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Research Article

# Treatment of dairy industry wastewater by variations of coagulation-flocculation and ozonation

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## ABSTRACT

In this study, COD removal efficiencies were investigated in dairy industry wastewater by using variations of coagulation-flocculation and ozone oxidation in laboratory. Coagulation-flocculation (CF), ozonation (O), ozonation following coagulation-flocculation (CF+O) and coagulation-flocculation following ozonation (O+CF) processes were applied to the wastewater. Optimum pH and optimum dosages were studied at various pH values and at various coagulant doses at room temperature (25°C). In ozone oxidation, wastewater was taken to ozonation reactor and ozone was given 1 g/hour dose and COD removal efficiencies were determined by taking samples in 15, 30, 45, 60, 90, 150, 210 and 240 minutes. Treatment performance of coagulation and flocculation and ozonation processes and their variations were evaluated by COD removal efficiencies. The optimum pHs for the $Al_2(SO_4)_3$ and $FeCl_3$ were found as 7 and 6 respectively. The optimum dosages were 1200 mg/L for $Al_2(SO_4)_3$ and 500 mg/L for $FeCl_3$. At optimum conditions, approximately 45% and 28% of COD were removed by using $FeCl_3$ and $Al_2(SO_4)_3$ respectively. When only ozonation was applied, COD removal efficiency was determined as 20% at the end of 240 minutes. COD removal efficiency was 65% with the application of ozonation after coagulation-flocculation (CF+O). When the coagulation-flocculation process was applied after the ozonation process (O+CF), COD removal efficiency remained at 52%.

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## INTRODUCTION

Milk is one of the essential nutrients needed in all areas of life. For this reason, the dairy industry is one of the most basic industrial areas accepted all over the world. A wide variety of products such as pasteurized milk, white cheese, cream cheese, yoghurt, etc. can be obtained after preliminary technological processes such as cleaning, separation of fat, processing, homogenization, standardization of cream, thermal processes and cooling applied to raw milk.

In the European Union countries, 158.2 million tons of raw milk was produced in 2019. According to Eurostat data, milk production is increasing by 1.4 million tons every year [1]. According to TURKSTAT data for 2019, the estimated milk production in Turkey is 23 million tons per year, 91% of which is cattle milk [2]. 60% of the milk produced is processed in dairy industries [3].

In the dairy industry, approximately 2.5 L of wastewater (0.2 - 10 L of wastewater) can be produced for 1 L of processed milk. The quantity and characterization of high volumes of wastewater, which generally consists of washing and cleaning processes, may be related to the production technology [4]. In milk and dairy products facilities, cleaning of milk transportation vehicles, washing of milk transportation boilers, cleaning the production equipment where production takes place, washing the materials such as the tank where the production is made, wastewaters with high pollution contents are formed. In addition, detergents, disinfectants, machine oils and cloth fibers used in cleaning are also included in the wastewater. The content of milk and dairy industry wastewater is generally organic. However, the wastewater flow rate is complicated by the hourly, daily and seasonal changes [5].

Wastewater from dairy industries contains large quantities of fat, casein, lactose, inorganic salts, and detergents, sanitizers which used for washing [6, 7]. General characterization of dairy industry wastewater is as follows; BOD: 1000-3000 mg/L, COD: 2000-5000 mg/L and TSS: 400-1000 mg/L [8]. In addition, dairy industry wastewater may contain various microorganisms, including pathogens [4, 9].

The environmental impacts of the dairy industry wastewaters can be very high, especially due to the discharge of large quantities of organic matter and nutrients (nitrogen and phosphate). Generally, in the treatment of these wastewaters; physical treatment processes are used to separate solids and oils, and then biological treatment systems are used for the removal of organic substances and nutrients. Aerobic processes, in particular activated sludge plants, are frequently used in the treatment of dairy industry wastewater [10].

Many problems can be encountered in the conventional treatment methods used. These can be listed as follows; excessive sludge formation, low sludge sedimentation rate, difficulties in N and P removal, oil breakdown problems [9, 11, 12].

