Indian Journal of Chemical Technology
Vol. 30, March 2023, pp. 211-216
DOI: 10.56042/ijct.v30i2.62849

Process parameters study of sanitary landfill leachate treatment using photo-Fenton-like systems

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Received 2May 2022; accepted 19 January 2023

In this study, the effects of the combination of classical Fenton (CFP) and modified Fenton (MFP) processes with UV light on the treatment of sanitary landfill leachate have been investigated. Iron (Fe^{2+} and Fe^0) dosage, hydrogen peroxide dosage, reaction time, pH and different UV lamps have been optimized to achieve high COD removal. In addition, zeroth, first and second order kinetic models are applied for all processes under optimum conditions. For CFP and MFP; optimum pH 3, reaction time 30 min, 4 g/kg TS Fe^{2+} and Fe^0 and 5 g/kg TS H_2O_2 have been determined. The COD removal efficiency is determined as 48.86% for CFP and 59.27% for MFP under optimum conditions. COD removal efficiencies increased in photo Fenton application under UV light. The efficiency is found to increase under UVA light source from 48.86% to 49.17% and from 59.27% to 70.72% in in CFP and MFP, respectively. In the kinetic study, the highest R^2 values are obtained in the CFP/UV process, while CFP and MFP are found to fit the 0th order kinetic model. In this study, it has been concluded that Fenton and photo Fenton applications are effective in reducing the COD values of landfill leachate.

Keywords: Advanced oxidation process, Fenton, Landfill leachate, Photo-Fenton

Today, landfill is a common way of municipal solid waste (MSW) disposal. However, wastewater from landfills, such as leachate, is very polluted and difficult to deal[1]. Landfill leachate is a mixture of different organic and inorganic compounds such as heavy metals, nitrogen compounds, etc[2]. Their amount and quality depend on factors such as the age of the landfill, the type of waste, climatic conditions and hydrogeological conditions[3].

Advanced oxidation processes (AOPs) such as Fenton, electro-Fenton, and photo-Fenton were used to improve landfill leachate quality in terms of COD and color removal[4-7]. AOPs have been of interest as suitable methods for reducing organic compound loads and increasing the biodegradability of persistent organic compound pollutants in leachate[8].

Traditional methods of leachate treatment such as coagulation, flocculation and settling have disadvantages such as energy requirements and frequent use of chemical additives[9]. AOPs such as UV/Fe^{2+} H_2O_2, UV/TiO_2 have been proposed in recent years as an effective alternative for the mineralization of refractory organic matter in wastewater and landfill leachate[10-12].

The mixture of hydrogen peroxide (H_2O_2) and Fe^{2+} involves the production of the hydroxyl radical (•OH), which is highly oxidative to organic compounds found in wastewater[13] and can non-selectively oxidize almost all polluting organic compounds[14]. The destiny of organic compounds and their degradation by products depends primarily on their reactions with •OH[15]. •OH attack organic pollutants and, as final products, lead to complete destruction of pollutants to CO_2, water and inorganic salts. Classic Fenton process (CFP) includes Equation 1-7 reactions[4, 15] and modified Fenton process (MFP) includes Equation 8-9 reactions[16].

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

$$Fe^{3+}+H_2O_2 \rightarrow Fe^{2+}+ HO_2+ H^+ \qquad \ldots (2)$$

$$\bullet OH + H_2O_2 \rightarrow HO_2 + HO_2 \qquad \ldots (3)$$

$$\bullet OH + Fe^{2+} \rightarrow Fe^{3+} + OH^- \qquad \ldots (4)$$

$$Fe^{3+} + HO_2 \rightarrow Fe^{2+} + O_2H^+ \qquad \ldots (5)$$

$$Fe^{2+} + HO_2 + H^+ \rightarrow Fe^{3+} + H_2O_2 \qquad \ldots (6)$$

$$2HO_2 \rightarrow H_2O_2 + O_2 \qquad \ldots (7)$$

$$Fe^0+ 2H^+ \rightarrow Fe^{2+}+ H_2 \qquad \ldots (8)$$

$$2Fe^{3+}+ Fe^0 \rightarrow 3Fe^{2+} \qquad \ldots (9)$$

Production of •OH(Eq. 1) is very rapid. The net reaction can be generally defined as the decomposition of H_2O_2 in the presence of iron as a catalyst.

$$2Fe^{2+} + H_2O_2 + 2H^+ \rightarrow 2Fe^{3+} + 2H_2O \qquad \ldots (10)$$

Equation (10) shows that the reaction is complete under acidic conditions, that is, the presence of H^+ ions is necessary for the dissociation of H_2O_2 ions. Iron acts as a catalyst in the above reactions.

