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Original Research Article

Landfill Leachate Treatment with Ozonation to Improve Biodegradability

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Abstract
Leachate is a liquid that comes from processed waste generation so that it contains compounds that are harmful or toxic to the environment. The quality of the leachate obtained included COD: 3,315 mg/l, BOD: 339 mg/l, TSS: 216 mg/l, and color 3,1210 Pt.Co. Waste processing site Temesi has a BOD/COD ratio of 0.11, this value is not suitable for using biological treatment because the biodegradability value is small so it is difficult for microorganisms to degrade existing pollutants. Therefore, it is necessary to have an pretreatment that is used to increase the value of biodegradability, one of which is oxidation using ozone. Ozone has a strong oxidizing value so that it can produce free radicals to break long chains of pollutants so that they become simpler. This research on ozone oxidation resulted in an increase in the BOD/COD ratio from 0,1 to 0.14 with a generator capacity of 36 g/hour. In this study, an ozone generator of 100 G/hour was used which increased the BOD/COD ratio to 0.46 with a final COD yield of 815 mg/l increasing BOD to 379 mg/l, TSS become 157,62 mg/L and color become 23.642 Pt.Co.

Keywords: Leachate, biodegradability, ozone

1. Introduction
Leachate is liquid waste that arises due to the entry of external water into the waste generation, dissolving and rinsing dissolved materials, including organic matter resulting from biological decomposition processes (Damanhuri E. , 2010). In general, leachate is a liquid that comes from piles of garbage that contain hazardous compounds. If leachate is left alone in the environment it will result in negative impacts such as environmental pollution which results in the health and quality of living things. The leachate in the landfill has different characteristics depending on the dominant type of waste in the landfill. Leachate does not only come from the waste decomposition process but can also come from rainwater and drainage channels that seep into the waste. So that the quantity of leachate is also influenced by the rainfall factor in the landfill area. Leachate has unique characteristics, that is the high content of organic matter, metals, acids, dissolved salts, and micro-organisms.

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methanogenesis (10-20 years), and the maturation phase (> 20 years) (Aziz, 2010). The required leachate treatment system must be able to process the characteristics of the leachate in the TPA. If there is no appropriate leachate treatment system, the leachate can enter the ground or empty into rivers so that it can contaminate groundwater and rivers.

Disposal of domestic or commercial solid waste in the landfill area produces leachate products derived from moisture in the waste and external infiltration fluids. Leachate characteristics change from time to time due to the age of the landfill and changes in the waste decomposition phase from aerobic to anaerobic. In anaerobic decomposition there are two phases, namely the acidogenesis phase and the methanogenesis phase. Leachate originating from the acidicgenesis phase has the characteristics of high organic pollutants. The high organic content contains molecules of volatile fatty acids (VFAs) derived from the decomposition of carbohydrate content in landfills. The carbohydrate content comes from household waste and food waste that goes to landfill. This molecule contains a low weight of organic acids (MW < 120) and is the majority content of leachate with a percentage of 95% (Johansen and Carlson, 2001). Due to the high content of VFAs, the leachate biodegradable level is high with a BOD/COD ratio of 0.4-0.5 or even higher (Diamadopoulos, 2004). The high content of VFAs in the leachate results in a pH value below 4. The acidic conditions in the leachate cause the heavy metal content in the leachate to increase (Johansen, 2001).

Meanwhile, for landfills originating from the methanogenesis phase, the methane gas content will produce bacteria which cause a decrease in the VFAs content and reduce the organic content in leachate. The remaining organic acids in this phase are substances with high molecular weight. These substances receive little degradation from microbes so they generally tend to be in old landfills (> 10 years). In this methanogenesis phase, BOD will be degraded more quickly so that the leachate BOD/ COD ratio will decrease to 0.1-0.3. The decrease in the VFAs content in the leachate resulted in an increase in the pH content in the leachate which is more than 7. The leachate treatment required for leachate characteristics from young landfills is biological treatment (aerobic and anaerobic) without the need for chemical treatment. Whereas leachate processing with old landfill characteristics will require chemical processing assistance to increase the biodegradability of the leachate.

