Reduction of Pollutant Parameters in Textile Dyeing Wastewater by Gambier (Uncaria gambir Roxb) Using the Multi Soil Layering (MSL) Bioreactor

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Abstract. Gambier (Uncaria gambir Roxb) is one of the sources of natural dyes that able to apply to the textile industry. Textile dyeing industrial wastewater using Gambier comes from several sources. Among them come from the dyeing process itself and from the fixation process wastewater using alum (Al2(SO4)3), lime (CaO) and tunjung (FeSO4). The wastewater was characterized by high levels of organic pollutants and dyestuffs as well as compositions of fixator raw materials. This study explored the performance of two types of Multi-Soil-Layering (MSL) bioreactors in order to reduce pollutant parameters in such a wastewater. The result revealed that MSL showed good performances. The more MSL layers, the higher the reduction. Reduction of BOD5, COD, and TSS in MSL type 1 could reach 98.6%, 95.3% and 87.1% respectively, while 98.1%, 93.8% and 84.9% in MSL type 2, respectively. The MSL bioreactor could neutralize pH from pH 4.7 to be pH 7.4 and pH 7.1 and able to reduced colour from 427 PtCo to 0.4 PtCo and 1.3 PtCo as well as reduced turbidity from 47 NTU to 0.8 NTU and 1.2 NTU by MSL type 1 and MSL type 2 respectively.

1. Introduction

Gambier is one of the natural potential local dyes with the largest production in West Sumatra Province. The Gambier production process produces about 25 percent wastewater in the form of brown to blackish brown liquid [1]. Utilization of Gambier wastewater as textile dyes has been widely studied. These dyes have been applied to dye cotton fabrics [1], rayon [2, 3], silk [4, 5] and weaving yarns [6]. The coloring process is also done on batik cloth [3, 7, 8].

The use of Gambier wastewater as textile dyes can reduce the using of synthetic dyes that will able to reduce environmental pollution and support environmental issues by creating environmentally friendly textile products. Besides that, nowadays the use of natural dyes as textile dyes is a trend because natural dyes have special, unique, ethnic and exclusive colors that synthetic textile dyes cannot have. Gambier wastewater can produce color varies, depending on the color fixation agent and the concentration of wastewater used [4].

Currently the utilization of Gambier is being developed as a textile natural coloring agent. However, this activity produces wastewater even though in the form of organic wastewater, but it can still damage the environment aesthetically. Although now a day it is still in small amounts, the accumulation also has the potential to pollute the environment. Textile dyeing wastewater with Gambier and Gambier wastewater are a combination of wastewater from mordan activity, dyeing
(coloring), color locking (fixation), and washing process. The characterization of textile dyeing wastewater by Gambier is determined by the composition of Gambier raw materials. They consist of organic substances as well as compositions of fixators raw materials. They are mainly such as tannins, catechins and dyes as Gambier component, Al derived from Al₂(SO₄)₃ as color generator towards yellow, Ca from CaO as color generator towards brownish to red, and Fe from FeSO₄ as color generator towards a green to blackish green [4], [9].

A Multi Soil Layering (MSL) bioreactor is one method of processing wastewater that utilizes soil as the main medium, by increasing the ability of the soil through its structural changes. It is composed of soil mixture layers and permeable layers that are arranged in a brick-layer-like pattern, and this structure keeps the water permeability high and reduces the risk of clogging of the system. MSL bioreactors consist of two processing zones, namely aerobic zones that occur in the permeable layer and anaerobic zone in the impermeable layer. The treatment process that occurs in this MSL system consists of filtration, adsorption, absorption, biodegradation, fixation, nitrification, and denitrification [10, 11]. The advantage of MSL bioreactor is that they are low-cost, maintenance is technically easy, but requires more land than a mechanical treatment system.

Research on wastewater treatment, especially organic wastewater with the MSL bioreactor has been widely reported. Among them are for domestic wastewater treatment [10], [12-15], polluted river water [11], medium scale food industry [16], livestock wastewater [17], dairy industry [18], tofu industry [19], edible oil refinery [20], Crumb rubber industry [21].

Gambier dyeing wastewater treatment has never been reported yet. While this wastewater contains high organic pollutant and colors. The purpose of this study is to explore the performance of two types of MSL bioreactors in treating Gambier textile dyeing wastewater and reducing these pollutant parameters. This is also aims to anticipation for the development of utilization of Gambier as a textile dye.

