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Advanced oxidation processes technology using the Fenton method in Bakung landfill leachate treatment

Dedi Teguh*[1], Devy Cendekia[1], Yeni Ria Wulandari[1], Cynthia Eka Putri[1], and Adityas Agung Ramandani[2]

[1]Departement Industrial Chemical Engineering Technology, Politeknik Negeri Lampung, Indonesia.
[2]Departement Chemical Engineering and Materials Science, Yuan Ze University, Taiwan.

* Corresponding author's e-mail: dediteguh@polinela.ac.id

ABSTRACT

The primary issue at the Bakung final processing location was the high organic and inorganic material concentration. Toxicity, high chemical oxygen demand (COD), a black hue, and an unpleasant smell can usually identify landfill leachate; it can be hazardous if spilled directly into a body of water. Consequently, the implementation of an efficient leachate treatment was necessary. One use of this cutting-edge oxidation process technology consists of the Fenton method for treating leachate. The goals of this study involve implementing the Fenton technique to leachate treatment and ascertaining the impact of reaction time (30, 60, 90, or 120 min) and Fenton molar ratio (1:150, 1:200, or 1:250) on color degradation and chemical oxygen demand (COD). A Fenton molar ratio of 1:150 and a 120-min reaction period were ideal, resulting in maximum color and COD degradation percentages of 91.4% and 92.4%, respectively. The study's findings demonstrate how well the Fenton approach works to reduce COD and color in leachate while also meeting leachate water quality requirements for enterprises and/or ultimate waste processing operations.

Keywords:
Bakung landfill; AOPs; wastewater treatment; fenton method; COD

Introduction

The final processing site is where waste goes after it has been created, collected, handled, and disposed of to the point where it has reached the end of the waste management process. One of the last locations for the open-dumping of waste that the Bandar Lampung City Government oversees is Bakung TPA. At this site, waste is dumped and left to decompose in the open air. The amount of rubbish piled up in Bandar Lampung itself is increasing daily, now totaling 750–800 tonnes per year or around 292,000 tonnes total (Phelia & Damanhuri, 2019). The primary issue will arise from the ever-growing volume of trash, specifically from enormous garbage at the final waste processing site (TPA), which may produce leachate and contaminate the surrounding environment (Rahmayanti et al., 2022). The resultant leachate is often brown, smells terrible, contains many organic materials, and is primarily poisonous (Prameswari et al., 2020). This is because leachate typically contains a wide range of contaminants, including heavy metals, and extremely high BOD and COD concentrations (Efendi et al., 2023; Pratama & Nufutomo, 2021).

Natural organic matter makes up the majority of the leachate; According to Singa et al. (2018), NOM is a mixture of dissolved organic and inorganic components and particulate debris. High and low molecular components comprise natural organic matter (NOM). Leachate has significant color qualities due to humic acid. The leachate may have been hazardous to the locals' health because it contained significant inorganic pollutants and heavy metals. The water has a brown and rusty odor, making it unfit for washing because it would turn the clothing's color dull and leave brown stains on them (Nufutomo, 2021). The fact that locals still rely on healthy water for their everyday needs makes this a significant concern.

In particular, leachate treatment has been the subject of several wastewater treatment methods designed to address issues with wastewater (Adriansyah et al., 2019). Aerating wastewater with microbubbles to break down Fe levels (Efendi et al., 2023) and waste processing can be carried out with adsorbents derived from natural resources (Herlambang et al., 2023). On the other hand, processing wastewater using aeration will reduce phosphate compounds (Fadilah et al., 2023). However, physical technology has been around for a long time, and this technique frequently has extremely high operational expenses related to chemical use, installation, and land use. Therefore, chemical methods of treating wastewater, such as the Advanced Oxidation Process (AOPs) technology, are comparatively more efficient than physical and biological methods (Deng & Zhao, 2015). AOPs are chemical wastewater treatment methods that decompose organic matter using hydroxyl radicals, proving highly effective in treating stubborn organic pollutants. AOPs generally refer to a set of chemical wastewater treatment procedures intended to break down organic materials in wastewater by reacting with hydroxyl radicals in an oxidation process (Deng & Zhao, 2015). When handling resistant organic contaminants in wastewater, AOPs are successful.