Advanced oxidation processes have been evaluated in numerous studies for the degradation of organic pollutants. The most important advantage of this method is that it can successfully oxidize most organic compounds found in wastes with high organic pollution such as the dairy industry, be able to completely mineralize, reduce their toxicity or increase their biodegradability [13, 14]. Ozone application, which is one of the advanced oxidation processes, is widely used in wastewater treatment. Ozone gas ($E_o$= 2.07 V) is transformed into a hydroxyl radical ($E_o$= 2.80 V), which has a higher oxidation potential than itself under certain conditions [15]. In recent years, several researchers have used ozonation technology alone or in combination with other technologies for the treatment of dairy industry wastewater [14].

In the study conducted by Sivrioglu and Yonar (2015) [3], coagulation flocculation using iron sulphate ($FeSO_4$) and ozone application as pre-treatment before biological treatment of dairy industry wastewater (6300 mg/L COD) with high organic load was evaluated. Ozonation studies were carried out between pH 7 and 12 with 2 g/h ozone application. The highest COD removal was achieved as 71% after 240 minutes of ozone application at pH 12. With the coagulation flocculation application, 60% COD removal efficiency was obtained. They stated that, technically, ozonation is more efficient than coagulation flocculation, but economically quite costly.

In their study, Pereira et al. (2018) [15] evaluated the decomposition effects of ozone combined with hydrogen peroxide and catalyzed with $Mn^{+2}$ for the treatment of synthetic dairy industry wastewater with a COD content of 2.3 g/L. They stated that no significant effect of $H_2O_2$ was determined in $H_2O_2/O_3$ application, and $Mn^{+2}$ catalyzed ozone application was more effective in organic matter removal (69.4%). Despite this positive result, it was stated that it is not possible to obtain disposable water with ozone application alone, and it is necessary to make evaluations on the basis of long-term ozone applications and other parameters.

In this study, COD removal efficiencies were investigated by using laboratory scale coagulation-flocculation and ozone oxidation processes in the treatment of dairy industry wastewater. The wastewater was treated with coagulation-flocculation (CF), ozonation (O), ozonation after coagulation-flocculation (CF+O), and coagulation-flocculation after ozonation (O+CF).

## MATERIALS AND METHOD

Wastewater was taken from a dairy industry located in Kırklareli province in Turkey, Thrace Region. White cheese, butter and curd cheese are produced in the plant. Products produced at the plant and annual usage amounts are given in Table 1. Wastewater samples used in this study were taken from equalization tank of wastewater treatment plant of the studied company. The samples were collected in 20 L plastic drums and stored at 4°C. Chemical oxygen demand (COD, mg/L) and oil-grease (mg/L) were determined according to the 5220 C-Closed Reflux Titrimetric Method and 5520 D Soxhlet Extraction Methods, which given in Standard Methods, respectively [16]. For each parameter characterization was made in duplicate and taking the mean values characterization was determined. The characterization of the wastewater studied was as follows: COD: 9430 mg/L, oil-grease: 346 mg/L and pH: 7.2.

**Treatment Methods**

***Coagulation–flocculation process***

In the study, ferric chloride ($FeCl_3$ - Merck, CAS No: 7705-08-09) and aluminium sulphate ($Al_2(SO_4)_3$ - Sigma-Aldrich, CAS No: 17927-65-0) were tested as conventional coagulants, and sodium hydroxide (NaOH -Merck CAS No: 1310-73-2) and sulfuric acid ($H_2SO_4$ - Merck, CAS No: 7664-93-9) were used for pH adjustment. A series of Jar-test experiments were conducted on wastewater using 2 min rapid mixing at 200 rpm, 15 min slow mixing at 45 rpm and 30 min settling using an anionic polyelectrolyte.

***Ozonation***

An ozone generator manufactured by Degremont with production rate of 1 g $O_3$ per hour was used to supply ozone. Ozonation system was operated in semi-continuous type, i.e., continuous with respect to the gas flow and batch with respect to solution. 2 liter wastewater was filled into 4 liter stainless steel reactor. Ozonation was carried out at the natural pH of the wastewater. Samples were taken in 15, 30, 45, 60, 90, 150, 210, 240 minutes for the wastewater studied. Excess ozone gas passed out through the top of the reactor into a gas-washing bottle containing KI solution. The concentration of ozone in the effluent gas was measured by taking samples from the KI trap during experimental run and titrating the iodine in the samples with $Na_2S_2O_3$ according to Standard Methods [16]. Two gas-washing bottles containing KI solution was connected to the system in parallel with reactor to determine the quantity of ozone applied to the reactor.

