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Effect of cellulase addition on leachate recirculation for leachate qualities using bioreactor landfill method

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Abstract. Lignocellulose material which consists of three main components, including cellulose, hemicellulose, and lignin, is known for its difficulty to be degraded using biological process using biological process. Cellulase has proven to catalyze the degradation of celullose by enzymatic hydrolysis. However, the addition of cellulase might affect leachate qualities that was generated from landfill. The aim of this research is to analyze the effect of cellulase addition on leachate qualities. Two 1.5 m-height bioreactors were provided for two different treatments including (1) leachate recirculation with cellulase addition (2) leachate recirculation only as control. The addition of cellulase at 15 x 106 U/tonne was resulting in lower concentration for COD (29,100 mg/L in cellulase addition and 31,900 mg/L in control), TS (17,800 mg/L and 22,100 mg/L, respectively), TDS (15,900 mg/L and 19,800 mg/L, respectively). This was likely caused by acceleration of hydrolysis using enzymatic process. However, BOD value higher when cellulase addition was conducted (16,100 and 11,600 mg/L, respectively) because the addition of cellulase supported formation of glucose, therefore escalated BOD value. pH value was increasing over time towards neutral, indicating landfill had been heading toward methanogenic phase. From the experiment, it can be concluded that addition of cellulase has impacts towards leachate qualities.

1. Introduction

Solid waste is all wastes derived from human and animal activities which are usually solid and disposed of because they are mostly considered as useless and undesirable [1]. Due to the increasing population, it directly increases the amount of waste generation. Approximately 1.3 billion tons of waste are generated annually across the world, this volume is expected to increase until 2025 to reach 2.2 tons [2]. If the amount of waste continues to grow and not accompanied by adequate waste treatment will cause serious problems. Wastes have caused problems such as supporting global warming because solid waste emits carbon dioxide along with degradation, waste also can degrade water body quality, release toxic gases, and leachate can contaminate groundwater [3]. Waste problem in Indonesia, especially in big cities, is one of the most challenging urban issues for the government [4]. Indonesia's waste problem is caused by many factors such as high waste generation that estimated to increase by about 2-4 percent per year, poor quality of waste management, limited land for landfill, lack of funds for waste management and bad waste management institution [5].

One of the problems of waste management in Indonesia is limited space of landfill. In big cities, landfill must be closed when it has exceeded the capacity, but the closure is not accompanied by new land clearing for landfill due to land availability, community prohibition, and land price [6]. Indonesia is still too dependent on landfill. Approximately 69% of the waste ends up in the landfill with a total of 200

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landfills in Indonesia, with poor conditions. A good landfill using sanitary landfill technology is only 10 percent of the total landfill in Indonesia [7].

The composition of waste landfill in Indonesia mostly consists of organic waste. TPA Cipayung in West Java consists of 54.014% [8]. Organic waste consists of the main components of lignocellulose. The existence of lignocellulose in waste affects biodegradability in anaerobic systems [9]. Lignocellulose material is a hard-to-break polysaccharide due to its chemical stability, insoluble in water or organic solvents such as acids or weak bases [10]. Lignocellulose has evolved for degradation resistance and this resistance comes from the crystalline form of cellulose, lignin hydrophobicity, and cellulose wrapping by the lignin-hemicellulose matrix [11]. Lignocellulosic materials consist of mainly 3 types of polymers that is cellulose, hemicellulose, and lignin [12].

The degradation of lignocellulosic waste and the products produced by the process are carried out by the activity of various enzymes, especially cellulase [13]. Cellulase is an enzyme that converts one of the components of Lignocellulose, which is cellulose into glucose. The addition of enzymes and leachate recirculation simultaneously can be applied as a TPA treatment [14]. Leachate recirculation has received more attention because it is easy to apply on small and large scale due to accelerated degradation so as to reduce landfill land requirements [15].

However, the addition of cellulase might affect leachate qualities that was generated from landfill. The aim of this research is to analyze the effect of cellulase addition on leachate qualities by analyzing leachate quality parameters for 90 days (BOD, COD, TS, VS, TDS, and TSS) which generated from lab-scale bioreactor landfills that was provided for this purpose.

