Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2020.26373

206 (2020) 108–115
December

Comparison of advanced oxidation methods of Fenton, UV/Fenton, and O_3/Fenton in treatment of municipal wastewater

Tarokh Khodadadi[a], Eisa Solgi[a,*], Samar Mortazavi[a], Hashmatollah Nourmoradi[b]

[a]*Department of Environment, Faculty of Natural Resources and Environment, Malayer University, Malayer, Iran, Tel. +98933339841; email: e.solgi@malayeru.ac.ir (E. Solgi), Tel. +98933339841; email: ktarokh@yahoo.com (T. Khodadadi), Tel. +98933339841; email: mortazavi.s@gmail.com (S. Mortazavi)*

[b]*Department of Environmental Health Engineering, School of Public Health, Ilam University of Medical Sciences, Ilam, Iran, Tel. +988432235733; email: h.nourmoradi2004@gmail.com (H. Nourmoradi)*

Received 13 December 2019; Accepted 23 July 2020

ABSTRACT

The present study aimed to evaluate the efficiency of advanced oxidation methods: Fenton, UV/Fenton, and O_3/Fenton in the treatment of municipal wastewater. The effect of H_2O_2:Fe^{2+} ratio, pH, and reaction time on chemical and biological parameters: chemical oxygen demand (COD), biological oxygen demand (BOD_5), total coliform (TC), and Fecal coliform (FC) were evaluated in different advanced oxidation processes (AOPs) methods (Fenton, UV/Fenton, and O_3/Fenton). All parameters were measured according to the standards methods. The results obtained from the present study showed that the highest removal of pollutants was achieved in H_2O_2:Fe^{2+} mole ratio of 0.7:1. Furthermore, the removal of COD, BOD_5 were increased when pH increased from 4 to 7. Moreover, reaction time influenced COD, BOD_5, TC, and FC until equilibrium time, however, its influence was not significant after equilibrium value. A comparative study on different AOP systems showed the order of highest to lowest removal efficiency of all parameters is as follows: $O_3/H_2O_2/Fe^{2+}$ > $UV/H_2O_2/Fe^{2+}$ > H_2O_2/Fe^{+2}. It can be concluded that Fenton and its modified methods have the promising potential for advanced municipal wastewater treatment.

Keywords: Fenton; UV/Fenton; O_3/Fenton; Advanced treatment; Wastewater

1. Introduction

The World Health Organization (WHO) estimates that half of people worldwide have limited access to water resources. Water restriction has forced authorities and scientists to reuse effluent as the main surrogate source of irrigation [1]. The proximity to agricultural fields is posed as one of the important benefits of effluent reuses [2]. Iran is an arid and semi-arid country; approximately 46 million (59%) of its population live in areas with less than the per capita water consumption (200 L/d per capita) [3,4]. Biological processes as conventional treatment methods are not efficient in the removal of recalcitrant and emerging pollutants, therefore, they are not able to meet the standards required for reusing wastewater [5,6]. Over recent years, significant attention has been devoted to effective non-biodegradable treatment methods for the elimination of biological pathogens found in wastewater [7]. The advanced oxidation processes (AOPs) are the method based on the generation of non-selective and strongly oxidizing radicals [8]. Hydroxyl radical ($^{\bullet}OH$) is well-known to strongly mineralize most organic waste [9]. Many different AOP methods have been developed for the generation of reactive radicals such as oxidation agents (O_3 and H_2O_2), radiation (ultraviolet or ultrasonic), and catalysts (such as Fe^{2+}). The Fenton and its modification have been widely used due to highly efficient, easy

* Corresponding author.

to purify, high react with organic matters, and producing less toxic compounds during the oxidation process [10–12]. The Fenton process is based on the following mechanism (Eqs. (1)–(5)) (Fig. 1) [13]:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^- \quad (1)$$

$$^{\bullet}OH + H_2O_2 \rightarrow HO_2^{\bullet} + H_2O \quad (2)$$

$$Fe^{2+} + {}^{\bullet}OH \rightarrow Fe^{3+} + OH^- \quad (3)$$

$$Fe^{3+} + HO_2^{\bullet} \rightarrow Fe^{2+} + O_2 + H^+ \quad (4)$$

$$^{\bullet}OH + {}^{\bullet}OH \rightarrow H_2O_2 \quad (5)$$

Previous studies revealed that Fenton and pseudo-Fenton processes in alone state and in combination with other oxidants are efficient for pretreatment of wastewaters and landfill leachate [14–17]. As wastewater contains recalcitrant pollutants and some pollutants cannot be degraded by the Fenton process alone, therefore combined Fenton and ultraviolet can improve degradation efficiency; increased hydroxyl radicals are generated. In addition, regeneration of ferrous ions leads to more radical generation (Eqs. (6)–(9)) (Fig. 1) [18].

$$Fe^{3+} + H_2O \rightarrow FeOH^{2+} + H^+ \quad (6)$$

$$FeOH^{2+} + h\nu \rightarrow Fe^{2+} + {}^{\bullet}OH \quad (7)$$

$$H_2O_2 + UV \rightarrow 2{}^{\bullet}OH \quad (8)$$

$$Fe^{3+} + H_2O_2 + UV \rightarrow Fe^{2+} + {}^{\bullet}OH + H^+ \quad (9)$$

Ozone (O_3) is classified as a promising advanced oxidation process and grabbed much attention by researchers due to its inherent strong oxidation. The incorporation of O_3 alone has some drawbacks: (a) less selectivity to degradation the contaminants, (b) low reactivity with certain organic matters such as inactivated aromatics or saturated carboxylic acids, and (c) in many cases, it does not completely mineralize the organic compounds [19,20]. Therefore, it is suggested to employ with other AOPs such as Fenton. The incorporation of combined O_3/Fenton can lower iron consumption in the Fenton process and less O_3 dosage. On the other hand, H_2O_2, iron, and hydroxyl ion can facilitate hydroxyl radical generation through the decomposition of ozone (Eqs. (10)–(12)) (Fig. 1) [21–23].

$$Fe^{3+} + O_3 + H_2O \rightarrow FeO^{2+} + {}^{\bullet}OH + O_2 + H^+ \quad (10)$$

$$O_3 + OH^- \rightarrow {}^{\bullet}O_3^- + {}^{\bullet}OH \quad (11)$$

$$O_3 + H_2O_2 \rightarrow O_2 + {}^{\bullet}OH + HO_2^{\bullet} \quad (12)$$

Since municipal effluents contain a wide range of pollutants including organic and inorganic matters, a comparison study on investigation of the removal efficiency of different AOP methods can help to choose the most effective treatment method [24,25]. Therefore, in the present work, the performance of Fenton (Fe^{+2}/H_2O_2), photo Fenton ($UV/Fe^{+2}/H_2O_2$), and ozone/Fenton ($O_3/Fe^{+2}/H_2O_2$) methods was compared in order to treat municipal wastewater after the biological process of the aerated lagoon in laboratory scale.

