

Research Article

**Effectiveness of separate ozonation process separately and in combination
with clay to remove sodium dodecyl sulfate from aqueous solutions**

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ABSTRACT

Today, large amounts of surfactants containing sodium dodecyl sulfate (SDS) are put in the domestic use and can be found in urban waste water treatment outputs and treatment inputs. Removing this containment requires high costs and sophisticated technologies. The purpose of this study was to determine the effectiveness of ozone disinfection process separately and in combination with clay to remove sodium dodecyl sulfate from aqueous solutions.

This study was an experimental-interventional research that was conducted at a laboratory scale and in three stages: First, initial concentrations of 10, 30 and 50 mg/L SDS with pH levels of 4, 7, and 10 were transferred to an ozonation reactor containing 0.05 g/hr ozone. Then, ozonation was performed at 2.5, 5, 10, 20, and 30 min. In the second stage, initial concentrations of 10, 30, and 50 mg/L SDS with 0.1, 0.25, and the adsorbent mass of 50 g/L montmorillonite at pH levels of 4, 7, and 10 were ozonized in full combination with air in 2.5, 5, 10, 20, and 30 min. Finally, in the third stage, initial concentrations of 10, 30, and 50 mg/L SDS mixed with 0.1, 0.25, and adsorbent mass of 0.50 g/L montmorillonite were transferred to the reactor and the ozonation was performed at contact times of 2.5, 5, 10, 20, and 30 min. The total SDS and COD values were measured in different stages.

The efficiency analysis of SDS removal by ozonation process indicated minor changes in various pH levels such that an increase in pH from 4 to 10 led to a very slight increase in the efficiency. The absorption process is also influenced by the mass of adsorbent and the initial SDS concentrations so that by increasing the adsorbent mass and decreasing initial concentrations, the absorption efficiency also increased. The efficiency of SDS removal by catalytic oxidation process was affected by pH levels.

Keywords: ozonation, surface adsorption, catalytic, montmorillonite, sodium dodecyl sulfate.

1. INTRODUCTION

Today, large amounts of surfactants containing sodium dodecyl sulfate (SDS) at the end of the current domestic and urban waste water treatment plants can be seen, and the input [1]. Several studies have shown that different amounts of surfactant in surface water and drinking water is measured, as well as low concentrations of surfactants in water levels and severe toxic

effects on aquatic organisms have a variety. Removing these contaminants requires high cost and complicated technology. Recently, it is believed that surface absorption is the simplest and most effective technology to treat water and wastewater and the access to this technology depends on the development and efficiency of absorption. To this end, activated carbon, clay,

zeolites, and some industrial solid waste are widely used to attract ions and organic materials in water and wastewater treatment [2]. Furthermore, studies have shown that the use of such materials reduces the efficiency of biological wastewater treatment system for the removal of organic matter. Ozone is a strong oxidant compound that has successfully been used to treat wastewater containing organic matter. Ozone oxidizes organic compounds by direct oxidation or through free hydroxyl radicals or a combination of these two processes. Organic compounds that have been exposed to ozone will change into intermediate oxygen-containing compounds that are more soluble and therefore are more capable of biodegradation [3]. Absorption is a process in which a soluble material accumulates on the surface of another suitable material. In fact, absorption refers to the transition from liquid to the solid phase [4]. An important factor in designing the system is to select the type of adsorbent material. Given that surfactants have large impact on eutrophication and algal growth and also due to the recent droughts water entering lakes has reduced significantly, resulting in destruction of lakes, finding an effective method for the removal of surfactants seems essential. Given the need for removing significant amounts of surfactants to achieve environmental standards and considering their toxicity effects, the application of methods to reduce surfactants concentrations in wastewater with high efficiency is necessary. As such, this study tries to examine the impact of two effective processes affecting the reduction of surfactants including oxidation with ozone and surface adsorption separately and collectively.

