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Effectiveness of Organic Compound Removal in Fishery Product Processing Wastewater Using Photocatalysis Method

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ABSTRACT: The development of the industry is increasingly rapid to support the needs of human life, one of which is in the food industry in the form of fish processing at the Fish Center Surabaya. The Fish Center Surabaya has a WWTP consisting of a storage tank, a settling tank and a biofilter. The photocatalysis method used in the study serves to alleviate the organic load contained in the wastewater before it is flowed to the biofilter unit. In addition, this study aims to determine the mass of ZnO compound, the optimal irradiation time and irradiation intensity to reduce the parameters of BOD, COD and NH_4 in the wastewater of the fishery product processing. In the photocatalysis method using ZnO compound have masses of 1 g, 1.5 g and 2 g. In addition, there are also irradiation intensity variables of 732±5,17 μW/cm^2, 743±3,56 μW/cm^2 and 849±1,92 μW/cm^2 with irradiation time variables of 8 hours which were sampled at each hour. After that, the results will be analysed and discussed to answer the research objectives. The photocatalysis process using ZnO is known to reduce the content of BOD, COD and NH_4 in fishery products wastewater by 27%, 31% and 35%.

KEYWORDS: fishery product processing wastewater, photocatalysis, ZnO, organic pollutants

I. INTRODUCTION

As the demand for fish consumption increases, it will increase the volume of wastewater produced from fishery product processing activities, one of which is the Fish Center in Surabaya. Wastewater from fisheries comes from water used for fish washing and cleaning fish stomachs to cutting fish with a fairly high content of organic matter (Anh, et al., 2021). Wastewater from fisheries has a high content of oil, fat and protein from the fish processing process including washing, fermentation, filleting, and canning. This causes high content of Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), and nutrients contained in wastewater from the fish processing industry (Khalatbari, et al., 2023). The Fish Center already has a Wastewater Treatment Plant (WWTP) consisting of a storage tank, a settling tank and a biofilter tank.

Several methods of processing wastewater from fisheries can be classified into three types of processing methods, namely physically, chemically, and biologically which can be in the form of adsorption, filtration, ion exchange, and even photocatalysis processes. In addition, it can be done using various methods such as phytoremediation that are able to set aside TDS, TSS and BOD levels (Widiyanti & Hamidah, 2021). Another method that has been carried out by Pangestika, et al. (2022) is the ozonization method that is able to set aside TDS, TSS, COD and BOD levels, in addition to that it can also use the electrochemical oxidation method which is able to set aside COD and BOD levels (Souli, et al., 2023).

This study uses physical and chemical processing methods in the form of a photocatalysis process using UV light with the help of semi-conducting photocatalytic chemical compounds such as ZnO, TiO_2, MnO_2, WO_x and $CaCu_3Ti_4O_{12}$ (Le et al., 2022). The photocatalysis method was chosen because it has cheaper operational costs when compared to other Advance Oxidation Process (AOPs) methods. In addition, it does not require advanced processing methods from the degradation results (Annisaputri, et al., 2020), can degrade organic compounds and reduce inorganic compounds into simple components that are safe for the environment (Wildan & Mutiara, 2019). The photocatalysis process is the process of converting light energy into chemical energy and in the process will produce hydroxyl radicals that will react redox with organic compounds (pollutants), so that the water will return to clarity because it is separated from wastewater (Sujatha, et al., 2020). This process can be accelerated by using photocatalytic materials, which are materials that are able to accelerate photochemical reactions. The photocatalyst method using catalyst ZnO, it has a sufficiently large band gap (E_g) energy of up to 3.37 eV which can only be activated through a relatively high-energy light source such as UV light. In general, when ZnO is exposed to UV light that has more energy than the energy band, it will produce electron pairs and holes. The UV light used in the photocatalysis process is UV C with a wavelength of 200 to 260 nm (Hammer Sr. & Hammer Jr., 2014). The use of the photocatalysis method in the treatment of wastewater in the fish industry functions as a treatment process before the wastewater is flowed to the biofiler unit which is used to ease the organic load before being treated using a biofilter. The purpose of this study is to determine the mass of ZnO compound, the optimal irradiation time and irradiation intensity to reduce the parameters of BOD, COD and NH_4 in fish processing wastewater.

