

Evaluation of Electro/Fenton Process Using Iron Electrode on Phenol Removal from Aqueous Solution

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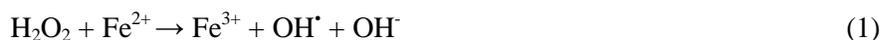
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Abstract: *This study examines phenol removal from aqueous solutions through electro-Fenton (EF) process using iron electrode. The effect of operational parameters such as initial pH, current density, initial concentration of phenol, hydrogen peroxide dosage on the removal of phenol was investigated. The results showed that the efficiency of phenol removal had a direct relationship with initial concentration of hydrogen peroxide, and was inversely correlated with the highly alkaline pH and elevated concentration of phenol. The removal efficiency of phenol was significantly increased with increasing H₂O₂ concentration from 0.1 mM to 0.4 mM, but there was little influence on the removal efficiency in greater quantities of H₂O₂. Ultimately, phenol was almost completely removed after 45 min in this process.*

Keywords: *Electro Fenton, Phenol removal, Oxidation process, Iron electrode.*

1. Introduction

One of the greatest concerns of the 21st century is water shortage. The shortage of the fresh water affects more than 25% of the world population. Consequently, 2.2 million people die every year as reported by the World Health Organization. On the other hand, a large amount of water is contaminated by domestic and industrial activities in developing countries. Therefore, evaluation of waste water quality is essential to avoid further contamination of the environment. The need for reuse of water resources is important in order to reduce fresh water consumption [1]. Overpopulation and the consequent increase of industrial activities generate high concentrations of pollutants leading to aquatic environment contamination. For example, phenol and its derivatives can be found in waste disposal of various industries including resins, plastics, paper-making and coal conversion factories. Although the toxicity and environmental impact of phenolic compounds depend on the number, type and the position of substitution groups of aromatic compounds, these chemicals is found to be toxic for various organisms, including humans, animals and plants. Hence, the removal of phenolic compounds is regarded as one of the greatest concerns in the world [3, 2]. There are several methods of treating wastewater containing phenol and the most important of these are advanced oxidation, chemical oxidation, adsorption, biological, filtration and a combination of these procedures [4]. High cost, long retention time and generation of toxic by-products are drawbacks to the widespread use of some of these elimination strategies [5, 6]. Advanced oxidation processes have attracted much attention today due to their ease of use, being economical and high efficiency [7-9]. Among the advanced oxidation processes, Electro/Fenton process can be noted. Fenton process is a process which Fe²⁺ ions and hydrogen peroxide are concurrently used to decompose and eliminate pollutants. This process is possible in the presence of Fe²⁺ ions, hydrogen peroxide and radical hydroxyl for oxidation to take place. H₂O₂ can be activated by Fe²⁺ to produce hydroxyl radical via Eq. (1).



If in the Fenton process, the Fe^{2+} ions produce by electrolytic method using iron electrode, it is called electrofenton (EF) process. This process has attracted much attention due to its characteristics such as low cost, easy operation, and high efficiency [10]. It has been proven that various types of organic compounds can be degraded by the Fenton process without producing any toxic substances in the aquatic environment. The whole process is very efficient and cost-effective compared to conventional chemical methods [11, 12]. If Fe^{2+} is produced by electrochemical method, its concentration will be low level and quenching of radicals does not occur. In the current study removal of phenol using electrofenton process was examined. The effect of different parameters was also evaluated in this process.

2. Materials and Methods

2.1. Materials

All chemicals, such as peroxide hydrogen (H_2O_2), phenol with 99.5% purity ($\text{C}_6\text{H}_5\text{OH}$), sulfuric acid (H_2SO_4), were purchased from Merck Company. Phenol absorbance measurement was accomplished by using UV-Vis Spectrophotometer and pH was measured using a pH meter manufactured by HACH Company.

2.2. Methods

A known amount of the phenol solution (100 mgL^{-1}) was poured to the reactor. Distillated water was used to prepare solutions. Adjusting of solution pH was performed using sulfuric acid (1 N) and NaOH (1N). After regulating of the voltage, hydrogen peroxide dosage, the iron electrodes were placed into the reactor and the run was done. Sampling was carried out at different times and then the samples were centrifuged (6000 RPM for 10 min) and absorption amounts of the remaining phenol in the samples were measured through direct photometry by using UV-Vis spectrophotometer at a wavelength of 500 nm [13,14]. The removal efficiency of phenol was calculated as follows [15].

2.3. Bench Scale Reactor

This experimental study was conducted using a batch processing reactor in water and wastewater laboratory in Hamadan University of Medical Sciences. The intended bench scale reactor whose schematic representation is given in Fig. 1 was made of a glass beaker with the volume of 1000 ml. Four iron electrodes each 200 mm length, 20mm width and 2 mm thickness at a distance of 1 cm are floating in the reactor. The electrodes are connected to a DC power supply so that cathode and anode were placed alternately. A magnetic stirrer is used in order to create a uniform mixing of the solution.

