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The effectiveness of the multi-soil-layering system in reducing pollutant parameters of crumb rubber industrial wastewater

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Abstract: Multi-soil-layering (MSL) is well-known as an emerging technology for wastewater treatment. However, limited studies have been conducted on the crumb rubber industry. Thus, this study aims to reduce the crumb rubber industrial wastewater using MSL system. Four models of MSL conducted it. They are (1) constructed by body size of gravel with a mixture of andosol soil and fine charcoal; (2) body size of gravel with a mixture of andosol soil, fine charcoal, sawdust, and iron particles; (3) tube size of gravel with a mixture of andosol soil and fine charcoal; and (4) tube size of gravel with a mixture of andosol soil, fine charcoal and sawdust. The construction models were installed in the variance of the box and each of them was flowed by the Hydraulic Loading Rate of wastewater. Aeration treatment is given to the maximum hydraulic loading rate. The result showed that MSL systems significantly minimize the crumb rubber industrial wastewater under requirements that cover Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Suspended Solid (TSS), Total Nitrogen (T-N) and Total ammonia (T-NH_3). It also significantly neutralizes the power of Hydrogen (pH) up to 6. The highest reduction was obtained by the MSL 3 system (tube size of gravel with a mixture of andosol soil and fine charcoal). The aeration significantly decreased the BOD, COD, TSS, and ammonia from 85% to 99%. Although the aeration process enhanced the performance of MSL system, however, MSL without aeration successfully treated the wastewater to meet the required standard. This study provides new insight into the effectiveness of MSL system in reducing pollutant parameters of crum rubber industrial wastewater.

Keywords: Adsorption; Wastewater treatment; Aerobic process; Anaerobic process.

1. Introduction

Many crumb rubber industries are located on the side of rivers. The industry uses a lot of water and chemicals, producing a lot of wastewater and effluent [1], [2]. The discharge of untreated wastewater into waterways or rivers causes water pollution, which has a negative impact on human life [3], [4]. With the new global trend toward sustainable development, the industry must focus on cleaner production technologies, wastewater reduction and minimization, water recycling and utilization, and resource recovery [5], [6], [7]. The crumb rubber industries produce wastewater of an average of 50-100 m^3/hour after a chemical treatment process. The wastewater could not be disposed of directly into the river since it has many pollutants [4]. According to the results of periodic monitoring and preliminary analysis of the quality of wastewater disposal by the industry, it does not comply with the quality standard of environmental requirements [8], [9].

Wastewater in the crumb rubber industry is sourced from raw materials and processes. The crumb rubber industry uses a lot of water to wash raw materials obtained from tappers, which are still dirty and even deliberately filled with wood, metal, and sand [10]. The wastewater becomes liquid and odorous waste [11], [12]. Generally, industrial wastewater from crumb rubber is acidic, containing dissolved and deposited suspended solids [13] The Biochemical Oxygen Demand (BOD) and Chemical Oxygen Demand (COD) values are quite high, the nitrogen content is mainly from ammonia which is used as a latex-fixing ingredient [14]. High BOD and COD in wastewater show high levels of organic matter [15]. Increasing levels of organic matter will further disrupt the environmental ecosystem that receives wastewater because oxygen is used by decomposing bacteria to destroy these organic materials. Lack of oxygen, the death of living things and the presence of organic material in wastewater generated the emergence of various microorganisms that have the potential to cause disease [16].

The discharge of liquid waste by land can take place in physical, chemical and biological processes, it has been known from the past as an effective method and has been used as one of the natural wastewater treatments [17]. Processing of this method is very cheap, but it requires a large area of land when compared to modern systems [18]. Where this land is unlimited and the price is cheap, this system can easily be applied for wastewater treatment. However, processing with this system has serious problems with permeability, where the processing capacity is limited per unit area. Sandy soils have high permeability with an infiltration rate of 1.8 to 3.6 m^2/day, but the chemical and biological ability is not as well as clay soils which have an infiltration rate of only 0.9-0.3 m^2/day [19]. An example of the average flow rate in a treatment system using land is 10-40 l/m^2 /day for traditional trench systems and 1-8 1/m^2/day for slow-rate land treatment systems [8].