II. MATERIALS AND METHOD

A. Study Area

The sampling location was carried out at the Fish Center, which is a center for processed fish products in Surabaya, East Java. Wastewater sampling was carried out as much as 30 L with a frequency of 2 times to meet the needs of the research. The sampling method used is grab sampling with procedures in accordance with SNI 6989.59:2008 concerning the method of billing wastewater samples. The grab sampling method is sampling that is carried out instantaneously and at a certain location using a dipper and the sample is stored in a plastic bottle.

B. Preparation of Tools and Materials

The research stage begins with the preparation of tools and materials which include sampling, providing catalysts, making reactors and preparing photocatalysis reactor designs. The implementation of the photocatalysis process was carried out in the ITS Environmental Engineering Workshop and characteristic testing was carried out in the Water Treatment Technology Laboratory, ITS Environmental Engineering Department. Details of the tools and materials used in the study are 30 L jerry can, measuring cup, UV-C lamp with AUV 03A 3 Watt, reactors made from 8 mm glass, mixing pump, aluminum foil, sample, ZnO. Sampling of wastewater from fishery products found at the Fish Center in Surabaya was 30 L. Then a research reactor made of glass with a thickness of 8 mm and dimensions of 25 cm in length, 15 cm in width and 25 cm in height. photocatalysis using UV C Lamp with AUV 03A 3 Watt specification as well as chemical compounds ZnO and TiO2 to determine the decrease in BOD, COD and NH3 parameters. UV C lamps were chosen because they are able to activate catalysts found in ZnO and UV C lamps have greater energy when compared to UV A and UV B lamps (Ramadhika, et al., 2021). Preliminary research was conducted to determine the initial characteristics of fishery wastewater with pH, NH_4, BOD and COD parameters used as a comparison in determining the percentage of removal efficiency.

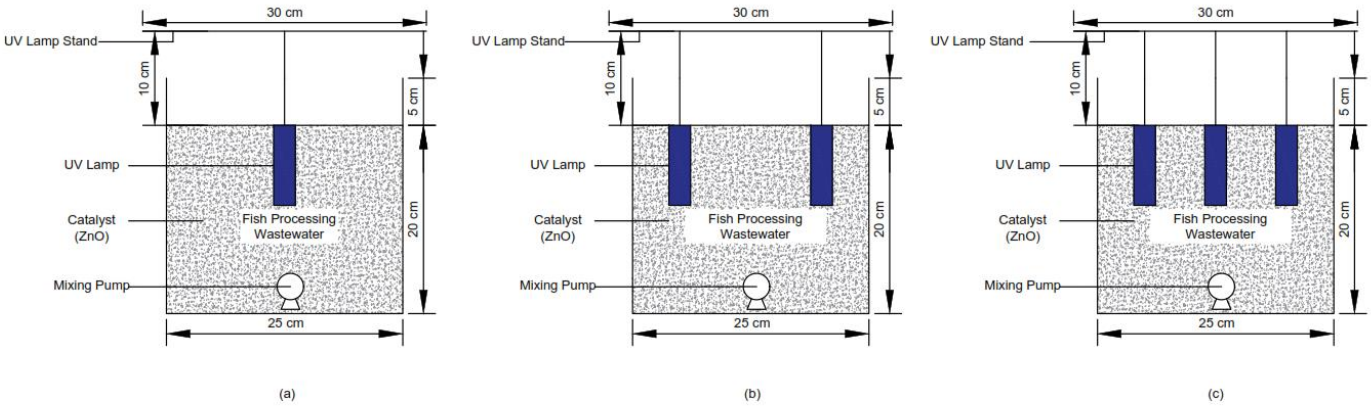

Figure 1: Photocatalysis reactor design (a) using UV lamp with irradiation intensity 732±5.17 μW/cm², (b) using UV lamp with irradiation intensity 743±3.56 μW/cm², (c) using UV lamp with irradiation intensity 849±1.92 μW/cm²

