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Efficiency of Electrochemical, Fenton and Electro-Fenton Processes on COD and TSS Removal from Leachate

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ABSTRACT

The development of human societies has led to increase in waste generation. On the waste disposal or composting facilities, the chemical and physical alterations produce a contaminated liquid called leachate. In this study, attempts were made to remove the Chemical Oxygen Demand (COD) and Total Suspended Solids (TSS) of the leachate using electrochemical, Fenton and electro-Fenton processes. Hence, we prepared leachate samples from a composting facility in Esfahan, Iran. Then, the effect of electrochemical, Fenton and electro-Fenton processes on COD and TSS removal investigated. The results confirm that the three mentioned methods can degrade the COD and TSS of the leachate. In most cases, the maximum removal rates were observed at first 40 min of the reaction. After that time, the removal process followed by a lower rate and decreased in some cases. Among the studied methods, the electro-Fenton process is the most efficient and is able to remove up to 73 and 92.4% of the COD and TSS, respectively. Also, the Fenton process is more effective than the electrochemical method in COD and TSS removal. Therefore, we suggest using the electro-Fenton method for the COD and TSS removal because of the higher efficiency and environmental advantages.

Key words: COD, TSS, electrochemical, Fenton, electro-Fenton

INTRODUCTION

In the last one hundred years, due to growth of population, development of urban and progresses of industry and technology, the urban and industrial waste increased considerably. The reports demonstrated that the rate of Municipal Solid Waste (MSW) was 1.3 billion t day^{-1} (666 g per person per day) in 1994. However, in 2008, the rate of MSW increased to 1.7 billion tonne day^{-1} (Foo and Hameed, 2009). The organization of recycling and conversion materials of Tehran municipality reported that Iran is the tenth largest producer of waste in the world (Yazdandad and Sadegh, 2011). The waste exposed to chemical, physical and biological factors in landfill sites or composting facilities and produce leachate (Kurniawan *et al*., 2006). This liquid has a dark colour and a foul odor and contains complex chemical and organic compounds (Galeano *et al*., 2011). The high concentration of ammonia nitrogen, heavy metals, inorganic salts and chlorinated organic and xenobiotic compounds reveal the severity of the contamination of the leachate (Gao *et al*., 2015; Ricordel and Djelal, 2014). As, the leachate is hazardous for surface water, groundwater and soils, we should be careful in control, collection, treatment and disposal (Moreira *et al*., 2015). Due to a variety of pollutants in the leachate, the treatment is a major concern in urban waste management (Shafieiyoun *et al*., 2012). Hence, leachate should be refined

Table 1: Experimental results of some previous studies on leachate treatment using electrochemical, Fenton and electro-Fenton processes

Methods	Optimal conditions	Initial COD (mg L^{-1})	COD removal (%)	References
Electrochemical	Current density: 348 A m^{-2}, Reaction time: 30 min, Iron electrodes	12860	33.0	Ilhan *et al*. (2008)
Electrochemical	Current density: 79.9 mA cm^{-2}, Reaction time: 4 h, Graphite Carbon electrodes	1414	68.0	Bashir *et al*. (2009)
Fenton	pH: 2.5, $[H_2O_2]/[Fe^{2+}]$: 3	8298-8894	89.0	Zhang *et al*. (2006a)
Fenton	pH: 3, $[H_2O_2]/[Fe^{2+}]$: 3,	1100-1300	61.0	Deng (2007)
Fenton	pH: 3, $[H_2O_2]/[Fe^{2+}]$: 3, Reaction time: 40 min	743	60.9	Cortez *et al*. (2010)
Electro-Fenton	pH: 3, $[H_2O_2]/[Fe^{2+}]$: 12, I: 2 A, d = 2.1 cm	5000	83.0	Zhang *et al*. (2006b)
Electro-Fenton	pH: 3, $[H_2O_2]$: 2000 mg L^{-1}, I: 3 A, Reaction time: 20 min, Iron electrode	2350	72.0	Zhang *et al*. (2006b)
Electro-Fenton	pH: 3, $[H_2O_2]/[Fe^{2+}]$: 1, Current density: 49 m A /cm^{-2}, Reaction time: 43 min, Aluminum electrode	2950	94.0	Mohajeri *et al*. (2010)

