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COMPARISON OF OZONATION (O_3) AND AOPs (O_3/H_2O_2) IN PURIFICATION OF WELL WATER IN KLUMPRIT VILLAGE, CENTRAL JAVA, INDONESIA

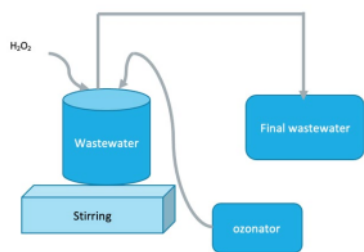
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Graphical abstract



Abstract

Water is a very important need for human life. The need for clean water has increased due to changes in people's behavior during the COVID-19 pandemic. Despite the increasing need for clean water during the pandemic, access to it remains unequal. One of the areas with poor water sources is Klumprit Village, Nusawungu, Cilacap, Central Java, where the water is cloudy, yellow in color, and a rustic smell. According to the Regulation of Ministry of Health of the Republic of Indonesia Number 32 of 2017, water that is suitable for sanitation needs is water that is odorless, tasteless, not cloudy or has a low level of turbidity. Ozone is an environmentally friendly technology because it does not produce harmful by-products. In addition to ozone, the production of clean water can use Advanced Oxidation Processes (AOPs) with (O_3/H_2O_2). The existence of H_2O_2 will increase the ability of ozone in the production of clean water or water purification. In this study, ozonation and AOPs (O_3/H_2O_2) were carried out to purify the yellow and cloudy well water in Klumprit Village, Cilacap, Central Java as an effort to improve public health during the COVID-19 pandemic. The research method was to compare ozonation and AOPs (O_3/H_2O_2). The study will include variations in pH levels, specifically neutral pH (untreated sample) and alkaline, as well as different concentrations of H_2O_2 . The experimental data included Fe and Mn content in water before and after the treatment process. Based on the results of this study, it was found that the yellow color of the groundwater in Klumprit village is caused by high levels of manganese (Mn). The lowest concentration of Mn was achieved after 60 minutes of ozonation at an alkaline pH of 9, with a concentration of 0.266 mg/L. The iron (Fe) content showed fluctuation for both treatment methods, ozonation (O_3) and advanced oxidation processes (O_3/H_2O_2).

Keywords: ozonation, AOPs, O_3/H_2O_2 , clean water, public health, covid-19

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1.0 INTRODUCTION

Humans really need water for their survival. About 68% of the human body consists of water, according to Rahayu and Gumilar (2017) [1]. During the COVID-19 pandemic, people's behavior has changed, leading to an increased need for clean water, such as frequent hand washing and bathing more than three times a day. Research conducted by the Indonesia Water Institute (IWI) between October 15 and November 20, 2020, found that the percentage of respondents who washed their hands more than ten times a day increased from 18% to 82%, while the percentage of those who showered more than three

times a day increased from 27% to 72%. The number of respondents who showered more than three times a day increased from 27% to 72%.

Despite the increasing need clean water has not been equally distributed. According to the Badan Pusat Statistik (BPS) [2], 9.79% of Indonesian households lacked access to clean water sources in 2020. The percentage was significantly higher in rural areas (17.26%) compared to urban areas (3.92%). The number of rural households that did not have access to clean water reached 17.26% compared to 3.92% in urban areas [2]. One of the areas with poor water sources is Klumprit Village, Nusawungu, Cilacap, Central Java, where the water is cloudy, yellow in color and smells of rust. The yellow color and odor of

the water indicate the presence of heavy metals such as Fe and Mn, as noted by Misa et al [3]. Organic substances that dissolve in water can undergo decomposition and decay, leading to water source contamination, as noted by Ningrum et al [4].

According to the Minister of Health of the Republic of Indonesia Number 32 of 2017, water that is suitable for sanitation needs to be odorless, tasteless, and not cloudy or have a low level of turbidity. The maximum allowable color content in water is 50 TCU, the maximum allowable turbidity is 25 NTU, the maximum allowable TDS is 1000 mg/l, the maximum allowable Mn content is 0.5 mg/l, and the maximum allowable Fe content is 1 mg/l [5].