The main research carried out was in the form of wastewater treatment of the Fish Center using the photocatalysis method using ZnO photocatalyst compounds with masses of 1 g, 1.5 g and 2 g. In addition, irradiation intensity variables of 732±5.17 μW/cm², 743±3.56 μW/cm² and 849±1.92 μW/cm² were also used and the irradiation time was 8 hours carried out in the batch method reactor. Reactors on Fig1 (a) is a research reactor consisting of 1 UV-C lamp which has an irradiation intensity of 732±5.17 μW/cm², a catalyst in the form of ZnO and a mixing pump. While in Fig1 (b) consists of 2 UV-C lamps that have an irradiation intensity of 743±3.56 μW/cm², a catalyst in the form of ZnO and a mixing pump and on the Fig1 (c) consists of 3 UV-C lamps that have an irradiation intensity of 849±1.92 μW/cm², a catalyst in the form of ZnO and a mixing pump. Sampling in the research reactor of 50 mL was carried out every 1 hour with a frequency of 8 times. Then pH, NH_4, BOD and COD tests are carried out in accordance with the procedures listed in the Standar Nasional Indonesia (SNI) as in Table 1.

Table 1: Parameter analysis method

No	Parameter	Satuan	Metode
1	pH	-	SNI 6989.11:2019
2	*Biological Oxygen Demand* (BOD)	mg/L	SNI 6989.72:2009
3	*Chemical Oxygen Demand* (COD)	mg/L	SNI 6989.2:2009
4	*Ammonium* (NH_4)	mg/L	SNI 06-2479-1991

III. RESULTS AND DISCUSSION

A. Characteristics of Fish Processing Wastewater

The initial characteristics of the wastewater of fish processing were used to determine the concentration of pH, BOD, COD and Ammonium parameters. Based on the results of the preliminary research, the parameters used in this study can be determined to analyze the decline using the photocatalysis method. The parameters used in this study are BOD, COD and Ammonium because they have a high concentration.

Table 2: Characteristics of fish processing wastewater

No.	Parameters	Unit	Result		Quality Standards *)
			22/5/24	15/6/24	
1	pH		6,8	5,9	6-9
2	BOD	mg/L	864	1248	100
3	COD	mg/L	1616,7	2335	200
4	NH4	mg/L	1,75	2,16	5

Note: *) Quality standards are in accordance with Lampiran XIV Peraturan Menteri Lingkungan Hidup No. 5 Tahun 2014 concerning Baku Mutu Air Limbah Bagi Usaha dan/atau Kegiatan Pengolahan Hasil Perikanan

B. Photocatalysis Method in Fish Processing Wastewater

The photocatalysis process used ZnO compounds with masses of 1 g, 1.5 g and 2 g accompanied by variations in irradiation intensity of 732±5.17 µW/cm^2, 743±3.56 µW/cm^2 and 849±1.92 µW/cm^2 and irradiation time of 8 hours to regulate BOD, COD and NH_4 parameters. Based on previous research, it shows that the irradiation intensity needed to activate the ZnO catalyst is 515.53 µW/cm^2 (Setiawan, 2018). The mechanism of organic compound elimination using the photocatalysis method that occurs can be seen as follows (Asri, et al., 2022).

$$ZnO + hv \rightarrow ZnO\ (e^-_{CB}, h^+_{VB}) \quad (1)$$

$$ZnO(h^+_{VB}) + H_2O \rightarrow ZnO + H^+ + OH^- \quad (2)$$

$$ZnO(h^+_{VB}) + OH^- \rightarrow ZnO + \cdot OH \quad (3)$$

$$O_2 + ZnO(e^-_{CB}) \rightarrow ZnO + \cdot O_2^- \quad (4)$$

$$2 \cdot O_2^- + 2H_2O \rightarrow O_2 + 2OH^- + 2 \cdot OH \quad (5)$$

$$\cdot OH + organic\ compounds \rightarrow degradation\ of\ organic\ compounds \quad (6)$$

The reaction shows that if ZnO is given light (hv) that has energy according to the need to activate the ZnO compound, then the electron (e^-) in the valence band (V_B) will move towards the conduction band (C_B). The electron transfer (e^-) produces e_{CB}^- so that it causes a void or hole (h_{VB}^+). The resulting electrons will react with electron acceptors such as O_2 adsorbed on the surface of ZnO or dissolved in water to form superoxide radical anions (•O_2). Then the resulting hole can oxidize organic compounds or react with OH^- or H_2O to produce hydroxyl radicals (•OH). Hydroxyl radicals (•OH) or superoxide (•O_2) play a role in the process of pollutant removal in wastewater (Sugiyana & Notodarmojo, 2015).

