

Comparison and Optimization of ozone – Based Advanced Oxidation Processes in The Treatment of Stabilized Landfill Leachate

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ABSTRACT-Leachate pollution is one of the main problems in landfilling. Among the most problematic parameters in stabilized leachate are COD, ammonia, and color. The treatment technology that can be used may differ based on the type of leachate produced. Even after treatment, the effluent characteristics are always hard to comply with the discharge standard. Ozonation is one of the chemical processes that can be used in the treatment of landfill leachate. However, its performance when use alone is low; its effectiveness can be improved using advanced oxidants. To date, application of Fenton and persulfate reagents separately to improve ozonation process in one ozone reactor was not well established. The study aimed to evaluate and compare the performance of the three treatment processes, namely ozone, ozone/Fenton and ozone/persulfate in treating stabilized leachate separately at different experimental conditions. The performance of the three methods in the treating stabilized leachate was compared. According to the results, the performance of ozone alone was poor, and utilizing new advanced oxidation material during ozonation of such leachate was required to improve leachate treatability. Ozone/Fenton process is a viable choice for degrading and decolourizing stabilized leachate. Furthermore, ozone/persulfate process has higher performance in ammonia removal as well as it has good removal efficiency of COD and color from stabilized leachate. Suitable data for establishing fully stabilized leachate treatment plant using ozone/Fenton and ozone/persulfate was suggested. The final effluent of ozone/Fenton process complied with the discharge standard for COD and colour.

Index Terms - Advanced oxidation process, Ozonation, Fenton, Persulfate, treatment efficiency.

I. Introduction

Growing population and industrial development have increased waste generated by urban areas and otherwise. In most countries, sanitary landfilling is the most common way of eliminating municipal solid waste (MSW) (Renou et al., 2008) [1]. MSW is waste from domestic, commercial, and industrial activities in urban areas (Bartone 1990) [2]. Sanitary landfilling is the most economical and environment-friendly method for disposing municipal and industrial solid waste (Tengrui et al., 2007)[3].

Malaysia generates about 6.2 million tons of solid waste per year, which equals approximately 25,000 tons per day. This amount is expected to increase to more than 31,000 tons per day by 2020 because of increasing population and per capita waste generation (Yahya 2012)[4]. Food, paper, and plastic constitute 80% of the overall weight of Malaysian waste (Manaf et al., 2009)[5]. The average amount of MSW generated in Malaysia is 0.5 kg/capita/day to 0.8 kg/person/day, and that in major cities is as high as 1.7 kg/capita/day (Kathirvale et al., 2003)[6]. Despite the many advantages of landfilling, the resulting highly polluted leachate has been a cause of significant concern, especially because landfilling is the most common technique of solid waste disposal (Ghafari et al., 2005)[7]. Landfill leachate is liquid that has seeped through solid waste in a landfill and extracted dissolved or suspended materials in the process. The environmental impact of leachate depends on leachate strength, proper leachate collection, and the efficiency of leachate treatment. Leachate contains high amounts of organic compounds, ammonia, and heavy metals and sometimes contaminates ground and surface water (Christensen et al., 2001)[8]. Landfill leachate usually contains a complex variety of materials and organic compounds, such as humic substances, fatty acids, heavy metals, and many other hazardous chemicals (Schrab et al., 1993)[9]. Researchers worldwide are still searching for a total solution to the leachate problem. Multiple-stage treatments are still required to remove leachate pollution thoroughly. No single method can effectively remove all pollutants simultaneously. Treatment by a conventional water treatment system (i.e., a combination of sedimentation, biological treatment, filtration, and carbon adsorption) cannot remove salts or organics, such as harmful recalcitrant compounds. This research aims to establish new technology and knowledge in stabilized leachate

treatment by using ozone – based advanced oxidation processes (ozone, ozone/Fenton, and ozone/persulfate) to reduce treatment time and improve the efficiency of treatment by increasing oxidation potential.

