



**Full Length Article**

## Post-treatment of Aerobically Pretreated Poultry Litter Leachate using Fenton and Photo-Fenton Processes

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### ABSTRACT

Fenton process is generally used to treat highly polluted wastewater or landfill leachate and is based on the addition of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and ferrous sulfate (FeSO<sub>4</sub>) at certain ratio. In this study, different doses of H<sub>2</sub>O<sub>2</sub> and FeSO<sub>4</sub> were applied as a post-treatment to aerobically pretreated poultry litter leachate to compare the treatment efficiency along with the application of UV radiation for removal of chemical oxygen demand (COD), color and hardness. The results revealed that the Fenton treatment of 0.1 mL H<sub>2</sub>O<sub>2</sub> in the presence of 10 mg FeSO<sub>4</sub> as a catalyst showed a maximum removal of COD, while increasing trend for COD removal was observed with the increase in H<sub>2</sub>O<sub>2</sub> concentration. Maximum removal of color and hardness was observed at 1.0 mL H<sub>2</sub>O<sub>2</sub> and 50 mg FeSO<sub>4</sub> per liter of leachate. In general, Fenton treatment 1.0 mL H<sub>2</sub>O<sub>2</sub> plus 50 mg FeSO<sub>4</sub> was found to be the best among all the treatments for removal of color, hardness and COD. Exposure of Fenton treated leachate (1.0 mL H<sub>2</sub>O<sub>2</sub> & 50 mg FeSO<sub>4</sub>) to UV light (Photo-Fenton process) accelerated the removal of COD, color and hardness of the leachate. These findings suggested that post-treatment of aerobically pretreated poultry litter leachate with low dose of Fenton reagent along with short exposure to UV radiation could be an effective tool for the treatment of poultry litter leachate. © 2011 Friends Science Publishers

**Key Words:** Fenton; Photo-Fenton; Poultry litter; COD; Color; Hardness

### INTRODUCTION

Poultry production plays a vital role in bridging the gap between supply and demand of animal protein foods for increasing human population. This sector showed a noticeable progress and development since 1960 and it reaches the growth rate of 8-10% per annum during the last decade. It is one of the largest, highly mechanized, vertically integrated, and intensified animal production industries in Pakistan (Khan, 2008). Several million tonnes of solid wastes are produced annually by poultry farm birds (Atuanya & Aigbirior, 2002). Poultry litter leachate contains a variety of organic and inorganic contaminants including feed additive metabolic wastes, topical pesticides, metals, antioxidants, and toxicants associated with the bedding material. That is why, a high value for COD, BOD (biological oxygen demand), VFA (volatile fatty acid), alkalinity and sulfate (up to 11000 mg L<sup>-1</sup>, 8000 mg L<sup>-1</sup>, 6000 mg L<sup>-1</sup>, 4000 mg L<sup>-1</sup> & 1425 mg L<sup>-1</sup>, respectively) in poultry leachate has been reported (Rao *et al.*, 2008). Nonetheless, very little attention has been given to dispose of such waste safely into the environment. As a result,

quality of the surface and ground water is deteriorated in the vicinity of poultry units due to leaching and runoff.

There are a variety of physicochemical and biological methods that can be used for the treatment of various leachate (Tabrizi & Mehrvar, 2004; Goi *et al.*, 2010; Umar *et al.*, 2010). Biological treatment is commonly used for the removal of the bulk of leachate containing high BOD and COD concentrations primarily due to its higher reliability, simplicity and cost effectiveness (Renou *et al.*, 2008). However, because of obstinate character and composite nature of poultry leachate, conventional biological processes alone may not be successful for complete removal of recalcitrant substances from stabilized leachate, and require a post-treatment for refining the biologically pre-treated leachate to bring its quality in accordance with the international standards (Yasar *et al.*, 2006; Li *et al.*, 2010).

