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Use of Zinc Oxide (ZnO) for Photodegradation of Fish Frozen Wastewater

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ABSTRACT. Increasing population growth has spawned numerous industries, including the fish industry. Liquid fish frozen waste has the potential to pollute the environment due to its high concentration of organic compounds, such as nitrogen, oil, and fat. Photodegradation is a cost-effective and efficient method for processing liquid waste. Photodegradation is the degradation of a substance by UV light's photon energy. This study aims to determine the variation in mass and contact time, as well as the efficiency of photodegradation using zinc oxide in reducing chemical oxygen demand (COD), total suspended solids (TSS), and pH changes in fish processing wastewater (ZnO). The variables included the ZnO masses (0.59, 0.75, and 1 g) and the contact times (3, 4, and 5 hours). According to the results, mass and contact time impact the efficiency of photodegradation. Photodegradation is 69.69% effective against COD and 81.7% effective against TSS.

1. Introduction

Population growth in Indonesia increases the number of industries, which includes the frozen fish industry. Fishing for frozen food is an industry that uses a great deal of water in its production processes; consequently, it generates enormous quantities of wastewater (Yuliasni et al., 2019; Piri and Mirwan, 2018). The liquid waste consists of fish-washing water that is discharged into local waterways (Widiyanti and Hafidah, 2021) (Kratky and Zamazal, 2020). One of the sources of liquid waste pollution resulting from the processing of fish is discharged directly into the environment, causing an offensive odor in the water and water and soil pollution (Rachmanto and Salamah, 2021). Waste treatment is one of the best ways to ensure the sustainability of fish production and processing waste management (Kurniasih et al., 2018). Due to a number of factors, improperly managed fish waste emits a pungent odor and attracts a large number of flies and mosquitoes (Kusuma and Yulianto, 2019). One way to determine the level of pollution in wastewater is to measure the biological oxygen demand (BOD) and chemical oxygen demand (COD) parameters (Rachmanto and Pebritama, 2021).

The photocatalyst method converts photon energy to chemical energy and generates hydroxyl radicals (OH) that react with organic compounds within pollutants as redox agents. These contaminants will then be converted into O2 and H2 (Sucahya et al., 2016). The photocatalyst technique has been applied to the treatment of textile dyeing waste (Arifiani and Hadiwidodo, 2007), laundry waste (Wahyuono et al., 2021), noodle industry wastewater (Dwiasi and Setyaningtyas, 2014), batik industry wastewater (Priantoro, 2020), and tempe industrial wastewater (Priantoro, 2020; Nisaa, 2018). However, fish processing byproducts have never been treated with photocatalysts.

Zinc oxide (ZnO) is a widely recognized photocatalyst due to its strong photocatalytic activity, chemical stability, biocompatibility, non-toxicity, and high photosensitivity. Compared to other semiconductor photocatalysts such as titanium dioxide (TiO_2), ZnO offers broader UV light absorption and superior quantum efficiency, which make it particularly effective in breaking down complex organic pollutants. Additionally, ZnO's intrinsic antimicrobial properties provide an advantage in addressing microbial contaminants often present in fish processing wastewater. While ZnO has been extensively studied for general wastewater treatment, its application as a photocatalyst for fish-frozen wastewater remains unexplored. This study aims to investigate the effects of ZnO mass variation and contact time on the reduction of chemical oxygen demand (COD), total suspended solids (TSS), and pH changes in fish processing wastewater through photodegradation.

2. Methods

2.1 Samples

The samples were collected from one of the fish processing industrial zones in Lampulo, Kuta Alam, Banda Aceh. Sampling was conducted using the grab sample method in accordance with the Indonesian National Standard (SNI 689.59.2008). At the fish processing facility, liquid waste samples were collected between 10:00 and 11:00 WIB, as this time coincided with peak fish processing operations. One liter of frozen fish wastewater was poured into a 1-liter beaker to determine the optimal deposition time. The samples were allowed to settle for four hours to allow for precipitation.

2.2 Experiment

For photodegradation experiments, 1 liter of the sample was transferred into a reactor tube. The apparatus used in the experiment is depicted in Figure 1. The sample was supplemented with ZnO in varying quantities (0.50 g, 0.75 g, and 1 g) and homogenized using a magnetic stirrerThe temperature was maintained at 26°C using a controlled water bath to ensure consistent reaction conditions. Irradiation durations were varied at three, four, and five hours. After irradiation, the sample was allowed to settle, and further analyses were conducted on the supernatant.

