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The performance of fluidized bed solar photo Fenton oxidation in the removal of COD from hospital wastewaters

A.S. Anjana Anand, S. Adish Kumar*, J. Rajesh Banu, G. Ginni

Department of Civil Engineering, Regional Centre of Anna University, Tirunelveli 627007, India, Tel. +91 7418444297; email: anandanjana64@gmail.com (A.S. Anjana Anand), Tel. +91 9841339016; Fax: 0462 2552877; email: adishk2002@yahoo.co.in (S. Adish Kumar), Tel. +91 9444215544; email: rajeshces@gmail.com (J. Rajesh Banu), Tel. +91 9942491180; email: ginnimuthuvel@gmail.com (G. Ginni)

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ABSTRACT

In this study, fluidized bed solar photo Fenton process was adopted for the treatment of hospital wastewater with an objective of improving the biodegradability and reducing the chemical oxygen demand (COD) of wastewater. The optimal conditions were observed at pH 3, Fe^{2+} dosage of 5 mM, H_2O_2 dosage of 50 mM and silica carrier amount of 40 g/L. Under such conditions, the maximum COD removal was 98% at a hydraulic retention time of 90 min. The enhancement of biodegradability, evaluated in terms of BOD_5/COD ratio increased from 0.16 to 0.7. It was also observed that in fluidized bed solar photo Fenton process, the COD efficiency was 92% whereas in solar photo Fenton process the COD removal efficiency was 67% at 60 min. The effluent COD and total suspended solid concentrations were found to be 30 mg/L, which met the requirements of the discharge standard.

Keywords: Hospital wastewaters; Fluidized bed solar photo Fenton; Biodegradability; COD removal

1. Introduction

According to World Health Organization there are 583 registered hospitals in India. Hospitals consume a significant amount of water in a day, ranging from 400 to 1,200 L/d/bed [1]. Wastewaters are the primary route of entry of pharmaceuticals in the environment and hospitals are considered important sources and significant contributors of pharmaceutical residues in influent municipal wastewater treatment plants [2]. Hospital wastewater contains a significant concentration of chemical oxygen demand (COD) in the range of 500–1,900 mg/L and biochemical oxygen demand (BOD_5) in the range of 400–700 mg/L. The mutagenicity value of hospital wastewater is high due to the presence of highly complex organohalogen compounds resulting from the disinfection of hospital wastewater. This wastewater if discharged directly into urban sewerage system without pretreatment, cause high risk which could have potential negative effect on the biological balance of natural environment [3].

The acute infection and the latent characteristics are very harmful. The magnitude of these pollutant shows that most of these leave the wastewater treatment plant without any degradation. Hospital wastewater has very low value of microbial load due to the high usage of disinfectant. Hence, these bactericides produce a negative impact in the biological process of

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*Corresponding author.

wastewater treatment plants. Even if this wastewater is diluted after discharge from wastewater treatment plant, the possibility of generating biological imbalance in the aquatic ecosystem is very high. Since some components of hospital wastewater, such as antibiotics, can be environmental problematic even at low concentrations. Release of these chemicals in the environment is of high concern for public health, and may have undesirable health effects on humans, animals and ecosystem. Antibiotics are such materials that can reach the environment via different routes like human or animal excretions, pharmaceutical manufacturing plants effluents, medical wastes, animal fertilizer, municipal wastewater treatment plants and hospital wastewater [4]. Hence, to reduce the toxicity of pollutants and enhance the biodegradability of the wastewater, onsite wastewater treatment is best.

