

## Performances of Polyethylene Terephthalate Plastic Bottles Waste as Supporting Media in Domestic Wastewater Treatment Using Aerobic Fixed-Film System

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### ABSTRACT

There has been a lot of research on domestic wastewater treatment utilizing polymer-based supporting media. The purpose of this study is to investigate the removal of organic compounds and nutrients, as well as the kinetics of substrate removal, in a batch aerobic fixed-film system that is fed by artificial domestic wastewater and uses Polyethylene Terephthalate (PET) plastic bottles waste as supporting media. The artificial domestic wastewater feeding contains  $C_6H_{12}O_6$ ,  $NH_4Cl$ , and  $KH_2PO_4$  as carbon, nitrogen, and phosphorus sources. Artificial domestic wastewater treatment was performed at COD levels of 200, 300, 400, 500, and 600 (mg/L). The findings demonstrated that an aerobic fixed-film system wastewater treatment with PET-supporting media could remove organics and nutrients. The removal for COD  $85.76 \pm 0.59\%$ , ammonia  $76.59 \pm 0.83\%$ , nitrite  $76.09 \pm 0.66\%$ , nitrate  $64.30 \pm 0.42\%$ , TN  $77.02 \pm 0.94\%$ , and TP  $86.54 \pm 0.68\%$ , with the Singh's method substrate removal kinetics ( $k_1$ ) is  $1.60 \pm 0.05$ /hour. The benefit of supporting media from PET plastic bottle waste is contributing to plastic bottle waste reduction in Indonesia.

**Keywords:** aerobic fixed-film system, artificial domestic wastewater, plastic bottles waste, supporting media, Polyethylene Terephthalate.

### INTRODUCTION

Wastewater treatment employing the biofilm method has gained popularity in recent years for lowering organic matter and nutrients in Wastewater Treatment Plants (WWTPs) (Barwal and Chaudhary, 2016; Xu et al., 1998; di Biase et al., 2019). A fixed film bioreactor, for example, has been found to have advantages over suspended growth systems (Huang et al., 2017). Biofilms produced on supporting media are more resistant to washing out from processing systems due to organic and hydraulic loads (Metcalf and Eddy, 2013; Di Biase et al., 2019), resulting in less sludge generation. In terms of organic matter and nutrient removal efficiency, the fixed-film system

outperforms suspended methods (Huang et al., 2017; Mannina et al., 2018). Based on these benefits, adding supporting media as a medium for attaching biofilms in biological wastewater treatment is a cost-effective approach (Sriwiryarat and Randall, 2005; Zhao et al., 2018).

In biological processing, the utilization of supporting media in connected growth reactors or biofilm technology has been widely used. The global development of biofilm technology in wastewater treatment is fairly rapid, with varied materials, forms, and sizes with the goal of obtaining a large specific surface area and cavity volume to attach large numbers of microorganisms and effect reactor performance. Tchobanoglous et al., (2003) established design criteria for

a specific surface area of 100–820 m<sup>2</sup>/m<sup>3</sup> and a void ratio of 15–98%, Grady et al., (2011) stated a surface area for bacterial growth of 100 m<sup>2</sup>/m<sup>3</sup> with a void fraction of 90–95%. Even the physical properties of a supporting media affect the rate of biofilm formation (Dias et al., 2018) as well as interactions and mass transfer in pollutant removal (Liu et al., 2019).

Initial biofilm adhesion plays an important role in biofilm growth systems on adhesive media (Mao et al., 2017; Tang et al., 2017). According to research (Habouzit et al., 2011; Almomani et al., 2019) shows that the adhesion of biomass is influenced by the physicochemical properties of the polymer media. PET has a small water contact angle of 77.20 (Peng et al., 2018), so PET is hydrophilic (Setiyawan et al., 2023). Media that have high surface energy (hydrophilic) can cause strong bacterial adhesion resulting in faster biofilm formation (Callow and Fletcher, 1994; Khan et al., 2013). So that PET can be used as a suitable medium as a place for biofilms to adhere properly. The time needed for the start-up period of biofilm formation is between 1 to 6 weeks (Bassin et al., 2016; Dong et al., 2015).

