

Potential of KMnO_4 and H_2O_2 in treating semi-aerobic landfill leachate

Nabihah Abdullah · Hamidi Abdul Aziz ·
N. N. A. N. Yusuf · Muhammad Umar ·
Salem S. Abu Amr

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Abstract Treatment of recalcitrant landfill leachate was carried out using hydrogen peroxide (H_2O_2) and potassium permanganate (KMnO_4). The treatment performance was reported in terms of colour, chemical oxygen demand (COD) and ammoniacal nitrogen ($\text{NH}_4\text{-N}$). The effect of oxidant dosages and pH was determined and optimum conditions were determined considering the removal of parameters of interest. The optimum dosage of both oxidants was similar in terms of COD removal; however, H_2O_2 gave better removal (42 %) and similar results were obtained for $\text{NH}_4\text{-N}$ (24 %). KMnO_4 proved better in terms of colour with a reduction of 74 % compared to 43 % by hydrogen peroxide at pH 7. Acidic conditions (pH 2–5) proved conducive towards the removal of all parameters with the exception of COD removal by KMnO_4 that exhibited a low removal at pH 5 and increased with increasing pH. The COD reduction at pH 3 was ~38 % for KMnO_4 and H_2O_2 , whereas the reduction in $\text{NH}_4\text{-N}$ was 22 and 28 % for KMnO_4 and H_2O_2 , respectively. The COD and $\text{NH}_4\text{-N}$ removal performance of H_2O_2 was better compared with KMnO_4 ; however, consistently higher colour removal was obtained for KMnO_4 .

Keywords Landfill leachate · Hydrogen peroxide · Potassium permanganate · COD · Ammoniacal nitrogen

Introduction

Landfills are widely accepted for controlled disposal of high quantities of solid waste at economical costs in many countries worldwide (Umar et al. 2010a, b). However, landfill leachate produced as a result of interaction of waste with percolating water is considered one of the major drawbacks of this disposal method. Combination of pollutants (organics, ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts) in higher concentrations makes landfill leachate a potential source of contamination to both ground and surface waters (Umar et al. 2010b). Additionally, the composition of leachate is highly site specific and varies widely depending on the age and design of landfill, type of waste, amount of rainfall and several other geographic and environmental factors. It has been widely recognized that depending on the nature of solid waste, the active microbial flora, characteristics of the soil, the rainfall patterns and the age of the landfill, the resultant leachate produced may require different treatment strategies depending mainly on the characteristics of leachate. Currently, several physico-chemical and biological processes are being applied to treat landfill leachate with varying degree of success (Li et al. 2010; Wang et al. 2012). Pre-treatment with physical technologies prior to biological treatment has been largely used via sedimentation, coagulation and flocculation or filtration to remove suspended solids. Biological processes are very effective in treating young leachate with higher biological oxygen demand (BOD_5) and chemical oxygen demand (COD); however, they are generally ineffective in treating mature leachate containing higher concentrations of recalcitrant compounds. Therefore, biological processes such as biological nitrogen removal, either through conventional autotrophic nitrification and heterotrophic denitrification or

N. Abdullah · N. N. A. N. Yusuf
Faculty of Chemical Engineering Technology, TATI University
College, Jalan Panchor, Teluk Kalong, 24000 Kemaman,
Terengganu, Malaysia

H. A. Aziz (✉) · M. Umar · S. S. A. Amr
School of Civil Engineering, Universiti Sains Malaysia,
Engineering Campus, 14300 Nibong Tebal, Penang, Malaysia
e-mail: cehamidi@eng.usm.my

completely autotrophic processes (e.g. Sharon + Anammox), preceded and/or followed by advanced oxidation processes (AOPs), are among recently studied treatment trains. AOPs have been proven effective in treating various recalcitrant wastewaters such as retentate (concentrate fraction) of membrane treatment and landfill leachate. A detailed literature review on various methods used for the treatment of landfill leachate is summarized by Renou et al. (2008) and Wiszniewski et al. (2006). AOPs are generally considered one of the most effective means of treating mature landfill leachate and many AOPs have been recently used for its treatment. Most of them use a combination of: two oxidants ($O_3 + H_2O_2$), catalyst plus oxidant ($Fe^{2+} + H_2O_2$), oxidant plus irradiation ($H_2O_2 + UV$), oxidant plus photo-catalyst ($H_2O_2 + TiO_2 + hv$), oxidants plus ultrasounds (US) ($H_2O_2 + US$) (Lopez et al. 2004).