2. Materials and methods

2.1. Design and installation of MSL

Figure 1 shows the two laboratory-scale MSL bioreactors structures used in this study. The MSL bioreactor type 1 were packed in W50 cm × H150 cm × D15 cm and the MSL bioreactor type 2 were packed in W50 cm × H100 cm × D15 cm acrylic boxes. They had differences in the number of soil mixture layers. 10 layers for MSL bioreactor type 1 and 7 layers for MSL bioreactor type 2 with arranged to form an alternative brick layer-like pattern. The main component of the soil mixture layers was Andesol type of soil and charcoal. They act as impermeable layers or the anaerobic layers. Andesol soil was obtained from the top surface soil of Mount Merapi at Padang Panjang city West Sumatera, Indonesia. Comparison of Andesol soil and charcoal was 2: 1. The composition was calculated through analysis of water content and bulk density. This mixture of soil with charcoal was then formed into a brick shape, with dimensions of 10 cm long, 15 cm wide and 5 cm high. The distance between the bricks was 2 cm and the distance between the first layer and the second layer was 5 cm. The aerobic layer or permeable layers of the two bioreactors were Perlite. Perlite, with a diameter of 3–5 mm, was used to fill the void spaces between adjacent soil mixture layers. Stone, with a diameter 2-4 cm was used to fill the bottom of the installations. The Bioreactors were equipped with an inlet pipe and outlet pipe. A porous inlet pipe for wastewater was placed in the top layer of perlite.
2.2. Wastewater level of contamination and HLR
Textile dyeing wastewater with Gambier comes from vive sources. One source comes from dyeing process, three sources from fixation process which is fixation using alum (Al₂(SO₄)₃), lime (CaO) and Tunjung (FeSO₄) and the other source comes from washing process. The characterization of it was done by analyzing the composition of the parameters contained in wastewater. The results of the analysis can be seen in Table 2. The wastewater from all fixation process and the washing process was collected in the wastewater equalization tank added by the spilled wastewater from the dyeing process. They were diluted by the washing process wastewater automatically. However, at a certain time, dyeing wastewater will be added into, because dyeing wastewater is still reused to a certain extent where it no longer provides good color quality. The wastewater used in this study was wastewater at the lowest level of contamination (Table 3). Wastewater was flowed in a cascade into the MSL bioreactor with a flow rate of 100 L/m² h.

2.3. Characterization and analysis
Main material using in bioreactor and the textile dyeing wastewater with Gambier were characterized and analyzed. Elemental composition was determined by XRF (X-ray fluorescence spectrometer) PANalytical Epsilon3, available surface functional groups were qualitatively determined by Fourier Transform Infrared (FTIR) spectroscopy (SpectrumTM One Spectrometer, Perkin Elmer). Properties of the main constituent material of the bioreactors shown in Table 1 and Figure 2. pH was determined by pH meter, Biological Oxygen Demand (BOD) was calculated by Winkler method, Chemical Oxygen Demand (COD) were measured by spectrophotometer, Total Suspended Solid (TSS) was determined by gravimetric method, color analysis with UV 1601 Shimadzu spectrophotometer, and turbidity with turbidity meter. The characterization of textile dyeing wastewater by Gambier is shown in Table 2.

3. Results
3.1. The characterization of materials and wastewater Analysis
Characterization of the main material build up the structure of the MSL bioreactor has high metal oxide and carbon content. Especially alumina and silica in Andesol soil and perlite and carbon content in charcoal (Table 1). Figure 1 shows that Andesol soil and Perlite also have some functional groups. The peak at 3189 cm⁻¹ – 3451 cm⁻¹ was stretching vibration mode of the O-H bond in surface hydroxyl groups. The peak at 1624 cm⁻¹ can be attributed to the stretching vibration mode of C=O bond or C=C
bond, and a peak was observed at 1025 cm\(^{-1}\) and at 1041 cm\(^{-1}\), indicating the presence of Si-O-Si or Si-O-C bond [22]. All of them assumed to play an important role in the colour adsorption process of Gambier dyeing wastewater.

Characterization of Textile dyeing wastewater with Gambier was acidic and has a high BOD, COD and TSS content (Table 2). It due to that Gambier was a natural organic material that was easily degraded by microorganisms and will produce acidic compounds [4]. Unless the fixation process wastewater used alkaline that fixation solution itself was the addition of 0.5% lime (CaO) which was dissolved into water to generate colour after the dyeing process with Gambier solution.

