collected from Taman Beringin landfill was analyzed to determine the characteristics of the leachate. The studies include:

i. pH and conductivity

These were measured using a pH and conductivity probe (Hanna Model, No. 8033).

ii. Total suspended solid (TSS)

TSS was determined with spectrophotometer, HACH Model (DR/4000)

iii. Alkalinity, hardness, chloride and sulphate

These were determined using Hanna Instruments HI 4817 Test Kit

iv. BOD_5, COD, TOC, Total-N and heavy metals were analysed according to Standard Methods [9]. TOC was analyzed using TOC analyser (Shimadzu 5000 Model) while heavy metals were determined using digested leachate with Inductive Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) Model Baird 2000.

Jar test Trials

A six-paddle flocculator from Stuart Scientific (Flocculator SW1) with a capacity of 500 ml was used. The samples were coagulated with different dosages, pH and mixing speed using Fenton's reagent, PAC, alum and $FeCl_3$. Chemical oxidation with the addition of Fenton's reagent consisting Fe to H_2O_2 ratio ranging from 1:2 to 1:8, the concentrations of PAC ranged from 0.2g/500 ml to 1g/500 ml, while alum and $FeCl_3$ were tested using concentrations of 3g/500 ml to 7g/500 ml. These were performed to get the optimum concentration of coagulants and flocculants. After establishing the optimum concentration, pH was tested at the range of 3 to 8 for all coagulants and flocculants. After determining the optimum pH, the mixing speed study was carried out. In this study, rapid mixing speed was fixed at 100 – 120 rpm based on earlier studies[10,11,12]. For all coagulants and flocculants, the slow mixing was conducted at the 20 - 30 rpm to 60 - 70 rpm. After settlement of the sludge, the supernatant was collected and was evaluated for BOD, COD, NH_3-N and heavy metal content.

RESULTS AND DISCUSSION

The waste type disposed into a landfill is one of the main factors that influenced the composition of the leachate formed [1]. Table 1 shows the characteristics of raw leachate from Taman Beringin landfill. The sample contained high concentration of COD (5400 mg/L), BOD (1900 mg/L), NH_3-N (3500 mg/L), chloride (260 mg/L), sulphide (68 mg/L), Pb (3.38 mg/L), Cu (0.48 mg/L), Al (35.8 mg/L), Zn (4.5 mg/L) and Fe (35.8 mg/L) and all parameters measured except Bo (0.25 mg/L) and Ni (0.01 mg/L) exceeded the regulatory standard limit of EQA 1974 (Standard A and B) [13]. The low values of Bo and Ni were probably due to the removal of the elements by ion exchange reactions as leachate travels through the soil [14].

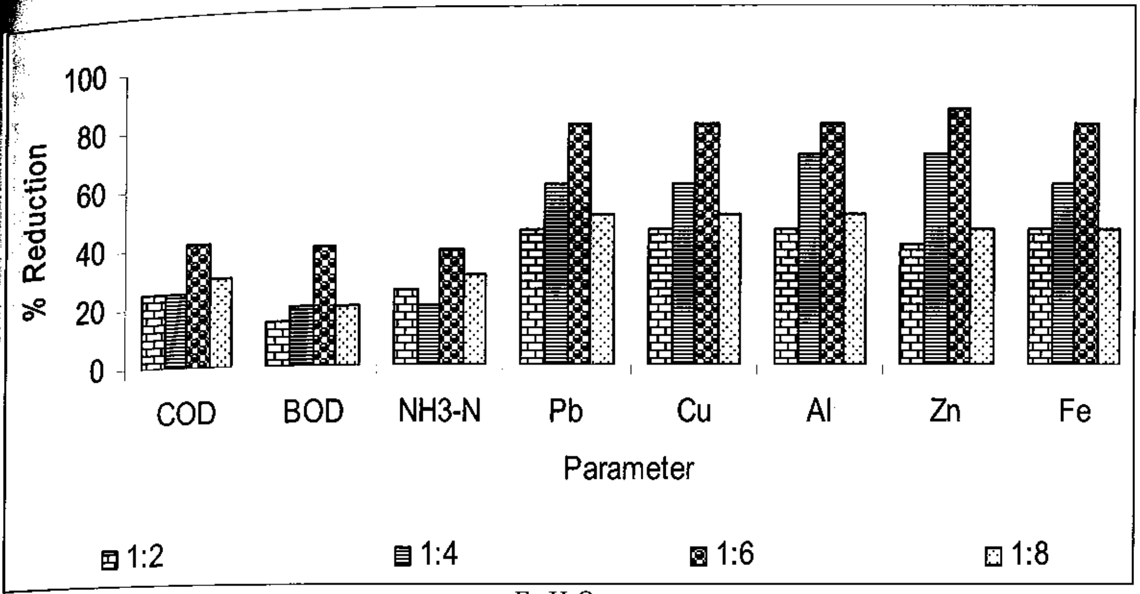
The age of landfill, limitation of solid generation and thus the degree of solid waste stabilization have a significant effect on the composition of leachate [15]. Reported data shows that Malaysian landfill leachate generally contains high quantities of Na, K and chloride. The BOD_5/COD ratio of raw leachate was approximately 0.4 and according to Tchobanoglous *et al.* [14], the ratio of 0.4 to 0.6 indicates that the organic matter in the leachate is readily biodegradable. High concentration of BOD_5 and COD were mainly from organic waste. Heavy metal pollutants are normally contributed by industrial waste. The high salt concentration in leachate could be attributed to the large quantity of garbage in the municipal waste. The characteristics of the garbage in turn reflect that the food waste disposed in Malaysia is of high salt concentration [1].

