Araştırma Makalesi / Research Article

Acid Red 17, Acid Yellow 11, Direct Yellow 12, Direct Blue 86 ve Mordant Violet 40 Boyalarının Karışımını İçeren Suyun Arıtılması İçin Gelişmiş Oksidasyon İşlemi

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Öz

Anahtar kelimeler
Acid Red 17;
Acid Yellow 11;
Direct Yellow 12;
Direct Blue 86;
Mordant Violet 40;
İleri oksidasyon

Boyaların atık sudan uzaklaştırılması hem çevre hem de üreticiler için tekstil endüstrisindeki en büyük sorunlardan biridir. Bu makale, beş farklı boya (Asit Kırmızısı 17, Asit Sarısı 11, Doğrudan Sarı 12, Doğrudan Mavi 86 ve Mordan Menekşe 40) içeren sentetik atık su karışımını arıtmak için ultraviyole (UV) ile birlikte ozon (O_3) ve O_3 uygulama olasılığı farklı kromofor ve ankraj gruplarına sahip boyaların renk giderme etkinliği için incelemiştir. Temas süresi, optimum çalışma koşullarını elde etmek için UV ile kombine O_3 veya O_3 akışı altında bir kesikli reaktörde incelenmiştir. Sonuçlar, O_3 uygulamasının UV destekli ozonlamadan daha etkili olduğunu göstermiştir. Bu karışımdaki her bir boyanın ileri oksidasyonu, aynı boyanın çözelti içinde ayrı ayrı çıkarılmasıyla karşılaştırıldı. Sonuçlar, boyayı karışımdan ve su çözeltisindeki tek boyayı çıkarmak için gereken sürede yalnızca küçük bir fark olduğunu kanıtlamıştır.

Advanced oxidation process for treatment of water containing mixture of Acid Red 17, Acid Yellow 11, Direct Yellow 12, Direct Blue 86 and Mordant Violet 40 dyes

Abstract

Keywords
Acid Red 17;
Acid Yellow 11;
Direct Yellow 12;
Direct Blue 86;
Mordant Violet 40;
Advanced oxidation

The removal of dyes from wastewater is one of the biggest problems in the textile industry for both environment and manufacturers. This study examined the possibility of applying ozone (O_3) and O_3 in combination with ultraviolet (UV) to treat a mixture of synthetic wastewater containing five different dyes (Acid Red 17, Acid Yellow 11, Direct Yellow 12, Direct Blue 86 and Mordant Violet 40) for the decolorization efficiency of dyes with different chromophore and anchor groups. The contact time was studied in a batch reactor under the flow of O_3 or O_3 combined with UV to achieve optimal operating conditions. The results showed that the application of O_3 was more effective than UV-assisted ozonation. The advanced oxidation of each dye in this mixture was compared with the removal of the same dye in the solution separately. The results proved that there is only a slight difference in the time required to remove the dye from the mixture and the single dye in the water solution.

1. Introduction

The textile industry is one of the most important industries in Egypt, considering the number of companies, local production costs and export costs. On the other hand, such an industry is considered a major contributor to pollution, currents, and especially wet processes that use hazardous chemicals in that industry. About 31 public sector employees and 3000 private sector companies and joint ventures are affiliated with the Egyptian textile industry (Sultan 2002, EPAP 2003, El Nemr 2012a). Most of the synthetic dyes used in the industry are distributed in different sectors such as textile dyes (60%), paper (10%) and plastics (10%) (Slokar and Majcen Le Marechal 1998, Oneill *et al.* 1999, Robinson *et al.* 2001, Guivarch *et al.* 2003, Sun and Yang 2003).
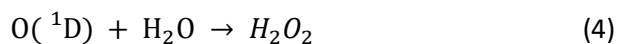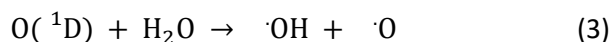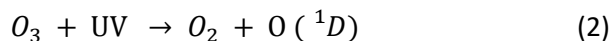
The presence of organic pollutants such as dyes, pesticides and surfactants in the environment poses a major threat to freshwater and marine environments, as they are carcinogenic and non-biodegradable (Mahmoodi and Arami 2010, Salem *et al.* 2013, 2014a, b, Amini *et al.* 2011, Ragab *et al.* 2016, El Nemr *et al.* 2016a, b, Asiri *et al.* 2020). Different methods have been used in the treatment of wastewater such as adsorption, advanced oxidation and photodegradation (El Nemr 2007, 2009, 2012b, El Nemr *et al.* 2010, Hassaan *et al.* 2017a, b, Helmy *et al.* 2018, Alghamd *et al.* 2019, El-Nemr *et al.* 2020a, b, Hassaan *et al.* 2020).