Table 1. Properties of main constituent material of the MSL bioreactors

| Parameter (%)          | Andesol | Charcoal | Perlite |
|------------------------|---------|----------|---------|
| Al\(_2\)O\(_3\)        | 22.618  | 1.803    | 15.759  |
| SiO\(_2\)              | 50.624  | 29.276   | 79.365  |
| CaO                    | 4.496   | 37.981   | 0.619   |
| SO\(_3\)               | 2.217   | 5.923    | -       |
| P\(_2\)O\(_5\)         | 3.158   | 11.077   | 0.519   |
| Fe2O3                  | 11.761  | 4.004    | 0.424   |
| C\(^+\)                | -       | 66.59-72.14 | -      |

Figure 2. FTIR spectra of main constituent material of the MSL bioreactors

Table 2. The characterization of textile dyeing wastewater by Gambier

| Parameters  | Dyeing Wastewater | Fixation Process Wastewater |
|-------------|-------------------|-----------------------------|
|             | (Al\(_2\)(SO\(_4\))\(_3\)) | Lime (CaO) | Tunjung (FeSO\(_4\)) |
| pH          | 4.8               | 3.48                      | 10.6          | 2.98           |
| BOD (mg/L)  | 3678.29           | 885.68                    | 729.31        | 592.43         |
| COD (mg/L)  | 84506             | 2925                      | 1791          | 3405           |
| TSS (mg/L)  | 1835              | 732                       | 692           | 997            |
3.2. Effect of treatment on pH values

![Figure 3](image-url)

**Figure 3.** Effect of treatment of the MSL bioreactors Type 1 and Type 2 on pH value.

Figure 3 shows that textile dyeing wastewater by Gambier was acidic with a value of pH 4.72 on the MSL inlet. After treated by flowing into the bioreactors, the pH values were changed into a Neutral pH (pH 7.1 in MSL type 1 and 7.4 in MSL type 2). The change in pH was due to the presence of soil layers in the MSL reactor. Soil can neutralize pH because of the ability of the soil to hold basic cations such as, Ca\(^+\), Mg\(^+\), Na\(^+\), K\(^+\) and acid cations such as H\(^+\) and Al\(^{+3}\). So that if the soil is in acidic conditions there will be acid exchange cations with alkaline cations. The existence of these exchanges able to cause changes in pH, both changes in pH caused by the soil itself, such as weathering or changes in soil pH caused by the presence of other substances that passing through the soil. Soil has a high buffering capacity for changes in chemical and physical conditions due to microorganism activity and physical reactions caused when the mechanism for treating wastewater occurs in the MSL system [11, 13].

3.3. The efficiencies of the MSL bioreactors

| Parameters     | Unit     | Concentration |
|----------------|----------|---------------|
| pH             | -        | 4.72          |
| Colour         | (PtCo)   | 427.5         |
| Turbidity      | (NTU)    | 46            |
| BOD            | (mg/L)   | 325           |
| COD            | (mg/L)   | 426           |
| TSS            | (mg/L)   | 232           |

Figures 4 and 5 show that both types of MSL ware efficient in reducing color, turbidity, BOD, COD and TSS pollutant parameters in textile dyeing wastewater with Gambier. MSL structure affects the efficiency of pollutant parameters reduction. The more MSL layers, the higher the reduction, it because of the improved contact between wastewater and soil mixture layers [12, 14].
adsorption process of color and turbidity occurs in layers of mixed soil and perlite layers repeatedly as well as the process of filtration and decomposition of organic pollutants. MSL 1 was able to reduce color up to 0.4 \( \text{PtCo} \) and turbidity up to 0.75 NTU, while MSL 2 can reduce color up to 1.2 \( \text{PtCo} \) and turbidity up to 1.12 NTU. Reduction of BOD, COD, and TSS in MSL type 1 could reach 98.6\%, 95.3\% and 87.1\% respectively, while 98.1\%, 93.8 \% and 84.9\% in MSL type 2 respectively. The possible process of treating wastewater in MSL bioreactors consists of decomposition, fixation, nitrification, filtration, adsorption, and absorption. That process occurred in the aerobic and anaerobic zone in the layer structure of the MSL bioreactors repeatedly [10, 11].

**Figure 4.** Efficiency of MSL bioreactors in reduces color and turbidity in textile dyeing wastewater by Gambier.

**Figure 5.** Efficiency of MSL bioreactors in reduces BOD, COD and TSS in textile dyeing wastewater by Gambier.
4. Conclusion

The Multi-Soil-Layering bioreactors able to use to treat textile dyeing wastewater by Gambier effectively. MSL structure affects the efficiency of pollutant parameter reduction. The more MSL layers, the higher the reduction of pollutant parameters. It could be able to use as an alternative treatment to anticipate the development of utilization of Gambier as a textile dye.

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