2. Materials and methods

2.1. Features of the effluent studied

Ilam city with 194,030 people is located at a latitude of 33.6350°N and longitude of 46.4153°E in western Iran. The wastewater per capita in this city was estimated to be 148.8l capi^{-1} d^{-1}. This large volume of domestic wastewater is treated by two parallel series of aeration lagoons after collecting and passing through a coarse and fine screen. The treated wastewater is discharged to the river after settling and chlorination process [26]. The samples were withdrawn from

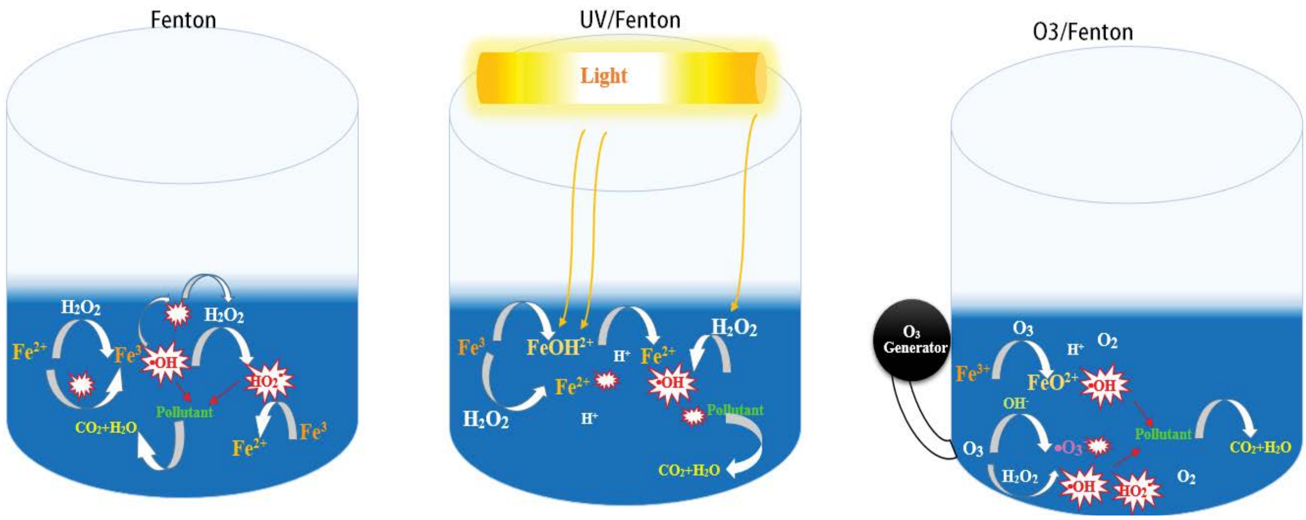

Fig. 1. Fenton, UV/Fenton, and O_3/Fenton mechanism.

the effluent every three days before entering into the river and immediately transferred to the laboratory at 4°C. The chemical and microbial characteristics of the effluent were measured according to the standard methods for the examination of water and wastewater [27,28]. The effluent characteristics of the treatment plant are summarized in Table 1.

2.2. Procedures

2.2.1. Fenton (H_2O_2/Fe^{2+}), photo/Fenton ($UV/H_2O_2/Fe^{2+}$), and ozone/Fenton ($O_3/H_2O_2/Fe^{2+}$) oxidation processes

The Fenton process was conducted in different molar ratios of H_2O_2/Fe^{2+} (0.35:1, 0.7:1, 1.06:1, 1.42:1, 1.78:1, and 2:1) using 0.003, 0.006, 0.009, 0.011, 0.015, and 0.017 mol/L H_2O_2 and 0.009 mol/L of $FeSO_4{\cdot}7H_2O$. The pH was adjusted using 0.1 M hydrochloric acid (HCl) and sodium hydroxide (NaOH) after adding H_2O_2 [29]. The optimal molar ratio of the Fenton process was determined at the initial pH of effluent (7.65) and contact time of 180 min in a beaker with an effective volume of 0.5 L (height: 0.220 mm and internal diameter: 120 mm). A magnetic stirrer (IKA® RH Basic2, Germany) at medium speed was used to create a uniform environment. The effect of contact time (30, 60, 90, 120, 150, and 180 min) and pH (7) were evaluated at optimal H_2O_2/Fe^{2+} molar ratio. The residual H_2O_2 was neutralized using manganese dioxide in order to prevent the interferences in BOD_5, DO, and COD values (Eq. (13)) [30].

$$MnO_2 + H_2O_2 \rightarrow MnO_2 + H_2O + ½O_2 \tag{13}$$

The combined photo/Fenton process was performed in a similar procedure to Fenton process with except of the presence of four low-pressure UV lights (Philips, 6 W, UV-C, 254 nm) placed vertically in the inner and outer part of the beaker and inside the transparent quartz glass. The beaker was covered with aluminum foil in order to prevent the harmful effects of UV. Similar to the Fenton process, Ozone/Fenton was conducted using two Impinger containers (Schott Duran, Germany) with an effective volume of 0.5 L in a series mode [31]. Ozone generator (model MOG-5G/H, Arda, France) and dry air source fed was used to transfer ozone to the reactor with a flow rate of 1.6 g/h. The amount of ozone was measured using standard method No. 2350E [27].

Table 1
Characteristics of the effluent of the treatment plant

Parameter	Range value
EC (μS cm^{-1})	240–265
TDS (mg L^{-1})	156–172
pH	6.62–7.85
COD (mg L^{-1})	112–336
BOD_5 (mg L^{-1})	70–197
TSS (mg L^{-1})	60–327
Total coliform (MPN/100 mL)	$1.10E^{+06}$–$6.30E^{+06}$
Fecal coliform (MPN/100 mL)	$1.12E^{+04}$–$3.60E^{+06}$
Turbidity (NTU)	59–150
Color (TCU)	40–200
DO (mg L^{-1})	2.7–5.7

2.2.2. Measurement methods and samples analysis

All BOD_5 (No. 5210A), COD (No. 5220C (Closed Reflux)), FC (No. 9221A, B, C (Multiple Tube)), TC (No. 2130), and pH (No. 4500-H^+) parameters were measured as per the standard methods for the examination of water and wastewater [27]. All experiments were performed under the same condition in triplicate with relative standard deviation (RSD) less than 4.5%. The removal efficiency of BOD_5, COD, FC, and TC parameters were calculated with the following equation:

$$\text{Removal}\left(\%\right) = \left[\frac{\left(C_i - C_f\right)}{C_i}\right] \times 100 \tag{14}$$

where C_i and C_f denote the initial and final concentrations of BOD_5, COD, FC, and TC, respectively.