Use of various oxidants for the oxidation of toxic compounds is widespread including potassium permanganate and hydrogen peroxide (H_2O_2) with respective oxidation potentials of 1.7 and 1.8 V. Potassium permanganate is capable of oxidizing organic compounds containing carbon–carbon double bonds, aldehyde groups or hydroxyl groups. As an electrophile, permanganate ion is strongly attracted to the electrons in carbon–carbon double bonds found in chlorinated alkenes, borrowing electron density from these bonds to form a bridged, unstable oxygen compound known as hypomanganate diester. This intermediate product further reacts by a number of mechanisms including hydroxylation, hydrolysis or cleavage. Potassium permanganate extends several advantages such as easy handling, and is a readily soluble solid and highly effective in water and wastewater treatment (Xu et al. 2005). H_2O_2 is also a strong oxidant and it can be used to generate hydroxyl radicals ($\cdot OH$) with reactivity second only to fluorine. It has been used to oxidize organic matter in domestic or industrial wastewaters for many years (Ksibi 2006). Amokrane et al. (1997) reviewed various oxidants such as chlorine, ozone, potassium permanganate and calcium hydrochloride for landfill leachate treatment and reported a COD removal of around 20–50 %. However, most of the studies reviewed are on mature landfill leachate and none of them looked at semi-aerobic landfill leachate. Although use of oxidants such as potassium permanganate may show low organic content removal, pre-oxidation can reduce the membrane fouling potential making membrane treatment applicable for further treatment (Amokrane et al. 1997). Therefore, instead of using oxidants in combination with UV or ozone, their use as single chemicals was investigated for the treatment of semi-aerobic landfill leachate. The effect of different dosages of H_2O_2 and $KMnO_4$, pH and settling time was studied with an aim to maximize the reduction treatment

efficiency and determining the optimum treatment conditions.

Materials and methods

Leachate sampling and characterization

Landfill leachate was collected from Pulau Burung Landfill Site (PBLs) which is located within Byram Forest Reserve at $5^{\circ}24'N$ and $00^{\circ}24'E$ in Penang, Malaysia. Leachate samples were collected in plastic containers and transported to the laboratory to be stored at $4^{\circ}C$ to minimize possible changes. A total of four sampling campaigns were carried out under dry and wet weather conditions to achieve representative leachate samples. Leachate was removed from the refrigerator and placed for about 2 h at about $22^{\circ}C$ for conditioning before analysis was carried out. Characterization of leachate was carried out for COD, BOD_5 , NH_4-N , TSS, colour and turbidity before and after treatment using standard methods (APHA 2005). The HACH DR/2010 spectrometer was used for determining COD, BOD_5 , NH_4-N and turbidity. COD was determined using the colorimetric method (5220-D). BOD_5 was determined using 5-Day BOD Test (5210-B). Ammoniacal nitrogen was determined using Nesslerization method (4500-NH₃). Turbidity was determined at a wavelength of 860 nm according to Method No. 8237 using DR 2010 HACH spectrophotometer. TSS were measured by Method 2540D. Colour measurements were reported as true colour (filtered using $0.45\ \mu m$ filter paper) assayed at 455 nm using DR 2010 HACH spectrophotometer. pH was measured by portable pH meter (Hanna). pH adjustments were made using 10 Normality of H_2SO_4 and 6 Normality of NaOH to achieve a range of working pH between 2 and 10.

Determination of optimum conditions

Potassium permanganate ($KMnO_4$ 158.03 g/mol) and hydrogen peroxide (H_2O_2 30 %, 34.01 g/mol) were used in this study to treat stabilized leachate. To determine optimum dosage of oxidants, a range of concentrations (50–500 mg/L, SD = 146.7, SE = 48.9) of $KMnO_4$ and H_2O_2 was used. For this purpose, 150 mL of leachate sample was added to a volumetric flask and the sample was mixed well (300 rpm for 60 min) and pH was adjusted to 7. COD, ammoniacal nitrogen and colour were measured and recorded as mentioned above. The sample pH was also measured and recorded as final pH. Percentage removal of COD, ammoniacal nitrogen and colour versus concentration of $KMnO_4$ and H_2O_2 were plotted and the dosage that gave the maximum removal was considered as the optimum dosage. Similarly, the effect of pH was studied by

Table 1 Characteristics of landfill leachate

Parameter	Value
COD (mg/L)	1,964 ± 150
BOD ₅ (mg/L)	396 ± 25
BOD ₅ /COD	0.2
Turbidity (FAU)	290 ± 20
TSS (mg/L)	243 ± 30
NH ₄ -N (mg/L)	1,152 ± 60
Colour (Pt.Co)	3,800 ± 190
pH	7.96 ± 0.5
Fe ²⁺ (mg/L)	9 ± 0.5

varying pH between 2 and 10 and optimum pH was determined at optimal dosages of KMnO₄ and H₂O₂ established previously. Optimum settling time was similarly determined using the optimum dosages and pH as discussed above.