The photo-Fenton process is a combination of Fenton's reagents (H_2O_2 and Fe^{2+}) and UV, which give rise to extra •OH by two additional reactions: (i) photo-reduction from Fe^{3+} to Fe^{2+} ions as shown in Equation 2, and (ii) more peroxide photolysis with short wavelengths (Eq. 12) (Ref. 17).

$$Fe(OH)^{2+} + h\nu \rightarrow Fe^{2+} + \bullet OH \quad \dots (11)$$

$$H_2O_2 + h\nu \rightarrow 2OH \quad \dots (12)$$

In photo-Fenton process, there is the formation of •OH from the decomposition of H_2O_2 in the presence of iron ions and under acidic conditions[4]. •OH reacts non-selectively with organic pollutants and oxidizes them to form more biodegradable intermediates[18,19].

Methods such as coagulation and flocculation[20-22], adsorption[23], chemical oxidation[7,24,25], reverse osmosisv[26-28], nanofiltration[29], biological process[30,31] and photo-fenton[32] are involved in the treatment of leachate. The Photo-Fenton process is emerging as an attractive and promising technology for pretreatment of hard-to-degrade compounds[31,33].

In this study, removal of organic pollutants from landfill leachate was carried out by Photo-Fenton systems. To maximize the efficiency of the process, it was first necessary to determine the optimum pH, UV power, oxidant and catalyst dosage[34]. Both classical fenton (Fe^{2+}) and modified fenton (Fe^0) were used in the study. The main objectives were to optimize the process and explore the factors affecting removal efficiency and to establish equations describing the efficiency of COD removal from leachate according to operating conditions.

Experimental Section

Sanitary landfill leachate

Based on age, leach ate is divided into three categories: young (less than one-year-old), medium (1-5 years) and old, or stabilized leach ate (older than 5 years). Stabilized leachate is very difficult to treat because of its low biodegradability. Young and medium leachates can be effectively treated by biological methods[35]. The leachate used in this study is stabilized leachate. The leachate samples were taken from Sivas/Turkey urban solid waste landfill with a waste age of more than 10 years. Samples were collected in 5 L plastic bottles and stored at 4°C to minimize oxidation. The leachate used in the study has *p*H 6.8, TS 35,000 mg/L and COD 40,550 mg/L.

Chemicals and analytical methods

$FeSO_4.7H_2O$ salt and stock solution of H_2O_2 (35% H_2O_2 solution) were used as Fenton reagent in the experiments performed with Fe^{2+}. To adjust the *p*H of the leachate to acidic conditions, 0.1 and 1 N H_2SO_4 solutions were used. The Fe^0 solution used in the study was obtained by the performed synthesis. For this, 5.34 g of $FeCl_2.4H_2O$ was mixed in 30 mL of solution (24 mL of ethanol + 6 mL of distilled water) in a magnetic stirrer. It was prepared by dissolving 3.05 g of $NaBH_4$ in 100 mL of distilled water. The prepared $NaBH_4$ solution was added dropwise to the Fe solution mixed in the magnetic stirrer. The resulting black sludge was separated by centrifugation, washed with 25 mL of ethanol, and then centrifuged again. It was dried at 50°C until completely dry[36].

Standard methods[37] were followed throughout the landfill leachate COD removal process. *p*H was measured with Adwa AD8000 brand *p*H meter.