Biological methods and mechanical methods have been used in the crumb rubber industrial wastewater treatment. Several studies have focused on biological methods that use microorganism like Pseudomonas sp., Arthrobacter sp., native purple nonsulfur bacteria (PNSB), and Rhodopseudomonas palustris specifically aimed at treating wastewater from the rubber industry [20]–[23]. Although biological methods, particularly anaerobic, aerobic, and facultative ponds, are widely used for the treatment of rubber wastewater, vast areas are required. Meanwhile, mechanical methods such as rotating discs, anaerobic filter beds, and aerated lagoons require little land, but the concentration of pollutants remains high [24]. Biological treatment presents a straight forward approach to address nitrogen compounds within rubber industry wastewater. However, it necessitates substantial space for installation and prolonged retention time. Conversely, membrane treatment offers a compact setup, yields high-quality water, and consumes minimal energy. Yet, it grapples with membrane fouling, impeding its broad and large-scale implementation [25]–[28]. Meanwhile, chemical treatments like coagulation/flocculation and AOPs prove less cost-effective and carry the risk of causing secondary pollution in water bodies [29]–[32]. Overall, the selection of advanced wastewater treatment methods hinges on several factors: (1) the extent to which a method is needed to elevate wastewater quality to an acceptable level, (2) the adaptability of the control method, (3) the expenses involved in the process, and (4) the compatibility of the method with environmental concerns. Therefore, this wastewater must be addressed using appropriate technologies such as Multi-Soil-Layering [24].

Multi-Soil-Layering (MSL) is one method of wastewater treatment that utilizes soil as the main medium by enhancing its function through its structure to clean wastewater. Soil aggregate structure is the key to purifying wastewater. The specific area of the aggregate surface area is between 1-10 m^2 [33]. Some advantages of MSL system are: (1) being able to decompose organic substances such as BOD, COD, nitrogen and phosphate from liquid waste simultaneously; (2) able to prevent blockages; (3) able to accept high hydrolysis loading and absorption rates; (4) the composition of the constituent coating material can be replaced by the available material [34], [35], [36]. The system was formed in the construction of a soil layers mixture with 10-35% iron particles and organic material with zeolite film in the form of bricks. Besides being able to prevent blockages, the system is equipped with two processing zones, namely the aerobic zone in the zeolite layer and the anaerobic zone in the soil mixture layer [37]. The MSL consists of two main processing zones, namely the aerobic zone and the anaerobic zone. The aerobic zone is in the zeolite layer as a permeable layer and the space between the zeolite layer and the mixed soil block. Anaerobic zones are found in mixed soil layers as unpermeable layers. The liquid waste treatment process in the Multi Soil Layering system consists of decomposition, fixation, nitrification, denitrification, adsorption and absorption [38]. The aerobic and anaerobic conditions in the MSL system should be regulated in such a way that all pollutants can be decomposed efficiently [39].

The efficiency of the MSL system also depends on the properties of the mixture coating material. Perlite and charcoal have a special area that has the same absorption power as organic material with zeolite. Gravel has the smallest area compared to other attachments, but organic waste liquid material can coat the surrounding gravel like a film (thin layer) [40], [41], [42]. Perlite, zeolite and charcoal have lower cation exchange capacity than zeolite, but higher than pearlite and gravel [43]. Therefore, ammonia and nitrogen absorb, and P fixation by pearlite, zeolite and charcoal is lower than by zeolite and gravel [44]. The lowest fixation occurs in layers that are mixed with wax and gravel [45]. If zeolite is replaced with zeolitized perlite, perlite, charcoal or gravel, the efficiency of the system in purifying wastewater changes depending on the content of the material. Zeolitized perlite, perlite, and charcoal have high specific areas so that they can absorb organic material as well as zeolite [46]. Gravel has a smaller specific area compared to the previous material but the organic material is placed around the gravel particles so that it can purify wastewater [47].