The reaction of ozone with wastewater compounds is in two different ways. These are either by direct molecular or indirect radical chain reactions (Glaze *et al.* 1987, Steahlin and Hoigne 1982, Arslan and Balcioglu 2000, Hassaan *et al.* 2016, 2017a). The reaction mechanisms of Ozone occurring at alkaline pH are listed below in five steps;

$$3 O_3 + H_2O \rightarrow 2 \cdot OH + 4 O_2 \tag{1}$$
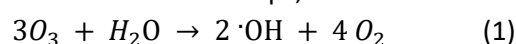
The following equations (Eq. 2 to 5) have been suggested for UV-assisted ozonation treatments (Hassaan *et al.* 2017b, Hassaan and El Nemr 2017, El Nemr *et al.* 2017, 2018). The presence of different oxidizing agents in the removal of dyes from wastewater can create a synergistic effect and increase the removal.

$$O_3 + UV \rightarrow O_2 + O(^1D) \tag{2}$$

$$O(^1D) + H_2O \rightarrow \cdot OH + \cdot O \tag{3}$$

$$O(^1D) + H_2O \rightarrow H_2O_2 \tag{4}$$

$$H_2O_2 + hv \rightarrow 2 \cdot OH \tag{5}$$

Pathogen-containing organic and inorganic compounds in wastewater react with ozone gas given to the water. The first reaction occurs with ozone with chromophore group dyes, where ozone reacts with organic compounds at the double bonds of nitrogen, carbon (N=N, C=C) and aromatic rings (Wijannarong *et al.* 2013). Textile effluents contain various dyes, so it is very important to remove the dye mixture using different new methods instead of a single dye solution removal with the existing methods. To the best of our knowledge, few studies have been reported on the degradation of a synthetic mixture of dyes using an advanced oxidation process (Wijannarong *et al.* 2013, Sancar and Balci 2013, Gupta *et al.* 2015). Therefore, this study aims to examine the effectiveness of O_3 and UV/O_3 application techniques for the removal of the synthetic dyes mixture. The effect of ozone and ozone + UV was studied at the initial dyes mixture pH 7. The second aim of this research work is to propose a simple technique to monitor the decay of the color during the reaction of ozone with a mixture of dyes.

2. Materials and Methods

A mixture of the five dyes was used without further purification. A stock solution of 1000 mg/L of each dye was prepared using double distilled water. By diluting this solution, working aqueous solutions (100 mg/L) of each dye were prepared. In this study, absorbance measurements were performed in a UV-VIS spectrophotometer (Analyticjena Spekol 1300 UV-VIS spectrophotometer (model no. 45600-02, Cole Parmer Instrument Co., USA)) to determine the dye concentrations of 5 different dyes (Fig. 1.) at the maximum wavelengths shown in Table 1. The

decolorization effect of the color removal rate was calculated as:

Color removal efficiency % $= \frac{A_0 - A_t}{A_0} \times 100$ (6)

Where A_0 and A_t are the initial and quantified absorbances of the samples at different time intervals.

