Leachate Treatment from Sarimukti Landfill Using Ozone with Sludge from Water Treatment Plant as a Catalyst

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Abstract. Leachate is the liquid waste from anaerobic decomposition in a landfill. The ozonation process can be used for leachate treatment. Sludge from sedimentation in water treatment plant contains 5.96% of Al and 9.35% of Si which can affect of its cation exchange capacity and affects the active site in the catalyst. This study aims to determine the effectivity of sludge in the ozonation process to treat leachate. A 1,5 L semi-batch reactor containing 1 L sample was used in this experiment with the rate of oxygen supply was at 4 L/min taken from ambient air. Two groups of sludge weighing 1.5 grams, 3.0 grams and 4.5 grams were used and activate with physically and chemically activated. The best result was obtained by the physically activated sludge with mass of 4.5 gram O_3-L-4,5 AF. The differences of removal efficiency between O_3-L-4,5 AF with the control (O_3) for turbidity were respectively 13.02% and 7.81%, for EC were 10.57% and 8.29%, for COD were 49.44% and 37.50%, and for residual ozone concentration at the end of contact time were 7.6 mg/L and 9.7 mg/L. It can be concluded that activaed sludge and ozonation can be used as a catalyst in leachate treatment.

1 Introduction

Population growth will lead to an increase in the amount of waste disposed off to a landfill. The waste disposed to the landfill undergoes several simultaneous physical, chemical, and biological changes that produce a liquid called leachate [26,12]. Leachate is the liquid from solid waste decomposition in a landfill. Leachate will be produced, with a high chemical oxygen demand (COD), low biodegradibility, heavy metals, pathogens [25], organic materials (biodegradable and non-biodegradable), and ammonia-nitrogen [27].

The commonly used techniques to treat leachate in Indonesia is the use of stabilization ponds [5]. This technique has the disadvantages of which is the long detention time, and required a large area. Stabilization ponds also used at Sarimukti landfill, and based on measurement data from the west java provincial government through the department of settlements and housing (Diskimrum), 2016, the results of leachate treatment with stabilization pond technique has a BOD/COD ratio of 0.050 [29], and the effectiveness of COD removal was only 13,94% [20]. The effectiveness of biological leachate treatment in Sarimukti landfill is less effective, because leachate is a wastewater that is difficult to treat by biological [4], due to the complex nature of the COD component [2]. Therefore, chemical processing alternatives are required, i.e. using ozone.

Ozonation processes are attractive means for the treatment of landfill leachates due to the high oxidative power that ozone possesses [25]. Ozonation is a chemical process that can oxidize many organic and inorganic materials in water. Ozone is a very selective oxidant, and unstable in water because it can decompose quickly, however the unique feature of ozone is its decomposition into hydroxyl radicals ($OH^{\bullet}$) which are the strongest oxidants can react fast with many dissolved compounds and the water matrix [9]. According to Nuriana, 2015, leachate treatment with ozone has a removal efficiency for turbidity of 91.47%, electrical conductivity (EC) of 20.41%, and COD of 35% [14]. The ozonation process can be optimized by various advanced oxidation process (AOP) such as the addition of peroxide (H_2O_2), UV, and catalytic ozonation. According to Sururi et.al, 2014, the addition of 0.6 gr/L H_2O_2 in diluted the leachate sample taken from Sarimukti landfill has a removal efficiency COD of 28.9%, and with the use of UV lamps within 10 cm of the ozone contactor has efficiency of 23.4% [23,12].

In the last few years heterogeneous catalytic ozonation has been successfully used for the removal of organic compounds [9]. Heterogeneous catalytic ozonation is one of the oxidant techniques used to improve the efficiency of ozonation process [10]. The application of ozonation technique by using a catalyst is a promising alternative for leachate treatment. The sludge from water treatment plant can be used as a

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catalyst, based on Muruganandham et.al, 2007, found that water treatment sludge powder was successfully used as an ozone decomposition catalyst in aqueous medium. Increase in the catalyst loading from 125 to 750 mg significantly promotes the percentage of ozone decomposition from 50.9% to 96.2% [13]. Thus, it is predicted that the use of sludge in the ozonization process of leachate treatment can accelerate the decomposition of ozone into OH$^\bullet$.