MSL system has been an emerging technology in wastewater treatment either domestic or industrial bonded with pollutant parameters such as BOD, COD, TSS, T-N, total ammonia, and high acidity [48]. Theoretically, it is suitable to be applied on crumb rubber industrial wastewater treatment. However, there is a limited study on crumb rubber industrial wastewater [49]. Therefore, this study aims to determine the effectiveness of the MSL system on crumb rubber industrial wastewater treatment. The effectiveness is considered from the effect of hydraulic loading rate, aeration, MSL model composite structure, and components on pollutant parameters such as BOD, COD, TSS, T-N, T-NH_3, and pH.

2. Material dan methods

2.1 Material

Sampling was collected from crumb rubber industrial wastewater near Padang, West Sumatera, Indonesia. The sampling is conducted according to SNI 6989.59:2008. Each treatment was taken triplo sample (inlet and outlet) every day. Each sample was taken before treatment (untreated) and after treatment by using MSL system (treated). The experimental model and its mechanism are presented in Figure 1.

The samples were then placed into the MSL system from the upper side. The treated wastewater output from bottom MSL system was collected and subjected to testing for pollutant parameters: BOD, COD, TSS, total nitrogen (T-N), total ammonia (T-NH_3), and pH specified in data analysis.

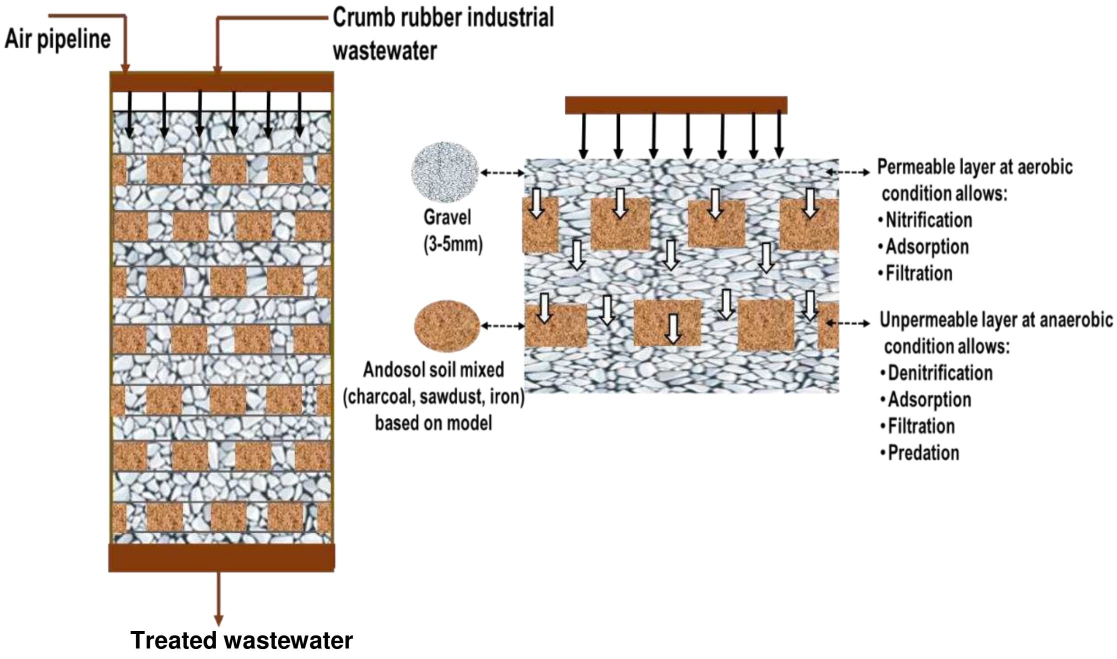

Figure 1: Laminate composite preparation

MSL system is classified into 4 models with different sizes and structures as follows:

(1) MSL 1, body size 50 x 75 x 100 cm, with gravel size 3 - 5 mm as compiler permeable layer and a mixture of andosol soil and fine charcoal with a ratio of 2:1 as unpermeable layers.
(2) MSL 2, body size 50 x 75 x 100 cm, with gravel size 3 - 5 mm as a compiler permeable layer and a mixture of andosol soil, fine charcoal, and sawdust, with the ratio of 2:0.5:0.5 and 10% iron particles as unpermeable layers.
(3) MSL 3, tube size 50 x 15 x 150 cm, with gravel size 3 - 5 mm as a compiler permeable layer and a mixture of andosol soil and fine charcoal with a ratio of 2:1 as unpermeable layers.
(4) MSL 4, tube size 50 x 15 x 150 cm, with gravel size 3 - 5 mm as a compiler permeable layer and a mixture of andosol soil, fine charcoal and sawdust with a ratio of 2:0.5:0.5 as unpermeable layers.

Each model of MSL system was tested with different hydraulic loading rates of 250, 500, 1000, 2000, 4000 and 8000 $l/m^2/day$. Aeration is carried out for the highest wastewater rate tested. The results of the analysis are compared with the crumb rubber industrial wastewater quality standards according to Indonesia Ministry Decree No. 51 / MenLH /10/1995. The tools used in this study consisted of a set of multi-soil-layering installation equipment, 5.5 mm and 2.5 mm sieves, gloves, buckets, and laboratory equipment for analysis. Further, untreated and treated crumb rubber wastewater were analyzed on pollutant parameters: BOD, COD, TSS, total nitrogen (T-N), total ammonia (T-NH_3), and pH.

2.2 Methods

Pollutant parameter BOD is analyzed by Winkler Titrimetric Method (SNI 06-6989.57-2008), COD is analyzed by Iodometric method closed reflux (SNI 6989.2: 2009), TSS is calculated by Gravimetric method with Whatman microfiber glass filter no 42 and accordance with SNI 6989.3:2019. Total nitrogen (T-N) and total ammonia (T-NH_3) were determined by Nessler using a spectrophotometer DR 2000/2010. Meanwhile, pH is measured by a pH meter which has been calibrated by a buffer solution and accordance with SNI 6989.11:2019. The test was conducted with 3 repetitions. The percentage of reduction (%R) is determined by the percentage of reduction between untreated concentration (UC) and treated concentration (TC) as follows:

$$\% R = \frac{UC - TC}{UC} \times 100\% \quad (1)$$

3. Results and discussion

3.1 Pollutant parameters of untreated wastewater

The results of the analysis of untreated crumb rubber industrial wastewater are shown in Table 1. None of the pollutant parameters of untreated wastewater comply with quality standards. Untreated wastewater contains severe organic content BOD of 85-255 mg/l and COD of 259-830 mg/1, and its colour is brown with high turbidity. This result was aligned with [15], [16], [50] and reported that crumb rubber industrial wastewater has high impurities. Theoretically, this is due to the source of the ingredients it is natural material containing organic compounds such as proteins, carbohydrates, lipids and organic salts that will be decomposed by the activity of microorganisms over time [24], [51].

Table 1: Pollutant parameters of untreated crumb rubber industrial wastewater

Pollutant parameters	Untreated wastewater	Quality standard
BOD, mg/l	85 - 255	60
COD, mg/l	259 - 830	200
TSS, mg/l	150 - 540	100
T-N, mg/l	15 - 25	10
T-NH_3, mg/l	14 - 25	5
pH	5.0 - 5.5	6-7

Besides that, it is also due to the addition of lumpy materials and latex stabilizers [50], [52]. Untreated wastewater is acidic liquid due to acetic acid or formic acid utilization during the clumping process and also because of the formation of free fatty acids as a result of the activity of microorganisms.

3.2 MSL system on reducing pollutant parameters of treated wastewater

The results of the analysis of treated crumb rubber industrial wastewater by using MSL system are shown in Table 2.