Chemical oxidation is a treatment unit used to degrade toxic content in wastewater. Chemical oxidation uses chemical elements in the process such as ozone (O_3), hydrogen peroxide (H_2O_2), permanganate (MnO_4), chlorine dioxide (ClO_2), chlorine (Cl_2) and even oxygen (O_2) without the role of microorganisms in the process (Eckenfelder, 2000) . Chemical oxidation is usually used to 1) increase the treatability of non-biodegradable (refractory) components, 2) remove disturbances for the development of microorganisms such as organic and non-organic compounds 3) remove or reduce the toxic content of organic and non-organic components for the growth of microorganisms and aquatic systems (Tchobanoglous, 2003).

One of the oxidizers used in chemical oxidation is ozone (O_3). Ozone is a strong oxidizing agent (EH > 2.0 volts) and is commonly used for the disinfection of wastewater treatment plants (Tchobanoglous, 2003). Ozone gas is stable at all temperatures and pressures. With the excess of ozone so that ozone is well for being oxidizer. Ozone is generated from an ozone generator which is derived from pure oxygen in the gas stream. The decomposition process of ozone in water, especially at high pH, can produce free radicals: $O_3 \rightarrow O\cdot + O_2$ (n = 1) (Eckenfelder W. , 2000). Then O· will react with water to form OH· (hydroxyl radical). This hydroxyl radical will function to break down the bonds of non-biodegradable chemical compounds. According to Eckenfalder (2000) there are three mechanisms in the ozonation process including:

1. Oxidation of alcohols to aldehydes and then to organic acids
   $RCH_2OH \rightarrow RCOOH$
2. Substitution by an oxygen atom into an aromatic ring
3. Termination of carbon double bonds

The process of forming OH· in the ozone oxidation process occurs in direct and indirect conditions (Malik, 2020). The indirect reaction process involves radical ions where ozone decomposition occurs which is accelerated by the initiator in the form of OH- to form hydroxyl radicals (OH·). There are three stages in the formation of hydroxyl radicals in the indirect process, namely the initiation stage, the radical stage and the termination stage. Meanwhile, in the direct reaction at the ozonization stage, a selective reaction occurs with (k = 1.0 – 103 M-1 s-1). The reaction that occurs in direct reaction ozonation is Cyclo Addition (Criegee Mechanism). In this mechanism, the dipolar structure, namely ozone, will experience cyclo addition with double bond compounds (unsaturated) to form ozonide. In the presence of a protonated solution such as water, ozonide (I) integrates into aldehydes, ketones, or zwitter ions. The presence of this ion will cause integration into hydrogen peroxide and carboxyl groups.

2. Methods

This research was carried out on a laboratory scale at the Diponegoro University Environmental Engineering Laboratory. This experiment uses an ozone generator with specifications 36G/Hour 220V 3 mgs-1. The reactor used was a 1L beaker with a volume of 600 ml of leachate. This study used a variation of the contact time between ozone and leachate with a difference in contact time every 15 minutes starting from the 15th minute to the 75th minute. The leachate used came from the waste processing site Temesi, Gianyar Regency, Bali Province. The characteristics of this leachate have already been tested at the Diponegoro University Environmental Engineering Laboratory.

In this experiment, small bubbling gas will be produced indicating that gas is coming out of the reactor (Nur Athikoh, 2021). Ozone produced from reaction pure oxygen and dry oxygen from corona discharge using an ozone generator. By using oxygen or enriching the process air in oxygen, the generating capacity of a given ozone generator can be increased by a factor ranging form 1.7 to 2.5 versus the production capacity with air (Langlais et al., 1991). Ozone gases will flow through diffuser paced at the bottom and producing small bubble gases. Gases will increase in proportion to the increase in ozone contact time with wastewater. The yield obtained when using an oxygen-enriched process gas is increased with a smaller gas space and an increased electrical current frequency. The next test is to test the results of ozone oxidation to find out whether ozone oxidation has succeeded in increasing the biodegradability of leachate. The parameters tested included COD, BOD, TSS, pH and color. In addition to these parameters, the temperature and pH values of the leachate before and after the ozone process were also checked to find out whether there were any changes caused by the ozone oxidation process.

