Jurnal Biodjati 9(2): 218-227, November 2024

e-ISSN : 2541-4208
p-ISSN : 2548-1606

JURNAL BIODJATI

http://journal.uinsgd.ac.id/index.php/biodjati

Effectiveness of Fruit Peels Eco Enzyme in Reducing Total Chromium from Yogyakarta Tanning Industry Waste

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Received: 27 November 2023
Revise from: 29 December 2023
Accepted: 03 September 2024

DOI: 10.15575/biodjati.v9i2.31077

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Abstract. *The leather tanning industry is essential in Indonesia's economic sector, specifically in the export of hides and leather goods. The high production of goods is often accompanied by a large amount of waste disposal, containing elevated levels of chromium, which hurts the environment. To address this problem, eco enzyme containing high carboxyl groups is expected to be an effective solution. Therefore, this study aimed to determine the functional groups in fruit peels eco enzyme and its effectiveness in reducing total chromium content in leather tanning waste. The experimental process included the preparation of eco enzyme formulations, testing functional groups with FTIR, and determining total chromium content using AAS. Furthermore, the characteristics of waste were determined, including such as BOD, pH, temperature, ammonia, TDS, and TSS, followed by incubation of eco enzyme in waste, as well as data analysis with ANOVA and Duncan test. The results showed that eco enzyme formulations contained hydroxyl (O-H) and carboxyl (-COOH) functional groups, carbon-carbon single bonds (C-C), carbon-oxygen single bonds (C-O), carbon-carbon double bonds (C=C), and carbon-hydrogen bonds (C-H). All formulations effectively reduced chromium levels in leather tanning waste to value lower than quality standards. Based on the results, the best formulation was found to be eco enzyme A, which comprised one type of fruit, namely mango peels.*

Keywords: *chrome, eco enzyme, tannery, waste, yogyakarta*

Citation
Putri, C. R. R., Jati, A. W., & Yulianti, L. I. (2024). Effectiveness of Fruit Peels Eco Enzyme in Reducing Total Chromium from Yogyakarta Tanning Industry Waste. *Jurnal Biodjati,* 9(2), 218-227.

INTRODUCTION

The Leather tanning industry is among the rapidly growing sectors in Indonesia, spreading across various regions. This widespread distribution is significantly observed on the island of Java, including provinces of East Java, Central Java, West Java, Banten, DIY, and DKI Jakarta (Kuncoro & Soedjono, 2022). According to APKI data, the production capacity of this industry is approximately 70,000 tons per year (Kementerian Perindustrian Republik Indonesia, 2019).

The leather tanning industry in Java Island is dominated by the Magetan, Garut, and Yogyakarta regions (Kuncoro & Soedjono, 2022). In August 2022, leather goods were estimated as one of the three main export commodities of the Yogyakarta Special Region, with a value of US$ 5.4 million, accounting for 10.83% of total exports from January to August 2022 (Badan Pusat Statistik, 2022). Other leather tanning industries in Yogyakarta include the Fajar Makmur Leather Factory, PT Adi Satria Abadi, and PT Sinar Obor.

The increasing development of the leather tanning industry hurts environmental quality degradation, particularly regarding water waste discharge without treatment. The operational process produces solid, gas, and liquid waste, constituting the largest waste (Murti et al., 2013). This waste containing chemicals such as sulfide, ammonia, and chromium that are irritating, corrosive, and carcinogenic (Nugraha et al., 2018).

Total chromium is the only heavy metal parameter that requires significant attention in the wastewater quality standard for leather tanning industry activities. Despite its significance, several industries are often indifferent to the dangers of total chromium content in production waste. This leads to direct waste disposal without passing through a specific processing process (Setiyono & Gustaman, 2017).

Chromium (Cr) is a heavy metal pollutant that is toxic to water, capable of causing chronic poisoning of aquatic organisms and reducing water quality. Furthermore, the accumulation of Cr in organisms can interfere with metabolism by inhibiting enzyme activity, potentially leading to the death of aquatic organisms (Setiyono & Gustaman, 2017). To address this problem, the eco enzyme has been proven effective as an alternative solution for treating leather tanning waste with high chromium content.