2. Literature Review

A study by Paterson et al., (1999) on linear alkylbenzene sulfonate (LAS) showed that surfactants are decomposed up to 98% in wastewater treatment systems, reducing the treatment efficiency and increasing the number of bacteria in the effluent [5]. Mehrvar et al., (2005)

studied the decomposition of linear alkylbenzene sulfonate (LAS) and found UV/H₂O₂ process would remove over 90% of linear alkylbenzene sulfonate while sodium dodecyl sulfate (SDS) would be removed up to 41% [6].

The results of a study by Isobe et al., (2004) showed that measurable SDS concentrations can be found in samples of surface water and even in drinking water and small SDS quantities after biodegradation have severe toxicity effects on aquatic organisms [7]. Rivera et al., (2006) showed that the efficiency of ozone/activated carbon powder is much higher than that of ozone or ozone/hydrogen peroxide in removing alkylbenzene sulfonate [8]. Olmez-Hanci et al., (2010) studied anionic, cationic, and nonionic textile surfactant degradation with the H₂O₂/UVC process. The results indicated that H₂O₂/UVC process plays a significant role in removing surfactants only under certain circumstances [9]. In a study by Rivera-Utrilla et al., (2010) on advanced oxidation of sodium dodecyl benzene sulfonate (SDBS) with hydroxyl radicals and sulfate, it was found that Fenton method showed a good efficiency in acidic conditions in SDBS removal [10]. A study by Rezai et al., (2007) showed that the amount of activated carbon and contact time are directly related to the percentage of removal of sodium dodecyl sulfate (SDS). It was also noted that this percentage would decrease by increasing the amount of surfactant [11]. Liu et al., (2011) studied the removal of surfactant by activated carbon catalytic ozonation and found that the use of the combined system (activated carbon/ozone) is much more effective in removing surfactants than activated carbon sorbent alone [12]. The results of a study by Nourmoradi et al., (2012) showed that montmorillonite as a type of clay can be regarded as one of the most effective choices for the removal of organic contaminants, benzene, toluene, ethyl benzene, and Xylene (BTEX) from aqueous solutions as a result of

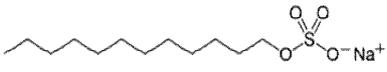
high contact area, low cost, non-toxicity, and high absorption capacity [13].

3. MATERIALS AND METHODS

3.1 SDS composition

Sodium dodecyl sulfate (SDS) or sodium lauryl sulfate (SLS) powder was produced by Merck Corporation, Germany and its chemical characteristics are given in Table 1:

Table 1: SDS chemical composition

| | |
|--------------------|---|
| Chemical structure |  |
| Color | White |
| Chemical formula | $C_{12}H_{25}NaO_4S$ |
| Molecular weight | 288.38 g/mol |

3.2 Montmorillonite composition

Montmorillonite was used in this study as an absorbent with the following physical and chemical properties as shown in Table 2:

Table 2: Montmorillonite physical and chemical composition

The composition of the montmorillonite used in this study.

| Chemical composition (%) | |
|--------------------------------|-------|
| SiO ₂ | 60 |
| Al ₂ O ₃ | 20.03 |
| Fe ₂ O ₃ | 2.31 |
| Na ₂ O | 3.02 |
| MgO | 4.02 |
| F ₂ O ₃ | 0.05 |
| K ₂ O | 0.13 |
| CaO | 1.46 |
| TiO ₂ | 0.23 |
| MnO | 0.03 |
| H ₂ O | 8.71 |
| Mineralogical analysis (%) | |
| Montmorillonite | 98 |
| Feldspar | 0.5-1 |
| Quartz | 0.5-1 |
| Other | |
| CEC (meq/100g) | 108 |
| Limit of ignition (%) | 7.03 |

3.3 Reactor properties

The overall design of the reactor used in this study is shown in Figure 3. As it can be seen, the required ozone was supplied by an ozone generator with the capacity of 3 gr/hr by using pure oxygen. Then the produced ozone entered into the reactor from the bottom through a diffuser (air stone) in the form of tiny bubbles. In this study, a tubular column made of Plexiglas with an internal diameter of 4 cm and a height of