## RESULTS AND DISCUSSION

In this study, COD removal efficiencies in dairy industrial wastewater were investigated using variations of coagulation-flocculation and ozone oxidation. Coagulation-flocculation (CF), ozonation (O), ozonation following coagulation-flocculation (CF+O) and coagulation-flocculation following ozonation (O+CF) processes were applied to the wastewater.

**Table 1.** Products produced at the plant (2011)

| Products | Annual amount |
|---|---|
| White cheese | 1029 tons/year |
| Butter | 7839 tons/year |
| Curd cheese | 38250 tons/year |

**Coagulation–Flocculation Process (CF)**

In the coagulation-flocculation process, the optimum pHs were determined by $FeCl_3$ and $Al_2(SO_4)_3$ chemicals. The optimum doses were determined after the optimum pHs were determined. As shown in Table 2, Optimum pHs were determined as pH:6 and pH:7 for 100 mg/L $FeCl_3$ and 600 mg/L $Al_2(SO_4)_3$, respectively. As can be seen from Figure 1, COD removal efficiencies were the highest at pH 7 in the CF process using 94.7 mg/L $Al^{3+}$ and the optimum dose for $Al_2(SO_4)_3$ was determined as pH: 7. When Figure 1 was examined for $Fe^{3+}$, COD removal efficiencies increased to pH: 6 to 32% and then decreased to 28%. Accordingly, the highest removal efficiency was obtained at pH: 6.

Optimum dosage studies were performed at pH: 6 for $FeCl_3$ and at pH 7 for $Al_2(SO_4)_3$. The results of the optimum dosage study are shown in Table 3. As can be seen from the Table 3, the optimum dose for $Al^{3+}$ was 188.4 mg/L and the COD removal efficiency at this dose was determined as 33% (Figure 2). As seen from Table 3, the optimum dosage for $Fe^{3+}$ were found as 172.5 mg/L and COD removal efficiency was found as 45% at this dosage (Figure 3). At these optimum dosages, COD values decreased to 6333 mg/L and

**Table 2.** Results of optimum pH study

| | COD Removal Efficiencies (%) | |
|---|---|---|
| **pH** | **94.7 mg/L $Al^{3+}$** | **34.5 mg/L $Fe^{3+}$** |
| 4 | 27 | 29 |
| 5 | 27 | 29 |
| 6 | 26 | 32 |
| 7 | 28 | 28 |

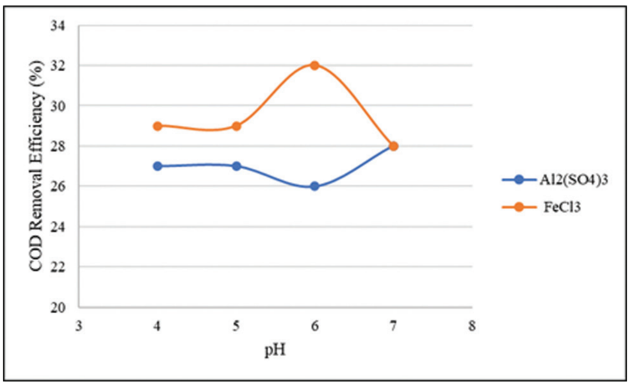


**Figure 1.** Results of optimum pH study (94.7 mg/L $Al^{3+}$, 34.5 mg/L $Fe^{3+}$).

5231 mg/L for $Al_2(SO_4)_3$ and $FeCl_3$, respectively. As a result of coagulation-flocculation study, better removal efficiency was obtained with $FeCl_3$ than $Al_2(SO_4)_3$. As stated in the studies in the literature, iron coagulants generally give better results than aluminum coagulants in wastewater treatment. Similar results were found in this study [17, 18].

