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PRE-TREATMENT AMMONIA REMOVAL OF SCHEDULED WASTE LEACHATE WITH HYDRATED LIME AND CAUSTIC SODA

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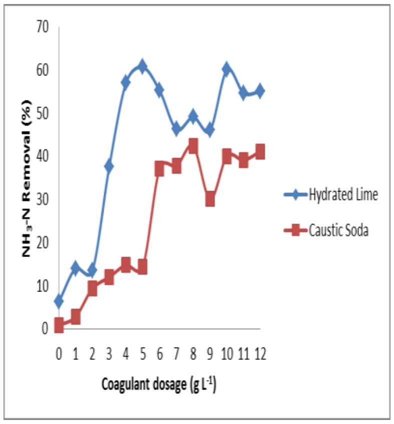
Graphical abstract



Abstract

Leachate is known to be hazardous wastewater in terms of its organic matter and ammonia content. Various methods have been studied extensively for the treatment of different types of leachate. However, scheduled waste leachate is rarely studied due to its complex characteristics and highly contaminated nature compared to other types of landfill leachates. In this study, hydrated lime ($Ca(OH)_2$) and caustic soda (NaOH) as softener agent were used for increasing the pH of the treatment process of scheduled waste leachate to remove ammonia-nitrogen (NH_3-N). Chemical oxygen demand (COD) and colour removal were also evaluated. Jar test experiments was performed using $Ca(OH)_2$ and NaOH at varying dosages ranging from nil to 12 g L^{-1}. A Historical Data Design (HDD) of Response Surface Methodology (RSM) was employed to optimize the parameters affecting NH_3-N, COD and colour removal efficiency. The optimal conditions obtained from desirable response were predicted at 5.9 g L^{-1} of $Ca(OH)_2$ dosage, where the maximum NH_3-N, COD and colour removal efficiency would be 49%, 18% and 66%, respectively. Meanwhile for NaOH, dosage of 6.4 g L^{-1} was predicted to remove NH_3-N, COD and colour up to 32%, 4% and 42%, respectively. The predicted values concurred with the experiments, in which $Ca(OH)_2$ successfully reducing NH_3-N, COD and colour up to 52 %, 17.5 % and 65 %, while NaOH could only remove NH_3-N, COD and colour by up to 35 %, 2 % and 49 %, respectively. The results obtained from this study suggest that hydrated lime ($Ca(OH)_2$), is more feasible to be used for the pre-treatment process of scheduled waste leachate as less dosage is required with higher removal efficiencies of NH_3-N, COD and colour. This study demonstrates that RSM was a reliable tool to predict the optimum dosage and suitable types of chemicals for the removal of NH_3-N, COD and colour from the available data.

Keywords: Ammonia-nitrogen, caustic soda, leachate, hydrated lime, scheduled waste

Abstrak

Air larut resap dikenali sebagai air sisa yang berbahaya dari segi kandungan bahan organik dan ammonia. Pelbagai kaedah telah dikaji secara meluas untuk merawat pelbagai jenis air larut resap. Walau bagaimanapun, air larut resap sisa terjadual jarang dikaji kerana ciri-ciri kompleks dan mengandungi bahan yang sangat tercemar berbanding air larut resap tapak pelupusan yang lain. Dalam kajian ini, kapur terhidrat ($Ca(OH)_2$) dan soda kaustik (NaOH) sebagai agen pelembut telah digunakan untuk meningkatkan pH proses rawatan air larut resap sisa terjadual untuk menyingkirkan ammonia - nitrogen (NH_3-N). Permintaan kimia oksigen (COD) dan penyingkiran warna juga telah dinilai. Eksperimen ujian jar telah dilakukan dengan menggunakan $Ca(OH)_2$ dan NaOH pada dos yang berbeza-beza antara sifar hingga 12 gL^{-1}. Rekabentuk data sejarah (HDD) melalui tindak balas permukaan metodologi (RSM) telah digunakan untuk mengoptimumkan parameter yang mempengaruhi kecekapan penyingkiran NH_3-N, COD dan warna. Keadaan optimum yang diperoleh dari tindak balas wajar telah diramalkan pada dos 5.9 g L^{-1} $Ca(OH)_2$, di mana kecekapan penyingkiran maksimum NH_3-N, COD dan warna diramalkan 49%, 18% dan 66%. Sementara itu, bagi soda kaustik (NaOH), dos 6.4 g L^{-1} telah diramalkan untuk menyingkirkan NH_3-N, COD dan warna sehingga 32 %, 4 % dan 42 %. Nilai ramalan menepati nilai eksperimen, di mana $Ca(OH)_2$ telah Berjaya menyingkirkan NH_3-N, COD dan warna sehingga 52 %, 17.5 % dan 65 %, sementara NaOH hanya boleh menyingkirkan NH_3-N, COD dan warna sehingga 35 %, 2 % dan 49 %. Keputusan yang diperolehi daripada kajian ini mencadangkan bahawa kapur terhidrat ($Ca(OH)_2$), adalah lebih sesuai untuk digunakan bagi proses pra-rawatan air larut resap sisa terjadual kerana dos yang kurang diperlukan dengan kecekapan penyingkiran NH_3-N, COD dan warna yang lebih tinggi. Kajian ini menunjukkan bahawa RSM adalah alat yang boleh dipercayai untuk meramalkan dos yang optimum dan jenis bahan kimia yang sesuai untuk penyingkiran NH_3-N, COD dan warna dari data yang ada.