COD: Chemical oxygen demand

to achieve discharge standards, before released on the environment (Mohan and Gandhimathi, 2009). However, there is no comprehensive approach in the treatment, because of the variation of leachate composition in different places (Galeano *et al*., 2011). Some leachate treatment methods include: biological treatment (Mohtashami *et al*., 2008), reverse osmosis process (Rukapan *et al*., 2015), coagulation and flocculation process (Calli *et al*., 2005) and Advanced Oxidation Processes (AOPs) (Deng and Englehardt, 2006; Amr *et al*., 2015). The AOPs convert non-biodegradable pollutants into non-toxic and biodegradable materials (Catalkaya and Kargi, 2007; Salari *et al*., 2009). Then, the biodegradable pollutants converted to carbon dioxide and inorganic materials using the following reaction (Kishimoto *et al*., 2008):

$$AOP_S \rightarrow OH^0 \xrightarrow{\text{Pollutant}} CO_2 + H_2O + \text{Inorganic ions} \quad (1)$$

The AOPs such as Fenton and related processes are based on hydroxyl radical and used for treatment and degradation of pollutants (Poyatos *et al*., 2010). Recently, the AOPs have been used successfully for wastewater treatment of textile (Naumczyk *et al*., 1996), tannery (Rao *et al*., 2001), phenol (Canizares *et al*., 2002), cyanide (Lanza and Bertazzoli, 2002) and leachate (Deng and Englehardt, 2007). Table 1 lists the reaction conditions and Chemical Oxygen Demand (COD) removal of some experimental studies on leachate treatment using electrochemical, Fenton and electro-Fenton processes.

As illustrated in the Table 1, leachate treatment by electrochemical, Fenton and electro-Fenton processes can effectively reduce the concentration of persistent organic pollutants. However, there is no comprehensive comparison between the three mentioned processes on COD and Total Suspended Solids (TSS) removal efficiency. Hence, in this research, the leachate samples were prepared from a composting facility in Esfahan, Iran. Then, the COD and TSS degraded with electrochemical, Fenton and electro-Fenton processes. Furthermore, the efficiency of these processes on COD and TSS removal investigated.

MATERIALS AND METHODS

The studied samples were prepared from a composting facility in Esfahan and transported into the laboratory. The samples were kept at 4°C before testing procedure to prevent changes in the characterization (Rice *et al*., 2012). The typical characterization of the leachate was analysed and listed in Table 2.

Table 2: Typical characteristic of the studied leachate before testing procedure

Parameters	Values
pH	7.5
COD (mg L^{-1})	48500
BOD_5 (mg L^{-1})	14820
BOD_5/COD	0.3
TS (mg L^{-1})	44900
TSS (mg L^{-1})	14900
EC (ms cm^{-1})	4.95
Alkalinity (mg L^{-1} $CaCO_3$)	3400
Turbidity (NTU)	883.33
Sulfate (mg L^{-1})	600

COD: Chemical oxygen demand, BOD: Biological oxygen demand, TS: Total solids, TSS: Total dissolved solids, EC: Electrical conductivity

In the laboratory procedures, the contents of the container were mixed by shaking. Then, 500 mL of the sample was added into a Plexy glass reactor and pH decreased to 3 using sulfuric acid. During the processes, the contents of the reactor were mixed with a magnetic stirrer. Afterwards, the initial COD and TSS were measured. Next, the COD and TSS reduced using electrochemical, Fenton and electro-Fenton processes according to standard methods for the examination of water and wastewater (Rice *et al*., 2012). Finally, the experimental results were analyzed statistically by a two-way ANOVA. It should be noted that, the processes were carried out at 25±5°C. The manual of the Fenton, electrochemical and electro-Fenton processes are expressed as follows:

Fenton process: In the Fenton process, the required $FeSO_4.7H_2O$ was added into the samples. Then, the hydrogen peroxide (30%) added gradually in 1000, 2000 and 3000 mg L^{-1} dosage and sampling was conducted at first 20, 40 and 60 min of the experiment. In COD measurement, the samples were settled for 20 min. Later, we decanted supernatant and diluted. Subsequently, the COD test was performed using the closed reflux method. For removing excess hydrogen peroxide, the pH was increased to 8 with NaOH (Zhang *et al*., 2006a). For the exact determination of TSS, the Total Solids (TS) were measured using 2540B method and, Total Dissolved Solids (TDS) determined according to 254°C method. Finally, the TSS calculated from the following equation:

$$TSS = TS\text{-}TDS \tag{2}$$

Electrochemical process: In the electrochemical process, six iron electrodes with a surface area of 15.5×2.8 cm^2 (surface submerged was 2.8×6 cm), a thickness of 1 mm and 1.5 cm spacing were inserted into the reactor. Then, the direct currents of 0.5, 1 and 1.5 A were applied to the electrodes using ZhaoXin R×N-605D DC Power Supply 60V 5 A apparatus (Fig. 1). Subsequently, the COD and TSS determined at first 20, 40 and 60 min according to the methods mentioned for the Fenton process.