Several methods have been used for water purification, such as chlorine, ozone, and others. Ozone is an environmentally friendly technology because it does not produce harmful by-products. According to research by Abdi et al [6] (2017), water treatment using ozone is quite effective in reducing the intensity of color from red to a much clearer state. Ozone, as a common oxidizing agent, has been applied in many fields in water and wastewater treatment [7]. Ozone is a strong oxidizing agent that has been widely used in water treatment as an oxidizing dye [8]. The advantage of ozone is that it has a fast reaction, and the final product, which is safe, is oxygen [9] (Summerfeilt, 2002). In addition to ozone, AOPs (O_3/H_2O_2) also could be used in the production of clean water. The existence of H_2O_2 increases the ability of ozone in the production of clean water or water purification. Chen and Wang [10] (2020) conducted research on wastewater treatment using ozonation with H_2O_2 . The results showed that the treatment using the ozonation process with H_2O_2 had the better result in degradation of ofloxacin compared to ozonation alone with degradation percentage was 55% in the ozonation process with H_2O_2 and 30% in ozonation alone.

Several studies have been conducted for the production of clean water or water purification, as follows: the first study was ozonation conducted by Al jibouri et.al. (2015) with the research using ozonation in treatment of water contaminated with methylene blue dye. The results showed that the dye content was reduced by 71% after the ozonation process. The second study was conducted by Chen and Wang [10] (2020) with the research was AOPs of O_3/H_2O_2 for ofloxacin wastewater. The result was using AOPs of O_3/H_2O_2 for ofloxacin wastewater, increases the efficiency by 25% compared to ozonation alone.

The ozonation process is a well-developed technology that has been widely applied throughout the world. Due to its strong oxidative properties, O_3 can be used in many fields, such as oxidation of inorganic/organic compounds and disinfection (or pathogen control) [11](Chen and Wang, 2019). Ozone is a strong oxidizing agent capable of reacting with many organic substances in water. During ozonation, a compound can be removed through direct oxidation by O_3 or indirect reactions with active radicals, such as OH produced by the decomposition of O_3 [11] (Chen and Wang, 2019). Today, ozone is still used for disinfection, but because of its superior oxidizing ability, it is also used for several other purposes, including taste and odor control, color removal, iron and manganese oxidation, turbidity removal, algae removal, enhancing biodegradability, and the oxidation of certain micropollutants [12].

AOPs have received a lot of attention in the field of water purification because of their potentially significant effectiveness and minimal adverse effects on water quality

[13]. The existence of H_2O_2 will increase the effectiveness of the ozonation process [14]. Chen and Wang (2020) [10] conducted a study using ozone- H_2O_2 in the treatment of liquid waste containing organic compounds. The presence of H_2O_2 will increase the formation of OH radicals, which are strong oxidizing agents.

H_2O_2 is an important oxidizing agent in industry usually use in various disinfection applications and bleaching processes in textile industries [15]. This compound can increase the direct decomposition of ozone into reactive hydroxyl radicals or enhance direct oxidation by molecular ozone [16]. As shown in Table 1, the hydroxyl radical (OH radical) has a high oxidation potential.

Table 1. Oxidation Potential [17]

Oxidizing Type	Oxidation potential, eV
Fluorine	3.06
Hydroxyl radicals (radikal OH)	2.80
Ozone	2.07
Hydrogen Peroxide	1.77
Chlorine	1.36

The previous research that has been carried out is water purification using only ozonation, AOPs of O_3/H_2O_2 for wastewater, so the innovation in this research will be AOPs of O_3/H_2O_2 for the purification of well water. The ozonation process is influenced by pH conditions [18], so in this study, the pH conditions will also be varied. Based on the presented background, this research aimed to carry out AOPs of O_3/H_2O_2 to purify the yellow and cloudy well water in Klumprit Village, Cilacap, Central Java, Indonesia, as an effort to increase public health during the COVID-19 pandemic.