Advanced oxidation process (AOP), an emerging and very promising technology based on the oxidation of hazardous organic compounds in several wastewater [5,6]. AOPs generate hydroxyl radicals ($HO^{\bullet}$) in sufficient quantity to oxidize the complex pollutants. $HO^{\bullet}$ have an oxidation potential of 2.80 eV. The highly reactive $HO^{\bullet}$ can be produced by many routes including heterogeneous photocatalytic, homogeneous photo and non-catalytic processes [7]. Among these methods, Fenton process (homogeneous photo-catalytic) is commonly used due to its low capital cost, easy operation and non-toxic byproducts. Fenton process consists of a catalytic reaction between ferrous ion (Fe^{2+}) and hydrogen peroxide (H_2O_2) usually called "Fenton's reagent". According to simplified mechanism, Fe^{2+} is oxidized to Fe^{3+} and H_2O_2 is reduced to hydroxide anions and hydroxyl radicals explained by Eq. (1).

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

$$Fe^{3+} + H_2O_2 \rightarrow Fe\text{–}OOH^{2+} + H^+ \quad (2)$$

$$Fe - OOH^{2+} \rightarrow Fe^{2+} + HO_2^{\bullet} \quad (3)$$

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

$$Fe^{2+} + HO^{\bullet} \rightarrow OH^- + Fe^{3+} \quad (5)$$

$$H_2O_2 + HO^{\bullet} \rightarrow H_2O + HO_2^{\bullet} \quad (6)$$

From Eqs. (2) and (3), we can see that, Fe^{3+} gets reduced back to Fe^{2+} by H_2O_2 [8]. From all the above equations, it can be seen that proper composition of Fenton's reagent is required to optimize the process. If the concentration of Fenton's reagent is too high, it quenches the $HO^{\bullet}$, as shown in Eqs. (5) and (6). The $HO^{\bullet}$ produced during the reaction combines with the organic contaminant to produce low carbon compounds CO_2 and H_2O as shown in Eq. (7).

$$HO^{\bullet} + RH \rightarrow H_2O + R^{\bullet} \overset{\text{Chain propagation}}{\rightarrow\rightarrow} CO_2 + H_2O \quad (7)$$

The main disadvantage of this process is the substantial production of sludge $Fe(OH)_3$ which needs to be separated and disposed. This disadvantage could be overcome with the help of fluidized-bed Fenton process. Here, the carriers induce the crystallization and/or precipitation of iron into their surface. This reduces the puffy sludge production. In fluidized-bed reactor, many types of reaction occurs (a) homogeneous chemical oxidation (H_2O_2/Fe^{2+}), (b) heterogeneous chemical oxidation (H_2O_2/iron oxide), (c) fluidized-bed crystallization and (d) reductive dissolution of iron oxides [9].

The main objective of this work is to study the performance of fluidized bed solar photo Fenton oxidation in the removal of COD from hospital wastewaters. This study aims to enhance the degradation of pollutants from the wastewater in order to meet the minimal national standards for discharge. This work also evaluates the feasibility of enhancing the biodegradability thereby making the wastewater biocompatible and suitable for subsequent biological treatment.

2. Materials and methods

2.1. Wastewater source

Hospital wastewater was obtained from a hospital near Karakonam, Kerala, India. The treatment system consisting of a plain sedimentation tank as primary treatment extended activated sludge process as secondary treatment, sand filter beds and carbon filter as tertiary treatment has a capacity of 500 m^3/d. The wastewater used for the experiment was collected at the outlet of the plain sedimentation tank. The sample was collected continuously for 5 d at regular times due to large variations in concentration. The sample was collected in plastic cans that were transported to laboratory and stored at 4°C.

2.2. Chemicals

All reagents used in this experiment were of analytical grade and used as received without further purification. The chemicals used in this study are ferrous sulphate heptahydrate ($FeSO_4 \cdot 7H_2O$), hydrogen peroxide (H_2O_2 30% w/w), sodium thiosulphate ($Na_2O_3S_2$), sulphuric acid (H_2SO_4), potassium dichromate ($Cr_2K_2O_7$), mercuric sulphate ($HgSO_4$), ferrous ammonium sulphate ($Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$), sodium hydroxide (NaOH) and sodium sulphite (Na_2SO_3) were purchased from Merck (India).