II. Materials and methods

A. ozone oxidation

Ozone experiments were conducted in a 2 L sample using an ozone reactor with a height of 65 cm and an inner diameter of 16.5 cm. The reactor was supported by a cross column ozone chamber to enhance the ozone gas diffusion (Figure 3.8 A and B). The water bath and cooling system supported the ozone reactor in keeping the internal reaction temperature at $<15^{\circ}\text{C}$ as an optimal half-life of the dissolved ozone (30 min) in water (Lenntech, Water treatment solution, 2012). Ozone was produced by a BMT 803 generator (BMT Messtechnik, Germany) fed with pure dry oxygen with recommended gas flow rate of 200–1000 ml/min $\pm 10\%$ under 1 bar pressure. The recommended input ozone gas concentration (30 – 80 in g/m^3 non-thermal plasma (NTP) $\pm 0.5\%$) was measured by an ultraviolet gas ozone analyzer (BMT 964). The initial pH of leachate samples was adjusted at different pH values ranges between 3 and 11, in order to investigate an optimal initial pH in treating stabilized leachate by ozone. The reaction time was varied between 10 and 120 min to determine an optimal ozonation time (Tizaoui et al., 2007)[10].

B. Ozone/Fenton in the advanced oxidation process

Fenton reagent ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$) was employed in the advanced oxidation during the ozonation of stabilized leachate. H_2O_2 (30%) and ferrous sulfate heptahydrate ($\text{Fe}_2\text{SO}_4 \cdot 7\text{H}_2\text{O}$, 278.02 g/mol) were used in preparing the Fenton reagent, which was then added to the leachate sample in the ozone reactor as one reaction process.

C. Ozone/persulfate in the advanced oxidation process

Persulfate ($\text{S}_2\text{O}_8^{2-}$) as sodium persulfate ($\text{Na}_2\text{S}_2\text{O}_8$, $M = 238$ g/mol) was employed in the advanced oxidation during the ozonation of stabilized leachate, which was added to the sample in the ozone reactor as one reaction process

D. Biodegradable and soluble COD fractions

The effects of the three ozonation treatment processes such as ozone alone, ozone/Fenton and ozone/persulfate on biodegradable and soluble characteristics of stabilized solid waste leachate were investigated in this research. The fractions of biodegradable COD_(bi), non-biodegradable COD_(ubi), soluble COD_(s), biodegradable soluble COD_(bsi), non-biodegradable soluble COD_(ubsi), and particulate COD (PCOD) were examined and calculated before and after each ozonation treatment processes.

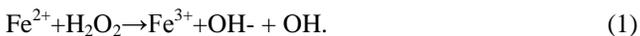
III. Results and dissection

A. Comparison of the three oxidation processes

The comparison of different ozone oxidation processes is of interest to determine the best removal performance of COD, colour and ammonia, as well as enhancing of biodegradability and their effects on COD fractions of stabilized leachate. The aim of this study was to evaluate the above mentioned approaches in terms of reduced organic load and ammonia, decreases colour, and enhances the biodegradable characteristics of stabilized leachate. To investigate the performance of combined ozone application and two advanced oxidant reagents, stabilized leachate was treated with ozone oxidation alone, ozone/Fenton and ozone/persulfate in the AOPs, respectively.

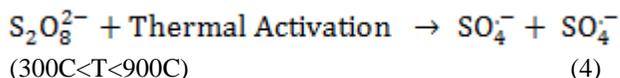
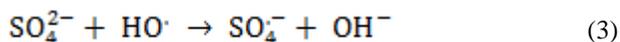
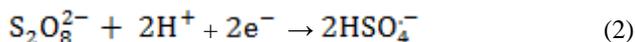
1. Comparison on COD, colour and NH₃-N removal

The three ozone oxidation processes are compared in Figure 1 in terms of COD, colour, and ammonia reduction based on optimal operational conditions. In the O₃/H₂O₂/Fe²⁺ system, the Fenton ions reacted with H₂O₂, resulting in the formation of hydroxyl radicals (·OH) (Eq. (1)). ·OH has the potential to destroy and degrade organic pollutants (Hermosilla et al., 2009)[11].