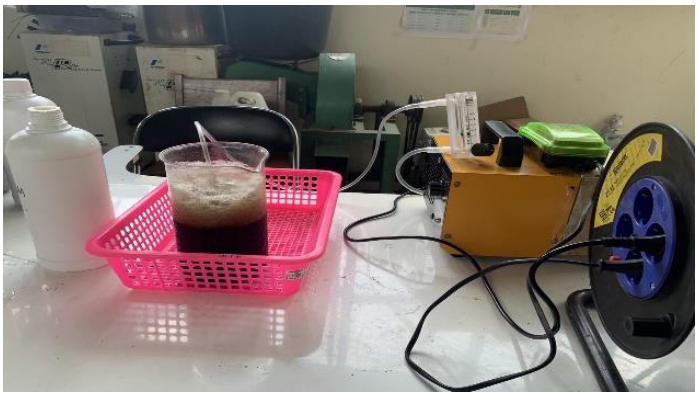

Figure 1. Process oxidation ozone

Temesi landfill is one of the landfills located in Gianyar Regency, Bali Province. Currently the Gianyar Regency Government, Bali Province is carrying out the revitalization of the Temesi TPA, one of the plans in revitalizing the TPA is the planning for the Leachate Processing Installation. Based on the results of the characteristic test that has been carried out, the BOD/COD ratio in the leachate is very small, that is 0.11 with a BOD value of 339 mg/l and a COD of 3,315 mg/l. This is because the

biodegradability content of leachate is very low, making it difficult to apply biological treatment (Tchobanoglous, 2003). Therefore, in this study a pre-treatment was carried out which could increase the biodegradability content of leachate (Daniele, 2004). The pre-treatment carried out was oxidation using ozone. Ozone oxidation was chosen because the resulting effluent can quickly decompose into oxygen and also leaves no residue in the environment. In addition, oxidation using ozone can be easily carried out and does not require chemicals. In the characteristic test carried out using the following measurement methods:

Table 1. Leachate characteristics

No	Parameter	Result Test	Unit	Method
1	pH	7.89	-	SNI 6989.11:2019
2	Temperature	28.2	^{0}C	SNI 06-6989.23-2005
2	COD	3,315	mg/l	SNI 6989.2:2019
3	BOD	339	mg/l	SNI 6989.72.2009
4	TSS	216	mg/l	SNI 6989.3:2019
5	Colour	31,210	Pt.Co	SNI 6989.80:2011
6	Ratio BOD/COD	0.11		

3. Result and Discussion

a. Effect of Ozonization on pH and Temperature

In the oxidation process there is an increase in pH from the beginning and end of the oxidation process. An increase in pH in this process indicates the presence of compounds that can react with ozone (Fernando, 1995). The compounds in the leachate consist of organic compounds and inorganic compounds. Organic compounds such as humic compounds, COD, BOD, Color and for the inorganic compounds such as ammonia, nitrate, and nitrite. This reaction occurs between the ozone product, namely OH·, and the compounds present in the leachate. OH· will be produced form direct reaction and indirect reaction. The reaction that occurs in direct reaction ozonation is Cycle Addition (Criegee Mechanism). In this mechanism, the dipolar structure, namely ozone, will experience cyclo addition with double bond compounds (unsaturated) to form ozonide. In the presence of a protonated solution such as water, ozonide (I) integrates into aldehydes, ketones, or zwitter ions. The presence of this ion will cause integration into hydrogen peroxide and carboxyl groups (Gunten,2003). Whereas the indirect reaction process involves radical ions where ozone decomposition occurs which is accelerated by the initiator in the form of OH- to form hydroxyl radicals (OH·). There are three stages that occur in the indirect reaction process, namely the initiation stage, the radical stage and the termination stage. The following is the mechanism of the indirect reaction in the ozone oxidation process.