Because the leachate treatment at Sarimukti landfill which has a very low BOD/COD ratio, it needs chemical processing with ozone. Ozone is a powerful oxidant that can treat waste that is difficult to degrade such as leachate. In addition, this study aims to determine the effectiveness of the addition of sludge from drinking water treatment to treat leachate which is assumed to accelerate the decomposition of ozone to OH$^\bullet$.

2 Materials and Methods

2.1. Materials

Grab sample were taken from the Sarimukti landfill leachate collection points. The sludge that will be used as a catalyst was collected from sedimentation unit of Tirtawening water treatment plant in Bandung. The sludge was previously examined using X-ray Flourescence (XRF) which showed that the sludge consist of 9.35% Si, and 5.96% of Al, such that the Si/Al ratio is 1.56, this Si/Al value is dominated by silica content rather than aluminium, while Sururi et.al, 2016, says that aluminium acts as a catalyst. After that, the sludge as a catalyst was sieved based on ASTM D 421-85 to determine the coefficient of uniformity [22], which was 5.67, indicating a much more sand. Because the sludge was too fine sand, it is necessary to solidified, by compacting, to get a greater specific gravity than before. If the sludge is not solidified, it will cause the sludge to carried away during the ozonation process at the time of measurement. Egg whites were used for solidification as an adhesive, which serves to attached the sludge.

The catalyst is then physically and chemically activated. Physical activation was done by heating at 105°C for 24-36 hours [28]. For chemical activation, the solidified sludge was heated at 105°C for 24-36 hours, and impregnated with 24.5 ml of 3 mol/L per 10 gr of sample with ZnCl$_2$ for 24 hours at room temperature; after which the supernatant liquid was removed, and the sludge was dried at 105°C for 24 hours. For chemical activation, the sludge was than washed using 3 mol/L HCl to remove inorganic impurities, rinsed with de-ionized water and dried at 105°C for 24 hours [28]. A pH$_{PZC}$ measurements was taken for the activated sludge, to know the pH where the catalyst is uncharged or the positive and negative charges are equal (isoelectric point) [7]. A pH$_{PZC}$ in this study was measured to understand the mechanism of catalytic ozonation of the adsorption phenomenon from the catalyst when it is used in the ozonation process. Measurement of pH$_{PZC}$ was done by titration [16]. Firstly the sludge as catalyst were washed with de-ionized water and dried in an oven at

108°C for 2 hours. Subsequently, sludge as much as 0.1 gr was added to 25 mL of 10^{-3}mol/L NaCl with continues stirring. The pH changed gradually and become constant at the point of zero charge (pH$_{PZC}$) [16]. Physically activated sludge has a pH$_{PZC}$ value of 6.46-7.05 pH, while chemically activated with ZnCl$_2$ has a pH$_{PZC}$ value of 4.32-4.42 pH.

2.2 Materials and Methods

This study was conducted in a semi-batch reactor with continuous ozone gas supply to the contactor filled with leachate sample. The equipment used in this studies consisted of an aerator, a flow meter, an ozone generator, and an ozone contactor that can be seen in Figure 1.

Fig. 1. Reactor Configuration.

The flow rate of oxygen supply was at 4 L/min taken from ambient air as connected to an ozone generator. The Ozone generator which changes the O$_2$ into O$_3$, has a spesification of OZF-1G, 200 W, and 220 V. The ozone generated was applied to an ozone contactor (1500 ml) which had previously been filled with 1000 mL of leachate sample. Sludge as a catalyst was then added to the ozone contactor with different activation and weight. Physically activated abbreviated as AF and chemically activated abbreviated as AK, have a weight variation of 1.5 gr (O$_3$-L-1,5 AF/AK); 3.0 gr (O$_3$-L-3,0 AF/AK); and 4.5 gr (O$_3$-L-4,5 AF/AK) was prepared. To make a homogeneous ozone condition through out the leachate sample, the contactor was equipped with magnetic stirrers. The ozonation process is carried out for 180 min, with samples taken ± 30 ml every 30 minutes. Parameter such as pH, dissolved oxygen (DO), residual ozone concentration (ROC), turbidity, electrical conductivity (EC), and chemical oxygen demand (COD) were measured every 30 minutes.

3 Results

3.1. Leachate Characteristics and Sludge as Catalyst

The results of measurement of the initial characteristics of leachate sample from Sarimukti landfill can be seen in Table 1.

Table 1. Leachate Characteristic.

