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Performance Analysis of Ozonation on Sludge Filterability of Coal Mine Runoff

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ABSTRAK

Air limpasan tambang batu bara mengandung padatan tersuspensi tinggi dan koloid persisten yang menghambat pemisahan padat–cair serta menyebabkan rendahnya *sludge filterability*. Penelitian ini mengevaluasi ozonisasi sebagai pra-pengolahan untuk meningkatkan *sludge filterability* melalui modifikasi stabilitas koloid dan pembentukan flok yang lebih kompak. Eksperimen laboratorium menggunakan generator ozon berkapasitas 3 g/jam dengan dosis ozon 0; 1,2; 1,5; 2; 3; dan 6 g/jam/L selama 60 menit. Lima sampel lumpur sintetis dengan total padatan tersuspensi (TSS) 3.000–50.000 mg/L disiapkan untuk merepresentasikan variasi kondisi lapangan. pH dan *Oxidation-Reduction Potential* (ORP) dipantau selama ozonisasi dan filtrasi vakum. Ozonisasi menggeser nilai ORP dan sedikit menurunkan pH, yang menandakan oksidasi parsial dan destabilisasi partikel koloid. Dosis moderat (1,2–3 g/jam/L) menggeser zeta potensial mendekati netral sehingga meningkatkan kekompakan flok dan kinerja filtrasi, sedangkan oksidasi berlebih pada 6 g/jam/L melepaskan fragmen humat dan koloid halus yang menurunkan kemampuan *dewatering*. Dosis optimum adalah 1,2 g/jam/L pada lumpur dengan TSS sekitar 10.000 mg/L, yang menurunkan kekeruhan filtrat sebesar 86,87% dan waktu filtrasi sebesar 34,53%, dengan pH yang stabil dan peningkatan ORP yang moderat. Temuan ini menunjukkan bahwa ozonisasi terkendali efektif meningkatkan *sludge filterability* air limpasan tambang batu bara dan menawarkan pendekatan yang menjanjikan untuk pengelolaan air limbah yang lebih efisien di sektor pertambangan.

***Kata kunci*:** Advanced Oxidation Process (AOP), Air limpasan tambang batu bara, Oxidation-Reduction Potential (ORP), Ozonisasi, Sludge filterability

ABSTRACT

Coal mine runoff contains high suspended solids and persistent colloids that hinder solid–liquid separation and result in poor sludge filterability. This study evaluated ozonation as a pre-treatment to enhance sludge filterability by modifying colloidal stability and promoting the formation of more compact flocs. Laboratory experiments used an ozone generator with a capacity of 3 g/h at ozone doses of 0, 1.2, 1.5, 2, 3, and 6 g/h/L for 60 minutes. Five synthetic sludge samples with total suspended solids (TSS) of 3,000–50,000 mg/L were prepared to represent varying field conditions. pH and Oxidation-Reduction Potential (ORP) were monitored during ozonation and vacuum filtration. Ozonation shifted ORP values and slightly reduced pH, indicating partial oxidation and destabilization of colloidal particles. Moderate doses (1.2–3 g/h/L) shifted the zeta potential toward neutral, thereby enhancing floc compactness and filtration performance, whereas excessive oxidation at 6 g/h/L released humic fragments and fine colloids that impaired dewatering. The optimum dose was 1.2 g/h/L for sludge with approximately 10,000 mg/L TSS, which reduced filtrate turbidity by 86.87% and filtration time by 34.53%, with stable pH and a moderate increase in ORP. These findings demonstrate that controlled ozonation effectively improves the sludge filterability of coal mine runoff and offers a promising approach to more efficient wastewater management in the mining sector.

***Keywords*:** Advanced Oxidation Process (AOP), Coal mine runoff, Oxidation-Reduction Potential (ORP), Ozonation, Sludge filterability

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1. INTRODUCTION

Mining activities are a strategic sector in global economic development. According to the World Mining Data report (2023), global mineral production reaches more than 17 billion tons per year, encompassing primary commodities such as coal, metal ores, and other industrial minerals. This sector plays a crucial role in providing raw materials for the

energy, construction, and technology industries, thus supporting infrastructure progress and global industrialization. On the other hand, mining activities also have a serious impact on the environment, particularly through the generation of large amounts of waste that pollutes land, air, and water.

Within this sector, coal mining is particularly important for Indonesia. As the world's third-largest coal producer after China and India, Indonesia recorded coal production of 687 and 775.2 million tons in 2022 and 2023, respectively (Ministry of Energy and Mineral Resources, 2024). This sector's contribution to Indonesia's Gross Domestic Product (GDP) reached more than 9% in 2022 (Badan Pusat Statistik, 2023). Despite its role in meeting national energy needs and exports, mining activities also have environmental impacts.

Coal mining activities naturally produce various wastes, including solid waste in the form of overburden and tailings, gaseous waste in the form of dust and greenhouse gases, and liquid waste. One of the most common forms of liquid waste is mine runoff, a surface flow generated by rainfall that hits open mining areas. Rainwater dissolves material from open mining areas, carrying sediment, heavy metals, and dissolved chemicals from the mining process, which can pollute the environment if not managed properly (Bian et al., 2010). Coal mine runoff generally contains contaminants such as Total Suspended Solids (TSS) at concentrations that can exceed 1000 mg/L, heavy metals such as Fe (up to 50 mg/L) and Mn (10–15 mg/L), as well as low pH, depending on the rock characteristics and hydrological conditions of the mining area (O'Sullivan, 2005). If this waste is not properly treated, it will certainly cause water pollution, eutrophication, and the death of aquatic life. Humans will also suffer from skin irritation, kidney damage, and digestive problems due to consuming water contaminated with mining wastewater, especially heavy metals.

Most mining companies have implemented runoff water treatment systems, either actively using chemicals to neutralize and precipitate heavy metals, or passively using settling ponds, constructed wetlands, or vegetative systems. However, these treatment processes generally produce large amounts of sludge, which is often difficult to treat. This sludge is sticky, contains fine colloidal particles, and has a high water content, making the dewatering process inefficient and expensive (Madzivire et al., 2011). Consequently, poorly managed sludge can cause secondary pollution, increase storage space requirements, and increase mining companies' operational costs.

The sludge produced by mine runoff treatment, with these characteristics, presents unique challenges in the dewatering process. Inefficient dewatering will increase waste volume, increase management costs, and pose a risk of further pollution. The main problem in sludge management is low filterability, namely the ability of the sludge to be filtered and separated from water. Low filterability is caused by the high stability of the colloidal system in the sludge, which prevents particles from aggregating and forming large, easily settleable flocs. Technical parameters directly related to this phenomenon are pH and Oxidation Reduction Potential (ORP), which affect zeta potential. Zeta potential reflects the electrical charge on the particle surface, where values approaching zero indicate decreased colloidal stability, making particles more likely to clump and settle (Yu et al., 2019). In coal mine runoff sludge, ozone is also expected to oxidize dissolved Fe^{2+} and Mn^{2+} into insoluble precipitates such as $Fe(OH)_3$ and MnO_2 (Gunten, 2003). These precipitates may act as in-situ coagulants that destabilize colloids and promote floc formation; therefore, this study hypothesizes that ozonation improves sludge filterability, as indicated by shorter filtration time and lower filtrate turbidity.

To address this problem, most mining companies typically employ a sludge conditioning process aimed at increasing the efficiency of solids and water separation. One commonly used method is coagulation-flocculation, using a combination of chemicals such as $FeCl_3$ and polyacrylamide (PAM). Research by Zhai et al. (2012) showed that the optimal dosage of $FeCl_3$ and PAM was able to reduce the Specific Resistance to Filtration (SRF) by more than 90%, indicating a significant improvement in dewatering performance. Furthermore, the approach of using fly ash as a conditioning agent was also proven effective.

A study by Lee et al. (2010) reported that the addition of fly ash up to 50% dry weight can reduce the sludge water content from 80% to 29%, as well as reduce the filtration resistance from 2.9×10^{14} to 2.9×10^{13} kg/m. Another study by Liu et al. (2014) also showed that the addition of 20–30 g of fly ash per 100 mL of sludge reduced the specific resistance from 10^{13} to 10^{10} m/kg, thereby significantly increasing the sludge filtration rate. On the other hand, physical approaches such as thermal conditioning also provide promising results. For instance, Han et al. (2016) showed that the combination of $FeCl_3$ and steel slag at a temperature of 52°C can reduce the Capillary Suction Time (CST) value to 7.8 seconds, as well as improve the floc structure based on SEM observations. While these various methods are effective in improving dewaterability, each has limitations: the use of chemicals increases secondary sludge production, fly ash increases the volume of solid residue, and thermal conditioning requires high energy consumption. Therefore, exploration of alternative technologies that are more technically efficient, economically feasible, and environmentally sustainable for sludge conditioning prior to dewatering is still needed.

One promising method is ozonation, which uses ozone as a strong oxidizing agent in the sludge conditioning process. Ozone has an oxidation-reduction potential (ORP) of +2.07 V, making it one of the most reactive oxidizers used in water and

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wastewater treatment. Ozone works by attacking organic bonds and disrupting the polymeric structure of sludge components, increasing the tendency of particles to flocculate and separate from water (Gottschalk et al., 2009).

Technically, ozonation can affect critical parameters related to colloidal stability, namely pH and ORP. According to a study by Yu et al. (2019), ozonation lowers pH by producing organic acids, such as carboxylic acids, from the oxidation of organic compounds. A decrease in pH combined with an increase in ORP creates chemical conditions that favor colloid destabilization. This results in the zeta potential approaching zero, favoring the formation of larger, denser, and more easily separated flocs. Thus, ozone functions not only as an oxidizing agent but also as a trigger for changes in the chemical environment that directly contribute to increased sludge filterability.

Various studies have shown that ozonation is effective in improving sludge dewatering performance, particularly for domestic and industrial wastewater sludge. Furthermore, ozone offers several advantages over conventional chemicals. Unlike heavy metal coagulants, which can produce secondary sludge and leave harmful residues, ozone naturally decomposes into oxygen after reaction, thus not adding new pollutant loads to the system (Gunten, 2003). In addition, ozone is produced on-site through an ozone generator, eliminating the need for transportation and storage of hazardous or corrosive chemicals. From a sustainability perspective, ozonation technology aligns with the principles of green chemistry and supports the transition to sustainable mining, which is increasingly becoming a focus within environmental regulatory frameworks in many countries, including Indonesia.

However, most research related to ozonation is still limited to sludge generated from the treatment of domestic, textile, and food industry wastewater. Studies on the application of ozone to sludge from coal mine runoff treatment, particularly in Indonesia, are still very rare. This is despite the fact that the characteristics of mine sludge differ significantly from those of domestic wastewater sludge, including lower pH and heavy metal content. Therefore, an in-depth scientific study is needed to test the effectiveness of ozonation in the context of mine runoff sludge, particularly regarding changes in pH and ORP parameters, and their correlation with increased sludge filterability.

Considering the potential of ozonation and the need for more environmentally friendly solutions in mine sludge management, this study aims to evaluate the effect of varying ozone dosage on the sludge filterability of coal mine runoff sludge. The focus of the study is to analyze changes in pH and ORP values as key indicators of the mechanism of colloid destabilization and increased dewaterability. The results of this study are expected to make a significant scientific contribution to the development of effective mine wastewater treatment technologies.

2. METHODOLOGY

The experimental methodology of this study is illustrated in Figure 1.

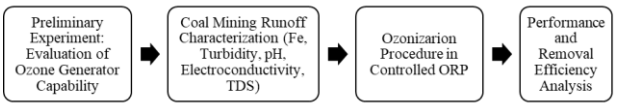


Figure 1. Schematic Method of This Study

Coal mine runoff samples were collected from four sampling points (coded D, E, F, and G) at a coal mining site in Kalimantan, Indonesia, and stored in closed containers prior to analysis. The initial characterization included TSS (gravimetric method), pH, and ORP, measured using a multiparameter meter. To obtain controlled variations in solids loading, the samples were diluted with tap water to prepare five engineered samples (M1–M5) with TSS concentrations of approximately 50,000, 30,000, 20,000, 10,000, and 3,000 mg/L.