Eco enzyme is a complex organic liquid resulting from the fermentation of organic matter, sugar, and water. The organic materials used in eco enzyme production include vegetable or fruit waste (Pratamadina & Wikaningrum, 2022), which is applied to treat domestic waste effectively by reducing TDS, BOD, COD, ammonia, nitrogen, and phosphate pollutants (Agustina, 2021). According to Wei et al. (2020), eco enzyme could reduce Cd levels in *Salvia miltiorrhiza* (Bge.) by approximately 25.31% due to microbial activity.

Zainudin & Kesumaningwati (2022) showed that solutions from pineapple, dragon fruit, banana, papaya, and orange wastes with concentrations of 0%, 5%, 10%, 15%, and 20% provided analysis results for heavy metals As, Pb, Hg, and Cd. Low et al. (2021) showed that fruit and vegetable eco enzyme were not effective in reducing Ca^{2+}, Na^{+}, and K^{+} concentrations. Pratamadina & Wikaningrum's study (2022) showed that domestic waste samples given a 10% solution had a detergent concentration of 0.3019 mg/L on the seventh day from an initial concentration of 1.9385 mg/L.

The content of eco enzymes obtained from citrus peels include flavonoids, alkaloids, quinones, saponins, amylase, protease, lipase, OH groups, and COOH groups (Vama & Cherekar, 2020). The presence of the carboxyl group (COOH) facilitates binding of metal cations in solution (Arifiyana & Devianti, 2020). Under higher pH conditions, carboxyl group tends to have a negative charge due to the ability to form carboxylate anions, thereby increasing absorbed Cr metal cations (Mawardi et al., 2014). Carboxyl groups are found in pectin polymers (Ciriminna et al., 2015), which are abundant in vegetable and fruit, constituting the main component of the primary cell wall. Specifically, the C-6 carboxyl unit serves as the backbone of pectin (Ciriminna et al., 2015).

In this study, eco enzyme formulation uses waste from five types of fruit with high pectin content, including orange, lemon, banana, and mango peels, with apple pulp. Through the fermentation process, pectin from fruit peels breaks down into simpler compounds, thereby enhancing the ability to absorb metals. Despite the promising potential of eco enzyme in metal processing, there is no report regarding Cr reduction in waste using eco enzyme. Therefore, this study aimed to evaluate the potential of eco enzyme to reduce Cr levels in leather tanning waste in Yogyakarta.

MATERIALS AND METHODS

Preparation of Eco Enzyme Formulations

A total of five eco enzyme formulations were developed, namely Eco Enzyme A (mango peels), Eco Enzyme B (mango and lemon peels), Eco Enzyme C (mango, lemon, and banana peels), Eco Enzyme D (mango, lemon, banana, and orange peels), and Eco Enzyme E (mango, lemon, banana, and orange peels, with apple pulp). Initially, fruit waste was cut into small pieces, followed by weighing of all ingredients with the ratio of palm sugar, fruit peels waste, and water, at 1 : 3 : 10 (100 gram : 300 gram : 1000 mL). The fermentation process was carried out for three months in a tightly closed container, with the lid opened once every day for approximately 10 seconds in the first week without further intense opening. Subsequently, filtration was carried out after 90 days and tests were conducted on eco enzyme 10 days after filtration (Patel et al., 2021).

Functional Group Testing with FTIR

Power cable was connected to the power source and the computer was turned on, followed by double-clicking of the Onmic desktop icon. The Experiment Set Up option was clicked and the menu settings on Acquisition Parameters were determined as follows: Wavenumber: 4000-650 cm-1; Resolutions: 8; Number of Scans: 32; and Final Format: Absorbance/Transmittance. Subsequently, the air background reading was carried out by clicking the Col Bkg option. After completion, the sample was positioned to cover the Diamond ATR and pressure was applied using a Press Knob. Sample reading was carried out by clicking the Col Smp option, followed by scanning and data processing.

Total Chromium Level Testing

Total chromium content was tested following the SNI 6989.17:2009 method using an Atomic Absorption Spectrophotometer (AAS) type iCE 3000 Series brand Thermo Scientific. The testing stages included the preparation of washing solution, diluent, and test solution, as well as creating standard 10 ppm chromium metal solution and chromium metal standard series solution. Additionally, a calibration curve was established, followed by calculating the total chromium content, which was tested before and after treatment.