Figure 1 shows that maximum removal of pollutants was at the ratio of Fe to H_2O_2, 1:6. Referring to Figure 2, PAC was most effective in removing pollutants at 0.4g/500 ml. The optimum concentrations of alum and $FeCl_3$ in reducing the percentage of pollutants were 5 g/500 ml and 4 g/500 ml respectively (see Figures 3 and 4). Figures 5 – 8 show the effects of pH on various parameters after treatment with Fenton's reagent, PAC, alum and $FeCl_3$. Figure 5 shows maximum removal of pollutants using Fenton's reagent was at pH 4 while the minimum removal was at pH 7. At pH 4, Fenton's reagent was effective in removing 100% of Pb, Cu and Zn. This might be due to greater stability of hydrogen peroxide and ferrous ions in acidic medium [16]. At pH 4, Fenton's reagent was effective in removing 100% of Pb, Cu and Zn. This is true according to Lin and Lo [5], who stated that a good working condition for Fenton's reagent is at pH 3 - 6. The drop in efficiency at higher pH is attributed to the transition of iron from a hydrated ferrous ion to colloidal ferric species and the fragmenting of organic materials into organic acids [16].

Table 1. Characteristics of raw leachate from Taman Beringin Landfill compared to Environmental Quality Act (EQA) 1974 Standard A and B [13].

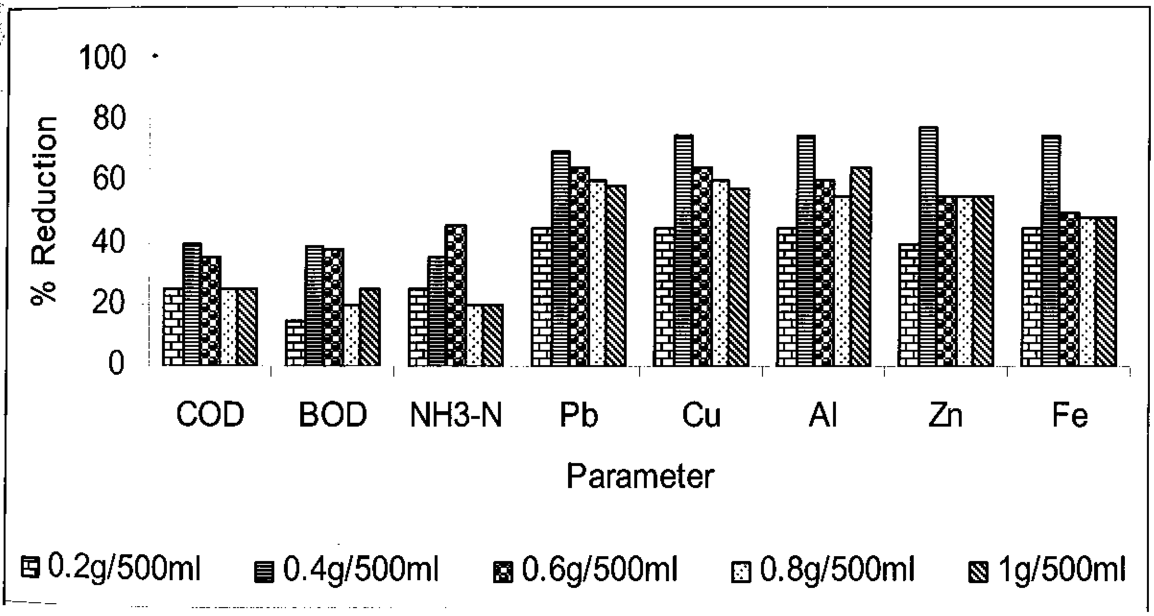
PARAMETER	UNIT	TAMAN BERINGIN LANDFILL LEACHATE	EQA 1974 STANDARD[1]	
			A	B
Temperature	^0C	26^0C	40	40
pH		8.75	6.0 - 9.0	5.5 - 9.0
BOD_5 at 20^0C	mg/L	**1910**	20	50
COD	mg/L	**5380**	50	100
Total Solid, TS	mg/L	13 650	N.A.	N.A.
NH_3-N	mg/L	**3500**	50	100
Turbidity	NTU	6900	N.A.	N.A.
Total Alkalinity	mg/L	5210	N.A.	N.A.
Hardness ($CaCO_3$)	mg/L	930	N.A.	N.A.
Conductivity	uS	35.4	N.A.	N.A.
Salinity	mg/L	7.05	N.A.	N.A.
Cadmium (as Cd)	mg/L	**0.2**	0.01	0.02
Arsenic (as As)	mg/L	**ND**	0.05	0.10
Lead (as Pb)	mg/L	**3.38**	0.01	0.5
Chromium (as Cr^{6+})	mg/L	**0.7**	0.05	0.05
Copper (as Cu)	mg/L	**0.48**	0.2	1.0
Aluminium (as Al)	mg/L	**29.3**	NA	NA
Nickel (as Ni)	mg/L	0.01	0.2	1.0
Zinc (as Zn)	mg/L	**4.5**	1.0	1.0
Boron (as B)	mg/L	0.25	1.0	4.0
Iron (as Fe)	mg/L	**18.53**	1.0	5.0
Chloride	mg/L	**262**	1.0	2.0
Sulphide	mg/L	**68**	0.50	0.50
Total-P	mg/L	25	N.A.	N.A.
Oil & Grease	mg/L	ND	N.D.	10.0