Kata Kunci: Ammonia-nitrogen, soda kaustik, air larut resap, kapur terhidrat, sisa berjadual

1.0 INTRODUCTION

Malaysia is one of the developing countries in this world that experienced rapid economic growth, urbanisation and industrialisation. With such rapid development in Malaysia, more solid and industrial waste is generated daily. Numerous waste disposals methods had been widely practiced such as open dumping, sanitary landfill, composting, incineration, dumping, milling, hog feeding, grinding, reduction and anaerobic digestion. However, landfilling has been the most popular waste disposal method in Malaysia due to its low cost and simplicity [1]. Even though landfilling is the most widely adopted method of waste disposal, the production of highly polluted wastewater called leachate may pose a great threat to the environment [2].

Leachate is a concentrated liquid with several characteristics which make it difficult to treat. It is produced from the percolation of precipitation through the waste deposited in a landfill. Generally, leachate consists of biodegradable organic matter, recalcitrant organic matter (such as humic substances), heavy metals, inorganic salts as well as a high concentration of ammonia-nitrogen [3-6]. Leachate pollutes the soil and infiltrates into the ground resulting in groundwater contamination [3]. With the advancement of modern technology, there has been a swift increase in the variation and amount of wastes generated. This has led to an increasing complexity of components in the leachate, which has considerably increased the difficulty of leachate treatment.

Scientific literature documents many studies of leachate treatment from municipal solid waste (MSW) and sanitary landfill, but there is limited information on scheduled waste leachate [7, 8]. Scheduled waste landfill leachate is rarely studied due to its complex characteristics and hazardous materials compared to other leachate types. Leachate treatability is often dependent on its composition and the organic matter present in it [9]. Based on the actual leachate treatment plant facilities that have been commissioned, the treatment processes of leachate involve complex procedures and a high-cost of operation and maintenance. In fact, leachate takes many years to be stabilised even after the landfill has been closed upon reaching full storage capacity. Therefore, a proper leachate treatment technique which is efficient, cost-effective and environmental friendly is crucial.

Among the leachate constituents, the most problematic pollutant in the long term is ammonia and its concentration may vary up to several thousand mg L^{-1} [10, 11]. The release of leachate containing high level ammonia-nitrogen (NH_3-N) into water may enhance the growth of algae and aquatic plants, cause eutrophication in lakes, cause deterioration of the delicate balance of the ecosystem and toxicity of living organisms in surface waters if released without proper treatment [12, 13]. Besides, it may decrease the performance of conventional biological treatment systems [13]. Thus, removing high ammonia concentration from leachate has become a critical issue recently. In Malaysia, scheduled waste leachate is categorised under Environmental Quality (Industrial Effluent or Mixed Effluent) Regulations 2009, Fifth Schedule (Standard B) and the permissible limit standards of ammonia-nitrogen (NH_3-N) into the water bodies must not exceed 20 mgL^{-1} [14]. However, most of treatment plant facilities cannot meet the allowable limit standards as the concentration of NH_3-N in the treated effluent still remains high (>20 mgL^{-1}).

Leachate can be treated with biological, chemical and physical methods. Numerous methods have been studied in order to determine the most effective technique of ammonia removal from leachate, including chemical precipitation, magnetic field separation, adsorption, chemical oxidation, coagulation-flocculation and electrocoagulation (EC) [15]. Recently, chemically enhanced leachate treatment has created substantial interest, especially for the leachate that difficult to be treated by conventional biological treatment due to waste materials content in leachate which is toxic to biological growth. High concentration of total ammoniacal nitrogen (TAN) about 1000 mg L^{-1} may inhibit the activity of microorganisms, hence decreases the effectiveness of biological processes for leachate treatments [11]. Since it is difficult to achieve high and reliable ammonia and COD removal efficiencies with biological treatment alone, especially for stabilised leachate, the combination of both biological and physical-chemical techniques is generally practiced. Consequently, chemical pre-treatment could be introduced prior biological process treatment.

Among currently employed chemical unit processes in leachate treatment, coagulation-flocculation has received significant attention as it yields high pollutant removal efficiency. The coagulation-flocculation process is generally employed for the removal of non-biodegradable organic compounds, ammonia-nitrogen (NH_3–N) and heavy metals from landfill leachate due to its capability, simplicity of the process and inexpensive equipment requirements [7, 8, 16-18]. There are few factors that may influence the efficiency of the coagulation-flocculation process such as the pH, mixing speed and time, type and dosage of coagulants, temperature and retention time [19-23]. Coagulation is one of the vital steps in wastewater and leachate treatment. The most difficult part is to determine the dosage of chemicals that are necessary to be used for the treatment processes. Any faulty aspect of this process may cause a significant increase in the operating costs and an inability to meet the effluent quality objectives. Hence, the optimisation of the types and dosage of chemicals may significantly increase the process efficiency. Over the past few years, several authors have investigated the potential of few chemicals such as hydrated lime or Calcium Hydroxide ($Ca(OH)_2$), aluminium sulphate ($Al_2(SO_4)_2$, calcium carbonate ($CaCO_3$),magnesium hydroxide ($Mg\ (OH)_2$), Polyaluminium Chloride (PaCl) and sodium hydroxide or caustic soda (NaOH) for the treatment of most water, wastewater and leachate [9, 24-28].

In the treatment of municipal waste leachate and sanitary leachate, ammonia transfer rate is enhanced by converting most ammonium ions (NH_4^+) to a gaseous form (NH_3), normally at pH ranged from 10.5 to 12 [16, 29-30]. The addition of chemicals is crucial to increase the pH for optimum treatment processes. Out of the chemicals examined above, $Ca(OH)_2$ and NaOH were preferred due to its effectiveness in the removal of suspended solids, ammonia-nitrogen (NH_3-N), colour, heavy metals and other substances [7-9, 24-26].