1 meter was used to implement the process. Ozone flow into the reactor was controlled via a rotameter (gas flow meter). The gas flow from the reactor may also have some ozone which is hazardous and can pollute the air and expose people to some dangers. Therefore, to prevent the release of ozone in ambient air and also measure the concentration of ozone, the ozone output was directed to a 1-liter glass tank containing 2% potassium iodide solution as shown in Fig.1:

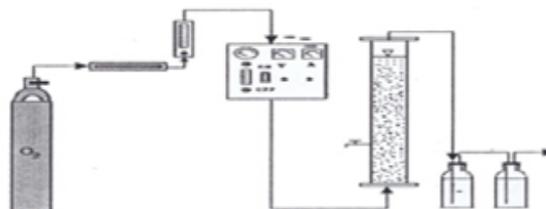


Fig. 1 Reactor used in the study

3.4 Method

The present study is an experimental study that was conducted discontinuously at a laboratory scale in three stages: ozonation, surface absorption with montmorillonite, and catalytic oxidation or a combined process (ozone/ surface adsorption). In addition, three absorbent doses (0.1, 0.25, and 0.5) and three SDS concentrations (30, 10, and 50) during the 5 time periods (2.5, 5, 10, 20, and 30 min) were used. Besides, the experiments were performed with three iterations at three pH levels (4, 7, and 10) to determine the optimal pH level. The collected data were analyzed using SPSS 13 by ANOVA. Besides, the impact of independent variables on the removal efficiency was determined using regression analysis.

3.5 Tests

To perform SDS removal tests, SDS initial concentrations (10, 30, and 50 mg/L) were prepared and were treated alternatively at three time periods (2.5, 5, 10, 20, and 30 min) in a reactor with a volume of 1 L. To measure SDS levels, a Dr/2000 Spectrophotometry was used. To adjust initial pH levels of solutions, a Mi 151

Wagtek pH meter was used. In the ozonation process, initial concentrations were transferred to the ozonation reactor. Then SDS concentrations were prepared at the determined time periods. In the surface absorption in the presence of montmorillonite, initial concentrations in contact with 0.1, 0.25, and 0.50 g/L montmorillonite were transferred to the reactor. To ensure adequate mixing at the determined times, COD and SDS concentrations were prepared after montmorillonite particles removed from the solution using a 0.45 μ filter. In the mixed catalytic oxidation process (ozone and surface absorption), montmorillonite was mixed and transferred to the reactor. After removing montmorillonite particles from the solution using a 0.45 μ filter, COD and SDS concentrations were measured using standard water and wastewater testing method.

4. Results

4.1 Effects of pH and SDS initial concentrations on ozone oxidation process

The impact of pH on ozonation to remove SDS with 0.05 gr/h ozone at intervals of (2.5, 5, 10, 20, and 30 min) was measured as shown in Fig. 2. As can be seen, the SDS removal efficiency at 30 min has increased from 59% and 64% by an increase in pH level from 4 to 10.

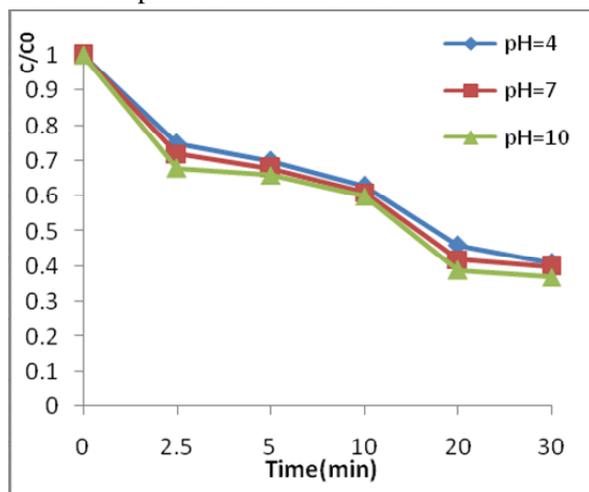


Fig. 2 Impact of pH level on ozonation process in SDS removal in terms of initial concentrations