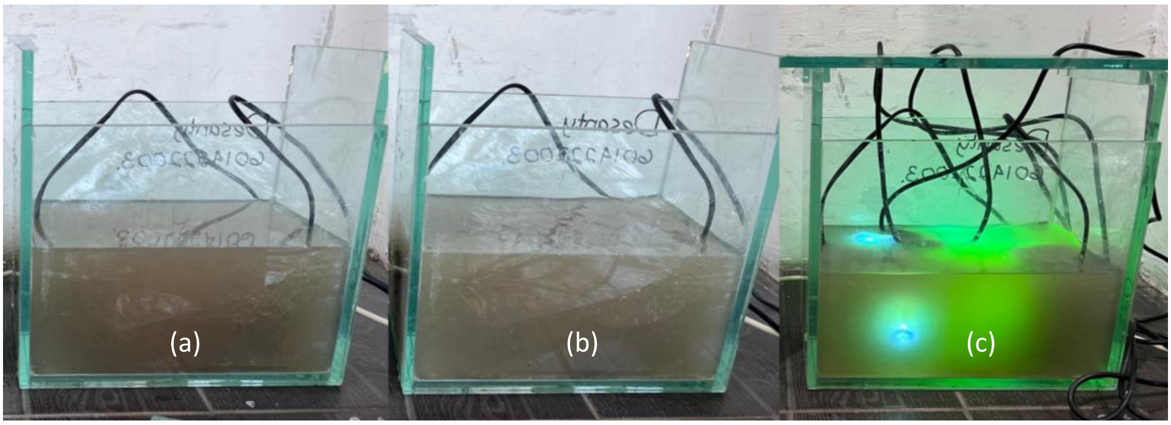

Figure 2: Photocatalysis process, (a) sample without treatment, (b) sample with catalyst ZnO, (c) sample with photocatalysis process

The photocatalysis process using ZnO compounds with masses of 1 g, 1.5 g and 2 g with irradiation intensity variations of 732±5.17 μW/cm^2, 743±3.56 μW/cm^2 and 849±1.92 μW/cm^2 in fishery wastewater resulted in a change in pH value from an acidic condition (pH < 7) before treatment to an alkaline condition (pH > 7) after treatment. The wastewater from fisheries products added with 1 g of ZnO with an irradiation intensity of 732±5.17 μW/cm^2 and an irradiation time of 8 hours had a pH in the range of 7.53 to 8.05. Meanwhile, the addition of ZnO of 1.5 g has a pH in the range of 7.03 to 7.79, and the addition of ZnO of 2 g has a pH range of 7.46 to 8.06. The wastewater from fisheries added with 1 g of ZnO with an irradiation intensity of 743±3.56 μW/cm^2 and an irradiation time of 8 hours had a pH in the range of 6.43 to 7.45. Meanwhile, the addition of ZnO 1.5 g has a pH in the range of 6.62 to 7.43, and the addition of ZnO 2 g has a pH range of 6.71 to 7.52. Fishery wastewater added with 1 g of ZnO with an irradiation intensity of 849±1.92 μW/cm^2 and an irradiation time of 8 hours had a pH in the range of 6.75 to 7.54. Meanwhile, the addition of ZnO 1.5 g has a pH in the range of 6.63 to 7.93 and the addition of ZnO 2 g has a pH range of 7 to 8.01.