Therefore, this study was performed using jar test experiments to evaluate the feasibility of $Ca(OH)_2$ and NaOH precipitation as a pre-treatment process and to find the optimum dosage used in removing ammonia-nitrogen (NH_3-N), chemical oxygen demand (COD) and colour of Scheduled Waste Landfill (SWL) leachate. The usage of $Ca(OH)_2$ and/or NaOH as pretreatment may facilitate their applicability and efficiency in removing NH_3-N for further treatment such as for air stripping. Finding the most suitable chemicals and its optimum dosage is crucial as this can reduce the cost of the overall treatment process and produce high effluent quality which is within the permissible limit standards.

2.0 METHODOLOGY

2.1 Leachate Collection and Characterization

Leachate samples were collected from a Scheduled Waste Landfill (SWL) facility in Peninsular Malaysia. The scheduled waste landfill started its operation in the year of 2000. The major constituents of the scheduled waste are the incinerated ashes of industrial and hazardous waste from factories, hospitals and others. The facility is equipped with a leachate collection pond. The collection and preservation of samples was performed in accordance with the Standard Methods for the Examination of Water and Wastewater [31]. The leachate samples were collected from the same collection point in a 25 L container, and stored at 4 °C. The samples were characterised in terms of temperature, pH, COD, BOD_5, colour, ammonia-nitrogen (NH_3-N), Total Suspended Solids (TSS), heavy metals, and others. Table 1 represents the general characteristics of the raw leachate from the Scheduled Waste Landfill (SWL) used.

2.2 Experimental Set-up

The hydrated lime used was $Ca(OH)_2$, supplied by scheduled waste facility. Caustic soda, NaOH was purchased (Merck, Germany). $Ca(OH)_2$ and NaOH were preferred based on their performance in the preliminary studies, which concluded that these chemicals was applicable and effective for removing NH_3-N from scheduled waste leachate [7-8]. Other studies also reported the effectiveness of these two chemicals in the removal of suspended solid, colour, heavy metals and other substances [24-26].

Coagulation studies were performed using a jar test apparatus, with six impellers equipped with six beakers (VELP-Scientifica, Model: JLT6, Italy). Each beaker containing 100 mL leachate was dosed with 0 gL^{-1} (as control) to 12 gL^{-1} of $Ca(OH)_2$, respectively [32]. The operating parameters were set at rapid mixing speed of 80 rpm for a period of 2 minutes, followed by a slow mixing speed of 30 rpm for 30 minutes with an additional 120 minutes of settling time [32-33]. The experiment was repeated with caustic soda (NaOH) and each of the experiments was carried out thrice [3].

After the settling period, supernatant was withdrawn from the beaker using a plastic pipette for further analysis. The pH was measured using a portable instrument (Hanna Instruments, Model: 991003, Romania). The ammonia-nitrogen (NH_3-N) in the sample was measured using Ammonia Ion Selective Electrode (HACH, Intellical). COD was determined using the colorimetric method (5220-D). Colour was measured as true colour based on APHA Platinum-Cobalt Standard Method using a DR 890 HACH colorimeter. Method Number 8025 reports the colour in platinum–cobalt (Pt–Co). The removal efficiency for the parameters was obtained using the following formula:

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

where C_i and C_f are the initial and final concentrations of the parameters measured in the leachate sample in mgL^{-1}, respectively [34].

2.3 Validation with Response Surface Methodology (RSM)

Response surface methodology (RSM) is a tool of accurate statistical methods in mathematic for optimization of one or more response variables according to change of several independent variables. The ammonia-nitrogen (NH_3-N), chemical oxygen demand (COD) and colour removal efficiency results reported earlier was used in RSM study. The input parameter is the type of chemical, whether hydrated lime ($Ca(OH)_2$) or caustic soda (NaOH) and dosage of chemical used ranging from 0 to 12 g L^{-1}. The responses of the experiment were NH_3-N, COD and colour removal efficiency (%). Historical data design (HDD) in response surface methodology (RSM) was used to analyse the interactions among the independent variables and further optimize operating parameters of NH_3-N, COD and colour removal efficiency from scheduled waste leachate. HDD was used for importing data that already exists. Statistical software package embedded in Design Expert 7.1.5 was used for the experimental data analysis and polynomial regression model was built [35].

3.0 RESULTS AND DISCUSSION

3.1 Characteristics of Scheduled Waste Leachate

Table 1 shows the characteristic of scheduled waste leachate (SWL). The average pH 10.0 was higher compared to the pH of other municipal leachates (pH 5 to 8) [36]. Sani *et al*. (2014) reported about similar pH value for scheduled waste leachate, that is, 9.92. pH value of leachate also depends on the landfill gas generation stage and the age of the landfill [37]. Generally, young leachate has pH value less than 6.5 while old landfill leachate has pH higher than 7.5 [36].

SWL contained a high amount of BOD_5 (2053 mgL^{-1}), COD (4127 mgL^{-1}), arsenic (13.01 mgL^{-1}), boron (29.30 mgL^{-1}), and NH_3-N (2267 mgL^{-1}) compared to other municipal leachates with BOD_5 (48-515 mgL^{-1}), COD (599-1788 mgL^{-1}), arsenic (0 mgL^{-1}), boron (not measured), and ammonia-nitrogen (503-1380 mgL^{-1}) [7, 35]. Similarly, previous studies on SWL also observed fairly high amount of BOD_5 (2873 mgL^{-1}), COD (4633 mgL^{-1}), and NH_3-N (2403 mgL^{-1}) [7].

The BOD_5 /COD ratio describes the degree of biodegradation and gives information about the landfill age. Generally, low BOD_5/COD ratio shows the high concentration of non-biodegradable organic compounds, which makes the leachate difficult to be biologically degraded. For the SWL, the BOD_5/COD ratio was 0.50. Scheduled waste landfill was considered as young (<5 years) since the BOD_5/COD ratio value was more than 0.3 [38]. The concentration of TSS was 313 mg/L which indicated the presence of organic and inorganic solids. A greater concentration of colour was mainly contributed by the dissolved organics. These organic compounds may be present in the form of recalcitrant material mainly composed of humic-like substances [32].