Ozonation was carried out in a laboratory-scale batch reactor. Ozone was produced by an ozone generator at a constant output of 3 g/h and introduced at the bottom of the reactor through a gas diffuser. Ozone dose variations of 6, 3, 2, 1.5, and 1.2 g/h/L were obtained by treating sample volumes of 0.5, 1.0, 1.5, 2.0, and 2.5 L, respectively, under the same contact time, while an untreated sample served as the control (blank). ORP and pH were recorded periodically during ozonation.

After ozonation, sludge filterability was evaluated by vacuum filtration of the same volume of each sample through filter paper placed in a Büchner funnel under identical vacuum conditions. Filtration time was recorded as the time required to filter the entire sample volume, and filtrate turbidity was measured using a turbidimeter. The pH and ORP were also measured before and after filtration. Differences among ozone doses were evaluated using single-factor ANOVA.

3. RESULTS AND DISCUSSION

3.1. Characteristics of Coal Mine Runoff Water Samples

Mine runoff is a type of wastewater with complex physical and chemical characteristics. Runoff is surface flow caused by rainfall or water flowing over mining areas. The runoff sludge samples used in this study came from a mine site in Kalimantan and were sent to the laboratory of the Department of Civil and Environmental Engineering, Gadjah Mada University, for further analysis.

Prior to ozonation treatment, several basic water quality parameters were measured to determine the sample's initial condition. These parameters included

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pH, TSS, and ORP. These measurement results served as the basis for designing further treatment and as a reference for evaluating the effectiveness of the ozonation process on the physical characteristics of the runoff sludge. The received runoff samples, coded as D, E, F, and G, came from Slurry Pump A, Slurry Pump B, Dragflow D2, and Dragflow RTM, respectively. The initial measurement data for the runoff samples are presented in Table 1.

Based on Table 1, the coal mine runoff samples coded D, E, F, and G represent four sample groups that have been diluted with tap water. The pH and ORP values were obtained from the average of seven sample variations: D1-D7, E1-E7, F1-F7, and G1-G7. Furthermore, the TSS value is the average of three repeated measurements for each group without dividing the sample variations. In this case, all samples had pH values within the quality standard range, namely 6-9. The average values for samples D and E were slightly acidic, at 6.1 and 6.05, respectively. Samples F and G were closer to neutral to slightly alkaline, at 7.12 and 7.31, respectively. This indicates variations in chemical conditions at the runoff intake locations.

Meanwhile, the TSS values for all samples far exceeded the quality standard of 400 mg/L. The average TSS levels for samples D and E were 86,540 mg/L and 88,233 mg/L, respectively, while those for F and G reached 123,087 mg/L and 533,100 mg/L. These very high TSS levels indicate that runoff from the mining area carries a large suspended solids load, potentially complicating further treatment and impacting the quality of the receiving water body if not properly managed. The differences between samples also reinforce the rationale for TSS engineering in this study, to achieve more controlled variations in sludge concentration for ozonation treatment.

The average ORP values measured across the four samples ranged from 279.48 mV to 311.86 mV, with the highest value in sample E and the lowest in sample G. The ORP was in the positive range, indicating moderate oxidative activity. This ORP value serves as a benchmark for assessing changes in oxidation levels after ozonation treatment.

Then, to obtain controlled variations in TSS levels in the study, a TSS concentration engineering process was carried out using the dilution method. The runoff water samples were diluted with tap water using a V2:V1 ratio of 10:1, namely 1 part of the original sample mixed with 9 parts of tap water to produce five new sample groups, namely M1, M2, M3, M4, and M5. The TSS target for each sample was determined as follows.

Table 2 shows the characteristics of the TSS-treatment samples. All samples remained within the pH standard, with a tendency for the pH to increase as the TSS concentration decreased. The initial ORP values varied but remained within the positive range, reflecting the oxidative conditions prior to ozonation.

The ozone dose variation in this study was engineered by adjusting the sample volume to be ozone-treated, while the ozone injection rate from the generator was maintained at a constant 3 grams per hour. This approach is based on the principle that ozone dose is a critical parameter describing the amount of ozone (oxidizing agent load) delivered to the system per unit time and per unit solution volume. The ozone dose (g/h/L) is highly dependent on two main variables: the ozone production rate and the sample volume.

By maintaining a constant ozone injection rate, adjusting the sample volume directly changes the ozone concentration received per unit volume of water. A smaller sample volume causes the ozone produced over an hour to accumulate in a smaller reaction chamber, resulting in a higher ozone dose. This is the basis for adjusting the ozone dose variation through solution volume manipulation. The ozone dose concentration variation is shown in Table 3.

Table 1. Initial Characteristics of Coal Mine Runoff Samples

Runoff Sample	TSS (mg/L)		pH		ORP (mV)	
	Quality Standard	Test Result	Quality Standard	Test Result	Quality Standard	Test Result
D	400	86,540	6 – 9	6.11	*	308.62
E	400	88,233	6 – 9	6.05	*	311.86
F	400	123,087	6 – 9	7.12	*	285.48
G	400	533,100	6 – 9	7.31	*	279.48

Table 2. Characteristics of Runoff Water Samples After TSS Treatment

Runoff Sample	TSS (mg/L)		pH		ORP (mV)	
	Quality Standard	Test Result	Quality Standard	Test Result	Quality Standard	Test Result
M1	400	50,000	6 – 9	7.60	*	260.67
M2	400	30,000	6 – 9	7.78	*	334.00
M3	400	20,000	6 – 9	7.26	*	263.67
M4	400	10,000	6 – 9	7.52	*	340.00
M5	400	3,000	6 – 9	8.23	*	321.00

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Scientifically, the values expressed in g/h/L should therefore be interpreted as nominal volumetric ozone loading rates calculated from the generator output divided by the treated liquid volume, rather than as directly measured transferred or absorbed ozone doses. Changing sample volume can also alter liquid depth, bubble residence time, and gas–liquid contact conditions; consequently, ozone transfer efficiency may not be identical among treatments. The volume-based approach was used as a controlled laboratory screening method to compare oxidant loading per unit liquid volume at a constant nominal generator output, while this mass-transfer limitation should be considered when extrapolating the results or comparing them with systems operated at a constant reactor volume.

Table 3. Variation of Ozone Dose Concentration

Sample Volume (L)	Ozone Rate (g/h)	Ozone Dose (g/h/L)
Blank	0	0
0.5	3	6
1.0	3	3
1.5	3	2
2.0	3	1.5
2.5	3	1.2

The ozone dosage variations used in this study ranged from low to high to identify the optimum dosage that provided the best performance in the solid-liquid separation process. Previous research has shown that excessively high ozone dosages can cause over-oxidation, which actually worsens floc quality and slows down the filtration process (G. Erden and A. Filibeli, 2010).

3.2. ORP Change Profile During the Ozonation Process Series

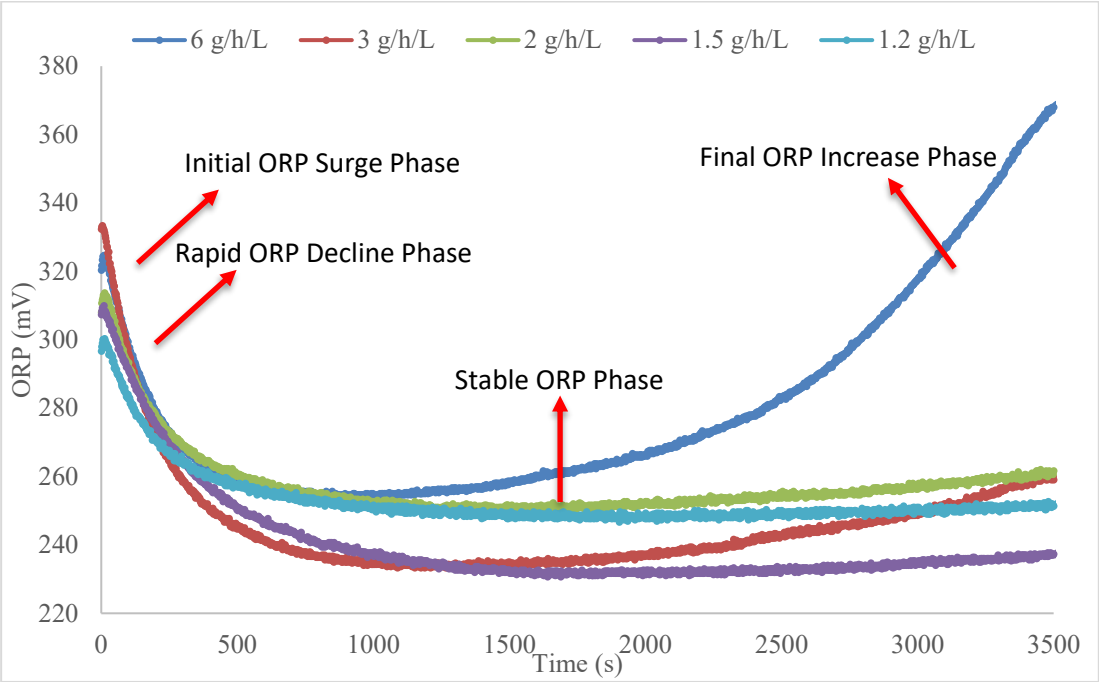
Oxidation-Reduction Potential (ORP) is an important indicator that reflects the redox state within a system and can be used as an indirect proxy for the presence of dissolved ozone and the dynamics of oxidation reactions during wastewater treatment. In this study, ORP was monitored continuously for 60 minutes at two-second intervals to fully capture the dynamics of the ozonation reaction occurring in the coal mine runoff sludge system. This duration was chosen because the ozonation reaction is not linear and consists of several interrelated kinetic stages, including a rapid reaction phase, a decline and stabilization phase, and a final oxidant accumulation phase. By monitoring ORP for an hour, these change patterns can be mapped in detail, revealing when ozone is actively adsorbed by contaminants, when the system begins to stabilize, and when ozone begins to accumulate.

Changes in ORP values within the system are also closely related to particle destabilization in the sludge, a prerequisite for improving sludge filterability. Ozone, as a strong oxidizing agent, has the ability to alter the surface charge of suspended particles, break down colloidal organic compounds, and deconstruct polymeric compounds that maintain suspension stability. This process will cause a decrease in the zeta potential and weaken the repulsive forces between particles, thus encouraging clumping. Figure 2 displays a graph of changes in ORP values during the ozonation process.

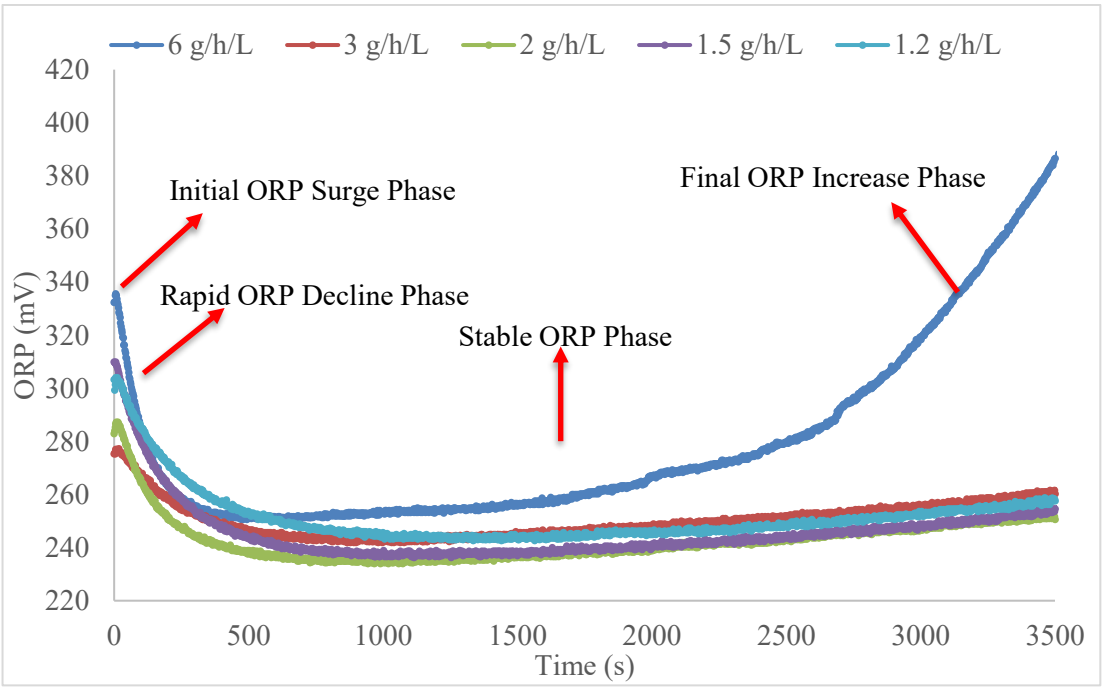
A closer look at graphs M1–M5 (Figure 2) reveals a spike in ORP values within the first 0–60 seconds after ozone injection, even though the initial ORP value was already in an oxidative state. This initial spike is likely due to ozone's high reactivity as a strong oxidizing agent, rapidly reacting with reduced compounds in water (Gottschalk et al., 2009). This rapid oxidation reaction produces a spike in ORP due to the release of electrons from the reduced substance to the oxidizing agent, thereby strengthening the redox state of the system. This reflects the initial characteristics of the ozonation process, which is highly reactive to easily oxidized contaminants.