3. Results and discussion

3.1. Effect of molar ratio (H_2O_2:Fe^{2+})

The molar ratio of Fenton reagent (H_2O_2:Fe^{2+}) is one of the most important parameters influencing the Fenton, UV/Fenton, and O_3/Fenton processes [32]. Fig. 2a shows the effect of H_2O_2/Fe^{2+} molar ratio in the removal of COD, BOD_5, total coliform (TC), and fecal coliform (FC) in different processes. Results indicated that increasing the ratio of H_2O_2:Fe^{2+} from 0.7:1 to 1.8:1, decreased the removal efficiency of COD and BOD by 32.15% and 34.56%, respectively. While TC and FC removal were constant in all H_2O_2:Fe^{2+} ratio. Results indicated that maximum removal efficiency for all parameters was observed at 0.7:1 ratio (H_2O_2:Fe^{2+}). The decrease of COD and BOD removal at the high H_2O_2:Fe^{2+} ratio is due to the scavenging effect of hydroxyl radicals by excess hydrogen peroxide (Eqs. (15) and (16)):

$$H_2O_2 + {}^{\bullet}OH \rightarrow HO_2^{\bullet} + H_2O \tag{15}$$

$$HO_2^{\bullet} + {}^{\bullet}OH \rightarrow H_2O + O_2 \tag{16}$$

Besides, according to the Eq. (16), the recombination of hydroxyl radicals could occur at a high concentration of hydrogen peroxide which leads to reduced removal efficiency of organic matters [33]. This phenomenon was confirmed by K_{obs} of BOD and COD removal (Fig. 2b).

On the other hand, at the high H_2O_2:Fe^{2+} molar ratio, there are no adequate iron ions to decompose H_2O_2 and generate ${}^{\bullet}OH$ radicals [34].

Wu et al. [35] reported that the removal efficiency of humic acid is landfill leachate was decreased when hydrogen peroxide concentration was increased at a constant concentration of ferrous due to quenching the hydroxyl radicals by H_2O_2. The study of Murray and Parsons [36] revealed that the removal of natural organic matter from drinking water

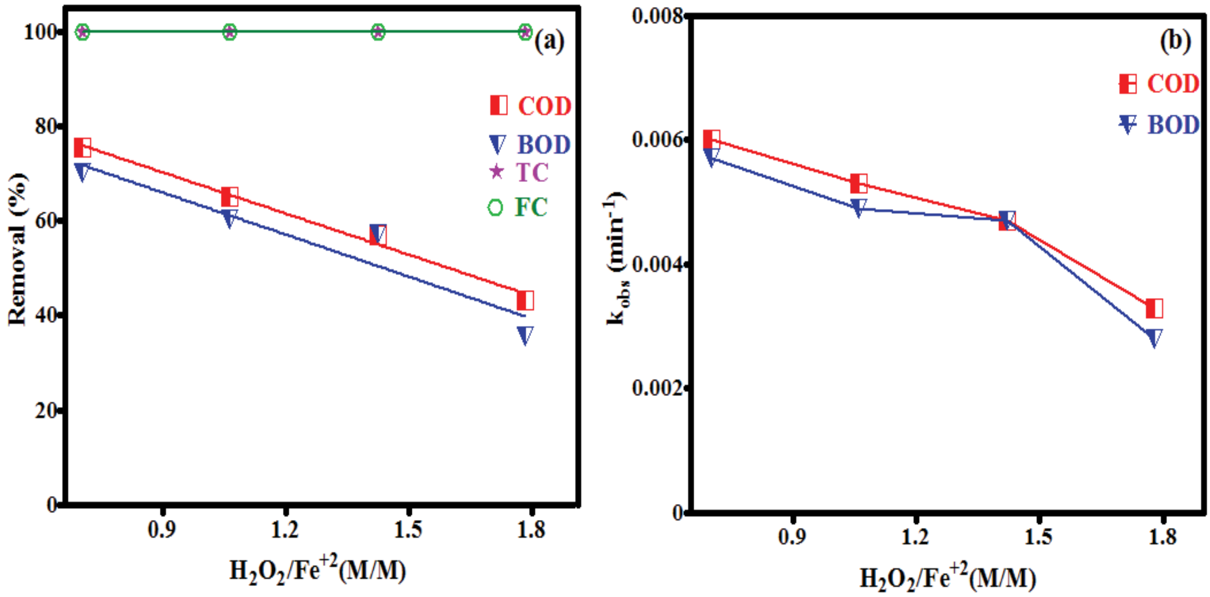

Fig. 2. (a) Effect of molar ratio (H_2O_2/Fe^{2+}) on COD, BOD, TC, and FC removal, (b) K_{obs} of BOD and COD.

was decreased at H_2O_2 concentrations higher than 20:1 ratio. Chen et al. [37] demonstrated that excess hydrogen peroxide prevents the decomposition of organic materials.

3.2. Effect of pH

pH is the main influencing parameter in the chemical reactions. Fig. 3 shows the removal efficiency of COD, BOD, TC, and FC in Fenton, UV/Fenton, and O_3/Fenton processes at pH 4–9. Results showed that the removal efficiency of all parameters excepted (TC and FC) was raised for all processes when pH was increased from 4 to 7 (Fig. 3 and Fig. S1). According to Fig. 3, the highest removal efficiency of COD (92.37%, 95.44%, and 98.37%) and BOD (94.12%, 96.64%, and 99.12%) were observed in Fenton, UV/Fenton, and O_3/Fenton oxidation processes, respectively.

The high efficiency of TC and FC at acidic condition is contributed to the effect of acidic conditions on the microorganism. Because TC and FC bacteria will survive only in a limited pH near the neutral.