Ammonium indicates the main proportion of total nitrogen. The release of soluble nitrogen from waste into leachate last longer than soluble organic [2]. Therefore, as the age of the landfill increases, the ammonia nitrogen concentration will also increase due to fermentation and hydrolysis of nitrogenous fractions of biodegradable refuse substrates [36]. The stability of ammonia under anaerobic conditions makes it as a major long-term pollutant. Different types of landfills produced different ranged of ammonia concentration in leachates. The concentration may varied from tens or hundreds of mg L^{-1} to few thousands (10 000 mgL^{-1}) [2, 36]. Li and Zhao (2002) reported that in stabilized leachate, ammonia nitrogen might ranged between 3000 to 5000 mg L^{-1}. Previous studies reports that high ammonia-nitrogen is toxic to living organisms, promotes eutrophication and may disrupt the biological leachate treatment [16, 36]. Hence, scheduled waste leachate should be treated prior discharge into the environment.

Table 1 Characteristics of raw leachate from Scheduled Waste Landfill (SWL)

Parameter	Units	SWL leachate		DOE Standard*
		Range	Average	
pH Value	-	9.7-10.2	10.0	5.5-9.0
Biochemical Oxygen Demand (BOD_5)	mgL^{-1}	1610 - 2670	2053	50
Chemical Oxygen Demand (COD)	mgL^{-1}	3430 - 4610	4127	200
Total Suspended Solids (TSS)	mgL^{-1}	0 - 870	313	100
Copper	mgL^{-1}	0.18 – 1.04	0.55	1.0
Arsenic	mgL^{-1}	9.24 - 16.9	13.01	0.10
Boron	mgL^{-1}	27.2 – 31.0	29.30	4.0
Phenols (Total)	mgL^{-1}	0.730 – 8.08	3.26	1.0
Ammoniacal-Nitrogen	mgL^{-1}	1790 - 2570	2267	20
Colour (pH Natural)	ADMI	52-1590	1004	200
Colour (pH adjusted to 7.6)	ADMI	50-1550	913	200

***** DOE Standard: Environmental Quality (Industrial Effluent) Regulations 2009, Fifth Schedule (Standard B).

3.2 Effect of Dosage and Types of Chemicals on Ammonia-nitrogen (NH_3-N) Removal Efficiency

The results of Ammonia-nitrogen (NH_3-N) removal at various dosages of hydrated lime, $Ca(OH)_2$ and caustic soda (NaOH) was presented in Figure 1. The dosage of chemicals plays a vital role in the removal of target pollutants in coagulation-flocculation studies. An optimum dose of chemicals can be defined as a value above which there is no significant increase in removal efficiency with further addition of the chemicals [27]. In this study, the concentration of $Ca(OH)_2$ and NaOH ranged from 0 gL^{-1} to 12 gL^{-1}. The removal of NH_3-N (between 6.2 % and 0.8 %) occurred even without the addition of chemicals to the leachate. This was due to the release of ammonia as the sample was stirred in the experiment [7]. The formation of insoluble solid precipitates was observed during the treatment with $Ca(OH)_2$. However, no precipitates formed when NaOH was applied.

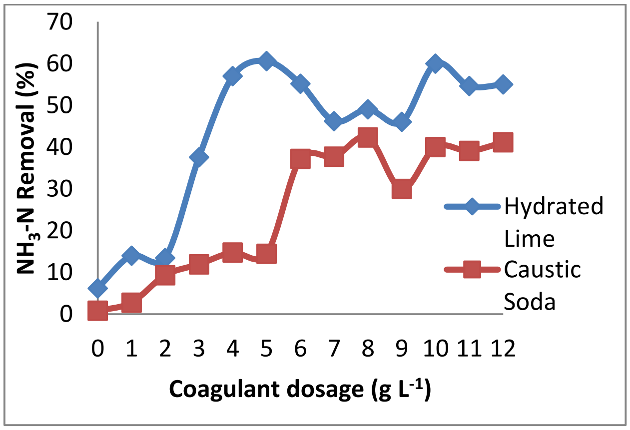


Figure 1 Removal of ammonia- nitrogen (NH_3-N) at various dosages of hydrated lime, Ca $(OH)_2$ and caustic soda (NaOH).

As depicted in Figure 1, the maximum removal of NH_3-N was observed at $Ca(OH)_2$ concentrations of 5 gL^{-1} and NaOH at 8 gL^{-1} with 60.6% and 42.3%, respectively . The study showed that less dosage of $Ca(OH)_2$ was needed (i.e 5 gL^{-1}) to obtain the optimum NH_3-N removal efficiency compared to NaOH (i.e 8 gL^{-1}). The pH value recorded at the optimum dosage of 5 gL^{-1} $Ca(OH)_2$ and 8 gL^{-1} NaOH were pH 12.17 and 12.83, respectively. In other study [7], it was reported that 4 gL^{-1} of $Ca(OH)_2$ dosage may remove up to 54% of NH_3-N at pH=12.39. Sani *et al*. (2014) remarked that the addition of $Ca(OH)_2$ of more than 6 gL^{-1} does not show any significant effect on NH_3-N removal, which was similar to the findings in this study. Further addition of $Ca(OH)_2$ than the optimum dosage showed a slight decrease in the NH_3-N removal due to restabilization of colloids and re-dispersion of the colloidal particles [27]. Another study by Nurul Hanira *et al*. (2015) found that the percentage of NH_3-N removal was relatively the same for both $Ca(OH)_2$ and NaOH which was up to 45% and 48%, with optimum dosage of 6 gL^{-1} and 8 gL^{-1}, respectively. The recorded pH for $Ca(OH)_2$ and NaOH was pH=12.40±0.02 and pH=12.83±0.02, respectively. Since there is a small difference in the removal of NH_3-N, $Ca(OH)_2$ is preferred for the pre-treatment process as it required less dosage [8].