The ORP spike indicates that ozone can cause a rise in ORP values within seconds when reacting with reduced metal ions or simple organic compounds. This increase occurs before ozone decomposes or reacts further with complex contaminant compounds. Ozone, with a redox potential of +2.07 V, tends to react readily with reductive wastewater components, resulting in a rapid and significant ORP response.

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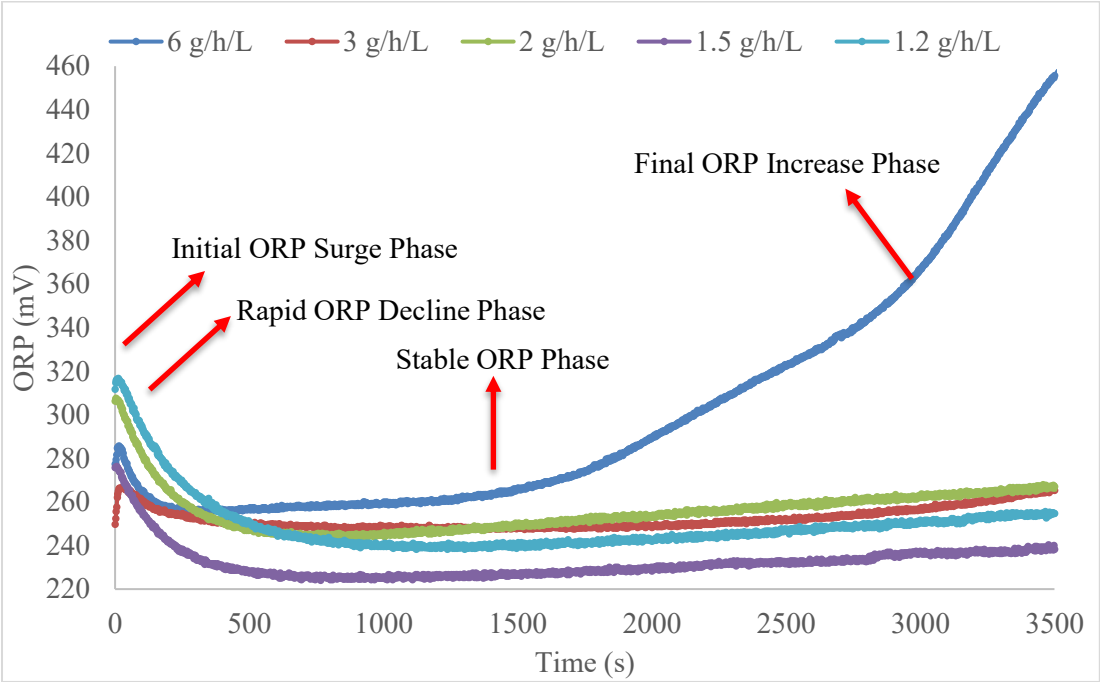


(a)

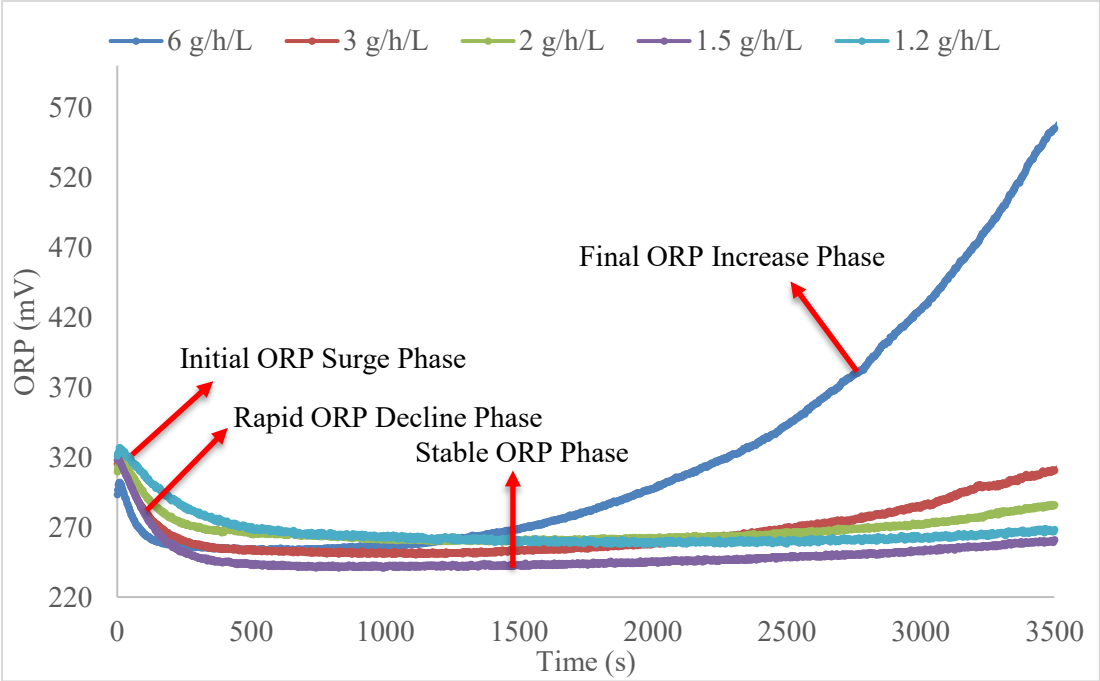


(b)

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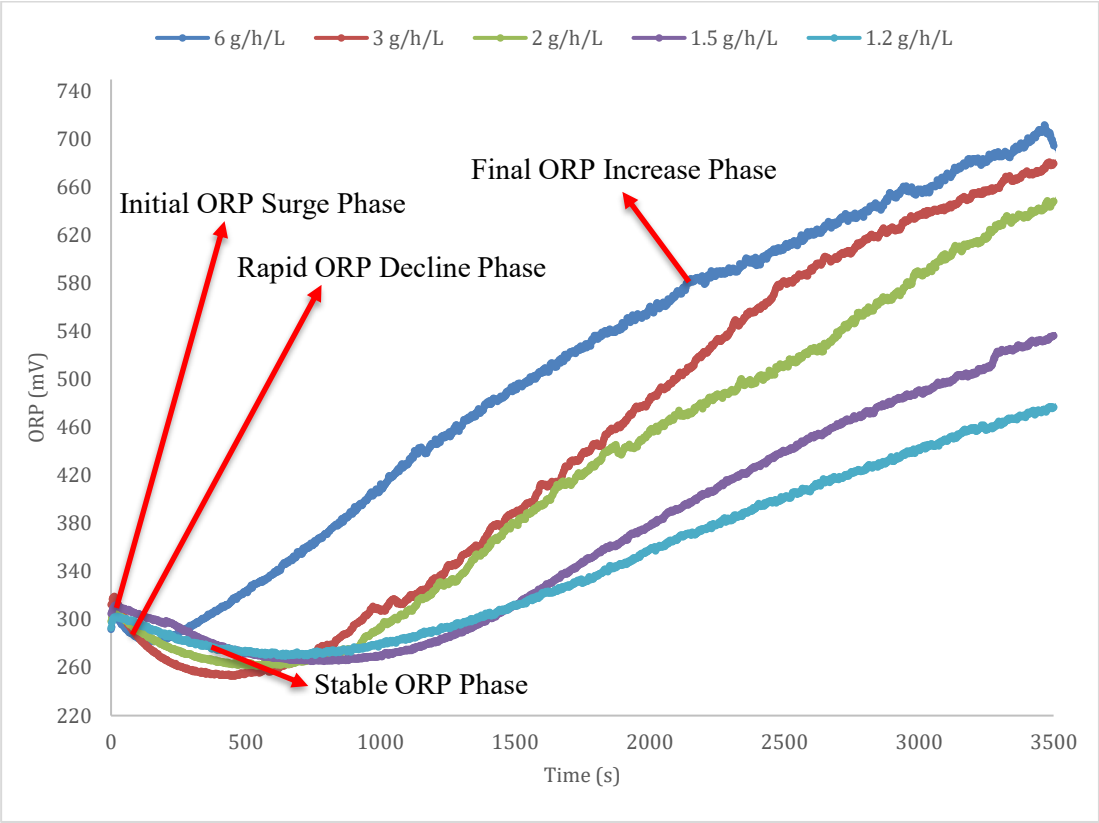


(c)



(d)

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(e)

Figure 2. ORP Variation Profile During the Ozonation Process for Samples (a) M1, (b) M2, (c) M3, (d) M4, and (e) M5

After the initial spike, the ORP values across all samples tended to decrease and then entered a stable phase. This decrease was caused by two main factors. First, ozone was consumed by contaminants in the system. Oxidation reactions proceed intensively and form oxidation products, especially in the initial phase, when pollutants such as metals and organic compounds are still quite high. Second, there was a decrease in system reactivity. After easily oxidized contaminants were depleted, only more stable compounds, or flocs, that are less reactive to ozone, remained. This phase, as explained by Gottschalk et al. (2009), is referred to as the redox steady-state phase, when ozone enters the system and reacts immediately, preventing ozone accumulation in the liquid phase.

The increase in ORP values again at the end of the ozonation phase indicates that some ozone is no longer consumed by pollutants because the pollutant load has decreased. The accumulation of ozone in the liquid phase causes the ORP value to gradually increase, eventually becoming significant. Gottschalk et al. (2009) emphasized that in semi-batch systems, such as the one in this study, the increase in ORP that occurs at the end of the process often reflects the accumulation of unreacted ozone, especially when the ratio between ozone dose and pollutant mass is too high. This finding is further supported by the findings of Beltran (2003), who stated that in wastewater with low TSS content, ozone is more difficult to consume efficiently, resulting in its accumulation in the system, causing the ORP value to increase.

As shown in the Figure 2(a), the initial ozonation process in sample M1 showed a sharp decrease in ORP after an initial spike at all doses in the range of 296-332 mV, reaching 230-253 mV. Then, at 500-1000 seconds, a stable phase occurred for all doses in sample M1. After passing through the stable phase, the ORP graph for sample M1 at ozone doses of 6 and 3 g/h/L increased to 374 and 261 mV, respectively. Ozone doses of 2-1.2 g/h/L showed a slow and low increase in ORP.

The ORP graph for sample M2 (Figure 2 (b)) shows a sharp decline in all curves, with initial values ranging from 275-332 mV to 234-250 mV. After reaching a low point, the curves for each ozone dose stabilized between 500-1500 mV. Between 1500-3600 mV, the ORP increased, varying between ozone doses. The ozone dose of 6 g/h/L showed the sharpest increase in ORP, reaching 407 mV. Meanwhile, ozone doses of 3-1.2 g/h/L produced a relatively slow increase in ORP, resulting in a low final value.