Electro-Fenton process: In this process, the H_2O_2 (30%) added gradually in a concentration of 1000, 2000 and 3000 mg L^{-1} and six iron electrodes similarity to electrochemical process were inserted into the reactor. Then, the direct currents of 0.5, 1 and 1.5 A were applied to the electrodes using the same apparatus as electrochemical process (Fig. 1). Finally, the COD and TSS established at first 20, 40 and 60 min according to the methods as described for the Fenton process.

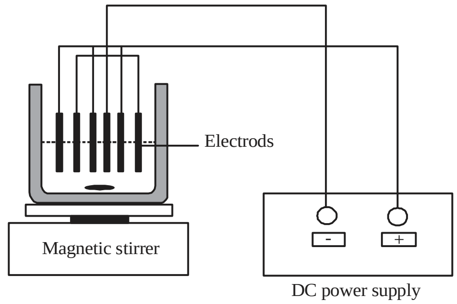

Fig. 1: Schematic of electrochemical and electro-Fenton procedures

RESULTS AND DISCUSSION

Electrochemical process: The COD and TSS removal using an electrochemical process at different electrical current and reaction times is illustrated in Fig. 2. The maximum COD removal of 22.7% was found in the process in electrical current 1 A and in the first 40 min (Fig. 2a). Likewise, the most TSS removal of 41.7% was observed in electrical current 1 A and in the first 60 min (Fig. 2b). Total statistical analysis demonstrated that the increasing electrical current has significant effects on COD and TSS removal percentage. However, increasing the electrical current from 1-1.5 A has no significant effects on the COD and TSS removal with 95% confidence interval. Therefore, in the present study, the electrical current 1 A and reaction time 40 min are optimum conditions in both COD and TSS removal in the process. Nevertheless, TSS removal percentage in the first 60 min (41.7%) is higher than in the 40 min (35.4%) in electrical current 1 A, nonetheless, this change has no essential effect on the test results. It should be noted that, Ilhan *et al.* (2008) removed 33% of the COD by electrochemical process at reaction time 30 min and current density 348 A m^{-2} whereas the initial COD was 12860 mg L^{-1}.

Fenton process: The efficiency of Fenton process on COD and TSS removal percentage is shown in Fig. 3a-b. In this process, both COD and TSS removal percentage are increased by increasing the hydrogen peroxide dosage. We observed maximum COD removal of 38% in hydrogen peroxide dosage of 3000 mg L^{-1} and reaction time 60 min. Moreover, the highest TSS removal of 60.3% was determined in hydrogen peroxide dosage of 3000 mg L^{-1} and reaction time 40 min. The statistical analysis confirmed that increasing the hydrogen peroxide dosage from 1000-2000 mg L^{-1} and 2000-3000 mg L^{-1} and reaction time from 20-40 min have significant effects on both COD and TSS removal percentage with 95% confidence interval. Similarity, the optimum $[H_2O_2]/[Fe^{2+}]$ molar ratio 3 was observed for COD removal using the Fenton process in Zhang *et al.* (2006a), Deng (2007) and Cortez *et al.* (2010) studies. It should be noted that in the present research, hydrogen peroxide dosage of 1000, 2000 and 3000 mg L^{-1} is equal to $[H_2O_2]/[Fe^{2+}]$ molar ratio of 1, 2 and 3, respectively. Hence, hydrogen peroxide dosage of 3000 mg L^{-1} and reaction time of 40 min is the most favorable conditions for COD and TSS removal using the Fenton process.

