

Enhancing Domestic Wastewater Treatment: A Study on the Efficacy of Tiered Filtration and a Combined Photocatalytic Ozonation Methods

Mega Gemala¹, Roni Saputra^{2*}, Annisa Fyona³, Ita Wijayanti⁴, Fedia Restu⁵, Ice Irawati⁶, Adhe Aryswan⁷, Hengky Oktarizal⁸

¹Metallurgy Engineering Departement, Politeknik Negeri Batam, Kota Batam, Indonesia

^{2,8}Environmental Health Department, Universitas Ibnu Sina, Kota Batam, Indonesia

^{3,4,5,7}Mechanical Engineering Departement, Politeknik Negeri Batam, Kota Batam, Indonesia

⁶Occupational Health and Safety Departement, Universitas Ibnu Sina, Kota Batam, Indonesia

ABSTRACT: This study aimed to construct an optimized reactor using photocatalytic ozonation, and its effectiveness was determined using PermenLHK No. 86/2016. The Tanjung Uma housing estate in Batam city was selected as the study sample site for domestic wastewater because of its role as a significant water body that receives liquid waste from a majority of activities across the city. The experiment employed a quantitative research approach and a true experimental approach, testing variables (catalyst weight, UV irradiation time, and ozonation duration) in a laboratory setting (25°C and 40-70% Humidity). The study found that the optimized parameter (a combination of 2 h of irradiation, with 2 g of TiO₂ along with 5 min of ozonation) resulted in a significant difference in water quality parameters compared to before treatment with 92% effectivity. It can be concluded that this Reactor prototype was highly successful in reducing environmental impacts based on its effectiveness.

KEYWORDS: Domestic wastewater, Photocatalytic, Ozonolysis, Reactor, Environment

I. INTRODUCTION

Industrial wastewater is a complex and pressing environmental problem in the modern industrial era (Berry & Rondinelli, 1998). Every year, industries produce large volumes of wastewater containing hazardous substances and pollutants (. Ahmed et al., 2021). Inadequate wastewater treatment can negatively impact water ecosystems, human health, and environmental sustainability (Saravanan et al., 2021). The main problem in industrial wastewater treatment is the presence of organic compounds that are difficult to degrade and toxic substances such as heavy metals, oil, detergents, and other hazardous chemicals (Rasheed et al., 2020). Conventional methods such as physicochemical and biological treatments may not be able to effectively remove pollutants (S. F. Ahmed et al., 2021). Therefore, it is important to develop new approaches that are more effective and efficient for industrial wastewater treatment (Saravanan et al. 2021). One promising approach is the combination of ozonation and photocatalysis, which has been proven to be effective in degrading organic pollutants and eliminating pathogenic microorganisms in water (Gomes et al., 2019).

Recent research by Malik et al. (2020) has emphasized the importance of developing more advanced ozonation and photocatalysis methods for industrial wastewater treatment. They highlighted the need to pay attention to the reaction mechanisms, influencing factors, and future perspectives of the process. Other studies have also

been conducted in this field, such as (Agustina et al. (2005), Mecha and Chollom (2020), Mehrjouei et al. (2015), Rekhate and Srivastava (2020a), and Xiao et al. (2015, 2020). These studies discussed the latest advances in ozonation and photocatalysis for wastewater treatment, including the reaction mechanisms and factors that affect processing efficiency. However, there is still a research gap in determining the optimal parameters, such as the effect of catalyst concentration, ozone contact time, and ozone concentration on the pH value, COD, BOD, TSS, oil and fat, heavy metals (lead), and turbidity levels in industrial wastewater.