1. Initiation Stage

   At this initiation stage, a reaction occurs between hydroxide ions (OH-) and ozone (O_3) to form radical superoxide anions (O_2·) and hydroperoxide radicals (HO_2·)

   $O_3 + OH^- \rightarrow O_2\cdot + HO_2\cdot$ (2- 8)

2. Radical Stage

   In this second stage, it is characterized by the production of radical ozone anion (O_3·) through the reaction between ozone and superoxide radical anion (O_3·) then rapidly decomposes into hydroxyl radical (OH·) with the following reaction

   $O_3 + O_2\cdot \rightarrow O_3\cdot + O_2$ (2- 9)

   The value of O_3· is comparable to HO_3·

   $HO_3\cdot \rightarrow OH\cdot + O_2$ (2- 10)

   Furthermore, the hydroxyl radical that is formed reacts with ozone through the reaction:

   $OH\cdot + O_3 \rightarrow HO_4\cdot$ (2- 11)

   $HO_4\cdot \rightarrow O_2 + HO_2\cdot$ (2- 12)

Then the hydroxyl radical reacts with the organic molecule R, as a chain carrier or also known as a promoter. The organic molecule R which reacts with OH· will form a radical organic component (R·).

$H_2R + OH· \rightarrow HR· + H_2O$ (2- 13)

If the above reaction meets with oxygen, peroxy radical organic molecules will be produced (ROO·).

$HR· + O_2 \rightarrow HRO_2·$

$HRO_2· \rightarrow R + HO_2·$

$HRO_2· \rightarrow RO + OH·$ (2- 14)

3. Termination Stage

In the final stage, namely the termination stage, organic and inorganic compounds react with OH· to form secondary radicals without being followed by the formation of $O_2·/HO_2·$. This inhibitor can terminate the chain reaction and prevent the ozone decomposition process from occurring.

$OH· + CO_3^{2-} \rightarrow OH^- + CO_3·^-$

$OH· + HCO_3^- \rightarrow OH^- + HCO_3·$ (2- 15) (Malik, 2020)

An increase in pH from the range of 8-9 will accelerate the ozone decomposition process that occurs in leachate. From the process that has been carried out, it shows that the highest increase in pH occurred in the 60th minute of 8.98. Increasing the pH indicates that more and more hydroxyl radicals are produced. The increase in pH in the process indicates that the dominant reaction mechanism is the presence of OH. Based on the graph below, the 60th minute produces the highest hydroxyl radicals. Then the temperature parameter does not change every minute of the experiment. The constant leachate temperature is at 25^{0}C.

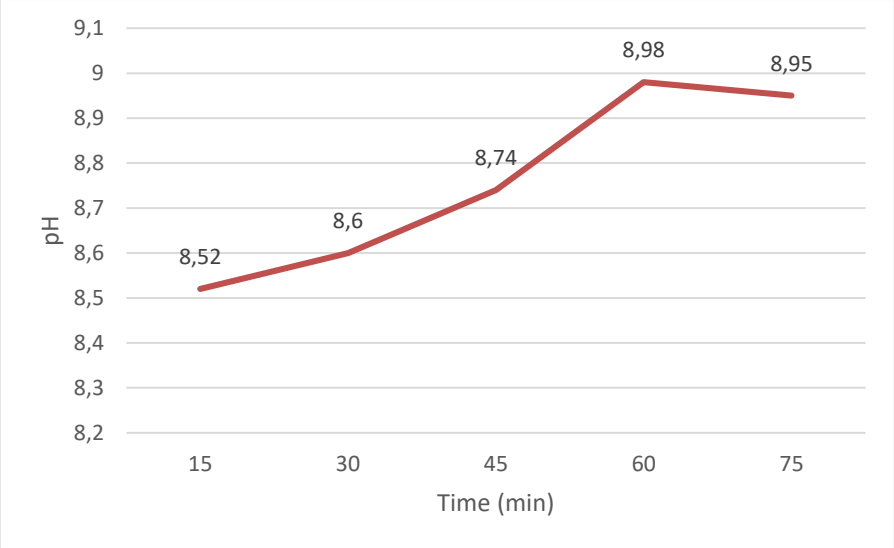

Figure 2. Effect of ozonization on Ph

b. Effect Ozonization on Color

For color removal results in a decrease in color concentration in proportion to the increase in time. This decrease in color concentration is caused by a decrease in organic content in the leachate as the basis for the formation of color, the color will be oxidized into compounds with simpler molecular structures (Nur Athikoh, 2021). In the experiment, the results of the color concentration at 0 minutes were 31,043 Pt.Co, 15 minutes produced 2,954.33 Pt.Co, 30 minutes produced 28,110 Pt.Co, 45 minutes produced 27,476 Pt.Co, 60 minutes produced 27,410 Pt.Co and at 75 minutes produced 27,176 Pt.Co. For the efficiency produced, namely the 15th minute produced an efficiency of 4.83%, 30 minutes produced 9.44%, 45 minutes produced 11.48%, 60 minutes produced 11.770% and 75 minutes produced 12.45%. The increase

in contact time between ozone and leachate causes more ozone to be supplied in leachate. The graph of decreasing color concentration is shown in the graph below.