Electro-Fenton process: In the electro-Fenton process, both hydrogen peroxide and electrical current are used in COD and TSS removal, simultaneously. The effect of electro-Fenton process on COD and TSS removal is shown in Fig. 4. In general, increasing hydrogen peroxide dosage, electrical current and reaction time have led to increase in both COD and TSS removal percentage. According to Fig. 4a, the most COD removal of 72.9% was observed in hydrogen peroxide dosage

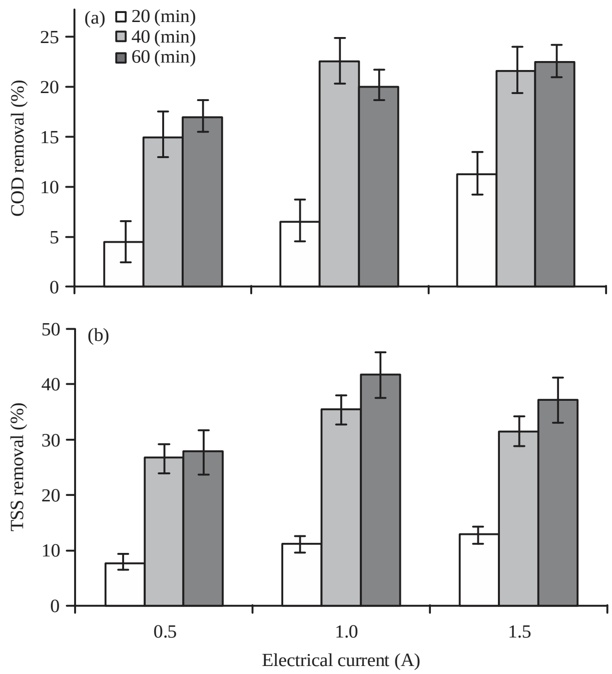

Fig. 2(a-b): (a) COD and (b) TSS removal percentage using electrochemical process at different electrical current and reaction time

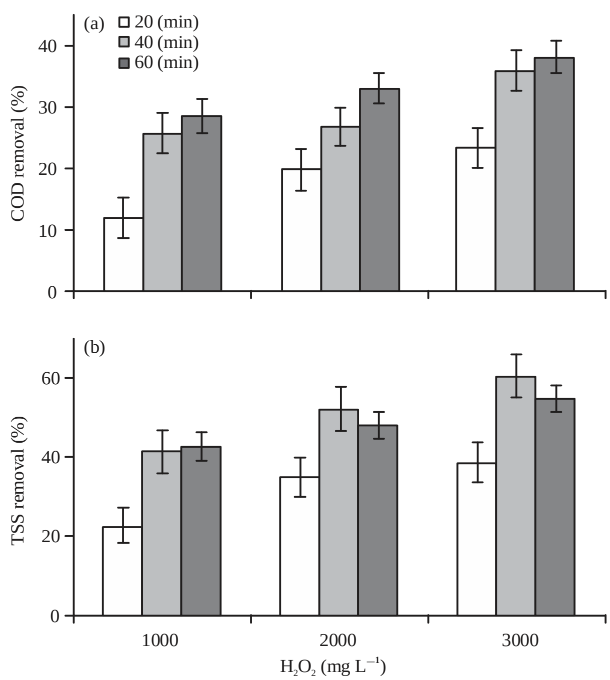

Fig. 3(a-b): (a) COD and (b) TSS removal percentage using Fenton process at different hydrogen peroxide dosage and reaction time

1000 mg L^{-1}, electrical current 1 A and 40 min reaction time. Moreover, the maximum TSS removal of 92.4% was found in hydrogen peroxide dosage 2000 mg L^{-1}, electrical current 1.5 A and 40 min

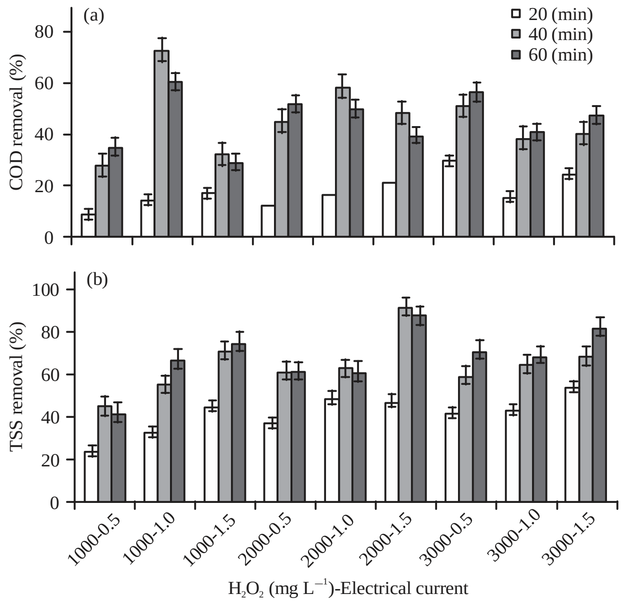

Fig. 4(a-b): (a) COD and (b) TSS removal percentage using electro-Fenton process at different electrical current, hydrogen peroxide dosage and reaction time