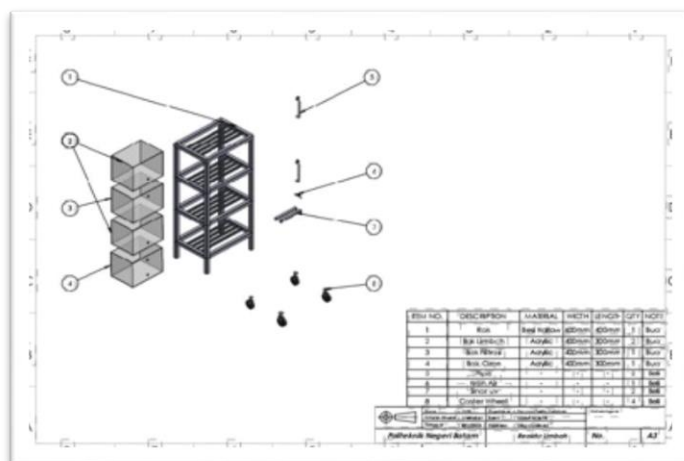


Figure 1. Design of the photocatalytic ozonation reactor

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This article aims to address this research gap by determining the optimal parameters using a laboratory-scale ozonation photocatalytic reactor to optimize the interaction between ozone, photocatalyst, and wastewater. This research will focus on determining the optimal parameters to achieve high degradation levels and efficient pollutant removal using photocatalytic and ozonolysis methods. This study is in line with the Center of Excellence (CoE) research roadmap in the field of industrial wastewater treatment, which encourages the development of innovative technologies that are more effective in industrial wastewater treatment. The results of this study are expected to contribute to solving industrial wastewater problems and support sustainable environmental management.

METHOD

The research method used in this study is divided into several stages.

A preliminary Analysis was conducted before any treatment was applied, and an initial analysis was conducted to determine the baseline values of the parameters. This served as a comparison of the samples that had undergone the treatment. The initial parameters analyzed included pH, COD, BOD, TSS, turbidity, oil, and total coliforms.

The Reactor Design Stage began with the preparation of a Photocatalytic Ozonation Reactor framework. The fabrication process involves machining through cutting and the use of machine tools. The stages in the Reactor Fabrication process included the following: a. A 3D design was created for the photocatalytic ozonation reactor (Figure 1). The main rack material was cut using galvanized hollow iron.

The experiment was conducted in collaboration with research partners including lecturers and partner students. Partner students collected samples, and experiments were conducted with partner lecturers. The materials used in this stage were wastewater samples (20-35 L), a TiO₂ catalyst, zeolite, activated charcoal, gravel, and cheesecloth.

This study uses a quantitative approach and employs a true experimental approach. The quantitative research was based on scientific rules, and this research aimed to determine the pH, COD, TSS, turbidity, oil, and total coliforms in domestic wastewater before and after treatment with a combination of photocatalytic and ozonolysis methods. The true experimental approach involved the use of a laboratory scale to test engineered variables and observe the influence between two variables using a control variable.

RESULT AND DISCUSSION

Result

Based on the test parameters, we can compare the results of the laboratory examination with the maximum standards according to the Regulation of the Minister of

Environment and Forestry No. P68/Menlhk-Setjen/2016 as follows. Table 1. The results showed that the pH test parameter before treatment was 5.2, while the maximum standard was 6-9. Therefore, this falls under the category of not meeting the criteria. The laboratory results for the COD test parameter before treatment were 296.84 mg/L, and the quality standard was 100 mg/L; thus, this fell into the category of not meeting the criteria. The laboratory results for the ammonia test parameters before treatment were 2.94 mg/L, while the maximum standard is 10 mg/L, respectively. Therefore, this falls into the category of meeting criteria. However, the total coliform parameter is far above the quality standard; thus, it does not meet the criteria for clean water.

Table 1. Sample Test Before Treatment

No	Test Parameter	Result	*Standard	Criteria
1	pH	5,2	6 – 9	Not Passed
2	TSS	254	25 NTU	Not Passed
3	COD	296,84 mg/L	100 mg/L	Passed
5	Oil-Fat	70 mg/l	5	Not Passed
4	Amoniak	2,94 mg/L	10 mg/L	Passed
5	Total Coliform	17.000	50 MPN/100 ml	Not Passed

It can be inferred that the domestic wastewater sample did not meet the standard criteria for several parameters prior to treatment. Specifically, pH, Total Suspended Solids (TSS), oil and fat content, and Total Coliform count did not meet the standard criteria. However, the Chemical Oxygen Demand (COD) and ammonia content were within the acceptable ranges. From the physical examination of domestic wastewater before treatment, it was observed (Figure 2) that the water had an unpleasant odor and a murky blackish color.