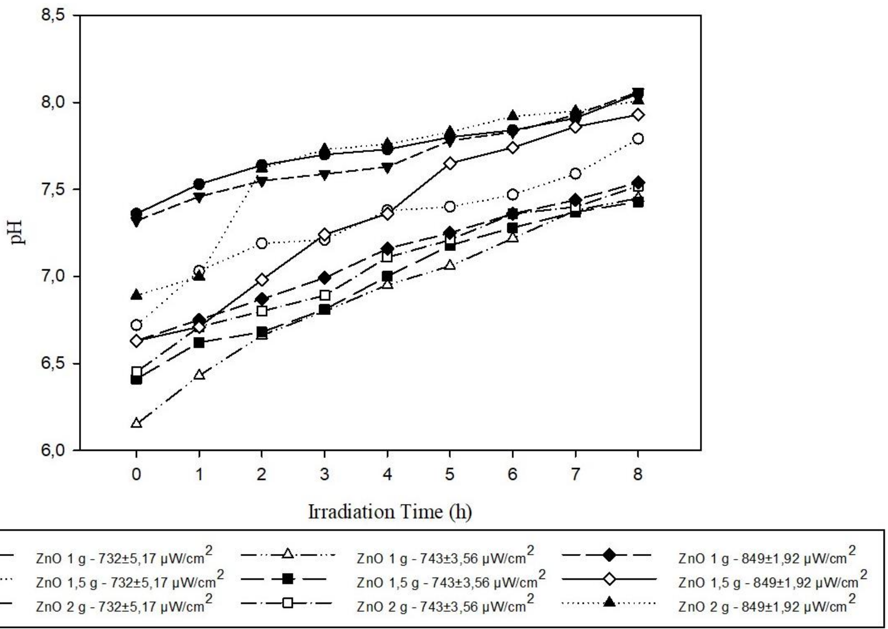

Figure 3: pH values from photocatalysis process

In the photocatalysis process using ZnO, the wastewater from fisheries is found in alkaline conditions caused by the large amount of OH bound by ZnO, causing the surface of ZnO to be negatively charged. The photocatalyst surface is negatively charged under the period condition and positively charged under the acid condition (Lee, Kim, Danish, & Jo, 2023). Wastewater from fisheries before the photocatalysis process has a pH value of 6.8 which is classified as acidic (pH < 7), so it is positively charged. Meanwhile, the photocatalysis process using ZnO produces an alkaline pH value (pH > 7), so it is negatively charged. The difference in the load found in the wastewater of fishery products and the surface of ZnO can result in pollutants in wastewater being more easily degraded by ZnO (Lee, Kim, Danish, & Jo, 2023). Each additional irradiation time in the photocatalysis method using ZnO produces an increasingly alkaline pH value so that it can increase photocatalysis activity. A high pH value can give hydroxyl ions higher, so they can react with holes (h_{VB}^+) and form hydroxyl radicals (Kazeminezhad & Sadollahkhani, 2016).

1. Ammonium (NH_4) Removal

Ammonium (NH_4) can cause harm to aquatic ecosystems if it has concentrations that exceed the set quality standards (Ngibad, 2019). Ammonium (NH_4) can be found in wastewater with pH concentrations in the range of 6-8 (Malovanyy, et al., 2019). Ammonium (NH_4) is one of the parameters that indicates the decomposition of organic matter that occurs in wastewater from fishery products and can interfere with water quality if it has a value that exceeds the quality standard. The photocatalysis process using ZnO had the highest NH_4 clearance efficiency of 35% at a mass of 1 g with an irradiation time of 8 hours and an irradiation intensity of 849±1.92 μW/cm², an initial concentration of 0.96 mg/L to 0.62 mg/L. The lowest NH_4 clearance efficiency was 1% at a mass of 1.5 g with an irradiation time of 3 hours and an irradiation intensity of 743±3.56 μW/cm². with an initial concentration of 0.78 mg/L to 0.77 mg/L.