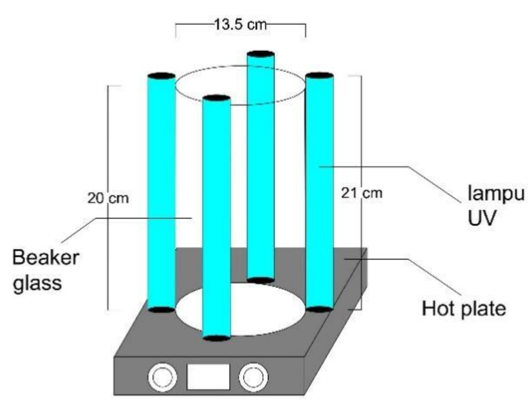

Figure 1. The experiment apparatus.

2.3 Measurements

In accordance with SNI 6989:11:2004, the pH values were measured directly with a multiparameter instrument in order to determine the amount of sour existing materials in the waste. Using electrode-dried tissue paper that had been washed with distilled water, the pH of each stage was measured. The test sample was used to wash the electrode. Afterward, the electrode was immersed in the sample until a constant reading was obtained. pH meter scale or reading numbers are displayed and recorded (SNI 06-6989.11-2004).

COD measurements were performed to determine the amount of organic matter in the waste laundry water (refer to SNI.6989.02.2009). Using a dropper, add 1.5 mL of $K_2Cr_2O_7$ and 3.5 mL of H_2SO_4 to a reaction tube containing up to 2.5 mL of waste water that has been labeled in accordance with an experiment of variation. The COD reactor of the Hanna brand was then heated to 150 °C. The tube reaction was then regulated for two hours in the reactor before being transferred to the rack tube reaction for cooling.

TSS measurements were carried out with measurements in a manner called gravimetry, which refers to SNI 6989.3.2004. This measurement is performed to determine the rate at which solids suspended in waste, organic or inorganic, are good ingredients. The TSS measurement steps, specifically the filter on the equipment, were vacuum moistened with aquades, then the paper filter Whatman number 42 was washed with aquades for sterilization, then allowed to dry before weighing. The tool vacuum is then subjected to a paper strain. The paper filter already used in accordance with the variation treatment dried in the oven for 1 hour at 103 °C. Paper strain cooled in desiccator; weighing completed until heavy constant The TSS value of the sample can be calculated by using **Equation 1**.

$$\text{TSS} = \frac{(\text{A}-\text{B})\text{x } 1000}{\text{Test sampel volume}} \quad (1)$$

In this study, the data analysis phase determined the percentage reduction in pollutant load in fish processing wastewater for each of the parameters tested before and after treatment with ZnO photodegradation. To determine the efficiency of the photocatalyst process, use the formula in **Equation 2** below.

$$\%\ \text{Effectiveness} = \frac{\text{Initial level}-\text{final level}}{\text{Initial level}} \text{ x } 100 \quad (2)$$

3. Results and Discussion

Change physical processes occurring in the waste fish processing before the experiment showing dark and dark color. After the experiment with photocatalyst ZnO color waste fish processing was clearer. **Figure 2** shows the appearance of physique waste fish processing (a) before the experiment, (b) after the photocatalyst process, and (c) after precipitated. The results of measurements are shown on **Table 1**.

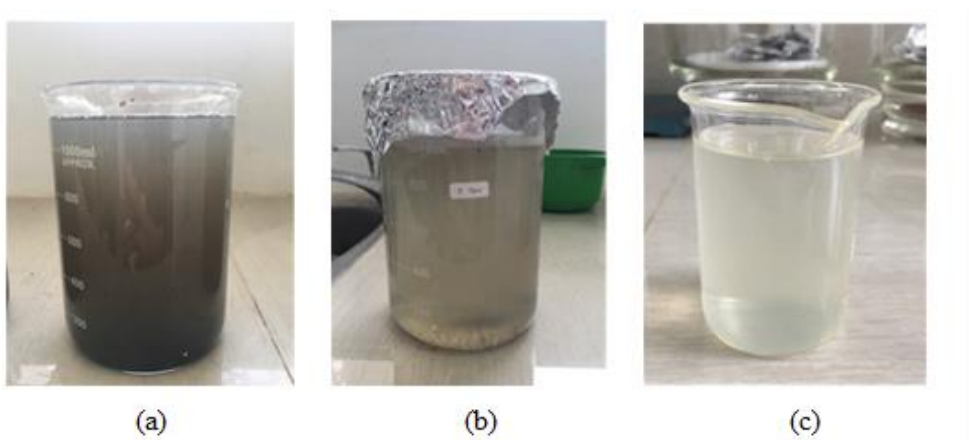

Figure 2. waste fish processing (a) before the experiment, (b) after deposition, and (c) after the photocatalyst process.