2.0 METHODOLOGY

The study was using method of AOPs (O_3/H_2O_2) to purify the water sample. The variables in this study include control, dependent, and independent variables. The control variables, the variables that were kept constant in this study were the well water sample of Klumprit Village, the specification of the ozonator (ozonator capacity 400mg/hour). The ozone dosage used in this study was 400 mg/hour. The dependent variable in this study was the metal content of Fe and Mn. The independent variables in this study were pH (6.61 and 9) using H_2SO_4 and NaOH to reach the desired pH, H_2O_2 dosage (0; 2.5; 5; 7.5; 10% volume), and ozonation time (0, 15, 30, 45, 60 minutes). The materials used in this study were well water samples from Klumprit Village Indonesia, H_2O_2 , NaOH, HCl, and distilled water. The series of AOPs (O_3/H_2O_2) devices can be seen in Figure 1.

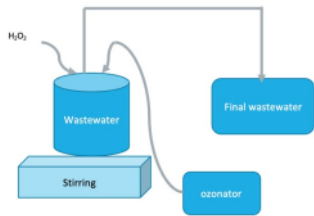


Figure 1. AOPs of O₃/ H₂O₂ Device Circuit

The results of this study were the metal content of Fe and Mn in water samples before and after processing. The metal content of Fe and Mn was analyzed using Atomic Absorption Spectrophotometry (AAS). The AAS instrument was adjusted followed by the gas was ignited as required in the test. The test method and cathode lamp were adjusted according to the test parameters; as follows, Mn=279.5 nm and Fe= 248,3 nm [29].

3.0 RESULTS AND DISCUSSION

3.1 Effect of Ozonation on Fe and Mn Levels in Well Water

The content of Fe metal in the well water sample before ozonation was below 1 mg/L, as per the standard set by the Ministry of Health Regulation No. 32 of 2017. This indicates that the yellow color and smell of rust in the well water in Klumprit village are not caused by the metal content of Fe. The removal of Fe levels by ozone can be seen in Figure 2.

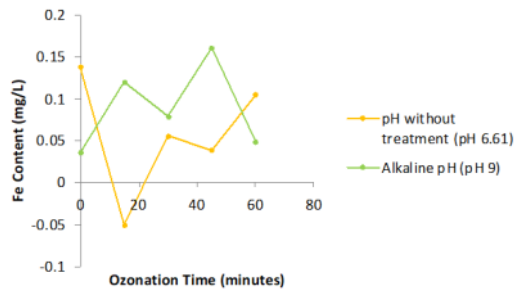


Figure 2. Effect of pH Variations on Fe Levels

The test results show fluctuating data for the Fe levels, which can be attributed to compounds that bind to amphoteric compounds, meaning they can act as acids or bases depending on the reaction conditions [19]. The contact time of ozonation does not affect the removal of iron (Fe) by ozone because ozone is a strong oxidant that rapidly oxidizes iron (II) to iron (III) [20]. However, when the hardness in groundwater increases, iron oxidation decreases because ozonation can form other organic compounds that may interfere with Fe removal [20, 21].

Figure 2 shows the results of the Fe removal test in well water samples at different pH levels and ozonation times. The optimal time for Fe removal in untreated samples is 45 minutes, with a removal result of 0.039 mg/L, meeting the water quality standards in Permenkes No. 32 of 2017. However, there was a measurement of Fe content of -0.05 mg/L at the 15th minute, indicating a concentration lower than the detection limit of the device used [22, 23]. While the

removal of Fe in samples with alkaline pH, the optimal results of Fe removal after ozonation were shown at a time of 60 minutes with a Fe content of 0.049 mg/L. It is also seen that the smallest value of Fe concentration in the alkaline pH sample is at 0 minutes of ozonation with a Fe concentration of 0.036 mg/L. The increase in Fe concentration after ozonation from its original value before ozonation is possible due to the addition of Fe which is formed as a by-product of bacterial metabolism that may be present in the water [24].