Table 1: The maximum wavelength (λ max) of some dyes that used in this study.

Dye	λ_{max}	M.W.	M.F.
A.R.17	518 nm	502.43	$C_{20}H_{12}N_2Na_2O_7S_2$
A.Y.11	407 nm	380.35	$C_{16}H_{13}N_4NaO_4S$
D.Y.12	389 nm	680.66	$C_{30}H_{26}N_4Na_2O_8S_2$
D.B.86	594 nm	780.17	$C_{32}H_{14}CuN_8Na_2O_6S_2$
M.V.40	510 nm	400.77	$C_{16}H_{10}ClN_2NaO_5S$

A cylindrical 200 mL glass reactor was used in the ozonation process (Fig. 2). The ozone generator was connected to reaction vessels with Teflon tubing. A fresh dye sample (200 mL) was used in each study due to the small reactor volüme. The ozone fed to the system was carried out with an ozone generator (model N 1668A, power 18 W, volume AC 220 V/50 Hz) with a flow rate of 500 mg O_3/h. The ozone leaving the reactor was captured in two consecutive bubblers with potassium iodine (KI) (2%) aqueous solution (Hassaan *et al.* 2017a,b). In addition, further tests were performed utilizing the AOPs (O_3/UV), therefore a Horizontal Clean Bench/Laminar Flow Cabinet (Bw-LFH1300) with a power of 30 W and a UV low-pressure mercury lamp (254 nm) was used. The pH measurements were made using a pH meter from JENCO Electronics, LTD (model 6173, serial number JC 05345).
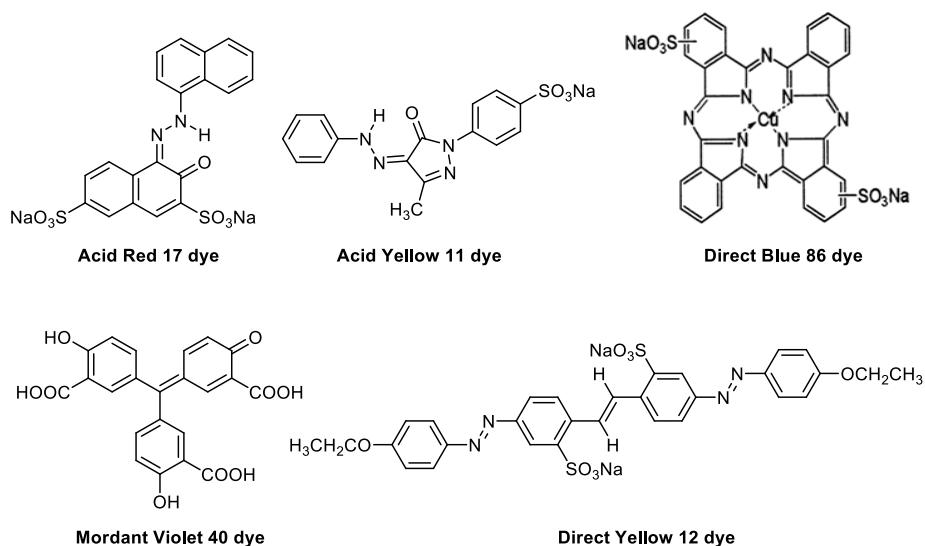
Fig. 1. Structure of Acid Red 17 dye, Acid Yellow 11 dye, Direct Blue 86 dye, Mordant violet 40 dye, and Direct Yellow 12 dye.
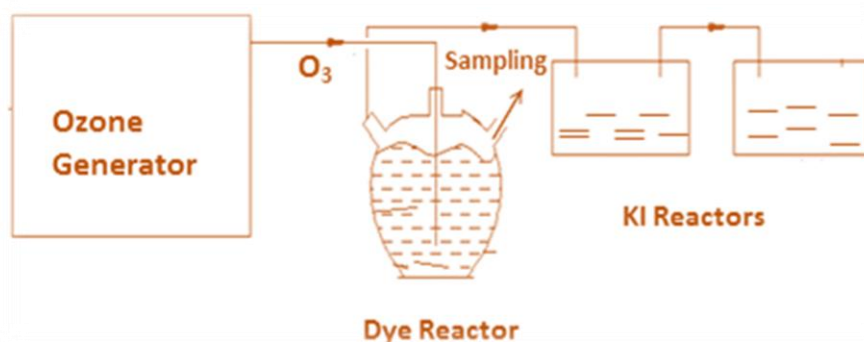
Fig. 2. Design of the ozonation experiment.

