

Physical and oxidative removal of organics during Fenton treatment of mature municipal landfill leachate

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Abstract

Municipal landfill leachate, especially mature leachate, may disrupt the performance of moderately-sized municipal activated sludge wastewater treatment plants, and likewise tend to be recalcitrant to biological pretreatment. Recently, Fenton methods have been investigated for chemical treatment or pre-treatment of mature leachate. In this paper, the results of laboratory tests to determine the roles of oxidation and coagulation in reducing the organic content of mature leachate during Fenton treatment are presented. The efficiencies of chemical oxygen demand (COD) oxidation and coagulation were tested, and the ratio of COD removal by oxidation to that by coagulation was assessed, under various operating conditions. Low initial pH, appropriate relative and absolute Fenton reagent dosages, aeration, and stepwise addition of reagents increased COD removal by oxidation and the importance of oxidation relative to coagulation. Simultaneous aeration and stepwise reagent addition allowed comparable treatment without initial acidification pH, due to the generation of acidic organic intermediates and the continuous input of CO₂. On the other hand, high COD oxidation efficiency and low ferrous dosage inhibited COD removal by coagulation. At significantly high oxidation efficiency, overall COD reduction decrease slightly due to low coagulation efficiency. Under the most favorable conditions (initial pH 3, molar ratio [H₂O₂]/[Fe²⁺] = 3, [H₂O₂] = 240 mM, and six dosing steps), 61% of the initial COD was removed, and the ratio of COD removal oxidation to coagulation was 0.75. Results highlighted the synergistic roles of oxidation and coagulation in Fenton treatment of mature leachate, and the role of oxidation in controlling the efficiency of removal of COD by coagulation.

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Keywords: Fenton treatment; Landfill leachate; Oxidation; Coagulation; Organic contaminants

1. Introduction

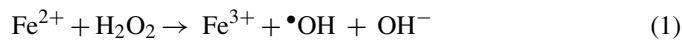
Sanitary landfills have been suggested to be the most economical and environmentally acceptable method for disposal of municipal solid wastes, in the United States and throughout the world [1]. However, leachate produced from landfills is a high strength organic wastewater which, when discharged directly to a municipal wastewater treatment plant, may cause corrosion of the pump station, difficulty in maintaining constant effluent chlorine residual, and sludge bulking and settling problems. Biological methods are typically applied for treatment of young leachates (e.g., from landfills of less than 1–2 years age), characterized by high 5-day biochemical oxygen demand (BOD₅)/chemical oxygen demand (COD) ratios (>0.6) and high

concentrations of low molecular weight organics [2]. However, such methods are not effective for treatment of mature leachates (e.g., from landfills of more than 5–10 years age), due to their low BOD₅/COD ratios (<0.3) and high fraction of high molecular weight, refractory organics. Hence, several physicochemical processes have been studied or used for pretreatment or full treatment of mature leachate [3–8].

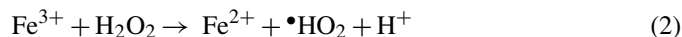
Among potential physicochemical technologies for leachate treatment, the Fenton process has been extensively studied in recent years [9–17], and analyses indicate Fenton process to be one of the most cost-effective alternatives for this application [18]. Organic removal efficiency depends upon several operating parameters, including reaction pH, dosages of Fenton reagents, aeration, coagulation pH, mode of reagent addition, temperature, UV irradiation, and subsequent pH adjustment for coagulation. A detailed discussion on the effects of these operational factors is available somewhere [19]. The typical Fenton wastewater treatment process includes four stages: oxidation,

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neutralization, coagulation/flocculation, and solid–liquid separation [20,21]. Hence, organics are removed by both oxidation and coagulation. The oxidation is generally ascribed to generation of hydroxyl radical ($\bullet\text{OH}$) according to Eq. (1), a strong and indiscriminant oxidant. However, other species including ferryl moieties are also candidate oxidants [22,23].



Some of Fe^{3+} produced can be reduced to Fe^{2+} through Eq. (2), although the rate is several orders of magnitude slower than that of Fe^{2+} to Fe^{3+} conversion through Eq. (1).