However, increasing pH > 8 had no significant effect on the removal of the parameters (COD, BOD, TC, and FC). Therefore, pH = 7 seems to be well-suited for the removal and oxidation. Many studies have proven that the most oxidation occurred at pH 3–5, while, conversely this study showed that maximum oxidation was obtained at pH = 7 [38]. Yang et al. [39] confirmed the hydroxylated form of $Fe(OH)^+$ could be formed in the presence of organic

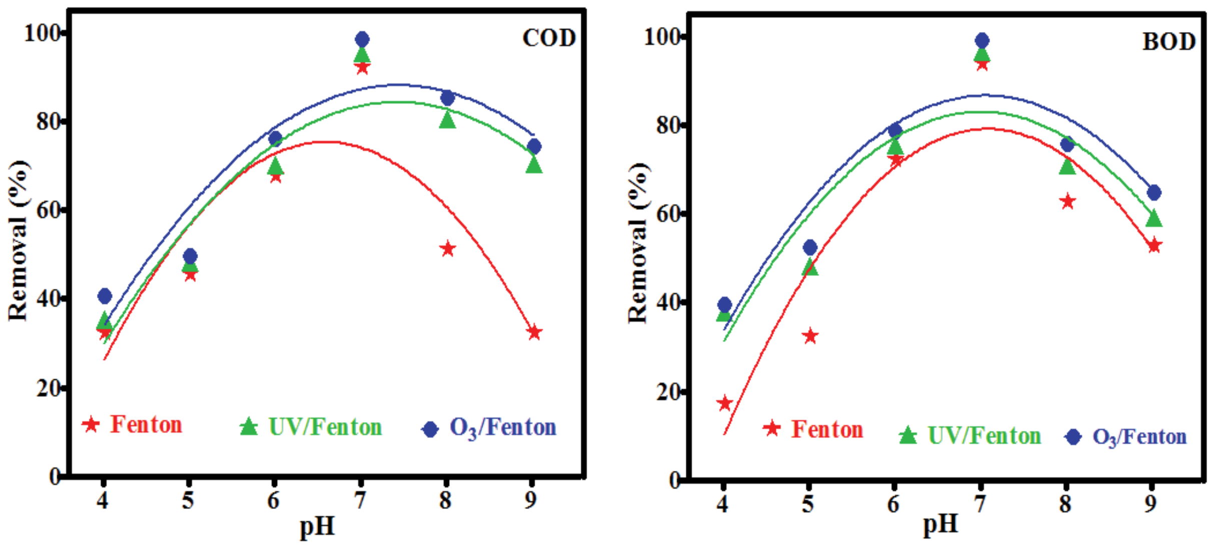

Fig. 3. Effect of pH on COD and BOD removal in different processes (Fenton, UV/Fenton, and O_3/Fenton).

compounds which has much faster than other species reaction with H_2O_2 at pH = 7.

Moreover, Chen et al. [37] reported that the effect of pH on the oxidation process is restricted to the first few minutes of reaction. Therefore, the COD and color removal was increased by increasing the pH from 3 to 7 (Eqs. (4) and (17)). The reaction of Eqs. (4) and (17) are important due to the regeneration of ferrous ions at natural pH and hydroxyl radical generation by ferric ion and hydrogen peroxide at pH higher than 5 (Fig. 1).

$$H_2O_2 + Fe(OH)^{+} \rightarrow FeOH_2^{+} + {}^{\bullet}OH \quad (17)$$

On the other hand, decreased removal efficiency in the Fenton process at low pH range is due to the creation of foam in the acidic aqueous which accordingly prevents UV light penetration to the wastewater [40]. Also, there are some limitations (e.g., sulfides or cyanides gases emission, increase of sludge, salinity, and coasts) to utilize Fenton and its modified process in acidic pH [41].

3.3. Effect of reaction time on H_2O_2/Fe^{2+}, $UV/H_2O_2/Fe^{2+}$, and $O_3/H_2O_2/Fe^{2+}$

Fig. 4 and Fig. S2 illustrated the effect of reaction time on COD, BOD, TC, and FC removal in Fenton, UV/Fenton, and O_3/Fenton processes. Reaction time is one of the most important influencing factors in Fenton process which has been reported to be between 30 and 180 min [42–44] and also the optimum reaction time was reported to be between 60 and 120 min in $UV/H_2O_2/Fe^{2+}$ and $O_3/H_2O_2/Fe^{2+}$ processes [17,45]. Reaction time can be very helpful in saving energy and chemicals usage in the oxidation processes and the entrance of materials into aquatic environments and ecosystems [46,47]. The results showed that COD, BOD_5, TC, and FC removal were increased as the time preceded. Equilibrium time for removal of COD and BOD_5 in H_2O_2/Fe^{2} process was found to be 60 min, however, this value in $UV/H_2O_2/Fe^{2+}$ and $O_3/H_2O_2/Fe^{2+}$ processes were obtained 90–120 min. The removal efficiency of COD, BOD_5, TC, and FC rates were not significant after equilibrium time due to the resistance of refractory materials against the ${}^{\bullet}OH$ radical, which is not degraded after this time and the yield remains stable [48]. On the other hand, the amount of H_2O_2, Fe^{2+}, and O_3 will be decreased with increasing the reaction time. Therefore, there is no oxidant agent for radical production. Ferreira et al. [49] observed the removal > 90% of the dye in the olive mill wastewater using the UV/Fenton method at H_2O_2/Fe^{2+} molar ratio of 0.2 and pH = 4.2 within 90 min. Amr and Aziz [50] also reported that removal efficiency over 98% was achieved for removal of dye from leachate using the $O_3/H_2O_2/Fe^{2+}$ method at 90 min, pH = 7, and molar ratio of $H_2O_2/Fe^{2+} = 1$ (0.05 mol/L H_2O_2 and 0.05 mol/L Fe^{+2}).

3.4. Comparison of different processes

Results clearly illustrated the differences between the removal efficiency of three methods. The highest removal efficiency for all parameters was obtained as follows: $O_3/H_2O_2/Fe^{2+} > UV/H_2O_2/Fe^{2+} > H_2O_2/Fe^{+2}$. As shown, the efficiency of the O_3/Fenton process is more than other processes. The explanation for higher corresponding removal efficiency to O_3/Fenton process is a combination of two oxidants that have a synergic effect on each other. Although previous studies reported that the O_3 alone has low efficiency in removing COD, ozone combined with Fenton leads to more generation of reactive radicals that influence the organic matter degradation. Hagman et al. [51] reported only 22% of COD through O_3 process alone. While the conjunction of Fenton oxidation and ozonation are famed as effective oxidation processes in wastewater treatment.