3. RESULTS AND DISCUSSION

3.1. Different standard curves

Figure 3 shows the standard curve for the five different dyes in the mixture and single water solution at appropriate λ_{max} for each dye.

3.2. Effect of Ozone

Figure 4 shows the dye removal efficiency of the dye mixture of the 5 studied dyes. The color removal efficiency for the studied dyes (Acid Red 17, Acid Yellow 11, Direct Yellow 12, Direct Blue 86, and Mordant Violet 40) with an initial concentration of 100 mg/L were 99.8, 98.9, 98.9, 100 and 99.8% at 180, 210, 240, 180, and 180 minutes, respectively for O_3 only.
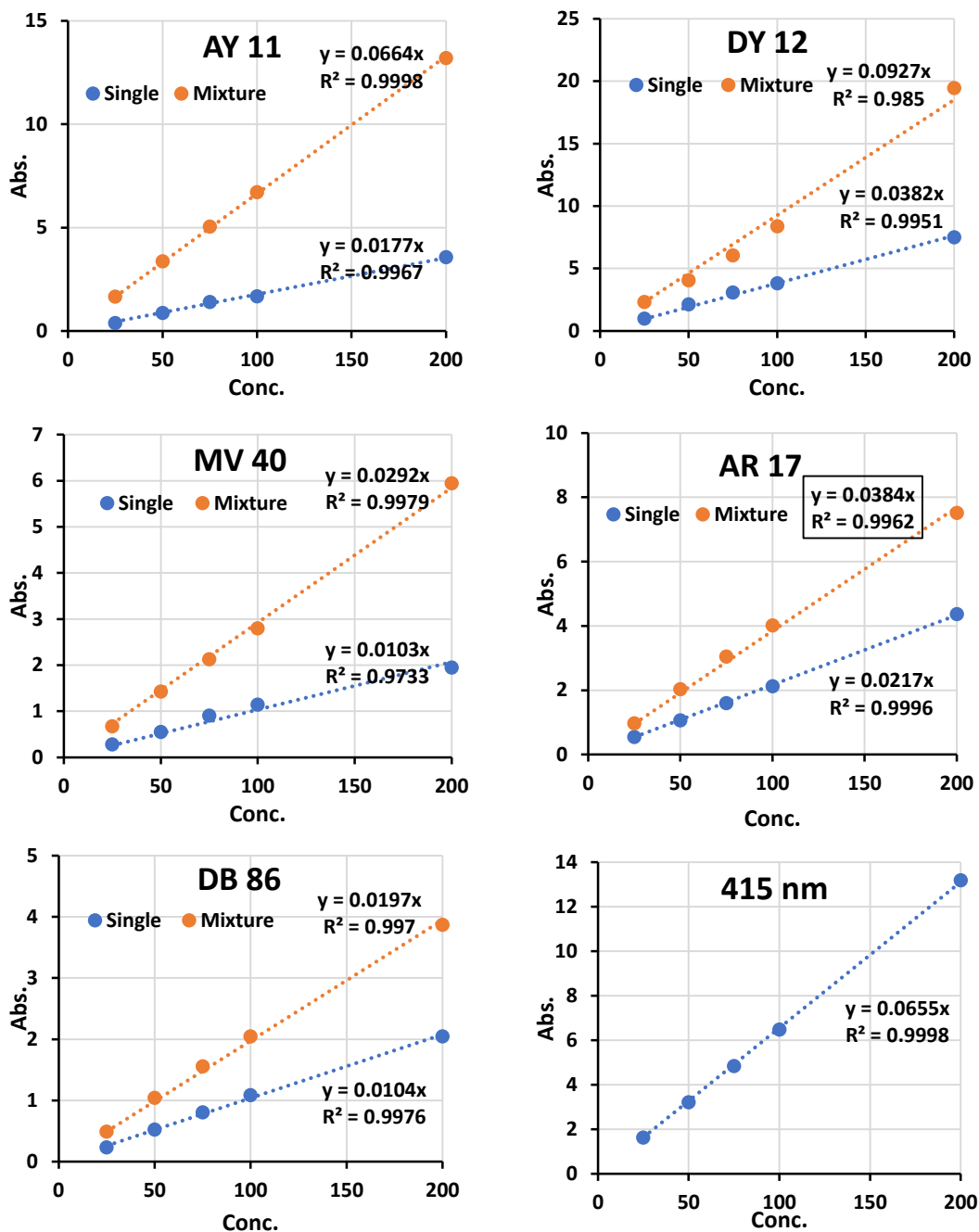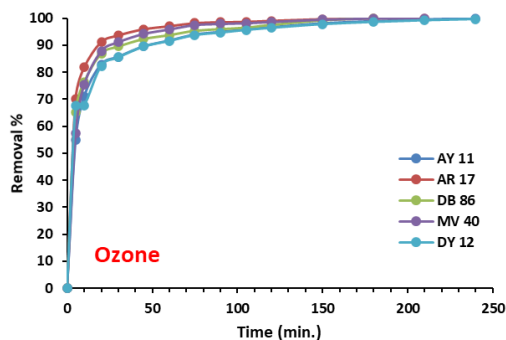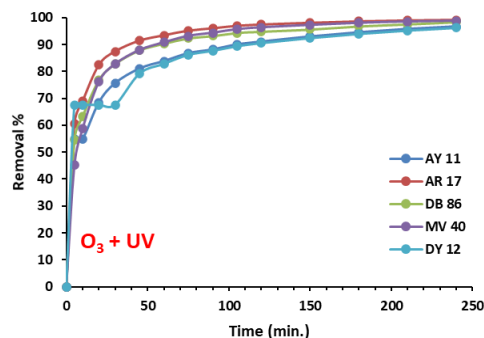