The Fe^{2+} generated in Eq. (2) may react with H_2O_2 to produce more $\bullet\text{OH}$ through Eq. (1). Coagulation is then due to precipitation of ferric-oxyhydroxides after the neutralization stage. However, most previous investigations focused primarily on the effects of operating parameters on overall organics removal efficiencies; how these parameters impact oxidation and coagulation and whether oxidation or coagulation predominates in terms of treatment under various conditions are not well recognized, and such understanding as pertains to treatment of landfill leachate is still more limited. Yoon et al. [24] concluded that coagulation played a primary role in Fenton removal of organics from leachate, based only on the observation that both Fenton treatment and simple coagulation removed high molecular organics more readily and selectively. Wang et al. [25] and Lau et al. [26] reported that oxidation and coagulation were responsible for approximately 20 and 80% of overall COD removal, respectively, in Fenton treatment of a biologically stabilized leachate. Kang and Hwang [27] found that pH and absolute dosages of Fenton reagents significantly affected the removal of COD from leachate by oxidation and coagulation, and that coagulation pH influenced the efficiency of coagulation. However, more required relationships particularly between oxidation and coagulation in terms of removal efficiency have not been clear.

In this paper, overall COD removal efficiency, and COD reduction by oxidation and by coagulation under various operating conditions are reported for a mature leachate. The ratio of COD removal by oxidation to that by coagulation was used to determine whether oxidation or coagulation played a predominating role under the tested conditions. Specifically, the effects of initial pH, molar ratio and absolute dosages of Fenton reagents, aeration, and mode of stepwise reagent addition on oxidation and coagulation are described. In addition, the interaction between oxidation and coagulation in terms of treatment efficiency is characterized. Finally, the interactive effects of aeration and stepwise addition of reagents on both oxidation and coagulation are described.

2. Experimental

Landfill leachate was collected from the Polk County North Central Landfill collection tank (Winter Haven, FL, USA). The mean BOD_5/COD was less than 0.05, typical for mature leachate. The collected leachate was stored in a zero headspace plastic bottle in refrigerator at 4°C until use. The average composition of the tested landfill leachate is listed in Table 1. All chemicals were at least analytical grade and were used as received, except as noted. The raw landfill leachate was pre-filtered through a glass filter paper (934-AH, Whatman, Cifton, VA, USA) to remove large particles and debris, and maintain uniformity of tested samples. All runs were conducted at room temperature and atmospheric pressure. Initial leachate pH was adjusted to the desired value with concentrated sulfuric acid (H_2SO_4 , 96%, FishChemical, Fair Lawn, NJ, USA). Subsequently, 200 mL of prefiltered leachate was dispensed to a 1-L beaker. The leachate was stirred thoroughly with a magnetic stirrer. In the aerated Fenton tests, injected air was supplied by a small air pump (20 L/h, Tetrac, China). The height from the leachate table to the beaker brim adequately avoided the overflow due to foaming occurring at initial stage of reaction under aeration. In the experiments to investigate effects of aeration and stepwise feeding of Fenton reagents, Fenton oxidation proceeded for 9 h, and the total reagent dose was added incrementally at each designated addition time. In other experiments, Fenton oxidation proceeded for 2 h. Reagents addition was as follows. First, granular ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, heptahydrate, FishChemical, Fair Lawn, NJ, USA) was added. Subsequently hydrogen peroxide solution (H_2O_2 , 30% w/w, VWR, West Chester, PA, USA) was added. After the designated oxidation time, NaOH pellets were added to the rapidly stirred solution, to increase the pH to approximately 6.5. A solution of 10 M NaOH solution was then added dropwise, to a pH of 8.0. The beaker with leachate was transferred to a Phipps & Bird Stirrer (Model 7790, Richmond, VA, USA) for a period of 20 min flocculation at 20 rpm. Overall COD removal, and COD removal by oxidation and by coagulation were measured by a slight modification of the method of Kang and Hwang [27]. Accordingly, 100 mL aliquot of uniformly mixed solution was immediately dispensed to a glass cylinder and heated in a 50°C water bath (8851, Cole Parmer, Chicago, IL, USA) for 30 min to remove any residual H_2O_2 in solution, and the sample was brought to the room temperature for a 90 min sedimentation period. Then, the volume of the settled iron sludge was recorded. After that, the COD values of the supernatant and the settled sludge samples were measured separately. The former indicated COD in the effluent after the overall process, and the latter indicated the COD of the solid phase that was contributed from these organics coagulated. COD removal by oxidation was the difference between COD reduction by the overall process and the COD