The reaction of ozone with H₂O₂ generates ·OH radicals. H₂O₂ is also dissolved in water and dissociates into the hydroperoxide ion (HO²⁻), which rapidly reacts with ozone to initiate a radical chain mechanism that generates hydroxyl radicals (Stahelin et al., 1982; Glaze et al., 1987)[12,13]. The removal efficiency of the target parameters was generally decreased with increasing Fenton molar ratio.

In Ozone/persulfate reaction; the Na₂S₂O₈ dosage was fixed as a COD/S₂O₈²⁻ ratio (g/g), namely, 1/1 to 1/10 during 60 min ozonation of leachate (Fig. 3), to evaluate the role of S₂O₈²⁻ in ozonation improvement. Persulfate oxidation can be enhanced by the release of sulfate radicals, which have powerful effects on the oxidation of organics (Watts 2011)[14]. The generation of sulfate radicals during oxidation can be significantly enhanced by catalysts, such as heat and UV radiation (Eq. 2 - 4), which were found to improve the persulfate oxidation potential (Gao et al., 2012; Abu Amr and Hamidi 2012)[15, 16].



Shiyong et al. (2009)[17] initiated sulfate radical by activating persulfate, using Microwave (MW)–activated persulfate oxidation. The effectiveness of the process was evaluated by the degradation of organics in wastewater; 83% to 95% of COD removal was obtained.

Consequently, to investigate the performance of ozone in initiating sulfate radical formation from Persulfate during the ozonation process, the influence of Na₂S₂O₈ concentration on COD, color and NH₃-N was investigated with the optimal ozone conditions fixed at 15 °C, max O₃ dosage 80 g/m³, reaction time 60 (min), and high pH (8.5) (Abu Amr et al., 2013)[18].

The results revealed that the proposed ozone/Fenton process was much more effective in removing COD and colour compared with other treatment processes, whereas ozone/persulfate was much more effective in removing NH₃-N. Furthermore, the final effluent concentrations of the ozone/Fenton process for COD (392 mg/L) and colour (60 Pt-Co) met the acceptable discharge levels (COD 400 mg/L, colour 100 Pt-Co) according to Environmental Malaysian Regulations 2009 for solid waste landfill control. Although the rate of removal of NH₃-N by using ozone/persulfate was much higher than that of other processes, the final effluent concentration of NH₃-N (198 mg/L) did not meet the maximum acceptable level (5 mg/L).

The reaction time of treatment processes is an important factor for saving energy and reducing treatment cost. The optimal reaction time consumed during the ozone/Fenton process was very short (90 min) compared with the time consumed by the ozone/persulfate process (210 min) (Figure 1). Although the optimal reaction time for ozone alone (60 min) was shorter than that of ozone/Fenton, the treatment efficiency of ozone alone was very low. Based on the comparative experiments, the ozone/Fenton process reduced the reaction time and improved the effectiveness of ozone in reduction of organic load and decolorizing stabilized leachate.

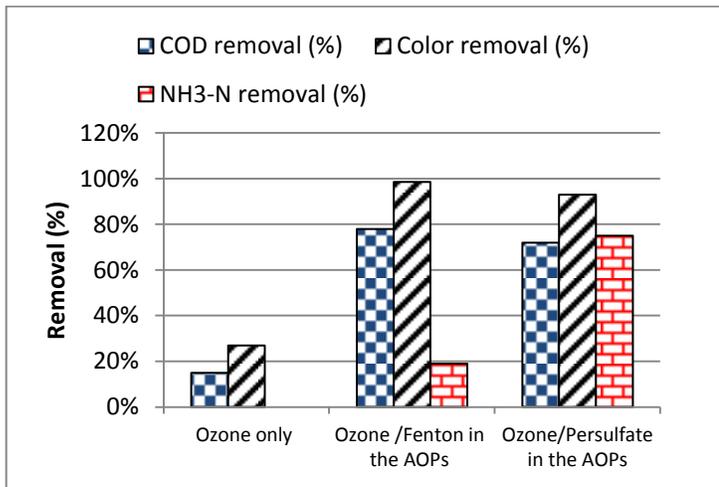


Figure 1: Comparing the performance of persulfate advanced oxidation of ozone with other applications for the treatment of stabilized leachate