Experimental procedures

Optimal doses in the Fenton process can be determined in one of the following ways; (i) varying the Fe^{2+} concentration at a fixed H_2O_2 dosage and then optimizing the H_2O_2 dosage at that Fe^{2+} concentration[38], (ii) choosing the best combination of H_2O_2 and Fe^{2+} in terms of COD removal from various combinations[39], (iii) Finding the optimal ratio of H_2O_2 and Fe^{2+}, and then using optimal dosages at established rates[7]. In this study, dosages were determined in accordance with item (i).

Photo-Fenton experiments were carried out in the photoreactor whose schematic representation is given in Fig. 1. Three different UV light sources, namely UV-A (365 nm), UV-B (302 nm) and UV-C (256 nm), were

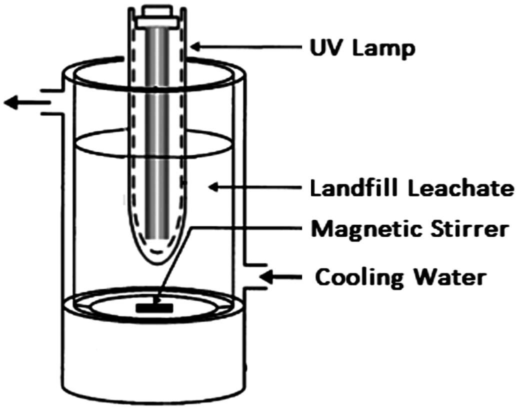

Fig. 1 — Photo-Fenton oxidation application reactor

used in the study. Each of the Fenton optimization experiments was carried out in 250 mL bottles using 100 mL of leach ate. The optimum amounts of iron and H_2O_2 obtained as a result of optimization were used in the Photo-Fenton experiments.

Results and Discussion

Fenton application

Effect of iron amount

In order to determine the effect of Fe^{2+} and Fe^0 amount on treatment efficiency, 40 g/kg TS H_2O_2, pH 3 and reaction time were kept constant for 30 minutes at different dosages (2, 4, 6, 8, 10, 20, 30, 40, 50 g/ kg TS) iron was applied to Fenton oxidation (Fig. 2).

Since the amount of iron present affects the initial rate of the reaction, an appropriate dosage of iron is required for the reaction to start. With the increase in the amount of iron ions present in the system, the COD removal efficiency and speed increase, but as seen in Fig. 2, the removal rate and efficiency decrease after the iron ions reach a certain concentration[40, 41].

As a result of the experiments, the COD removal efficiency varies between 21.40 and 33.71% for Fe^{2+} and between 33.71 and 47.91% for Fe^0. The optimum amount of iron for Fe^{2+} and Fe^0 was determined as 4 g/kg TS. Similar results were obtained in different studies[42, 43].

The mass ratio of H_2O_2 and Fe^{2+} is crucial for the overall cost of the process and the removal efficiency[7]. An increase in iron dose increases COD removal[40]. In addition, the iron concentration causes an excessive increase in the $Fe(OH)^{2+}$ complex ion after a certain value, this ion absorbs UV light and causes a significant decrease in oxidation power[43].

Effect of H_2O_2

H_2O_2 is the main source of •OH radicals produced as a result of the Fenton reaction and oxidation. However, its excessive use both increases the cost of treatment and causes scavenging of •OH radicals that carry out oxidation, so it needs to be optimized[45]. Within the scope of the study, *p*H 3, Fe^{2+} and Fe^0 4 g/kg TS and H_2O_2 1, 3, 5, 10, 20, 40, 60 g/kg TS dosages were studied at different H_2O_2 concentrations to determine the optimum amount of H_2O_2 in the Fenton process (Fig. 3) .

As a result of the experiments, the optimum amount of H_2O_2 was determined as 5g/kg TS with 48.86% yield for Fe^{2+} and 59.27% yield for Fe^0. Similar results were obtained in different studies[43].

