

The Effectiveness of Gambir Leaves (*Uncaria gambir roxb*) as a Biocoagulant in Reducing Pollutants in Palm Oil Mill Effluent

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Abstract—Crude Palm Oil (CPO) production has been increasing and reached 46.82 tons in 2022 (BPS), and the wastewater from its manufacturing process is harmful to the environment. Palm oil mill effluent (POME) with high organic content can cause eutrophication, siltation, and death of aquatic organisms. One great method of treating this wastewater can be carried out through coagulation. In water treatment, chemical coagulants are typically used to remove impurities. However, biocoagulants from Gambir leaves offer a natural and environmentally friendly alternative. These biocoagulants can effectively treat water while reducing the need for synthetic chemicals, presenting a sustainable solution. Gambir leaves contain polyphenolic compounds that are effective as biocoagulants. This study examines the effectiveness of Gambir leaves as a biocoagulant in reducing palm oil effluent pollutants, including parameters such as TSS, BOD, COD, Oil and Grease (O&G), and Total Nitrogen. This study presents the initial steps in making biocoagulants and its application for POME wastewater. From the results of this study, it was found that Gambir leaves extract has the potential to reduce pH and remove TSS, BOD, O&G, and Total Nitrogen contained in POME with concentration variations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L, respectively.

Keywords: Biocoagulants, Gambir extract, POME, Physical treatment, Wastewater.

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1. Introduction

According to the Central Statistics Agency of Indonesia 2022 [1], crude palm oil (CPO) production has increased to 46.82 tons from the previous year. During the process of producing CPO, there are by-products in the form of water, oil, and organic solids from the remaining processing of fresh fruit bunches (FFB). This wastes will harm the ecosystem if discharged directly into the environment. The Minister of Environment Regulation No. 5 of 2014 concerning Wastewater Quality Standards stated the wastewater quality standards for the palm oil industry for the parameters Biological Oxygen Demand (BOD) of 100 mg/L, Chemical Oxygen Demand (COD) of 350 mg/L, Total Suspended Solid (TSS) of 250 mg/L, Oil and Grease (O&G) of 25 mg/L, and Total Nitrogen of 50 mg/L.

Palm oil mill effluent (POME) still needs to be considered by the palm oil industry so that palm oil wastewater is produced that does not meet quality standards when discharged into the environment. POME has a high organic content that can result in eutrophication in waters, causing siltation and death of aquatic organisms [2]. One of the most effective wastewater treatment methods is the coagulation

process. Coagulation is the agglomeration of colloidal particles that form sediment. The occurrence of coagulation will cause dispersed substances to no longer form colloids. The principle of coagulation is the process of destabilizing suspended particles by adding coagulants and flocculants to reduce the repulsive forces between particles because the process of floc formation in stable conditions will not occur. The unstable particles then collide with each other and form larger flocs that settle quickly. Chemical coagulants and flocculants are often used in wastewater treatment because of their proven high efficiency in removing suspended particles [3]. The use of chemical coagulants in the coagulation process has the potential to produce sediment or sludge-containing chemicals. The emergence of these problems require innovation to properly treat POME, namely by using biocoagulants derived from Gambir leaves (*Uncaria gambir roxb*).

The potential for utilizing Gambir leaves as a biocoagulant is strengthened by its abundance in Indonesia, one of which is in West Sumatera because almost 90% of Gambir production comes from West Sumatra [4]. Gambir contains functional compounds in the polyphenolic compounds, resulting from plant secondary metabolites that comprise the tannin group [5]. The performance of biocoagulants is related to their functional groups. Hydroxyl groups are the functional groups that contribute most to the coagulation process in biocoagulants, followed by amine and carboxyl groups and proteins. Hydroxyl and carboxyl groups represent ionized groups present in biocoagulants. Hydroxyl and carboxyl groups can be released during coagulation, resulting in positively charged biocoagulant particles. These functional groups facilitate the charge neutralization mechanism that occurs during coagulation [6]. This research discusses the effectiveness of Gambir leaves as a biocoagulant in reducing the pollutants contained in POME. It is expected that through this research, the effectiveness of Gambir leaves extract in removing pollutant parameters such as TSS, BOD, COD, oil and grease, and total nitrogen can be well understood.