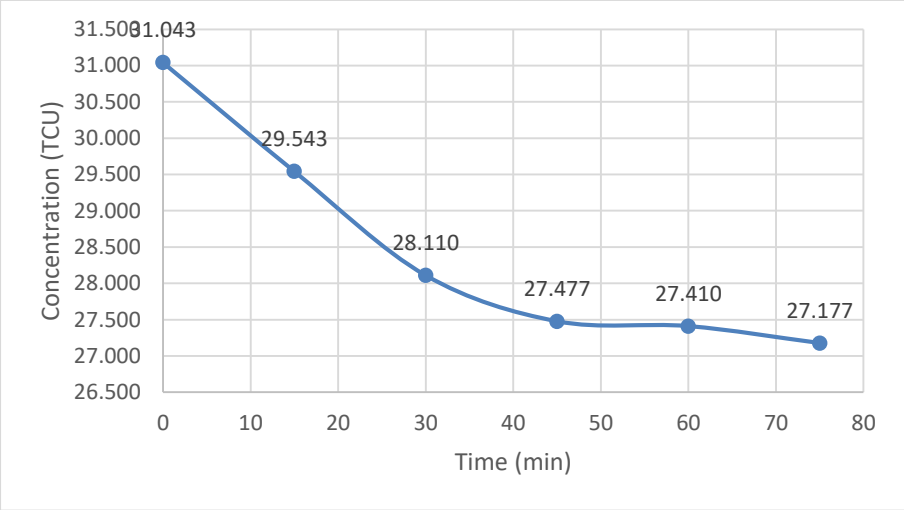

Figure 3. Effect of ozonization on color

c. Effect of Ozonization on TSS

The concentration of TSS in leachate results in a decrease proportional to the addition of time. The decrease in TSS was due to the reduced levels of organic and inorganic suspended solids retained on the paper. The higher the concentration of ozone contacted in the leachate, the more flocs that can absorb the colloids in the leachate (Purwadi A, 2006). In this experiment, the TSS concentration at 0 minutes was 216 mg/l, 15 minutes produced 208 mg/l with an efficiency of 3.7%, 30 minutes produced 196 mg/l with an efficiency of 9.2%, 45 minutes produced 192 mg/l l with an efficiency of 11.11%, 60 minutes produces 188 mg/l with an efficiency of 12.96% and 75 minutes produces 168 mg/l with an efficiency of 22.22%.