Table 1
Average composition of the tested landfill leachate

Parameters	COD (mg/L)	BOD_5/COD	Electrical conductivity (ms/cm)	pH	$\text{NH}_3\text{-N}$ (mg/L)	Alkalinity (CaCO_3 mg/L)
	1100–1300	<0.05	8.30	8.18	300	3050

coagulated to the solid phase. Sample pH was measured by pH meter. COD was measured colorimetrically following digestion (20–1500 mg/L range, HACH, Loveland, CO, USA). Error bar in the figures represent one standard deviation ($n=3$).

3. Results and discussion

3.1. Initial pH

The effect of initial pH on COD removal efficiency is shown in Fig. 1(a). At a H_2O_2 concentration of 23.5 mM and Fe^{2+} concentration of 14 mM, maximum COD removal efficiencies by oxidation and coagulation occurred at pH 2.5–3.5, such that overall COD removal efficiency peaked above 50%. These observations are consistent with those of Kang and Hwang [27]. At extremely low pH (<2.5), COD oxidation efficiency and overall COD removal efficiency decreased sharply, due principally to the lower reaction rate of $[\text{Fe}(\text{H}_2\text{O})]^{2+}$ and H_2O_2 [28], the increased scavenging of $\bullet\text{OH}$ by H^+ [29], and the increased inhibit on the reaction between Fe^{3+} and H_2O_2 due to H^+ [30]. On the other hand, COD removal dropped significantly with

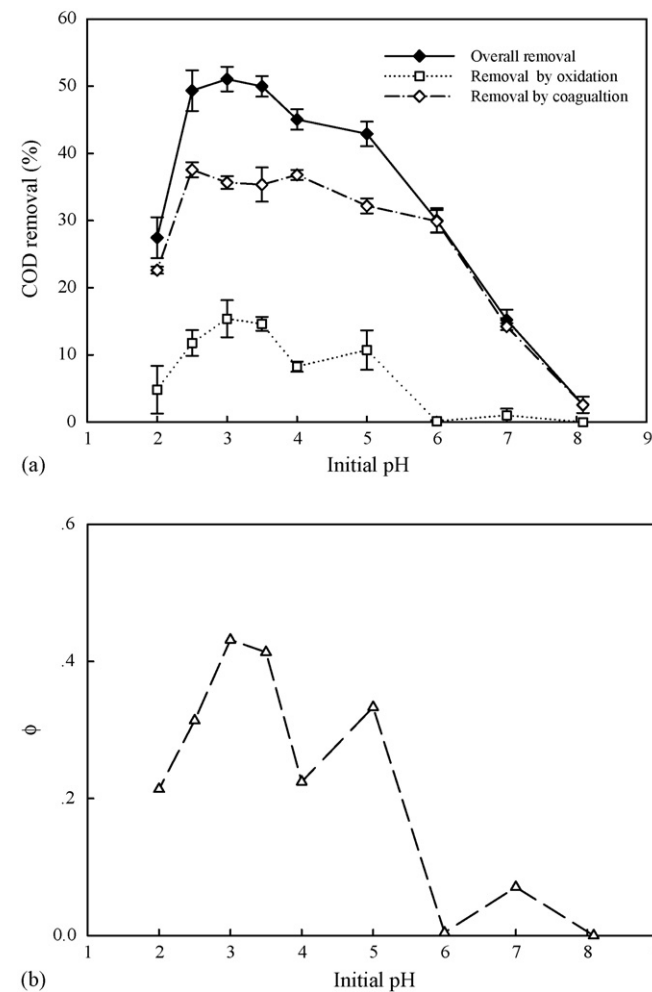


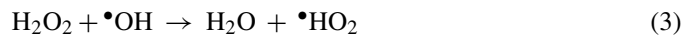
Fig. 1. The effect of initial pH on: (a) COD removal efficiencies; and (b) ratio ϕ of COD removal by oxidation to that by coagulation, in Fenton treatment of leachate (conditions: $\text{Fe}^{2+} = 14$ mM; $\text{H}_2\text{O}_2 = 23.5$ mM; mean initial COD = 1166 mg/L).