H_2O_2 in water creates the hydroperoxide ion (HO_2^-) which quickly reacts with O_3 to initiate a radical chain mechanism (Eqs. (18) and (19)) [52,53].

$$H_2O + O_3 \rightarrow {}^{\bullet}OH + 3O_2 \quad (18)$$

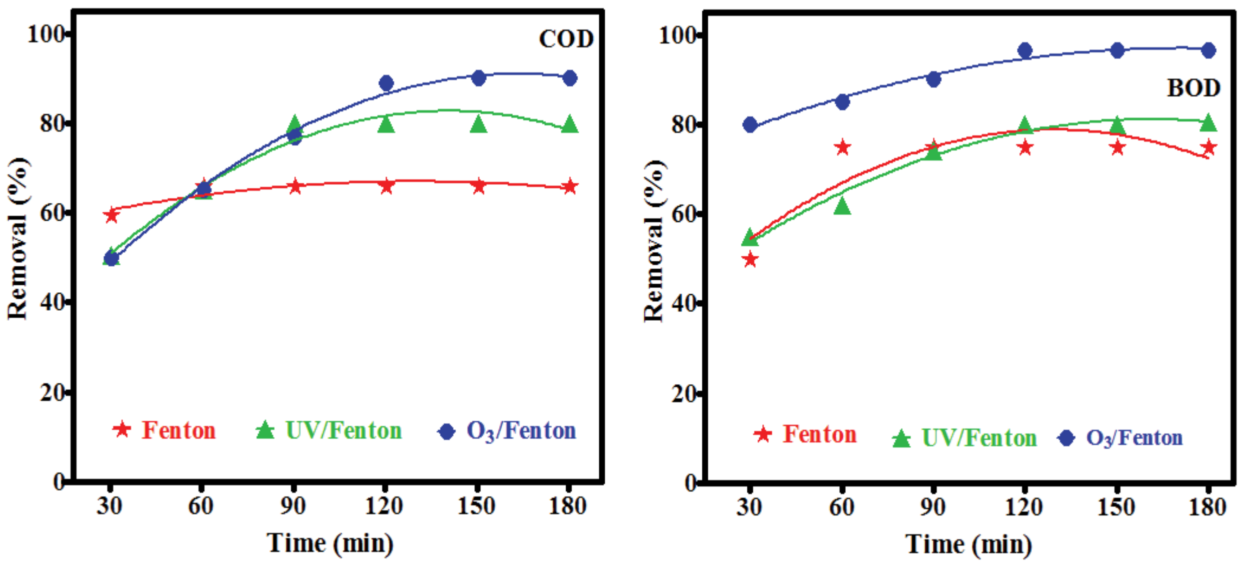

Fig. 4. Effect of reaction time on COD and BOD removal in different processes (Fenton, UV/Fenton, and O_3/Fenton).

$$HO_2^- + O_3\ HO^\bullet{}_2 \rightarrow 2O_3 + {}^\bullet OH + H^+ \quad (19)$$

Moreover, as illustrated in Eq. (10), the ferric ions which are predominant in natural pH react with O_3 and generate hydroxyl radicals. Besides, the oxidation ability of O_3 is effective at high pH values [54].

Results indicated that the UV/Fenton process is more effective than Fenton process. The use of H_2O_2/Fe^{2+} in conjunction with the UV light leads to more degradation of organic matters [55]. The major benefits of UV are associated with the regeneration of ferrous ions and direct decomposition of H_2O_2 through the photolysis process (Eqs. (7) and (8)). Therefore, these mechanisms causes an increase in the amount of $^\bullet$OHs in UV/Fenton process [56]. Many studies [17,50,57] revealed that the use of O_3 and UV combined with the H_2O_2/Fe^{2+} method are more effective in the removal of pollutants from wastewater and leachate compared with the use of the H_2O_2/Fe^{2+} method alone.

4. Conclusions

Fenton, photo/Fenton, and ozone/Fenton are the AOP methods that are based on $^\bullet$OH generation facilities the mineralization of most organic compounds. Maximum removal efficiency for all parameters were obtained at 0.7:1 ratio of H_2O_2:Fe^{2+}, while the removal efficiency of COD and BOD were decreased with increasing the ratio of H_2O_2:Fe^{2+} (TC and FC removal were constant). The removal efficiency of all parameters were raised with as pH increased from 4 to 7 (excepted TC and FC). Also, COD, BOD_5, TC, and FC removal increased with increasing reaction time. Equilibrium time for degradation of COD and BOD_5 in H_2O_2/Fe^2 process was found to be 60 min and in UV/H_2O_2/Fe^{2+} and $O_3/H_2O_2/Fe^{2+}$ was 90 to 120 min. A comparison of different processes showed that the highest removal efficiency of all parameters are as follows: $O_3/H_2O_2/Fe^{2+}$ > UV/H_2O_2/Fe^{2+} > H_2O_2/Fe^{+2}. The highest removal efficiency of COD (92.37%, 95.44%, and 98.37%) and BOD (94.12%, 96.64%, and 99.12%) were obtained for Fenton, UV/Fenton, and O_3/Fenton processes respectively. Since the effluent of biological wastewater treatment methods cannot mainly be incorporated directly for agricultural purposes, therefore, advanced municipal wastewater treatment using Fenton, UV/Fenton, and O_3/Fenton methods can meet the required standards for limited and unlimited application in different parts of the urban environment, the irrigation of plants with food and non-food consumption, the agricultural irrigation, fish farming, and injecting into the ground or in water bodies. Moreover, these efficient methods can be considered as a promising approach to alleviate the water shortage crisis in Iran and most areas of the world.

Acknowledgments

This article is extracted from a doctoral thesis in the field of Environment-Environmental Pollution at the Malayer University. The authors thank the Malayer University for their financial and ethical support and the Medical Sciences University of Ilam for their assistance in conducting the tests.

References

[1] T. Khodadi, Investigation of Performance on the Oxidation Processess in Urban Wastewater Supplementary Tretment, Ph.D Thesis, Department of Environmental Sciences, Faculty of Natural Resources and Environment, Malayer University, Malayer, Iran, 2019.

[2] S. Ricart, A.M. Rico, Assessing technical and social driving factors of water reuse in agriculture: a review on risks, regulation and the yuck factor, Agric. Water Manage., 217 (2019) 426–439.