reaction time (Fig. 4b). The statistical analysis revealed that increasing hydrogen peroxide dosage from 1000 mg L^{-1} and electrical current from 1 A has no essential effect on the COD removal. Moreover, the optimum conditions for TSS removal were observed in hydrogen peroxide dosage of 2000 mg L^{-1} and electrical current 1.5 A. Likewise, Zhang *et al.* (2006b), Atmaca (2009) and Mohajeri *et al.* (2010) removed 83, 72 and 94% of the leachate COD, respectively. The most favorable conditions of these studies are listed in Table 1.

The experimental studies and comparisons revealed that the electro-Fenton process is more ideal process in COD and TSS removal than the electrochemical and Fenton processes. The statistical analysis revealed that increasing the reaction time from 40-60 min has no significant effect on the electrochemical, Fenton and electro-Fenton results with 95% confidence interval. Also, we found the ideal reaction time 40 min for all processes. The pH is another essential factor affecting the efficiency of the Fenton and electro-Fenton processes. Due to growth of reaction rate or regeneration of hydrogen peroxide, production of hydroxyl radicals is high in the pH range of 2-4 (Sedlak and Andren, 1991). Hence, pH 3 is ideal for Fenton and electro-Fenton related processes. Likewise, amounts of hydrogen peroxide and Fe^{2+} are very important for COD and TSS removal using Fenton and electro-Fenton processes (Umar *et al.*, 2010). Excess hydrogen peroxide decomposed and could products iron sludge floatation or declined sludge sedimentation because of the O_2 released (Kim *et al.*, 2001). Moreover, excess Fe^{2+} results extra sludge and increase the TDS and electrical conductivity (Gogate and Pandit, 2004) . Hence, we suggest hydrogen peroxide dosage of 3000 mg L^{-1} or $[H_2O_2]/[Fe^{2+}]$ molar ratio 3 for COD and TSS removal using Fenton process and hydrogen peroxide dosage of 1000 and 2000 mg L^{-1} for COD and TSS removal using electro-Fenton process, respectively. Although, the electrochemical process is less effective than electro-Fenton process for COD and TSS removal, we found that the electrical current 1A is appropriate for the removal. Since, the initial COD at the present study was very high (48500 mg L^{-1}), we suggest that the electrochemical, Fenton and electro-Fenton processes are suitable as pre-treatment.

CONCLUSIONS

In this study, the effect of electrochemical, Fenton and electro-Fenton processes on COD and TSS removal percentage investigated. The results indicate that with an increase in hydrogen peroxide dosage, electrical current and reaction time, the COD and TSS removal efficiency will increase. In most cases, the favorable COD and TSS removal percentage observed at reaction time 40 min. In the electrochemical process, the highest COD and TSS removal observed at electrical current 1 A and reaction time 40 and 60 min, respectively. Additionally, maximum COD and TSS removal percentage using Fenton process obtained at hydrogen peroxide dosage of 3000 mg L^{-1} and reaction time 60 and 40 min, respectively. Moreover, we attained the best COD removal percentage (73%) at hydrogen peroxide dosage of 1000 mg L^{-1} and electrical current 1 A and the most TSS removal (92.4%) at hydrogen peroxide dosage of 2000 mg L^{-1} and electrical current 1.5 A, both in reaction time 40 min and using electro-Fenton process. Although, Fenton process has higher efficiency than electrochemical process, however, is limited to use of chemical materials and production of iron (III) sludge. Hence, an alternative method as electro-Fenton is suggested. The combination of Fenton and electrochemical processes has led to increase the oxidation capacity within the system. Therefore, the electro-Fenton has low cost and high potential for decomposition and removal of organic pollutants into harmless compounds as CO_2, water and inorganic salts.

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