[1]EQA (1974), N.A. = Not Available, N.D. = Not Detected
Bold – indicates concentrations higher than EQA 1974 standard limit



$Fe:H_2O_2$

Figure 1. Effect of various concentrations of Fenton's reagent ($Fe:H_2O_2$) on the removal of pollution parameters in the leachate



PAC concentration

Figure 2. Effect of various concentrations of PAC on the removal of pollution parameters in the leachate

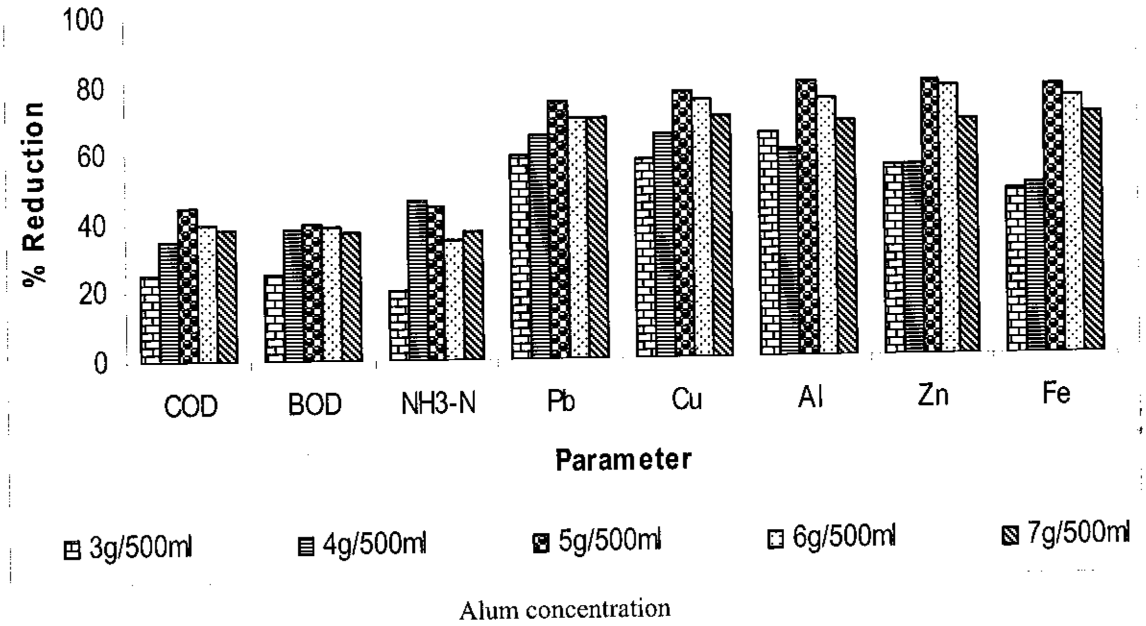


Figure 3. Effect of various concentrations of alum on the removal of pollution parameters in the leachate