In comparing the optimal conditions for the ozone/Fenton process (90 min in one treatment stage with 78% COD removal) with those of Fenton oxidation followed by ozonation, the results showed that around 50% of the reaction time was reduced, and removal efficiency increased from 58% and 95% to 78% and 98.5% for COD and colour, respectively. Goi et al. (2009) obtained a 77% COD removal rate after 240 min of ozonation after Fenton oxidation.

2. Comparison on Ferrous and Sulfate residual

Fenton and persulfate were used as advanced oxidant reagents during ozonation. Both reagents release ferrous and sulfate ions, respectively, during ozone oxidation. Thus, iron ions as Fe^{2+} mg/L and sulfate ions as SO_4^- mg/L were examined to evaluate the final effluent after each of the three experimental processes

(Figures 4.42 and 4.43). The raw leachate used in the experiments had significant concentrations of ferrous (9 mg/L) and sulfate (187 mg/L) before treatment. The ferrous level was exceeding the acceptable level (5 mg/L) set by the Environmental Regulations (2009) [19] for solid waste control and discharge. Mohajeri (2010)[20] reported that iron ranged from 6.3 mg/L to 6.6 mg/L for the same type of leachate.

Ferrous can be removed from wastewater by employing different physical and chemical processes, such as aeration, softening, chlorination, electro-Fenton, and filtration (Arslan and Bayat, 2009; Aziz et al., 2004; Mohajeri et al., 2010) [21 – 23]. However, ozone alone reduced iron only from 9.0 mg/L to 7.5 mg/L. The optimal amount of iron ions used in the ozone/Fenton experiments was 1,160 mg/L, and the residual iron concentration was 3.5 mg/L. The major part of the iron content precipitated as iron sludge during settling after ozonation.

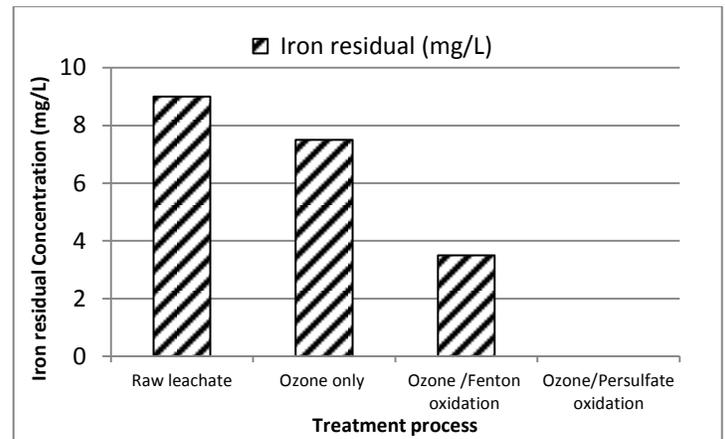


Figure 2: Iron ions residual in the effluent of the three ozonation process

Eren and Acar (2006)[24] reported that all iron species precipitate as ferric hydroxide at pH above 8. The iron ions were completely removed by using the ozone/Fenton process under optimal conditions of pH 10, 1g/1g COD/S_2O_8 ratio, O_3 30g/m³, and reaction time of 210 min. Thus, the ozone/persulfate process is an efficient method for removing iron from stabilized leachate.

As shown in 3, stabilized leachate contains a specific amount of sulfate ions (187 mg/L). Around 20% removal rate of sulfate was achieved by the ozone-alone process, whereas the ozone/persulfate scheme increased the concentration of sulfate (520 mg/L) because of the

sulfate ions released during oxidation. Based on different national environmental standards (Lebanon National Standard for Environmental Quality, 2001[19] the maximum acceptable level of sulfate in treated wastewater effluent is 1,000 mg/L. Nevertheless, sulfate ions can be uptaken by bacteria during aerobic and anaerobic treatment conditions (Monheimer, 1975; Meulepas et al., 2009)[25, 26].