2.3. Fluidized bed solar photo Fenton reactor

The experimental apparatus operated in this study was fluidized bed solar photo Fenton reactor (Fig. 1). The total volume of reactor was 1.5 L. The reactor consists of a cylindrical vessel of 0.053 m diameter and 1.33 m height, with an inlet, outlet and recirculation sections. Silica granules used as carriers had a particle diameter of 0.42–0.59 mm. The suspension and expansion of silica carriers was controlled using recirculation pump with a recirculation rate of 12.5 L/min. Carriers were fluidized by adjusting internal circulation at 50% bed expansion. Batch recirculation mode was used in this study.

2.4. Experimental methods

All experiments were carried out in Regional Centre of Anna University, Tirunelveli campus, India (8˚44´ N 77˚44´ E). The reactor was exposed to solar radiation from January to April (ultraviolet intensity $32 \pm 2\ W/m^2$). Experiments were conducted at 27 ± 3˚C. One litre of wastewater sample was poured into reactor with silica carriers. The recirculation pump was started to suspend the silica carriers (varied in the range of 20–60 g/L) to desired bed expansion level. Initially, the pH of the wastewater was adjusted to using sulphuric acid. The effect of pH was studied by varying the pH from 2 to 5 while maintaining Fe^{2+}, H_2O_2 and silica carriers constant at 4, 40 mM and 60 g/L, respectively. The effect of Fe^{2+} was studied by varying Fe^{2+} from 1 to 6 mM, while maintaining pH, H_2O_2 and silica carriers constant at 3, 40 mM and 60 g/L, respectively. The effect of H_2O_2 was studied by varying H_2O_2 from 10 to 60 mM, while maintaining pH, Fe^{2+} and silica carriers constant at 3, 5 mM and 60 g/L, respectively. The effect of silica carriers was studied by varying the silica carriers from 20 to 60 g/L, while maintaining pH, Fe^{2+} and H_2O_2 constant at 3, 5 mM and 50 mM, respectively. Solar photo Fenton experiments were carried out without silica carriers supplemented in the reactor. Samples were withdrawn from the reactor every 15 min for COD analysis. Immediately after collecting each sample, sodium sulphite solution (approximately 0.5/10 mL wastewater sample) was added to quench the oxidation reaction for H_2O_2 decomposition [10] and the pH was raised by adding sodium hydroxide to precipitate iron salt. The total suspended solids (TSS) of the samples were carried out as per standard methods. Changes in COD were determined by means of the dichromate standard method. Biodegradability was measured by 5 d BOD_5 test according to standard methods (5210 B) by seeding procedure under controlled temperature [11].

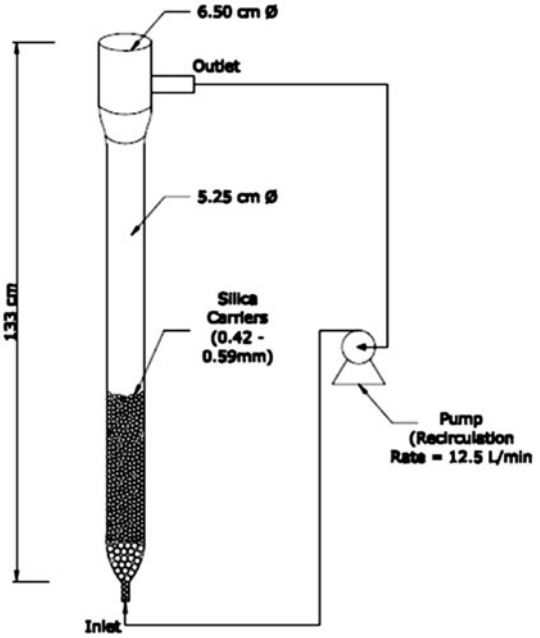

Fig. 1. Schematic diagram of fluidized bed solar photo Fenton reactor.

3. Results and discussion

3.1. Wastewater characterization

The physicochemical characteristics of the primary treated wastewater were determined using standard methods (Table 1). After the primary treatment, the ratio of BOD_5 to COD ratio was 0.16, indicating the non-biodegradable character of the wastewater and the possible presence of minimally biodegradable chemical substances, which decrease the effectiveness of biological treatment. Therefore, upgrade of the existing hospital wastewater treatment system is needed.