increasing pH (>5.0), due to the increasing rate of autodecomposition of H_2O_2 , deactivation of iron ion into iron oxyhydroxides, the increased scavenging effect of carbonate and bicarbonate on $\bullet\text{OH}$, and the decreased oxidation potential of $\bullet\text{OH}$. Therefore, COD removal by oxidation, coagulation, and overall process depended strongly on initial pH.

The effect of initial pH on the ratio ϕ of COD removal by oxidation to that by coagulation is shown in Fig. 1(b). This ϕ is proposed as a simple but useful indicator to evaluate the relative importance of oxidative degradation of organics in Fenton treatment. As shown, the ratio peaked around 0.43 at pH 3.0, thus suggesting an optimal initial pH for oxidative Fenton treatment. In contrast, at $\text{pH} \geq 6.0$, the ratio of removal by oxidation to overall COD removal was less than 0.10, indicating that coagulation predominated in Fenton treatment of mature leachate under these conditions. Hence, initial pH also significantly influenced importance of oxidation relative to coagulation.

3.2. Dosages of Fenton reagents

The effect of molar $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio on COD removal efficiencies at a fixed H_2O_2 dosage of 180 mM at initial pH 3.0 is shown in Fig. 2. The maximum oxidative COD removal efficiency of 27% occurred at a molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] = 3$. Oxidation efficiency dropped sharply to one half of the maximum when the molar $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio increased to 12. This result is attributed to the scavenging effect of peroxide on hydroxyl radicals, which presumably became stronger as the relative ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ rapidly increased, as shown in Eq. (3).



Oxidation efficiency also dropped a little at a molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] < 3$, due to the increasing scavenging effect of Fe^{2+} on hydroxyl radicals, shown in Eq. (4).

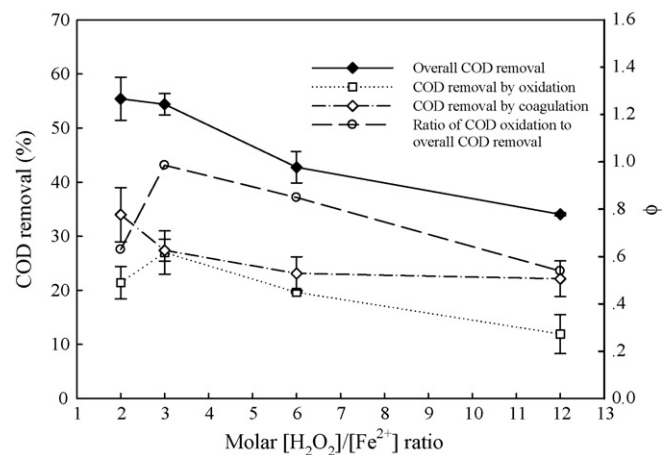


Fig. 2. The effect of molar $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio on COD removal efficiencies and ratio ϕ of COD removal by oxidation to that by coagulation, at a fixed peroxide dosage in Fenton treatment of leachate (conditions: initial pH 3; $[\text{H}_2\text{O}_2] = 180$ mM; mean initial COD = 1133 mg/L).

Results indicated the optimal molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ to be approximately 3.0, for oxidation of this leachate. On the other hand, from molar ratios $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ of 2 to 12 under a fixed H_2O_2 dosage of 180 mM, COD removal by coagulation decreased slightly from 34 to 22%, primarily because the content of ferrous ion added, which was eventually transferred to ferric-oxyhydroxides as the coagulant after neutralization, decreased by 83%. Thus, Fe^{2+} dosage strongly influenced the treatment efficiency of coagulation. Overall COD removal of ca. 55% resulting from the interaction between oxidation and coagulation was almost constant at molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] = 2\text{--}3$, gradually decreasing to 34% at molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] = 3\text{--}12$. Additionally, the ratio φ was related to molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$. The maximum $\varphi \approx 1.0$ occurred at $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] = 3.0$.