Overall, the final results of the well water samples after ozonation showed that the Fe content had met the standards of Indonesian Government, which is below 1 mg/L. The best Fe removal results were observed at 45 minutes of ozonation under alkaline pH conditions, with a removal rate of 71.74%. These findings suggest that higher pH levels during ozonation lead to greater levels of Fe removal in the sample. Previous studies have shown that under alkaline pH conditions, iron (II) is oxidized to iron (III), which subsequently forms iron hydroxide [25]. During the ozonation process, high pH levels are associated with ozone decomposition, leading to the generation of hydroxyl radicals (OH⁻), which are highly reactive and react with iron [26]. At lower pH levels, the Fe²⁺ content is still oxidized by ozone, but the rate of ozone decomposition is slower, resulting in slower formation of Fe(OH)₃ than at alkaline pH [19].

The metal content of Mn in the well water sample before ozonation was 3.293 mg/L, which exceeds the quality standards of Indonesian Government, with a maximum allowable level of 0.5 mg/L. This indicates that the yellow color and smell of rust in the well water in Klumprit village may be caused by the high content of Mn metal. The removal of Mn levels by ozone can be seen in Figure 3

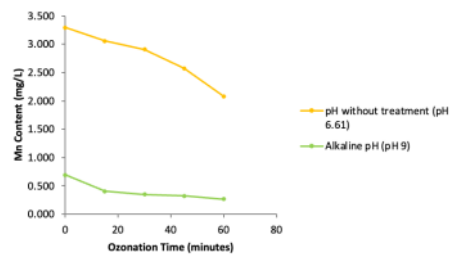


Figure 3. Effect of pH Variations on Mn Levels

In contrast to the data from the removal of Fe, the results of the removal of Mn in well water samples at pH without treatment and at alkaline pH both showed a significant decrease. It can be seen in Figure 3. that the removal curve decreases with increasing ozonation time. The Mn content in the untreated sample decreased from 3.293 mg/L to 2.080 mg/L after 60 minutes of ozonation, indicating that the longer the contact time, the higher the ability of ozone in degrading Mn. The best decrease in Mn levels was obtained at 60 minutes of ozonation at alkaline pH. The level of Mn at minute 0 was 0.693 mg/L decreased to 0.266 mg/L at minute 60 with a percentage allowance of 61.62%, so it was able to meet the quality standards of Indonesian Government. pH and time of ozonation affect the ability of ozone in degrading Mn. Manganese is oxidized rapidly by ozone in the absence of organic matter, and the rate of this reaction increases with increasing pH [27].

3.2 Effect of AOPs (O_3/H_2O_2) on Fe and Mn Levels of Well Water

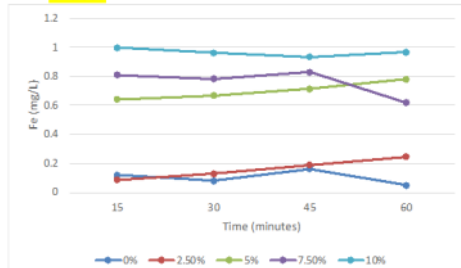


Figure 4. Effect of H₂O₂ concentration Variation on Fe Content

Based on the test results shown in Figure 4, the average shows an increase in both the addition of H₂O₂ and every increase in ozonation time. This is presumably due to the addition of H₂O₂, which can result in the percent reduction in iron not being optimal. In Fenton's reagent, if the water contains iron (II), then H₂O₂ is added, and the two substances will react to form Fe (III), OH⁻, and OH. If there is excess H₂O₂, then the OH radicals formed will react with H₂O₂ and produce HO₂, so that OH radicals are no longer effective in breaking the complex bonds of iron with organic substances [28].