**Ozonation Following Coagulation-Flocculation (CF+O)**

The effluent wastewater of the chemical treatment with $FeCl_3$ was used for the ozonation study (CF+O). COD removal efficiencies are given in Table 4 and Figure 4 for CF+O process. As seen in Figure 1, COD removal efficiencies increase with time. Although the removal efficiencies

**Table 3.** COD removal efficiencies of coagulation-flocculation processes with $Al_2(SO_4)_3$ and $FeCl_3$

| Initial COD (mg/L) | $Al^{3+}$ doses (mg/l) | COD removal eff. (%) | COD (mg/L) | $Fe^{3+}$ doses (mg/L) | COD removal eff. (%) | COD (mg/L) |
|---|---|---|---|---|---|---|
| | 62.8 | 24.8 | 7090 | 34.5 | 32.0 | 6414 |
| | 94.7 | 28.3 | 6764 | 68.9 | 40.5 | 5612 |
| 9430 | 125.6 | 26.9 | 6890 | 103.5 | 43.3 | 5347 |
| | 157 | 29.2 | 6679 | 138 | 42.7 | 5403 |
| | 188.4 | 32.8 | 6333 | 172.5 | 44.5 | 5231 |

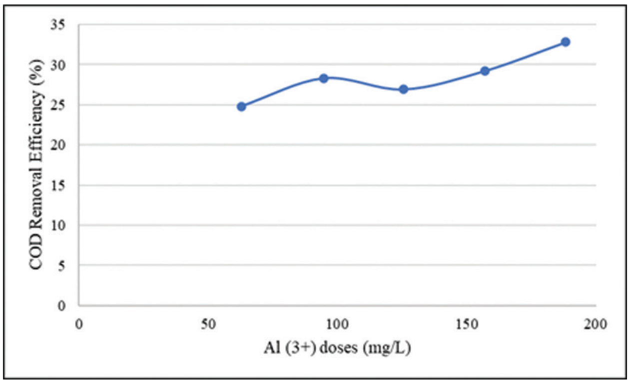


**Figure 2.** Results of optimum dosage study for $Al_2(SO_4)_3$ (pH:7).

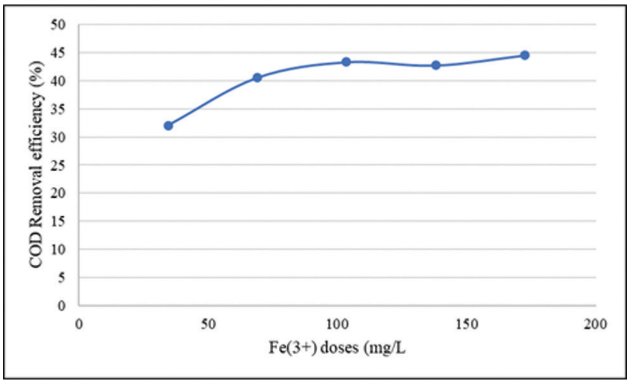


**Figure 3.** Results of optimum dosage study for $FeCl_3$ (pH:6).

**Table 4.** COD removal efficiencies of ozonation process after coagulation-flocculation (Ozone dose applied after 240 minutes; 3856 mg/14462 mg COD = 0.37 mg $O_3$/mg COD)

| Initial COD (mg/L) | Wastewater volume | | 2 L | | | |
|---|---|---|---|---|---|---|
| | **Air Flow Rate** | | **10 L/min** | | | |
| | **Ozone produced by ambient air** | | **1 g/h** | | | |
| | Time (min) | Applied Ozone (mg) | Ozone kept in washing bottle (mg) | Used ozone (mg) | COD (mg/L) | COD Removal Efficiency (%) |
| | 15 | 250 | | | 5180 | 1 |
| | 30 | 500 | | | 5103 | 2.4 |
| | 45 | 750 | | | 4876 | 6.8 |
| 5231 | 60 | 1000 | 144 | 3856 | 4574 | 12.6 |
| | 90 | 1500 | | | 4020 | 23.2 |
| | 150 | 2500 | | | 3685 | 29.6 |
| | 210 | 3500 | | | 3461 | 33.8 |
| | 240 | 4000 | | | 3286 | 37.2 |