Overall COD removal, COD removal by oxidation and coagulation, and the ratio φ versus dosages of Fenton reagents are shown in Fig. 3(a)–(c), respectively. As shown in Fig. 3(a), for any particular $[\text{Fe}^{2+}]$, overall COD removal increased with increasing peroxide dosage, though the increase was insignificant at molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] > 3.0$ (for $[\text{Fe}^{2+}] = 30\text{ mM}$, $[\text{H}_2\text{O}_2] > 90\text{ mM}$; for $[\text{Fe}^{2+}] = 60\text{ mM}$, $[\text{H}_2\text{O}_2] > 180$; and for $[\text{Fe}^{2+}] = 90\text{ mM}$, $[\text{H}_2\text{O}_2] > 270$). Furthermore, overall COD removal increased with increasing Fe^{2+} concentration over the interval 30–60 mM, but did not increase significantly over the interval 60–90 mM, indicating that the optimal Fe^{2+} dosage for overall COD reduction was near 60 mM.

Two characteristics of the interaction between oxidation and coagulation during Fenton treatment can be noted in Fig. 3(b). First, as peroxide dosage and oxidation efficiency increased, COD coagulation decreased gradually, implying that remaining oxidative by-products were not as amenable to coagulation. This explanation would coincide with the observation that Fenton oxidation and coagulation both remove high molecular weight organics preferentially [31,32,24]. Second, at high peroxide dosages, higher Fe^{2+} dosage led to higher efficiency of coagulation due to higher concentrations of ferric coagulant after neutralization; however, this effect was not obvious at low peroxide dosages where low oxidation efficiency occurred. That is, ferrous dosage greatly influenced COD removal by coagulation at high oxidation efficiency, but not at low oxidation efficiency. Therefore, oxidation efficiency apparently controls coagulation efficiency, such that high oxidation efficiency may cause relatively low coagulation efficiency.

As shown in Fig. 3(c), the ratio φ increased with increasing $[\text{H}_2\text{O}_2]$ for any particular $[\text{Fe}^{2+}]$. At $[\text{H}_2\text{O}_2] > 90\text{ mM}$, higher ferrous dosage apparently resulted in a lower ratio, though the phenomena was not observed at $[\text{H}_2\text{O}_2] < 90\text{ mM}$. The ratio φ was increased to ca. 2.10 only at an extremely high molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] = 10$.

3.3. Aeration and stepwise addition of Fenton reagents

The effects of aeration alone, stepwise addition alone, and the combination of the two on COD removal were investigated, as shown in Fig. 4. The addition of reagents in nine steps