Results and discussion

Characteristics of leachate

The characteristics of landfill leachate are given in Table 1. The average concentration of COD in PBLs leachate was 1,964 mg/L which is in agreement with Ghafari et al. (2010). Based on literature, this value of COD represents the methanogenic phase within the landfill body (Christensen et al. 2001; Jokela et al. 2002; Kostova 2006). Various BOD₅/COD ratios in the range of 0.043–0.67 have been reported for mature landfill leachate (Aghamohammadi et al. 2007; Canziani et al. 2006; Salem et al. 2008; Weiner and Matthews 2003). The recorded value in this study is in close agreement with Aghamohammadi et al. (2007) who reported a BOD₅/COD ratio of 0.17 for PBLs leachate. The low BOD₅/COD ratio represents the leachate stability and hence its recalcitrance to biological degradation (Jokela et al. 2002). Therefore, physico-chemical treatment is generally considered effective for the treatment of stabilized landfill leachate (Ghafari et al. 2010; Kurniawan et al. 2006). The value of turbidity for PBLs was in agreement with Ghafari et al. (2010). The concentration of TSS was 243 mg/L which was higher than that reported by Bashir et al. (2009), whereas it was lower than that measured by Aziz et al. (2010). This could be due to the variations in sampling from different leachate ponds. As shown in Table 1, the value of ammoniacal nitrogen (1,152 mg/L) is in close agreement with Ghafari et al. (2010) who reported an average value of 1,184 mg/L. The value of colour was calculated as 3800Pt.Co which agrees well with the average value of 3869Pt.Co reported by

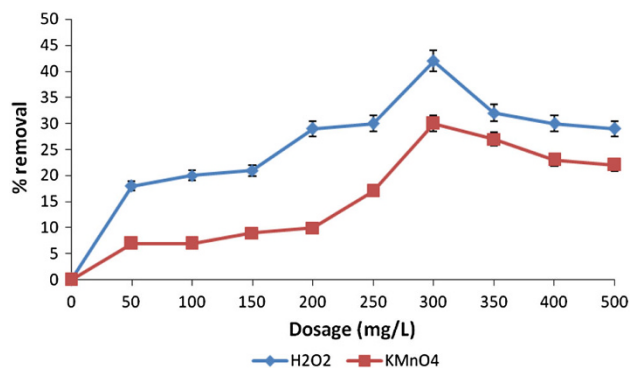


Fig. 1 Removal of COD with H₂O₂ and KMnO₄ at various dosages and pH 7

Ghafari et al. (2010). The value of pH recorded represents a typical mature leachate as also reported by others (Salem et al. 2008; Zhong et al. 2009).

Optimum dosages of KMnO₄ and H₂O₂

Removal of COD, ammoniacal nitrogen and colour

The results of optimum dosage determined at pH value of 7 are given in Fig. 1. It can be seen that H₂O₂ was more effective in removing COD than KMnO₄. The maximum COD removal was 42 % at 300 mg/L of H₂O₂, whereas it was 30 % for 300 mg/L KMnO₄. Different removal by each oxidant can be attributed to the different type of organic matter targeted by each of the oxidant and the amount of generation of hydroxyl radicals. Moreover, hydrogen peroxide improved oxygen production and oxidation rate (Eq. 1) (Chen et al. 1996)



Potassium permanganate is highly reactive under conditions found in the water industry. It will oxidize a wide variety of inorganic and organic substances. Potassium permanganate (Mn⁷⁺) is reduced to manganese dioxide (MnO₂) (Mn⁴⁺) which precipitates out of solution (CRC 1990). The half reaction of potassium permanganate at natural pH is illustrated in Eq. 2



A chemical oxidation can remove organic matter and heavy metals (Urase et al. 1997) by absorption and development of complex solution (Trebouet et al. 2001; Urase et al. 1997). Lower COD removal can be partly attributed to higher ammonia concentration, which is a difficult inorganic matter to oxidize as also reported by Vogel et al. (2000). Generally, the amount of COD removed depends on the reaction of organics with inorganics or inorganics with inorganics (Kylefors et al.