Even though the same dosages of $Ca(OH)_2$ and NaOH were used, both have a different effect on the NH_3-N removal. The type and dosage of chemicals/coagulants have an influence on the efficiency of the coagulation-flocculation process [20]. NaOH has a wider range in pH compared to Ca $(OH)_2$ as excessive additions of NaOH and $Ca(OH)_2$ will result in the pH of the leachate reaching 14 and 12.5, respectively [39]. When $Ca(OH)_2$ alone is added as precipitant, the clarification will combine with all the free carbonic acid to produce calcium carbonate, which acts as the coagulant. $Ca(OH)_2$ also may react with sulphates in the wastewater producing hard precipitates. Meanwhile, the addition of NaOH is mainly to raise the pH. The high pH also promotes the precipitation of the metals hydroxide. However the presence of ammonia may interfere with copper precipitation [29]. Additionally, the optimisation based on other factors such as temperature and retention time, pH, mixing speed and time may significantly increase the effectiveness of the coagulation-flocculation process [40].

Ammonia can be removed from wastewater by volatilization of gaseous ammonia (NH_3). The rate of ammonia transfer is enhanced by converting most of ammonia to a gaseous form at high pH (10.5 to 11.5), by the addition of alkaline chemicals [29]. Ammonia in the aqueous phase is found in two equilibrium forms, ionic (NH_4^+) and the gaseous state (NH_3).The process of ammonia removal is based on the following Equation 2 [41].

$$NH^{4+}\ (aq) + OH^- \rightleftharpoons NH_3\ (g) + H_2O\ (aq) \qquad (2)$$

Based on Equation 2, ammonium ions (NH_4^+) and ammonia gas (NH_3) concentrations are dependent on the pH. When pH is low, ammonium and water are the dominant species. However when pH increases, the equation shifts to the right and increases the ammonia gas concentration. As $Ca(OH)_2$ and NaOH are alkaline, the addition of $Ca(OH)_2$ and NaOH increases the pH of the leachate. The increase of pH shifts the reaction to the right, producing ammonia gas. At pH 11, most ammonium ions are in the form of ammonia gas, thus resulting in a higher removal of ammonia [16]. Besides, higher removal efficiencies was observed in this study when using $Ca(OH)_2$ instead of NaOH. This is due to the higher number of hydroxide ion (OH^-) that $Ca(OH)_2$ possessed. 1 molecule of $Ca(OH)_2$ comprised of two OH^- ions, while one NaOH molecule only have one OH^- ion. This OH^- ion reacts with ammonium ion (NH_4^+) to produce ammonia gas (NH_3). As more OH^- ion reacts with NH_4^+ ion, more NH_3 gas will be produced, hence reducing the concentration of ammonia in aqueous phase (leachate). This explains why $Ca(OH)_2$ was more effective in removing NH_3-N than NaOH even less dosage was used.

3.3 Effect of Dosage and Types of Chemicals on Chemical Oxygen Demand (COD) Removal Efficiency

Chemical oxygen demand (COD) indicates the mass of oxygen consumed per litre of solution to determine the amount of organic pollutants found in wastewater. The result of COD removal was presented in Figure 2.

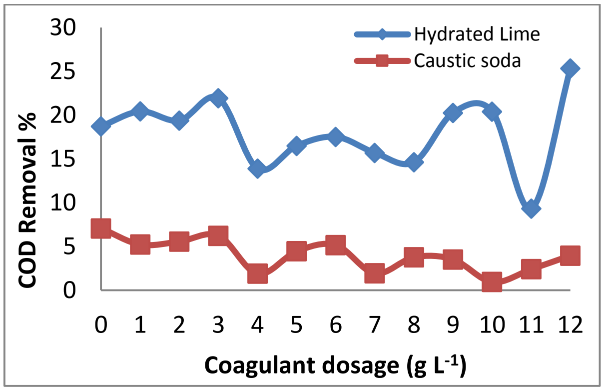


Figure 2 Removal of chemical oxygen demand (COD) at various dosages of hydrated lime, Ca $(OH)_2$ and caustic soda (NaOH)

Based on Kostova (2006), scheduled waste leachate can be categorised in the methanogenic phase as the COD values ranged between 100 and 3500 mg L^{-1} [42]. Figure 2 indicates at the optimum dosage of 5 gL^{-1} of Ca $(OH)_2$ for NH_3-N removal, the COD removal recorded was 16.5 % while at the optimum dosage of 8 gL^{-1} of NaOH for NH_3-N removal, it was observed that the value of COD removal was 3.8 %. However, higher COD removal efficiencies were obtained at 12 gL^{-1} dosage of $Ca(OH)_2$, and 0 gL^{-1} dosage of NaOH with 25.3 % and 7.1 % COD removal, respectively. Generally, there was a reduction in COD removal efficiency for both $Ca(OH)_2$ and NaOH with an increase in the amount of chemicals. However, in this study, inconsistent readings were obtained. This was probably due to the complex characteristics of the scheduled waste leachate itself, which may need advanced treatment strategies. Besides, $Ca(OH)_2$ and NaOH might be a poor chemicals and not very efficient in removing COD from scheduled waste leachate. An extended settling period might be needed to decrease the concentration of the precipitates which contributed to the COD in the leachate [9].