3.2. Effect of initial pH

To determine the effect of initial pH, the degradation of hospital wastewater was investigated at pH values ranging from 2 to 5. After 1 h of solar irradiation, the maximum COD removal of 66% was observed at pH 3 (Fig. 2a). This could be due to the formation of dominating species of $Fe(OH)^+$ at acidic pH as shown in Eq. (8). $Fe(OH)^+$ species are reported to have higher activity than the non-complex form of Fe^{2+} in Fenton oxidation [12].

$$Fe^{2+} + HO^- \rightarrow Fe(OH)^+ \tag{8}$$

Above pH 4.0, the degradation efficiency decreases markedly because at this pH, the formation of $HO^{\cdot}$ radicals in Eq. (1) is suppressed according to the Le Chatelier's principle [13]. At pH 2, COD removal efficiency is reduced because H_2O_2 is solvated to form oxonium ion ($H_3O_2^+$). The oxonium ion reduces the reactivity of H_2O_2 with ferrous ion, thereby reducing the concentration of $HO^{\bullet}$ radical. Hence, low COD removal efficiency was observed.

3.3. Effect of Fe^{2+} dosage

To determine the effect of Fe^{2+} dosage, the degradation of hospital wastewater was investigated by varying Fe^{2+} dosage from 1 to 6 mM. Maximum COD removal of 75% was observed for Fe^{2+} dosage of 5 mM (Fig. 2b). When the Fe^{2+} dosage was increased from 1 to 5 mM, the COD removal efficiency increases from 25% to 75% it may be due to increased production of hydroxyl radical [14]. This indicates Fe^{2+} as catalyst can significantly accelerate the decomposition of H_2O_2 to form $HO^{\cdot}$ radicals. Further increase in Fe^{2+} dosage above 5 mM, COD removal efficiency remains constant and plateau. This may be due to the scavenging effect of Fe^{2+} and consequently, the $HO^{\cdot}$ radicals concentration decreased dramatically. The low COD removal at low Fe^{2+} concentration might be due to the side reaction between H_2O_2 and $HO^{\cdot}$ given by Eq. (6) which explains that not much Fe^{2+} react with the H_2O_2. Thus, the amount of $HO^{\cdot}$ reacting with the other organic pollutants in the wastewater was reduced [6].

Table 1
Characteristics of the primary treated hospital wastewater

S. no.	Parameters	Value
1.	pH	6.4 ± 3
2.	TSS (mg/L)	590 ± 30
3.	BOD_3 (mg/L)	200 ± 50
4.	COD (mg/L)	1,200 ± 100
5.	(BOD_3/COD)	0.16

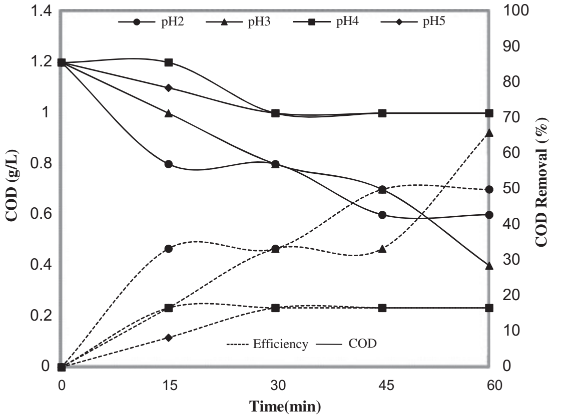

Fig. 2a. Effect of pH on COD removal (Fe^{2+} = 4 mM, H_2O_2 = 40 mM, silica carrier = 60 g/L).