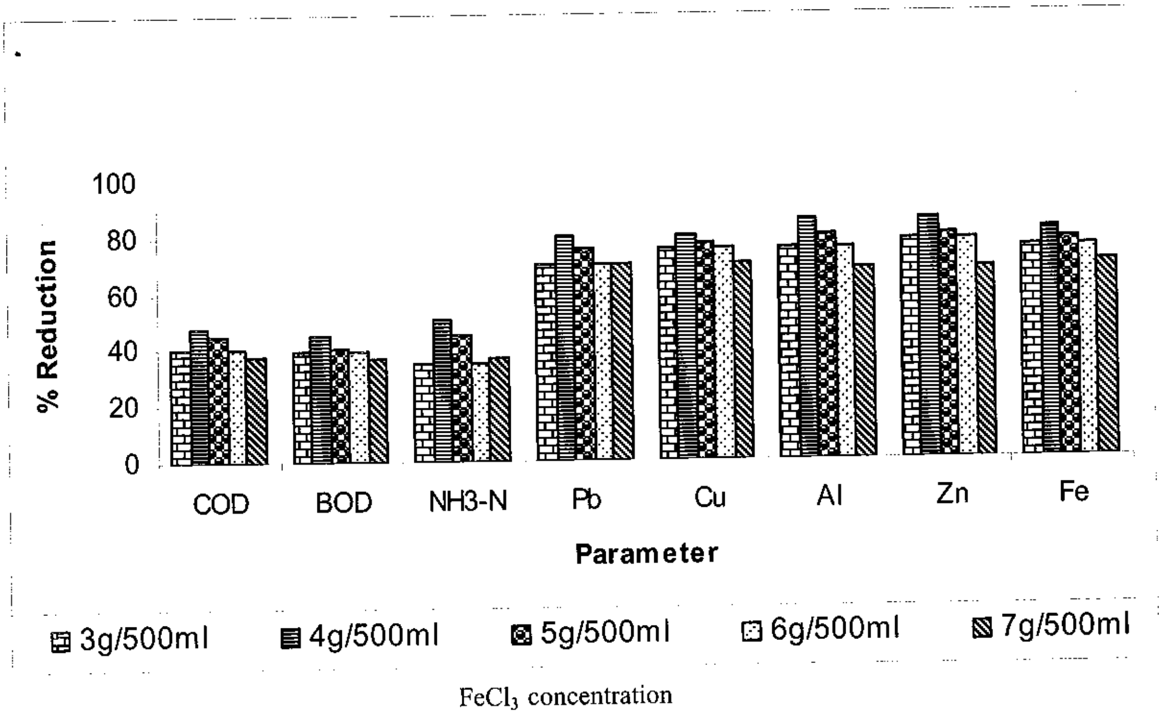


Figure 4. Effect of various concentrations of $FeCl_3$ on the removal of pollution parameters in the leachate

Figure 6 shows the optimum pH for PAC was at pH 7. Figures 7 and 8 show the optimum pH in reducing pollutants by alum and $FeCl_3$ were 4 and 7, respectively. The optimal pH value is dependent on the leachate characteristics, and it is normally about 4 - 6 for alum and 7 - 9 for ferric chloride [17]. The results of Briley and Krappe [18] showed that ferric sulphate hydrolysis products neutralized surface charges less than alum hydrolysis products when equimolar coagulant doses were added at a given coagulation pH. Thus, poorer charge neutralization may also help explain the lower heavy metal removal efficiency with $FeCl_3$ than alum at equimolar doses.

Figures 9 – 12 show the effect of mixing speed on various pollution parameters after treatment with Fenton's reagent, PAC, alum and $FeCl_3$. Rapid mixing ensures the total mixing of the coagulant in the solution whereas slow mixing causes the agglomeration of the flocs produced during the rapid mixing. At optimum concentration of Fenton's reagent at a ratio of Fe to H_2O_2 (1:6), with rapid mixing at 100 – 120 rpm and slow mixing from 20 - 30 rpm to 60 - 70 rpm, Fenton's reagent was capable of reducing 61% of COD as compared to PAC, alum and $FeCl_3$ which reduced 59%, 53% and 38% respectively. The reduction of BOD by Fenton's reagent, PAC, alum and $FeCl_3$ were 58%, 55%, 49% and 42% accordingly. For NH_3-N, the percentage of reduction was 65% with Fenton's reagent, 59% with PAC, 63% using alum and 41% using $FeCl_3$. Fenton's reagent and PAC gave complete removal of Pb, Cu and Zn, while alum gave complete removal of Pb only. For Fe concentration, PAC showed higher reduction percentage (87%) as compared to Fenton's reagent (82%) This can be caused by the existence of Fe ion in the ferrous sulphate components that would contribute towards higher value of Fe in the supernatant [16].

Alum reduced about 99% of Pb, 100% of Cu, 99% of Zn, 81% of Fe and 91% of Al as compared to $FeCl_3$ which reduced 78% of Pb, 80% of Cu, 74% of Zn, 70% of Fe and 80% of Al. When compared with $FeCl_3$, alum was more effective in reducing the pollutants parameter. One possible explanation for better performance of alum is that sulphate accelerated the precipitation of aluminium hydroxide [19], yielding a larger floc volume fraction. In the absence of sulphate addition from the coagulant, a smaller floc volume fraction may have resulted with ferric chloride at equimolar metal ion doses because cationic hydrolysis products irreversibly reacted with reactive sites of particles, algae and dissolved organic matter. Consequently, the reacted fraction of the coagulant would not have been available for precipitate formation [17].

Comparative optimum conditions are given in Table 2 which shows that the concentration, pH and other parameters differ for each chemical.