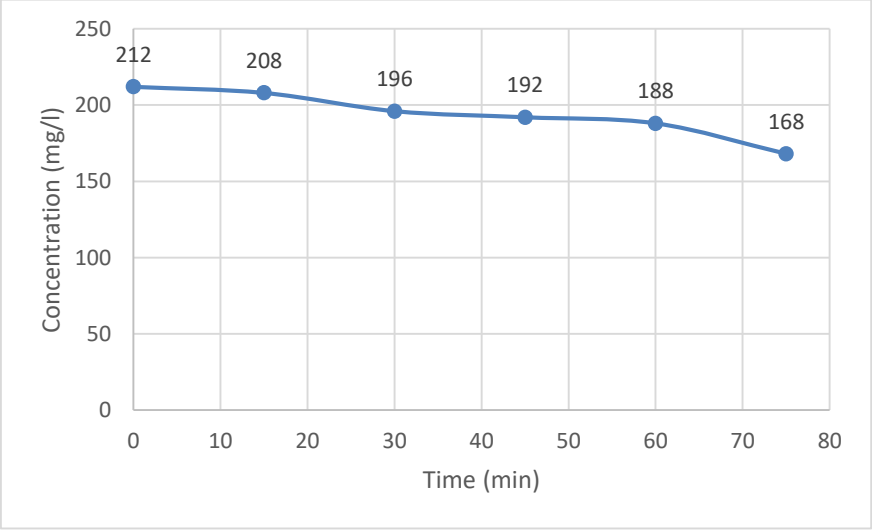

Figure 4. Effect of ozonization on TSS

d. Effect of Ozonization on COD

The greatest decrease in COD levels in leachate occurred in the 60th minute. The decrease in COD concentration was due to the content of hydroxyl radicals which simplified the non-biodegradable content in leachate. The higher the production of hydroxyl radicals, the greater the degree of simplification of the non-biodegradable concentration, which causes a decrease in the COD concentration (Susana Cortez, 2010). The decrease in COD concentration for contact time of 15 minutes,

30 minutes, 45 minutes, 60 minutes and 75 minutes was 29.86%, 31.37%, 29.36%, 37.9% and 34.38%. COD concentrations produced at 0, 15, 30, 45, 60, 75 minutes were 3308.83 mg/l, 2325 mg/l, 2275 mg/l, 2341.67 mg/l, 2058.33 mg/l, and 2175 mg/l.

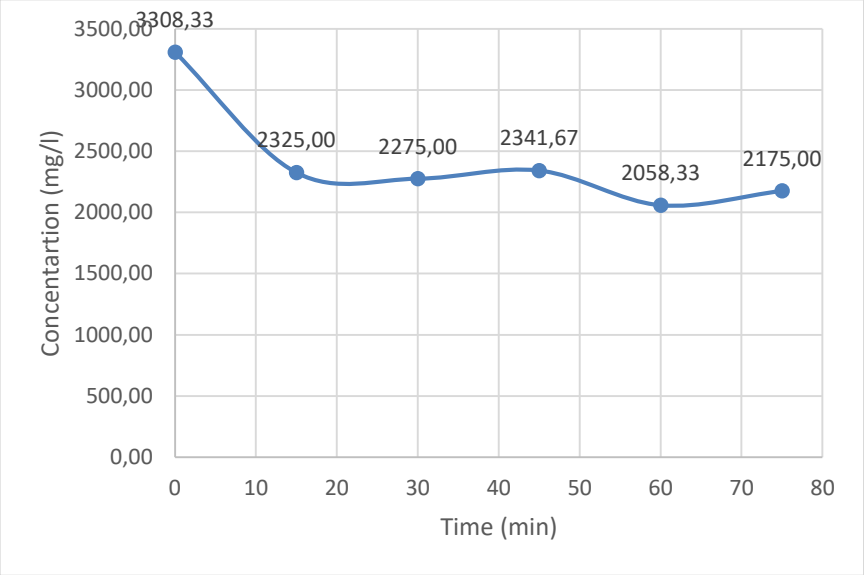

Figure 5. Effect of ozonization on COD

e. Effect of Ozonization on BOD

This experiment will result in an increase in BOD caused by hydroxyl radicals which can break down non-biodegradable ingredients into biodegradable ingredients. So that by increasing the biodegradable content, the BOD concentration will increase (Nuno Amaral & Silva, 2016). The highest increase in BOD content occurred in the 60th minute of 6.41%. The increased ozone concentrations produced in this experiment at 0, 15, 30, 45, 60, and 75 minutes were 277 mg/l, 278 mg/l, 279 mg/l, 289 mg/l, 296 mg/l, and 292 mg/l. The graph below illustrates the increase in BOD concentration in each time variation of the experiment.