2003). Moreover, it depends on the chemical reaction in raw water (Kylefors et al. 2003) due to its complex matrix which may result in scavenging hydroxyl radicals. Higher initial colour can also reduce the efficiency of chemical oxidation due to internal quenching. According to Kylefors et al. (2003), one-third of the COD in leachate was influenced by inorganic matters such as Fe(II), manganese(II), sulphide, ethanol, acetic acid, ammonia and chloride. In their research, Kylefors et al. (2003) concluded that Fe(II) and sulphide were the main contributors to inorganic matters that influenced COD reduction.

Figure 2 shows the removal of $\text{NH}_4\text{-N}$ at different dosages of both oxidants. The highest removal of ammoniacal nitrogen achieved was about 24 % at 300 mg/L H_2O_2 and decreased thereafter. On the other hand, the highest removal of $\text{NH}_4\text{-N}$ was 19 % at 500 mg/L KMnO_4 . Hence, the removal using H_2O_2 was higher as compared to KMnO_4 at dosages below 400 mg/L, but became higher at dosages of 400 and 500 mg/L.

The effect of different dosages of H_2O_2 and KMnO_4 for colour removal at pH 7 is shown in Fig. 3. As noted earlier (Table 1) leachate was high in colour with concentration of 3800Pt.Co. At optimum dosage of KMnO_4 (400 mg/L), the

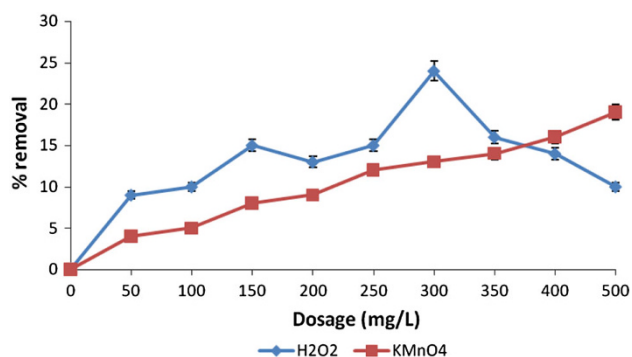


Fig. 2 Removal of ammoniacal nitrogen with H_2O_2 and KMnO_4 at various dosages and pH 7

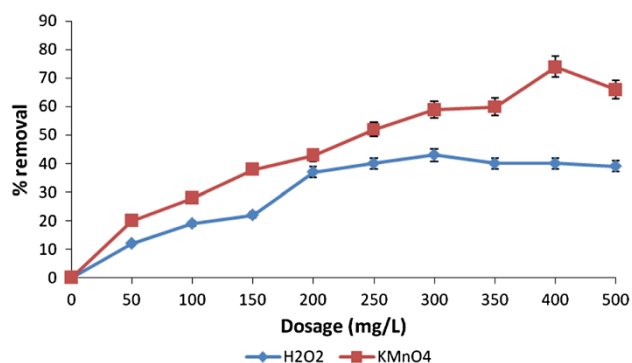


Fig. 3 Removal of colour with H_2O_2 and KMnO_4 at various dosages and pH 7

removal of colour was 74 %. For H_2O_2 , the optimum dosage for colour removal was 300 mg/L with 43 % removal. Increasing the dosage of oxidants turned the leachate from black to brown and finally light brown, which was observed particularly for KMnO_4 . The colour of leachate was observed to change to brownish after the optimum dosage was exceeded.

Generally, the percentage of removal increased with an increase in the amount of oxidant. This could be explained in terms of the increased amount of photons that was absorbed and reacted with the oxidant at high concentration. Although a higher dose of oxidant implies more concentration of hydroxyl radicals that react with organics and inorganics, an optimum dosage is required to be established to avoid both chemical consumption and therefore cost of the treatment process. In addition to that, excess amount of an oxidant can lead to decrease in removal efficiency as reported by Wang et al. (2000). The authors noted scavenging effect of excess H_2O_2 due to its reaction with HO^\cdot , inhibiting the oxidation process.

Determination of optimum pH

An optimum pH is generally required to be established to achieve maximum removal efficiency. According to the previously established results, the dosage of KMnO_4 was fixed at 350 mg/L, whereas the dosage of H_2O_2 was fixed at 300 mg/L. The range of pH of samples was kept between 2 and 10 at fixed contact time of 30 min.