Recently, advanced oxidation processes (AOPs) have been widely applied to enhance the biotreatability of wastewaters containing different organic compounds (Martin *et al.*, 2009; Derco *et al.*, 2010; Li *et al.*, 2010). Among the AOPs, Fenton (catalyst plus oxidant) and Photo-Fenton (oxidant plus radiation) processes are more

convenient than others as they are carried out at room temperature (Altin, 2008; Lee & Shoda, 2008; Zhang *et al.*, 2009). Strong OH radicals are formed in the Fenton oxidation process and the oxidation of  $\text{Fe}^{+3}$  to  $\text{Fe}^{+2}$  ions takes place. The  $\text{Fe}^{+2}$  and  $\text{Fe}^{+3}$  ions also act as the coagulants, so Fenton process has a dual function i.e., oxidation and coagulation in treatment process (Badawy & Ghali, 2006). This process has been found successful in increasing both degree and rate of biodegradation (Yasar *et al.*, 2006). Therefore, such processes can be used as a post treatment of biologically pretreated poultry leachate to enhance the degradation process in terms of COD and color removal (Yetilmezsoy & Sakar, 2007).

Since biocalcitrant nature of the poultry litter leachate does not allow the application of biological treatment alone, Fenton and Photo-Fenton processes were applied to a poultry leachate as a post-treatment to accelerate the degradation process. Although AOPs have been used widely for the treatment of wastewater or landfill leachate but very little work has been done for the treatment of poultry leachate by using this technology. The main objective of present study was to optimize the concentration of  $\text{H}_2\text{O}_2$  (an oxidant) and  $\text{FeSO}_4$  (a catalyst) along with UV exposure time for efficient removal of COD, color and hardness from the poultry litter leachate.

## MATERIALS AND METHODS

**Preparation of poultry litter leachate:** Poultry litter in solid form was collected from poultry houses of the Murree hills. A leachate was obtained by adding distilled water in the poultry litter at a ratio of 1:4 (w/v). After thorough mixing, a settling time of about 1 h was given to the sample for maximum dissolution of the solid litter. The mixture was passed through a sieve and the leachate was collected. A locally fabricated unit comprises of two sieves was used. The poultry leachate was stored at 4°C for subsequent studies.

**Fenton and Photo-Fenton experiments:** Aerobically treated sample of poultry litter leachate was used to compare the efficiency of different doses of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$ . The  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  (Merck, Germany) was applied at the concentration of 10, 50 and 100  $\text{mg L}^{-1}$ , while  $\text{H}_2\text{O}_2$  (30% aqueous solution having a density of 1.11  $\text{kg L}^{-1}$ ) was applied at 0.1, 0.5 and 1.0  $\text{mL L}^{-1}$  of leachate. The dose of  $\text{H}_2\text{O}_2$  and the concentration of  $\text{Fe}^{2+}$  are two relevant factors affecting the Fenton process (Bautista *et al.*, 2008; Umar *et al.*, 2010). The  $\text{H}_2\text{O}_2$  dose was correlated with the initial pollutant concentration measured as COD. The above doses of catalyst and oxidant were applied in all possible combinations with each other. In this way, the experiment was performed with a total nine treatments. Low doses of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  were used to reduce the residual effect of these chemicals on the treated water. The Fenton reaction was carried out in the glass beakers (1 L) at room temperature. During the Fenton reaction, pH was maintained at 3.0, because degradation of organic matter is

most effective at pH 3 (Kochany & Lipczynska-Kochany, 2009). First,  $\text{FeSO}_4$  was fed to the aerobically treated leachate. Then, required doses of  $\text{H}_2\text{O}_2$  were fed by a micropipette during the period of 15 min. A reaction time of 30 min was given to each Fenton treatment. All the treatments were replicated thrice.

Then the coagulation experiment was conducted with Jar test method. For this purpose, pH was adjusted to 8.5 by using 1 N NaOH solution. Jar test was carried out by rapid mixing of the Fenton treated effluent at 100 rpm for 5 min and then slow mixing at 40 rpm for 30 min. Then, a settling time of 30 min was provided. After settling, the supernatant was collected and filtered through 0.45  $\mu\text{m}$  filter. This process was repeated for all combinations. The doses of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  exhibiting the highest removal rate for the selected parameters were used in Photo-Fenton process.