2. Methods

2.1. Bioreactor landfill

Batch test were conducted for 90 days to observe the effect of cellulase addition into leachate recirculation (1) and leachate recirculation only on leachate quality. Two air-tight bioreactors from Polyvinyl Chloride (PVC) cylinder were provided for this purpose. The dimensions of the columns were as follows: thickness = 9,2 mm, internal diameter = 299 mm and height = 1,85 m (volume = 0.13 m3). The reactor consisted of four kinds of pipes. One port (bottom) served as leachate collection pipe while the other two ports (top) served as a leachate recirculation pipe and gas collection pipe, respectively. Five ports on each sides were served as sample collection pipes. To prevent clogging, 15 cm depth gravels were filled at the bottom reactor. 53 kg waste was compacted to 500 kg/m3 density and was filled on top of gravel for 150 cm height. Rest of the reactor space was filled with cover soil to prevent gas leak.

Leachate recirculation flow rate is 5 Liter/tonne waste as suggested by Environment Agency (2009) for recirculation that was conducted using batch system and categorized as low recirculation rate. Therefore, 265 ml leachate from leachate collection tank was recirculated for 11 times into bioreactor on 90 days period using batch system.

Water addition was conducted to represent the amount of water that percolated through landfill Determination of water addition based on rainfall condition of Depok City taken from Rain Station of FT UI Depok, West Java, Indonesia with average rainfall 0.086 dm / day. Therefore the addition of water is 0.6 L / day.

Cellulase addition was carried out simultaneously with leachate recirculation using batch system [14]. The amount of cellulase added is equivalent to 15 million U / tonne waste was suggested to be added along with leachate recirculation [16]. The cellulase was added into reactor, hence was named Reactor A and reactor without cellulase addition was named Reactor B. The cellulase was produced from cultivation and extraction from *Trichoderma reesi*. The cellulase used is a CEL 150 with an activity of 1,500,000 U / gram was purchased from Sinobios, Shanghai. Therefore, the amount cellulase added for 53 kg waste sample were 0.53 gr.

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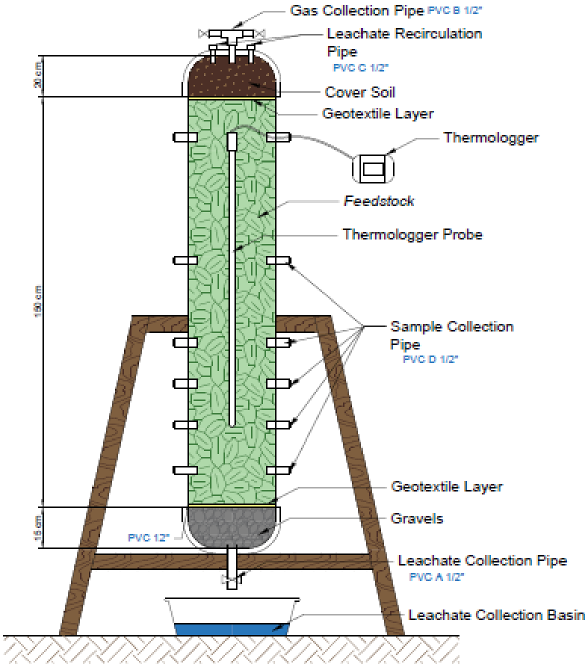

Figure 1. Bioreactor landfill cross section

2.2. Feedstock

Whole 100% organic waste samples were collected from TPS Kemiri Depok, West Java. Both bioreactors contained 53 kg waste samples and its density was assumed as 300 kg/m3 as density before treatment in landfill [1]. Manual compacting was conducted until 500 kg/m3 was reached as an ideal density in landfills [1]. Waste samples were shredded to particles in 5 – 10 cm.

2.3. Analysis method

Leachate was collected from leachate collection at the bottom of bioreactors. To obtain representative leachate characteristic, leachate was characterized in terms of pH, biochemical oxygen demand (BOD5), BOD/COD Ratio chemical oxygen demand (COD), total solid (TS), total dissolved solid (TDS), total suspended solid (TSS), and volatile solid (VS). BOD analysis was conducted using titrimetric method which determined the amount of oxygen loss through the biological decomposition inside winkler bottle.