Furthermore, the ORP graph for sample M3 (Figure 2 (c)) also exhibited a similar curve pattern, with a fairly sharp decline. Initial ORP values ranged from 249-311 mV, dropping to 224-255 mV. After reaching a low point, the ORP values tended to stabilize between 500-1500 mV. Entering the final phase of ozonation, which saw an increase in ORP values, the ozone dose of 6 g/h/L showed the highest

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increase, reaching 469 mV. Ozone doses of 3-1.2 g/h/L showed a slow and low increase in ORP.

Sample M4 (Figure 2 (d)) then showed an ORP graph with a curve showing a decrease in ORP values from 193-321 mV to 241-259 mV. After reaching its lowest point, the entire curve entered a stable phase between 250-1500 seconds, during which the ORP values changed very slowly. The duration of this stable phase was relatively uniform across all ozone dose variations. Entering the final phase of ozonation, between 1500-3600 seconds, the ORP values showed a striking difference between the ozone doses. The ozone dose of 6 g/h/L, the highest dose, experienced a drastic increase in ORP values between 1500-3600 seconds, reaching 582 mV. While the ozone dose of 3-1.2 g/h/L indicates the ORP increase was much slower and lower.

Furthermore, sample M5 (Figure 2 (e)) exhibited a similar pattern on the ORP graph, with the curve showing a decrease in ORP values from 292-312 mV down to a range of 253-281 mV over 0-800 seconds, depending on the ozone dose. The steady-state phase in sample M5 was also very short, with varying durations depending on the ozone dose. After reaching a minimum, the graph showed a very sharp increase in ORP compared to the previous samples. The increase in ORP began to appear from 250-800 seconds and continued to increase until the end of ozonation. The highest ozone dose resulted in the highest ORP spike, reaching 712 mV. Ozone doses of 3 and 2 g/h/L also showed a fairly sharp increase in ORP, reaching 688 and 658 mV, respectively. This also indicates that despite the lower ozone concentration compared to the 6 g/h/L dose, the oxidation value was still high due to the relatively light pollutant load in the system in sample M5. Furthermore, at ozone doses of 1.5 and 1.2 g/h/L, the ORP increased sharply but gradually, reaching maximum values of 543 and 286 mV, respectively. These ORP values are equivalent to or higher than the ORP values in samples M1-M4 at the highest ozone dose of 6 g/h/L.

Furthermore, the phenomena observed in graphs M1-M5 align with the principles of ozonation reaction kinetics in heterogeneous systems. As explained by Gottschalk et al. (2009), in wastewater ozonation, the ozone concentration in the liquid phase can increase over time if the target compound is depleted or the reaction proceeds more slowly. This causes an increase in ORP values as ozone begins to dominate the reaction system. Furthermore, Gottschalk et al. (2009) also emphasized that in semi-batch experiments, the increase in ORP often reflects the accumulation of ozone not consumed by the pollutant load, especially when the ratio of ozone dose to pollutant mass is too large. Therefore, understanding the ORP profile is crucial in evaluating the ozonation process.

Overall, the data shows that the combination of an ozone dose of 6 g/h/L and samples with a low pollutant load, namely TSS ± 3,000 mg/L, produces a highly oxidative environment, resulting in ORP values reaching 712 mV. Conversely, in samples with a high TSS load, more ozone is consumed by the initial oxidation reaction, limiting ozone accumulation and ORP increase. Therefore, selecting the appropriate ozone dose relative to the pollutant load is key to optimizing the oxidation process.

3.3. Changes in ORP During the Filtration Process Series

After the ozonation process is complete, the wastewater is not immediately discharged but instead flows through a filtration stage to separate suspended particles and floc formed during the oxidation process. To understand the dynamics of the post-ozonation redox system, ORP measurements were performed before and after the filtration process using a vacuum pump filter with 10-micron filter paper. This observation aims to evaluate how the separation of the solids fraction affects the stability of the oxidative environment in the liquid phase.

The ORP value before filtration reflects the total redox state of the system, including contributions from dissolved ozone, oxidation products, and reduced compounds that have not been fully eliminated. During the filtration process, particles, including floc containing organic compounds, heavy metals, and residual active oxidants, will separate from the liquid phase. This can result in two scenarios for changes in ORP. First, if the filtered suspended particles contain a high concentration of oxidizing compounds or active reactants, the post-filtration ORP may decrease because most of the redox contributors have been removed from the system. Conversely, if filtration successfully removes reduced contaminants that consume ozone, the remaining dissolved ozone will dominate the system and cause an increase in ORP (Gottschalk et al., 2009).

Research by Beltran (2003) also shows that filtration after ozonation can significantly alter the system's redox profile, depending on the contaminant composition and floc characteristics. Environments with low TSS loads tend to retain greater amounts of active ozone after filtration, resulting in an increase in ORP. Therefore, monitoring ORP values before and after filtration is useful not only as an indicator of the success of the solids separation process but also as an evaluation parameter for the remaining oxidative potential in the system. Figure 3 presents changes in ORP values across all samples and treatment doses, comparing conditions before and after the filtration process. The ORP values before filtration in this graph are taken from the ORP mode values during the final phase of the ozonation process.

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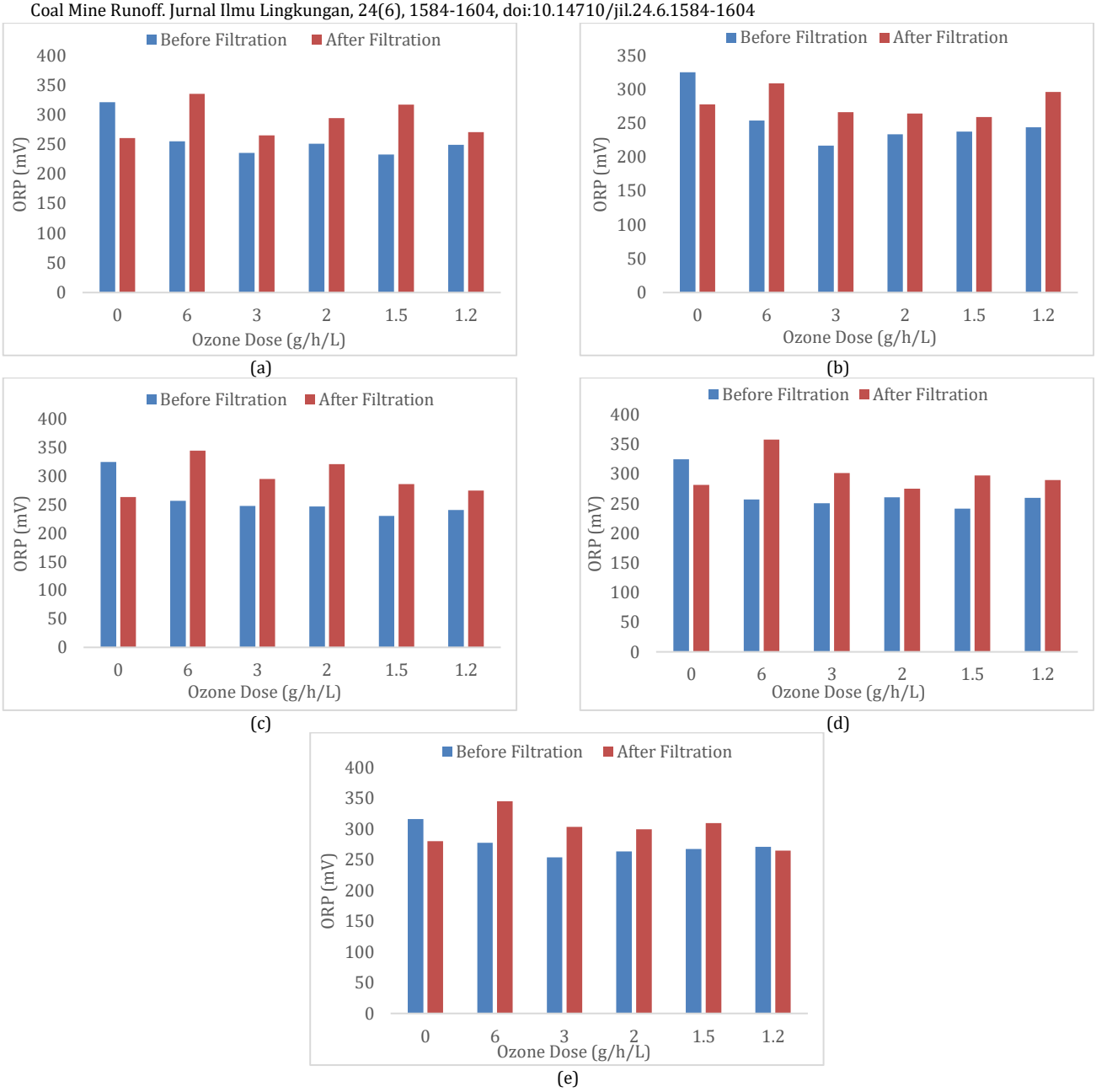


Figure 3. ORP Variations throughout the Filtration Process for Samples (a) M1, (b) M2, (c) M3, (d) M4, and (e) M5

Based on the bar chart (Figure 3), the ORP values of samples without ozone treatment (control) generally decreased after the filtration process. In samples M1, M2, M3, M4, and M5, the ORP values decreased by 60.33 mV; 47.33 mV; 61.33 mV; 43.33 mV; and 35.67 mV, respectively. This decrease was due to the dominance of reduced compounds that were not oxidized, as well as the loss of oxidative compounds during the filtration process. Conversely, in samples treated with ozone, the ORP values tended to increase after the filtration process. This contrasts with samples without ozone treatment (control).

3.4. pH Changes During the Ozonation and Filtration Process Series

pH is a fundamental parameter in evaluating changes in the chemical characteristics of water during the ozonation process. The pH value not only indicates the acidity or alkalinity of the solution but also directly influences the ozone reaction pathway in the water. Therefore, pH measurements are performed at three crucial time points: before ozonation to determine the initial condition of the wastewater, after ozonation to assess the results of oxidative reactions, and after filtration to observe changes following solids separation.

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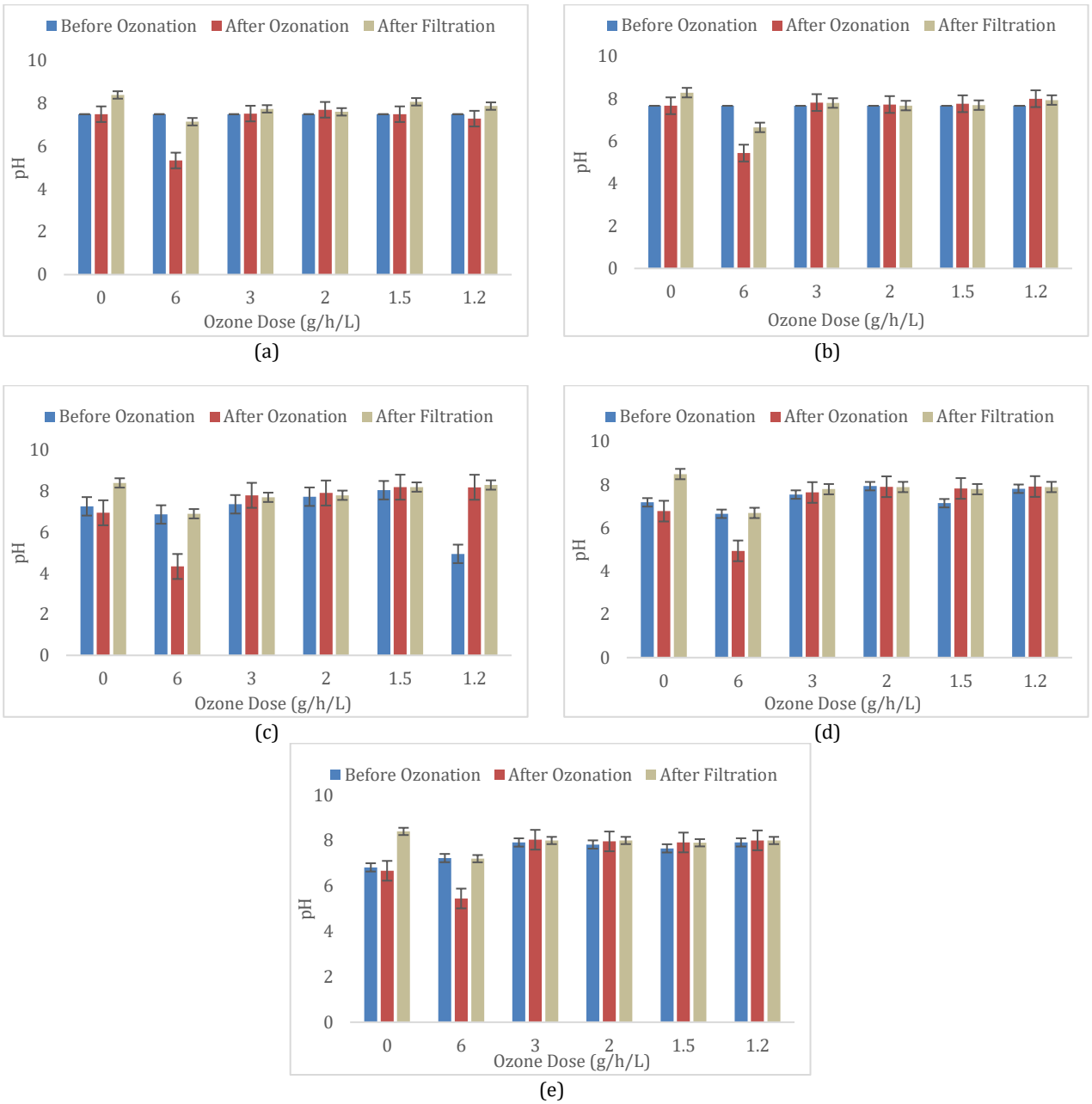