One advancement in AOP technology is the Fenton approach. In the Fenton process, Fenton's reagent serves as a source of hydroxyl radicals. It is a solution of hydrogen peroxide (H_2O_2) and iron [II] or iron [III] ($FeSO_4$) salt catalyst at an acidic pH (Leszczyński, 2018). In the Fenton process, iron ions (Fe^{2+}) function as a catalyst in a reaction with hydrogen peroxide (H_2O_2), an oxidant, to produce a radical hydroxy group that can oxidize both organic and inorganic substances (Wang et al., 2021). The Fenton technique is the most efficient AOP technology since it can save energy and space, is inexpensive, simple to use, safe, and has a quick and efficient processing time. Oxidation can also break down hazardous, non-biodegradable substances in wastewater (Efendi et al., 2023; Rezagama et al.). To achieve environmental quality standards, the purpose of this research is to treat Bakung landfill leachate using the Fenton technique and ascertain the impact of the Fenton molar ratio (1:150, 1:200, 1:250) and reaction duration (0, 30, 60, 90, 120 min) on color degradation and COD.

Materials and method

Equipment and apparatus

The equipment that was used included a stirred reactor, glass beaker, measuring cup, analytical balance (OHAUS, PA 224), Erlenmeyer (Pyrex), pH meter (Hanna, HI 11310), filter paper (Whatman, 40nm), and UV-VIS spectrophotometer (Aelab, Ae-S60-2U). All chemicals used are pro-analysis. The apparatus was distilled water, H_2O_2 (30% w/v) (Merck), $FeSO_4.7H_2O$ (Merck), $Na_2S_2O_3.5H_2O$ (Merck), H_2SO_4 (95-97% w/v) (Merck), and leachate collected from the Bakung landfill in West Betung Bay, Bandar Lampung City, Lampung, Indonesia.

Preparation of leachate sample

Leachate sampling uses a grab sample, where samples are taken directly from the initial pond from the final processing site in West Betung Bay, Bandar Lampung City, Lampung. Leachate sampling was carried out at three points. This sample only describes the characteristics at the time of sampling, so the number of leachate samples taken was 10 liters using a plastic ladle with a stem and put into a sterilized sample bottle.

Leachate processing using the Fenton method

Leachate from the Bakung landfill has been taken from the inlet channel and applied to the oxidation process technology. A total of 500 ml was put into the continuously stirred tank reactor (CSTR) and first analyzed for its COD value (MU.SS-UJI.90) and color (using a spectrophotometer UV Vis). Before adding the Fenton reagent, the pH of the initial leachate sample was adjusted to 3-5 by adding H_2SO_4. The optimum condition for the oxidation reaction with the Fenton reagent occurs in an acidic environment, specifically at a pH of 3-5, temperature of 30°C, and atmospheric pressure. Fenton reagent is added to the reactor with various ratios between $FeSO_4$ and H_2O_2 of 1:150 (4 mM $FeSO_4$: 600 mM H_2O_2), 1:200 (4 mM $FeSO_4$: 800 mM H_2O_2), and 1:250 (4 mM $FeSO_4$: 1000 mM H_2O_2) while adjusting the stirrer speed to 250 rpm. The reaction time of 0 min started once the Fenton reagent was added to the reactor. During the oxidation process, samples were taken at 0, 30, 60, 90, and 120 min reaction times. During the sampling interval, 0.5 mL of $Na_2S_2O_3$ solution was added to each sample for COD and color analysis using a UV-Vis spectrophotometer at 374 nm was calculated for equations 1 and 2, respectively. Before being discharged into the river body, the treated sample was neutralized to a pH of 7-9 by adding Na_2CO_3, as shown in Figure 1.