Figure 2. Sample Physical Examination

Based on Table 2, the parameters of pH, COD, TSS, turbidity, oil and fat, and Total coliform in domestic wastewater from the Tanjung Uma residential area, Batam city, exceed the quality standards set by the Regulation of the Minister of Environment and Forestry of the Republic of

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Indonesia Number: P68/Menlhk-Setjen/2016 Regarding Domestic Wastewater

Quality Standards and the Regulation of the Minister of Health Environment and Water Health Requirements for Hygienic Sanitation, swimming pools, and public baths.

Pre-treatment of the wastewater sample was conducted using a multi-stage filtration unit with an upflow system designed using an acrylic reactor measuring 40 cm x 30 cm x 31 cm. Inside the reactor, a multi-stage filtration media was used, arranged in the following sequence: zeolite sand with a thickness of 15 cm, charcoal with a thickness of 15 cm, and gravel with a thickness of 15 cm (Figure 3). Pre-treatment was carried out to remove interfering particles that could hinder the photocatalytic process, such as moss, fungi, plankton, and other contaminants with relatively large particles. The results of the wastewater filtration from the pre-treatment are shown in Figure 3.



Figure 3. Wastewater Pretreatment Reactor

Table 2. Test Result Of TiO2 Photocatalization with variations (UV Exposure & Catalyst Weight)

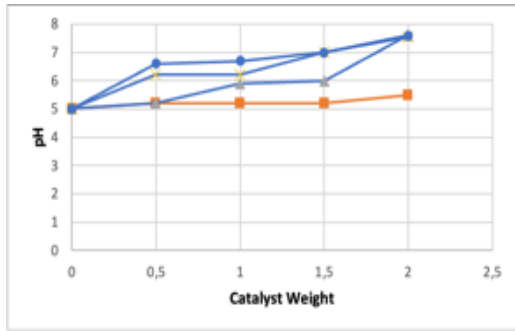
Experiment Parameters		Test Results					
UV Exposure Duration (Hours)	Catalyst Weight (grams)	pH	TSS	CO D (mg/l)	oils and fats (mg/l)	ammonia (mg/l)	Total e-coli (NTU)
1	0	5	250	286	67	2,90	17.000
	0,5	5,2	186	195,6	67	2,90	15.000
	1	5,2	180	182,4	60	1,87	14.200

	1,5	5,2	168	166,6	56	1,81	11.200
	2	5,5	162	150,8	50	1,80	9800
2	0	5	146	278	62	2,90	17.000
	0,5	5,2	127	140	47	1,85	7010
	1	5,9	100	100	44	1,80	6700
	1,5	6,0	60	55	40	1,78	3500
	2	7,6	59	51,7	39	1,77	2800
3	0	5	153	250	39	2,88	17.000
	0,5	6,2	110	165	39	1,85	2500
	1	6,2	93	94	34	1,80	1300
	1,5	7	60	54	33	1,79	950
	2	7,5	58	52	30	1,79	520
4	0	5	140	240	30	2,89	17.000
	0,5	6,6	100	156	27	1,65	1200
	1	6,7	89	94	27	1,64	560
	1,5	7	59	52	26	1,64	400
	2	7,6	59	51	20	1,62	320

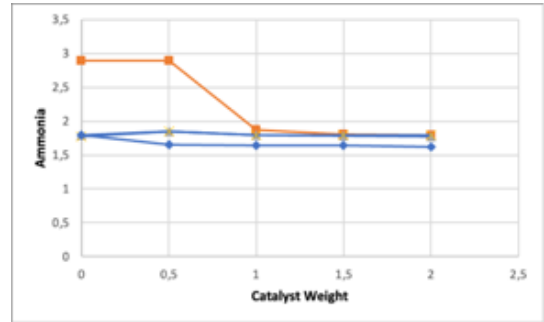
The experimental results show that the use of the TiO2 Photocatalyst method, preceded by a Pre Treatment using a tiered filter, successfully degrades and reduces pollutant values in domestic wastewater. As seen in Table 4.4, the pH value gradually increases with the addition of the catalyst amount and the contact time of the photocatalyst on a TiO2 catalyst mass of 1.5 grams and a contact time of 3 hours using a single UV lamp. Similarly, the values of TSS,