Testing General Characteristics of Waste

Testing the general characteristics of waste included BOD (modified SNI 6989.72:2009), TSS (gravimetry: modified SNI 06-6989.3-2004), TDS (TDS meter), ammonia (spectrophotometry), temperature (DO meter), and pH (pH meter) parameters. Measurements were made before and after treatment using a DO meter, Whatmann Grade 934 AH filter paper, TDS meter brand Portable E-1 TDS & EC Meter, spectrophotometer multidirect brand Lovibond, and pH meter.

Wastewater Treatment with Eco Enzyme

A total of 450 mL of waste sample was mixed with 50 mL of eco enzyme solution. This mixture produced a 10% eco enzyme solution, which was left for 50 days (Patel et al., 2021). The experiment was carried out 3 times for each formulation.

Data analysis

The data analysis was conducted using one-way ANOVA and Duncan's test with a 95% confidence level. Specifically, Duncan's test was carried out when the results of the ANOVA test showed a significant difference. Data analysis and processing were carried out using SPSS 15.0 software.

RESULTS AND DISCUSSION

Functional Group Content of Eco Enzyme

The finished eco enzyme was obtained after 3 months of fermentation with a volume of 500 mL for each formulation. The pH values of Eco Enzyme A, B, C, D, and E formulations are respectively 3.31; 3.86; 2.81; 4.99; and 4.98, with their appearance shown in Figures 1 and 2. The characteristics observed included yellow to brownish color, fermented aroma, and sediment at the bottom.

Figure 1. Eco Enzyme A (a), Eco Enzyme B (b), Eco Enzyme C (c) EcoEnzyme D (d), and Eco Enzyme E (e).

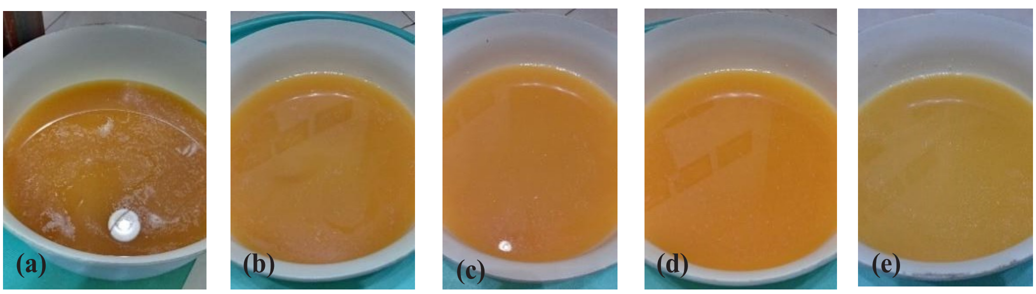

Figure 2. Filtering Results of Eco Enzyme A (a), Eco Enzyme B (b), Eco Enzyme C (c), Eco Enzyme D (d), and Eco Enzyme E (e).

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Fruit used in making eco enzyme was obtained from Pasar Buah dan Sayur Gamping. The five formulations were tested using FTIR to obtain their functional group content. The results obtained are shown in Figure 3.

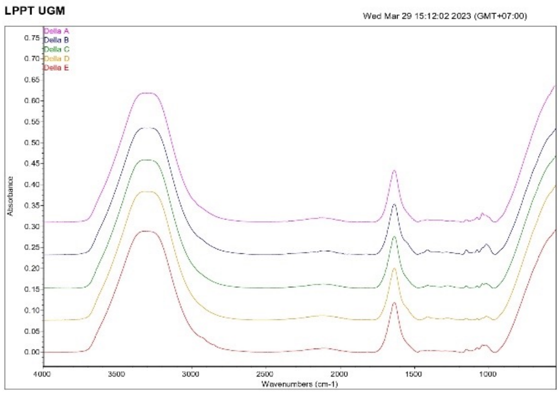

Figure 3. Fifth eco enzyme FTIR test.

The results in Figure 3 showed that all formulations contained relatively the same functional groups, including C-C, C-O, C-H, C=C, and O-H. This showed that the different types of fruit used in making eco enzyme does not affect functional groups content. According to Khaldun (2018), C-C, C-O, C-H (alkanes), and O-H groups could be found in absorption areas of 1000-1250 cm^{-1}, 1080-1300 cm^{-1}, 1350-1470 cm^{-1}, and 2500-3600 cm^{-1} (width), respectively. Nandiyanto et al. (2017) also reported that the C=C group (alkene) could be found in the absorption areas 1620-1680 cm^{-1} and 2100-2260 cm^{-1}.