Fig. 3. Standard curves of the single and mixture dyes.

Fig. 4. Effect of ozone on dyes mixture (Solution volume = 200 mL, O_3 = 500 mg/h, initial solution pH (6-7), 100 mg/L dye concentration and different λ_{max}).
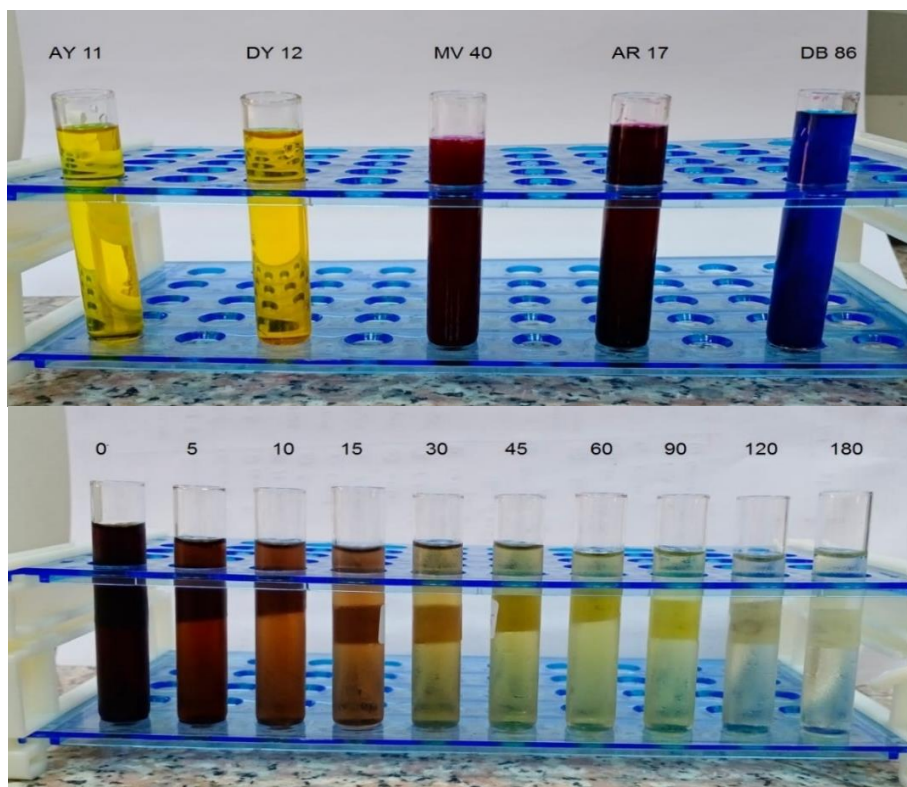