Removal of COD, ammoniacal nitrogen and colour

Figure 4 shows the removal of COD at various pH values for both oxidants. Most of the organic matter was removed under acidic condition for both the oxidants and the highest removal of 37 % was obtained using KMnO_4 at pH 3. A slightly higher removal (41 %) was obtained using H_2O_2 at

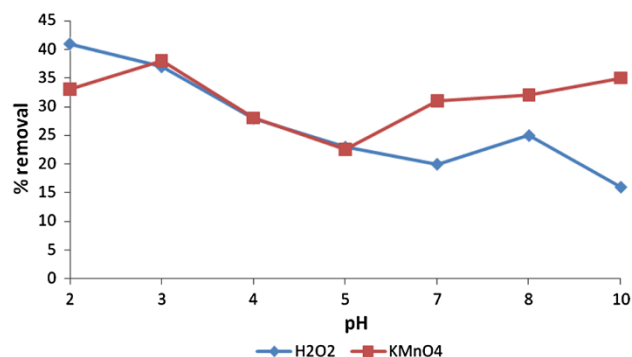
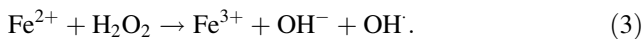


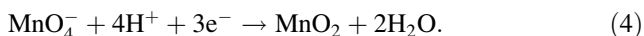
Fig. 4 Effect of pH values on COD removal using 400 mg/L of both H_2O_2 and KMnO_4

pH 2. At pH 5, the removal of COD was about 23 % for both oxidants. However, the removal increased under alkaline condition using KMnO_4 , whereas it decreased substantially to 16 % using H_2O_2 at pH 10 (Fig. 4).

In the H_2O_2 system, the dissolved iron ions in leachate (Fe^{2+} 9 mg/L, Table 1) reacted with H_2O_2 , resulting in Fenton reaction and formation of hydroxyl radicals ($\cdot\text{OH}$) (Eq. 3). $\cdot\text{OH}$ has the potential to destroy and degrade organic pollutants, while the optimum pH value for Fenton reaction ranged between 2.5 and 4 (Hermosilla et al. 2009).



Generally, the oxidation potential of KMnO_4 (Eq. 4) in acidic medium ($E^0 = 1.68$ V) is higher than that in alkaline medium ($E^0 = 0.60$ V) (CRC 1990)



Better removal of H_2O_2 under acidic conditions is in agreement with Lim et al. (1997), who concluded that the removal of organic matter in water is influenced by the pH of water. Generally, the optimum removal of organic matters happen at low pH (Urase et al. 1997). At low pH, carboxylic functional group and phenol from humic matter are protonized and decreased the charges of humic matter (Trebouet et al. 2001).

Depending on the solubility in alkali and acidic conditions, humic matter is divided into three categories. They are humic acid, fulvic acid and humine (Alvarez-Puebla et al. 2004). The humic matter contains aromatic and aliphatic components, especially carboxyl functional group and phenol (Trebouet et al. 2001). Some of the organic pollutants in leachate are directly oxidized to the end product (CO_2 and H_2O) in the chemical oxidation process and the same is firstly converted into the intermediate product (acetic acid) which is further oxidized to the final product.

Figure 5 shows the effect of pH on ammoniacal nitrogen removal. Oxidation process using H_2O_2 seems to be more effective in removing ammoniacal nitrogen as compared to

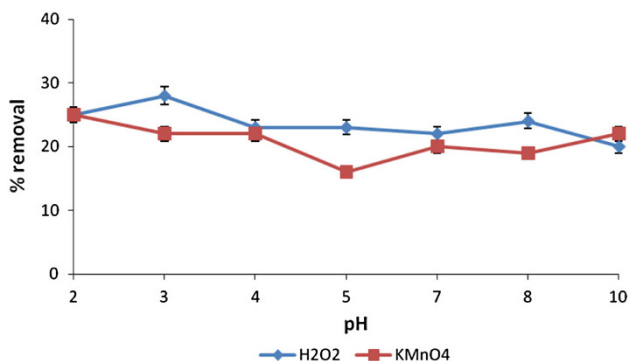


Fig. 5 Effect of pH values on removal of ammoniacal nitrogen using 400 mg/L of both H_2O_2 and KMnO_4

KMnO_4 , especially at pH 3. The maximum percentage removal of ammoniacal nitrogen was 28 % with H_2O_2 at pH 3 and the corresponding value for KMnO_4 was 22 %.