Fruit used in eco enzyme formulations contained high pectin. However, the finished eco enzyme contained a pectinase enzyme capable of breaking down this pectin into smaller hydrocarbon compounds, including galacturonic acid. After the reshuffling process, several clusters such as C-C, C-H, C=C, C-O, and O-H could be detected on the FTIR spectrophotometer. This was in accordance with the study by Latupeirissa et al. (2019), where pectin extracted from sweet orange peels contained the functional groups O-H, CH3-, C=O, C=C, and C-O.

Ismail (2012) in Megawati & Machsunah (2016) state that pectin compounds contain functional groups C=O, RCOOH, R-O-R, and cyclic C-C. Roikah et al. (2016) also identified pectin extracted from starfruit, containing O-H, C-H, C=O, CH_3, and C-O. According to Valdivia-Rivera et al. (2021), the compounds obtained from mango fruit peels include C-O, C-H, C-C, and -COOH.

The five eco enzyme formulations obtained in this study contained carboxyl groups due to the presence of O-H stretching at the absorption peak of 2500-3600 cm^{-1}. The wide signal showed the presence of strong hydrogen bonds from the acid. According to Khaldun (2018), O-H group in the absorption area of 2500-3600 cm^{-1} (width) showed the type of alcohol, phenol, or carboxylic acid compound. Dachriyanus (2004) also reported that the O-H bond with a signal width of 2500-3300 cm^{-1} could be easily identified as an acid.

Characteristics of Tannery Waste

Tannery waste was tested for general characteristics, such as BOD, TSS, TDS, ammonia, temperature, and pH, after being treated with eco enzyme formulations. The measurements were carried out twice, before and after treatment. The results of all formulations presented in Table 1. The results showed that the levels of chromium, BOD, TDS, temperature, and pH in the treatment of all formulations were below the quality standards of Perda DIY, while TSS levels were relatively high. Ammonia levels in the treatment of eco enzyme A, B, C, and E were below the quality standard, while eco enzyme E was higher. Although the control treatment reduced the value of the parameters, a greater reduction was observed after treating waste with eco enzyme formulations.

Table 1. Characteristics of waste treatment eco enzyme A, B, C, D, and E.

No.	Test	Eco Enzyme					Control	Quality Standards
		A	B	C	D	E		
1	Chrome (mg/L)	0.1493	0.2144	0.1923	0.2384	0.1595	0.7721	0.5
2	BOD (mg/L)	0.23	0.83	0.30	0.77	1.03	1.7	50
3	TSS (mg/L)	60	66.67	73.33	80	67	80	50
4	TDS (ppm)	626	624	712.33	699	655	710	2000
5	Ammonia (mg/L)	0.043	0.023	0.103	0.027	11.53	0.3	0.5
6	Temperature (°C)	25.43	24.97	24.43	24.27	24.23	24.7	±3°C to air temperature
7	pH	7.51	7.32	6.64	7.43	7.80	7.39	6.0-9.0

Table 2. Chromium content in leather tannery wastewater.

Test	Before Treatment (mg/L)	After Treatment of Eco Enzyme (mg/L)					Control
		A	B	C	D	E	
Chrome	0.9637	0.1493[a]	0.2144[bc]	0.1923[b]	0.2384[c]	0.1595[a]	0.7721[d]

Table 2 showed that the five eco enzyme formulations reduced chromium content of leather tanning waste to below the quality standards of 0.5 mg/L. All waste that was treated had a value below 0.5, ranging from 0.1493 to 0.2384 mg/L. This decrease in chromium content was attributed to the presence of carboxyl group in all formulations. This was in accordance with Arifiyana & Devianti (2020), where the carboxyl group (COOH) could bind metal cations in solution. Chromium metal binding process was supported by the environmental pH conditions. Based on the results, the five eco enzyme formulations had a fairly high pH value, ranging from 6.64 to 7.80. These relatively high values caused the carboxyl group to form anions that tended to be negatively charged, thereby increasing the absorption of chromium. Similarly, Mawardi et al. (2014) stated that higher pH facilitated the formation of carboxylic anions, thereby increasing the absorption of metal cations will increase.