Table 2: Analysis of treated crumb rubber industrial wastewater by using MSL

Pollutant parameters	Treated wastewater	Quality standard
BOD, mg/l	0.1 – 13	60
COD, mg/l	3 -76	200
TSS, mg/l	15 – 80	100
T-N, mg/l	0.2 – 9.4	10
T-NH_3, mg/l	0.04 – 4.9	5
pH	6	6-7

All of the pollutant parameters of treated wastewater comply with quality standards. MSL systems can reduce pollutant parameters of crumb rubber industrial wastewater, especially BOD and COD, which are the most critical in wastewater by more than 90%. Other parameters of TSS, T-N and T-NH_3 also showed a high decrease and an increase in pH to the specified limit and fulfilled the quality standard requirements. The comparison of MSL system's effect on minimizing wastewater between the crumb rubber industry and domestic waste is almost the same but higher than the palm oil industry [20], [21]. The decrease in BOD and COD of palm oil wastewater by MSL was 80% and 65%, respectively [53], [54]. The high ability of the MSL system in treating industrial crumb rubber wastewater is due to crumb rubber industrial wastewater containing only organic substances and suspended solids but not containing oils and fats such as those contained in the oil palm industrial wastewater, so the process of biodegradation of organic substances by microorganisms decomposers that live in a layer of soil mixture, the layers of gravel can run perfectly, as well as the process of absorption of suspended solids by other constituent components in the MSL installation. This is supported by the statement of [33] which states that the presence of fat oil along with dissolved suspensions tends to accumulate and cover the surface of the condenser while the substance is insoluble which results in blocking the activity of microorganisms [55]. Further comparison between untreated and treated wastewater, and the percentage of reduction is shown in Table 3.

Table 3: Analysis of treated crumb rubber industrial wastewater by using MSL

Pollutant parameters	Untreated wastewater	Treated wastewater	% Reduction
BOD, mg/l	85 - 255	0.1 – 13	94.90 - 99.88
COD, mg/l	259 - 830	3 -76	90.84 - 98.84
TSS, mg/l	150 - 540	15 – 80	85.18 - 90
T-N, mg/l	15 - 25	0.2 – 9.4	62.4 - 98.67
T-NH_3, mg/l	14 - 25	0.04 – 4.9	80.4 - 99.71
pH	5.0 - 5.5	6	Comply the standard

Table 3 shows that the MSL system effectively reduces all pollutant parameters in crumb rubber industrial wastewater, especially BOD and COD. This is supported by the study [11], [24]. The change in pH from 5.0 - 5.5 to 6 is due to the presence of a soil layer on the MSL system. Theoretically, soil can neutralize pH due to the soil's ability to hold basic cations such as Ca^{2+}, Mg^{2+}, Na^+, K^+ and acidic cations such as H^+ and Al^{3+}, thus when the soil is in an acidic condition, there will be an exchange of acidic cations with basic cations and vice versa [56]. The existence of the exchange can cause a change in pH, both a change in pH caused by the soil itself and a change in soil pH caused by the presence of other substances in the soil or passing through the soil. This study is supported by [57] that soil has a high buffering capacity against changes in chemical and physical conditions due to the activity of microorganisms and physical reactions caused when the liquid waste treatment mechanism occurs in the MSL system.