Table 1. Analysis method

Leachate Quality Parameter	Method	Standard
pH	pH meter	-
BOD5	Titrimetric	SNI 6989.72:2009
COD	Spectrophotometric	SNI 6989.2:2009
TS	Gravimetric	SNI 06-6989.26-2005
TDS	Gravimetric	SNI 06-6989.27-2005
TSS	Gravimetric	SNI 06-6989.3-2004
VS	Gravimetric	SNI 06-6989.26-2005

COD was determined with HACH DR 2000 Spectrophotometry COD test kits (500–10,000 mg/L). In Total Solids (TS) analysis, leachate samples were dried in an oven at 105^{0}C for 3 hours and weighted. Dried samples then were dried in a furnace at 550^{o}C for 1 hour and weighted for Volatile Solids (VS) analysis. Total suspended solids (TSS) were conducted by filtering a certain amount of leachate through glass micro fibre filter paper (200 nm). The filter was then dried in an oven at 105^o C for 1 hour and weighted. The liquid that percolated through the filter was collected at a known amount for total suspended solids (TDS) analysis. The filtrate was dried in an oven at 180^{o}C for 1 hours and weighted.

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3. Results and discussion

3.1. pH

pH value in leachate depends on activities occuring inside. It is caused by the processes of aerobic, acidogenesic, acetogenesic, and metanogenesic, which affect the composition content of leachate such as volatile fatty acid and asetic acid. [17]. Typical leachate usually had pH value between 4.5 to 9 [18]. pH value of leachate is also usually affected by landfill age, in which leachate from new landfill usually more acidic (below 6,5). As for leachate from older landfill, it was more alkaline (above 7.5). pH value range for Reactor A was between 5.5 – 7. As for Reactor B, it was between 5.6 – 7.3. It was suitable with pH value typical for new landfill is between 4.5 – 7.5 [1].

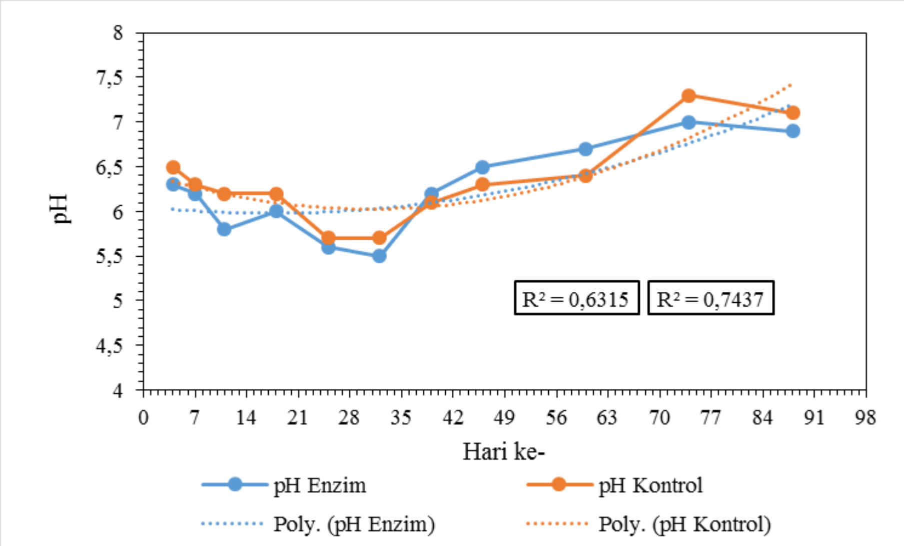

Figure 2. pH during the experiment

pH value on acidogenic stage was influenced by the concentration of volatile fatty acid (VFA). On the fourth day to thirty-second day, the pH values for both reactors was decreased. It was caused by the high production of Volatile Fatty Acid (VFA) and partial pressure of CO_2 in acidogenic phase which usually have typical pH value of 4.5 – 6 [19]. pH value returned to increase towards neutral over time, which indicated that concentrations of VFA's free ions was decreasing [20]. It occurred in initial methanogenic phase, where acid converted into methane and caused formation of acid was decreased and pH value of leachate was increased to between 6.8 – 8 [1]. Therefore the increase of pH value indicated acidogenic phase moved into methanogenic.