Table 2. Summary of the optimum conditions for use of coagulants/flocculants

	FENTON'S REAGENT ($Fe : H_2O_2$)	**PAC**	**ALUM**	**$FeCl_3$**
Concentration	1:6 (w/w)	0.4g/500 ml	5g/500 ml	4g/500 ml
pH	4	7	4	7
Slow mixing speed (rpm)	30 - 40	30 - 40	40 - 50	40 - 50

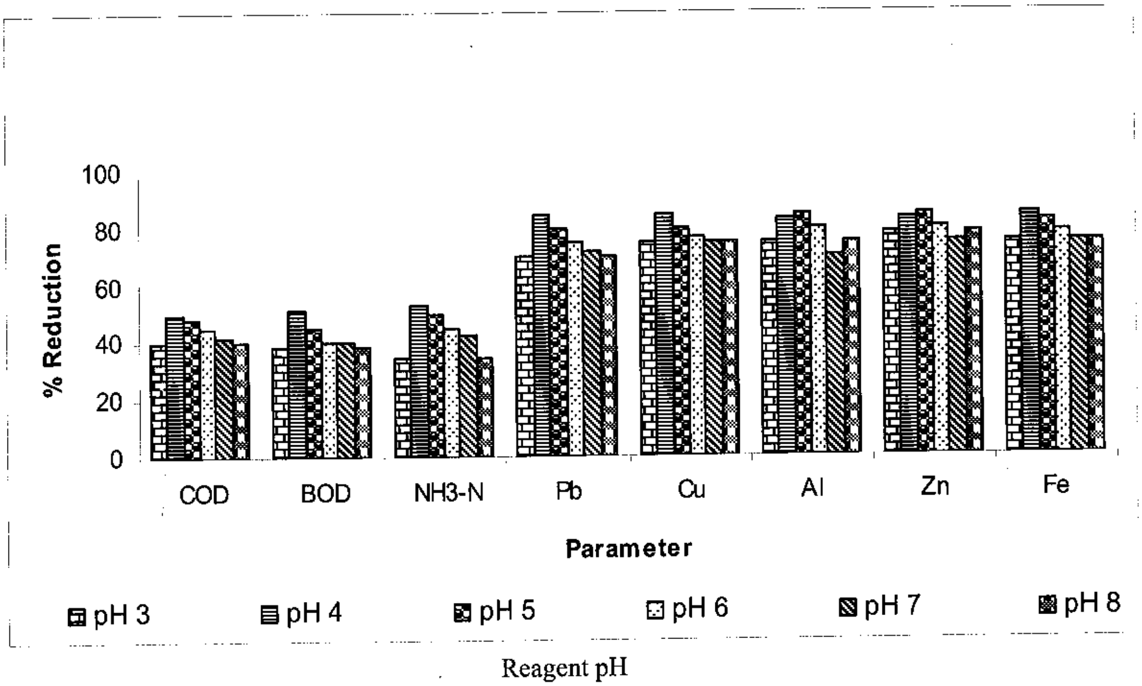


Figure 5. Effect of pH on Fenton's reagent to reduce the pollution parameters in the leachate

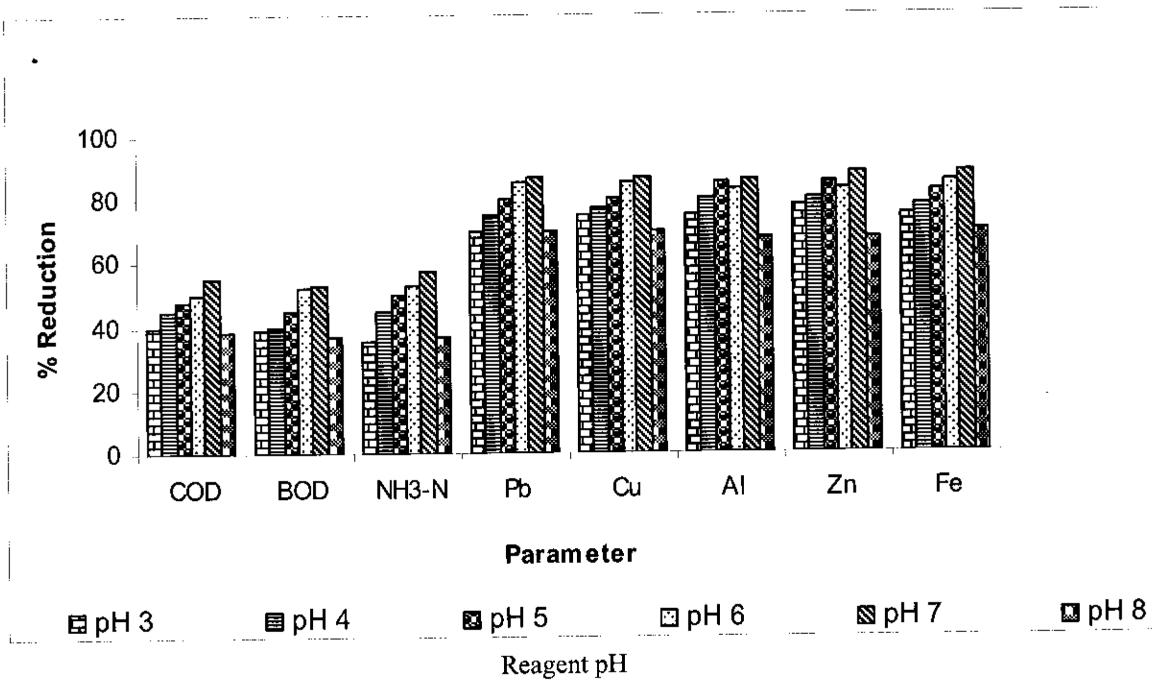


Figure 6. Effect of pH on PAC on the removal of pollution parameters in the leachate