2. Materials and Method

2.1 Materials

This study used Gambir leaves and extracted them to make biocoagulants. The water sample used in this study is POME, which was taken from a plantation in West Sumatra. Measurements of pH, TSS, BOD, COD, oils and grease, and total nitrogen were carried out with pH meters, gravimetry, titrimetry with Winkler titration, titration using Ferro Ammonium Sulfate (FAS) solution using ferroin, gravimetry, and Kjeldahl indicators, respectively. All the experiments are conducted according to the standard methods (Please cite the reference)

2.2 Extraction of Gambir

Gambir leaves were washed with running water and dried in the open air. The dried samples were then pulverized with a blender. The sample powder weighed as much as 500 grams, was soaked using 3 liters of ethanol solvent (allowed to stand for 3-4 days), and then filtered. The extract was evaporated by the solvent to obtain pure Gambir leaves extract. The pure Gambir leaves extract was converted from liquid to solid with a freeze dryer [7]. Then, Gambir leaves extract with varied concentrations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L were prepared.

2.3 Jar Test Method

POME samples were prepared in 1 liter of beaker and tested for selected parameters. Gambir leaves extract as a biocoagulant was put into the sample, stirred quickly at 120 revolutions per minute (rpm) for 30 seconds, and stirred slowly at 60 rpm for 30 minutes using a magnetic stirrer. The coagulation process was stopped and allowed to stand for 60 minutes until a precipitate was observed. After the sediment was formed, the treated water was tested according to the standard methods for selected parameter. The efficiency value of Gambir leaves extracts as a biocoagulant against POME was determined by the formula [8]:

$$\text{Efficiency (\%)} = \frac{X_{\text{before}} - X_{\text{after}}}{X_{\text{before}}} \times 100\% \quad (1)$$

Description:

X_{before} = Pollutant concentration before the jar test

X_{after} = Pollutant concentration after the jar test

2.4 pH Test

pH testing on water samples was carried out based on the Indonesian National Standard (SNI) 6989.11:2019 using a pH meter [9]. The sample is put into a beaker and measured using a pH meter.

2.5 BOD Test

The method used in the BOD test is the titrimetric method with Winkler titration. Oxygen will oxidize Mn^{2+} in an alkaline atmosphere to form MnO^{2-} precipitate. Furthermore, adding sodium iodide in an acidic atmosphere will free iodine. BOD measurement consists of sample dilution, incubation for five days at 20°C , and measurement of dissolved oxygen during incubation to show the amount of oxygen required by the water sample. Dissolved oxygen is measured by the Winkler titration method [10].

2.6 COD Test

The method used in the COD test is titration using a Ferrous Ammonium Sulfate (FAS) solution using a Ferroin indicator. Organic compounds in water were oxidized by potassium dichromate solution in an acidic atmosphere at 150°C for 2 hours. Excess potassium dichromate was titrated by a FAS solution with a ferroin indicator [11].

2.7 TSS Test

The sample was homogenized and filtered using filter paper (Whatman Grade 934 AH, with a pore size of $1.5 \mu\text{m}$). The filter paper containing the sediment was heated for one hour at 105°C . The filter paper was then weighed, and the TSS value of the sample was obtained [12].

2.8 Determination of Oil and Grease Concentration

Oil and grease in water samples were extracted with organic solvents in a separating funnel, and to remove the remaining water, anhydrous Na_2SO_4 was used. In the extraction process, two layers will form; the top layer is collected and dried at room temperature, and the residue left behind is weighed as oil and grease [13].

2.9 Determination of Total Nitrogen Concentration

Kjedahl's method utilizes POME samples digested by adding sulfuric acid and catalysts. During this process, the acid breaks down the waste in the sample by oxidation and reduced nitrogen in the form of ammonium sulphate is liberated. Then, the extract was diluted, and NaOH was added. The ammonia formed was collected and titrated with an indicator, and total nitrogen was calculated [14].

3. Results and Discussions

The study results are summarized in Table 1. which shows the concentration per parameter in the sample before and after coagulation-flocculation using biocoagulant from Gambir leaves extract with the percentage removal value given in Table 2.