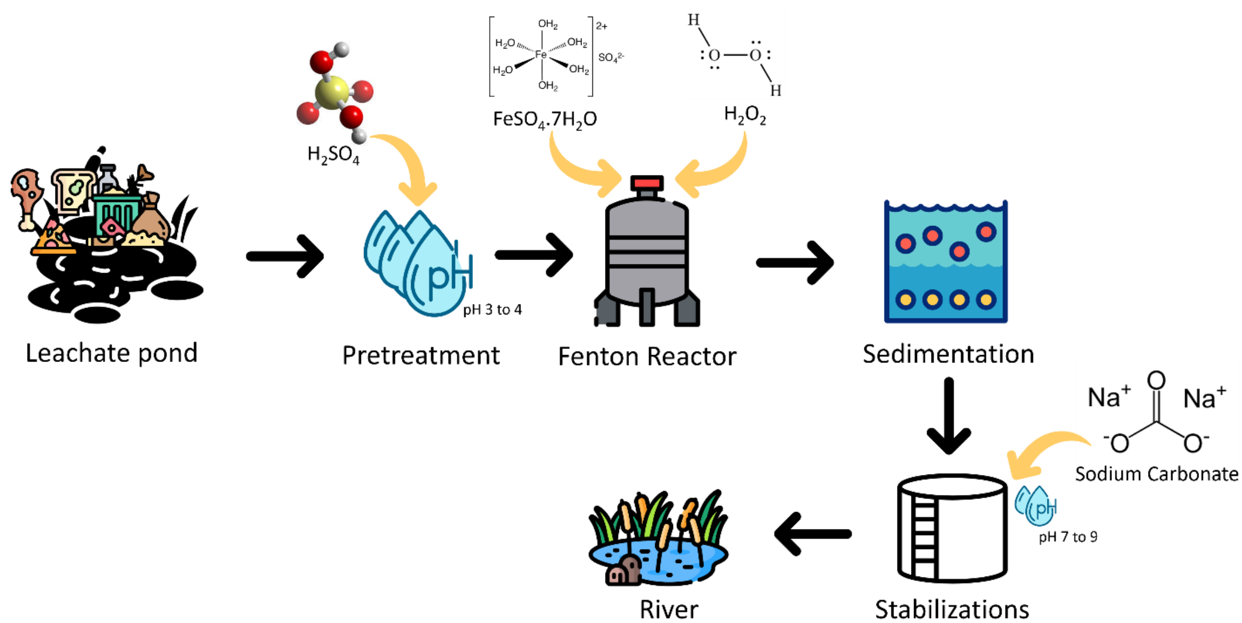

Figure 1. Process flow of Bakung landfill leachate treatment with the Fenton method

Assessment of COD and color degradation

For the COD analysis, a linear relationship was observed between the reaction time and the absorbance of the samples.

$$COD\ degradation\ (\%) = \frac{Initial\ COD - Final\ COD}{Initial\ COD} \times 100\% \tag{1}$$

$$Color\ degradation\ (\%) = \frac{Initial\ absorbance - Final\ absorbance}{Initial\ absorbance} \times 100\% \tag{2}$$

The initial COD and absorbance values are obtained from the initial sample analysis before treatment. The final COD and absorbance values are obtained from the analysis results after the sample is treated with the Fenton process

Statistical analysis

The statistical analysis in this research was carried out using one-way ANOVA with a two-tailed t-test (with significance $p < 0.05$) using IBM SPSS statistics version 26.

Results and discussion

Advanced oxidation process technology for wastewater treatment

Advanced oxidation process technology for wastewater treatment is increasingly important for removing emerging contaminants. These processes involve the generation of highly reactive hydroxyl radicals to break down organic pollutants into harmless byproducts. This technology is particularly effective in treating pharmaceuticals, personal care products, and pesticides not easily removed by conventional treatment methods. Advanced oxidation process technology is also advantageous because it does not produce harmful byproducts or sludge, making it a more environmentally friendly option. Additionally, these processes can be tailored to target specific contaminants, providing a versatile solution for various types of wastewater treatment needs. Overall, advanced oxidation process technology offers a sustainable and efficient approach to wastewater treatment, potentially significantly reducing the presence of harmful contaminants in water sources (Giwa et al., 2021). Its ability to target specific pollutants makes it a valuable tool for industries looking to improve their environmental impact and comply with regulations. By utilizing advanced oxidation processes, industries can meet regulatory requirements and contribute to preserving water quality for future generations (Babuponnusami et al., 2023; Miklos et al., 2018). This technology could revolutionize wastewater treatment practices and create a more sustainable approach to environmental stewardship.