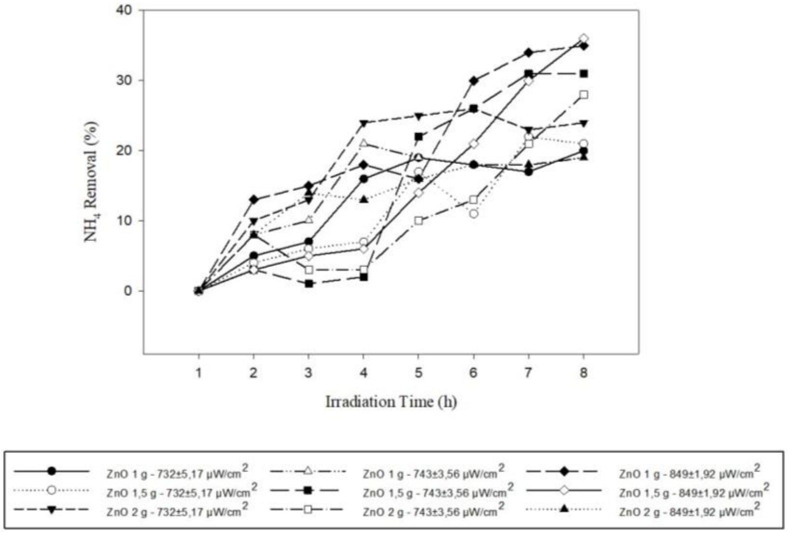

Figure 4: NH_4 removal

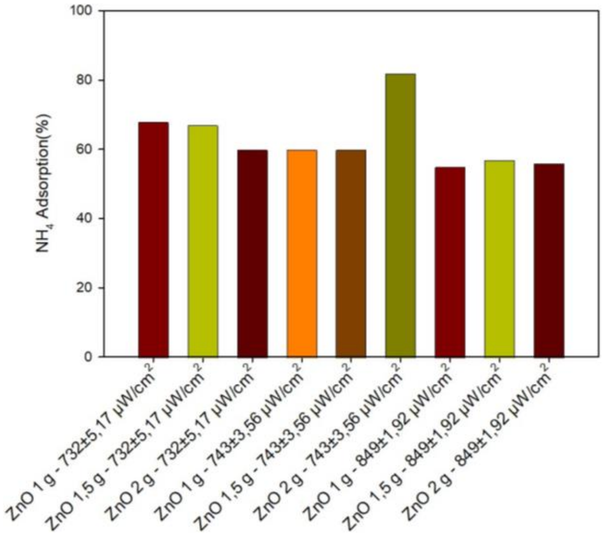

Figure 5: NH_4 adsorption

By Fig4 It can be seen that the longer the irradiation time will result in a higher removal efficiency, but the removal efficiency will decrease as the optimum time passes. This is supported by research conducted by Wulansari (2018) which stated that when the optimal time has passed, the degradation of wastewater will decrease, which is suspected because the active side of the catalyst is saturated. The percentage of NH_4 removal using ZnO shows that the highest value is 35% so that the photocatalysis process is considered not effective enough in NH_4 removal in fishery wastewater. In the processing process using this photocatalysis method, it is estimated that the adsorption process by the catalyst surface occurs. The percentage of adsorption using ZnO against NH_4 exclusion can be seen in Fig5. The adsorption process that occurs can result from a fairly large surface area of ZnO. The adsorption process can occur if the available surface area of ZnO is relatively high, because the organic compounds contained in the wastewater are relatively small in size and the mass of ZnO used is quite large (Kholidah, et al. 2021). The surface area of the ZnO catalyst used in this research is 611.827 m²/g.

2. BOD Removal

Biological Oxygen Demand (BOD) is one of the parameters in wastewater that shows the amount of dissolved oxygen used by microorganisms to oxidize organic matter (Eddy M., 2003). Biological Oxygen Demand (BOD) is a parameter used to determine the total oxygen demand by microorganisms for the degradation process of pollutants (Sincero Sr. & Sincero, 2003). High levels of BOD concentrations can cause ecosystem damage to waters, as the life of organisms that need oxygen becomes disrupted. In addition, it can also result in the depletion of oxygen in wastewater and the presence of anaerobic conditions in wastewater (Astuti & Rosemalia, 2022). Fig6 It can be seen that the largest BOD exclusion process in the ZnO catalyst is 1.5 g mass and irradiation time for 8 hours with an irradiation intensity of 743±3.56 μW/cm² with an initial concentration of BOD of 275 mg/L to 202 mg/L with an exclusion percentage of 27%. Meanwhile, the lowest percentage of BOD removal was found at ZnO 1.5 g with an irradiation intensity of 849±1.92 μW/cm² at the 6th hour. The decrease in the percentage of removal in the photocatalysis process using ZnO is influenced by several factors, one of which is the turbidity of the sample so that it inhibits the penetration of UV in the wastewater of fishery products which makes ZnO completely inactive.