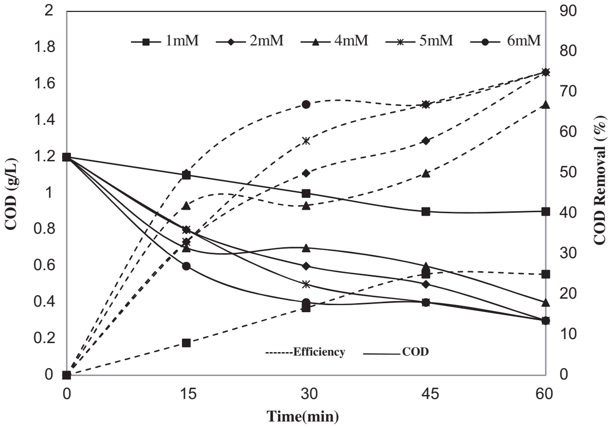

Fig. 2b. Effect of Fe^{2+} on COD removal (pH 3, H_2O_2 = 40 mM, silica carrier = 60 g/L).

3.4. Effect of H_2O_2 concentration

To maintain efficiency, it is necessary to choose the optimum concentration of H_2O_2. The effect of addition of 10–60 mM H_2O_2 on the COD removal is shown in Fig. 2c. From the figure, it can be seen clearly that efficiency was increased from 75 to 83% when H_2O_2 concentration is increased from 10 to 60 mM. Above this critical concentration, the COD removal decreases or remains constant as a result of the scavenging effect given by Eqs. (9)–(11) [15]. Also more H_2O_2 molecules are available for Fe^{2+} ions to react, which increase the number of $HO^{\bullet}$ radicals [16]. The excess H_2O_2 reacts with the hydroxyl radicals earlier formed and hence acts as an inhibiting agent of degradation by

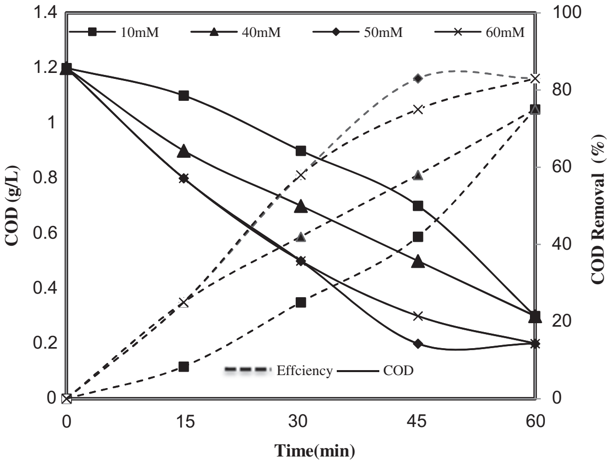

Fig. 2c. Effect of H_2O_2 on COD removal (pH 3, Fe^{2+} = 5 mM, and silica carrier = 60 g/L).

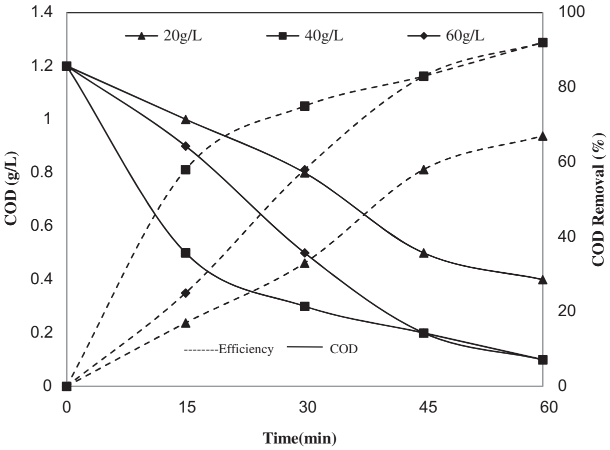

Fig. 2d. Effect of silica carriers on COD removal (pH 3, Fe^2 = 5 mM, H_2O_2 = 50 mM).

consuming the hydroxyl radical. It was also observed that the process was very fast in the beginning of the reaction and it was due to the exhaustion of H_2O_2 [17]. There was no significant difference in the COD removal efficiency of 50 and 60 mM H_2O_2 dosage. Therefore, it was not worth taking of large amounts of H_2O_2 dosage for increasing degradation. Hence, lower dose of 50 mM of H_2O_2 was taken as the optimum dosage in which 83% COD removal were achieved.