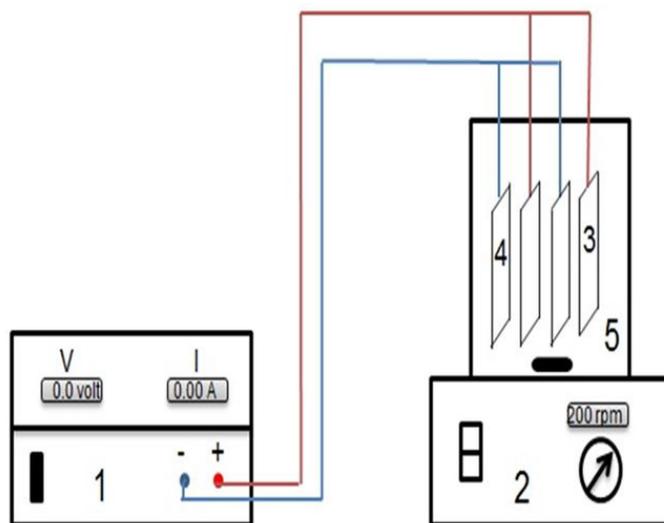


Fig. 1. Experimental Setup for EF Process: 1. DC power supply, 2. Stirrer 3. Anode, 4. Cathode, 5. Electrochemical cell.

3. Results and Discussion

3.1. Effect of pH

Fenton process is heavily dependent on pH solution [16]. In this study, the effect of pH changes in the range of 3-10 in EF process was studied and the results are shown in Fig. 2. Results indicate that the pH changes are fairly significant in phenol removal, so that the highest removal efficiency in pH = 3 was 93.99 %. pH has a direct effect on the stability of H_2O_2 , the amount of producing $\text{OH}\cdot$, and finally determines the type and state of iron in the solution [17-19]. Studies have shown the oxidation potential of $\text{OH}\cdot$ increases as pH decreases [12, 20]. As mentioned the maximum percentage of phenol removal occurred at pH= 3 because of in acidic pH the surface of iron over time would be refreshed so production of free electron can be faster than other pH [21]. Also the lowest phenol removal efficiency was observed at pH= 10. In alkaline pH Fe^{2+} precipitate to the form of $\text{Fe}(\text{OH})_2$ and leaves the catalytic cycle.

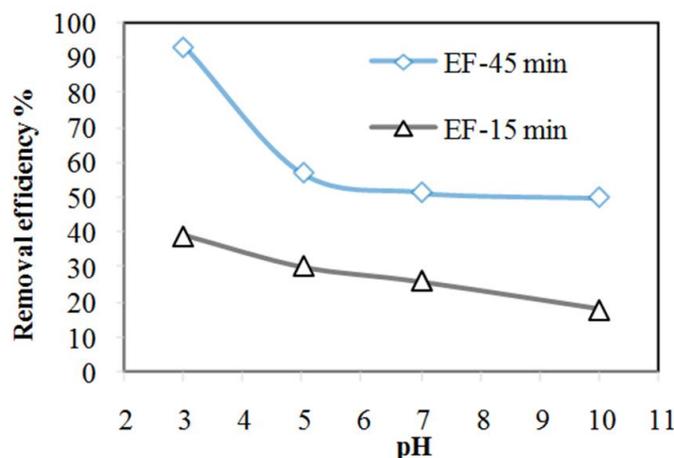


Fig. 2. Effect of pH on removal of phenol in EF process ($[\text{phenol}]_0 = 100 \text{ mg.l}^{-1}$, $[\text{H}_2\text{O}_2]_0 = 0.4 \text{ mM}$ and $\text{CD} = 0.17 \text{ mA cm}^{-2}$)

3.2. Effect of Current Density

In all electro-chemical processes, current density (CD) is the main parameter to control the reaction rate [22]. According to Faraday's law, intensifying the applied current, increases producing metal ions on a sacrificial anode surface [22]. In most electrochemical studies a constant current density was used or potential difference was fixed [23]. To control the effect of iron concentration on the process, constant current density enters the electrode that iron can be produced at a constant rate by sacrificial anode and the system can be steered under controlled condition [24]. Increasing current density results a rapid ferrous generation and ferric reproduction so phenol removal efficiency soars [12, 25]. When the current density in the process increases from 0.12 to 0.17 mA.cm^{-2} phenol degradation increases from 47.54 to 93.18%. This would in turn boost the efficiency of phenol removal, so that the maximum phenol removal efficiency is observed when the current density is 0.17 mA.cm^{-2} . As a result, according to Fig. 3 in the current density of 0.24 mA.cm^{-2} phenol removal efficiency has declined.