Figure 4. The pH Variation Profile throughout the Series of Ozonation and Filtration Processes for Sample (a) M1, (b) M2, (c) M3, (d) M4, and (e) M5

In general, ozone is more stable and reacts as an intact molecule at low to neutral pH. Under these conditions, ozone acts as a selective oxidizer and tends to attack reduced compounds such as metal ions and certain organic compounds. However, when the system pH increases to alkaline conditions above pH 8, ozone can decompose into hydroxyl radicals, reactive species with a much stronger oxidizing potential than ozone itself but are not as selective. This means that hydroxyl radicals can attack almost any type of organic or inorganic compound in water, including those that should not be oxidized. Therefore, their use must be carefully considered to avoid excessive degradation or the formation of undesirable byproducts (Gottschalk et al., 2009).

In the context of mining wastewater, a decrease in pH during ozonation is actually normal. This decrease occurs because the ozone oxidation reaction of contaminants often produces acidic products such as carboxylic acids, acetic acid, or dissolved carbon dioxide, which forms carbonic acid. Therefore, the higher the ozone dose and the more contaminants successfully oxidized, the greater the likelihood of a decrease in pH in the system. This finding is supported by Beltran (2003), who stated that the ozonation process of humic compounds also produces mild acidic compounds. Figure 4 shows the pH changes during the ozonation and filtration process.

Based on Figure 4, changes in pH values before and after the ozonation process in the five samples, a consistent pattern is seen that the administration of ozone at a high dose of 6 g/h/L tends to reduce the pH value more than the lower dose, in each sample the initial pH value ranged from 7.5 to 8.1 but after

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ozonation at a dose of 6 g/h/L there was a decrease to reach a pH range of 4.33-5.45. This decrease is in line with the findings of Gottschalk et al. (2009) who explained that the reaction of ozone with organic compounds in wastewater produces carboxylic acids and other acidic compounds. This condition occurs due to intensive oxidation reactions, especially at high ozone doses which accelerate the formation of acidic byproducts. Zhang et al. (2009) also noted that ozonation of sludge can reduce pH by 0.5–0.7 due to increased dissolved compounds, while Ferrari et al. (2020) showed a decrease in pH of more than two units due to increased dissolved metals and disintegration of particle structures. Thus, the results of this study confirm that excessively high ozone doses can cause a drastic decrease in pH, which can affect colloidal stability and sludge treatment efficiency.

The initial pH of the engineered samples (7.26–8.23, Table 2) indicates that, unlike the highly acidic mine drainage commonly reported in the literature, the coal mine runoff sludge in this study was near-neutral to slightly alkaline. Under these conditions, both oxidation pathways are likely to occur: direct (molecular) ozone reactions dominate at near-neutral pH, whereas hydroxyl radical formation becomes increasingly important as the pH exceeds 8, as in sample M5 (pH 8.23) (Gottschalk et al., 2009). At the highest dose (6 g/h/L), the pH decreased to 4.33–5.45, shifting the system toward the molecular ozone pathway, which is more selective toward reduced metal ions. The rapid ORP increase within the first 0–60 seconds of ozonation (Figure 2) is consistent with the oxidation of dissolved Fe^{2+} and Mn^{2+} into insoluble $Fe(OH)_3$ and MnO_2, which can act as in-situ coagulants and promote floc formation; this explains why moderate ozone doses improved filterability. However, at excessive doses, the strong pH decrease and over-oxidation may increase the solubility of metal hydroxides and break flocs into finer particles, which is consistent with the turbidity rebound observed at 6 g/h/L (Section 3.5.1). Measurement of dissolved Fe and Mn before and after ozonation is recommended to confirm this mechanism.

Conversely, at lower ozone doses, namely 3-1.2 g/h/L, the pH value tends to stabilize and remains close to the initial value before treatment. This is because, at lower ozone doses, the buffer system in the runoff water tends to neutralize the effects of acidity, keeping the pH within the neutral range. This is crucial for maintaining an optimal reaction environment, particularly in the context of floc formation and filtration processes.

Changes in pH during the ozonation process not only reflect the dynamics of chemical reactions but also play a significant role in influencing the surface charge of particles through the zeta potential mechanism. Suspended particles in wastewater generally have a negative charge due to the dissociation of functional groups such as hydroxyl and carboxylate. At high (alkaline) pH conditions, these groups dissociate further, increasing the negative charge on the particle surface. Consequently, electrostatic repulsion between particles occurs, resulting in a stable suspension that is difficult to coagulate. Conversely, when the pH decreases toward neutral or acidic, the hydrogen ions in the solution begin to neutralize the negative charge, weakening the repulsive forces. This condition causes the particles to become electrostatically unstable and tend to approach each other, forming flocs. This process is called particle destabilization, the initial stage of the formation of larger, denser aggregates or flocs that are easily separated through filtration.

After the filtration stage, the pH values of all samples returned to near neutrality, with an initial range of 7.52–8.15 and after filtration 6.66–8.45. All values are within the pH standard range of 6–9. This phenomenon indicates that most of the acidic compounds resulting from ozonation were likely adsorbed or trapped in suspended solids and then filtered out during the filtration process. This is in line with the principle that acidic compounds resulting from oxidation, such as humic acid or lower fatty acids, can bind to the surface of suspended particles through charge interactions or hydrogen bonds, and are eliminated during the filtration process.

3.5. Effect of Ozone Dose on Sludge Filterability

Sludge filterability is a crucial parameter in evaluating the ease of separating sludge solids from water through filtration and directly impacts the efficiency of the sludge dewatering process. In wastewater treatment systems, sludge dewatering is a crucial step in reducing sludge volume by removing as much water as possible, making the sludge easier to handle, transport, or dispose of. Good filterability means the sludge can be filtered more quickly and produces a clear filtrate, indicating a stable and easily separated floc structure. Therefore, in this study, sludge filterability is defined through two main parameters: filtrate turbidity and filtration time. Both were chosen because they represent the efficiency of water separation from solids on a laboratory scale, relevant to field dewatering practices. Filtration was performed using filter paper under vacuum conditions (Section 2). Because the same sample volume was filtered under identical conditions for all treatments, filtration time is directly comparable among treatments and can be converted into an average filtrate flow rate (mL/min). Although SRF and CST are the standard indicators of sludge dewaterability, they were not measured in this study; therefore, filtration time and filtrate turbidity are used as practical comparative indicators of filterability under identical filtration conditions.

In this context, ozonation acts as a pretreatment that can improve filterability through the oxidation process of particle surfaces and organic compounds. The reaction of ozone with reduced compounds or colloids can cause a decrease in zeta potential, making particles more likely to flocculate. However,

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excessively high ozone doses can break down flocs into fine particles or produce dissolved materials that are difficult to filter, thus reducing dewatering performance. Therefore, evaluating sludge filterability through filtration time and filtrate turbidity in this study is not only relevant in the laboratory but also reflects the potential dewatering performance in the field.

3.5.1. Effect of Ozone Dose on Reducing Turbidity Levels

Turbidity is a crucial parameter in assessing the effectiveness of solid-liquid separation processes, particularly in the treatment of wastewater with high suspended solids content, such as coal mine runoff. The turbidity of the filtration results also indicates the extent to which fine particles or colloidal compounds were retained during the filtration process. A clear filtrate indicates that the flocs are large and dense enough to be retained by the filter media, while a cloudy filtrate indicates that fine particles escaped due to poor floc destabilization or breakup. In this study, the effect of varying ozone dosage on post-filtration turbidity was tested across all samples and is presented in Figure 5.

The horizontal axis shows the variation in ozone dosage, while the vertical axis shows the turbidity values of the filtrates after ozonation and filtration. The blue bars in each graph represent the average turbidity values for each ozone dosage treatment. The lower the blue bar, the clearer the filtrate, indicating good filterability. Additionally, each graph features a red horizontal line indicating the turbidity value of the control sample, the sample without ozone treatment. If the height of the blue bar is below the red line, the ozone treatment successfully reduced turbidity compared to the control, indicating improved sludge filterability.

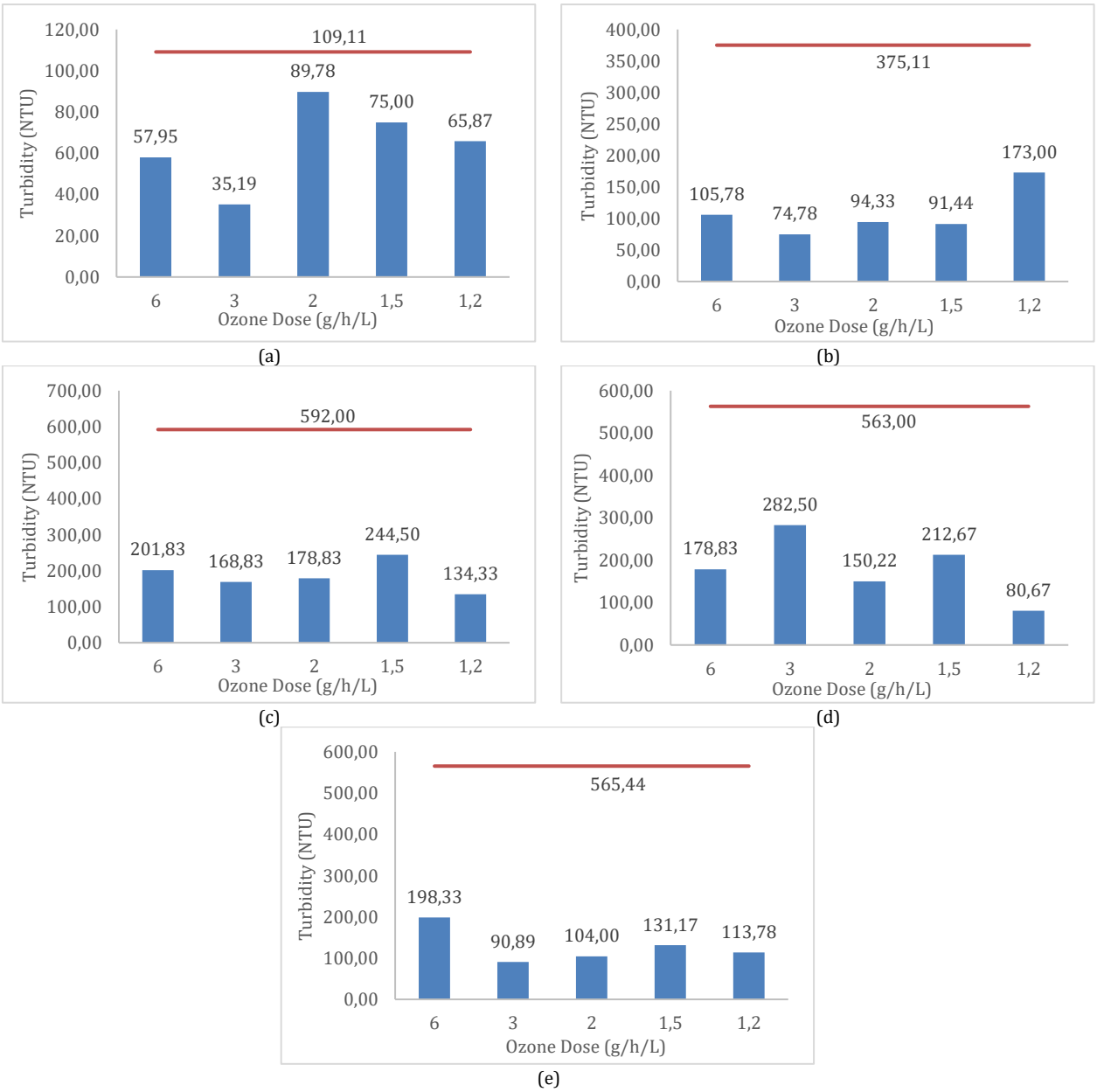


Figure 5. Turbidity Variations after Ozonation and Filtration for Samples (a) M1, (b) M2, (c) M3, (d) M4, and (e) M5