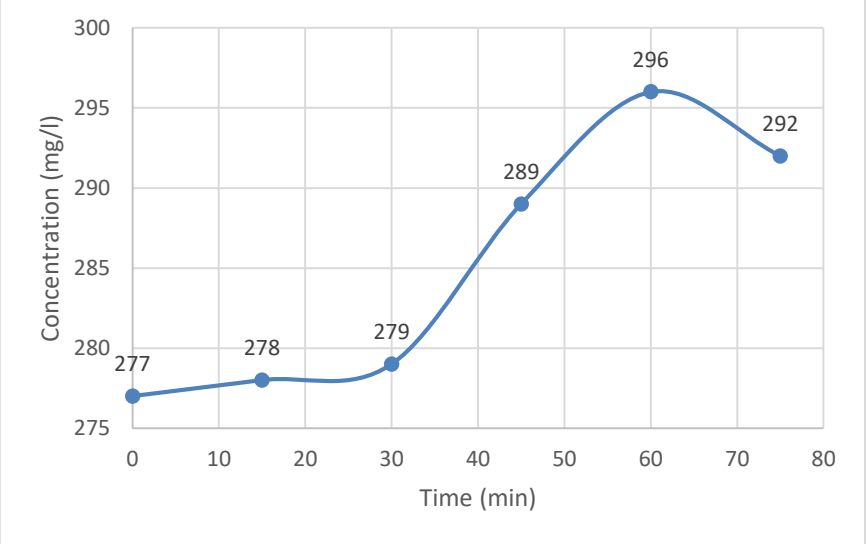

Figure 6. Effect of ozonization on BOD

f. Effect of Ozonization on Ratio BOD/COD

From the results of the oxidation that has been carried out, the results show an increase in the BOD/COD ratio with a decrease in COD concentration and an increase in BOD concentration. The initial BOD/COD ratio of the leachate was 0.11 and after oxidation it became 0.144 in time 60 min. So that in

this experiment it succeeded in increasing the level of the BOD/COD ratio in leachate. To achieve the desired level of biodegrability, you can increase the dose of ozone used so that it can produce more hydroxyl radicals (Daniele M.Bila, 2004). In this experiment the required ratio level to be able to treat leachate using a biological treatment unit is 0,4-1. According to Tcbanoglous, the ratio of BOD/COD ≥ 0.4 is safe to use in biological treatment units.

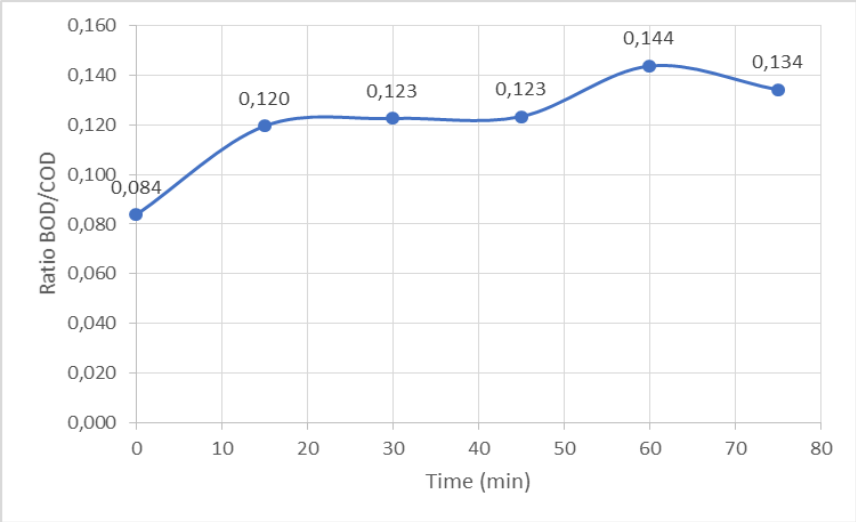

Figure 7. Effect of ratio BOD/COD

Based on the tests that have been carried out, the results show that the optimal time for ozone oxidation in an effort to reduce COD concentrations and increase BOD occurs at the 60th minute so that the removal efficiency used in this plan is at the 60th minute. In the tests carried out using an ozone generator with a capacity of 36 G with a flow rate of 3 mgs-1 which can degrade COD by 37.91%. The BOD/COD ratio obtained from the ozone experiment is 0.14. This value still does not meet the level of biodegrability for biological processing so as to achieve the desired leachate biodegrability value, the ozone concentration is increased. This increase in ozone concentration will result in a greater removal of COD concentrations (Tao Dong, 2015).