A Pyrex glass photo-reactor (with the internal diameter of 5 cm & height of 30 cm) was set up in a batch model for Photo-Fenton experiment. A UV lamp model (PENRAY 3SC9 of UPLAND, USA) with radiation intensity of 5  $\text{mW/cm}^2$  and wavelength of 254 nm was positioned in the center of the reactor (Muhammad *et al.*, 2008). UV radiation time varied with time interval of 10 min. One liter of the leachate was poured in the Pyrex reactor, which was placed on a magnetic stirrer for mixing at 250 rpm. The pH was adjusted to 3. Thereafter, optimal doses of  $\text{FeSO}_4$  (50  $\text{mg L}^{-1}$ ) and  $\text{H}_2\text{O}_2$  (1  $\text{mL L}^{-1}$ ) as determined in the Fenton process were applied to the solution and the UV lamp was switched on immediately. UV exposure time of 10, 20 and 30 min was given. A temperature of  $25 \pm 2^\circ\text{C}$  was maintained throughout the experiment. Treated samples were given a settling time of 2 h. Then, the supernatant was collected and filtered through 0.45  $\mu\text{m}$  membrane filter before analysis.

**Analytical methods:** The filtrates were analyzed for color, COD, hardness, alkalinity, total dissolved solids (TDS), and total solids (TS). All chemicals used were of analytical reagent grade. The change in color of treated leachate was measured at 450 nm by spectrophotometer (Hongve & Akesson, 1996). The COD was determined by closed reflux colorimetric method (APHA, 2005). Absorbance of the wet-digested sample was determined at 600 nm by spectrophotometer. Hardness, TDS and TS of the leachate were determined by following the standard methods (APHA, 2005). Alkalinity was determined by titrimetric method i.e., neutralization of radicals by standardized  $\text{H}_2\text{SO}_4$  (Jenkins & Moore, 1997).

**Statistical analysis:** The data regarding COD and color removal were subjected to standard deviations using the statistical functions in Microsoft® Excel 2007 (xlsx). Data were entered in a Microsoft® Excel 2007 spreadsheet, and means and standard deviations were calculated. ANOVA was performed for the set of data on hardness, TDS and TS. The means were compared using Duncan's multiple range test at  $P < 0.05$ , using the MSTATC software (Michigan State University, USA).

## RESULTS

The COD removal was significantly affected by the application of different treatments of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  (Fig. 1). Application of Fenton treatments improved the COD removal rate in the aerobically pretreated poultry litter leachate, which ranged between 14-35% compared to the aerobically pretreated leachate. Maximum COD removal (35%) was achieved by using 10 mg  $\text{FeSO}_4$  and 0.1 mL  $\text{H}_2\text{O}_2$ . Next three the most effective Fenton treatments were:

(10 mg  $\text{FeSO}_4$ +0.5 mL  $\text{H}_2\text{O}_2$ ) = (100 mg  $\text{FeSO}_4$ +0.1 mL  $\text{H}_2\text{O}_2$ ) > (50 mg  $\text{FeSO}_4$ +1.0 mL  $\text{H}_2\text{O}_2$ ).

At 50 mg  $\text{FeSO}_4$  concentration, an increasing trend in COD removal was observed with the increase in  $\text{H}_2\text{O}_2$  concentration. Fenton oxidation caused 18-50% color removal as compared to pretreated leachate (Fig. 1). The maximum color removal was observed at 50 mg  $\text{FeSO}_4$  plus 1.0 mL  $\text{H}_2\text{O}_2$ . Then, 10 mg  $\text{FeSO}_4$  plus 0.1 mL  $\text{H}_2\text{O}_2$  removed color of the leachate up to 35%.

A random decrease in alkalinity was observed after the application of Fenton treatment (data not shown). Maximum alkalinity removal was achieved, when the concentrations of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  were maximum i.e., 100 mg and 1.0 mL per liter leachate, respectively. Maximum decrease in hardness of the leachate was 67%, which was achieved by using 100 mg  $\text{FeSO}_4$  and 0.5 mL  $\text{H}_2\text{O}_2$  (Table I). However, TS and TDS were increased significantly at all Fenton treatments (Table II). The highest TS (1081 mg  $\text{L}^{-1}$ ) and TDS (3138 mg  $\text{L}^{-1}$ ) were observed at 10 mg  $\text{FeSO}_4$  and 0.5 mL  $\text{H}_2\text{O}_2$ .

Based on the efficiency of the process for removal of COD, color and hardness, Fenton treatment such as 50 mg  $\text{FeSO}_4$ +1.0 mL  $\text{H}_2\text{O}_2$  was chosen as an optimum oxidation post-treatment. It resulted in 31.5% COD and 50% color removal of the poultry litter leachate. It removed alkalinity up to 16%, while hardness removal was 46%.