3.2. Organics

3.2.1. BOD. Leachate from early acidogenic phase contains organic material biodegradable in high number. It occurred because in acidogenic phase the fermentation of complex organic component was occurred and produced VFA and amino acid [21]. Therefore, typical BOD on this phase is rather high (>10.000 mg/L). BOD value will decrease by time, because stabilization process of solid waste keep increasing [22]. The lower BOD value indicated that the remaining of the organic component only consisted of final products from degradation process [21].

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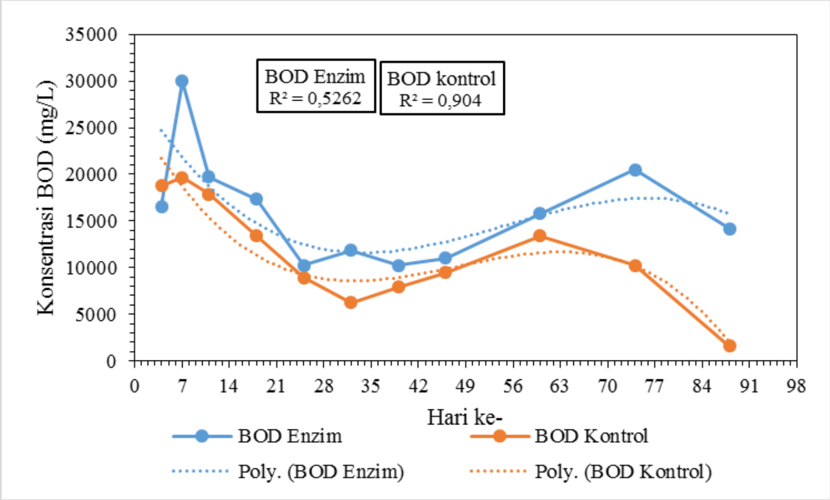

Figure 3. BOD during experiment

The reactor with cellulase addition (Reactor A) had higher BOD value than reactor without cellulase with 4542 mg / L difference. The BOD value describes the amount of organic biodegradable in leachate. Therefore BOD leachate in the Reactor A had higher biodegradibility than the Reactor B. Higher BOD value of the Reactor A was due to increased hydrolysis processes and the production of volatile fatty acids [23] The addition of cellulase has increased cellulose hydrolysis to glucose, so the glucose concentration in reactor with cellulase addition is higher than the Reactor B, thereby increasing the BOD value.

3.2.2. COD. In many cases COD concentration will increase on initial phase in a short time, and will decrease by time [24]. The decrease of COD Value can also be caused by faster solid waste's degradation in laboratory scale by using anaerobic landfill [25]

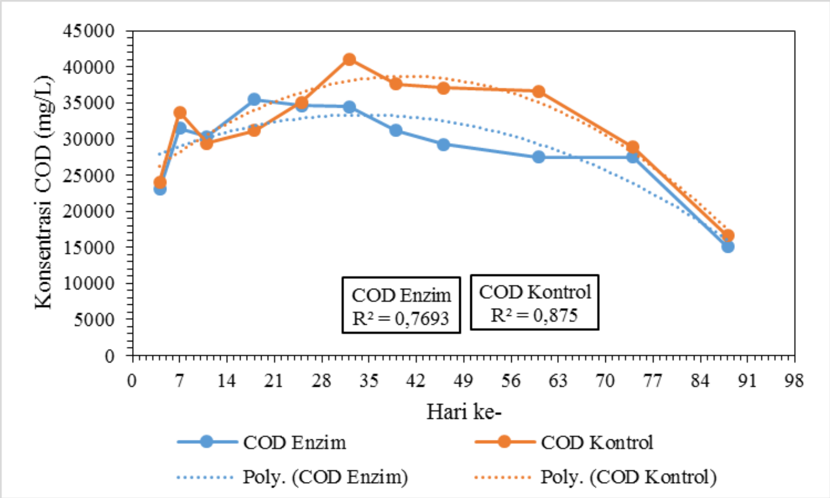

Figure 4. COD during the experiment

Maximum COD value was shown to occur early on Reactor A which reached its maximum value on the day-81 than Reactor B which occurred on the day-32. It was similar with the statement that cellulose hydrolysis influenced COD value [26]. Therefore the escalation of COD value on initial phase indicated that hydrolysis process occurred faster on Reactor A. pH value for Reactor A was decreased until the ninety-first day with the value of 15.100 mg/L.