$$H_2O_2 + HO^{\bullet} \rightarrow HO_2^{\bullet} + H_2O \qquad (9)$$

$$HO_2^{\bullet} + HO^{\bullet} \rightarrow H_2O + O_2 \qquad (10)$$

$$HO^{\bullet} + HO^{\bullet} \rightarrow H_2O_2 \qquad (11)$$

3.5. Effect of silica carriers

To determine the effect of silica carriers, the degradation of hospital wastewater was investigated by varying silica carrier concentration ranging from 20 to 60 g/L, and the results obtained are shown in Fig. 2d. The precipitation/crystallization of iron oxide onto carriers is a surface phenomenon, hence it depends on the surface availability. It was found that COD removal efficiency increases from 67 to 92% when the silica concentration was increased from 20 to 60 g/L. This is due to the increase in the availability of surface for crystallization. The fluidized bed solar Fenton process causes ferric ions, produced in the Fenton reaction, to be transformed into iron oxide on the surface of carriers by crystallization or sedimentation. This process not only provides a high COD removal efficiency but also reduces the large amount of iron sludge. The iron oxide immobilized onto silica carrier could be used as catalyst for catalytic degradation of hospital wastewater. But the COD removal efficiency remains constant after 40 g/L concentration of silica. This may be due to the settling of excess silica carriers [18]. The results is supported by Diz and Novak [19], they crystallized $Fe(OH)_3$ in fluidized bed reactor using quartz sand as the media.

3.6. Performance of fluidized bed solar photo Fenton process

In order to study the performance of fluidized bed solar photo Fenton process in the removal of COD from hospital wastewaters, reactors were operated with silica carriers (fluidized bed solar photo Fenton) and without silica carriers (solar photo Fenton) at optimum conditions. The results of the experiment are depicted in Fig. 3. It can be observed that in fluidized bed solar photo Fenton process, the COD removal efficiency was 92% whereas in solar photo Fenton process the COD removal efficiency was 67% at 1 h of treatment. In solar photo Fenton process the COD reduction decreases due to the absence of Fe^{2+} ion, whereas in fluidized bed solar photo Fenton process the reaction continues as the carriers enable the availability of Fe^{2+} ion by crystallization. A continuous decrease in COD can be observed for fluidized bed solar photo Fenton process in the whole treatment time unlike the conventional solar photo Fenton process whose COD removal activity almost stopped after 45 min of operation.

3.7. Effect of hydraulic retention time

To determine the effect of hydraulic retention time (HRT), the degradation of hospital wastewater was investigated at optimum condition and the results obtained are shown in Fig. 4. The maximum COD removal efficiency of 98% was observed at 90 min. Table 2 shows the characteristics of fluidized bed solar photo Fenton treated wastewater. On increasing the HRT, it was observed that the BOD_5 and COD of wastewater were reduced drastically. This may be because, on increasing the HRT the wastewater gets exposed to sunlight more and this allows to utilize more energy from sunlight and generates more hydroxyl radicals. From Table 2, it is clear that at 90 min of treatment time, COD was reduced to 30 mg/L and BOD_5 to 20 mg/L which meets the required minimal national standards (MINAS, India 2008) for discharge. The hospital wastewater is non-biodegradable in nature and they contain many recalcitrant compounds. The biodegradability of hospital wastewater was evaluated through the evolution of BOD_5/COD ratio. If BOD_5/COD ratio is higher than 0.6, it indicates a readily and rapidly degradable solution while ratio below 0.6 involves the presence of slowly biodegradable compounds [20]. Result of experiment conducted was as shown in Fig. 4. After the treatment in fluidized bed solar photo Fenton reactor for 60 min the biodegradability was increased to 0.6. The result indicates that this treatment could break down or rearrange molecular structures of highly complex organic matter and convert the non-biodegradable organics to more biodegradable forms. Enhancement in biodegradability suggests that further degradation could be achieved by coupling fluidized bed solar photo Fenton and biological treatment processes, thereby reducing the cost of treatment [21]. First-order kinetic model (Eq. (12)) was utilized for fluidized bed solar photo Fenton and solar photo Fenton processes.