Photo-Fenton treatment was applied on aerobically pretreated poultry litter leachate by varying the UV exposure time from 10-30 min and by using the selected Fenton treatment (50 mg  $\text{L}^{-1}$ +1.0 mL  $\text{L}^{-1}$ ). The UV exposure enhanced the efficiency of the treatment process.

Fig. 2 represents COD and color removal from the pretreated poultry leachate, when Fenton treated leachate was exposed to UV radiation. The UV exposure enhanced COD removal than that achieved by the Fenton oxidation process. Photo-Fenton treatment removed COD efficiently at 10 min UV exposure, which was 71% compared to a maximum of 37% with Fenton treatment alone. The UV exposure of 20 and 30 min resulted in 64% COD removal, which revealed that with the increase in exposure time of UV, efficiency of the treatment for COD removal decreased. The same trend was observed in case of color removal; however, there was a gradual decrease in the efficiency of the process with the increase in UV exposure time from 10 to 30 min.

**Table I: Effect of Fenton treatment on hardness of the aerobically pretreated poultry litter leachate**

Treatment ( $\text{FeSO}_4$ mg $\text{L}^{-1}$ + $\text{H}_2\text{O}_2$ mL $\text{L}^{-1}$ )	Hardness ( $\text{CaCO}_3/\text{mg}$ ) <sup>a</sup>	
	Before	After
10 + 0.1	400 b <sup>**</sup>	373 c
10 + 0.5	400 b	240 e
10 + 1.0	300 d	240 e
50 + 0.1	300 d	230 e
50 + 0.5	240 e	190 f
50 + 1.0	240 e	130 h
100 + 0.1	240 e	200 f
100 + 0.5	480 a	160 g
100 + 1.0	480 a	193 f

<sup>a</sup>Data are means of (n= 3)

<sup>\*\*</sup> Mean values sharing same letters do not differ significantly at  $p<0.05$ , according to Duncan's multiple range test

**Table II: Effect of Fenton treatment on total dissolved solids (TDS) and total solids (TS) of poultry litter leachate**

Treatment ( $\text{FeSO}_4$ mg $\text{L}^{-1}$ + $\text{H}_2\text{O}_2$ mL $\text{L}^{-1}$ )	TDS (mg $\text{L}^{-1}$ ) <sup>a</sup>		TS (mg $\text{L}^{-1}$ ) <sup>a</sup>	
	Before	After	Before	After
10 + 0.1	585 e <sup>**</sup>	945 b	2045 b	3100 a
10 + 0.5	585 e	1081 a	2045 b	3138 a
10 + 1.0	574 e	943 bc	1020 i	1465 d
50 + 0.1	574 e	889 cd	1020 i	1360 e
50 + 0.5	610 e	1070 a	745 k	870 j
50 + 1.0	610 e	1040 a	745 k	1185 g
100 + 0.1	610 e	1040 a	745 k	1530 d
100 + 0.5	502 f	877 d	1110 h	1650 c
100 + 1.0	502 f	866 d	1110 h	1280 f

<sup>a</sup>Data are means of (n= 3)

<sup>\*\*</sup> Mean values sharing same letters in a parameter do not differ significantly at  $p<0.05$ , according to Duncan's multiple range test

**Table III: Effect of photo-Fenton treatment on hardness, total dissolved solids (TDS) and total solids (TS) of poultry litter leachate**

UV exposure <sup>a</sup> (min)	Hardness ( $\text{CaCO}_3/\text{mg}$ ) <sup>**</sup>	TDS (mg $\text{L}^{-1}$ ) <sup>**</sup>	TS (mg $\text{L}^{-1}$ ) <sup>**</sup>
0	360 a <sup>***</sup>	526 b	1180 a
10	260 b	615 a	870 c
20	230 c	626 a	970 b
30	230 c	637 a	975 b

<sup>a</sup>UV irradiation was applied to poultry leachate treated with 50 mg  $\text{L}^{-1}$   $\text{FeSO}_4$  + 1.0 mL  $\text{L}^{-1}$   $\text{H}_2\text{O}_2$

<sup>\*\*</sup>Data are means of (n= 3)