Higher COD removal was resulted from Reactor A. It had 34% removal, higher than Reactor B which only had 30,1% removal. This may be caused by when decomposition of cellulose and hemicellulose started, COD and BOD value start to decrease [27]. This also suitable with the statement that the

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hydrolysis of cellulose influenced COD value [26]. So the bigger decrease of COD value was caused by decomposition process of cellulose and hemicellulose which became faster by using enzymatic process with cellulase.

Similar research was conducted by Frank, et. al (2016) with eighty-days of observation. From that research, he obtained COD value for Reactor A for about 1943 mg/L and Reactor B produced a higher COD value which about 2065 mg/L. For leachate COD removal, Reactor A had bigger removal value which is 42%. Higher than Reactor B which had a value of 35%. However, COD removal in this research was smaller than [28] research which obtained COD removal above 95% when recirculation of leachate was implemented.

3.2.3. BOD/COD ratio. Biodegradability of leachate varies over time in processes within the landfill. Changes in biodegradability of leachate can be evaluated using the ratio of BOD / COD [29]. The BOD / COD ratio is a good indicator in measuring the proportion of biologically degradable organic material in all organic materials. The BOD / COD ratio is also an indicator of phase change from acetogenic to methanogenic in the waste stabilization process [30].

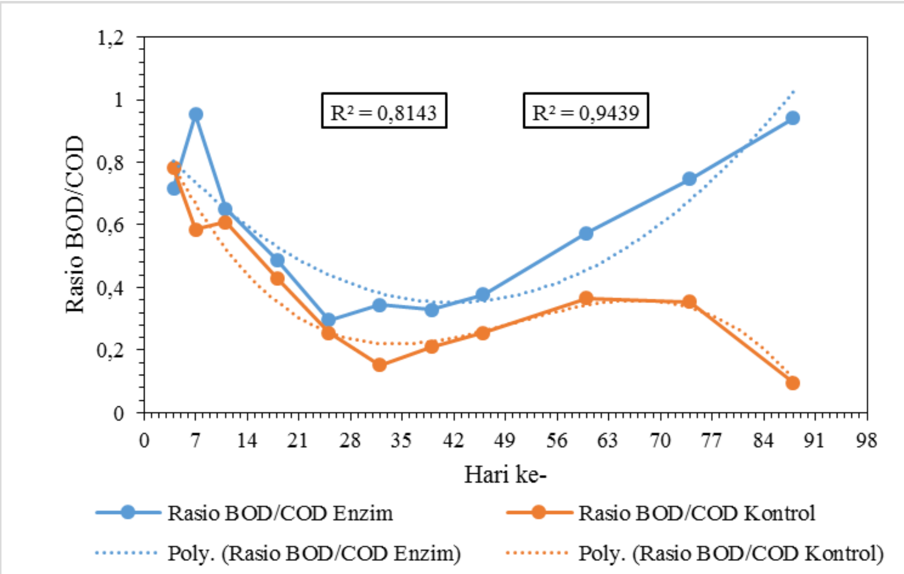

Figure 5. BOD/COD ratio during the experiment

The BOD / COD ratio for both reactors continued to decrease in halfway of bioreactor operation. The decrease in the BOD / COD ratio concludes that the remaining organic component is the final product of the degradation process [21]. Decrease in BOD / COD ratio occurred at the Reactor B until the last day with the number 0.09. A low BOD / COD ratio indicates a low biodegradation rate and contains less easily degradable organic material [31]. This is due to the content of cellulose and lignin which are difficult to degrade under anaerobic conditions [32].

3.3. Solids

The solid parameters measured in this study included Total Solid (TS), Volatile Solid (VS), Total Suspended Solid (TSS), and Total Dissolved Solid (TDS). All measurements were conducted using gravimetric method. The amount of total solids supposed to be equal with sum of total suspended solids (TSS) and total dissolved solids (TDS) [33]. However, this value couldn't be reached in this study, therefore these errors should be taken into consideration

3.3.1. Total solids (TS). TS value between Reactor A and Reactor B had significant differences after evaluated using statistical analysis. Lower TS values obtained by Reactor A, with 4,225 mg/L differences compared with Reactor B.