3.4 Effect of Dosage and Types of Chemicals on Colour Removal Efficiency

The effects of amount and types of chemicals on colour removal efficiency were shown in Figure 3. The colour values for SWL indicated a reduction of 15 % and 24.2 % even without the addition of coagulant. According to Aziz *et al*. (2007), the presence of a high concentration of colour in landfill leachate is due to the presence of high levels of organic substances [43]. Typically, stabilised landfill leachate contains high molecular weight substances i.e., humic and fulvic compounds which are not easily degradable [3, 16]. The stirring process might release some of the compounds and thus remove some of the colour from the leachate [7].

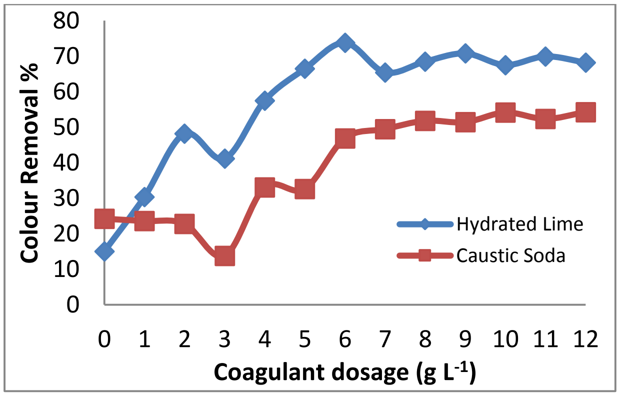


Figure 3 Removal of colour at various dosages of hydrated lime, Ca $(OH)_2$ and caustic soda (NaOH)

In Figure 3, at the optimum dosage of 5 gL^{-1} of $Ca(OH)_2$ for NH_3-N removal, the colour removal efficiency recorded was 66.5 % while at the optimum dosage of 8 gL^{-1} of NaOH for NH_3-N removal, it was observed that the colour removal efficiency was 51.8 %. However, higher colour removal efficiencies were obtained at 6 gL^{-1} dosage of $Ca(OH)_2$, and at 12 gL^{-1} dosages of NaOH with 73.8 % and 54.3 %, respectively.

The optimum dosages for colour removal were 6 gL^{-1} for $Ca(OH)_2$ and NaOH as any further increase in dosage did not have a significant effect on the colour removal. Generally, this study has shown that $Ca(OH)_2$ gives a higher colour removal efficiency than NaOH. A study performed by Malakootian and Fatehizadeh (2010) discovered 12 g L^{-1} of lime gives the best efficiency of colour removal for synthetic drinking water samples at 86.68% and 94% for methylene blue and eriochrome black T, respectively [25]. In particular, Liao and Randtke (1986) showed that excellent elimination of humic substances and proteins can be achieved during lime precipitation due to the substantial dissociation of the functional groups at high pH [44]. The removal of COD and colour was due to the removal of the particles that took place because of their attraction to the coagulants due to the positive zeta potential of these chemicals [32].

3.5 Statistical analysis performed by Response Surface Methodology (RSM) for treatment with hydrated lime, Ca $(OH)_2$ and caustic soda (NaOH)

To find the optimum parameters for NH_3-N, COD and colour removal efficiency from scheduled waste leachate, historical data design (HDD) in response surface methodology (RSM) was used. Table 3 summarized the results obtained from the HDD regarding the experimental factors: dosage of chemicals (X_1) and the types of chemicals (X_2). The responses for experiment: NH_3-N removal efficiency (%), COD removal efficiency (%) and colour removal efficiency (%) are denoted by Y_1, Y_2 and Y_3, respectively. The highest NH_3-N removal efficiency (%) for treatment with hydrated lime ($Ca(OH)_2$) was 60.6% with 5 g L^{-1} of dosage. Meanwhile, the highest COD removal efficiency (%) and colour removal efficiency (%) for treatment with $Ca(OH)_2$ were 25.3% and 73.8%, at dosage 12 gL^{-1} and 6 gL^{-1}, respectively. For the treatment with caustic soda (NaOH), it was observed the highest NH_3-N, COD and colour removal efficiency (%) were 42.3 %, 7.1% and 54.3% at dosage 8 gL^{-1}, 0 gL^{-1}, 12 gL^{-1}, respectively. The regression equations Y_1, Y_2 and Y_3 for each treatment with $Ca(OH)_2$ and NaOH demonstrated that removal efficiency have three empirical equations of independent variables (factors) in actual units, as shown in Table 2 below.

Table 2 Mathematical equation of regression model in the terms of actual factors for NH_3-N, COD and colour removal efficiency (%)of treatment with hydrated lime ($Ca(OH)_2$) and caustic soda (NaOH)

Removal Efficiency (%)	Equation for treatment with hydrated lime ($Ca(OH)_2$)	Equation for treatment with caustic soda(NaOH)
NH_3-N	$Y_1= 10.14874 +9.08520 X_1- 0.42585 X_1^2$	$Y_1= -7.13128 +8.81088 X_1-0.42585X_1^2$
COD	$Y_2= 20.13251 - 0.97133 X_1 + 0.073647 X_1^2$	$Y_2= 7.53368 -1.20199X_1 + 0.073647 X_1^2$
Colour	$Y_3= 25.19627 + 8.62310 X_1 -0.39577 X_1^2$	$Y_3= 10.21200 + 8.13923 X_1 - 0.39577X_1^2$