In order to reduce the negative impact of used plastic bottle waste, various types of technologies for utilizing plastic bottles have been developed, including the use of plastic bottles as a supporting medium, namely treating waste with waste or what is known as „waste treat waste” (Carrasco et al., 2016; El Essawy et al., 2017). Based on research by Ahn et al., (2004); Espinoza et al., (2019); di Biase et al., (2019) plastic bottles have the desired properties as a supporting medium, namely not easily biodegradable, lightweight, relatively large surface area, high void volume fraction, water-loving (hydrophilic), water-resistant durable, not easily damaged, does not corrode and is able to attach large numbers of microorganisms with a small risk of stalemate.

Although countries worldwide have not widely used research on PET plastic bottle waste as a supporting media, in Indonesia it has begun to develop since 2014 and is applied to communal WWTPs (Nur et al., 2021). Utilizing used PET plastic bottles as a supporting media in treating domestic wastewater is the right step because it can reduce plastic waste in Indonesia. This is very profitable because Indonesia is the 4<sup>th</sup> largest producer of plastic bottle waste in the world (PT. Chandra Asri Petrochemical, 2017), so it can minimize the generation of plastic waste.

## MATERIALS AND METHODS

### Reactor and supporting media preparation

This study used an aerobic fixed-film system made of transparent acrylic material with a working volume of 39 L. The reactor was equipped with a pump so that the substrate and seed sludge were well mixed so that the biomass attached to the PET formed a biofilm during the start-up process. In addition, the pump also injects air into the reactor to obtain the dissolved oxygen concentration in aerobic conditions. Microorganisms will grow and stick on the supporting media from used PET bottles with a diameter of 6.4 cm, and transparent color.

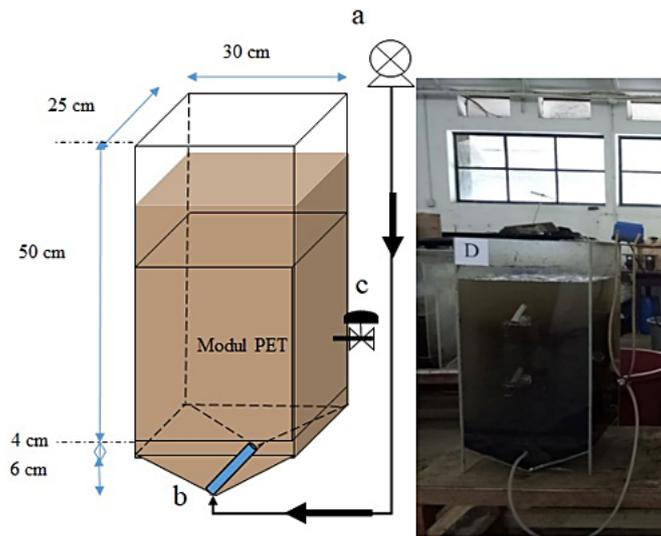
The media used in this reactor is a combination of 600 mL and 1.5 L of mineral water PET bottles waste. The 1.5 L plastic bottle waste media is modified by cutting it to a height of 14.5 cm and a diameter of 2 cm, then rolling it to form a zig-zag, and put in a 600 mL plastic bottle waste with a height of 14.5 cm. Furthermore, it is arranged according to the shape of the reactor which forms a surface area of 444 m<sup>2</sup>/m<sup>3</sup>. The schematic of the reactor and the PET supporting media formed can be seen in Figure 1 and Figure 2.

### Artificial domestic wastewater and seed sludge

Artificial domestic wastewater is made from C<sub>6</sub>H<sub>12</sub>O<sub>6</sub>, NH<sub>4</sub>Cl, KH<sub>2</sub>PO<sub>4</sub> (Bratachem) as a source of carbon, nitrogen, phosphate, and tap water at the Environmental Engineering Water Quality Research Laboratory of Institut Teknologi Bandung is used to mix the materials used. Before tap water is used in this research process, organic matter, and nutrients are checked. The results show that tap water does not contain organic substances and nutrients. The ratio of C: N: P substrate used was at the optimum conditions for bacterial growth, namely 100:10:1 under aerobic conditions (Tchobanoglous et al., 2014). Seed sludge comes from domestic septic tanks in the city of Bandung.