increased as the reaction continued, the study was terminated in 240 minutes, considering that the cost of ozonation will also increase over time. As seen in the Table 4 and Figure 4 COD removal efficiency of ozonation after CF process was at most 37%. Since the amount of ozone given by the ozonation system used is 1 g per hour, only 4 grams of ozone could be given in 4 hours. Therefore, although the reaction time of 4 hours seems long, the yield obtained in this process could be obtained in roughly 1 hour, for example, with a generator that gives 4 grams of ozone per hour. As seen in Table 4 by using ozonation system, the obtained ozone dose is 0.37 mg $O_3$/mg COD and this value seems insufficient to oxidize all organic matter. Therefore initial COD of 9430 mg/L decreased to 3286 mg/L after the CF+O process. As a result, the COD removal efficiency of the CF+O process was 65% in total.

**Ozonation**

The COD removal efficiencies obtained after only ozonation process are given in Table 5. As seen from Table 5, despite long ozonation times of 240 min COD removal efficiency was at most 20% (Figure 5). As discussed in the (CF+O) part, the removal efficiencies obtained were low due to the low ozone production capacity of the ozonation system used. Besides this low ozone dose low reaction rate can be attributed to the presence of low and resistant substances with ozone, such as aldehyde, ketone and organic acids [18, 19]. In addition, the presence of hard-to-mineralize materials such as proteins and amino acids in the dairy industry wastewater has prevented the achievement of high COD removal efficiency by ozonation. After 240 minutes, the applied ozone dose was calculated as 0.203 mg $O_3$/mg COD. As seen from the

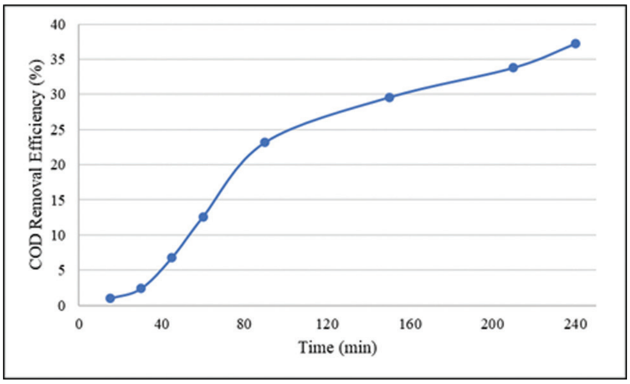


**Figure 4.** COD removal efficiencies of CF+O process (Ozone dose applied after 240 minutes; 3856 mg/14462 mg COD = 0.37 mg $O_3$/mg COD).

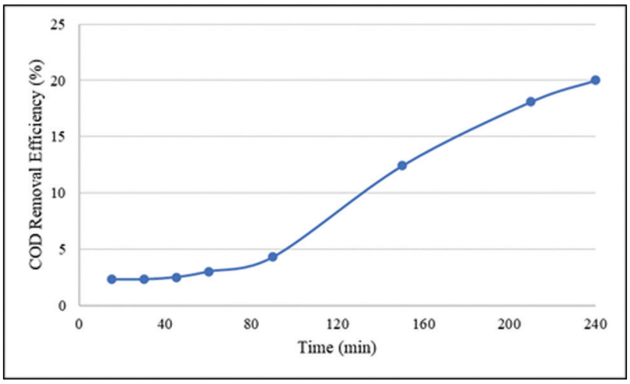


**Figure 5.** COD removal efficiencies of ozonation process (Ozone dose applied after 240 minutes; 3832 mg $O_3$/18860mg COD=0.203 mg $O_3$/mg COD).