Table 3 Design used for the models

Run	Dosage (g L^{-1}), X_1	Types of chemical, X_2	NH_3-N removal efficiency, Y_1	COD removal efficiency, Y_2	Colour removal efficiency, Y_3
1	0	$Ca(OH)_2$	6.1952	18.7021	15.0000
2	1	$Ca(OH)_2$	13.9938	20.4043	30.3333
3	2	$Ca(OH)_2$	13.4526	19.3617	48.1667
4	3	$Ca(OH)_2$	37.5624	21.9149	41.1333
5	4	$Ca(OH)_2$	56.9883	13.8723	57.5000
6	5	$Ca(OH)_2$	60.6248	16.4894	66.4667
7	6	$Ca(OH)_2$	55.2017	17.5106	73.8000
8	7	$Ca(OH)_2$	46.2334	15.6809	65.4667
9	8	$Ca(OH)_2$	49.0528	14.6170	68.5000
10	9	$Ca(OH)_2$	46.0301	20.2553	70.8000
11	10	$Ca(OH)_2$	63.3739	20.3830	67.5333
12	11	$Ca(OH)_2$	54.6423	9.3192	69.9667
13	12	$Ca(OH)_2$	60.4254	25.3191	68.2333
14	0	NaOH	0.7897	7.0591	24.2228
15	1	NaOH	2.7627	5.2158	23.5751
16	2	NaOH	9.3191	5.5509	22.7979
17	3	NaOH	11.9463	6.2212	13.7306
18	4	NaOH	14.7733	1.8852	33.0311
19	5	NaOH	14.4208	4.4407	32.6425
20	6	NaOH	37.2041	5.1529	46.8912
21	7	NaOH	37.7063	1.9271	49.4819
22	8	NaOH	42.3051	3.7704	51.8135
23	9	NaOH	20.5761	3.5191	51.4249
24	10	NaOH	45.7024	0.94261	54.1451
25	11	NaOH	39.1003	2.4089	52.3316
26	12	NaOH	41.1338	3.9589	54.2746

Table 4 showed the results of the regression models for NH_3-N, COD and colour removal efficiency (%) in form of analysis of variance (ANOVA). Analysis of Variance (ANOVA) was used to estimate the quality of regression. Based on data given in Table 4, the model F-value of for NH_3-N, COD and colour removal efficiency were 24.64, 34.69 and 36.26, respectively. This value implies the model is significant. Moreover, since the calculated F-Value of the model is higher than the tabulated F-Value, the model predicts the experimental data correctly. Table 4 indicates that computed F-Value (24.64, 34.69 and 36.26) is much greater than tabulated F-value (2.82) at the level of 5%. It was demonstrated that the models was significant at the 5% confidence level since probability of error (*P*) value was less than 0.05. Table 4 indicates "Probability > F" for all models to be less than 0.0001, which is significantly lower than 0.05. The accuracy of the model was checked by the determination of correlation coefficient (R^2) of 82.43%,86.86% and 87.35% for NH_3-N, COD and colour removal efficiency models, respectively. In this case, R^2 value for three models (0.8243, 0.8686 and 0.8735) indicated that the sample variation for NH_3-N, COD and colour removal efficiency 82.43%, 86.86% and 87.35% were attributed to the independent variables and only 17.57%, 13.14% and 12.65% of the total variation cannot be explained by the model. Predicted R-squared for the three models (0.7507, 0.7628 and 0.7765) also in reasonable agreement with the Adjusted R-Squared (0.7909, 0.8435 and 0.8494). Adequate Precision measures the signal to noise ratio. The ratio for three models in this study (16.203, 12.977 and 19.607) is greater than 4 that indicate an adequate signal. All these parameters show that this model can be used to navigate the design space, which is the multidimensional combination and interaction of input variables (e.g. material attributes) and process parameters that have been demonstrated to provide assurance of quality.

Table 4 ANOVA for quadratic response surface model for raw leachate treated with hydrated lime, ($Ca(OH)_2$) and caustic soda (NaOH) at different dosages

Response	F	P	R^2	Predicted R^2	Adjusted R^2	Adequate Precision	Standard Deviation	Coefficient of Variance
NH_3-N Removal	24.64	< 0.0001	0.8243	0.7507	0.7909	16.203	9.24	27.27
COD Removal	34.69	< 0.0001	0.8686	0.7628	0.8435	12.977	3.08	27.97
Colour Removal	36.26	<0.0001	0.8735	0.7765	0.8494	19.607	7.21	14.95

The main aim of using response surface methodology (RSM) in this study was to find the optimum process parameters that favour NH_3-N, COD and colour removal efficiency (%). Thus, the function of desirability was applied using Design-Expert software version 7.1.5. In the optimization analysis of numerical optimization in RSM, the target criteria response: Y_1= NH_3-N removal efficiency (%), Y_2 = COD removal efficiency (%) and Y_3= colour removal efficiency (%), was set as maximum values for the two factors of experimental process: dosage (X_1) and types of chemical (X_2). The dosage (X_1) was set as minimum values as our aim is to choose the chemicals that required less dosage but higher removal efficiencies. Hence, the optimum operating conditions for NH_3-N, COD and colour removal efficiency (%) for both treatment with Ca $(OH)_2$ and NaOH were presented in Table 5. From RSM result, it was suggested that the most optimum dosage of $Ca(OH)_2$ used in the pre-treatment was: 5.9 g L^{-1}, while for NaOH: 6.4 g L^{-1}. Verification experiment was conducted thrice and the result was described in table below.

Table 5 The optimum $Ca(OH)_2$ and NaOH dosage and removal efficiencies obtained by RSM for scheduled waste leachate

Lime dosage (g L^{-1})	NH_3-N Removal (%)		COD Removal (%)		Colour Removal (%)	
	Actual	Predicted	Actual	Predicted	Actual	Predicted
5.9 [$Ca(OH)_2$]	52	49	17.5	18	65	66
6.4 [NaOH]	35	32	2	4	49	42

The maximum value for NH_3-N, COD and colour removal efficiencies with $Ca(OH)_2$ was estimated as 49%, 18% and 66%, respectively. The selected combinations resulted in 52% for NH_3-N removal efficiencies, 17.5% for COD removal efficiencies, and 65% for colour removal efficiencies. In contrast, the pre-treatment with NaOH estimated that NH_3-N, COD and colour removal efficiencies were 32%, 4% and 42%, respectively. However, verification experiment results in 35%, 2% and 49% of NH_3-N, COD and colour removal efficiencies, respectively. The model prediction from the regression equation agreed well with the data from validation experiments. This verified that RSM approach was appropriate to optimize the operational conditions of the treatment in NH_3-N, COD and colour removal efficiency of scheduled waste leachate.