### Seeding and running

The seeding stage was carried out to breed and increase the concentration of mixed culture bacterial biomass with glucose substrate because it is easily degraded by bacterial cells (Dworkin et al., 2006). VSS and COD are tested to assess



a = pump, b = air stone, c = sampling point

Figure 1. Aerobic fixed-film system



Supporting Media



Structure of Supporting Media

Figure 2. PET supporting media

bacterial development using Standard Methods 2540 E and 5220 I (APHA, 2017). During the bacterial development phase, the pH is kept at optimum levels, that is 6.5–7.5 on a regular basis (Tchobanoglous et al., 2014; Ahn et al., 2004) and at 25–27 °C at room temperature. The reactor is kept aerobic by injecting DO levels through a pump. Besides that, it aims to keep the sludge mixed evenly in the reactor. Seeding lasted for 40 days and continued with running using artificial domestic wastewater with concentrations of 200, 300, 400, 500, 600 (mg/L).

### Identification of organics and nutrient compounds

The concentration of organic and nutrient compounds was measured using the Standard

Methods, COD (5220 I), total nitrogen (4500-W), nitrite (S4500-NO<sub>2</sub><sup>-</sup>), nitrate (4500-NO<sub>3</sub><sup>-</sup>), ammonia (4500-NH<sub>4</sub><sup>+</sup>) and total phosphate (4500-P-B-D). The efficiency of the wastewater treatment system can be calculated by the formula (Tchobanoglous et al., 2003), where E is the efficiency (%), Co is the initial concentration (mg/L) and C is the final concentration (mg/L).

$$E = \frac{C_o - C}{C_o} \times 100\% \quad (1)$$

### Kinetics of substrate degradation

The first-order kinetic equation is the most frequently applied model in determining the rate of decrease in the concentration of organic compounds and the growth of attached biomass (Droste, 1997). This model is applied by

determining the change in substrate concentration in the system by connecting it to the pseudo-steady state conditions as described below.

$$\frac{-dC_s}{dt} = k_1 \cdot C_s \quad (2)$$

$$\ln \frac{C_s}{C_{s0}} = -k_1 \cdot t \quad (3)$$

The first-order constants are calculated based on the time relationship with  $\ln C_s/C_{s0}$  plotted on the  $x$  and  $y$  axes. After carrying out linear regression on the equation, the organic compound degradation constant ( $k_s$ ) will be obtained.

The kinetic equation of the Singh model is a modification made by Singh (Kureel et al., 2017) from the first-order model to Equation 2.  $k_{si}$  is the removal rate of this model calculated from the slope of the straight line equation obtained from the plot  $\ln C_s/C_{s0}$  to  $\ln 1+t$ , so that the substrate degradation constant will be obtained.

$$\frac{-dC_s}{dt} = \frac{K_{si} \cdot C_s}{1+t} \quad (4)$$

$$\ln \frac{C_s}{C_{s0}} = -k_{si} \cdot \ln 1+t \quad (5)$$

In addition, there is a second-order model that, in general, can also be applied to determining the trend of removal of organic compounds in aerobic reactors (Ardestani, 2011). The second-order substrate degradation kinetics model is the most frequently applied model in evaluating the rate of biodegradation of organic compounds and nutrients (Grau et al., 1975; Mansouri et al., 2014). The substrate removal rate using this model does not depend on the concentration of microorganisms, but only on the 2<sup>nd</sup> power of the substrate shown in Equation 2. If plotted into a

graph between  $1/C_s$  and  $t$ , a linear line will be obtained with the slope being the value of  $k$ .