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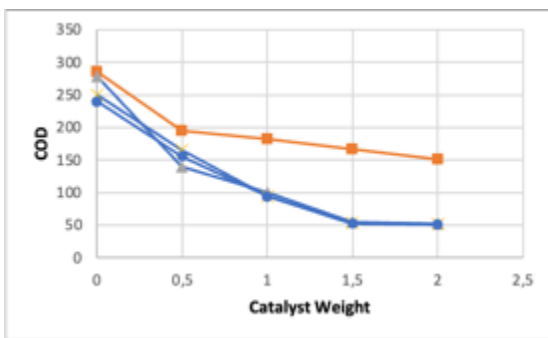
COD, Oil and Fat, and Total Coliform continue to decrease, approaching 80%.



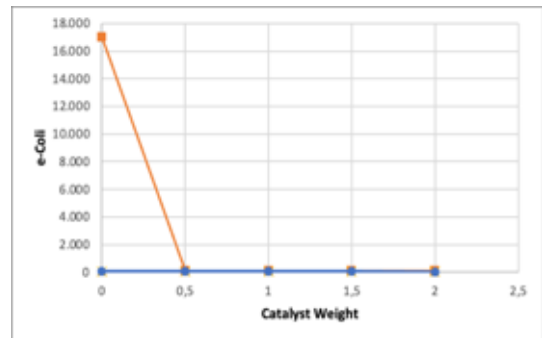
(a)



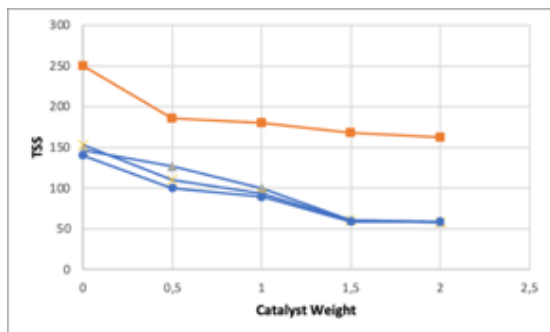
(e)



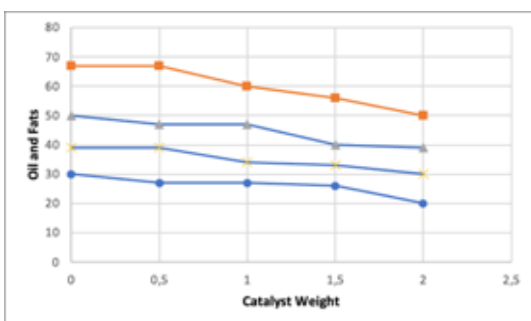
(b)



(f)



(c)



(d)

Figure 4. Catalyst Weight vs pH (a) , COD (b), TSS (c), Oil & Fats (d), Ammonia (e), and e-Coli (f) with Exposure Duration at 1 hour, 2 hours, 3 hours, 4 hours

The pH changes that affected the physical, chemical, and biological processes of living organisms in it (Maulidia et al., 2023). The pH value of domestic waste before the photocatalyst was 5.2. The research data for the pH parameter could be seen in Figure 4 (a), where the pH value showed a significant change when the contact time reached 3 hours and the mass of catalyst used was 1.5 grams. Figure 4 (b) showed that there was an optimal condition where the Chemical Oxygen Demand (COD) value decreased significantly. This condition was achieved by using a contact time of 3 hours and a TiO₂ catalyst mass of 1.5 grams, which resulted in an effectiveness of 82% in reducing COD. These results indicated that photocatalytic reactions with TiO₂ were very effective in oxidizing organic compounds that caused high COD in water or waste.