<sup>\*\*\*</sup> Mean values sharing same letters in a column do not differ significantly at  $p<0.05$ , according to Duncan's multiple range test

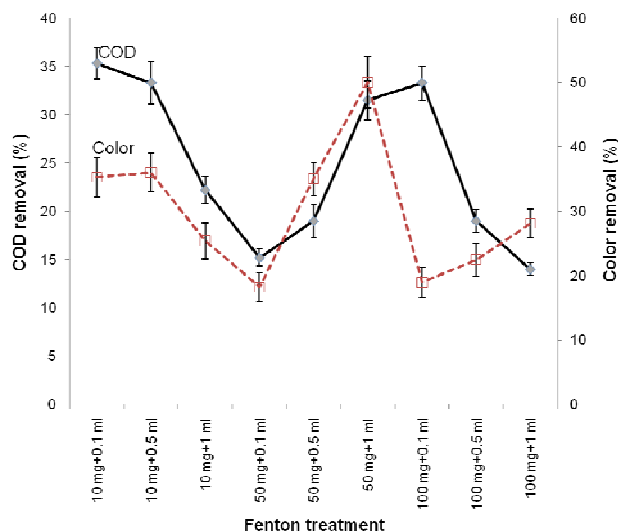
At 10 min of UV exposure, the observed decrease in hardness of the leachate was 28% (Table III). The UV exposure of 30 min further increased the hardness removal by 8% as compared to Fenton treatment alone. At 10 min of UV exposure, an increase in TDS concentration was observed to be 17% (Table III). Further exposure to UV increased the TDS concentration slightly as the further increase of up to 4% was observed at 30 min of exposure. At 10 min UV exposure, 26% TS removal was observed. The UV exposure of 20 and 30 min decreased the TS removal by 9%.

## DISCUSSION

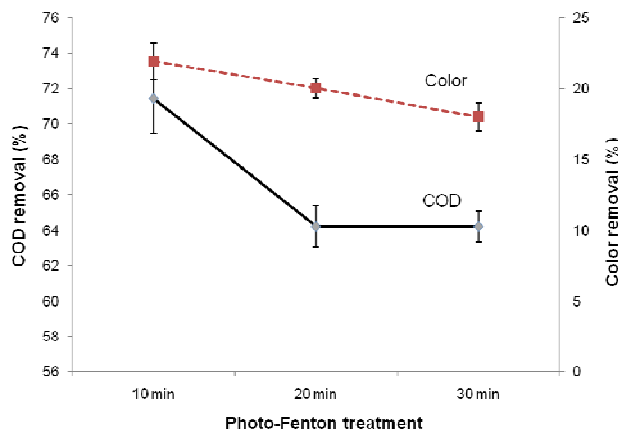
This study reports the comparative effectiveness of different doses of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  as a Fenton treatment as well as Photo-Fenton treatment for the removal of COD, color and hardness from poultry litter leachate. The highest rate of COD removal (up to 35%) was achieved, when 10 mg of  $\text{FeSO}_4$  was used in combination with 0.1 mL  $\text{H}_2\text{O}_2$ . However, Hermosilla *et al.* (2009) reported 80% removal of COD from young leachate after Fenton treatment. In general, the efficiency of Fenton oxidation for COD removal varied between 14–35%. The COD removal varied in different Fenton treatments most likely due to difference in proportion of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$ . Rate of COD removal showed a link with the concentration of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  and an increase in  $\text{H}_2\text{O}_2$  concentration affected the rate of COD removal significantly. When the amount of  $\text{FeSO}_4$  was constant at 10 mg  $\text{L}^{-1}$  leachate, the tendency of COD removal decreased with the increase in concentration of  $\text{H}_2\text{O}_2$  from 0.1 mL to 1.0 mL. This could be attributed to a low concentration of  $\text{FeSO}_4$  that may not be enough to accelerate the oxidation and the rate of COD removal decreased due to some un-reacted  $\text{H}_2\text{O}_2$  (Gulkaya *et al.*, 2006). Derco *et al.* (2010) observed more effective COD removal (57%) in leachate where the higher amounts of  $\text{Fe}^{2+}$  catalyst were used. By keeping the concentration of  $\text{FeSO}_4$  constant at 50 mg, an increase in  $\text{H}_2\text{O}_2$  amount showed an increase in the COD removal. Similar to our findings, Sun *et al.* (2009) found an increase in the COD removal efficiency from 20.3 to 53.0% as the dosage of  $\text{H}_2\text{O}_2$  increased from 10 to 80 mmol. Availability of an appropriate concentration of  $\text{Fe}^{2+}$  in the presence of higher concentration of  $\text{H}_2\text{O}_2$  enhances oxidation process (Yetilmezsoy & Sakar, 2007; Aris *et al.*, 2008). An improved COD removal was observed by Li *et al.* (2010) when  $\text{Fe}^{2+}$  concentration increased. However, an increase in  $\text{Fe}^{2+}$  concentration beyond the optimum region resulted in decreasing the COD removal. This is mainly due to the fact that the excess of  $\text{Fe}^{2+}$  could negatively affect the coagulation–flocculation process and scavenge hydroxyl radicals generated through the reaction of Fenton's reagents (Li *et al.*, 2010).