Table 1. Wastewater Quality Standards for Fishery Product Processing Businesses and/or Activities that Perform One Type Processing Activities

Experimental Variations		pH				COD (mg/L)					TSS (mg/L)				
Contact Time (hours)	Catalyst Mass (grams)	Quality standards	Preliminary measurement results	Measurement results after deposition	Measurement results after photocatalyst	Quality standards	Preliminary measurement results	Measurement results after deposition	Measurement results after photocatalyst	Photocatalyst Effectiveness	Quality standards	Initial measurement results	Measurement results after deposition	Measurement results after photocatalyst	Photocatalyst effectiveness
3	0.0	6-9	6.8	6.8	7.6	200	424	386	455	15.16	100	443	224	212	5.36
4	0.0				7.7				351	9.07				210	6.26
5	0.0				7.8				345	10.62				206	8.04
3	0.50				7.5				310	19.69				178	20.54
	0.75				7.2				248	35.75				138	38.39
	1.0				7.1				211	45.34				113	49.55
4	0.5				7.9				245	36.53				107	52.23
	0.75				7.7				198	48.70				97	56.70
	1.0				7.7				134	65.28				93	58.48
5	0.5				7.9				185	52.07				82	63.39
	0.75				7.8				129	66.58				57	74.55
	1.0				8				117	69.69				41	81.70

Based on **Table 1** results analysis initial waste parameters liquid has passed raw quality stipulated in the Regulation of the Minister of the Environment Life and Forestry (PERMIT LHK) Number 5 of 2014 concerning Wastewater Quality Standards. After conducting processing use the method of photodegradation with variation mass catalyst 0.50, 0.75, and 1 g with variation times 3, 4, and 5 hours. Waste parameters liquid fish processing experienced a decline and have a sufficient standard raw quality that has stipulated by PERMEN LHK Number 5 of 2014.

3.1 Effect of deposition in the experiment

Precipitation aims for separate solids that settle on the surface of the water-style gravity (Indrayani and Rahmah, 2018). Precipitation in the experiment this conducted for knowing the influence of the deposition process on the percentage decrease in the parameters tested. Based on **Figure 2**, the COD and TSS parameters are experiencing a decline after conducted deposition. Profit from deposition because going on in a manner gravity without existing additional substance chemistry, so more friendly environment because generated waste does not contains chemistry ingredient. However, deposition needs many times and require a tub for deposition.

Based on **Figure 3** shows that the experienced TSS values decline of 443 mg/L fell to reach 213 mg/L, the same case with the experienced COD parameter decline starts from 424 mg/L to 386 mg/L. However, the results of the deposition obtained not yet fulfill the standard raw quality based on PERMEN LHK Number 5 of 2014.