**Table 5.** COD removal efficiencies of ozonation process (Ozone dose applied after 240 minutes; 3832 mg $O_3$/18860mg COD=0.203 mg $O_3$/mg COD)

| Initial COD (mg/L) | Wastewater volume | 2 L | | | | |
|---|---|---|---|---|---|---|
| | **Air Flow Rate** | **10 L/min** | | | | |
| | **Ozone produced by ambient air** | **1 g/h** | | | | |
| | Time (min) | Applied Ozone (mg) | Ozone kept in bottle (mg) | Used ozone (mg) | COD (mg/L) | COD Removal Efficiency (%) |
| 9430 | 15 | 250 | 168 | 3832 | 9215 | 2.3 |
| | 30 | 500 | | | 9210 | 2.3 |
| | 45 | 750 | | | 9190 | 2.5 |
| | 60 | 1000 | | | 9146 | 3.0 |
| | 90 | 1500 | | | 9026 | 4.3 |
| | 150 | 2500 | | | 8259 | 12.4 |
| | 210 | 3500 | | | 7723 | 18.1 |
| | 240 | 4000 | | | 7544 | 20.0 |

**Table 6.** COD removal efficiencies of O+CF process

| pH | Initial COD (mg/L) | COD after ozonation process (mg/L) | COD value after CF process (mg/L) | COD Removal efficiency after only CF process(%) | COD Removal efficiency in total(%) |
|---|---|---|---|---|---|
| 6 | 9430 | 7544 | 4527 | 40 | 52 |

**Table 7.** The results of CF, O, CF+O and O+CF operations

| Processes | CF | O | CF+O | O+CF |
|---|---|---|---|---|
| COD Removal efficiency, % | 44.5 | 20 | 65 | 52 |
| Remaining COD, mg/L | 5231 | 7544 | 3286 | 4527 |

Table 5, although ozonation contact time was 240 minutes, it was observed that the ozone dose remained low due to the very high initial COD value. Initial COD of 9430 mg/L decreased to 7544 mg/L after the ozonation process.

***Coagulation-Flocculation Following Ozonation (O+CF)***

In the coagulation-flocculation process after the ozonation process (O+CF), optimum pH and optimum $FeCl_3$ dosages which used in raw wastewater were used. O+CF results are given in Table 6. As can be seen from the table, COD of 7544 mg/L after ozonation decreased to 4527 by coagulation-flocculation process using pH 6 and 500 mg/L $FeCl_3$. Initial COD of 9430 mg/L decreased to 4527 mg/L after O+CF treatment. As a result, it is seen that COD removal efficiency of O+CF operation was 52% in total.

**Comparison of the processes**

The Table 7 shows the results of CF, O, CF+O and O+CF operations. As a result, CF+O operations were found to be more efficient than O+CF operations in total COD removal. It is thought that high molecular oil-grease and TSS were removed using CF initially, thus increasing the ozonation efficiency. The COD removal efficiency of the wastewater directly in CF application (44.5%) was higher in CF application after ozonation (40%). It can be said that ozonation is effective on particulate COD, therefore the efficiency of coagulation-flocculation after ozonation is lower. The COD value of wastewater after 240 minutes of long ozonation time after the CF process was still high. Therefore, due to the high cost that may occur due to the ozonation process, additional studies on biological treatment or biological treatment after CF+O process should be carried out for the treatment of this industry wastewater.

## CONCLUSIONS

In this study, coagulation-flocculation and ozonation alternatives and their variations were used for COD removal in dairy industry wastewater. Only coagulation-flocculation (CF), only ozonation (O), ozonation after coagulation-flocculation (CF+O) and post-ozonation coagulation-flocculation (O+CF) were applied to the wastewater.

As a result of the study, it was found that CF+O processes are more efficient than O+CF processes in total COD removal. In the treatment of wastewater with coagulation-flocculation and ozonation, first chemical treatment with coagulation-flocculation and then ozonation was found to be more efficient in total COD removal than coagulation-flocculation after ozonation. Here, it can be thought that oil-grease and TSS were removed with coagulation-flocculation and thus the efficiency of ozonation increased. The high COD removal efficiency of the raw wastewater in direct coagulation-flocculation application was lower than the coagulation-flocculation application applied after ozonation. In this case, it can be assumed that the ozone formed in the wastewater by ozonation has a negative effect on the coagulation-flocculation efficiency. In addition, it can be said that intermediate and by-products resulting from ozone oxidation change the zeta potential.

## AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

## DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

## CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

## ETHICS

There are no ethical issues with the publication of this manuscript.

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