Understanding the roles of H_2O_2 and iron in the removal of organic compounds by the Fenton process helps determine optimal reagent dosages. Of the two reagents, H_2O_2 is the more critical because it directly affects the theoretical maximum mass of •OH produced[46]. In general, it can be said that the rate of degradation of organic compounds increases as the H_2O_2 dosage increases up to a critical hydrogen peroxide concentration. When a higher-than-critical concentration is used, H_2O_2 itself contributes to the •OH scavenging capacity[47]. In this case, the reaction of hydrogen peroxide with hydroxyl radicals prevents further degradation of the organic matter[48]. Low H_2O_2 concentration causes insufficient •OH to be produced for the oxidation of organic substances[32]. In addition, it can cause sludge flotation due to the spontaneous decomposition of excess H_2O_2 and the release of O_2 gas[38].

Effect of pH

In the Fenton process, the *p*H parameter generally changes in the range of 2-5. At *p*Hs outside this range, oxidation decreases due to the formation of lower hydroxyl radicals[49]. Since $[Fe^{2+}(H_2O)]^{2+}$ occurs at low *p*H, less hydroxyl radicals are formed[50].

The stability of the Fe^{2+}, Fe^0 and H_2O_2 reaction

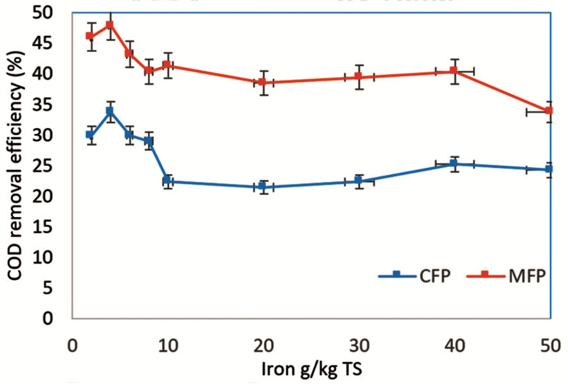

Fig. 2 — Effect of iron content on COD removal

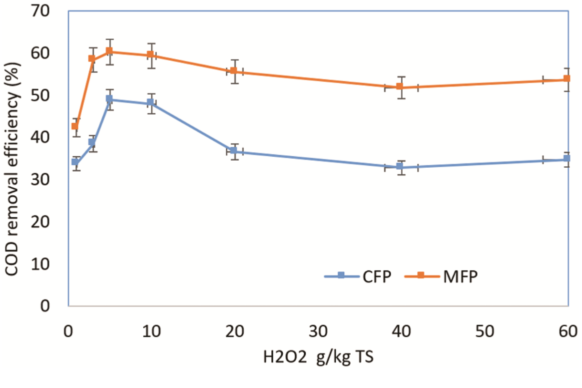

Fig. 3 — Effect of H_2O_2 amount on COD removal

during Fenton oxidation is very important and depends on *p*H. In order to determine the optimum *p*H value of the leachate, the Fenton process was applied at different *p*H ranges (*p*H 1-8) with keeping the other parameters to be constant (30 min of reaction time, 4g/kg TS of Fe^0, 4g/kg TS of Fe^{2+} and 5g/kg TS of H_2O_2). Figure 4 shows the effect of different *p*H conditions on COD removal efficiencies.

The removal efficiencies obtained at different *p*Hs were determined as 48.86% and 60.22% for Fe^{2+} and Fe^0, respectively. Accordingly, the optimum *p*H of the Fenton process was found to be 3 for CFP and MFP. Similar results have been obtained in different studies[51,52].

The pH value affects the formation of •OH radicals and thus the oxidation efficiency. The reason why the removal process is inefficient at *p*H >3 is that the rate of spontaneous decomposition and degradation of H_2O_2 is very high[44]. High *p*H values can cause the automatic decomposition of H_2O_2 and a decrease in the oxidation potential of •OH[53]. However, At very low *p*H, the hydronioma ion $(H_3O)^+$ becomes dominant and the H_2O_2 stability increases, thus limiting the formation of the •OH and consequently less pollutant is oxidized[44]. Production of •OH is higher in the *p*H range of 2-4 due to H_2O_2 regeneration or increase in reaction rates[54].