Furthermore, implementing advanced oxidation processes can lead to cost savings for industries by reducing the need for expensive filtration systems. This innovative technology offers a promising solution for addressing water pollution and promoting a cleaner, healthier environment. By incorporating advanced oxidation processes into their operations, industries can significantly reduce their environmental footprint and improve their overall sustainability. This proactive approach to wastewater treatment benefits the environment and enhances the reputation of companies as responsible corporate citizens. One of the methods of advanced oxidation processes technology used in this study is the Fenton method. Fenton is a reagent consisting of a mixture of solutions of hydrogen peroxide (H_2O_2) and iron (II) sulfate ($FeSO_4$), which has a high oxidizing ability in oxidizing contaminants or wastewater. Oxidation with Fenton reagent is an oxidation method using hydrogen peroxide (H_2O_2) as an oxidizer and iron as a catalyst. This oxidation reaction is a complex reaction involving the decomposition of H_2O_2 with the help of the Fe^{2+} catalyst. The reaction mechanism begins with Fe^{2+} initiating the reaction and catalyzing the decomposition reaction of H_2O_2 so that hydroxyl radicals ($OH^\bullet$) are produced.

Characteristic of Bakung landfill leachate

Testing of the leachate's initial characteristics is conducted to ascertain the parameters of leachate contamination by leachate quality criteria for enterprises and/or activities at final waste processing sites. The testing includes analysis for heavy metals, organic compounds, and other pollutants. While the COD value is one of the first features seen in leachate samples, The COD concentration is still very high, at 1,715 mg/L, based on the results of the leachate's initial characteristics test, even though the quality standard is only 300 mg/L. Because leachate contains complex organic compounds, a high COD value in the Bakung landfill leachate samples suggests that a significant amount of oxygen is needed for the organic components to be chemically oxidized (Phelia & Damanhuri, 2019). The leachate's dark brown color can identify this.

Another characteristic of leachate is the color, which is also important (i.e., dark brown, black, and cloudy). The presence of contaminants, such as humus, lignin, tannin, and other organic acids, is typically indicated by the dark color of the water. In addition, water can also be colored by sand, soil, and microorganisms like moss and algae (Pratama & Nufutomo, 2021). Although the color concentration value is not specified in the quality criteria, this is a unique issue since colors that contaminate water bodies might hinder photosynthesis by preventing sunlight from reaching the right intensity. The absorbance value can be used to calculate the color concentration value; the Bakung landfill leachate's initial absorbance is 2.12. Variations in absorbance values will reveal variations in the leachate's color concentration (Figure 2).

Figure 2. Leachate samples before and after processing using the Fenton method at various time variations (a) 0 min, (b) 30 min, (c) 60 min, (d) 90 min, and (e) 120 min

Characteristic of color degradation after advanced oxidation processes technology Bakung landfill leachate with Fenton method

Color degradation and COD characteristics in the Bakung landfill leachate are shown in Figures 3a and b, respectively. Figure 3a shows that the color degradation process is relatively steady, with noticeable color loss occurring at nearly all Fenton molar ratios between 30 and 120 min. At a 1:150 Fenton molar ratio and a 120-minute reaction duration, the most significant percentage of color degradation (91.4%) happens. Nevertheless, the percentage of color loss dropped at Fenton molar ratios of 1:250 and 1:200. This is due to an excessive $FeSO_4$ to H_2O_2 ratio, where an excessive amount of H_2O_2 is consumed during the color degradation process, which reduces the efficiency of the degradation process. The more hydroxyl radicals ($OH^{\bullet}$), the more dyes are degraded. This is because H_2O_2, in low concentrations, enhances dye degradation due to more efficient hydroxyl radical formation. The hydroxyl radicals ($OH^{\bullet}$) formed will break double bonds in leachate water into simpler compounds, so color degradation will occur in leachate water, which is initially brown to clear. H_2O_2 will initially make color degradation more effective, but too much will not make it more efficient. Efficiency will somewhat decline. As such, studies including adding H_2O_2 at high ratios have shown that this no longer enhances color degradation efficiency. Adding $FeSO_4$ and H_2O_2 will increase the ferric hydroxide precipitate created in the Fenton process, increasing the concentration value

(Gautam et al., 2019). Reduced color degradation efficiency results from this disruption of hydroxyl radical production.