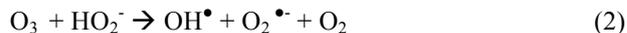| No	Parameter	Unit	Concentration
1	pH	-	7.86-8.40
2	DO	mg/L	1.34-5.10
3	Turbidity	NTU	45.25-116.75
4	EC	mS/cm	4.31-29.57
5	COD	mg/L	4286-8378

Table 1 shows that the pH is neutral or slightly alkaline. This high pH value indicates that there are many hydroxide ions (OH⁻) that can act to initiate the process of ozone decomposition [8]. The value of DO in the leachate sample were at 1.34-5.10 mg/L, indicating the amount of oxygen (O_2) available in the leachate. Oxygen fluctuations can affect to other chemical parameters, especially during oxygen-free conditions, which can lead to changes in the solubility properties of some chemical elements in the water [11, 6]. Sarimukti landfill and Jatibarang landfill is a landfill that has been operating for more than 10 years, and has similarities in treating lechate with stabilization pond. Sarimukti and Jatibarang landfills has a slightly similar pH were respectively 8.4 and 8.5, and for COD were 4286-8378 mg/L and 4000 mg/L [19].

The turbidity of the sample were 45.25-116.75 NTU indicate the amount of organic and inorganic content suspended in the leachate. The value of turbidity from Sarimukti landfill is lower than Jatibarang landfill of 300 NTU [19]. The EC of the sample were at 4.31-29.57 mS/cm indicating the inorganic content of the leachate sample. The value of EC is very high as compared to other leachates in other landfills in Indonesia which ranges from 1000-40000 µS/cm [24]. The concentration of COD were 4286-8378 mg/L indicating the number of organic compounds that were biodegradable and non-biodegradable. The value of COD is very high when compared with the effluent quality standard of leachate treatment plant in Indonesia, about the effluent leachate of 300 mg/L [17].
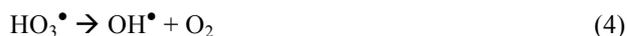
Sludge is predicted can used as a catalyst for ozone decomposition and also be utilized as an adsorbent. According to Ikhlaq et.al, 2012, the pH of a solution plays an important role in understanding the mechanisms of catalytic ozonation, since it is affected by ozone decomposition. At basic pH (pH>pH $_{PZC}$) the surface is negatively charged (no surface hydroxyl groups present), at acidic pH (pH<pH$_{PZC}$) its surface is positively charged (presence of hydroxyl groups), while at pH=pH$_{PZC}$ the surface is neutral [9]. Sludge as the catalyst used in this process has different pH$_{PZC}$ between activation variations. pH$_{PZC}$ on physically activated catalysts of 6.46-7.05 pH and pH$_{PZC}$ on chemical activated catalysts of 4.32-4.42 pH. Because the pH of the solution is higher than pH$_{PZC}$, it is predicted that the sludge will act as adsorbent of O_3 and organic matter, so that the reaction of ozone and organics will occur at the surface of sludge, see Tabel 1.

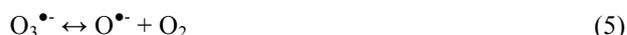## 3.2. Response to The Ozonization Process Based on pH and DO

pH is used as an indicator for hydrogen ion activity [21]. A high pH value indicates more hydroxide ions (OH⁻) acting as an initiator in the decomposition of ozone [8]. In the ozonation system pH usually gives a positive effect on the removal rate of COD [3]. A high pH value is important because the hydroxide ions will start the ozone decomposition involving the following reactions [8]:

$$O_3 + OH^- \rightarrow O_2 + HO_2^- \tag{1}$$

$$O_3 + HO_2^- \rightarrow OH^\bullet + O_2^{\bullet-} + O_2 \tag{2}$$

if pH ≤ 8:

$$O_3^{\bullet-} + H^+ \leftrightarrow HO_3^\bullet \tag{3}$$

$$HO_3^\bullet \rightarrow OH^\bullet + O_2 \tag{4}$$

if pH ≥ 8:

$$O_3^{\bullet-} \leftrightarrow O^{\bullet-} + O_2 \tag{5}$$

$$O^{\bullet-} + H_2O \rightarrow OH^\bullet + OH^- \tag{6}$$

$$OH^\bullet + O_3 \rightarrow HO_2^\bullet + O_2 \tag{7}$$

These reactions will increase pH by time as can be seen in Figure 2. The pH of all variations increase with increasing contact time as indicated by the increasing of hydroxide ions. The physically activated catalysts resulted in a higher pH than the control condition, and the other variations. The more catalyst was added during the ozonation process will result in higher pH values. Based on the reaction (1) and (2) the initiation of ozone decomposition can be accelerated by increasing the pH or by the addition of hydrogen peroxide [1].
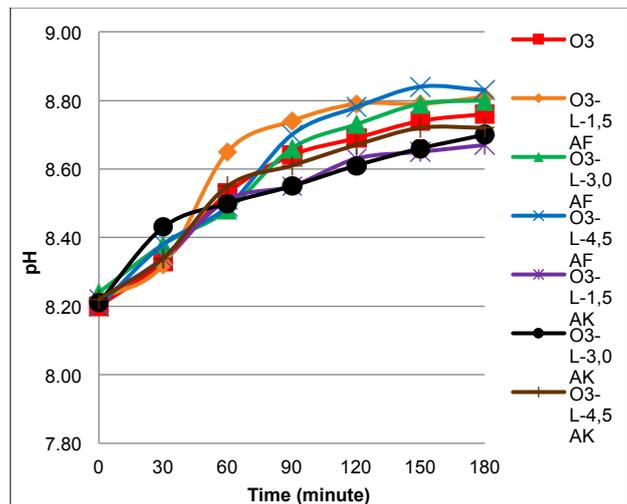

Figure 2. pH Change by Time.

Dissolved oxygen is one of the important parameters in leachate after going through ozonation process. Figure 3 shows that the DO fluctuates during the 180 minutes of the process. Increased DO occured because of the decomposition process of ozon based on reactions (1) and (2); and its decrease, occurred because oxygen acts

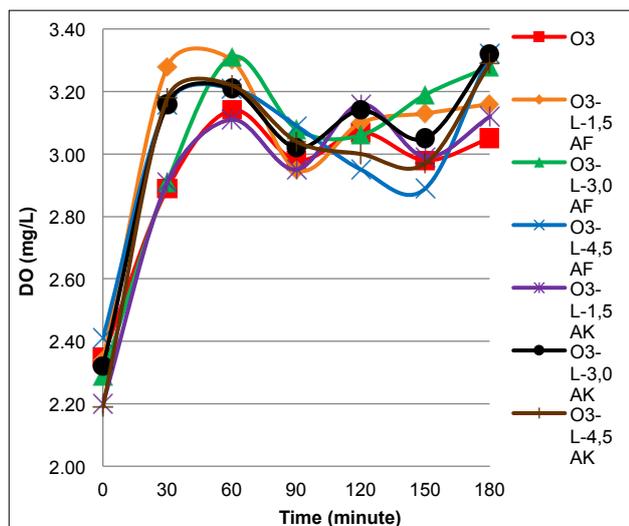as an oxidizing agent which react with the organic material in the leachate.

Figure 3. Dissolved Oxygen by Time.

3.3. Residual Ozone Concentration (ROC)

ROC measurements were conducted to find out the more dominant type of reaction (direct reaction and indirect reaction) occuring between ozone and the organic, and inorganic constituent in the sample. A direct reaction is a selective reaction that causes high ROC value due to the low ozone decomposition into OH$^\bullet$. The indirect reaction, however, is non-selective indicated by the low ROC value due to the high ozone decomposition into OH$^\bullet$. Measurements were made at the last 3 contact points because they were a critical points, and there has been decoloration on leachate sample [20].

Figure 4 shows a fluctuating ROC for all variations. The chemically activated catalysts resulted in higher ROC values than the physically activated, but it is lower than the control condition. The more catalyst was added, the faster the ozone decomposition takes place, which indicates an indirect reaction by OH$^\bullet$, evidenced by low ROC values due to the high ozone decomposition into the OH$^\bullet$. Based on the ROC data, it can be concluded, that the addition of sludge as catalyst can accelerate the decomposition of ozone into OH$^\bullet$.
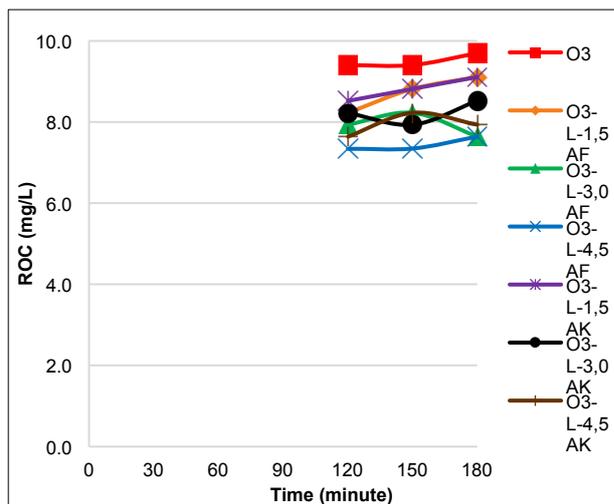