The effectiveness of the MSL system in reducing all pollutant parameters because the MSL mechanism consists of two treatment zones, the aerobic zone and the anaerobic zone. The aerobic zone is found in the rock layer (gravel) and the space between the rock layer and the andosol soil mixture block, while the anaerobic zone is in the soil mixture layer. The wastewater treatment process in the MSL system consists of decomposition, fixation, nitrification, denitrification, filtration, adsorption, and absorption [11]. The process of decomposition of organic material occurred in the aerobic and anaerobic zones. The adsorption process is on the surface of the energetic mixed layer in the aerobic zone and the rock layer while the absorption process is in the anaerobic zone in the soil mixed layer. The organic material of the wastewater is adsorbed in the upper layer of the andosol soil mixture and the surface of the rock layer will then be decomposed by the aerobic microorganisms that stick there. While adsorbed into the soil mixture layer will be decomposed by anaerobic microorganisms that live in the soil mixture layer. Filtration of suspended and dissolved substances occurs when liquid waste enters the layer of the MSL system. The reference states that the organic material of the liquid waste is adsorbed in the top layer of the andosol soil mixture as well as the surface of the gravel. Microorganisms in the soil and in the biofilm formed on the zeolite decompose the adsorbed and absorbed organic material [58], [59].

3.3 Effect of hydraulic loading rate on pollutant parameters

The results of the analysis of the effect of variations in Hydraulic Loading Rate (HRL) on the percentage of decreasing levels of parameters (BOD, COD, TSS, T-N, and T-NH_3) and pH are presented in Figure 2.

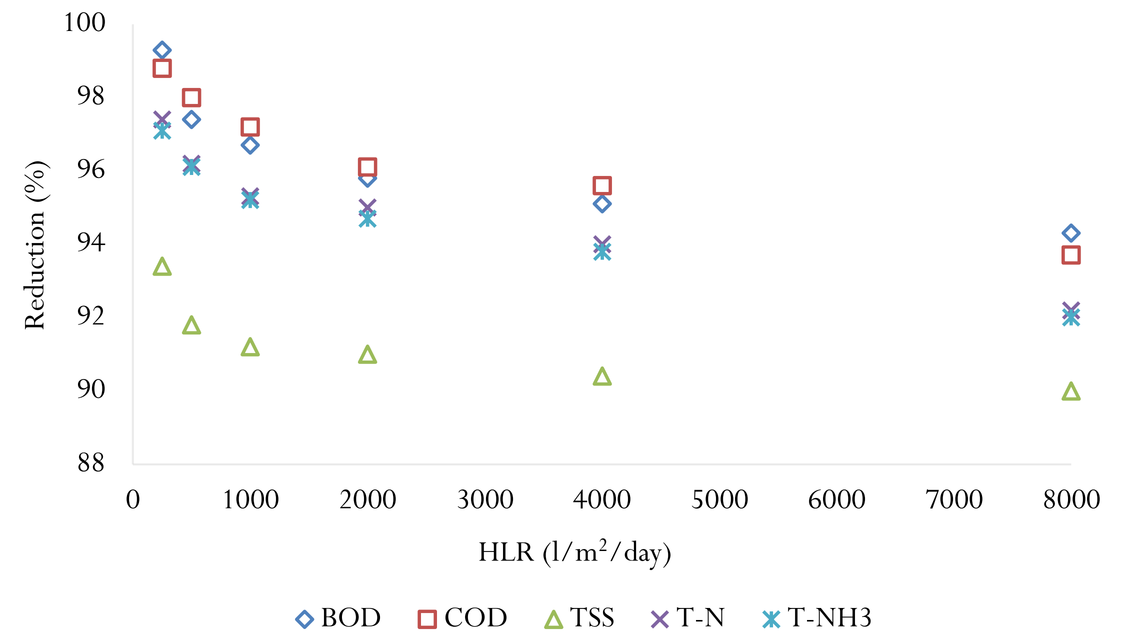

Figure 2: Effect of HLR on pollutant parameters in reduction percentage

HRL affects the percentage decrease in pollutant parameters (BOD, COD, TSS, T-N, and T-NH_3). The percentage is inversely proportional to HRL, the higher the percentage of decline and vice versa. This is aligned with [23] that the HRL is inversely proportional to the percentage of reduction in the treated wastewater. The process that occurs in MSL which consists of the filtration process, decomposition, nitrification, and denitrification, organic substances and suspended solutions in wastewater will be filtered and degraded by a population of microorganisms that attach to the surface of gravel and layers of soil mixture. Further absorbed into the biofilm as a plant food ingredient. While nitrogen as ammonia will be broken down by the Nitrobacter bacteria in the nitrification process that occurs in the layers of gravel, nitration will be converted to nitrogen gas in the soil mixture [58]. The lower the HRL, the process will take place slowly too, so the residence time will be longer. With longer contact at each layer, the process runs perfectly. Likewise, HRL is higher, processes are fast and partial. Hydraulic loading rate significantly influences wastewater treatment, especially in the rate of decomposition and denitrification.