Fig. 5. Effect of advanced oxidation processes (O_3 + UV) on Dyes mixture (Solution volume = 200 mL, O_3 = 500 mg/h, initial solution pH (6-7), 100 mg/L dye concentration and different λ_{max}).

3.3. Effect of Ozone + UV

Fig. 5 shows the color removal efficiency of the 5 studied dyes mixtures. The color removal efficiency for the studied dyes (Acid Red 17, Acid Yellow 11, Direct Yellow 12, Direct Blue 86 and Mordant Violet 40) with an initial concentration of 100, mg/L were 98.4, 92.6, 88.4, 98 and 98.3 %, respectively, after 240 minutes contact time of UV assisted O_3 treatment.

Fig. 6 shows the color of the five studied dyes and the color removal of the dyes mixture after 180 minutes of contact time. The studies showed that the AY11 and DY12 were more resistant to the advanced oxidation process than the other three dyes MV 40, AR 17 and DB 86 dyes.

Fig. 6. Coloring the five studied dyes and removing color from the mixture of pigments with a time of up to 180 minutes from the time of contact.

3.4. Absorbance measurement at different λ_{max} for mixture and single dyes

Fig. 7 shows the effect of O_3 and UV + O_3 on the degradation of the dyes mixture. Fig. 7a shows the effect of ozonation and advanced oxidation processes (AOP) on the decolorization of Acid Yellow 11 (AY11). (O_3 = 500 mg/h, solution volume = 200 mL, initial mixture solution pH (6-7), initial single solution pH (9), 100 mg/L dye concentration and λ_{max} = 407 nm).