$$\frac{-dC_s}{dt} = -k_1 \cdot C_s^2 \quad (6)$$

$$\frac{1}{C_s} = \frac{1}{C_{s0}} + k_1 \cdot t \quad (7)$$

## RESULTS AND DISCUSSION

### Effect of seeding to concentration of organic compounds and VSS

The seeding stage was carried out to grow microorganisms until the VSS reached 2,000–2,500 mg/L (Goswami and Maxumder, 2016), while according to Tchobanoglous et al (2013) around 3,000–4,000 mg/L. The increase in suspended VSS content was accompanied by a decrease in substrate concentration. The value of the biomass concentration is represented by the VSS concentration, while the value of the attached biomass concentration can be assumed from the attached VSS value. The seeding process can be stopped when the biomass concentration has exceeded 2000 mg/L and continued with the running process using artificial wastewater. In this study, the seeding process was stopped after the VSS content attached on day 40 was 3,330 mg/L and the suspended VSS was 2,190 mg/L which can be seen in Figure 3. This is directly proportional to previous studies which concluded that the seeding process in aerobic conditions lasted for 1–6 weeks (Leyva-Díaz et al., 2013; Tang et al., 2016). The COD content value decreased, then increased again after the addition of substrate. On the last day of seeding, the COD value was 20 mg/L.

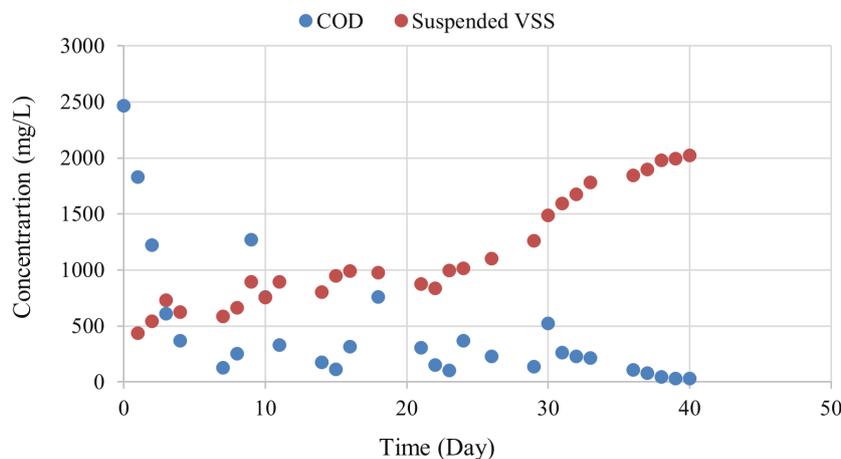


Figure 3. Concentration of COD and suspended VSS on seeding

### Removal of organic and nutrient compounds

COD concentrations of 200, 300, 400, 500, and 600 mg/L at running were selected based on the characteristics of domestic wastewater in Indonesia. Table 1 shows the results of evaluating the WWTPs. The removal efficiency produced at COD 200, 300, 400, 500, and 600 mg/L respectively were 85.71%, 85.71%, 86.67%, 85.71%, and 85.00%. If the average efficiency of this aerobic reactor is  $85.76 \pm 0.59\%$ . The final concentration of COD in each variation was 28.32, 42.48, 56.64, 70.80, 84.96 mg/L. Overall it can be concluded that the greater the substrate, the greater the removal efficiency. Huang et al. (2008) stated in their research that the greater the carbon source added to the sample, the greater the efficiency of organic removal. PET plastic bottle waste has a higher organic removal effectiveness as compared to other media. Table 2 shows a comparison of this supporting media with other plastics media. Types of plastics such as PS, PP, and HDPE have lower efficiencies, ranging from 30–80%. However, the type of LDPE plastic in the study by Espinoza et al. (2019) tends to have similar COD removal

efficiency with PET. Based on a review by Fauzi et al. (2023) also showed that the PET plastic type has a higher organic removal efficiency when compared to PS, HDPE, and LDPE. Removal of organic compounds using PET-supporting media has high efficiency. This is proven because the final concentration of running is in accordance with the quality standard of PermenLHK Number 68 of 2016 of 100 mg/L with an organic load of 0.86-2.57 kg/m<sup>3</sup>.day and specific surface area of 444 m<sup>2</sup>/m<sup>3</sup>. The effect of variations in organic load on the efficiency of substrate removal can be seen in Figure 4. The specific surface area formed on a series of supporting media is quite large, so more microorganisms are attached and grow on the media. This causes a greater number of microorganisms in the biomass layer to degrade the organic compounds in the wastewater (Dias et al., 2018; di Biase et al., 2019; Tchobanoglous et al., 2014).