3.3. Effect of Hydrogen Peroxide Dosage

In order to determine the optimal dosage of hydrogen peroxide, different H_2O_2 values in the range of 0.1-0.5 mM was added to each prepared sample and the pH was set to 3, and then the experiment was performed. The amount of hydrogen peroxide is the main factor affecting operation cost and process efficiency [6]. According to Fig. 4 phenol removal efficiency in this process will increase with increasing hydrogen peroxide concentration from 0.1 to 0.4 mM, from 49.71 % to 93.18%. It was noted that H_2O_2 is the only source of $\text{OH}\cdot$ in the process. Increasing the amount of H_2O_2 to a certain extent increases $\text{OH}\cdot$ concentration and will increase the process efficiency [26]. By increasing H_2O_2 concentration as much as 0.5 mM, an opposite trend was observed in the removal of phenol. When hydrogen peroxide concentration escalates, it is converted into oxygen and water and hydroxyl radicals are combined together, which is followed by a decline in the process efficiency.

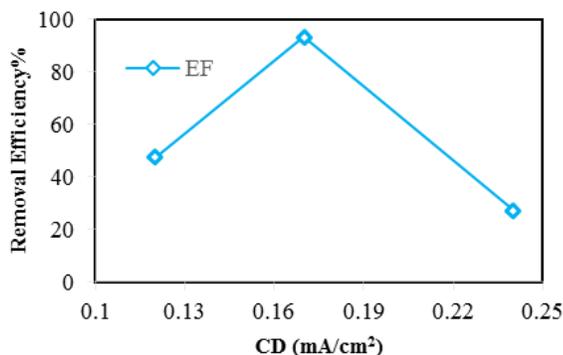


Fig. 3. Effect of different CD current densities on phenol removal efficiency ($[\text{phenol}]_0 = 100 \text{ mg.l}^{-1}$, $[\text{H}_2\text{O}_2]_0 = 0.4 \text{ mM}$ and $\text{pH} = 3$)

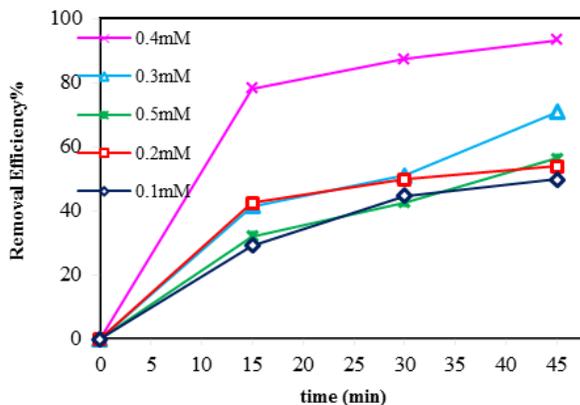


Fig. 4. Effect of different hydrogen peroxide concentrations in EF process ($[\text{phenol}]_0 = 100 \text{ mg.l}^{-1}$, $\text{pH} = 3$ and $\text{CD} = 0.17 \text{ mA.cm}^{-2}$)

3.4. Effect of Phenol Concentration

Effect of phenol initial concentration in the process was examined in the range of 50-200 mg.l^{-1} that the results illustrated in Fig. 5. The figure shows that with increasing of phenol concentration from 50 to 200 mg.l^{-1} , removal efficiency decreases from 93.18 to 15% after 45 minutes. It is clear that increasing the initial concentration of pollutants, needs more oxidant so in the constant amount of oxidant the efficiency of the process would be decreased. The intermediate compounds generated during the reaction would be rise in high initial concentration which consume hydroxyl radicals. This may lead to competition between pollutant molecules and intermediate in reacting with hydroxyl radicals. Thus decomposition rate drops in high initial concentrations [26].

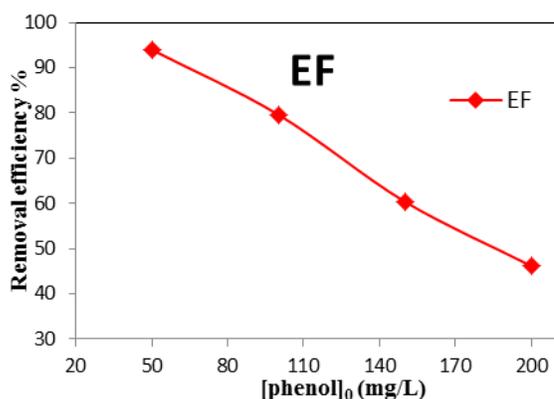


Fig. 5. Effect of phenol initial concentrations on EF process in optimal conditions efficiency ($[\text{phenol}]_0 = 100 \text{ mg.l}^{-1}$, $[\text{H}_2\text{O}_2]_0 = 0.4 \text{ mM}$ and $\text{pH} = 3$)

4. Conclusion

The present study investigates EF process using iron electrode. In the optimum condition removal efficiency of this process was obtained 93.18%. The results showed that phenol removal efficiency has a direct relationship with an increase in contact time and current density and had an inverse relationship with phenol initial concentration. Phenol removal efficiency is reduced when hydrogen peroxide concentration is much more than the optimum level. To reach the maximum efficiency, electrolyte and electrical current density must be present.

5. Acknowledgment

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6. References

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