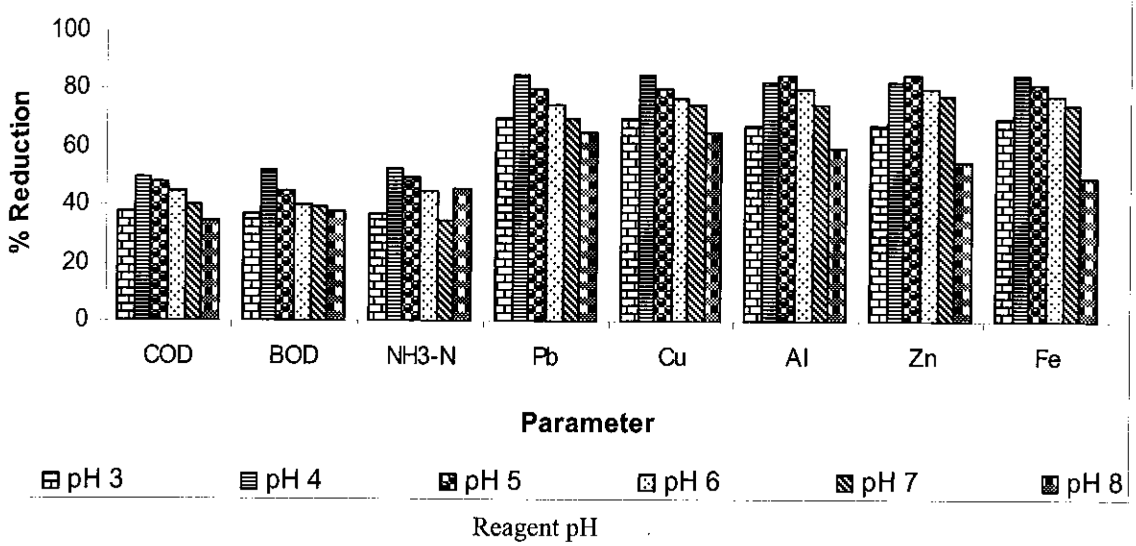


Figure 7. Effect of pH on alum on reduction of pollution parameters in the leachate

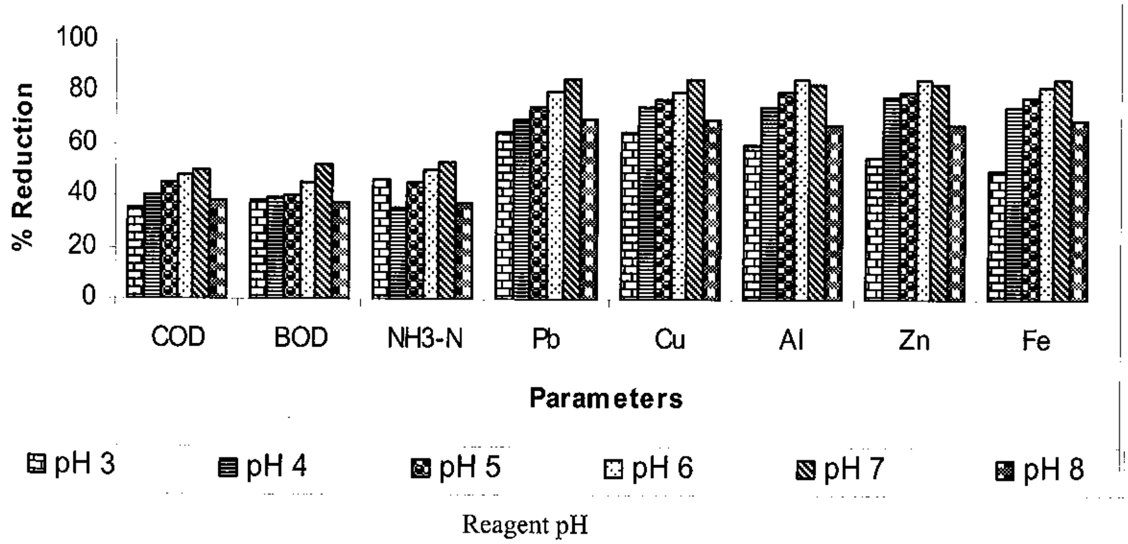


Figure 8. Effect of pH on $FeCl_3$ on the removal of pollution parameters in the leachate