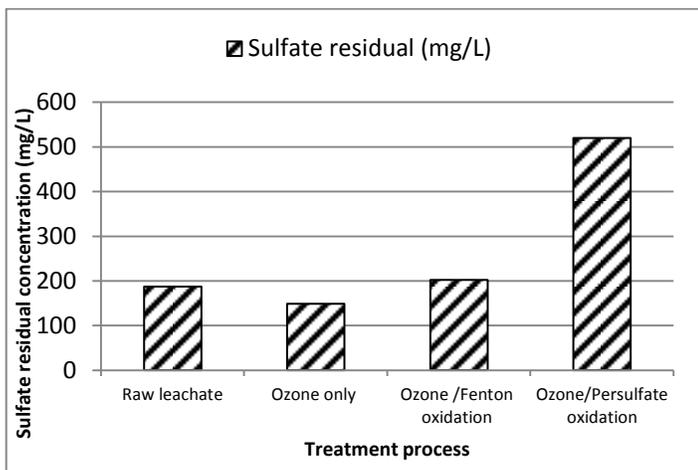


Figure 3: Sulfate residual in the effluent of the three ozonation processes

3. Comparison on biodegradability

The variation of biodegradability (BOD₅/COD) in leachate is attributed to the different types of leachate according to landfill age and leachate decomposition. The BOD₅/COD ratio in young (<5 years), intermediate (5 to 10 years), and stabilized (>10 years) leachate is >0.3, 0.1 to 0.3, and <0.1, respectively (Schiopu et al. 2010). Stabilized leachate with very low biodegradability (BOD₅/COD=0.034 to 0.05) and very strong organics made the biological treatment difficult. Different ozone applications have been used to enhance the biodegradability of landfill leachate (Tizaoui et al., 2007; Bila et al., 2004; Cortez et al., 2011b; Cortez et al., 2011a) [10, 27 – 29]. However, the performance of ozone alone in improving the ratio was still very low. Based on the results, the ozone/persulfate process is an efficient method for enhancing the biodegradability of stabilized leachate (Table 1).

TABLE 1
Comparison the effect of the three ozone applications on biodegradability

Process	BOD ₅ /COD
Raw leachate	0.034 – 0.05
Ozone alone	0.06
Ozone/Fenton	0.14
Ozone/Persulfate	0.29

4. Comparison the effect of COD fractions

COD fractionation is the most important parameter for leachate quality. However, the effects of ozone applications on these fractions have not been evaluated. The effects of the three ozone applications on COD fractions in stabilized leachate are compared in this section (Table 2). The quantity of biodegradable and soluble COD fractions in raw leachate was relatively low, whereas that of non-biodegradable, non-soluble, and particulate fractions was high. As shown in Table 2, each treatment process improved the biodegradable soluble and biodegradable soluble COD fractions but reduced the non-biodegradable, particulate, and non-biodegradable soluble fractions. Based on the results, ozone/persulfate is an efficient process to improve biodegradability and solubility of organics in stabilized leachate.

TABLE 2
Comparison the effect of the three ozone applications on COD fractions

Fraction \ Process	Process			
	Raw leachate	Ozone Only	Ozone/Fenton	Ozone/Persulfate
Biodegradable COD (%)	24	28	36	39
Non-biodegradable COD (%)	76	72	68	61
Soluble COD (%)	59	65	72	72
Particulate COD (%)	41	35	28	28
Biodegradable soluble COD (%)	40	43	51	55
Non-biodegradable soluble COD (%)	60	57	49	45