Table 1. Results of pH, TSS, Oil & Grease, Total Nitrogen, BOD, COD Parameters

Sample	Parameters						
	pH	TSS (mg/L)	O&G (mg/L)	Total Nitrogen (mg/L)	BOD (mg/L)	COD (mg/L)	
Standard Quality PermenLH No.5 2014	6 to 9	250	25	50	100	350	
Palm Oil Waste Water (POWW)	9.6 ± 0.00	226.00 ± 19.67	73.60 ± 17.26	714.4±0.00	15.33± 1.30	90.67 ± 9.00	
POWW +	Biocoagulant (1000 mg/L)	9.5 ± 0.00	102.89 ± 30.67	41.73 ± 3.52	140.0±0.00	15.33± 1.10	154.67 ± 9.00
	Biocoagulant (2000 mg/L)	9.4 ± 0.00	86.22 ± 21.49	44.40± 7.61	112.0±0.00	15.33± 1.30	240.00 ± 0.00
	Biocoagulant (3000 mg/L)	9.4 ± 0.00	101.56 ± 24,54	39.73 ± 5.68	98.06±0.00	15.47± 1.21	256.00± 0.00
	Biocoagulant (4000 mg/L)	9.3 ± 0.00	127.11 ± 12,31	39.87 ± 12.73	84.05±0.00	15.33± 1.30	256.00 ± 0.00

Table 2. Percentage of Pollutant Parameter Removal by Biocoagulant

Contents	Percentage						
	pH	TSS	O&G	Total Nitrogen	BOD	COD	
Palm Oil Waste Water (POWW)	Biocoagulant (1000 mg/L)	1.04%	54.47%	43.30%	80.39%	0.00%	-70.59%
	Biocoagulant (2000 mg/L)	2.08%	61.85%	39.67%	84.31%	0.00%	-164.71%
	Biocoagulant (3000 mg/L)	2.08%	55.06%	46.01%	86.27%	-0.87%	-182.35%
	Biocoagulant (4000 mg/L)	3.12%	43.76%	45.83%	88.24%	0.00%	-182.35%