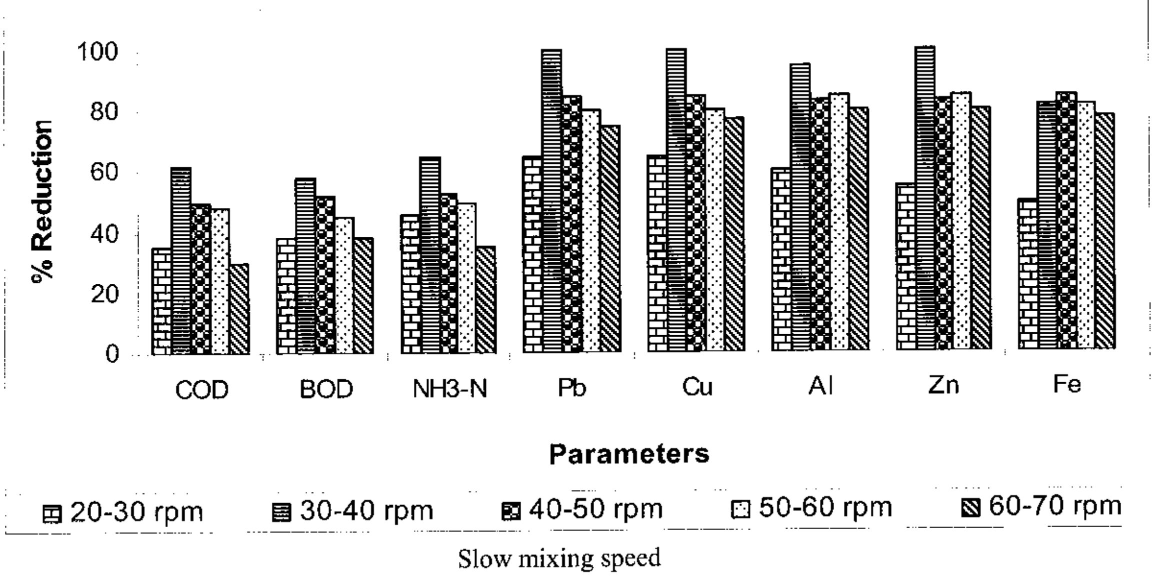


Figure 9. Effect of slow mixing of Fenton's reagent on the removal of pollution parameters in the leachate