5. Comparison on ammonia removals during aeration as a post treatment stage

The removal of ammonia from leachate before and after ozonation during batch aeration as a post-treatment stage was documented to evaluate and compare the performance of the three ozonation processes. In raw stabilized leachate, full removal of ammonia was obtained after 7 days of aeration, but ammonia removal did not significantly improve after the ozone-alone process (Figure 4). In the ozone/Fenton process, ammonia was completely removed after 4 day of aeration, and removal significantly improved during the first day of aeration (69%) (Figure 5). Therefore, the performance of Fenton oxidation alone in ammonia removal was much poorer. In the ozone/persulfate process, ammonia was completely removed after only 2 days of aeration (Figure 6). Around 92% removal rate of ammonia was also achieved after the first 24 h of aeration compared with the 52% and 69% after the ozone-alone and ozone/Fenton processes, respectively. Moreover, the performance of persulfate oxidation alone in ammonia removal was much poorer. Different applications have been reported for ammonia removal during aerobic and anaerobic biological processes of leachate. Gotvajn et al. (2009)[30] obtained an 86% removal rate for ammonia after 50 h of aeration at pH of 11.

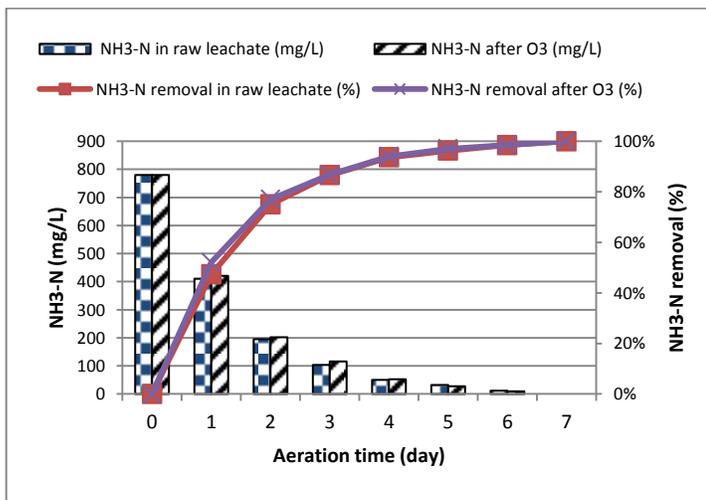


Figure 4: Ammonia removal from leachate before and after ozone alone by aeration process

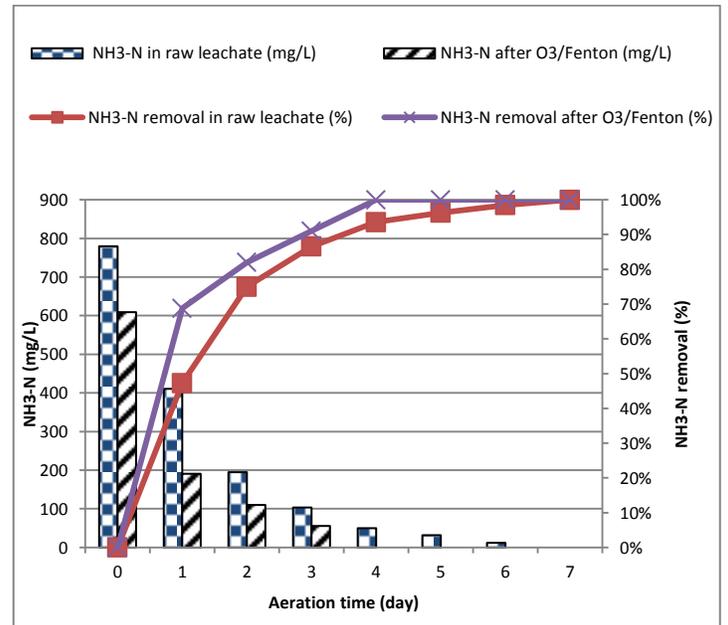


Figure 5: Ammonia removal from leachate before and after ozone/Fenton treatment by aeration process