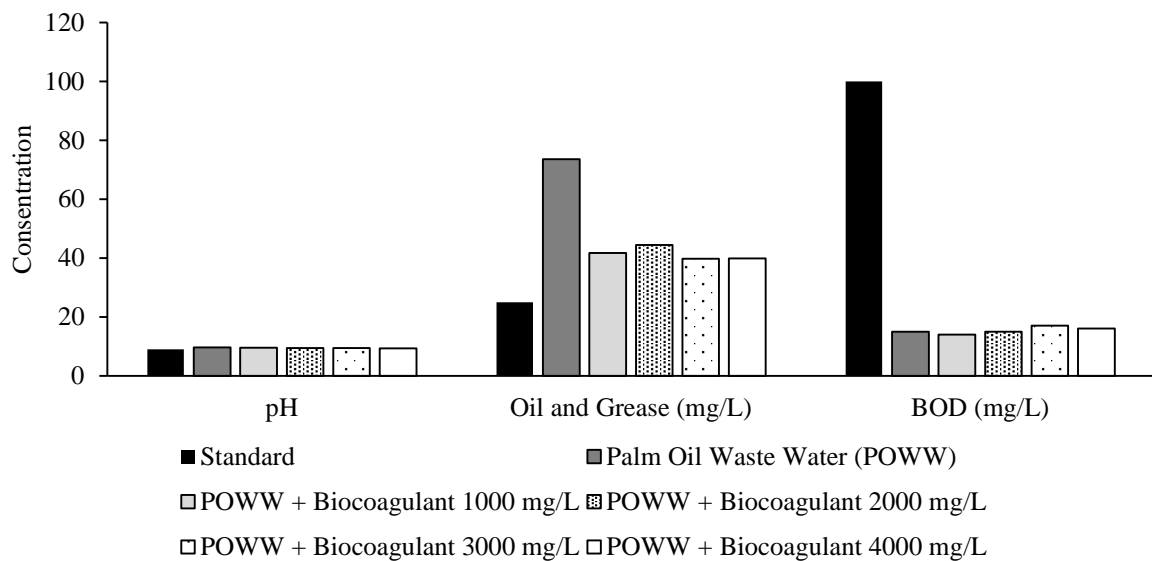


Figure 1. Results of pH, Oil & Grease, and BOD Parameters in Each Samples

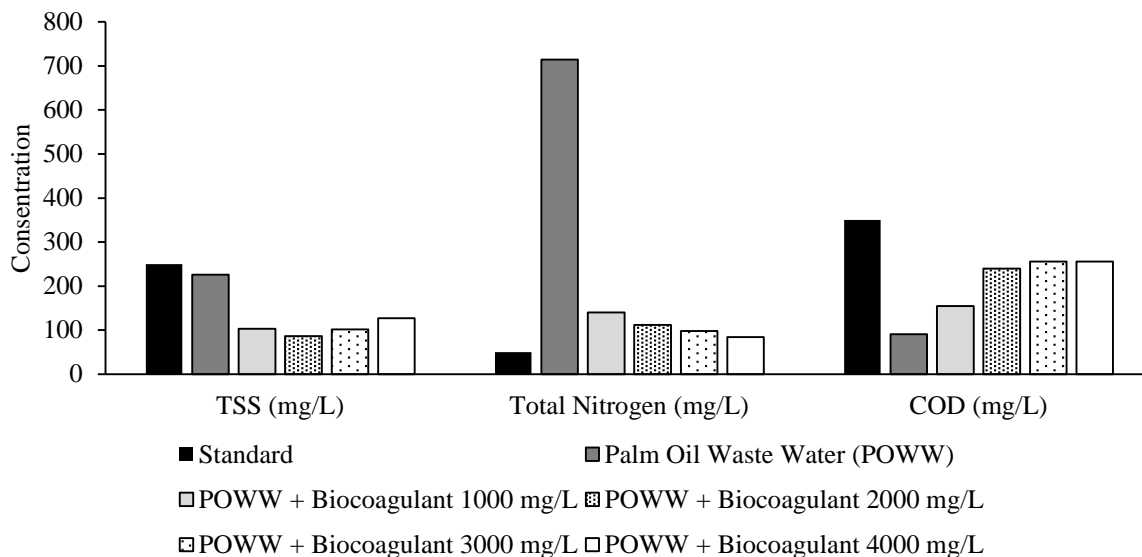


Figure 2. Results of TSS, Total Nitrogen, and COD Parameters in Each Samples

Figure 1. and Figure 2. show the test results of pH, BOD, COD, TSS, oil and grease, and total nitrogen in palm oil wastewater samples, palm oil wastewater with 1000 mg/L biocoagulant treatment, palm oil wastewater with 2000 mg/L biocoagulant treatment, palm oil wastewater with 3000 mg/L biocoagulant treatment, and palm oil wastewater with 4000 mg/L biocoagulant treatment. The color difference in the diagram represents the difference in samples, as stated in the figure. Based on Figure 1. and Figure 2. it can be observed whether the value of polluting parameters before and after coagulation-flocculation has decreased or increased.

The pH value of POME samples before and after coagulation did not show a significant decrease, as seen in Table 1 and Figure 1. The pH value of palm oil wastewater samples before coagulation was 9.6 ± 0.00 . After coagulation-flocculation with the addition of biocoagulants with variations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L, the pH value of the sample decreased to 9.5 ± 0.00 ; 9.4 ± 0.00 ; 9.4 ± 0.00 ; and 9.3 ± 0.00 with a percentage decrease of 1.04%; 2.08%; 2.08%; and 3.12%, respectively. The optimum biocoagulant concentration to reduce pH is 4000 mg/L with a percentage decrease of 3.12%.

TSS values in POME samples before and after coagulation-flocculation using Gambir leaves extract biocoagulant with variations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L were 226.00 ± 19.67 mg/L, respectively; 102.89 ± 30.67 mg/L; 86.22 ± 21.49 mg/L; 101.56 ± 24.54 mg/L; and 127.11 ± 12.31 mg/L with the percentage of TSS removal by biocoagulant for each variation of 54.47%; 61.85%; 55.06%; and 43.76%. Adding Gambir leaves extract biocoagulant to wastewater samples can reduce TSS levels, as shown in Figure 2. The most optimum concentration of biocoagulant from Gambir leaves extract to remove TSS is 2000 mg/L with a removal percentage of 61.85%.