The presence of organic matter, in particular the humics, contributes to the colour of water. Colour that is caused by turbidity is known as true colour, whereas that caused by metallic ions or suspended solid is known as apparent colour (APHA 2005). Because of this reason, the removal of suspended solids and turbidity also influences the colour removal. Figure 6 indicates the removal of colour at various pH values. The results revealed that the removal of colour was less influenced by the change of pH using KMnO_4 with colour reduction of 57–76 % at various pH values. Similarly, acidic conditions improved the removal of colour using H_2O_2 with 54 % reduction in colour at pH 4 and 39 % at pH 10. Observation during experiment showed that a decrease in pH increased the sludge content, which can be attributed to better oxidation of the organic matter. Almost 20 % of the total volume (150 mL) of leachate became sludge at pH 3 using KMnO_4 . With increase in pH, the colour of leachate changed from black to dark yellow and became black in colour at pH 7. KMnO_4 is an effective oxidant of manganese and the change in colour with change in pH can be attributed to the precipitation of manganese and its different forms depending on the pH. The manganous (Mn^{2+}) form is oxidized to manganic (Mn^{4+}) which is blackish in colour giving rise to the black colour of leachate as observed in this study.

It can be concluded from the above discussion that the removal performance was better under acidic conditions. pH is one of the critical parameters in achieving better removal performance using chemical oxidation and therefore, selection of optimum pH is important to be established. Considering the fairly low removal of studied parameters the need of polishing treatment is felt before leachate is discharged to surface water. Some of the potential post-treatment options include membrane treatment, provided the process is cost-effective. Another

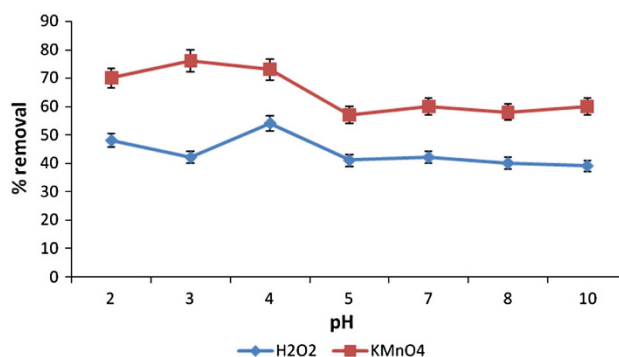


Fig. 6 Effect of pH values on colour removal using 400 mg/L of both H_2O_2 and KMnO_4

possibility is to use strong AOPs (UV/H₂O₂, UV/O₃, etc.) with a view to achieve better treatment performance followed by biological treatment.

Operating cost estimates for treating 1 m³ of stabilized leachate by using H₂O₂ and KMnO₄ were calculated and compared. The total treatment cost was allocated for purchase of chemical reagents. For H₂O₂, 1.47 mL was consumed to treat 150 mL of leachate, which means around 10 L of 30 % H₂O₂ is required for the treatment of 1 m³ of leachate. For KMnO₄, 3.16 g was consumed to treat 150 mL of leachate, and around 21 kg of KMnO₄ is required for treatment of 1 m³ of leachate. The cost of 1 L 30 % H₂O₂ was around 10 USD and that of 1 kg KMnO₄ around 14 USD. The estimated costs for treating 1 m³ of stabilized leachate by H₂O₂ and KMnO₄ were 100 and 294 USD, respectively. Thus, H₂O₂ is cheaper than KMnO₄ for treatment of stabilized leachate.

Conclusions

Treatment of landfill leachate was optimized using H₂O₂ and KMnO₄. At the optimum dosage conditions and pH 7, fairly moderate removal of COD (30 and 42 %) and ammoniacal nitrogen (19 and 24 %) removal were obtained, whereas the removal of colour was considerably lower (43 %) for H₂O₂ compared with KMnO₄ (74 %). The removal of COD was maximum at pH 2 for both oxidants, and further increase in pH resulted in lower removal performance; however, the removal performance improved gradually for KMnO₄ at neutral and basic pH values. The removal of ammoniacal nitrogen was better at pH 2 and 3 for KMnO₄ and H₂O₂, respectively; however, the removal remained fairly similar for H₂O₂ for higher pH values, whereas it decreased until pH 5 and then increased at neutral pH using KMnO₄. The removal of colour was higher at pH 3 for KMnO₄ and pH 4 for H₂O₂, but it remained almost similar for pH higher than 5.

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