According to Wang et al. (2016) the optimal ratio of nitrogen and phosphate for absorption by microbes is between 6 and 10. This study used a ratio of nitrogen to phosphate of 9.2. The process of removing phosphate from wastewater can occur due to the consumption of orthophosphate by microorganisms to provide energy and develop cell biomass. Under aerobic conditions, bacteria will accumulate phosphorus so that it is active in absorbing phosphate in the form of polyphosphate or orthophosphate from wastewater (Biyashyna et al., 2010). In addition, phosphate removal can occur due to the assimilation process by microorganism cells (Shen et al., 2017). Under aerobic conditions, phosphate will be absorbed into the bacterial cells and then converted to polyphosphate by the cells, resulting in a high efficiency of phosphate removal.

Nitrogen concentration is generally removed by biological processes, namely in the nitrification process ammonia is converted to nitrate through nitrite compounds under aerobic conditions,

**Table 1.** Domestic wastewater characteristic on WWTP in Indonesia

Num	Influent	Concentration of COD (mg/L)
1	WWTP 1	316
2	WWTP 2	459
3	WWTP 3	152
4	WWTP 4	286
5	WWTP 5	607
6	WWTP 6	493
7	WWTP 7	562
Average		410.71
Standart deviation		164.30

**Table 2.** Comparison of this supporting media with other plastics media

Num	Polymer	Type	Process	Wastewater type	Removal	Sources
1	LDPE, PET, HDPE	<i>Fixed-Bed</i>	Aerobic	Domestics wastewater	COD = 80% (LDPE and PET) COD << (HDPE)	Espinoza et al., 2019
2	PS	<i>Fixed-Bed</i>	Anaerobic	Domestics wastewater ( <i>greywater</i> )	TSS = 68% BOD = 64% COD = 31%	Radityaningrum and Kusuma, 2017
3	PP dan PET	<i>Fixed-Bed</i>	Aerobic	Domestics wastewater of hospital	BOD = 84.85% COD = 31.73% Ammonia = 50.60%	Juniarta et al., 2018
4	PET	<i>Fixed-Bed</i>	Aerobic	Artificial domestics wastewater	COD = $85.76 \pm 0.59\%$	This research

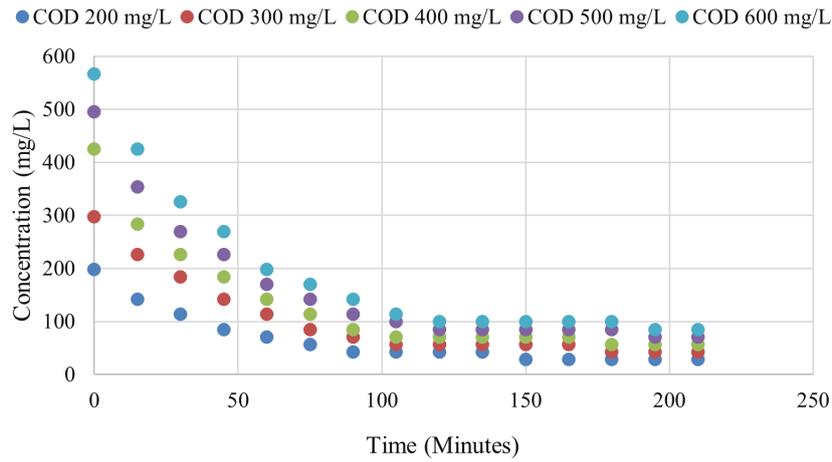


Figure 4. Removal of organic compound COD

which is followed by a denitrification process to convert nitrate into nitrogen gas under anoxic conditions. This causes the removal efficiency of ammonia and nitrite to be higher when compared to nitrate. The processing phases that occur are hydrolysis, ammonification, acid formation, acetogenesis, denitrification, phosphatation (phosphate