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Based on Figure 5 and the results of the single-factor ANOVA test in the appendix, there is a significant effect between ozone dose and the turbidity level of the filtrate in all samples M1 to M5. The P-value was <0.05 for each sample, all of which were far below the 0.05 significance level. This indicates that variations in ozone dose significantly affect changes in the turbidity of the sludge filtration.

In sample M1 (Figure 5 (a)), the optimum dose was found at 3 g/h/L, with the lowest turbidity of 35.19 NTU, compared to an initial value of 109.11 NTU. However, at a dose of 6 g/h/L, the turbidity level exceeded 3 g/h/L, reaching 57.95 NTU. This indicates over-oxidation, which could potentially release dissolved organic compounds (G. Erden and A. Filibeli, 2010). A similar thing happened in sample M2 (Figure 5 (b)) with the best turbidity reduction at a dose of 3 g/h/L, namely 74.78 NTU from 375.11 NTU and increased again at a dose of 6 g/h/L. This over-oxidation effect was also increasingly evident in M3 (Figure 5 (c)), where the lowest dose of 1.2 g/h/L actually gave the best results with a value of 134.33 NTU from 592 NTU. Conversely, the ozone dose of 6 g/h/L which is the highest ozone dose produced quite high turbidity, namely 201.83 NTU. In samples M4 (Figure 5 (d)) and M5 (Figure 5 (e)), the same pattern was observed. Ozone doses of 3-1.2 g/h/L tend to be optimal. The ozone dose of 1.2 g/h/L in sample M4 produced the best turbidity reduction with a final value of 80.67 NTU from 563 NTU. Furthermore, an ozone dose of 3 g/h/L on sample M5 produced the best turbidity reduction results, achieving 90.89 NTU from 565.44 NTU.

Test results on the five engineered samples (M1–M5) with varying ozone doses of 6–1.2 g/h/L showed that the effect of ozone dose on sludge filtration capacity showed varying patterns depending on the initial TSS concentration of each sample. In general, an ozone dose of 3 g/h/L demonstrated optimal performance in reducing filtrate turbidity levels in most samples M1, M2, and M5, with turbidity values of 35.19 NTU, 74.78 NTU, and 90.89 NTU, respectively.

Excessively high doses (6 g/h/L) generally caused a rebound in turbidity levels, suspected to be due to over-oxidation, which breaks flocks into fine, difficult-to-filter particles. Meanwhile, in some samples, such as M4 and M3, the best performance was achieved at low to moderate ozone doses of 1.2–2 g/h/L, indicating that ozonation effectiveness is significantly influenced by the solids load (TSS) and initial sludge characteristics.

The non-linear filtration response can be interpreted as a competition between ozone demand, mineral/metal transformation, and floc restructuring. In coal mine runoff sludge, reduced metal species and particle-associated organic matter can consume ozone. Where Fe(II) is present, oxidation toward Fe(III) can promote hydrolysis and formation of Fe(III)-bearing precipitates that may contribute to particle enmeshment or sweep-coagulation-type behavior. At the same time, partial oxidation of organic coatings can weaken steric and electrostatic stabilization of fine particles, facilitating aggregation. Conversely, excessive oxidation can fragment organic coatings or existing flocs and generate smaller colloidal or soluble products, which is consistent with the rebound in filtrate turbidity observed at the highest loading. The accompanying ORP increase and pH decrease at high ozone loading are also consistent with stronger oxidative conditions and acid-forming oxidation reactions. However, because Fe speciation, zeta potential, particle-size distribution, and dissolved ozone were not measured directly, these pathways should be regarded as physicochemical interpretations of the observed trends rather than directly verified transformation mechanisms.

These results support the findings of Demir & Filibeli (2012) and G. Erden and A. Filibeli (2010), who emphasized that ozonation is effective in improving sludge filterability, but must be administered at an optimum dose to avoid the negative effects of excessive oxidation. Therefore, an ozone dose of 3 g/h/L can be recommended as the starting point for sludge treatment with a TSS range of 3,000–50,000 mg/L, with additional evaluation for specific sludge conditions in the field.

3.5.2. Effect of Ozone Dose on Filtration Time

The filtration process is a crucial step in the separation of solids from liquids in sludge wastewater from coal mining operations. One of the performance indicators of this process is the time required to filter the sludge until a certain volume is reached, also known as the filtration time. In this context, ozonation is used as a pretreatment that aims to improve the filterability of the sludge by changing the floc structure, oxidizing organic compounds, and increasing redox activity. The filtration time is used to measure the resistance of liquid flow through the filter media, namely 10-micron filter paper. The shorter the time required to filter a certain volume, the lower the flow resistance, which means the floc structure does not clog the filter pores, and water can be separated more easily, thus indicating good dewatering performance. In this study, the effect of ozone dose variations on filtration time during the filtration process was specifically tested on samples M1, M2, M3, M4, and M5 with engineered TSS values. To illustrate this phenomenon in more detail, a graph of the filtration time of ozonized sludge at various ozone doses is presented and analyzed in the form of a bar chart. Figure 6 shows the effect of varying ozone doses on reducing filtration time.

The horizontal axis shows the variation in ozone dose, while the vertical axis shows the filtration time in seconds. The blue bars represent the average filtration time for each treatment. The shorter the blue bar, the faster the filtration process, indicating better sludge filterability. The horizontal red line shows the filtration time of the control sample (without ozone). If the blue bar is below the red line, ozonation has successfully increased filtration efficiency.

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The experimental results showed that ozone dosage significantly affected sludge filtration time for samples M1, M4, and M5, with a P-value <0.05 (see appendix). In sample M1, ozone administration reduced filtration time from 1749 seconds to 1189 seconds at an optimum dose of 2 g/h/L. However, ozone dosages that were too high or too low actually increased filtration time, indicating over-oxidation or a lack of oxidative reactions. This phenomenon aligns with a study by Demir & Filibeli (2012), which stated that moderate doses of ozone are effective in increasing filterability, while overdoses risk releasing humic compounds that interfere with the filtration process.

Samples M2 and M3 showed a pattern of decreased filtration time, but this was not statistically significant with a P-value >0.05. However, the 2 g/h/L dose in sample M2 resulted in a filtration time of 647 seconds, compared to 905 seconds for the untreated sample. In M3, the best reduction in time was achieved at a dose of 1.2 g/h/L, with a reduction of 394 seconds compared to the 441 seconds for the untreated sample. Excessively high ozone doses in both samples tended to increase filtration time, which is suspected to be caused by over-oxidation and the release of low-molecular organic compounds into the solution (G. Erden and A. Filibeli, 2014).

Sample M4 had the lowest filtration time at an ozone dose of 1.2 g/h/L, at 182 seconds, compared to the 281 seconds for the untreated sample. Even at an ozone dose of 6 g/h/L, the filtration time remained low, at 194 seconds. This indicates that at lower TSS concentrations, the potential for over-oxidation is minimal. Therefore, the initial characteristics of the sludge also determine the effectiveness of ozonation in increasing sludge filterability. In sample M5, the fastest filtration time was achieved at a dose of 1.2 g/h/L, reaching 86 seconds, compared to the 104 seconds achieved for the untreated sample. High ozone doses, such as 6 g/h/L, were also quite effective, producing a filtration time of 95 seconds. However, medium doses of 3-2 g/h/L showed higher filtration times than the others. This indicates that at low TSS levels, medium doses can actually worsen filtration due to suboptimal floc fragmentation.

Based on the analysis of sludge filtration times for each sample, it appears that the effectiveness of ozone doses in accelerating the filtration process varies depending on the initial TSS concentration. For samples M1 and M2, the optimum ozone dose was 2 g/h/L, with the lowest filtration times of 1189 seconds and 647 seconds, respectively. In samples M3, M4, and M5, the optimum dose shifted to 1.2 g/h/L, with filtration times of 394 seconds, 182 seconds, and 86 seconds, respectively. This pattern indicates that the lower the TSS content in the sludge, the lower the ozone dose tends to be more effective in accelerating filtration. Conversely, at high TSS levels such as in M1 and M2, a slightly higher dose is required to achieve optimal floc disintegration. Thus, the optimum ozone dose to improve sludge filterability is not a single factor, but depends on the initial characteristics of the sample, particularly the TSS content. These results also indicate that the effectiveness of ozonation in accelerating the sludge filtration process is not always linear with increasing ozone dose. The optimal dose tends to be in the low to medium range.

3.6. Ozonation Process

3.6.1. Ozonation Mechanism

According to Gottschalk et al. (2009) in their book Ozonation of Water and Waste Water: A Practical Guide to Understanding Ozone and Its Applications, the ozonation process is a strong oxidation method used in water treatment to remove organic and inorganic contaminants through two primary pathways: direct and indirect reactions. Direct reactions occur between ozone molecules and organic compounds in a selective and relatively slow manner. In contrast, indirect reactions involve the formation of hydroxyl radicals (·OH) generated from ozone decomposition, which subsequently attack target compounds with extremely high reactivity. These mechanisms are briefly presented here because they are directly used to interpret the experimental observations of this study, namely the ORP spike in the early phase of ozonation (Section 3.2), the pH decrease at high ozone doses (Section 3.4), and the non-linear changes in filtrate turbidity and filtration time (Section 3.5).

3.6.1.1. Indirect Reactions

Indirect reactions involve radicals molecules that contain unpaired electrons. Most radicals are highly unstable and immediately react with other molecules to regain electron stability. The radical-chain mechanism of ozone can be divided into three stages: initiation, chain propagation, and termination.