Optimum *p*H values for conventional landfill leachate treatment, reported for photo-Fenton processes, range from 2.0 to 4.5 (Refs.5, 41, 46, 54 & 55). A *p*H below optimum can inhibit oxidation in three ways. First, $[Fe(H_2O)]^{2+}$, which is formed at extremely low *p*H values, reacts relatively slowly with H_2O_2, producing fewer •OH radicals[56]. Second, the scavenging effect of H^+ on •OH becomes more significant at a lower *p*H[57]. Third, extremely low *p*H can inhibit the reaction between Fe^{3+} and H_2O_2[58]. In addition, amorphous iron oxyhydroxide sludge, which can accumulate at *p*H above 5.0, prevents UV light from passing through the reactor[55]. A *p*H in the neutral-alkaline range also inhibits Fenton oxidation[4].

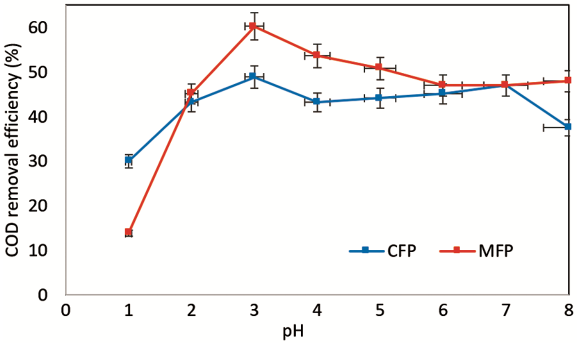

Fig. 4 — Effect of *p*H on COD removal

Effect of reaction time

COD removal efficiencies were investigated at 5, 10, 15, 20, 25, 30, 45 and 60 min with keeping the optimum *p*H 3, iron 4 g/kg TS and H_2O_2 5 g/kg TS constant (Fig. 5).

The organic matter degraded rapidly during the first 30 min of the reaction time and then slowed down. The initial rapid degradation is largely due to readily degradable organics. Current productivity also increased in the first 15-30 min, but gradually decreased thereafter[41]. In Fig. 5, it can be seen that most of the organic compounds are formed in the first 30 min, during the first stage where easily oxidizable compounds are rapidly destroyed by hydroxyl radicals. As time went on, the increase in COD removal slowed down. It is thought that when the refractory materials in the leachate are oxidized, the residual materials are not easily degraded by •OH, thus the efficiency becomes stable.

According to the experiments, the most appropriate reaction time was determined as 30 minutes with 47.91% efficiency for Fe^{2+} and 58.33% for Fe^0. The high yield of the reaction in a short time indicates that the oxidation reactions took place at a large rate and very quickly at the beginning.

Effect of different UV light sources

In the photo-Fenton oxidation application, optimum conditions obtained in Fenton oxidation were used. Experiments were carried out at 3 different wavelengths, namely UV-A, UV-B and UV-C, and their COD removal efficiencies were examined. The results are given in Fig. 6.

As seen in Fig. 6, while the highest COD removal was obtained in UVB with 74.58% in MFP, the highest efficiency was obtained in UVA with 49.17% in CFP. In the study, it is seen that the UV light source has a positive effect on the COD removal efficiency. The

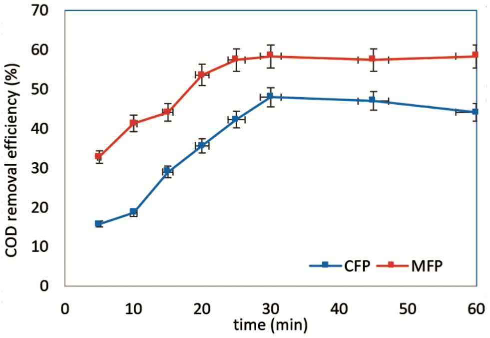

Fig. 5 — Time based COD removal efficiencies

Table 1 — Kinetic parameters in Fenton processes

Process	Zero Order Kinetics		1st Order Kinetics		2nd Order Kinetics	
	k_0 (mg/L.min)	R^2	k_1 (1/min)	R^2	k_2 (L/mg.min)	R^2
CFP	0.549	0.64	0.018	0.60	-0.0007	0.55
MFP	0.416	0.62	0.009	0.59	-0.0002	0.56
CFP/UV	-0.339	0.70	-0.008	0.71	0.0002	0.72
MFP/UV	0.1142	0.27	0.005	0.32	-0.0001	0.36