released into wastewater by microorganisms), and gas formation (Blyashyna, et al., 2018). The efficiency of nutrient removal can be seen in Table 3. The results of this study indicate that PET plastic bottles waste as supporting media are able to remove nutrient compounds properly. This media is easy to find in Indonesia because Indonesia is a producer of plastic bottle waste in the world. Not only from domestic sources (Fauzi et al., 2022), but the food industry (Dewilda et al., 2023), hotel (Dewilda et al., 2022), and restaurant (Dewilda et al., 2019) also produce plastic bottle waste. So that it has an opportunity to reduce plastic bottle waste in Indonesia. In previous studies, the use of PET also has the potential to eliminate microplastics because microplastics were found attached to

Table 3. Efficiency of nutrients removal

Num	Parameter	Removal efficiency (%)
1	Total phosphate (TP)	86.54 ± 0.68
2	Ammonia	76.59 ± 0.83
3	Nitrite	76.09 ± 0.66
4	Nitrate	64.30 ± 0.42
5	Total nitrogen (TN)	77.02 ± 0.94

Table 4. Recapitulation of the kinetics of the three models

Kinetics model	Runing	Equation	R <sup>2</sup>	k <sub>1</sub> (/hour)
First order	1 (COD 200 mg/L)	Y=0.989x + 0.053	0.995	0.989
	2 (COD 300 mg/L)	Y=0.964x + 0.011	0.998	0.964
	3 (COD 400 mg/L)	Y=1.018x + 0.078	0.992	1.018
	4 (COD 500 mg/L)	Y=0.960x + 0.069	0.992	0.960
	5 (COD 600 mg/L)	Y=0.926x + 0.051	0.991	0.926
Singh	1 (COD 200 mg/L)	Y=1.625x - 0.042	0.989	1.625
	2 (COD 300 mg/L)	Y=1.577x - 0.078	0.983	1.577
	3 (COD 400 mg/L)	Y=1.674x - 0.020	0.988	1.674
	4 (COD 500 mg/L)	Y=1.583x - 0.026	0.994	1.583
	5 (COD 600 mg/L)	Y=1.528x - 0.041	0.994	1.528
Second order	1 (COD 200 mg/L)	Y=0.011x + 0.003	0.959	0.011
	2 (COD 300 mg/L)	Y=0.007x + 0.002	0.963	0.007
	3 (COD 400 mg/L)	Y=0.005x + 0.001	0.955	0.005
	4 (COD 500 mg/L)	Y=0.004x + 0.001	0.977	0.004
	5 (COD 600 mg/L)	Y=0.003x + 0.001	0.983	0.003

biofilms that grow on the surface of this media (Nur et al., 2022).

### Kinetics of substrate removal

To find out the kinetics of decreasing organic concentrations, it can be done in Equations 2– 7.

After carrying out linear regression on the equation, the organic compound degradation constant (ks) will be obtained. Ks values based on the first-order, Singh and second-order models can be seen in Figure 5, Figure 6, and Figure 7.

Based on Figure 5, Figure 6, and Figure 7, the constant values resulting from the three models

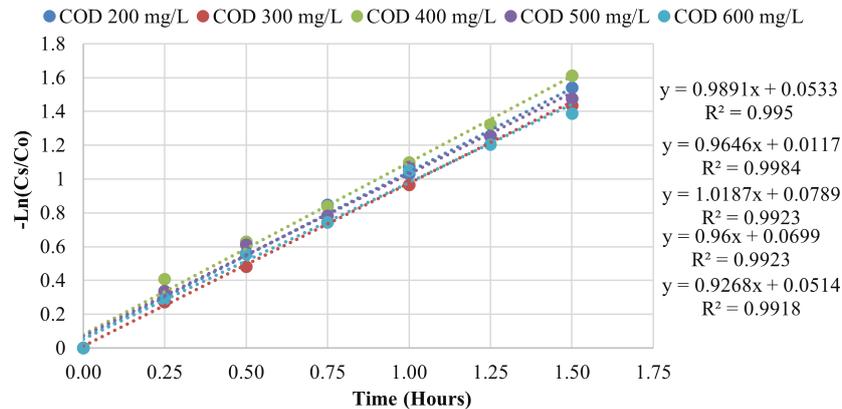


Figure 5. Linier regression determination of organic compounds using First-order model