Compared with COD removal, the Fenton process was more effective in color removal (up to 50%) for treating biologically pretreated poultry leachate. The color removal efficiency also enhanced with the increase in concentration of  $\text{H}_2\text{O}_2$  from 0.1 to 1 mL  $\text{L}^{-1}$  leachate. Fenton oxidation process considerably reduced the alkalinity for pretreated poultry leachate and the maximum alkalinity removal was achieved at high concentrations of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$ . Maximum alkalinity removal was just 28%, because of the reason that the sufficient availability of  $\text{FeSO}_4$  is necessary to produce hydroxyl radicals and maximum degradation can only be achieved if it will be coupled with proper concentration of  $\text{H}_2\text{O}_2$  (Aris *et al.*, 2008). Negative and random results were obtained for TS and TDS after the application of Fenton treatment to aerobically treated

**Fig. 1: Effect of different Fenton treatments on COD and color removal of poultry litter leachate (Data are means of n= 3)**



**Fig. 2: Effect of photo-Fenton treatments on COD and color removal of poultry litter leachate (data are means of n= 3)**



poultry litter leachate. Negative removal for TS can also be attributed to the fact that there may be unutilized quantity of iron left in the treated effluent (Gogate & Pandit, 2004). Fairly positive results were observed in case of hardness removal after application of Fenton process. Maximum hardness removal was 66%, achieved by using 50 mg  $\text{FeSO}_4$  and 1.0 mL  $\text{H}_2\text{O}_2$ . No supporting evidence was found for hardness removal but the possible reason can be that concentrations of both chemicals were optimum to carryout sufficient degradation.

Photo-Fenton treatment further improved the efficiency of COD and color removal from aerobically pretreated poultry leachate. Maximum COD removal (71%) was achieved at 10 min UV exposure time. The rate of COD removal became constant i.e., 64% when the UV exposed time increased beyond 10 min. The enhancement of COD

removal rate was due to an increase in hydroxyl radical concentration with increased UV exposure time (Rehman *et al.*, 2006). Primo *et al.* (2008) suggested Photo-Fenton process as an efficient alternative for the treatment of biologically pre-treated landfill leachate and the authors attained 86% COD and total color removal under optimum conditions. After Photo-Fenton treatment only 15, 16 and 17% increase in TDS concentration was observed at 10, 20 and 30 min exposure to UV. Rapid regeneration of iron ions promoted by UV could be responsible for an increase in concentration of dissolved solids as well as for total solids (Hermosilla *et al.*, 2009). In Photo-Fenton process, the hardness removal was also significant. The removal efficiency was increased with the increase in UV exposure time. No supporting results were found in case of hardness; however, UV exposure probably enhanced the degradability of pollutants in leachate.

## CONCLUSION

This study demonstrated that Fenton oxidation process can be used effectively as a post-treatment to enhance the treatment efficiency of poultry litter leachate. In this study low dose (50 mg+1 mL) of  $\text{FeSO}_4$  and  $\text{H}_2\text{O}_2$  efficiently removed COD, color and hardness from the biologically pretreated poultry litter leachate. Efficiency of the process can be further enhanced by exposing the leachate to UV radiation for 10-20 min. However, cost of the process should be considered prior to its large scale application for poultry leachate treatment.

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(Received 28 September 2010; Accepted 27 January 2011)