The first stage involves ozone degradation accelerated by initiators such as hydroxide ions (OH^-), producing secondary oxidants such as hydroxyl radicals (·OH). These radicals react non-selectively and very rapidly with target molecules, with reaction rate constants of $k = 10^8–10^{10}\ M^{-1}\ s^{-1}$. During electron loss, the target molecule becomes a radical and continues further reactions, extending the chain reaction. However, if radicals react with other radicals, both unpaired electrons neutralize each other, causing the chain reaction to stop. In the initiation stage, hydroxide ions react with ozone to form superoxide anions ($O_2{\cdot}^-$) and hydroperoxyl radicals ($HO_2{\cdot}$):

$$O_3 + OH^- \rightarrow O_2^{o-} + HO_2^o \quad (1)$$

Both species exist in acid–base equilibrium:

$$HO_2^o \leftrightarrow O_2^{o-} + H^+ \quad (2)$$

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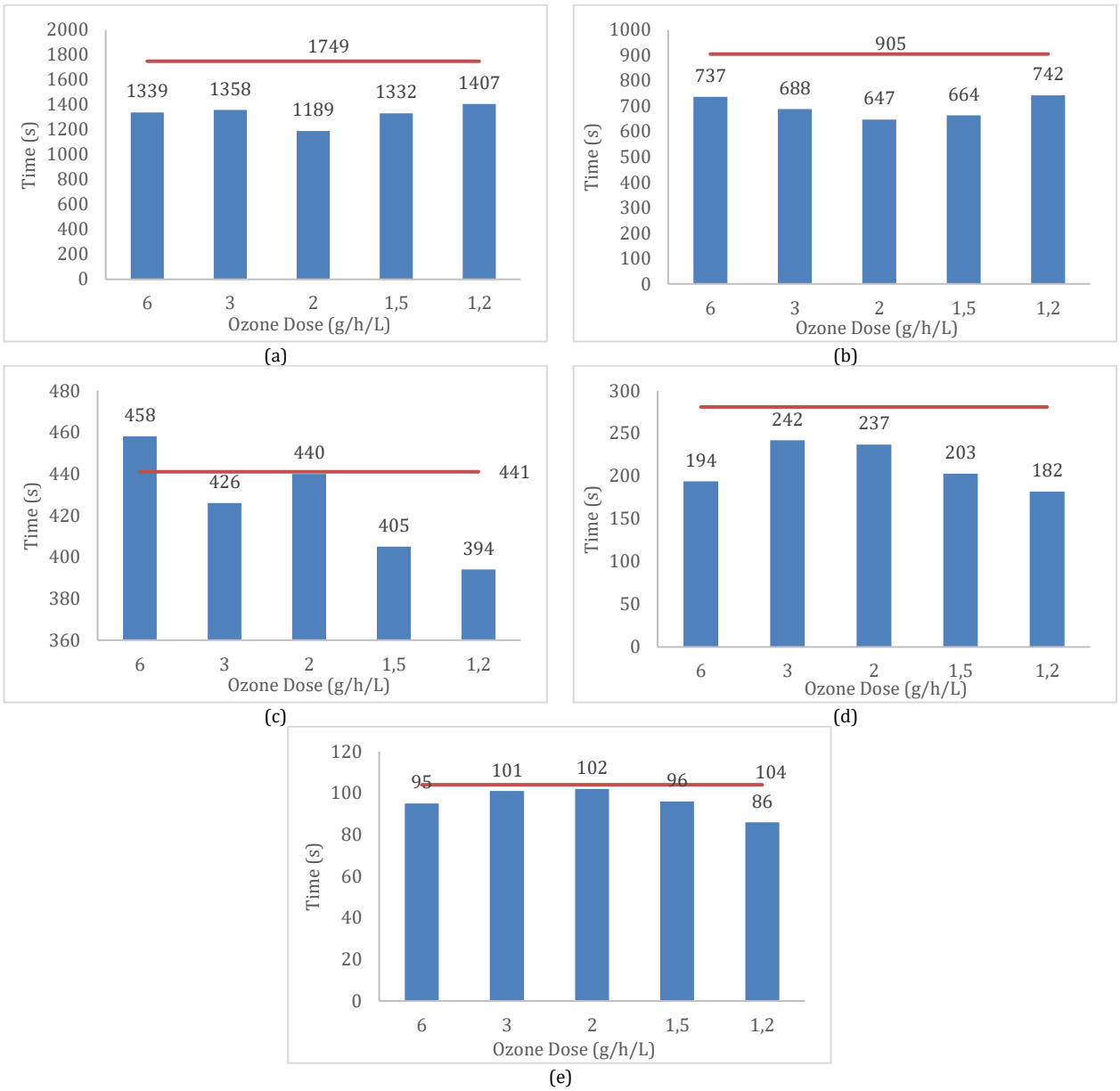


Figure 5. Effect of Ozone Dose on Sludge Filtration Time for Sample (a) M1, (b) M2, (c) M3, (d) M4, and (e) M5

During chain propagation, the superoxide anion reacts with ozone to form the ozonide anion ($O_3 \cdot^-$). This species rapidly decomposes via the formation of hydrogen trioxide ($HO_3 \cdot$), leading to the generation of hydroxyl radicals (·OH):

$$O_3 + O_2^{o-} \rightarrow O_3^{o-} + O_2 \quad (3)$$
$$HO_3^o \leftrightarrow O_3^{o-} + H^+ \quad (4)$$
$$HO_3^o \rightarrow OH^o + O_2 \quad (5)$$

This reaction may continue as follows:

$$OH^o + O_3 \rightarrow O_4^{o-} \quad (6)$$
$$HO_4^o \leftrightarrow O_2 + HO_2^o \quad (7)$$

Overall, two ozone molecules are consumed to produce two hydroxyl radicals. Substances that convert ·OH into superoxide-type radicals, such as $HO_2 \cdot$, accelerate this reaction and act as chain promoters. Organic molecules may also act as promoters if they contain functional groups that can react with ·OH to form organic radicals (R·):

$$H_{2R} + OH^o \rightarrow HR^o + H_2O \quad (8)$$

In the presence of oxygen, these organic radicals can transform into organic peroxy radicals (ROO·), further extending the chain reaction:

$$HR^o + O_2 \rightarrow HRO_2^o \quad (9)$$
$$HRO_2^o \leftrightarrow R + HO_2^o \quad (10)$$
$$HRO_2^o \rightarrow RO + OH^o \quad (11)$$

The termination stage occurs when certain organic or inorganic substances react with ·OH to form secondary radicals that do not produce superoxide. These substances, known as inhibitors or scavengers, stop the chain reaction and slow down ozone degradation:

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$$OH^o + CO_3^{2-} \rightarrow OH^- + CO_3^{o-} \quad (12)$$
$$OH^o + HCO_3^- \rightarrow OH^- + HCO_3^o \quad (13)$$

Radical–radical reactions may also terminate the chain:

$$OH^o + HO_2^o \rightarrow O_2 + H_2O \quad (14)$$

Overall, the combined reactions show that three ozone molecules yield two hydroxyl radicals:

$$3O_3 + OH^- + H^+ \rightarrow 2OH^o + 4O_2 \quad (15)$$

Hydroxyl radicals exhibit extremely reactive and non-selective behavior, preferentially attacking molecular sites with the highest electron density (electrophilic characteristics). These radicals have very short lifetimes–less than 10 milliseconds at an initial concentration of 10^{-4} M. According to Buxton et al. (2006) in Gottschalk et al. (2009), the reaction rate between hydroxyl radicals and aromatic compounds approaches the diffusion limit, meaning they react almost instantaneously upon collision.

3.6.1.2. Direct Reactions

Ozone can also react directly with organic compounds through selective, slower reactions. The reaction rates range from kD = 1.0–10^6 M^{-1} s^{-1}. The direct mechanism, known as the Criegee mechanism, involves ozone addition to unsaturated bonds. These reactions occur more rapidly with highly nucleophilic compounds, such as aromatics and aliphatics with electron-donating substituents. Direct ozonation becomes important when radical-based reactions are suppressed–for example, when water lacks chain-initiating compounds or contains abundant scavengers. Increased scavenger concentration shifts the oxidation pathway toward direct reactions. Thus, both inorganic and organic carbon play important roles. Generally, direct reactions dominate under acidic conditions (pH < 4), whereas indirect (radical-based) reactions become dominant above pH > 10. In groundwater and surface water (pH ≈ 7), both pathways may contribute significantly. In some wastewater types, indirect oxidation remains relevant even at pH = 2, depending on the contaminants present. Therefore, both pathways must always be considered in treatment process design.

3.6.2. Ozonation Effect

According to Gottschalk et al. (2009), ozonation can lower wastewater pH due to the formation of carboxylic acids during organic oxidation and the oxidation of ammonia to nitrate. This pH decrease depends on water composition, particularly alkalinity. In high-buffer systems, pH changes tend to be smaller.

This phenomenon is supported by several studies. Zhang et al. (2009) found that ozonating sludge at a dose of 25–80 mg/g DS decreased pH by 0.5–0.7 and increased soluble components such as SCOD and total nitrogen. Similarly, Ferrari et al. (2020) reported pH reduction from 6.17 to 3.6, accompanied by increased dissolved metal content and particle size changes. These pH shifts influence particle surface charge and colloidal stability.

On the other hand, ozone has a very high redox potential (+2.07 V), resulting in a significant increase in ORP during ozonation (Gottschalk et al., 2009). High ORP indicates strong oxidative conditions beneficial for organic degradation, microbial inactivation, and more stable floc formation. Lv et al. (2019) showed that Differential ORP (DORP) correlates strongly with sludge dewaterability, where an optimal DORP of ~387 mV resulted in lower CST and SRF values, indicating improved filterability. However, excessively high ORP may generate intermediate compounds that hinder flocculation.

Changes in pH and ORP during ozonation are closely related to the zeta potential of sludge particles, which indicates surface charge and governs colloidal stability. When pH decreases and ORP increases, particle surface charge tends to decrease toward neutrality. Gottschalk et al. (2009) noted that this reduces inter-particle repulsion, enabling floc formation. Yu et al. (2019) found that ozonation combined with Zero-Valent Iron (ZVI) decreased CST from 10.45 seconds to 1.63 seconds and reduced cake moisture to 52.67%, associated with zeta potential shifting toward neutrality.

Thus, ozonation can improve sludge quality by decreasing pH, increasing ORP, and adjusting zeta potential. These changes weaken colloidal stability and enhance flocculation effectiveness, making ozonation a promising pretreatment step for sludge management in wastewater systems, including coal mine runoff.

3.7. Optimum Ozone Dose

Determining the optimum ozonation dose is a crucial aspect in improving the effectiveness of the sludge separation process in coal mine runoff sludge wastewater. In this study, ozonation effectiveness was evaluated based on three main parameters: reduced turbidity, reduced filtration time, and changes in water chemical characteristics such as pH and ORP.

Data obtained from five samples showed that each ozone dose had a different effect on these parameters. Therefore, Table 4 shows the optimum dose based on turbidity level. Furthermore, Table 5 shows the optimum dose based on filtration time for samples M1 to M5.

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Table 4. Summary of the Optimum Ozone Dose for Turbidity Reduction

Sample Code	TSS (mg/L)	Optimal Ozone Dose (g/h/L)	$T_{Control}$ (NTU)	T_{Final} (NTU)	Effectiveness (%)
M1	± 50,000	3	109.11	35.19	67.75
M2	± 30,000	3	369.11	74.78	79.74
M3	± 20,000	1.2	592	134.33	77.31
M4	± 10,000	1.2	614.33	80.67	86.87
M5	± 3,000	3	617.33	113.78	85.28

Table 5. Summary of the Optimum Ozone Dose for Reducing Filtration Time

Sample Code	TSS (mg/L)	Optimal Ozone Dose (g/h/L)	$t_{Control}$ (s)	t_{Final} (s)	Effectiveness (%)
M1	± 50,000	2	1749	1189	32.02
M2	± 30,000	2	779	647	16.94
M3	± 20,000	1.2	441	394	10.66
M4	± 10,000	1.2	278	182	34.53
M5	± 3,000	1.2	110	86	21.82

Based on the results in Table 4 and Table 5, the ozonation process significantly reduced turbidity levels and sludge filtration time in coal mine runoff samples. Ozone acts as an oxidizing agent capable of disintegrating flocs and damaging the structure of complex organic compounds in the sludge, thereby improving solid-liquid separation performance.

Based on Table 4, regarding turbidity parameters and data on changes in ORP and pH values after the filtration process, it can be concluded that the optimum ozone dose was achieved in sample M4 at a dose of 1.2 g/h/L. This sample had an initial TSS level of ±10,000 mg/L and demonstrated an 86.87% turbidity reduction effectiveness. Furthermore, based on the results in Table 5, the optimum ozone dose for filtration time was also achieved in sample M4 at the same dose of 1.2 g/h/L. Under these conditions, filtration time decreased from 278 seconds to 182 seconds, resulting in an effectiveness of 34.53%.