To test the effect of initial SDS concentrations on ozonation process; pH and ozone levels were kept fixed at 10 and 0.05 gr/h. The results showed that increased initial SDS concentrations would reduce the efficiency in a way that when SDS concentration increased from 10 to 50 over 30 min, the efficiency increased from 64.2% to 62%. The COD removal was then measured after the reaction at a 30 min interval. The results suggested that increased SDS concentrations would result in reducing the SDS removal efficiency and resulting COD such that increasing SDS concentrations from 10 to 150 mg/L, the SDS removal efficiency decreased from 37% to 20%.

4.2 Effects of pH, absorbent mass, and initial SDS concentrations on surface absorption

Fig. 3 shows the impact of pH on SDS absorption on montmorillonite surface at 2.5, 5, 10, 20, and 30 min. as can be seen, pH variations had no considerable impact on SDS absorption by montmorillonite as the SDS absorption efficiency has reached from 67% to 70% because of an increase in pH level from 4 to 10.

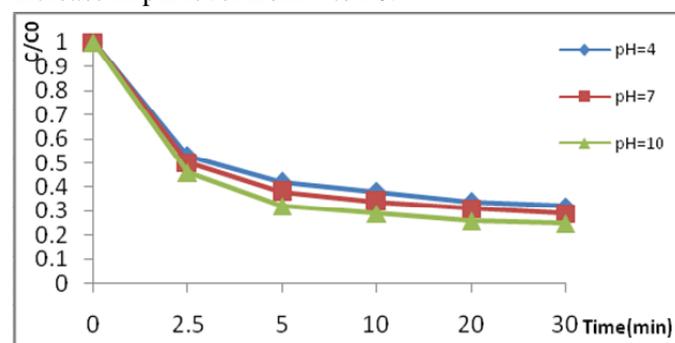


Fig. 3 Impact of pH level on SDS absorption in terms of initial concentrations

To investigate the impact of absorbent mass on SDS absorption, various absorbent masses (0.1, 0.25, and 0.50 g/L) at pH = 7 and concentration of 10 mg/L SDS were tested. The results indicated that increasing the absorbent mass would increase the removal efficiency as in a 30 min contact time, increasing the absorbent mass from 0.1 to 0.5 g/L the removal efficiency would

increase from 59% to 92%. The results for 2.5, 5, 10, 20, and 30 min intervals are shown in Fig. 4:

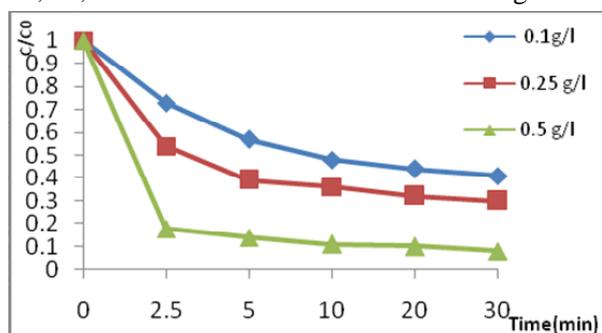


Fig. 4 Impact of pH level on SDS absorption in terms of initial concentrations

The COD removal was tested after the reaction time (30 min) and the results indicated that increasing the absorbent mass would increase the efficiency of SDS absorption and COD removal from the solution as increasing the absorbent mass from 0.1 to 0.5 g/L the removal COD efficiency would increase from 60% to 82%.