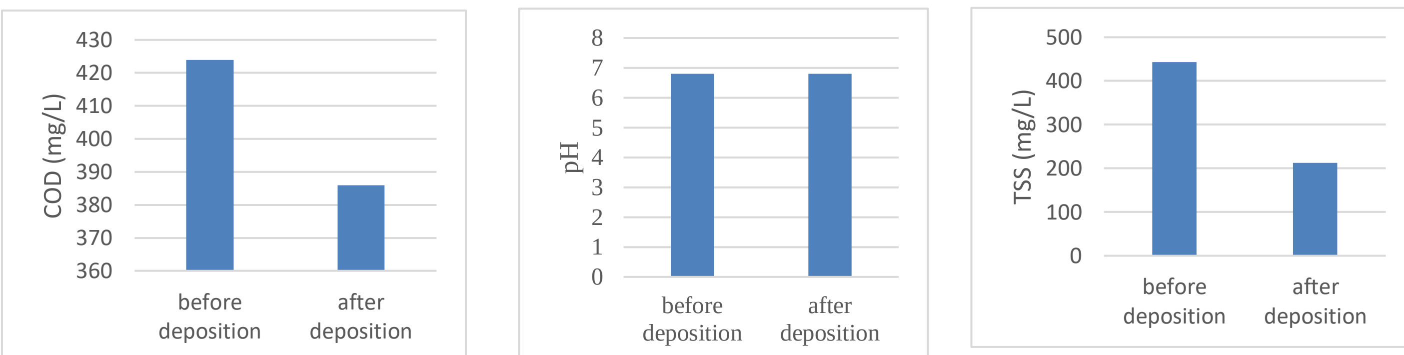

Figure 3. Parameter values before and after deposition

3.2 Effect of ZnO mass

Graph of catalyst mass test results for pH values in fish frozen wastewater are shown on **Figure 4**. Experimental results of photodegradation show that pH value is not influenced by the masses ZnO. pH value has a very important role in the formation of radical hydroxyl. Amount radical the resulting hydroxyl (·OH). influenced by the condition of the pH of the solution at conditions acid (<7), formation electrons - *holes are* blocked so that produce little ·OH. At alkaline pH, ·OH will in amount many (Dwiasi and Setyaningtyas, 2014) .

The addition of mass catalyst caused the pH to increase more language, p this caused surface photocatalyst ZnO will be loaded negative at alkaline pH. this is because will more easily degrade loaded waste positively charged ZnO negative (Permata et al., 2015). A higher pH base will cause level reduction the higher, so the level of absorption will be higher anyway. At high pH, there are excess OH anions that will make it easy for OH radical photodegradation (Dini, 2019).

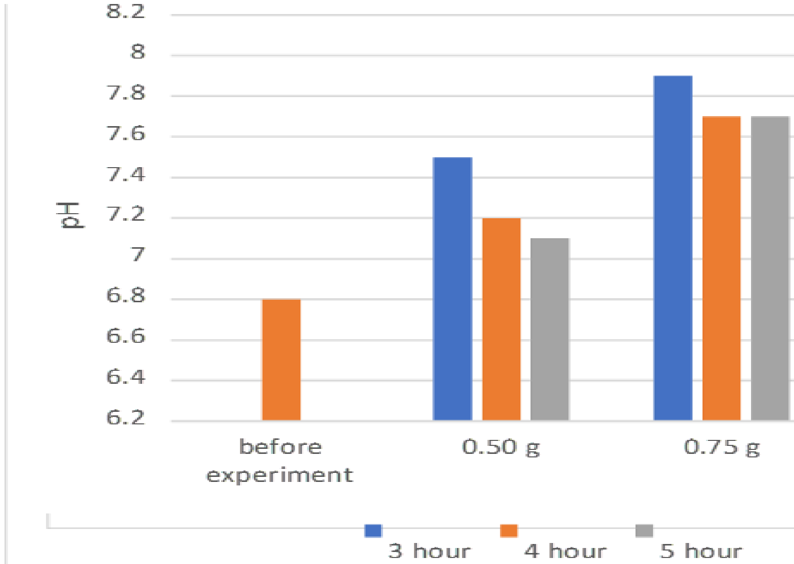

Figure 4. Graph of catalyst mass test results for pH values in fish frozen waste water

Graph of test results for the effect of catalyst mass on COD values in fish processing wastewater is shown on **Figure 5**. Experiment photodegradation showing results that mass catalyst ZnO used influence COD parameter degradation. Effectiveness reduction in COD reached 69.69% with mass catalyst ZnO 1 g with a time contact of 5 hours. Degradation results from the lowest seen in the addition of 0.50 g of catalyst ZnO with a time contact of 3 hours are 19.69%. Using ZnO 1 g can see that the percentage allowance will the higher, starting from time contact 3 hours to 5 hours. Reaction photocatalyst could be accelerated with the presence of ZnO material and assistance from photons, next the corresponding photon with the band gap will stimulate electrons contained in ZnO and will form *the hole.* Electrons stimulated on energy conduction will tie oxygen dissolved, and will produce radical oxygen ($O_2\cdot$) where as *holes* formed on the level energy valence will tie electrons contained in the OH- ions found in the waste liquid, so will form ·OH. Furthermore, $O_2\cdot$ and OH· will be a role in deciphering causative agent pollution in waters (Mohar et al., 2021).