Figure 4. Residual Ozone Concentration.

3.4. Process Efficiency

3.4.1 Physical Parameter: Turbidity Removal

Turbidity, expressed in NTU (nephelometric turbidity units), is caused by material suspended from colloidal size to dispersed coarse material [21].

Figure 5 shows that turbidity can decreased by all variations; indicating an increase in the effluent discharge efficiency. The chemically activated catalysts removed turbidity the most, by 13.83-14.92% which were higher than the control of 7.81%, and other physically activated catalysts of 11.58-13.02%. This phenomenon occured because $ZnCl_2$ can bind water molecules contained in the sludge, which causes the pores of the sludge to increase its surface area [15], so that many pollutants can be adsorbed by the chemically activated catalysts.
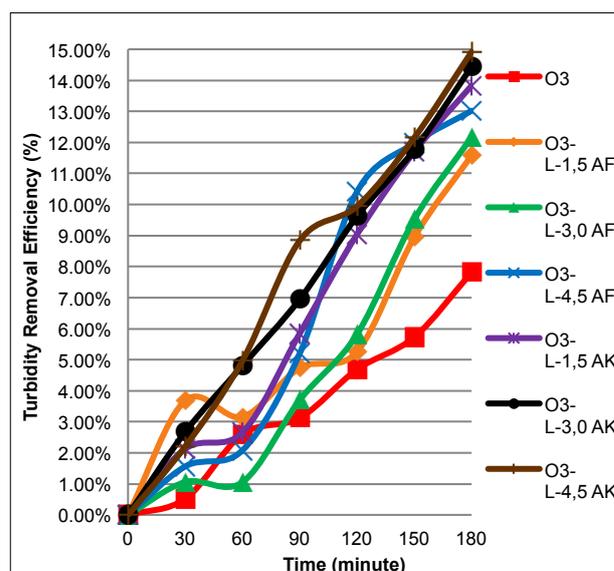

Figure 5. Turbidity Removal Efficiency.

Figure 5 also shows that the more catalyst was added, the higher the rate of turbidity removal. The highest level

of removal was found for the O_3-L-4,5 AK, which decreases the turbidity from 45.25 NTU to 38.50 NTU or a removal efficiency of 14.92%. The decrease in turbidity occurs because ozone can degrade compounds that cause turbidity during the ozonation process. Continuous ozonization process create negatively charged colloidal particles which can be neutralized by ozone [18]. This phenomenon can be attributed to the high pH value of this variation as shown in Figure 2 and low ROC value as shown in Figure 4. The alkaline pH is indicated by the amount of OH^- ions, and the low ROC indicates the amount of ozone decomposed into $OH^{\bullet}$, causing the turbidity of the sample to decrease.

3.4.2 Physical Parameter: EC Removal

In this study the ion measured as electrical conductivity was estimate that to represent the inorganic content in the leachate sample.