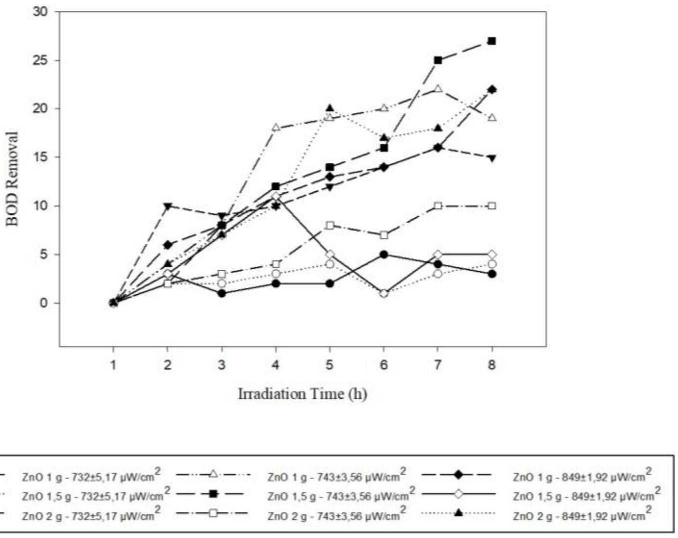

Figure 6: BOD removal

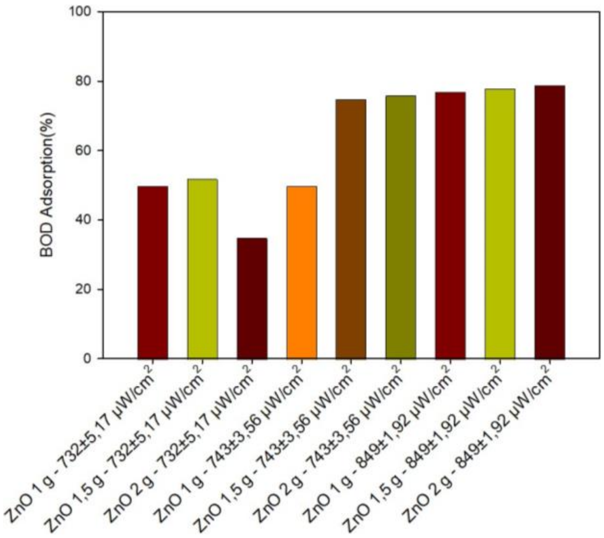

Figure 7: BOD adsorption

Based on the research that has been carried out, there is a decrease in BOD at the 0th hour, but at that time the wastewater from fisheries products is not treated with UV irradiation. The decrease in BOD that occurs in treatment without UV light can occur due to the adsorption process of organic compounds by the surface of ZnO. The percentage of BOD reduction caused by the adsorption process can be seen in Fig7. By Fig7 It can be seen that there is a BOD removal that reaches 79% in the use of ZnO 2 g with an irradiation intensity of 849±1.92 μW/cm^2. The percentage of BOD removal estimated due to the adsorption process has a larger removal than the photocatalysis process using ZnO which is only 27%. The adsorption process that occurs can result from a fairly large surface area of ZnO. The surface area of the ZnO catalyst used in this research is 611.827 m^2/g.

3. COD Removal

Chemical Oxygen Demand (COD) is the amount of oxygen needed to chemically decompose organic materials in wastewater (Eddy M., 2003). Chemical Oxygen Demand (COD) is a parameter used to determine the total amount of oxygen needed to chemically oxidize organic compounds. The concentration of COD in wastewater will increase as the content of organic compounds available in wastewater increases. High concentrations of COD can cause harm to the environment, as it can reduce the dissolved oxygen content in water (Harahap, et al., 2020). Based on the results of the study, in this photocatalysis process there was a decrease in COD parameters caused by the formation of hydroxyl radicals (•OH) that oxidized organic matter in the wastewater of fishery products. By Fig8 It can be seen that the highest percentage of exclusion in the photocatalysis process using ZnO is 31% found in ZnO with a mass of 1.5 g and an irradiation time of 8 hours at an irradiation intensity of 743±3.56 μW/cm^2, the initial COD concentration is 825 mg/L to 571.7 mg/L. While the lowest percentage of exclusion is 1% found in ZnO 1.5 g with an irradiation time of 8 hours and an irradiation intensity of 732±5.17 μW/cm^2, the initial COD concentration was 808.3 mg/L to 801.7 mg/L. The decrease in the percentage of removal in the photocatalysis process using ZnO was influenced by several factors, one of which was the turbidity of the sample so that it inhibited UV penetration in the wastewater of fishery products which made ZnO completely inactive.