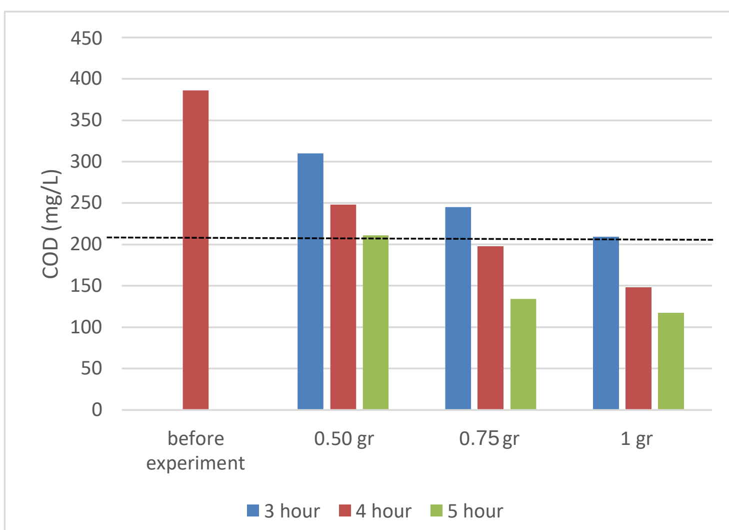

Figure 5. Graph of test results for the effect of catalyst mass on COD values in fish processing wastewater

Graph of test results for the effect of catalyst mass on TSS values in fish processing wastewater are shown on **Figure 6**. Experimental results photocatalyst showing that addition of mass ZnO influence TSS degradation. Initial TSS value before conducted processing of 443 mg/L, after processing of the resulting TSS up to 41 mg/L. TSS parameter degradation can see in **Figure 4**, the TSS parameters are experiencing a decline. Effectiveness TSS parameter degradation of 81.70% in mass catalyst 1 g with time contact 5 hours. This showing that ZnO has a role important in photodegradation because the catalyst could produce *a hole* ($h^{+)}$ to react with subsequent hydroxyl ions ($OH^{-)}$. will form radical hydroxyl (·OH) which will degrade the substance organic thus the degradation process becomes faster. The taller formation of radical hydroxyl, the bigger the ability of photocatalysts for degrading (Sibarani et al., 2016).

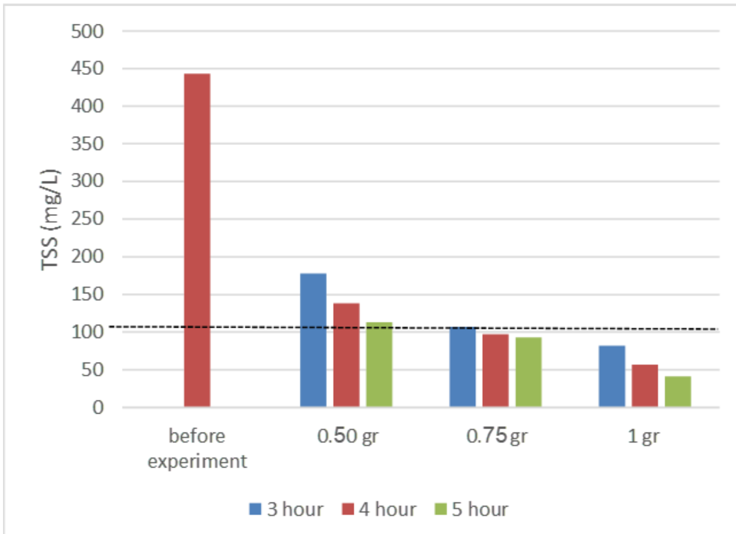

Figure 6. Graph of test results for the effect of catalyst mass on TSS values in fish processing wastewater

The observed lower efficiency in COD reduction compared to TSS can be attributed to the inherent differences in the nature of these parameters. TSS primarily consists of particulate matter that can be physically separated or degraded more readily during photodegradation and precipitation. In contrast, COD reflects the presence of dissolved and complex organic compounds, which require more energy and prolonged reaction times to break down into simpler molecules. The complexity and chemical stability of these organic compounds reduce the effectiveness of photodegradation within the same experimental timeframe.

Additionally, the COD reduction process relies heavily on the generation of reactive oxygen species (ROS), such as hydroxyl radicals (·OH), which interact with organic pollutants. However, factors like limited ZnO surface area, insufficient photon absorption, and potential recombination of electron-hole pairs may hinder the generation of sufficient ROS to degrade the more complex compounds contributing to COD. These results align with previous findings (Mohar et al., 2021), which state that while ZnO is effective in degrading simpler organic pollutants, higher doses or longer exposure times are often required for more significant COD reduction.