Tabel 2. Treatability results in the 60th minute

Parameter	Concentration (t=0)	Concentration (t=60)	Efficiency (%)	Increase/Decrease
COD	3308	2058	37,78	1250
BOD	277	296	6,86	19
TSS	216	188	12,96	28
Color	31043	27410	11,70	3633
BOD/COD	0,14			

Planning to increase the leachate BOD/COD ratio

b. The need for O3 in increasing biodegrability
Is known :
- BOD/COD ratio of leachate characteristics = 0.11
- BOD/COD ratio of oxidation results (X0) = 0.14
- Planning to increase the BOD/COD ratio (X1) = 0.4 (0.4-0.8) (Tchobanoglous, 2014)
- Process treatability ozone concentration (C0) = 36 G/hour.l

c. Wanted : The need for ozone concentration to produce a BOD/COD ratio of 0.4 (C1)
Resolution following equation (1)

$$\frac{X_0}{C_0} = \frac{X_1}{C_1} \quad \text{.....(1)}$$

$$\frac{0{,}14}{36\frac{G}{hour.l}} = \frac{0{,}4}{C_1}$$

$$C_1 = \frac{36\frac{G}{jam.l}\, x\ 0{,}4}{0{,}14}$$

= 102.85 G/hour.l

In the calculations performed to achieve the desired BOD/COD ratio, an ozone concentration of 102.85 G/hour.liter is required. Based on the ozone concentration that has been obtained, the removal efficiency results can be calculated using the generator used based on the loading concentration obtained in this experiment. Based on the results of the calculation of the loading load generated by ozone oxidation with a factor safety of 25%, it can be calculated the magnitude of the parameter concentration allowance according to the leachate characteristic data that has been carried out. Data related to the parameter allowance is presented in the table below. This data will also be used as the basis for calculations in the next unit.

Table 3. Results of ozone oxidation planning

Parameter	Concentration	Ozonization rate	Concentration Ozone	Oxidation Result	Remaining concentration
BOD	339 mg/l	4 x 10^{-4} $g.l^{-1}$/$G.hour^{-1}$	100 G/hour	40 mg/l	379 mg/l
COD	3315 mg/l	0,025 $g.l^{-1}$ /$G.hour^{-1}$		2,500 mg/l	815 mg/l
TSS	216 mg/l	5.8375 x 10^{-4} $mg.l^{-1}$/$G.hour^{-1}$		58.375 mg/l	157.62 mg/l
Color	31.210 Pt.Co	75.68 Pt.Co/$G.hour^{-1}$		7,568 Pt.Co	23,642 Pt.Co
BOD/COD	0,46				

Based on the calculations that have been done, the value of the BOD/COD ratio is 0.46 so that the leachate is safe for biological processing. The purpose of using biological treatment for the processing unit is because there are three zones based on the value of the BOD/COD ratio. These zones consist of a toxicity zone, biodegradability zone, and stable zone (Ganjar , 2010). The toxicity zone is the limit of organic compounds that can interfere with the growth process of microorganisms. Then there is the biodegradable zone, which is the limit of organic compounds that can be degraded by microorganisms. The last one is the stable zone, which is the limit of organic compounds that are safe to be disposed of into the environment. Due to the characteristics of the leachate produced at the Temesi landfill, it is in a toxic zone, so preliminary treatment is used, namely oxidation using ozone to increase biodegradability so that it is in the biodegradable zone. Then after obtaining the desired ratio, biological processing is used so that the leachate that is disposed of is safe for the environment.

4. Conclusions

Based on the results of experiments that have been carried out, the most effective results occur at 60 minutes. The reduction in COD, TSS and colour concentrations at 60 minutes was 37.78%, 12.96% and 11.7%. In addition, there was also an increase in the concentration of BOD, which was 6.85%. The increase in the BOD/ COD ratio produced in this ozone oxidation process was from 0.1 to 0.144. Based on these experiments, the results showed an increase in leachate biodegrability as evidenced by an increase in the ratio of BOD/ COD of leachate. To achieve the desired BOD/COD ratio, you can increase the

concentration of the ozone dose used to oxidize leachate. In this study, an ozone generator of 100 G/hour was used which increased the BOD/COD ratio to 0.46 with a final COD yield of 815 mg/l increasing bod to 379 mg/l, and decrease TSS become 157.62 mg/L and color become 23,642 Pt.Co. The treatability test showed that ozone can increase BOD/COD in the characteristic mature leachate and the ratio is safe for biological processing in the next unit.

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