Figure 6 shows that all catalyst variations increase the efficiency of removing the inorganic content of the leachate. The chemically activated catalysts removed EC by 7.82-8.77% which were higher than the control of 8.29%, but it is lower than the physically activated catalysts of 10.44-10.57%. The low removal efficiency for the chemically activated catalysts can be caused by saturation of it's surface area, as this variation was able to decrease turbidity more than the physically activated catalyst.
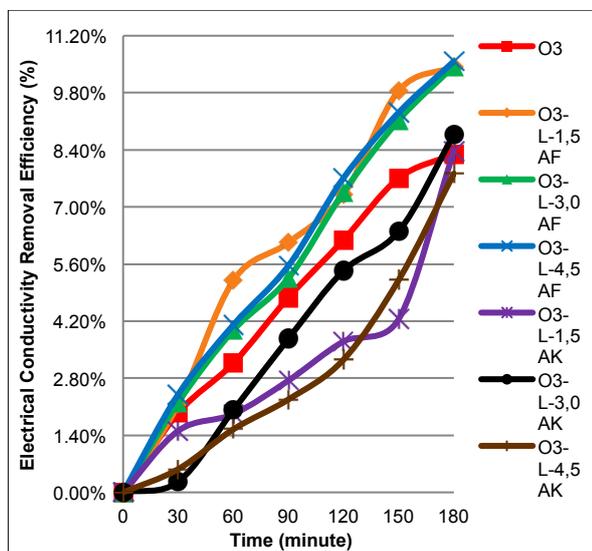

Figure 6. Electrical Conductivity Removal Efficiency.

The highest EC removal was found for of the O_3-L-4.5 AF which decreased the EC from 19.96 mS/cm to 17.85 mS/cm, or a removal efficiency of 10.57%. This phenomenon once again can be attributed to the high pH value of this variation as shown in Figure 2, and has a low ROC value as shown in Figure 4. High pH, and low ROC indicate the amount of $OH^{\bullet}$ formed that will react with the inorganic compound in the leachate sample. The low removal of inorganic compounds might be due to a material that is quite difficult to degrade, even using ozone which is a strong oxidizing agent.

3.4.3 Organic Parameter: COD Removal

COD removal was measured to find out how much organic compound can be eliminated, especially the non-biodegradables.

Figure 7 shows that each variation increases in COD removal by time. The chemically activated catalysts removed COD by 27.27-36.36% which were higher than the control of 30.0 0%, but the physically activated catalysts has a removal efficiency for COD of 27.27-40.00% higher than the other variations.

The highest COD removal was found for of the O_3-L-4.5 AF which decreased the COD from 7616 mg/L to 4570 mg/L, or a removal efficiency of 40.00%. This phenomenon can also be attributed to the high pH value of this variation as shown in Figure 2. In addition, this variation has a low ROC value as shown in Figure 4, the low ROC indicates that ozone decomposes to $OH^{\bullet}$. The decrease in COD concentration is caused by a non-biodegradable organic substance which is oxidised either directly with ozone which is selective or indirect by $OH^{\bullet}$ which has a non-selective property and converts it into an inorganic substance such as CO_2.
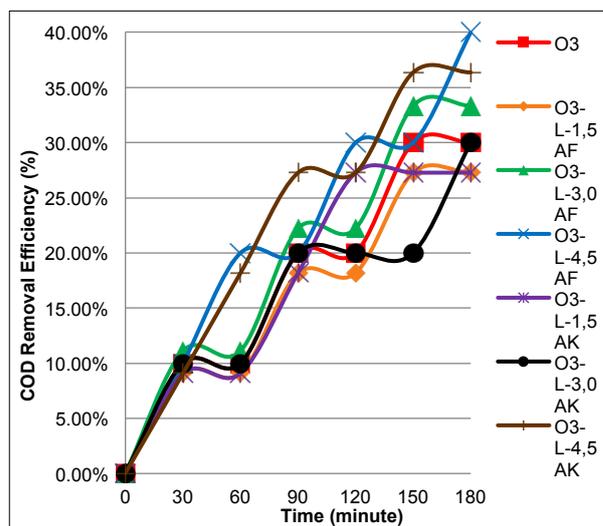

Figure 7. COD Removal Efficiency.

4 Conclusion

Several important conclusion can be drawn as follows:
1. The addition of sludge as a catalyst in the ozonation process can increase the ozone decomposition into $OH^{\bullet}$, thereby increasing the efficiency for removing organic and inorganic materials in leachate.
2. Sludge as a physically and chemically activated catalyst, in this study, showed a slightly different role in removal of pollutants. The physically activated catalysts were best to remove EC and COD. The chemically activated catalysts were best to remove turbidity.
3. The difference in sludge weight used during the research had an effect on the rate of removal, with more catalyst was added resulting in the higher the rate of removal, and the faster the decomposition of ozone.

4. Catalyst efficiency is an important parameter for commercial use. Further research is needed to determine the effect of catalyst if it is used more than once, and for optimum weight variation in leachate processing. Eventually the efficiency of a catalyst determine its practical applications.

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