This difference highlights the need for optimizing both catalyst mass and contact time to improve COD removal while maintaining the efficiency of TSS reduction. Future research could focus on enhancing ZnO's photocatalytic activity through doping or by using hybrid photocatalyst systems to address the limitations observed in COD degradation.

3.3 Effect of contact time

Graph of the effect of contact time with a decrease in pH parameters is shown on **Figure 7**. Effect of contact time to the pH parameter no influence ever time contact. on time 3-hour contact with mass catalyst 0.50, 0.75 and 1 g ie 7.5, 7.2, 7.1. on time 4 hours of contact with a catalyst mass of 0.50, 0.75, and 1 g, namely 7.9, 7.9, and 7.7. Whereas time 5 hours of contact with a catalyst mass of 0.50, 0.75, and 1 g, namely 7.9, 7.8, 8. Changes score this characteristic fluctuating, changing score not stable with increased time contact. At a long time 5 hours of irradiation pH reached 8, p this is supported by Palupi, (2006) state that the more ever time irradiation, then the more many the photon hits catalyst ZnO, so the resulting ·OH from the photodegradation process the more increase.

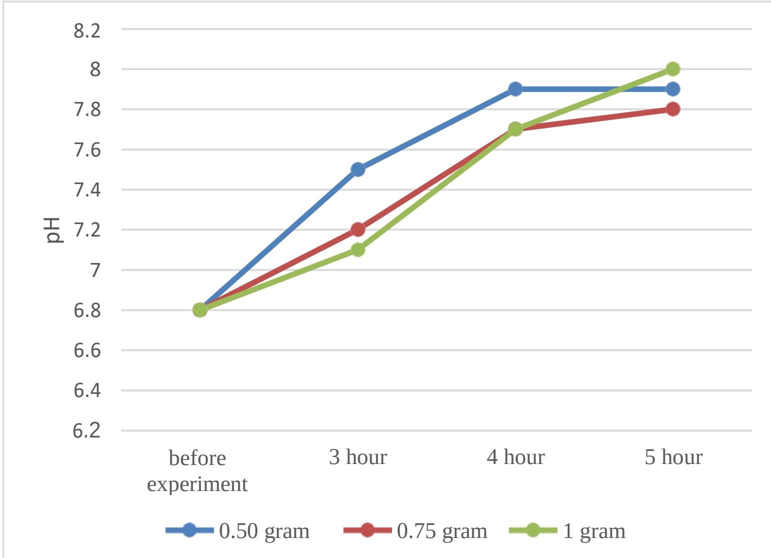

Figure 7. Graph of the effect of contact time with a decrease in pH parameters

Graph of the effect of contact time with a decrease in the COD parameters is shown on **Figure 8**. Experimental results photocatalyst showing that the COD parameter affects variation time contact. COD decreased along increase time contact. COD content contained in waste fish processing before he did photodegradation catalyst ZnO. on time a 5 h contact with a catalyst mass of 1 g was obtained percentage photocatalyst reached 69.69%. Longer the time contact between waste liquid with photocatalyst so the percentage of COD reduction will be the bigger. The consequence of time brief contact causes a clash Among substances organic with the photocatalyst too short. This triggers the surface photodegradation photocatalyst suboptimal because substance-driven other organics that will pass photocatalyst, so things that affects the degradation process yet perfectly causes a decrease in COD to low. Compared backward with time-long contact, so obtain results in maximum degradation (Aditya et al., 2012).