A significant reduction in Total Suspended Solid (TSS) concentration was shown in Figure 4 (c) where when the contact time reached 3 hours and the mass of catalyst used was 1.5 grams, the effectiveness value was up to 76.4%. This result was consistent with the literature which stated that photocatalysis using TiO₂ could be used to reduce TSS concentration in water (Chong et al., 2010; Khan et al., 2021). Figure 4 (d) showed that in the photocatalytic method using

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TiO₂ as a catalyst, the reduction of oil and grease parameters in domestic wastewater reached the optimal point with a contact time of 2 hours and a catalyst mass of 1.5 grams. The effectiveness of oil and fat reduction under these conditions reached about 43%. This effectiveness showed that the photocatalytic reaction was able to reduce the concentration of oil and fat in the effluent, but was not yet optimal.

The value of ammonia parameters before treatment had met the requirements or applicable quality standards. After treatment using the photocatalyst method, the reduction of ammonia parameters in wastewater reached the optimal level as shown in Figure 4 (e) with an exposure duration of 1 hour and a catalyst mass of 1.5 grams. The effectiveness of ammonia reduction in this condition reached 39%. This effectiveness showed that the photocatalytic reaction was able to reduce ammonia concentration in wastewater, although it was not optimal. Figure 4 (f) showed that in the photocatalysis method with the use of TiO₂ catalyst, the reduction of Total Coliform parameters in wastewater reached the optimal level at a contact time of 3 hours and with a catalyst mass of 1 gram. The effectiveness of reducing total coliform under these conditions reached a high level, which was around 92%. This high effectiveness showed that photocatalysis with TiO₂ was a very effective method in reducing the amount of total Coliform in wastewater, which was an indicator of water hygiene and water quality.

Table 2. Test Results of Combined Photocatalytic Ozonation Treatment

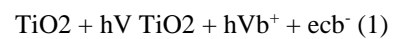
Exposure Duration (Minutes)	Test Parameter					
	pH	TSS	COD	Oil and Fats	Ammonia	Total Coliform
0	5,8	75	95	25	1,5	450
25	6,0	50	88	10	1,5	100
35	6,5	23	60	4	1,2	50
45	6,9	27	55	5	1,0	45
55	7,5	30	52	5	1,0	42

Table 3 showed that in the combination of the Ozonolysis method with photolysis using a TiO₂ catalyst of 1.5 grams and a contact time of 3 hours (optimal parameters in the previous treatment), the reduction of all pollutant parameters in wastewater reached the optimal level at an exposure duration of 35 minutes with an effectiveness value of about 94%. The addition of a longer exposure time (up to 55

minutes) did not show a significant decrease (tended to stagnate) in the test parameters.

DISCUSSION

The photocatalytic method used in this study involved variations in catalyst mass and contact time, which influenced the degradation of parameters such as pH, Chemical Oxygen Demand (COD), Total Suspended Solids (TSS), oil and fat, ammonia, and Total coliform. The following mechanism describes the photocatalytic process of the TiO₂ semiconductor through the subsequent reaction (Cheng et al., 2020) :



This resulted in when TiO₂ was exposed to UV light that matched or exceeded the band gap energy in the titanium oxide, the electrons were excited from the valence band to the conduction band (producing ecb⁻), causing vacancies or holes (hvb⁺) in the valence band that could act as positive charges (Guo et al., 2019). The hole (h+vb) reacted with the metal hydroxide, in this case titanium oxide hydroxide, to form metal hydroxide radicals.

These radicals acted as strong oxidizers capable of oxidizing organic compounds in domestic wastewater. This process occurred in the context of TiO₂ semiconductor photocatalysis with the assistance of UV light. When TiO₂ was exposed to UV radiation, electron excitation occurred from the valence band to the conduction band, producing electrons and holes (h+) (S. Chen et al., 2018). These holes then reacted with titanium oxide hydroxide to form metal hydroxide radicals. Electrons on the semiconductor surface were trapped in the metal hydroxide and could react with electron acceptors in the solution, such as H₂O or O₂, forming hydroxyl radicals (.OH). These hydroxyl radicals acted as strong oxidizing agents capable of degrading organic compounds in domestic wastewater. This process was effective in oxidizing organic compounds and reducing pollution in domestic wastewater (Xia et al., 2020). In addition, this process was also utilized for the degradation of color substances in waste, with the degradation product molecules being adsorbed and re-oxidized by hydroxyl radicals until the radiation process concluded.