The sample with the lowest chromium content was found in the treatment with eco enzyme A formulation, which consisted of mango peels. Compared to other, mango peels contain the highest pectin content of 49.13% and more carboxyl groups, which facilitate the absorption of chromium metal. Azzahra & Taufik (2020) stated that higher concentrations of adsorbent led to increased adsorption efficiency. Additionally, mango peels contain cellulose, which is capable of generating negative particles interacting with the positively charged heavy metals in water. This was supported by Saiyidah et al. (2016), where 1 gram of mango peels could adsorb Pb with an efficiency of 93.67% at a contact time of 40 minutes. This absorption efficiency was greater than papaya peels at 65.82% and banana stems at 56.33%. Eco enzyme E also reduced chromium content, without significant difference from eco enzyme A due to the absence of apple pulp in both treatments. Apple pulp contains several functional groups and minerals that support the absorption of heavy metals. According to Enniya et al. (2018) in Sostaric et al. (2023), apple pulp contains carbohydrates, insoluble fiber, and cellulose, providing functional groups such as -CO, -COO, -OH, and -NH2, which have an affinity for cations. Sostaric et al. (2023) showed that apple pulp contains minerals K, Ca, Na, and Mg, playing a significant role in ion exchange mechanisms.

Despite the low levels of pectin, apple pulp has a relatively high cellulose content of 90.4%, which enhances the ability to bind chromium metal more effectively. This is in with the study by Ma et al. (2019), where 1,000 grams of apple pulp with a pectin content of 196 grams and a holocellulose of 243.9 grams contain 90.4% cellulose. Additionally, the relatively small size and wider contact area facilitate easier binding of metal ions. Syauqiah et al. (2020) stated that smaller particle size corresponded to greater absorbent surface area, thereby enhancing metal adsorption capacity. Sostaric, et al. (2023) also stated that apple pulp can reduce metal levels by half within one minute, twice faster than beetroot.

Chromium test results were analyzed using one-way ANOVA, showing that control treatment had the highest average content of 0.7721 mg/L. Meanwhile, the lowest average chromium content of 0.1493 mg/L was obtained in eco enzyme A treatment with a significant value smaller than 0.05. This showed that the treatment of different types of fruit in each formulation had a significant effect on chromium content in leather tanning waste, with a 95% confidence level. Subsequently, data analysis continued with Duncan's test using SPSS due to the significant difference observed in the one-way ANOVA results.

The results of Duncan’s test showed that eco enzyme A and E, B and C, as well as B and D treatment values, are in the same column, respectively. This showed that there was no significant difference between the treatments in the same column, with a 95% confidence level. However, only the control treatment values are in separate columns, showing a significant difference in results. The treatment with the lowest chromium content and the fewest types of fruit, specifically eco enzyme A was selected to ensure efficacy in reducing chromium levels in leather tanning waste, with a 95% confidence level.

All five eco enzyme formulations show effectiveness in reducing chromium content in leather tanning waste below the quality standard. There are also have no significant difference among formulations. However, eco enzyme A is identified as the most effective formulation, consisting of only mango peels, which facilitates easy procurement of ingredients.

CONCLUSION

In conclusion, this study showed that the five eco enzyme formulations contained hydroxyl (O-H) and carboxyl (-COOH) functional groups, as well as carbon-carbon single bonds (C-C), carbon-oxygen single bonds (C-O), carbon-carbon double bonds (C=C), and carbon-hydrogen bonds (C-H). Fruit eco enzyme proved effective in reducing chromium content in leather tanning waste to below the quality standard. Specifically, eco enzyme A was identified as the best formulation, requiring only one type of fruit to significantly reduce chromium content.

This study recommended that a microbial identification test should be carried out to further strengthen the discussion on the microbial content from incubation between waste samples and eco enzyme. Additionally, using alternative methods for ammonia tests could improve results accuracy, particularly for values close to zero. Future studies should also explore the efficacy of eco enzyme formulation made from different types of fruit, thereby allowing a comparison of metal absorption capacity.

AUTHOR CONTRIBUTION

C.R.R.P. designed, performed data collection, analyzed, and prepared the thesis manuscript. **A.W.N.J**. and **L.I.M.Y**. guided during the planning and implementation of this study, including the preparation of the thesis manuscript.

ACKNOWLEDGMENTS

The authors are grateful to PT Sinar Obor for providing waste from the leather tanning industry as the study material. Furthermore, the authors are grateful to LPPT UGM for providing assistance during the analysis of several parameters.

CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this study.

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