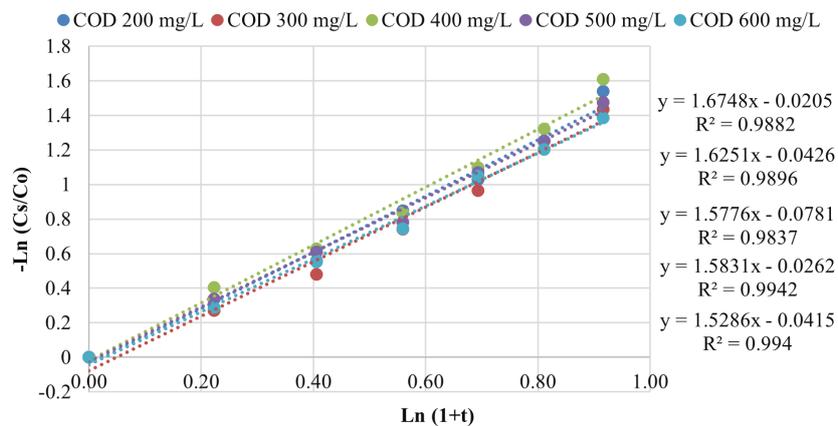


Figure 6. Linier regression determination of organic compounds using Singh model

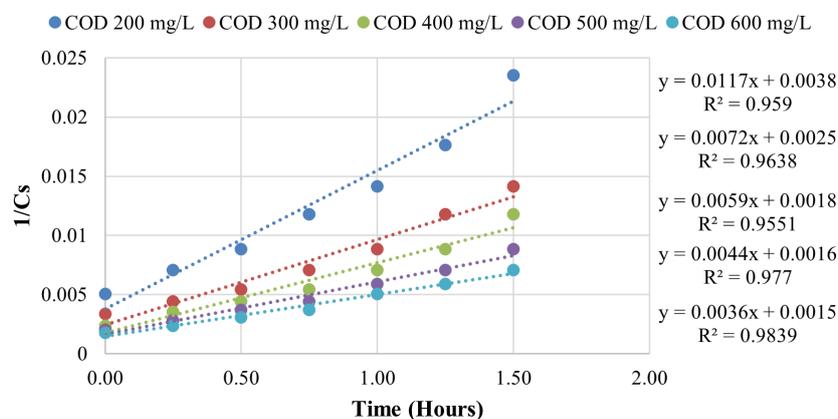


Figure 7. Linier regression determination of organic compounds using Second-order model

can be seen in Table 4. In the three models used, the Singh kinetics model produces a  $k_1$  value that is quite large compared to the other two models and provides a coefficient of determination close to number one which means that the Singh model approaches the reaction that occurs in the reactor used to degrade organic matter.

## CONCLUSIONS

Plastic bottles waste of the PET type as supporting media in the treatment of domestic wastewater using an aerobic fixed-film system is able to attach microorganisms well so that they affect reactor performance. The resulting removal efficiency of organic compounds and nutrients was COD  $85.76 \pm 0.59\%$ , ammonia  $76.59 \pm 0.89\%$ , nitrite  $76.09 \pm 0.66\%$ , nitrate  $64.30 \pm 0.42\%$ , TN  $77.02 \pm 0.94\%$  and TP  $86.54 \pm 0.68\%$ , with the selected substrate removal kinetics Singh method which produces a kinetics value ( $k_1$ ) of  $1.60 \pm 0.05$ /hour. The use of this media is very profitable for Indonesia which is one of the countries that produces the largest plastic bottle waste in the world.

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