[3] M. Minaei, H. Shafizadeh-Moghadam, A. Tayyebi, Spatio-temporal nexus between the pattern of land degradation and land cover dynamics in Iran, Land Degrad. Dev., 29 (2018) 2854–2863.

[4] M. Nouri, M. Homaee, M. Bannayan, Spatiotemporal reference evapotranspiration changes in humid and semi-arid regions of Iran: past trends and future projections, Theor. Appl. Climatol., 133 (2018) 361–375.

[5] M. Karimaei, K. Sharafi, M. Moradi, H.R. Ghaffari, H. Biglari, H. Arfaeinia, N.Fattahi, Optimization of a methodology for simultaneous determination of twelve chlorophenols in environmental water samples using *in situ* derivatization and continuous sample drop flow microextraction combined with gas chromatography-electron-capture detection, Anal. Methods, 9 (2017) 2865–2872.

[6] K. Sharafi, M. Fazlzadehdavil, M. Pirsaheb, J. Derayat, S. Hazrati, The comparison of parasite eggs and protozoan cysts of urban raw wastewater and efficiency of various wastewater treatment systems to remove them, Ecol. Eng., 44 (2012) 244–248.

[7] O. Sacco, V. Vaiano, L. Rizzo, D. Sannino, Photocatalytic activity of a visible light active structured photocatalyst developed for municipal wastewater treatment, J. Cleaner Prod., 175 (2018) 38–49.

[8] W.D.N. Júnior, R. Aquino, A. Barbosa, O. Rocha, Development of a new PET flow reactor applied to food dyes removal with advanced oxidative processes, J. Water Process. Eng., 31 (2019) 100823, doi: 10.1016/j.jwpe.2019.100823.

[9] Z. Zhang, Y.-H. Chuang, N. Huang, W.A. Mitch, Predicting the contribution of chloramines to contaminant decay during ultraviolet/hydrogen peroxide advanced oxidation process treatment for potable reuse, Environ. Sci. Technol., 53 (2019) 4416–4425.

[10] S. Hussain, J.R. Steter, S. Gul, A.J. Motheo, Photo-assisted electrochemical degradation of sulfamethoxazole using a Ti/$Ru_{0.3}Ti_{0.7}O_2$ anode: mechanistic and kinetic features of the process, J. Environ. Manage., 201 (2017) 153–162.

[11] R.R. Kalantary, A. Azari, A. Esrafili, K. Yaghmaeian, M. Moradi, K. Sharafi, The survey of Malathion removal using magnetic graphene oxide nanocomposite as a novel adsorbent: thermodynamics, isotherms, and kinetic study, Desal. Water Treat., 57 (2016) 28460–28473.

[12] C. Ji, C. Wang, J. Sun, Y. Yu, Degradation of 4-methylbenzylidene camphor (4-MBC) using Fenton, UV light irradiation and photo-Fenton, MATEC Web Conf., 100 (2017) 03006, doi: 10.1051/matecconf/201710003006.

[13] M.-h. Zhang, H. Dong, L. Zhao, D.-x. Wang, D. Meng, A review on Fenton process for organic wastewater treatment based on optimization perspective, Sci. Total Environ., 670 (2019) 110–121.

[14] V. Torretta, N. Ferronato, I.A. Katsoyiannis, A.K. Tolkou, M. Airoldi, Novel and conventional technologies for landfill leachates treatment: a review, Sustainability, 9 (2017) 1–39, doi: /10.3390/su9010009.

[15] G. Boczkaj, A. Fernandes, Wastewater treatment by means of advanced oxidation processes at basic pH conditions: a review, Chem. Eng. J., 320 (2017) 608–633.

[16] A. Klančar, J. Trontelj, A. Kristl, A. Meglič, T. Rozina, M.Z. Justin, R. Roškar, An advanced oxidation process for wastewater treatment to reduce the ecological burden from pharmacotherapy and the agricultural use of pesticides, Ecol. Eng., 97 (2016) 186–195.