In Figure 2, MSL system is capable of receiving high HRL up to 8000 l/m^2/day and is still able to reduce the levels of pollutants effectively. MSL system is better compared with other natural systems wastewater treatment methods such as constructed wetland, overland flow, and septic tank systems [8], [33]. This is due to MSL structure being made like the arrangement of bricks so that the permeability becomes high and the processing will take place repeatedly on each layer. The structure of MSL system (a brick structure) will be able to receive a high loading rate and can overcome blockages and allow them to occur repeatedly in each layer. However, HRL does not affect the pH of crumb rubber industrial wastewater. This is due to the nature of the soil can neutralize, thus, the soil has a high buffering capacity for changes in chemical and physical conditions due to the influence of micro-organism activity. The physical reactions caused when the wastewater treatment mechanism occurs in the MSL system, such as changes in pH caused by the quality of incoming water or the process of nitrification of ammonium, can be easily neutralized by ion-on H-exchange and other cations in the body and gravel surfaces.

3.4 Effect of aeration on pollutant parameters

The effect of aeration is considered as the percentage decrease in pollutants parameters of crumb rubber industrial wastewater showed significant results. The results showed that aeration had a significant effect on the percentage decrease in pollutant parameters, especially against BOD, COD, TSS, T-N, and T-NH_3. The percentage reduction in pollution levels increases with aeration, ranging from 85% to 99%. In theory, the MSL system's ability to decompose waste in anaerobic conditions is affected by aeration. According to [11], sufficient aeration treatment can help aerobic formation, however after aeration for some time anaerobic conditions will be formed by itself in the MSL system. This effect is due to aeration assisting in increasing the supply of dissolved oxygen needed by microorganisms to carry out biodegradation activities on organic substances contained in waste, especially in the middle and lower layers of the installation. In wastewater systems, aerobic microorganisms require dissolved oxygen to carry out activities, if oxygen is limited, it can cause the death of these microorganisms [60].

3.5 Effect of MSL model composite structure and components on pollutant parameters

The treatment of various models in the structure and constituent components of MSL on the percentage of decrease in pollutants parameters (BOD, COD, TSS, T-N, and T-NH_3) on the crumb rubber industrial wastewater in Figure 3.