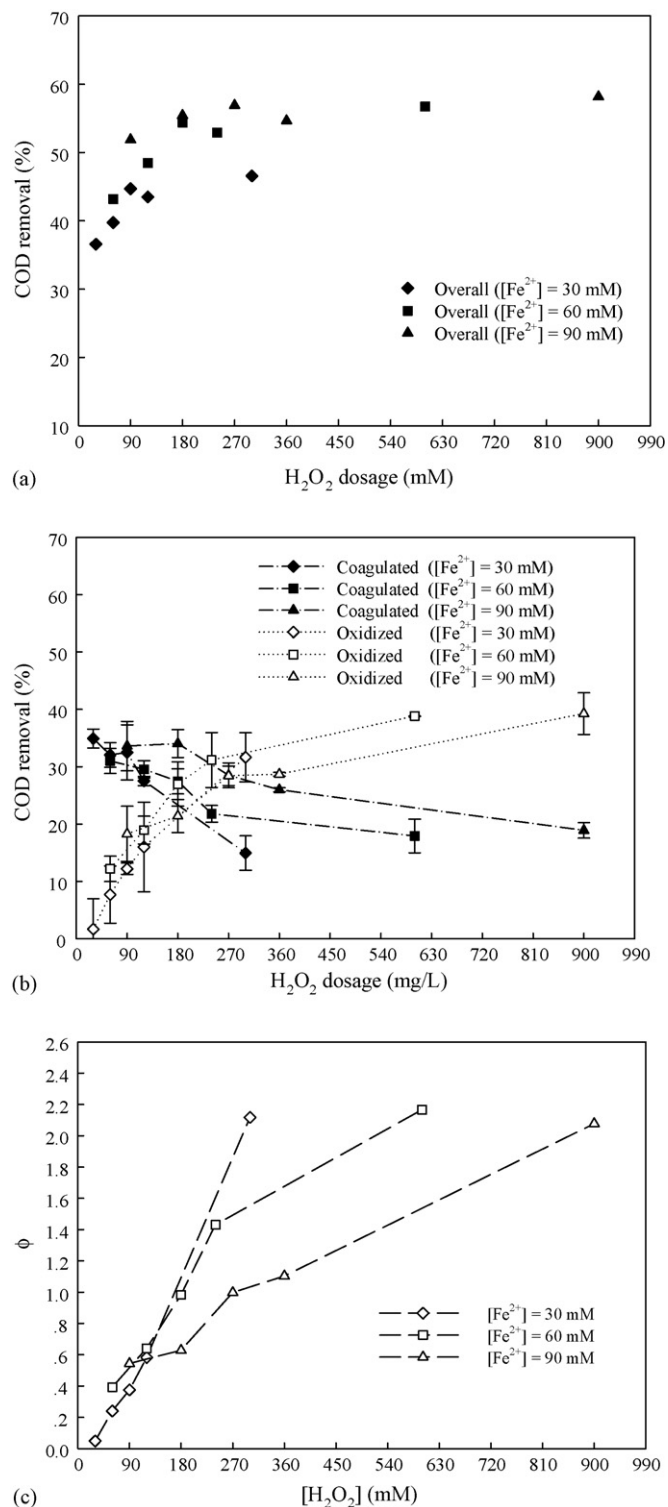


Fig. 3. Fenton treatment of leachate at various dosages of Fenton reagents: (a) overall COD removal; (b) COD removal by oxidation and coagulation; and (c) ratio φ of COD removal by oxidation to that by coagulation (conditions: initial pH 3; initial COD = 1133 mg/L).

increased COD removal by oxidation slightly, from 18 to 24%, and increased overall COD removal from 46 to 56%. Aeration alone similarly increased COD oxidation to 22%, but did not obviously improve overall COD removal. The combination of

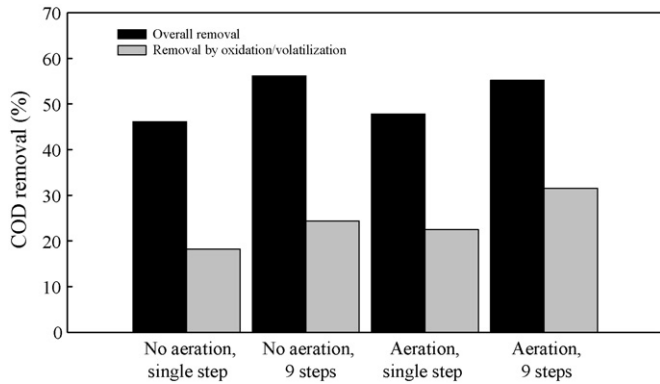


Fig. 4. Effects of aeration and stepwise addition of Fenton reagents on overall COD reduction and COD oxidation in Fenton treatment of leachate (conditions: initial pH 3; total reaction time = 9 h; mean initial COD = 1295 mg/L; $[H_2O_2] = 240$ mM; molar $[H_2O_2]/[Fe^{2+}]$ ratio = 3).

aeration and stepwise addition further improved COD oxidation to 32%, with an overall COD removal of 55%. Stepwise addition and aeration showed a clear synergistic effect on COD oxidation. The positive effect of stepwise addition of H_2O_2 may be due to a reduction in the autodecomposition of high-localized H_2O_2 concentrations at the point of injection, and to reduced scavenging of hydroxyl radicals by hydrogen peroxide [33,34]. Aeration may decrease ineffective consumption of H_2O_2 by maintaining high concentration of O_2 in water and then inhibiting the autodecomposition of H_2O_2 through Eq. (5).



Additionally, O_2 in water may rapidly and usually irreversibly react with carbon-centered radicals (R^\bullet), formed in the reaction of hydroxyl radicals and organic compounds, through Eqs. (6) and (7).