[17] X. Hu, X. Wang, Y. Ban, B. Ren, A comparative study of UV-Fenton, UV-H_2O_2 and Fenton reaction treatment of landfill leachate, Environ. Technol., 32 (2011) 945–951.
[18] W.-l. Li, G.-h. Peng, Experimental study on degradation of reactive red X-3B dye wastewater by heterogeneous UV/Fenton catalyst system, Liaoning Chem. Ind., 5 (2018) 8.
[19] E. Kusvuran, D. Yildirim, Degradation of bisphenol A by ozonation and determination of degradation intermediates by gas chromatography–mass spectrometry and liquid chromatography–mass spectrometry, Chem. Eng. J., 220 (2013) 6–14.
[20] F.J. Beltrán, A. Aguinaco, A. Rey, J.F. García-Araya, Kinetic studies on black light photocatalytic ozonation of diclofenac and sulfamethoxazole in water, Ind. Eng. Chem. Res., 51 (2012) 4533–4544.
[21] C.-H. Wu, Decolorization of CI reactive red 2 in O_3, Fenton-like and O_3/Fenton-like hybrid systems, Dyes Pigm., 77 (2008) 24–30.
[22] N. Kishimoto, S. Ueno, Catalytic effect of several iron species on ozonation, J. Water Environ. Technol., 10 (2012) 205–215.
[23] H. Arfaeinia, H. Rezaei, K. Sharafi, M. Moradi, H. Pasalari, S.E. Hashemi, Application of ozone/magnetic graphene oxide for degradation of diazinon pesticide from aqueous solutions, Desal. Water Treat., 107 (2018) 127–135.
[24] N. Mirzaei, H.R. Ghaffari, K. Karimyan, F.M. Moghadam, A. Javid, K. Sharafi, Survey of effective parameters (water sources, seasonal variation and residual chlorine) on presence of thermotolerant coliforms bacteria in different drinking water resources, Int. J. Pharm. Technol., 7 (2015) 9680–9689.
[25] N. Mirzaei, H.R. Ghaffari, K. Sharafi, A. Velayati, G. Hoseindoost, S. Rezaei, A.H. Mahvi, A. Azari, K. Dindarloo, Modified natural zeolite using ammonium quaternary based material for Acid red 18 removal from aqueous solution, J. Environ. Chem. Eng., 5 (2017) 3151–3160.
[26] H. Nourmoradi, H. Karimi, A. Alihosseini, A. Baghi, K. Farokhimoghadam, Survey on the performance of Ilam wastewater treatment plant in the removal of coliform, fecal coliforms and other parameters effected on water quality, J. Ilam Univ. Med Sci., 22 (2014) 77–83.
[27] WEF, APHA, Standard Methods for the Examination of Water and Wastewater, Water Environment Federation, American Public Health Association, Washington, DC, USA, 2005.
[28] K. Sharafi, M. Moradi, A. Karami, T. Khosravi, Comparison of the efficiency of extended aeration activated sludge system and stabilization ponds in real scale in the removal of protozoan cysts and parasite ova from domestic wastewater using Bailenger method: a case study, Kermanshah, Iran, Desal. Water Treat., 55 (2015) 1135–1141.
[29] A. Fischbacher, C. von Sonntag, T.C. Schmidt, Hydroxyl radical yields in the Fenton process under various pH, ligand concentrations and hydrogen peroxide/Fe(II) ratios, Chemosphere, 182 (2017) 738–744.
[30] E. Illés, A. Tegze, K. Kovács, G. Sági, Z. Papp, E. Takács, L. Wojnárovits, Hydrogen peroxide formation during radiolysis of aerated aqueous solutions of organic molecules, Radiat. Phys. Chem., 134 (2017) 8–13.
[31] R.R. Kalantary, M. Rahmatinia, M. Moradi, Data on modeling of UV/$Na_2S_2O_8$/FeS_2 process in amoxicillin removal using Box–Behnken methodology, Data Brief, 19 (2018) 1810–1815.
[32] M. Moradi, R.R. Kalantary, A. Esrafili, A.J. Jafari, M. Gholami, Visible light photocatalytic inactivation of *Escherichia coli* by natural pyrite assisted by oxalate at neutral pH, J. Mol. Liq., 248 (2017) 880–889.
[33] J.-S. Guo, A.A. Abbas, Y.-P. Chen, Z.-P. Liu, F. Fang, P. Chen, Treatment of landfill leachate using a combined stripping, Fenton, SBR, and coagulation process, J. Hazard. Mater., 178 (2010) 699–705.
[34] W. Jiang, D.D. Dionysiou, M. Kong, Z. Liu, Q. Sui, S. Lyu, Utilization of formic acid in nanoscale zero valent iron-catalyzed Fenton system for carbon tetrachloride degradation, Chem. Eng. J., 380 (2020) 122537, doi: 10.1016/j.cej.2019.122537.
[35] Y. Wu, S. Zhou, F. Qin, H. Peng, Y. Lai, Y. Lin, Removal of humic substances from landfill leachate by Fenton oxidation and coagulation, Process. Saf. Environ. Prot., 88 (2010) 276–284.
[36] C.A. Murray, S.A. Parsons, Removal of NOM from drinking water: Fenton's and photo-Fenton's processes, Chemosphere, 54 (2004) 1017–1023.
[37] Y. Chen, C. Liu, J. Nie, S. Wu, D. Wang, Removal of COD and decolorizing from landfill leachate by Fenton's reagent advanced oxidation, Clean Technol. Environ. Policy, 16 (2014) 189–193.
[38] M. Moradi, A. Esrafili, M. Pirsaheb, V.K. Gupta, H. Sharafi, H. Arfaeinia, R.R. Kalantary, The inactivation of fecal coliform using Fe_3O_4@Cu nanocomposite in real wastewater: emphasizing on synergic effect and inactivation mechanisms, Desal. Water Treat., 123 (2018) 41–51.
[39] S. Yang, H. He, D. Wu, D. Chen, X. Liang, Z. Qin, M. Fan, J. Zhu, P. Yuan, Decolorization of methylene blue by heterogeneous Fenton reaction using $Fe_{3-x}Ti_xO_4$ ($0 \leq x \leq 0.78$) at neutral pH values, Appl. Catal., B, 89 (2009) 527–535.
[40] N.H. Ince, Light-enhanced chemical oxidation for tertiary treatment of municipal landfill leachate, Water Environ. Res., 70 (1998) 1161–1169.
[41] A. Ruiz-Delgado, M. Roccamante, I. Oller, A. Agüera, S. Malato, Natural chelating agents from olive mill wastewater to enable photo-Fenton-like reactions at natural pH, Catal. Today, 328 (2019) 281–285.
[42] L. Tengrui, A. Al-Harbawi, Z. Jun, L.M. Bo, The effect and its influence factors of the Fenton process on the old landfill leachate, J. Appl. Sci., 7 (2007) 724–727.
[43] H. Zhang, H.J. Choi, C.-P. Huang, Optimization of Fenton process for the treatment of landfill leachate, J. Hazard. Mater., 125 (2005) 166–174.
[44] M. Ahmadian, S. Reshadat, N. Yousefi, S.H. Mirhossieni, M.R. Zare, S.R. Ghasemi, N. Rajabi Gilan, R. Khamutian, A. Fatehizadeh, Municipal leachate treatment by Fenton process: effect of some variable and kinetics, J. Environ. Public Health, 2013 (2013) 1–6.
[45] N. Azbar, T. Yonar, K. Kestioglu, Comparison of various advanced oxidation processes and chemical treatment methods for COD and color removal from a polyester and acetate fiber dyeing effluent, Chemosphere, 55 (2004) 35–43.
[46] M. Hadavifar, A.A. Zinatizadeh, H. Younesi, M. Galehdar, Fenton and photo-Fenton treatment of distillery effluent and optimization of treatment conditions with response surface methodology, Asia-Pac. J. Chem. Eng., 5 (2010) 454–464.
[47] F. Mahdad, H. Younesi, N. Bahramifar, M. Hadavifar, Optimization of Fenton and photo-Fenton-based advanced oxidation processes for post-treatment of composting leachate of municipal solid waste by an activated sludge process, KSCE J. Civ. Eng., 20 (2016) 2177–2188.
[48] A. Papadopoulos, D. Fatta, M. Loizidou, Development and optimization of dark Fenton oxidation for the treatment of textile wastewaters with high organic load, J. Hazard. Mater., 146 (2007) 558–563.
[49] F. Ferreira, L. Carvalho, R. Pereira, S. Antunes, S. Marques, F. Gonçalves, A. Duarte, T. Rocha-Santos, A. Freitas, Biological and photo-Fenton treatment of olive oil mill wastewater, Global Nest J., 10 (2008) 419–425.
[50] S.S.A. Amr, H.A. Aziz, New treatment of stabilized leachate by ozone/Fenton in the advanced oxidation process, Waste. Manage., 32 (2012) 1693–1698.
[51] M. Hagman, E. Heander, J.l.C. Jansen, Advanced oxidation of refractory organics in leachate-potential methods and evaluation of biodegradability of the remaining substrate, Environ. Technol., 29 (2008) 941–946.
[52] C. Tizaoui, L. Bouselmi, L. Mansouri, A. Ghrabi, Landfill leachate treatment with ozone and ozone/hydrogen peroxide systems, J. Hazard. Mater., 140 (2007) 316–324.
[53] M. Karami, K. Sharafi, A. Asadi, A. Bagheri, F. Yosefvand, S.S. Charganeh, N. Mirzaei, A. Velayati, Degradation of reactive red 198 (RR198) from aqueous solutions by advanced oxidation processes (AOPS): O_3, H_2O_2/O_3 and H_2O_2/ultrasonic, Bulg. Chem. Commun., 48 (2016) 43–49.
[54] R. Kıdak, Ş. Doğan, Medium-high frequency ultrasound and ozone based advanced oxidation for amoxicillin removal in water, Ultrason. Sonochem., 40 (2018) 131–139.