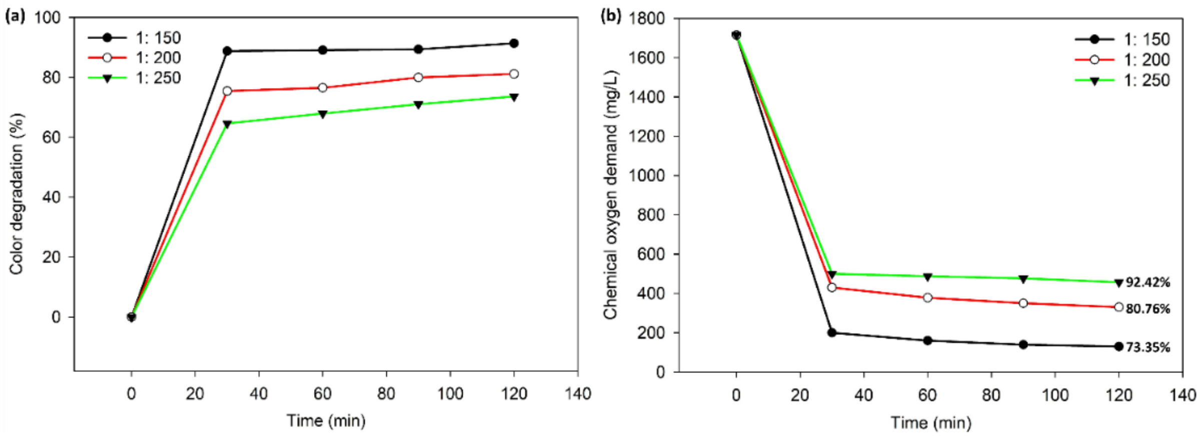

Figure 3. Effect of the Fenton method based on (a) color degradation, and (b) COD in the Bakung landfill leachate

Reaction duration impacts color degradation and the addition of Fenton's molar ratio. At a 1:150 Fenton molar ratio and a reaction duration of 120 min, Figure 3 illustrates the highest color deterioration that was attained. However, overall color degradation happens between 30 and 120 min into the reaction, meaning that the longer the reaction period, the more color degradation results. Oxidation time will affect the color removal efficiency. The Fenton process shows that the longer the reaction time, the higher the efficiency. So, the more significant the percentage of color degradation achieved. Overall color degradation occurred at various reaction times; the maximum results occurred at 120 minutes. It occurs due to the forming of more hydroxyl radicals with longer reaction times (Mahtab et al., 2021).

In this instance, the leachate's hue results from organic chemicals or contaminants degrading more readily with time. The findings of this study were consistent with those of (Adriansyah et al., 2019), whose research showed that 120 min was the most significant amount of degradation. The longer the reaction time, the more hydroxyl radicals are formed because the increase in time will provide more opportunities for the degradation of organic compounds or pollutants, which is the color of the leachate water. The percentage of color deterioration increases with longer reaction times; the higher the Fenton molar ratio, the smaller the percentage of color degradation. The relationship between Fenton's molar ratio and reaction time determines the maximum percentage of color degradation: the higher the percentage of color degradation produced, the lower the Fenton's molar ratio and the longer the reaction time.

Effect of advanced oxidation processes technology Bakung landfill leachate base on chemical oxygen demand (COD)

Figure 3b illustrates that the highest percentage of color deterioration, reaching 92.4%, happens at a 1:150 Fenton molar ratio after 120 min of the reaction period. Meanwhile, the percentage of COD degradation produced fell at Fenton molar ratios of 1:250 and 1:200. This indicates that the percentage of COD degradation produced decreases with increasing Fenton molar ratio. Because of the phenomena of $OH^{\bullet}$ reacting with H_2O_2 (Wang et al., 2021), which results in the generation of HO_2 radicals, which are less reactive than $OH^{\bullet}$ radicals, as shown in Equation 3 and 4 (Deng & Zhao, 2015), this is induced by the addition of excess H_2O_2. This will minimize the percentage of degradation.

$$OH^{\bullet} + H_2O_2 \longrightarrow H_2O + HO_2 \quad (3)$$
$$OH^{\bullet} + HO_2 \longrightarrow H_2O + O_2 \quad (4)$$