Fig. 7b shows the effect of ozonation and AOP on the decolorization of Acid Red 17 (AR17). (O_3 = 500 mg/h, solution volume = 200 mL, initial mixture solution pH (6-7), initial single solution pH (11), 100 mg/L dye concentration and λ_{max} = 518 nm). Fig. 7c shows the effect of ozonation and AOP on the decolorization of Direct Blue 86 (DB86). (O_3 = 500 mg/h, solution volume = 200 mL, initial mixture solution pH (6-7), initial single solution pH (11), 100 mg/L dye concentration and λ_{max} = 594 nm)
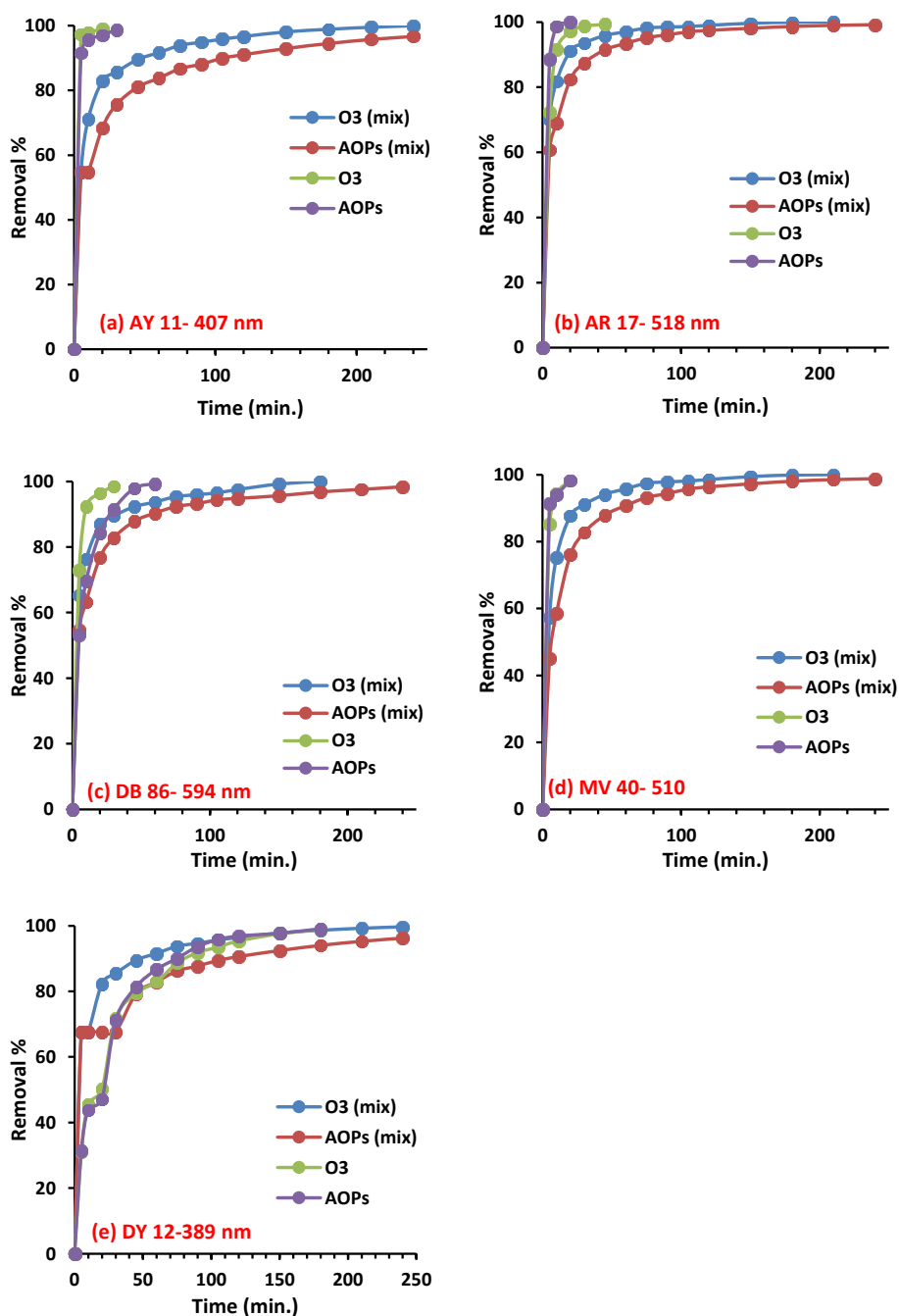

Fig. 7. The effect of O_3 and UV + O_3 on the degradation of dyes mixture.