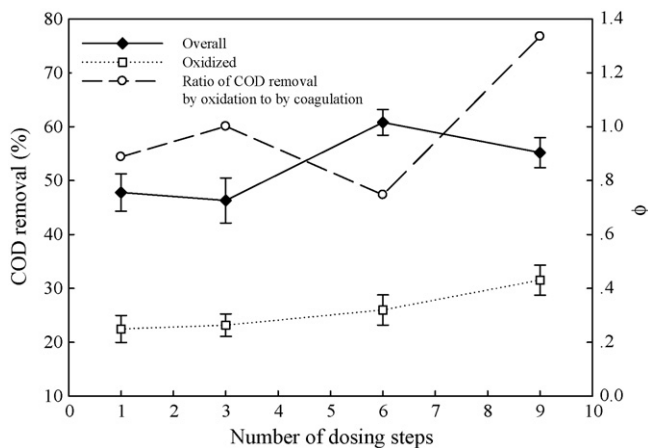
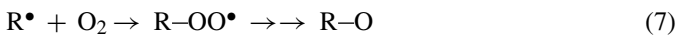
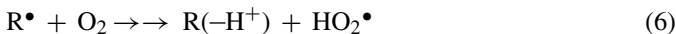


Fig. 5. The effect of number of dosing steps on COD removal and ratio ϕ of COD removal by oxidation to that by coagulation, in Fenton treatment of leachate (conditions: initial pH 3; total reaction time = 9 h; mean initial COD = 1295 mg/L; $[H_2O_2] = 240$ mM; molar $[H_2O_2]/[Fe^{2+}]$ ratio = 3).

These radicals R^\bullet , $R-OO^\bullet$, and $R-O^\bullet$ may couple or disproportionate to form relatively stable molecules, or react with iron ions [35]. The organic intermediates produced may continue to react with hydroxyl radicals and O_2 , thus leading to further decomposition and perhaps mineralization.

The effect of the number of dosing steps on overall COD reduction, COD removal by oxidation, and the ratio ϕ , is shown in Fig. 5. As the number of dosing steps increased from one to nine, COD removal by oxidation increased from 22 to 32%, and the ratio ϕ increased gradually from 0.90 to 1.33. The observed overall COD removal was higher for six steps than for nine, where the maximum oxidation efficiency and the highest ratio ϕ occurred, perhaps again because low molecular weight oxidative byproducts were less amenable to coagulation. As shown, the maximum overall COD removal of approximately 61% was achieved at initial pH 3, molar ratio $[H_2O_2]/[Fe^{2+}] = 3$, $[H_2O_2] = 240$ mM, and six reagent addition steps, in which case the ratio ϕ of removal by oxidation to coagulation was 0.75.

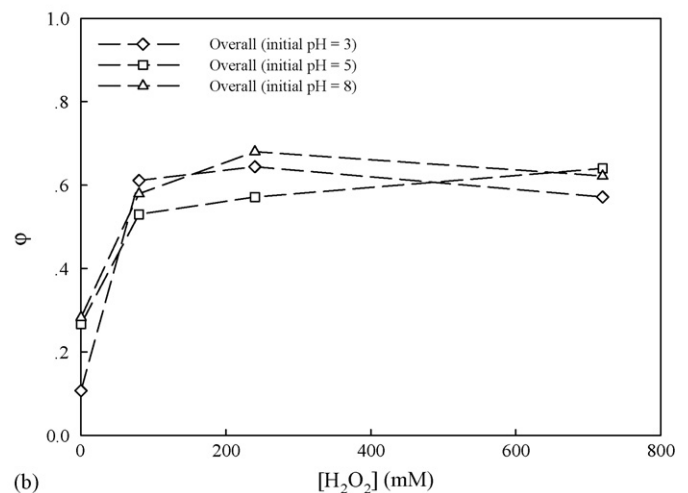
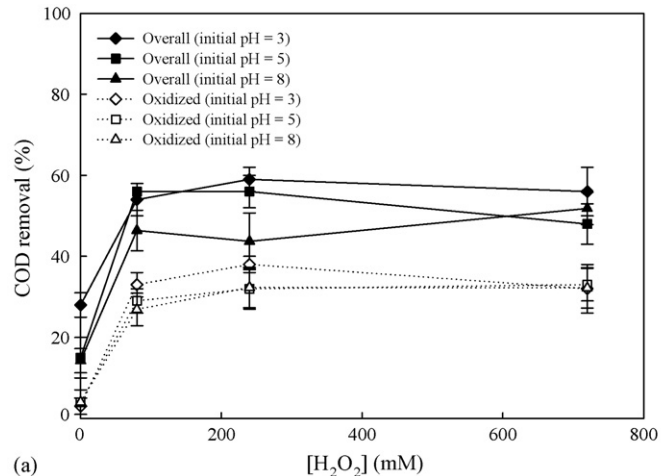