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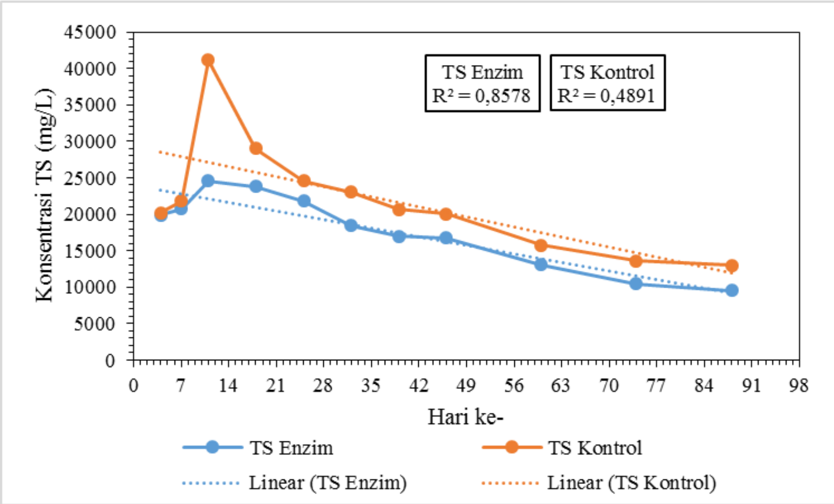

Figure 6. TS during the experiment

This was in contrast to the research of Frank, et al (2016) that has stated no difference in TS values in leachate when cellulase was added. Similar research stated that the concentration of TS values in leachate will decrease as the phase moved from asidogenic to methanogenic [34]. This indicated Reactor A has reached methanogenic phase earlier than Reactor B..

3.3.2. Volatile Solids (VS). VS removal for Reactor A and Reactor B is 45% and 21% respectively. This result was consistent with Frank et al. (2016) who have conducted similar research. The VS results were said to decrease in concentration and had small differences between the two reactors. Due to the biodegradable nature of waste, the organic component decreases faster than inorganic, therefore the VS value decreases over time [35]. Degradation process within bioreactor has led to decrease VS value caused reduction of organic in leachate. However, there was no difference in VS value between the two reactors.

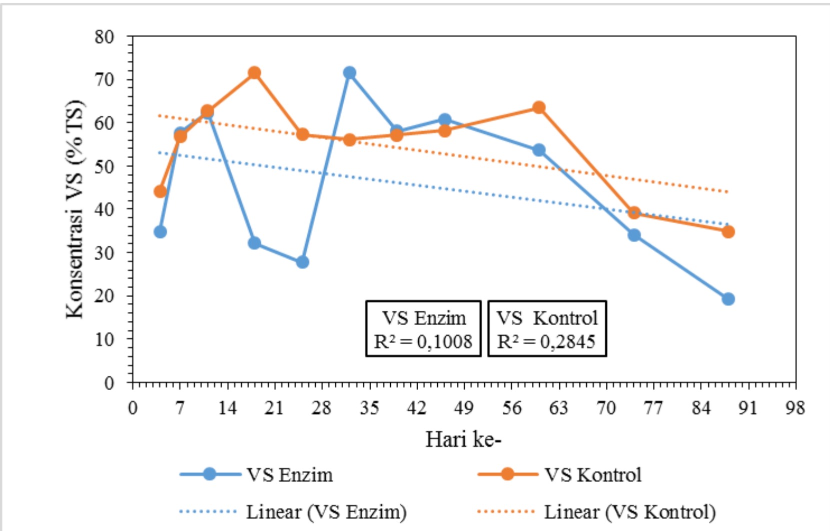

Figure 7. VS during the experiment

3.3.3. Total suspended solids (TSS). TSS removal for Reactor A and Reactor B were -650.6% and -373.6% respectively. This indicated escalation in TSS value before and after bioreactor landfill operation. This is in contrast to the results of the research by Frank et al. (2016), which stated decrease in TSS values in both reactor with or without cellulase addition due to the recirculation process. This may has been caused by leachate exposure to oxygen and lead to iron oxidation from Iron (II) to Iron (III). Hence forming colloidal iron hydroxide, which contributed to brown color in leachate samples and

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increased the value of TSS. Therefore while COD values decreased over time, TSS values continue to increase because of iron oxidation [34]. An increase in TSS value can also be caused by escalation of pH values which causes a decrease in the solubility for irons such as Sulfate Ferry [36].