The efficacy of COD degradation by the Fenton technique affects the concentration of H_2O_2. Because too much H_2O_2 can diminish hydroxyl radicals, the amount of H_2O_2 added must also be carefully considered (Costa et al., 2019). In addition, because it will raise waste processing's operating expenses, it will affect the industry's economic worth. However, a deficiency in H_2O_2 can also result in a deficiency in hydroxyl radicals, which means that not enough $OH^{\bullet}$ is present to complete the Fenton process (Hakika et al., 2019). Figure 2b demonstrates that even after a 30-min reaction time, the COD degradation was only marginal and did not reach the required quality criteria. This is because there were not enough hydroxyl radicals produced to completely oxidize all of the organic components in the leachate. Nevertheless, COD degradation started to happen at 60 and 90-min response periods, and it increased dramatically until 120-min reaction time. It is because there were not enough hydroxyl radicals to completely oxidize all of the organic components in the leachate.

Nevertheless, COD degradation started at 60 and 90-min response periods, increasing dramatically until 120-min reaction time. It demonstrates that a higher percentage of COD degradation is created the longer the reaction time. More hydroxyl radicals are produced due to the interaction between $FeSO_4$ and H_2O_2, which is more effective with longer reaction times. The oxidation process produces hydroxyl radicals that break double bonds in organic compounds, breaking complex compounds in the leachate into simpler ones and forming CO_2 and H_2O (Xu et al., 2020). The majority of the organic compound content in leachate has been reduced, as indicated by the declining COD values, indicating a decrease in the quantity of oxygen required for the chemical breakdown of organic compounds (Singa et al., 2018).

The advanced oxidation processes technology efficacy in handling the leachate from Bakung landfills

Table 1 presents the findings of an analysis of variance (ANOVA) study that looked at how reaction time and Fenton's molar ratio affected color deterioration and chemical oxygen demand (COD).

Table 1. Analysis of variance (ANOVA) ratio mol and time based on color degradation and chemical oxygen demand (COD)

Source	Type III Sum of Squares	df	Mean square	F	Sig.	Information
Color degradation						
Corrected model	22.795a	11	2.072	216.233	0.00	Significant
Intercept	196584.1	1	196584.100	20513123.5	0.00	Significant
Ratio mol	21.381	2	10.690	1115.522	0.00	Significant
Time	1.065	3	0.355	37.029	0.00	Significant
Ratio mol* time	0.349	6	0.058	6.072	0.004	Significant
Total	196607.010	24				
Chemical oxygen demand (COD)						
Corrected model	878.27a	11	79.84	3484	0.00	Significant
Intercept	184643.5	1	184643.5	8057174.5	0.00	Significant
Ratio mol	7.210	2	3.605	157.3	0.00	Significant
Time	866.0	3	288.66	12596.479	0.00	Significant
Ratio mol* time	5.053	6	0.842	36.752	0.00	Significant
Total	185522.130	24				

A significance level of 0.000 < 0.05 was assigned to the data. It is evident from a significant number that Fenton's molar ratio and time impact color deterioration and COD. The interaction between the ratio and time obtained significant data, indicating a real influence on the interaction of the ratio and reaction time used on color degradation and COD. These results suggest that both Fenton's molar ratio and reaction time are crucial in determining the extent of color deterioration and COD levels. Further research could explore the specific mechanisms through which these factors interact to influence the outcomes observed in this study. This study explored the effective Fenton molar ratio for COD degradation, such as a ratio of 1:150, and the most effective reaction time was 120 min.

Conclusion

The percentage of color degradation and COD can be affected by Fenton's molar ratio and reaction time. The proportion of color and COD degradation produced will increase with a smaller Fenton molar ratio and a more extended reaction period to achieve the highest degradation percentage. At a Fenton molar ratio of 1:150 and a reaction time of 120 min, ideal conditions were reached. The highest percentages of color degradation and COD generated were 91.4% and 92.4%, respectively. Because the Fenton approach may degrade color and COD and has satisfied leachate quality standards for businesses and activities at final waste processing facilities, the research results demonstrate its effectiveness in treating leachate.

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Conflicts of interest

The authors declare that there are no conflicts of interest.

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