The amount of oil and grease before and after coagulation-flocculation using Gambir leaves extract decreased as shown in Figure 1. The concentration of oil and grease before treatment using biocoagulant was 73.60 ± 17.26 mg/L. Treatment using Gambir leaves extract biocoagulant concentrations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L could reduce the value of grease oil in the sample to 41.73 ± 3.52 mg/L; 44.40 ± 7.61 mg/L; 39.73 ± 5.68 mg/L; and 39.87 ± 12.73 mg/L, respectively with a percentage removal of 43.30%; 39.67%; 46.01%; and 45.83%, respectively. The most efficient concentration of Gambir leaves extract biocoagulant to remove oil and grease was 3000 mg/L with a removal percentage of 46.01%.

The amount of total nitrogen after coagulation-flocculation using Gambir leaves extract biocoagulant shows a reasonably high removal, as seen in Figure 2. The total nitrogen value of palm oil wastewater samples before treatment using Gambir leaves extract was 714.41 ± 0.00 mg/L, which means it exceeded the quality standard value (50 mg/L). After treatment using Gambir leaves extract with a concentration variation of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L can reduce the value of total nitrogen respectively to 140.08 ± 0.00 mg/L; 112.06 ± 0.00 mg/L; 98.06 ± 0.00 mg/L; and 84.05 ± 0.00 mg/L with a percentage removal of 80.39%; 84.31%; 86.27%; and 88.24%, respectively. The higher the concentration given to the water sample, the higher the removal of the total nitrogen value in the water sample. The optimum concentration of Gambir leaves extract biocoagulant to remove total nitrogen was 4000 mg/L with a removal percentage of 88.24%.

The BOD value of palm oil wastewater samples before and after coagulation did not show significant changes, as shown in Table 1. and Figure 1. The value of palm oil wastewater samples before and after coagulation-flocculation using Gambir leaves biocoagulant with variations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L shows a value that meets the quality standards with each value of 15.33 ± 1.30 mg/L; 15.33 ± 1.10 mg/L; 15.33 ± 1.30 mg/L; 15.47 ± 1.21 mg/L; and 15.33 ± 1.30 mg/L with the percentage of BOD removal by biocoagulant for each variation of 0.00%; 0.00%; -0.87%; and 0.00%, respectively. The BOD concentration after coagulation did not show significant changes in the BOD value, although at a biocoagulant concentration of 3000 mg/L, there was an increase in BOD value from 15.33 ± 1.30 mg/L to 15.47 ± 1.21 mg/L.

COD values in wastewater samples before and after coagulation-flocculation using Gambir leaves extract biocoagulant show values that still meet the quality standard (350 mg/L). On contrary to the other parameters, after being treated using biocoagulants, it was found that the COD value increased as shown in Figure 1. COD values before and after coagulation-flocculation using Gambir leaves extract biocoagulant with variations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L were 90.67 ± 9.00 mg/L; 154.67 ± 9.00 mg/L; 240.00 ± 0.00 mg/L; 256.00 ± 0.00 mg/L; and 256.00 ± 0.00 mg/L, respectively, with a percentage increase by biocoagulant for each variation of 70.59%; 164.71%; 182.35%; and 182.35%, respectively. The higher the concentration of the biocoagulant, the more it will increase the COD levels in the sample.

4. Conclusion

POME has a high pH concentration, oil and grease, and total nitrogen that exceeds the quality standard, so it is not suitable to discharge into the environment. Gambir leaves extract with various concentrations of 1000 mg/L, 2000 mg/L, 3000 mg/L, and 4000 mg/L has the potential to reduce pH and remove TSS, BOD, COD, oil and grease, and total nitrogen contained in palm oil liquid waste. The concentration of 4000 mg/L Gambir leaves extract biocoagulant was the most efficient in reducing acidity (pH) and total nitrogen with a removal efficiency of 3.12% and 88.24%, respectively. The concentration of 3000 mg/L Gambir leaves extract biocoagulant was the most effective to removed oil and grease with a removal efficiency of 46.01%. The concentration of 2000 mg/L Gambir leaves extracts biocoagulant most efficiently removed TSS with a removal efficiency of 61.85%. Coagulation-flocculation of palm oil effluent samples using Gambir leaves extract had no significant impact on BOD concentration. On the other hand, coagulation-flocculation of palm oil liquid waste samples using Gambir leaves extract resulted in an increase in COD concentrations in the samples along with an increase in the concentration of Gambir leaves extract biocoagulant. More in-depth research needs to be conducted on biocoagulants from Gambir leaves extract to maximize the potential of Gambir leaves extract as a natural coagulant.

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