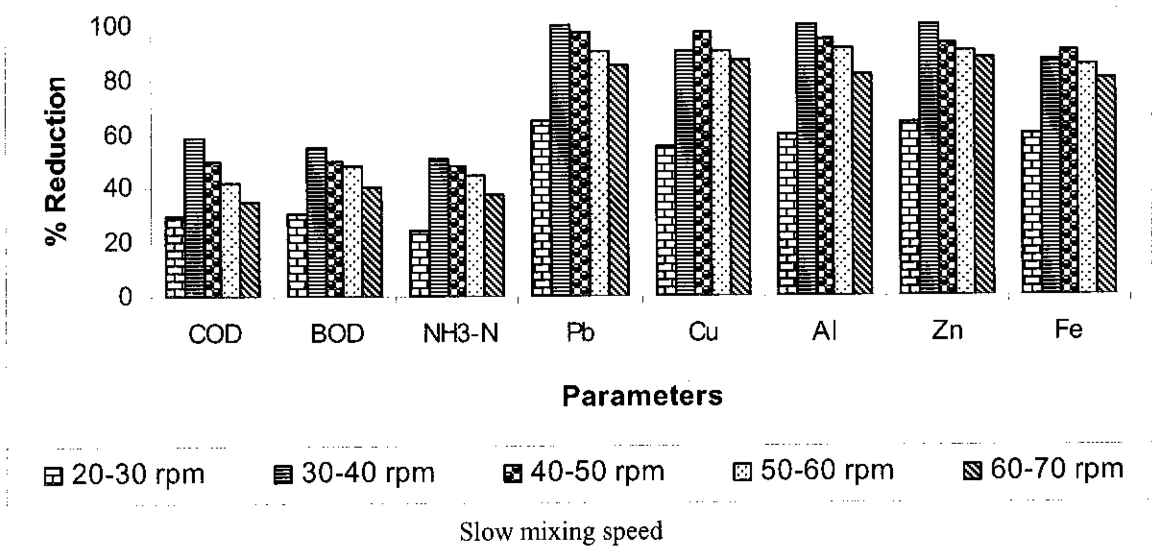
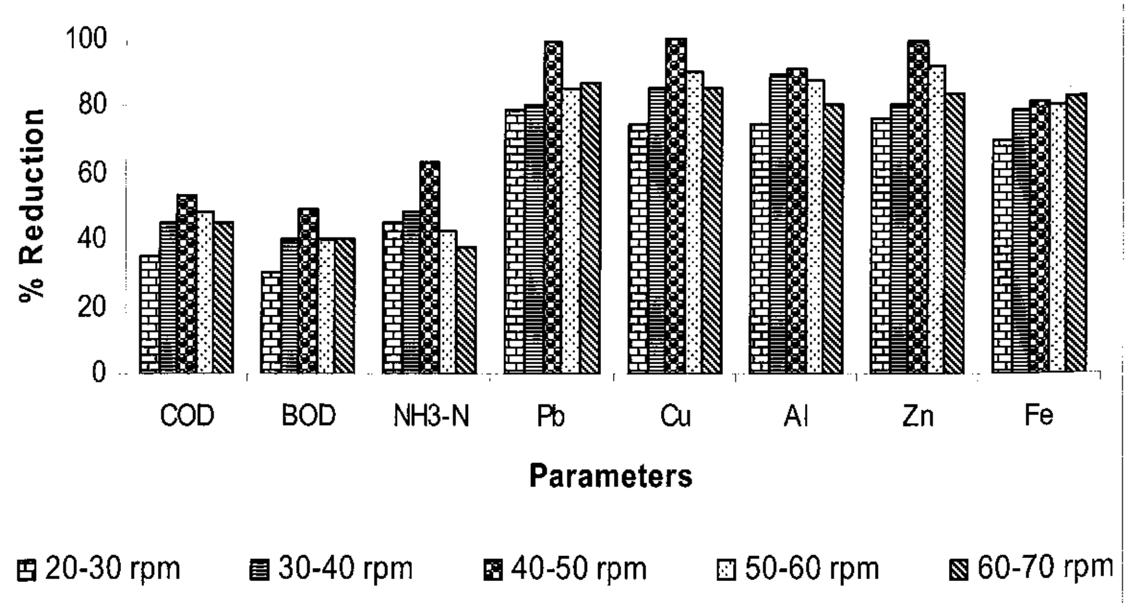
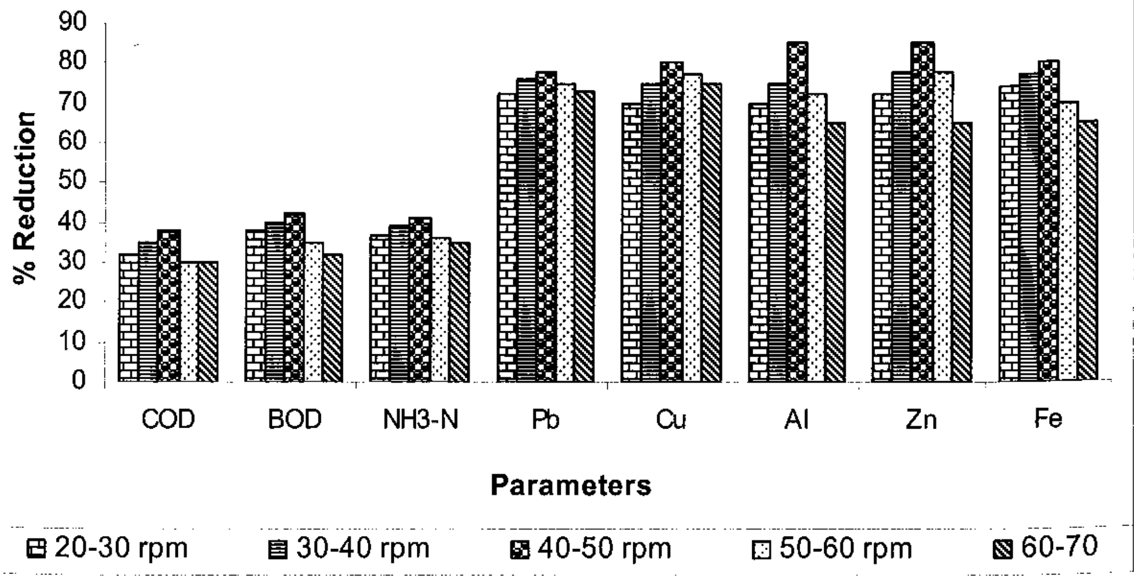


Figure 10. Effect of slow mixing of PAC on the removal of pollution parameters in the leachate



Slow mixing speed

Figure 11. Effect of slow mixing of alum on the removal of pollution parameters in the leachate



Slow mixing speed

Figure 12. Effect of slow mixing of $FeCl_3$ on the removal of pollution parameters in the leachate

CONCLUSION

From the characterization study, it can be concluded that leachate from the Taman Beringin landfill contains high amounts of COD, BOD, NH_3-N, sulphide, chloride and heavy metals such as Pb, Cu, Zn, Fe and Al. In general, the characteristics of this landfill leachate were found to be typical of other landfill leachate in the Klang Valley. All the parameters tested were above the EQA 1974 (Standard A and B) [13] limit. All coagulants and flocculants (Fenton's reagent, PAC, alum, $FeCl_3$) were able to cause significant reduction of COD, BOD, Pb, Cu, Zn. However, Fenton's reagent was the most effective coagulant/flocculant. The optimum activity of Fenton's reagent was at Fe: H_2O_2 ratio of 1:6 (w/w), at pH 4, while for PAC, the optimum concentration was at 0.4g/500ml with optimum pH of 7. The optimum slow mixing for both Fenton's reagent and PAC was 30 – 40 rpm. Optimum reduction of pollution parameter by alum was at concentration 5 g/500 ml at pH 4, while the optimum reduction by $FeCl_3$ was at concentration of 4 g/500 ml at pH 7. The optimum slow stirring for both alum and $FeCl_3$ was at 40 – 50 rpm. Since some of the parameters, such as COD, BOD, NH_3-N, Al and Fe still exceeded the limit of Standard A and B of the EQA (1974) [13], there is a need for further treatment to meet the requirements.

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