To test the impact of SDS initial levels on the surface absorption, pH and montmorillonite mass were considered fixed at 7 and 0.5 g/L, respectively. The results of the impact of SDS concentration levels on the removal efficiency at 2.5, 5, 10, 20, and 30 min were 83%, 86%, 89%, 90%, and 92%, respectively. In addition, increasing SDS concentrations would decrease the removal efficiency so that at SDS concentration levels of 10, 30, and 50 mg/L, the removal efficiency values at pH = 7 and a 30 min intervals were 92%, 87%, and 86%, respectively. The COD removal was tested after the reaction time (30 min) and the results indicated that increasing SDS concentrations would decrease the SDS removal efficiency and the resulting COD values in a way that increasing SDS concentration from 10 to 50 mg/L the removal COD efficiency would decrease from 60% to 82%.

4.3 The impact of pH, absorbent mass, and SDS initial concentration on the effectiveness of catalytic oxidation process (ozone/adsorption)

Fig. 6 shows the impact of pH on catalytic oxidation process at concentration level of 30 mg/L, montmorillonite 30 mg/L, and ozone level of 0.05 g/h at different time intervals (2.5, 5, 10, 20, and 30 min). As it is evident, increasing pH level from 4 to 7 has no effect on the removal efficiency but increasing pH from 7 to 10 increases the removal efficiency. Besides, the efficiency values for pH levels of 7, 4, and 10 are 84.1%, 84%, and 85%, respectively.

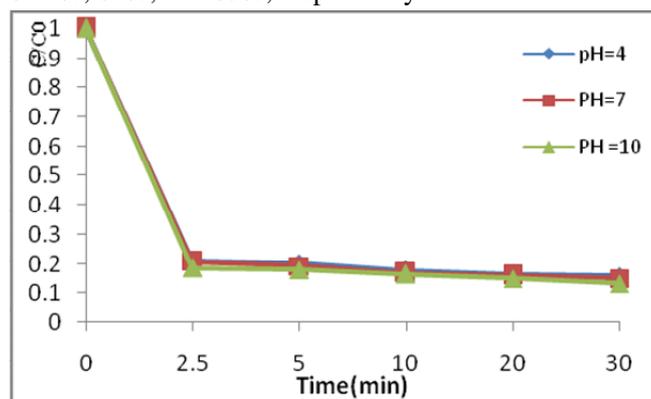


Fig. 5 Impact of pH level on SDS removal efficiency through catalytic oxidation (CO = 30 mg/L, absorbent mass = 0.5, ozone = 0.05 g/h)

The impact of absorbent mass on SDS removal in catalytic oxidation for different masses (0.1, 0.25, and 0.5 g/L) at initial concentration, pH, and ozone levels of 30 mg/L, 10, and 0.05 g/h was tested. The results indicated that the removal efficiency values at 30 min for absorbent masses of 0.1, 0.25, and 0.5 g/L were 75%, 81%, and 86.6%, respectively. To test the impact of SDS initial concentrations catalytic oxidation, pH, montmorillonite, and ozone levels were 10, 0.5 g/L, and 0.05 g/L, respectively. The results of SDS concentration on the removal efficiency at 2.5, 5, 10, 20, and 30 min show that the removal efficiency values at SDS concentrations of 10, 30, and 50 mg/L after 30 min reached 81.8%, 73.3%, and 69%, respectively. The removal efficiency at SDS concentrations of 10, 30, and 50 mg/L after 30 min reached 81.8%, 73.3%, and 69%, respectively.

4.3 Comparing SDS removal during absorption, ozonation, and catalytic oxidation

The removal of SDS concentrations through oxidation by ozone, absorption on montmorillonite, and catalytic oxidation is shown in fig. 6. Besides, a comparison of COD removal in these three processes is shown in Fig. 7:

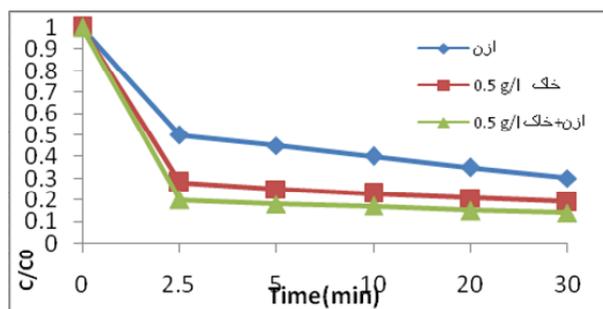


Fig. 6 SDS removal through oxidation by ozone, absorption on montmorillonite, and catalytic oxidation (SDS = 30 mg/L, ozone = 0.05 g/hr, pH = 10)

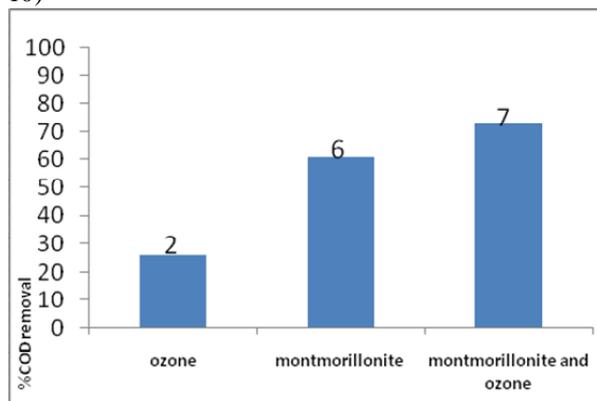


Fig. 7 Comparing COD removal through SDS in ozonation, absorption on montmorillonite, and catalytic oxidation (SDS = 30 mg/L, ozone = 0.05 g/hr, pH = 10)

5. DISCUSSION AND CONCLUSION

The results of many studies on ozonation have shown that pH plays a very important role in the oxidation process. However, the results of this study showed that an increase in pH would increase the removal efficiency. Given pH level in the solution in ozonation process, ozone reacts to the pollutant in two ways: Ddirect oxidation in

acidic conditions is a major factor in SDS decomposition while under alkaline conditions, oxidation indirectly remove SDS by hydroxyl radicals [14]. Yang et al., (2007) stated that the decomposition rate of nitrobenzene by ozone is very low when pH = 2 but the decomposition rate increases as pH raises [15]. Garoma et al., (2010) also found that increasing pH from 2 to 10 would speed up sulfamethoxazole removal [16]. Sirinvasan et al., (2009) stated that the increase in pH at short intervals improves the leather dye removal by ozone, but at long-term the removal in solutions with different pH levels shows very close values [17]. The results of the present study indicated that there is a negative relationship between the increase in the removal and the initial concentration of SDS. The removal efficiency values at 30 min and in the SDS initial concentrations of 10, 30, and 50 mg per liter were 64.2%, 62.8%, and 61%, respectively while COD removal efficiency at 30 min in the initial concentration of 10, 30, and 50 mg per liter is 37%, 26%, and 20%, respectively. The reason is that the removal of SDS by destroying its molecular structure has occurred very rapidly. However, the residuals of SDS organic structure that were measured via COD in this study have remained in the medium for longer times and their removal was more difficult. According to studies conducted by Gilbert (1988), Contreras (2003), and Sangave (2007), it seems that in ozonation process, color is removed faster and in greater amounts than COD does. A possible reason is the existence of compounds containing function groups sensitive to ozone which give color to the solution and their structure is changed when reacting to ozone, reducing the color saturation accordingly [18, 19, 20].

The results of several studies on the surface absorption indicated that pH in the reaction environment is one of the main factors that must be taken into account when considering the absorption process as it affects ionization of pollutants and the surface loading of absorbents.