Figure 4 shows the results of the removal of Fe content, where the H₂O₂ concentration is 2.5%. The best Fe removal mass is in the 15th minute with a Fe content of 0.085 mg/L, at the 30th minute the Fe content is 0.129 mg/L, at 45 minutes the Fe content was 0.188 mg/L, and at 60 minutes the Fe content was 0.246 mg/L. Then, at 5% H₂O₂ concentration, at 15 minutes the Fe content was 0.641 mg/L, at 30 minutes it was 0.668 mg/L, at 45 minutes it was 0.714 mg/L, and at 60 minutes it was 0.780 mg/L. While the percentage of H₂O₂ concentration is 7.5% by mass, at 15 minutes the Fe content is 0.81 mg/L, at 30 minutes it is 0.782 mg/L, at 45 minutes it is 0.828 mg/L, and it decreases at 60 minutes to 0.619 mg/L. And at the 10% mass H₂O₂ content, in the 10th minute, the Fe content was 0.998 mg/L, at the 30th minute it was 0.961 mg/L, at the 45th minute it was 0.932 mg/L, and at the 60th minute it was 0.967 mg/L.

From the data seen above, the ozonation process in the well water sample is quite influential in the reduction of Fe metal content. There was a decrease in the Fe content of the well water sample from 0.138 mg/L to 0.049 mg/L. However, in the AOPs (O_3/H_2O_2) process, where there is the addition of H₂O₂ in the ozonation process, it causes an increase in the content of Fe metal in the well water sample. At 2.5%-10% H₂O₂ concentration variation, each variation and ozonation time increased significantly.

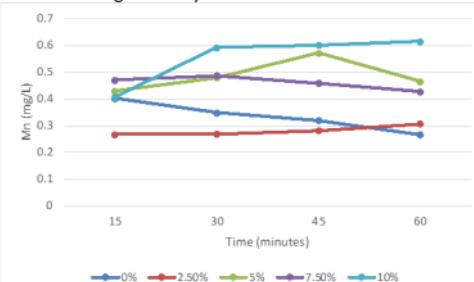


Figure 5. Effect of H₂O₂ concentration Variation on Mn Content

Based on Figure 5, it can be seen that Mn produces fluctuating data. This is presumably due to the presence of compounds that bind to amphoteric compounds, which can act as both acids and bases depending on the conditions of the solution being reacted with [19].

Figure 5 shows data on the results of Mn removal in water samples. When the percentage of H₂O₂ mass was 2.5%, the best Mn removal was achieved in the 15th minute, with a remaining Mn content of 0.267 mg/L. The levels increased at the 30th minute, where the Mn content remaining was 0.268 mg/L, and at the 45th minute, the Mn content increased to 0.281 mg/L. At the 60th minute, the Mn content was 0.301 mg/L. For the 5% mass H₂O₂ concentration, the results were fluctuating. The best results were achieved at 15 minutes of ozonation, with a remaining Mn content of 0.430 mg/L. Then, the levels increased at 30 minutes to 0.480 mg/L, increased again at 45 minutes to 0.572 mg/L, and decreased at 60 minutes to a remaining Mn content of 0.466 mg/L. At 7.5% mass percentage, the Mn content showed an increase over time. The content was 0.471 mg/L at 15 minutes, 0.487 mg/L at 30 minutes, 0.459 mg/L at 45 minutes, and 0.428 mg/L at 60 minutes. At 10% mass percentage, the data showed Mn content of 0.407 mg/L at 15 minutes, 0.593 mg/L at 30 minutes, 0.601 mg/L at 45 minutes, and 0.616 mg/L at 60 minutes. Based on the data above, it can be concluded that the higher the percentage of H₂O₂ concentration used, the smaller the Mn removal rate. However, the resulting level is <0.5 mg/L, which is still considered small and safe, and does not cause health problems.

4.0 CONCLUSION

Based on the results of this study, it was found that the yellow color of the groundwater in Klumprit village is caused by high levels of manganese (Mn). The lowest concentration of Mn was achieved after 60 minutes of ozonation at an alkaline pH of 9, with a concentration of 0.266 mg/L. The iron (Fe) content showed fluctuation for both treatment methods, ozonation (O_3) and advanced oxidation processes (O_3/H_2O_2).

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