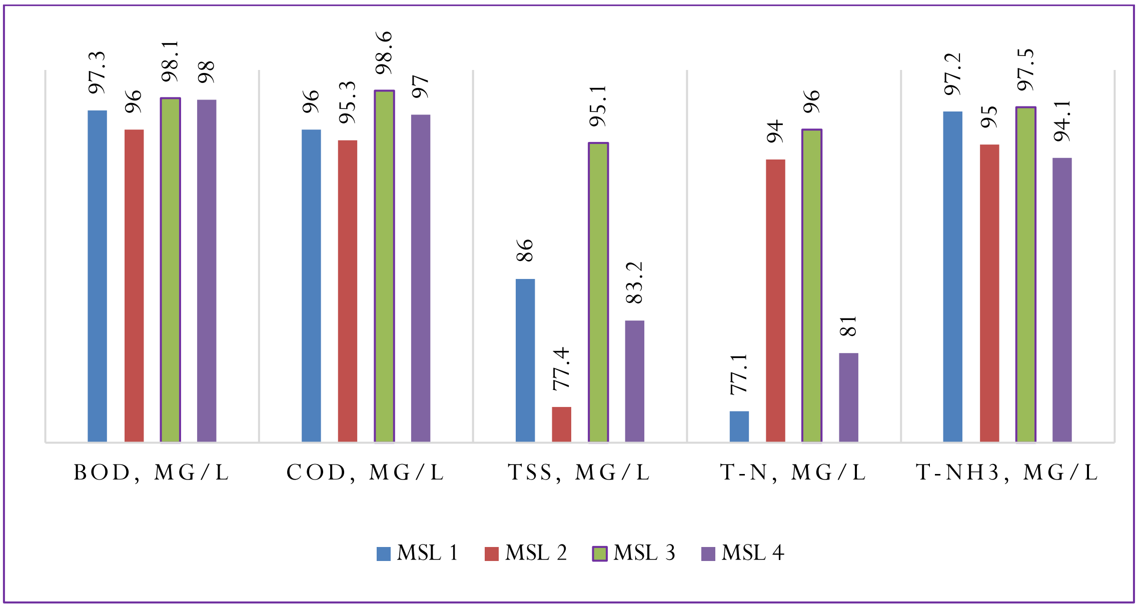

Figure 3: Effect of HLR on pollutant parameters in reduction percentage

Results showed that model MSL 3 reduces the highest percentage of pollutant parameters covering BOD, COD, TSS, T-N, and T-NH_3 compared to other MSLs models. MSL 3 is equipped with a tube size 50 x 15 x 150 cm, with gravel size 3 - 5 mm as a compiler permeable layer and a mixture of andosol soil and fine charcoal with a ratio of 2:1 as unpermeable layers.

4. Conclusion

In this study, wastewater from the crumb rubber industry has been minimized by using a Multi-Soil-Layering system to comply with quality standard requirements. The main findings of this study are summarized below:

1. MSL systems significantly minimize the crumb rubber industrial wastewater to comply with the quality standard requirement with pollutant parameters biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solid (TSS), total nitrogen (T-N) and total ammonia (T-NH_3).
2. Hydraulic loading rate (HLR) affects the percentage reduction in pollutant parameters (BOD, COD, TSS, T-N, and T-NH_3). The percentage is inversely proportional to HLR, the higher the percentage of decline and vice versa. MSL system is capable of receiving high HRL up to 8000 l/m_2/day and is still able to reduce the levels of pollutants effectively
3. MSL also significantly neutralizes the power of hydrogen (pH) up to 6.
4. The aeration significantly decreased the BOD, COD, TSS, T-N, and T-NH_3 ranging from 85% - 99%. However, MSL without aeration has complied with the quality standard requirement. Although MSL equipped with aeration process has better performance than without aeration, treated wastewater from the crumb rubber industry by MSL system without aeration has complied with the quality standard requirements.
5. The highest reduction was obtained by MSL 3 model (a tube size 50 x 15 x 150 cm, with gravel size 3–5 mm as a compiler permeable layer and a mixture of andosol soil and fine charcoal with a ratio of 2:1 as unpermeable layers).

In conclusion, MSL system has a significant effect on reducing pollutant parameters such as BOD, COD, TSS, T-N, and T-NH_3 in crumb rubber industry wastewater and complying with the standard based on Indonesia Ministry Decree No. 51/MenLH /10/1995. However, some recommendations for further studies could be provided for better results. For further studies, MSL could be applied to other industrial wastewater and consider another advanced process to improve MSL system to produce water supply for industrial processes. Another recommendation is physical pretreatment such as sedimentation or settling tanks prior to the MSL system to decrease the working load of MSL system, especially TSS. Further studies also could apply other carbon active such as coconut shell as unpremeable layers.

Author contribution

Sri Rizki Putri Primandari: research conceptualization & writing – original draft. Mulianti: data gathering; Methodology. Gusni Sushanti: data gathering, writing-result analysis. Mohamed Gabbasa: Writing – review & editing; Visualization. Muneer M. Ba-Abbad: Writing – review & editing.

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Competing interest

No potential conflict of interest was reported by the author(s).

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