The degree of acidity (pH) is a measure used to determine the acidic and basic properties of a solution. In the context of photocatalysis, significant pH changes are a result of the formation of highly reactive hydroxyl radicals (OH*) during the photocatalytic reaction. An increase in the concentration of H⁺ ions in the solution leads to a decrease in pH, which is consistent with previous research findings (Ding et al., 2019; Hasani et al., 2019). However, the change in pH becomes less significant with an increase in contact time and the amount of TiO₂ catalyst mass. This is due to several

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factors, including the increase in the number of hydroxyl radicals that no longer significantly affect pH and the influence of buffer compounds in the solution that can stabilize pH (Alkaykh et al., 2020a; Jung et al., 2009).

In the context of photocatalysis, the formation of OH radicals is believed to be a key step in photocatalytic oxidation in water. The pH dependence of OH radical formation has been studied, and it has been found that the current efficiencies of OH radical formation are smaller in alkaline solutions due to the deprotonation of the reaction intermediate (Bahnemann et al., 2018; Liu et al., 2020; Rao et al., 2022). However, the change in pH becomes less significant with an increase in contact time and the amount of TiO₂ catalyst mass. This is due to several factors, including the increase in the number of hydroxyl radicals that no longer significantly affect pH and the influence of buffer compounds in the solution that can stabilize pH. In the context of TiO₂ photocatalysis, significant pH changes primarily occur at the initial stage of the reaction, and after reaching a certain point (optimal point), the photocatalytic impact on pH may become less significant (Alkaykh et al., 2020b; Dariani et al., 2016; Wu et al., 2015). This is consistent with the understanding that the rate of a reaction can be influenced by the concentration of the reactants and the amount of catalyst available ((Liu et al., 2020)).

Total Suspended Solids (TSS) were defined as particulates that caused turbidity in water, being insoluble and unable to settle directly (Adjovu et al., 2023). Photocatalysis using TiO₂ was employed to reduce the concentration of TSS in water (Izadpanah et al., 2021). This reduction in TSS occurred due to a photocatalytic reaction triggered by the exposure of TiO₂ to ultraviolet light, which produced highly reactive hydroxyl radicals (OH^{*}). These hydroxyl radicals then interacted with the TSS particles in the water, breaking them down into simpler compounds that were easier to precipitate. The increase in contact time between TiO₂ and TSS resulted in a more significant decrease in TSS concentration. This was in line with the basic principle that the longer the TSS particles were in contact with the UV-activated TiO₂, the more photocatalytic reactions could occur, leading to a larger reduction in TSS concentration (Marín-Caba et al., 2021; Rekhate & Srivastava, 2020b; Sujatha et al., 2020). However, the data indicated that an increase in contact time and the amount of TiO₂ catalyst led to a decrease that was no longer significant. This could have been caused by several factors, including a decrease in the availability of unoxidized TSS particles, a decrease in photocatalysis efficiency over time, and the availability of sediment that had become saturated with TSS in the solution (Baby et al., 2019).

The photocatalytic reaction with TiO₂ was highly effective in oxidizing organic compounds that contribute to

high COD in water or wastewater. The decrease in COD value in TiO₂ photocatalysis occurred as TiO₂ was exposed to ultraviolet light, which activated TiO₂ and led to the formation of hydroxyl radicals (OH^{*}), a potent oxidizing agent. These hydroxyl radicals could oxidize oxygen-containing organic compounds, such as dissolved organic matter in water, which is a major component of COD. However, the change in COD became less significant with increased contact time and mass of TiO₂ catalyst. After reaching an effectiveness level of 82%, further increases in contact time and catalyst mass no longer provided significant improvements in reducing COD parameters. Factors such as the decrease in availability of oxidizable organic compounds, possible saturation of the catalyst surface, or an increase in side reactions may have played a role in this change (D. Chen et al., 2020; Zafar et al., 2021).

The research also indicated that an increase in contact time and the amount of TiO₂ catalyst mass did not significantly enhance the effectiveness of oil and fat reduction. In conditions where contact time was extended or catalyst mass was increased, the rate of effectiveness only slightly improved or even tended to stagnate. Factors such as the saturation of the catalyst surface or an increase in side reactions likely played a role in this change (Gardy et al., 2017; Iida et al., 2021).