Fig. 6. Aerated Fenton treatment of leachate with stepwise dosing as a function of peroxide dosage and initial pH: (a) overall COD removal, and COD removal by oxidation; and (b) ratio ϕ of COD removal by oxidation to that by coagulation (conditions: total reaction time = 9 h; dosing steps = 9; $[Fe^{2+}] = 60$ mM; aeration rate = 0.83 mL air/min mL sample; mean initial COD = 1288 mg/L).

A set of experiments was conducted to investigate the interaction between initial pH and peroxide dosage on aerated Fenton treatment of leachate with stepwise addition of reagents (Fig. 6(a) and (b)). As shown in Fig. 6(a), in terms of overall COD removal, the need for acidification could be overcome to some degree with increasing peroxide dosage. In particular, overall COD removal increased sharply for hydrogen peroxide addition rates increasing from 0 to 80 mM over the tested pH range, where the increase was minor or absent at higher hydrogen peroxide dosage. As shown, under the conditions tested, hydrogen peroxide dosage was even more dominant in terms of COD oxidation than was observed for overall removal, relative to initial pH. A similar trend was also observed in terms of the ratio ϕ , as shown in Fig. 6(b). This effect may have been enhanced by the stepwise addition of reagents. For example, even at an initial pH of 8, leachate pH during Fenton treatment decreased gradually with dosing steps, such that most of the Fenton oxidative treatment actually occurred at low pH; at hydrogen peroxide dosage equal to 80 mM, the final pH of the group with initial pH 8 was 2.523, approximately equal to final pH 2.312 of the group with initial pH 3. Reasons for this pH decrease during Fenton oxidation may be (a) the formation of organic acids as decomposition intermediates, and (b) the introduction of CO_2 that generates H_2CO_3 .

4. Conclusions

In this study of the traditional Fenton treatment of mature leachate, low initial pH, appropriate relative and absolute dosages of Fenton reagents, aeration, and stepwise addition of reagents all increased COD removal by oxidation as well as the oxidation/coagulation, ratio. When aeration was combined with stepwise addition of reagents, the effect of initial pH on COD removal became minor, due to the gradual decrease in leachate pH with the generation of acidic organic intermediates and continuous input of CO_2 . On the other hand, COD removal efficiency by coagulation was principally influenced by oxidation efficiency and ferrous dosage. High COD oxidation efficiency meant that residual organics probably contained high fraction of low molecular weight organics, most of which were oxidation products of high molecular weight organics originally in the mature leachate, and were not amenable to coagulation treatment. Significantly, high oxidation efficiency even might decrease overall COD reduction due to greatly decreased removal efficiency of coagulation. Hence, oxidation seemed to play a more active role in Fenton treatment, which itself contributed to COD reduction, and controlled the treatment behavior of coagulation. Moreover, high ferrous dosage increased the treatment efficiency by coagulation due to high concentration of coagulant, while the effect was not obvious at low oxidation efficiency. Under the most favorable conditions (initial pH 3, molar ratio $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}] = 3$, $[\text{H}_2\text{O}_2] = 240$ mM, and six dosing steps), 61% of the initial COD was removed, and the ratio of COD removal by oxidation to by coagulation was 0.75. Results highlighted the synergistic roles of oxidation and coagulation in Fenton treatment of mature leachate, and the role of oxidation in controlling the efficiency of removal of COD by coagulation.

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