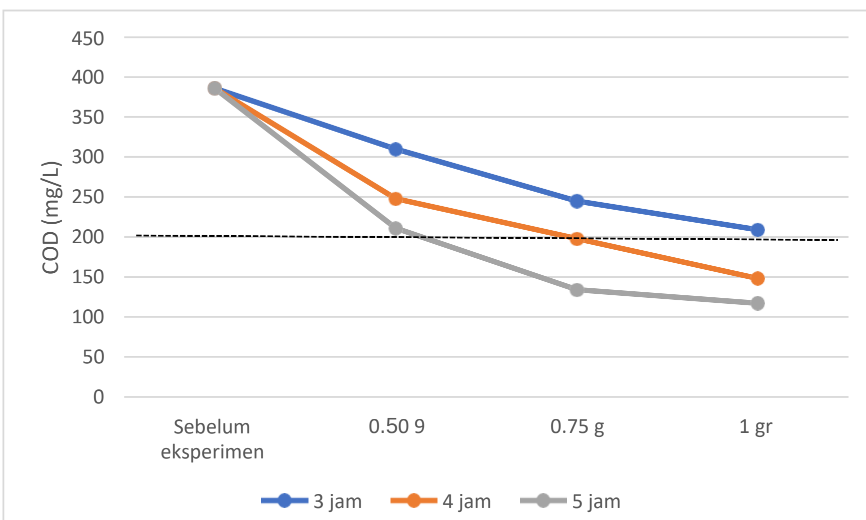

Figure 8. Graph of the effect of contact time with a decrease in the COD parameters

Experimental results photocatalyst showing that time contacts influence decline TSS value. TSS parameters have an effectiveness degradation of 81.70% on the mass of 1 g of catalyst ZnO with a time contact of 5 hours. TSS parameter degradation can be seen in **Figure 4** and **Figure 8** as experienced TSS levels decline score by the standard raw quality that has been set. kindly physical water that has been conducted in the experiment clearer. This is supported by a study earlier by Fauzi and Tuhu Agung, (2018) states that the more many time contact will cause percentage allowance will the more increase. Based on the study results, highest TSS allowance obtained was 94.5% with a time contact of 120 minutes and percentage the lowest TSS allowance of 43.8% with a time 10-minute contact from concentration early.

Graph of the effect of contact time with a decrease in TSS parameters is shown on **Figure 9**. The more ever time radiation to ingredient degraded organics. This is because the more ever time radiation so the more many the photon that hits catalyst ZnO, so cause an increased amount · OH, radicals · OH works with attack molecule ingredient organic matter and degrades it Becomes more simple (Sanjaya, 2017).

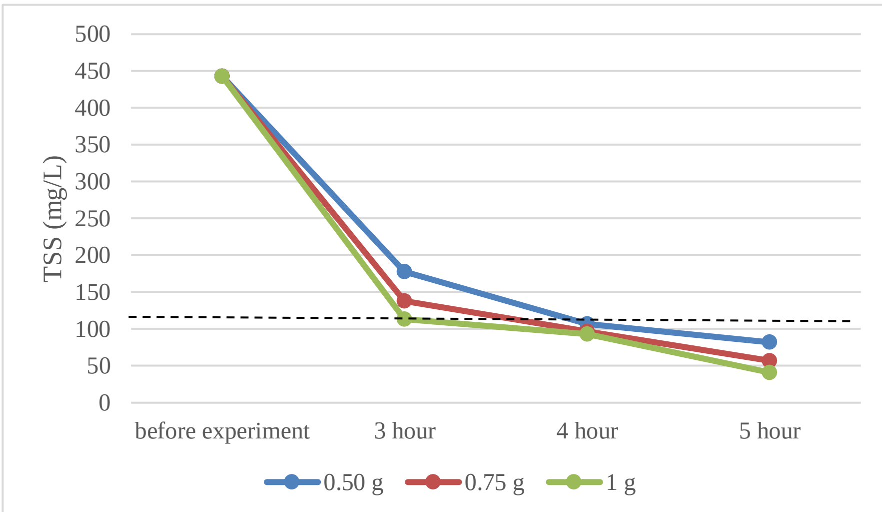

Figure 9. Graph of the effect of contact time with a decrease in TSS parameters

4. Conclusion

Based on the results and analysis, it can be concluded that the ZnO photodegradation method is effective for treating fish processing wastewater. The optimal conditions were achieved with a ZnO mass of 1 g and a contact time of 5 hours. Under these conditions, the pH reached 8, COD reduction efficiency was 69.69%, and TSS reduction efficiency was 81.70%. Statistical analysis using a T-test showed significant differences for COD ($p = 0.000$) and TSS ($p = 0.004$), while the pH parameter was not significantly affected ($p = 0.229$).

The findings suggest that ZnO photodegradation can effectively reduce both organic and physical pollutants in fish processing wastewater. Future research is recommended to scale up this process for industrial applications and explore combining ZnO with other advanced treatment methods, such as biological or chemical approaches, to enhance degradation efficiency and address more complex pollutants.

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