Table 6 shows the final result of this study and several other studies on removal of NH_3-N from different type of landfill leachate with various dosages and types of chemicals. The table shows that previous studies conducted for scheduled waste landfill leachate using the same chemical which is $Ca(OH)_2$ and NaOH only obtained 45% and 48% NH_3-N removal with 6 gL^{-1} and 8 g$L^{-1}$of dosage, respectively [8]. Compared to this study, 52% of NH_3-N was successfully removed at about the same dosage (5.9 g L^{-1}) of $Ca(OH)_2$. However, less dosage of $Ca(OH)_2$ with slightly higher NH_3-N removal efficiency was discovered by Sani *et al*. (2014), in which 4 gL^{-1} of $Ca(OH)_2$ was able to remove NH_3-N up to 54% from a scheduled waste leachate. In a study that utilized PolyAluminum Chloride (PACl) for the treatment of sanitary landfill leachate at two different landfill site [Ampang Jajar Landfill Site (AJLS) and Kulim Landfill Site (KLS)], it was found that 7.2 g L^{-1} and 4.5 g L^{-1} of PACl dosage successfully removed NH_3-N up to 26% (AJLS) and 45% (KLS), respectively [27]. This comparison shows that $Ca(OH)_2$ was more effective in the removal of NH_3-N compared to PACl.

Therefore, $Ca(OH)_2$ was selected to be use prior pre-treatment of scheduled waste leachate with optimum dosage of 5.9 g L^{-1}. The permissible limit standards of NH_3-N are 20 mg L^{-1} as stated in Environmental Quality (Industrial Effluent or Mixed Effluent) Regulations 2009, Fifth Schedule (Standard B) [14]. However, in this study, the effluent after current process still did not meet the discharge requirements (influent= 1790 mg L^{-1}, effluent= 859.2 mgL^{-1}). Hence, additional treatment or combination with biological treatment (using microbes) or physical treatment (air stripping) should be applied until the value of NH_3-N in scheduled waste leachate is below the permissible limit standards.

Table 6 Comparison between results obtained from previous studies and current study

Types of liquid treated	Types of chemicals used	Optimum dosage of coagulant used	Percentage removal of NH_3-N	Reference
Scheduled Waste Landfill Leachate	Calcium hydroxide ($Ca(OH)_2$)	4 g L^{-1}	54%	[7]
Scheduled Waste Landfill Leachate	Calcium hydroxide ($Ca(OH)_2$)	6 g L^{-1}	45%	[8]
	Caustic soda (NaOH	8 g L^{-1}	48%	
Leachate from closed landfill	Calcium hydroxide ($Ca(OH)_2$)	10 g L^{-1}	65-74%	[9]
Sanitary landfill leachate from Ampang Jajar Landfill Site (AJLS) and Kulim Landfill Site (KLS)	Polyaluminum chloride	7.2 g L^{-1} for AJLS 4.5 g L^{-1} for KLS	26% for AJLS 45% for KLS	[27]
Young municipal landfill leachate	Calcium hydroxide ($Ca(OH)_2$)	8 g L^{-1}	85%	[45]
Anaerobic digestion effluent	Calcium hydroxide ($Ca(OH)_2$)	27.5 g L^{-1}	78%	[46]
Scheduled Waste Landfill Leachate	Calcium hydroxide ($Ca(OH)_2$)	5.9 g L^{-1}	52%	This study
	Caustic soda (NaOH)	6.4 g L^{-1}	35%	

4.0 CONCLUSION

In this work, the coagulation-flocculation process with various dosages of hydrated lime ($Ca(OH)_2$) and caustic soda (NaOH) ranging from 0 to 12 gL^{-1} was applied for the treatment of scheduled waste leachate (SWL) using jar test apparatus. The ammonia-nitrogen (NH_3-N), Chemical Oxygen Demand (COD) and colour removal efficiency were evaluated. A Historical Data Design (HDD) of Response Surface Methodology (RSM) was employed to predict the optimum dosage and suitable types of chemicals for the removal of NH_3-N, COD and colour from the available data. The results revealed that the optimum conditions obtained from desirable response for $Ca(OH)_2$ were at 5.9 g L^{-1} of dosage, where NH_3-N, COD and colour were successfully remove up to 52 %, 17.5 % and 65 %. In contrast, the optimum conditions obtained from desirable response for NaOH were at 6.4 g L^{-1} of dosage, where NH_3-N, COD and colour were successfully remove up to 35 %, 2 % and 49 %, respectively. It can be concluded that treatment of scheduled waste leachate (SWL) by jar test experiments in the presence of $Ca(OH)_2$ was more effective in removal of NH_3-N, COD and colour compared to NaOH as a significant reduction in those three parameters measured was shown (NH_3-N, COD and colour removal). In fact, $Ca(OH)_2$ required less dosage in the study. Thus, $Ca(OH)_2$ could be used as an effective pre-treatment step prior to NH_3-N removal in a Scheduled Waste Landfill (SWL) leachate treatment plant. The results obtained in this study are useful for future research, in which the optimum value of $Ca(OH)_2$ could be applied to an ammonia stripping reactor for the removal of NH_3-N from scheduled waste leachate.

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