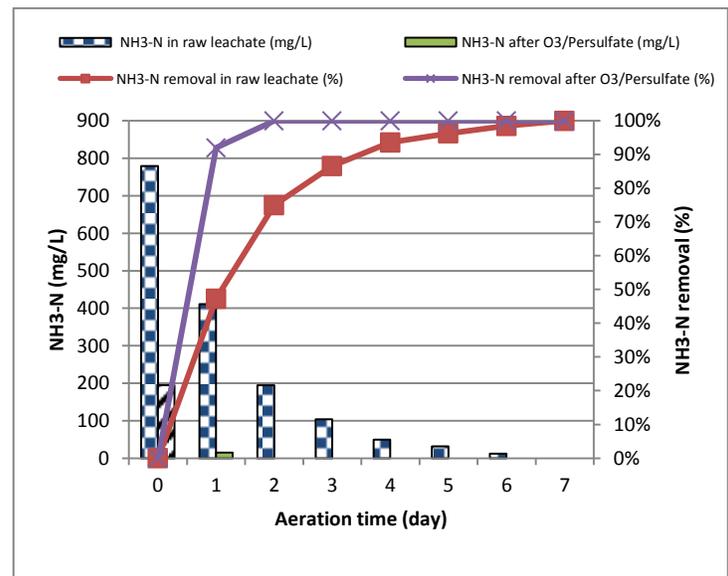


Figure 6: Ammonia removal from leachate before and after ozone/persulfate treatment by aeration process

Leachate is formed when water mainly from rain infiltrates deposited waste. As the liquid moves through the landfill, many organic and inorganic compounds, such as ammonia and heavy metals, are

transported into the leachate. The leachate then moves to the surface or base of the landfill cell and may pollute the surface and groundwater, which may affect human health and aquatic environment. Many factors affect the quality and quantity of leachate, such as seasonal weather variation, landfilling technique, waste type and composition, and landfill structure (Mohajeri, 2010)[20]. Leachate pollution in Malaysia is very serious, and the high generation of landfill leachate in tropical areas such as Malaysia is mainly attributed to the high amount of rainfall (Lema et al., 1988)[31].

IV. Conclusion

The performance of the three ozonation techniques in AOP, namely, ozone alone, ozone/Fenton, and ozone/persulfate treating stabilized leachate was investigated and compared. According to the results, the performance of ozone alone was poor, and utilizing new advanced oxidation material during ozonation of such leachate was required to improve leachate treatability. Ozone/Fenton in AOP is a viable choice for degrading and decolourizing stabilized leachate. This process was found to be ideal because it can achieve 99% of colour removal and 79% of COD removal and up to 50% reduction in treatment time compared with the classical combination of Fenton and ozone processes. The removal efficiency was also higher. The process achieved a desired OC value for COD removal (0.29 Kg/Kg COD) compared with other methods. Furthermore, the process reduced iron ions (3.5 mg/L) to lower than the maximum acceptable levels (5 mg/L). Moreover, biodegradability (BOD₅/COD ratio) was significantly improved, as were biodegradable and soluble organic fractions. Ozone/persulfate in AOP significantly removed NH₃-N, COD, and colour. The process achieved high biodegradability (BOD₅/COD; 0.29) compared with other treatment methods, which suggests further organic degradation via biological process as a post-treatment. The performance of the ozone/persulfate process to improving biodegradable and soluble organic fractions in stabilized leachate was better than that of other processes. Furthermore, the process completely removed iron ions from stabilized leachate and produced no undesirable sludge. Sulfate ions are not harmful to the environment, and sulfates can decompose in further biological processes. biodegradability (BOD₅/COD) enhanced from 0.034 to 0.05, 0.14 and 0.29, after ozone alone, ozone/Fenton and ozone/persulfate, respectively. The results reveal that ozone/persulfate in AOP achieved higher ratio among the three treatment process which recommended suggesting biological process as a post treatment for further organic degradation and ammonia removal. The effect of the three ozonation processes on COD fractions in stabilized leachate was documented in this research. Based on the results, the ozone/persulfate process is

the best choice for improving biodegradable and soluble fractions in stabilized leachate.

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