[55] E. Azizi, M. Fazlzadeh, M. Ghayebzadeh, L. Hemati, M. Beikmohammadi, H.R. Ghaffari, H.R. Zakeri, K. Sharafi, Application of advanced oxidation process (H_2O_2/UV) for removal of organic materials from pharmaceutical industry effluent, Environ. Prot. Eng., 43 (2017) 184–191.

[56] Y.-M. Kang, M.-K. Kim, K.-D. Zoh, Effect of nitrate, carbonate/bicarbonate, humic acid, and H_2O_2 on the kinetics and degradation mechanism of Bisphenol-A during UV photolysis, Chemosphere, 204 (2018) 148–155.

[57] P.R. Gogate, A.B. Pandit, A review of imperative technologies for wastewater treatment II: hybrid methods, ACS Environ. Res., 8 (2004) 553–597.

Supplementary information

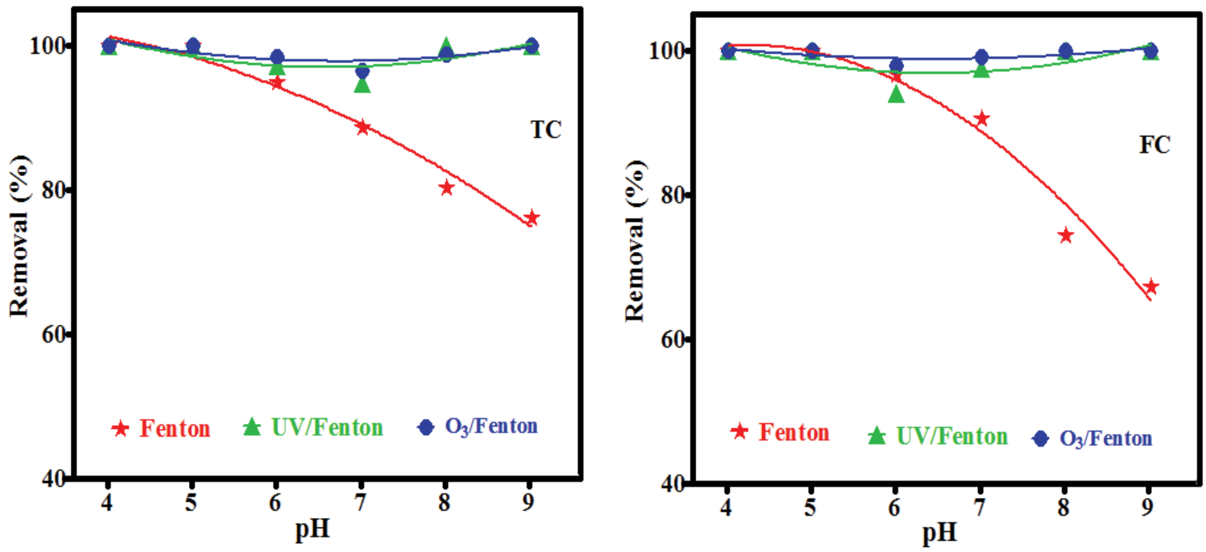

Fig. S1. Effect of pH on TC and FC removal in different processes (Fenton, UV/Fenton, and O_3/Fenton).

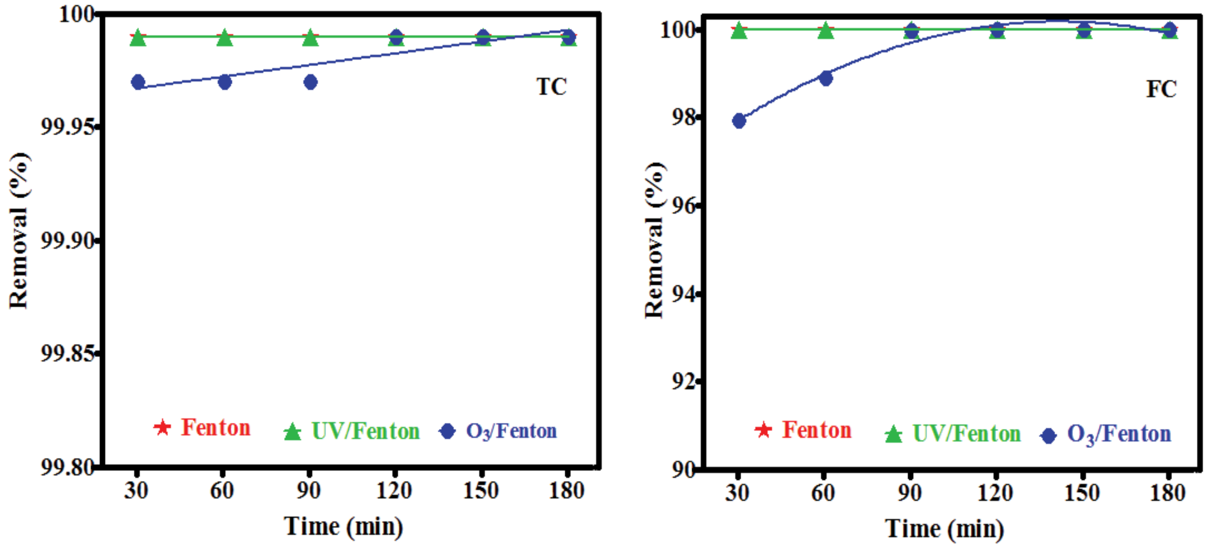

Fig. S2. Effect of reaction time on TC and FC removal in different processes (Fenton, UV/Fenton, and O_3/Fenton).