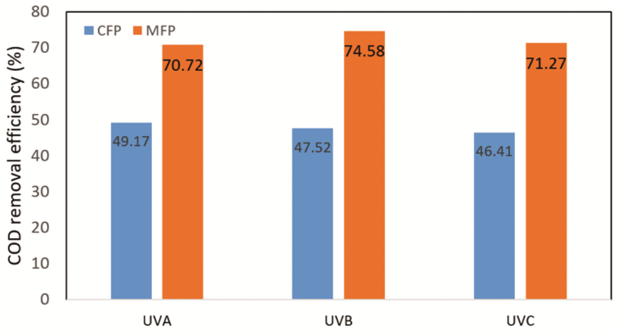

Fig. 6 — Effect of different UV light sources

efficiency, which was 48.86% in CFP under UVA light source, increased to 49.17% in photo-Fenton application and from 59.27% to 70.72% in MFP. In the photo-Fenton reaction that takes place under UV light source, more •OH radicals are formed as a result of decomposition of photoactive $Fe(OH)^{2+}$,[59-60] which leads to the formation of COD increases costs.

Kinetic study

For the optimum Fenton processes obtained in the study, zero (Eq. 13), first (Eq. 14) and second (Eq. 15) order kinetic models were applied to the time-dependent COD change data.[61] The calculated kinetic parameters are given in Table 1.

$$C = C_0 - k_0 . t \qquad \text{... (13)}$$

$$\ln C = \ln C_0 - k_1 . t \qquad \text{... (14)}$$

$$\frac{1}{C} = \frac{1}{C_0} + k_2 . t \qquad \text{... (15)}$$

In these equations, C_0 is the initial COD concentration (mg/L); C is the COD concentration at any time (mg/L); k_0, k_1 and k_2 are the kinetic constants of zero order, 1st order and 2nd order reaction kinetics, respectively; and t is reaction time (min).

As seen in Table 1, the highest R^2 values were obtained in the CFP/UV process (R^2=0.70-0.72). While this process complies with all three kinetics, the highest R^2 values (0.62-0.64) were obtained in zero order kinetics in CFP and MFP processes. The low R^2 values were obtained in MFP/UV.

Conclusion

In this study, the effects COD removal from sanitary landfill leachate has been determined by exposing the leachate to CFP, MFP and photo-Fenton oxidation. Fe^{2+} and Fe^0 were used in Fenton oxidation and optimum conditions are determined by applying different pH, H_2O_2 and iron amounts. Accordingly, the equilibrium time is investigated with the study performed in the range of 1-60 min. Based on the determined optimum conditions; Photo-Fenton oxidation is performed at different wavelengths (UV-A, UV-B and UV-C) for both Fe^{2+} and Fe^0. The optimum conditions determined in the study are *p*H 3, Fe^{2+} and Fe^0 4 g/kg TS, H_2O_2 amount 5 g/kg TS and 30 min as reaction time.COD removal efficiency from leachate was 48.86% in CFP and 59.27% in MFP. In the photo Fenton process, where different wavelengths are used, the highest efficiency was 75.58% in the MFP/UVB process, while the lowest removal efficiency was 47.52% in the CFP/UVB process. The efficiency increased from 48.86% in CFP to 49.17% under UVA light source, and from 59.27% to 70.72% in MFP. In the kinetic study, the highest R^2 values were obtained in the CFP/UV process. This process is suitable for all three kinetic models. In addition, CFP and MFP were found to fit the 0th order kinetic model. It was concluded that the Fenton and photo-Fenton processes applied for COD removal from landfill leachate successfully performed the COD removal, and the addition of UV light to the processes increased the COD removal efficiency.

Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

Conflict of interest

The authors declare that they have no conflict of interest.

Acknowledgement

This work is supported by the Scientific Research Project Fund of Sivas Cumhuriyet University under the Project number M-761. The authors sincerely thank CÜBAP Chairmanship for their endorsement.

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