The effectiveness of ammonia reduction reached 39%. This indicated that the photocatalytic reaction had the capacity to decrease the concentration of ammonia in liquid waste (Duan et al., 2021). However, it's important to note that this level of reduction was not yet optimal. In other words, there was still room to enhance the effectiveness of this method. Perhaps by adjusting the contact time and catalyst mass, or by modifying other methods, the effectiveness of ammonia reduction could be improved. Overall, these data demonstrated the potential of the photocatalytic method in processing liquid waste, particularly in reducing the concentration of ammonia. Despite this, further research is needed to optimize this method and ensure its effectiveness under various conditions.

The quantity of E. Coli in the samples significantly diminished with the escalation of catalyst weight and duration. At a catalyst weight of 1 gram and a duration of 1 hour, the E. Coli count peaked. However, with the further increase in catalyst weight and duration, the E. Coli count drastically declined.

The results of this study were consistent with other research indicating the effectiveness of photocatalytic methods in reducing contaminant concentrations in water. For example, a study demonstrated that photocatalysis based on titanium dioxide (TiO₂) could decrease the concentration of E. coli bacteria in water by up to 99.9% within a span of 60 minutes (Makropoulou et al., 2018). Other Previous research has indicated that photocatalysis can be employed to decrease the

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concentration of contaminants in water. In one particular study, researchers utilized a TiO₂-based photocatalyst to reduce the concentration of phenol in water. The findings revealed that photocatalysis was capable of reducing the phenol concentration by up to 90% within a 60-minute timeframe (Gul et al., 2023).

The ozonation method employing 1.5 grams of TiO₂ catalyst and a 3-hour contact time successfully reduced all pollutant parameters in liquid waste, achieving an effectiveness level of approximately 94%. However, increasing the contact time and TiO₂ catalyst mass did not result in significant improvements in pollutant reduction

effectiveness. This suggests that there are limitations to enhancing pollutant reduction effectiveness by extending contact time and increasing TiO₂ catalyst mass. Another study found that ozonation with TiO₂ catalysts was effective in reducing organic pollutants in wastewater (Duan et al., 2021). Further research considering variations in experimental conditions, such as temperature, pressure, or the type of catalyst used, could be beneficial. Additionally, expanding the range of measured parameters could provide a more comprehensive understanding of the effectiveness of ozonation with TiO₂ catalysts in reducing pollutants in liquid waste. Comparing ozonation methods with other techniques, such as the use of alternative chemicals or combinations with physicochemical processes, may also offer deeper insights into the effectiveness of liquid waste treatment methods. Overall, these findings indicate that photocatalytic methods can be employed as effective technologies for reducing contaminant concentrations in water. However, additional research is needed to optimize these processes and ensure their effectiveness under various conditions.

CONCLUSIONS

The photocatalytic method employed in this study, involving variations in catalyst mass and contact time, demonstrated significant potential in degrading various parameters such as pH, Chemical Oxygen Demand (COD), Total Suspended Solids (TSS), oil and fat, ammonia, and Total coliform in domestic wastewater. The process, which utilized TiO₂ semiconductor and UV light, was effective in oxidizing organic compounds and reducing pollution. However, the effectiveness of this method plateaued after reaching certain levels of contact time and catalyst mass, suggesting the existence of an optimal point beyond which further increases did not yield significant improvements. This was observed across various parameters including pH, TSS, COD, and oil and fat reduction. The effectiveness of ammonia reduction, while promising, was not yet optimal, indicating room for further enhancement. The study also demonstrated a significant reduction in E. Coli with increased catalyst weight and duration. Despite these promising results,

further research is needed to optimize this method and ensure its effectiveness under various conditions. Future studies could consider variations in experimental conditions, such as temperature and pressure, or the use of alternative catalysts. Comparisons with other techniques, such as the use of alternative chemicals or combinations with physicochemical processes, may also offer deeper insights into the effectiveness of liquid waste treatment methods.

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