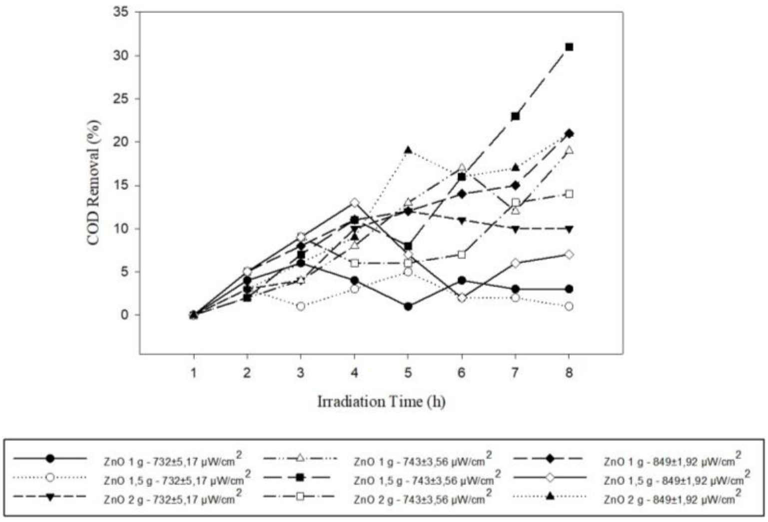

Figure 8: COD removal

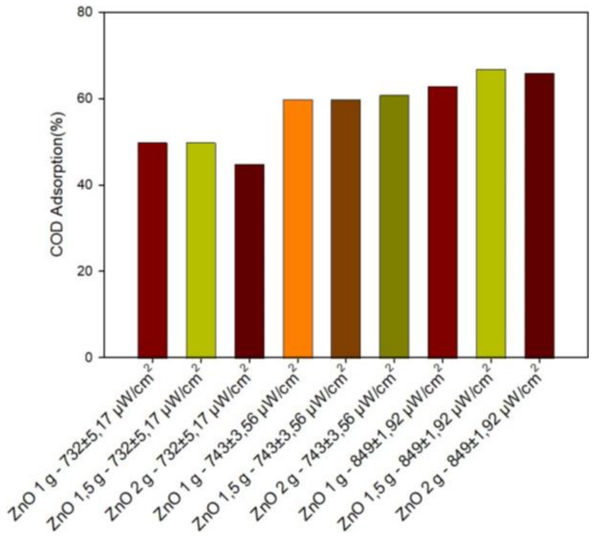

Figure 9: COD adsorption

By Fig9 It can be seen that there is a COD removal of 67% in the use of 1.5 g ZnO with an irradiation intensity of 849±1.92 µW/cm^2. The estimated percentage of COD removal due to the adsorption process has a larger removal than the photocatalysis process using ZnO which is only 31%. The adsorption process that occurs can result from a fairly large surface area of ZnO. The surface area of the ZnO catalyst used in this research is 611.827 m^2/g.

IV. CONCLUSION

The treatment of wastewater from fishery products using the ZnO photocatalysis process with catalyst mass variations of 1 g, 1.5 g and 2 g and irradiation intensity of 732±5.17 µW/cm^2, 743±3.56 µW/cm^2 and 849±1.92 µW/cm^2 resulted in a decrease in NH_4, BOD and COD parameters which were considered insufficiently effective in the degradation of organic compounds. This is due to the alleged adsorption process because the treatment of samples and catalysts without UV irradiation results in a significant decrease in NH_4, BOD and COD levels. The decrease in the content of these parameters is due to the fact that the organic compounds contained in wastewater are micro molecules with a relatively small size and the mass of ZnO used in this study is quite large, so that the availability of ZnO surfaces for adsorption is also quite high. The percentage of NH_4, BOD and COD exclusion using the photocatalysis process reached 35%, 27% and 31% respectively, while the suspected adsorption reached 82%, 79% and 67%.

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