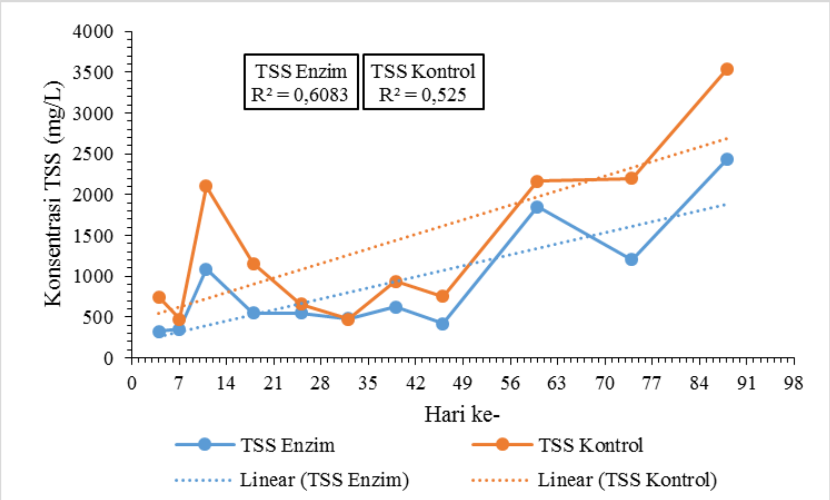

Figure 8. TSS during the experiment

The cellulase addition did not affect the TSS concentration [37], however in this study Reactor A had lower TSS concentration than Reactor B with difference of 485.18 mg / L. TSS consists mainly of leaf and wood particles, but in addition there are also soil particles [31]. TSS value in Reactor A is smaller than the Reactor B. This indicated addition of cellulase has supported hydrolysis of organic particles into water-soluble glucose.

3.3.4. Total dissolved solids (TDS). In general, for both reactors the TDS value continued to decrease constantly as the phase transfer from acidogenic to methanogenic.

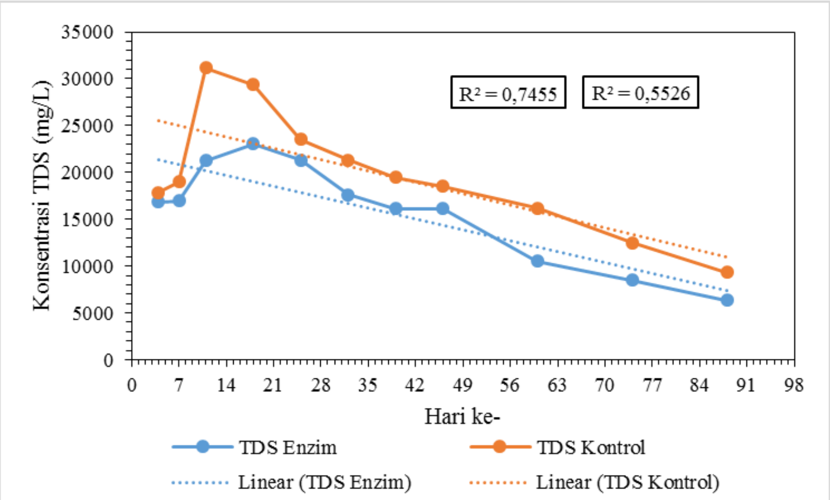

Figure 9. TDS during the experiment

This was occurring because low pH (acidogenic) has promoted heavy metal dissolution [34]. High metal solubility [38] and minerals such as Aluminum [39] at low pH cause high TDS. As the pH increases, the solubility of heavy metals and aluminum decreases.

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4. Conclusion

The addition of cellulase has influenced the concentration of pollutant leachate generated by waste. The values of COD, TDS, TSS, and TS had significantly lower caused by cellulase addition. This is because cellulase has accelerated the degradation process by hydrolyzing cellulose into glucose. BOD values are significantly larger when cellulase addition was implemented because glucose increases the load of BOD in leachate. There were no differences in pH and TSS after statistical tests.

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