Fig. 7d shows the effect of ozonation and AOP on the decolorization of Mordant Violet 40 (MV40). (O_3 = 500 mg/h, solution volume = 200 mL, initial mixture solution pH (6-7), initial single solution pH (9), 100 mg/L dye concentration and λ_{max} = 510 nm). Fig. 7e shows the effect of ozonation and AOP on the decolorization of Direct yellow 12 (DY12). (O_3 = 500 mg/h, solution volume = 200 mL, initial mixture solution pH (6-7), initial single solution pH (9), 100 mg/L dye concentration and λ_{max} = 389 nm). It is clear from the above-mentioned Fig. 7 for the dyes mixture that ozone is very effective in the removal of the color even in the dyes mixture and ozone alone is more effective than AOPs. The advanced oxidation processes (AOPs) showed that as the amount of dye increases, many different intermediates are formed during the degradation of the parent dye, which can interfere with the oxidation process. Such suppression would be more obvious in the existence of an increased amount of degradation intermediates formed at heightened dye concentration (Song *et al.* 2008). Therefore, full-color removal will take longer for dyes with higher initial dye concentrations. Another reason for this finding may be related to the structure of the dyes as mentioned by Hassaan *et al*. (2017a, b) in MV40, AY11 and DB 86 dyes degradation, the temperature that has a significant effect on the half-life of ozone. The reason why ozone decomposes rapidly in water is that its half-life is much shorter than in air (Hassaan *et al.* 2017a, b, El Nemr *et al.* 2020). Depending on the increase in temperature, the solubility of ozone decreases and becomes less stable. But the reaction speed increases by a factor of 2 to 3 per 10 ^{o}C. In principle, ozone dissolved in water cannot be used at temperatures above 40 ^{o}C, since the half-life of ozone is very short at this temperature (Hassaan *et al.* 2017a, b). In addition, the half-life of ozone decreases from 20 minutes at 20°C to 15 minutes at 25°C. This may explain why the O_3 time is shorter than O_3 + UV, and also the rate of decomposition of O_3 into hydroxyl radicals decreases due to the temperature increase from 22 to 27 C during treatment using O_3 + UV, as in MV40 and DB 86 degradation of dyes. Another significant reason is also the pH of the experiment, pH 7, which is not high enough to stimulate O_3 decomposition or generate hydroxyl radicals, compared to that of the previously treated dyes, where the pH was 11 , as in AR 17 dye and O_3 alone was lower than the UV-assisted O_3. It is also known that ozone stability is affected by the existence of salts, temperature and pH as reported by Mallevialle (1982).

Conclusion

The intention of this research was to be carried out with the nature of wastewater for dyeing factories and to simulate a mixture of dyes as close as possible to reality. Also, the other purpose of this study was to present a pattern to measure the disappearance of the color of the different dyes within a mixture of a group of dyes whose mixing leads to the formation of an unknown color. In order to simulate the closest class of liquid waste from textile dyeing factories five dyes of different types and colors were mixed (Acid Red 17, Acid Yellow 11, Direct Yellow 12, Direct Blue 86 and Mordant Violet 40 dyes). According to the results derived from the experimental study, it can be conveniently said that the ozone treatment was a very suitable method for removing the dyes from the reactive dyeing wastewater. When the experimental parameters were reviewed, ozone time was one of the highly efficient factors examined in the study. Discoloration efficiencies and rates of 20 to 180 minutes can be requested as sufficient time to eliminate all pigment types in the mixture consisting of five pigments of different colors and types. The study showed that using ozone alone gave better results than using ozone, along with ultraviolet rays. However, in general, 20 minutes was sufficient for the decolorization process for most colors and especially for the darker colors, depending on the type of dye and the percentage of dye flow. Critically, the decolorization efficiency differed depending on the chromophore group, the anchor group of the dyes, and the amount of dye (%) in the flux. As it is known, the reaction between ozone and the dyes took place in the chromophoric group of the dyes. This condition made the dye color and effluent content more important concerning the reactive dye. As the amount of dye in the effluent

increases, color removal becomes more difficult. In summary, the use of ozone has been a very effective means of preventing pollution from dyed effluents from textile mills, especially dye houses.

Acknowledgements

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