However, the results of the present study show that an increase in pH and alkaline solution does not increase significantly the removal efficiency (Fig.1). Nourmoradi et al., (2012) studied the removal of organic contaminants, benzene, toluene, ethyl benzene, and Xylene (BTEX) by montmorillonite and concluded that the increased pH does not have a great impact on the increased removal of BTEX. This shows the high stability of the adsorbent when pH is in the range of 2-12 [13]. According to findings from this study, absorption of pollutants increases as the adsorbent mass increases but closer examination reveals that while increasing the adsorbent mass would increase SDS removal, SDS absorption per unit mass of adsorbent would decrease when SDS concentration is fixed. A possible explanation is the non-saturation of active points in absorbing contaminants so that when the adsorbent mass increases, the active points on the absorbing surface are not used in full capacity and this reduces the absorption level per unit mass of adsorbent (Fig. 2). In a study by Khosravi et al., (2012) on the uptake of phenol from aqueous solutions using carbonated sesame tree bark, it was found that by increasing the mass of adsorbent from 5.0 grams per liter to 4 grams per liter, the adsorbent capacity decreased, but increased amounts of the adsorbent raised the removal efficiency [21]. Given that the reduction of COD occurs as a result of SDS molecules adsorbed onto the adsorbent, COD reduction in samples is in line with SDS reduction (Fig.3). Variations in SDS initial concentration are among other factors that affect the absorption rate in these systems. According to the results of this study, SDS absorbed per unit mass of adsorbent would increase as SDS concentration rises from 10 to 50 milligrams per liter while the removal efficiency decreases (Fig. 4). These results indicate that SDS absorption per unit mass of adsorbent is a function of SDS initial concentration, so that the absorption per unit mass of adsorbent increases at higher

concentrations. Such results have been supported by Asgari and Qanizade (2009) and Mehmet et al., (2004). The reason for why the absorption rate increases at higher concentrations can be due to the fact that in such cases the mass transfer force increases and it overcomes the force resisting against the absorption. This creates a strong driving force which transfers contaminants from the liquid phase into the fluid absorbing interface, thus increasing the rate of absorption per unit mass [22, 23]. If this is considered in terms of the efficiency of SDS removal, it can be found that by increasing SDS initial concentrations, the removal efficiency decreases. This depends on number of contaminant moles available and the active surface where absorption occurs so that at a constant mass of adsorbent, the active absorbing surface remains fixed but the number of contaminant moles available in the reaction medium increases and this in turn reduces the removal efficiency. In this study, at constant mass of adsorbent, by increasing the initial concentration of the contaminant from 10 to 50 mg per liter, ratio of the number of contaminant moles to the surface available increased which resulted in decreased absorption efficiency as supported by other studies (23, 24). The results of the present study indicated that catalytic oxidation efficiency is affected by pH so that its increase from 4 to 7 has no impact on the removal efficiency while a corresponding increase from 7 to 10 will rise the removal efficiency. Besides, higher removal efficiency in catalytic oxidation compared with that of ozonation can be attributed to the impact of soil on hydroxyl absorption. In a study by Nassiri et al., (2012) on leachate treatment using ozonation in the presence of activated carbon, it was concluded that an overall increase in pH from 3 to 10 would improve the efficiency of the oxidation process. Ozone in acidic conditions directly reacts with organic components but it decomposes in alkaline conditions and causes the formation of hydroxyl radicals with higher

oxidation power than ozone. It should be noted that the role of activated carbon in absorbing and removing contaminants and its catalytic ability should not be ignored. Chemical structure of the activated carbon affects the rate of decomposition of ozone and hydroxyl radical formation and thereby removing the organic loading [25].

The results of this study indicated that as the absorbent mass increases during catalytic oxidation, the removal efficiency improves significantly. Besides, it was noted that there is no considerable difference between SDS and COD removal efficacy rates. The results of a study by Nassiri et al., (2011) on the leachate treatment using ozonation in the presence of activated carbon showed when the treatment is performed in the absence of activated carbon, the rate of COD removal after 60 min was 57%, which increased to 73% and 84% when 50 and 100 grams per liter of carbon were added [26]. This is consistent with the results of this study. SDS initial concentration variations also affect catalytic oxidations so that an increase in SDS initial concentrations would reduce the removal efficiency. However, it was noted that there is no considerable difference between SDS and COD removal efficacy rates.

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