$$\ln C_0/C = kt \tag{12}$$

where C_0, C, t and k are the initial COD, final COD, degradation time (min) and the global reaction apparent rate constant (min^{-1}), respectively. The order of rate constants was fluidized bed solar photo Fenton ($0.043\ \text{min}^{-1}$) > solar photo Fenton process ($0.014\ \text{min}^{-1}$).

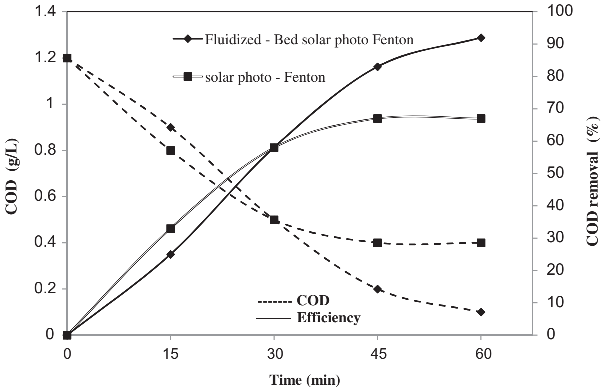

Fig. 3. Performance of fluidized bed solar photo Fenton process (pH 3, Fe^{2+} = 5 mM, H_2O_2 = 50 mM).

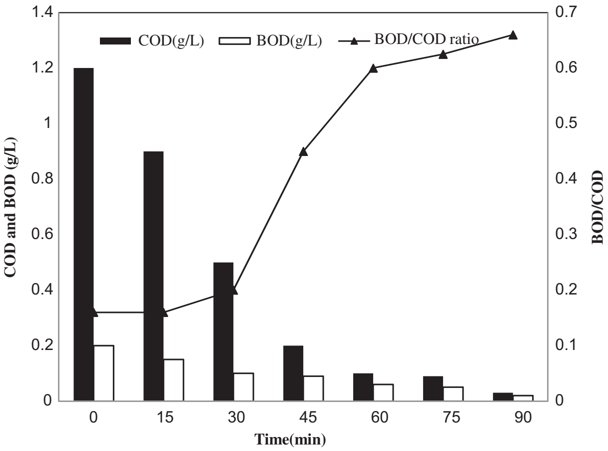

Fig. 4. Effect of HRT (pH 3, Fe^{2+} = 5 mM, H_2O_2 = 50 mM, silica carrier = 40 g/L).

Table 2
Treated characteristics of the hospital wastewater

	Fluidized bed solar photo Fenton treatment process		
Parameters	At 60 min of treatment	At 90 min of treatment	MINAS
pH	7	7	6.5–9.0
TSS (mg/L)	30	30	100
BOD (mg/L)	60	20	30
COD (mg/L)	100	30	250
BOD/COD	0.6	–	–

4. Conclusion

COD removal efficiency for the fluidized bed solar photo Fenton at the optimum conditions of pH 3, Fe^{2+}—5 mM, H_2O_2—50 mM was 98% after 90 min of treatment. Under the optimum conditions, 60 min of treatment enhanced the biodegradability value from 0.16 to 0.6. Thus, there is a possibility of coupling the fluidized bed solar photo Fenton process with biological treatment thereby the cost of treatment can be reduced. The order of rate constants was fluidized bed solar photo Fenton (0.043 min^{-1}) > solar photo Fenton process (0.014 min^{-1}). Use of carriers is very effective in the crystallization of iron and increases the COD removal efficiency from 67 to 92%. Economically, the employment of a natural resource, such as solar light, could be an interesting option due to zero input energy cost as an environmentally harmless photocatalytic treatment of industrial wastewater.

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