The success of the ozonation process in increasing filtration effectiveness is supported by an increase in the ORP value of 30 mV after filtration, indicating that residual ozone or other oxidative compounds are still active in the system but not excessive enough to trigger over-oxidation. This condition reflects that the oxidation reaction is proceeding efficiently, sufficient to disintegrate floc and improve sludge structure without producing fine colloidal particles that can clog the filtration media. Furthermore, the stability of the system is also reflected in the pH value, which remains near neutral, only changing from 7.82 to 7.92, indicating that there is no significant accumulation of acidic byproducts. This indicates that ozonation is working optimally in breaking down filtration-inhibiting compounds without disrupting the overall chemical balance of the system.

This finding aligns with a report by Demir & Filibeli (2012), which states that ozonation at an optimum dose can improve sludge dewaterability by destroying biopolymer compounds and strengthening the floc structure for filtration. Meanwhile, G. Erden and A. FIlibeli, (2010) added that too high an ozone dose can cause over-oxidation, which produces lowmolecularorganic fragments such as humic acid, which are colloidal and reduce filtration efficiency. the floc structure for filtration. Meanwhile, G. Erden and A. FIlibeli, (2010) added that too high an ozone dose can cause over-oxidation, which produces low molecular organic fragments such as humic acid, which are colloidal and reduce filtration efficiency.

Thus, the success of ozonation is highly dependent on the accuracy in determining the ozone dose that is appropriate to the initial pollutant load conditions or initial characteristics, so that the oxidation reaction takes place sufficiently to disintegrate the particles that cause turbidity, without producing by-products that worsen the quality of the filtrate.

3.8. Comparison of Research Results with Previous Studies

To assess the effectiveness of the ozonation method used in this study in improving sludge filterability, it is important to compare the results obtained with those of previous studies. This comparison aims to assess the consistency of the results, identify the method's strengths, and understand its limitations in the context of various sludge types, treatment doses, and indicators used.

In this study, sludge filterability was measured using two main parameters: filtrate turbidity (NTU) and filtration time (seconds). The results showed that ozonation at an optimum dose of 1.2 g/h/L reduced turbidity by up to 86.87% in the low TSS sample, M4, and reduced filtration time by up to 34.53% in the M4 sample. The primary focus of this comparison was the effectiveness of each method in improving filterability indicators such as filtrate turbidity, filtration time, capillary suction time (CST), and sludge moisture content. Furthermore, the comparison also considered practicality, the risk of over-oxidation, and the need for additional materials in the process. The following table presents a summary of relevant research results as a reference for comparing the effectiveness of the ozonation method developed in this study. Table 6 presents a summary comparison between this research and relevant previous studies.

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Table 6. Comparative Analysis of the Present Findings and Prior Studies

No	Authors	Research Title	Method	Filterability Indicators	Effectiveness
	This study	Performance Analysis of Ozonation on Sludge Filterability of Coal Mine Runoff	Ozonation (1.2-6 g/h/L)	Filtrate turbidity (NTU) & filtration time (s)	Turbidity reduction up to 86.87% (M4); filtration time decreased by 34.53% (M4); optimum dose 1.2 g/h/L
1	Yang dkk. (2024)	Response Surface Optimization of Sludge Dewatering Process: Synergistic Enhancement by Ultrasonic, Chitosan and Sludge-Based Biochar	Ultrasonic + CTS + Biochar	SRF, MC, EPS, FTIR, Calorimetry, TG–DTG	MC reduced to 76.81%, low SRF (5.41×10^{11} m/kg), EPS significantly decreased; FTIR shows hydrophilic groups reduced; sludge cake easier to dry, heating value increased by 17%.
2	Ge dkk. (2020)	An In-Depth Study on The Deep-Dewatering Mechanism of Waste Activated Sludge by Ozonation Pre-Oxidation and Chitosan Re-Flocculation Conditioning	Ozonation (60 mg/gTS) + Chitosan (20 mg/gTS)	CST, WC, SCOD, EPS, NMR, SEM, Heating Value	WC decreased from 82.1% to 56.5%; CST improved after chitosan; EPS solubilized by ozone then re-bound by chitosan forming larger flocs, enhancing dewatering and increasing heating value.
3	Ge dkk. (2019)	Insight Into A New Two-Step Approach of Ozonation and Chitosan Conditioning for Sludge Deep-Dewatering	Ozonation + Chitosan	CST, WC, Zeta Potential, EPS, Particle Size, Heavy Metals	CST drastically reduced from 196.3 s to 15.8 s; WC dropped to 57.5%; EPS decreased; zeta potential approached neutral, resulting in optimal flocculation. Metal speciation needs further evaluation.
4	Cai dkk. (2019)	Improving Dewaterability and Filterability of Waste Activated Sludge by Electrochemical Fenton Pretreatment	Electro-Fenton (ZVI + H_2O_2, electrical current)	CST, Water Content, Bound Water (DSC), Particle Size, EPS (3D-EEM, FTIR)	CST significantly reduced from 50.2 s to 11.9 s; WC from 83.1% to 65.7%. Hydroxyl radicals degraded EPS and converted bound water to free water, improving cake structure. Stable for 10 cycles; low cost (USD 59.78/ton DS).
5	Zhu dkk. (2018)	Conditioning of Sewage Sludge Via Combined Ultrasonication-Flocculation-Skeleton Building to Improve Sludge Dewaterability	US + CPAM + Rice Husk (RH)	SRF, TTF, MC, $d_{0.5}$, Porosity, EPS (Proteins, Polysaccharides)	SRF decreased from 1.39×10^{9} to 1.32×10^{8} s^2/g; MC from 91% to 62.15%; larger flocs and more porous cake structure; synergistic effect accelerated filtration and reduced water content.
6	Zhen dkk. (2014)	Enhanced Dewatering Characteristics of Waste Activated Sludge with Fenton Pretreatment: Effectiveness and Statistical Optimization	Fenton (H_2O_2 + Fe^{2+}), RSM	CST, CST Reduction Efficiency, EPS, Floc Microscopy	CST decreased from 1360 s to 24 s (98.25% improvement); EPS breakdown enhanced water release; statistically optimized for rapid dewatering
7	Demi1 (2012)	Fate of Return Activated Sludge After Ozonation: An Optimization Study for Sludge Disintegration	Ozonation (0.007–0.06 g O_3/gTS)	CST, SRF, SVI, TSS/VSS, SCOD, EPS, Particle Size	Effective for sludge reduction (TSS, VSS ↓ >70%) and disintegration (DD 56.2%), but CST increased → poor filterability at high ozone doses. Re-flocculation required for dewatering improvement.
8	Erden (2010)	Ozone Oxidation of Biological Sludge: Effects on Disintegration, Anaerobic Biodegradability, and Filterability	Ozonation (0.005–0.25 g O_3/g DS)	CST, DD, SCOD/DOC, SS/VSS, Particle Size, EPS Protein	Effective sludge disintegration (DD 51.1%), anaerobic biodegradability improved (+25% biogas), but filterability worsened at doses >0.1 g O_3/gDS (CST raised sharply). Suitable for digestion, not for direct dewatering.

This study demonstrates that ozonation without additives significantly improves sludge filterability, especially for sludge with low to moderate TSS. Its effectiveness is comparable to other combination methods, such as ozonation + chitosan from the studies of Ge et al. (2020) and Ge et al. (2019) or electro-Fenton from the study of Cai et al. (2019), which generally result in reductions in CST or water content of more than 40–90%.

Coal mine runoff sludge should not be expected to respond identically to waste activated sludge (WAS). WAS is predominantly composed of microbial biomass, extracellular polymeric substances (EPS), and associated bound water; consequently, ozonation can strongly affect dewaterability through cell/EPS solubilization and subsequent changes in floc structure. In contrast, coal mine runoff sludge in this study is characterized by a much larger inorganic suspended-solids fraction and metal-bearing mineral particles. Its response to ozone is therefore more strongly governed by solids loading, mineral surface chemistry, oxidation of redox-active metals, and possible precipitation/coagulation effects, in addition to oxidation of the smaller organic fraction. This compositional difference helps explain why the optimum ozone loading in the present system is sample-dependent and why percentage improvements reported for WAS should not be transferred directly to coal mine runoff sludge.

However, the advantage of this method lies in its simplicity—it does not require additional chemicals or special equipment—making it easier to implement in the field. Furthermore, the risk of over-oxidation can be controlled by selecting the optimum dosage appropriate to the TSS load. Thus, the low-dose ozonation applied in this study is proven to be a practical and efficient method for improving sludge filterability, while also making an important contribution to the development of simple oxidation-based wastewater treatment technologies.

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3.9. Limitations of the Study

This study has several limitations related to reactor design. The ozone dose was varied by changing the sample volume (0.5–2.5 L) at a constant generator output of 3 g/h, rather than by varying the contact time at a constant reactor volume. Because the reactor geometry was fixed, changes in sample volume altered the height of the water column, which affects the bubble residence time, the actual gas–liquid contact time, and the volumetric mass transfer coefficient (kLa). Consequently, the ozone transfer efficiency may not have been identical across treatments, and the doses reported in g/h/L represent nominal volumetric ozone loading rates rather than transferred or absorbed ozone doses. The results should therefore be interpreted as comparative laboratory screening of ozone loading, and comparisons between dose levels should take this hydrodynamic bias into account.

In addition, filterability was assessed using filtration time and filtrate turbidity rather than the standard dewaterability indicators, namely Specific Resistance to Filtration (SRF) and Capillary Suction Time (CST). Future studies should vary the ozone dose by adjusting the contact time (e.g., 10, 20, and 30 minutes) at a constant reactor volume, quantify the transferred ozone dose and kLa, and include SRF, CST, zeta potential, and dissolved Fe and Mn measurements to confirm the mechanisms proposed in this study.

4. CONCLUSION

This study demonstrates that ozonation induces measurable shifts in water chemistry and exerts a decisive influence on sludge filterability in coal mining runoff. The oxidation–reduction potential (ORP) exhibited a characteristic initial decline, reflecting rapid ozone consumption by reduced species, followed by a progressive increase as residual ozone accumulated in samples with lower solid loads. The highest post-ozonation ORP (712 mV in Sample M5 at 6 g/h/L) confirms that high-strength oxidation occurs primarily when the particulate and dissolved contaminant load is sufficiently low to permit ozone persistence.

Ozonation at elevated doses (6 g/h/L) caused a reduction in pH due to the formation of acidic oxidation intermediates, whereas moderate doses (1.2–3 g/h/L) maintained pH near neutrality. After filtration, pH typically returned toward its initial value, indicating that acidic byproducts were either co-removed with solids or neutralized through subsequent physicochemical interactions.

Ozone dosage had a significant and non-linear effect on sludge filterability. Moderate dosages (1.2–3 g/h/L) consistently improved turbidity removal and reduced filtration time across multiple samples, suggesting optimal partial oxidation that enhances floc compactness and promotes particle aggregation. In contrast, excessive ozonation (6 g/h/L) led to over-oxidation, the release of humic fragments, and the formation of finer colloids, all of which impaired filtration performance. Insufficient doses failed to restructure the sludge matrix effectively.

Integrating turbidity removal, filtration time, ORP behavior, and pH stability, the optimal operational condition was achieved in Sample M4 (initial TSS ~10,000 mg/L) with an ozone dose of 1.2 g/h/L. Under this condition, turbidity decreased by 86.87% (614.33 to 80.67 NTU) and filtration time improved by 34.53% (278 to 182 s), accompanied by a modest ORP increase of 30 mV and stable neutral pH (7.82 to 7.92). These results indicate that controlled, moderate ozonation effectively weakens flocs and enhances subsequent solid–liquid separation without generating oxidation byproducts that hinder filtration.

Overall, the findings confirm that ozonation is a viable pre-treatment strategy for improving sludge dewaterability in mining runoff, provided that the ozone dose is carefully optimized to balance oxidative breakdown and aggregation mechanisms.

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