



## Optimization and modeling of electro-Fenton process for treatment of phenolic wastewater using nickel and sacrificial stainless steel anodes



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### ABSTRACT

Sacrificial stainless steel and nickel electrodes were used as anodes in electro-Fenton (EF) process for degradation of phenol in controlled operating conditions. The influence of different operating parameters such as initial concentration of phenol, current intensity,  $\text{Fe}^{2+}$  dose, pH and anode material were studied. At optimal operating conditions, the degradation of phenol was 95.2% using sacrificial stainless steel anode in 90 min and 72% using nickel anode in 120 min. The favored current intensity was 900 mA. The optimal  $\text{Fe}^{2+}$  doses using stainless steel and nickel anodes were 5 mg/L and 20 mg/L respectively. The production and consumption of iron during the EF process was monitored to interpret the favorability of sacrificial stainless steel anode. Regression analysis was employed to develop a prediction model for phenol removal. The model is dependent on operational conditions and initial phenol concentration with coefficient of determination ( $R^2 = 0.9525$ ). In addition, artificial neural network (ANN) model was developed based on input layer of operating conditions and output layer of phenol degradation efficiency. The ANN model yielded a coefficient of determination ( $R^2 = 0.9742$ ) and ( $S_e/S_y = 0.16$ ).

### 1. Introduction

Phenol is one of the major bio-recalcitrant contaminants that exist in many chemical and biochemical industries. It is produced in the operation of Pharmaceutical plants, oil refineries, coke plants, resins industry, olive mill, and other food-processing industries [1,2]. It is preferred to perform degradation processes for phenol and other bio-recalcitrant pollutants from its source before the discharge of industrial wastewater to sewer network or nearby drains because the ability of degradation of bio-recalcitrant organics by conventional biological treatment is limited [3]. Without source treatment, these contaminant might exist in low concentrations in the aquatic environment which threaten the aquatic organisms and may develop the bacteria resistance to antibiotics and disinfectants [4].

Advanced oxidation processes (AOPs) have attracted a lot of attention as promising technologies for the degradation of various hazardous organic compounds such as pesticides, pharmaceuticals and phenolic compounds [5]. AOPs include the generation of very strong reactive species such as hydroxyl radicals which have the ability to break down the stable and resistant chains of organic molecules and degrade them to simpler compounds and benign end products [6]. Among AOPs, Fenton reaction is one of the favored methods for degradation of phenolic compounds because of simplicity and low toxic

residuals [7,8]. In Fenton reaction, powerful reactive hydroxyl radicals are generated from  $\text{H}_2\text{O}_2$  in presence  $\text{Fe}^{2+}$  ions. Fenton reaction could be carried out by different modified processes other than classical Fenton reaction to obtain better degradation efficiency, minimize residuals or save chemicals and energy. Modified processes include photo-Fenton, electro-Fenton, sono-Fenton and Fenton-like process [9–12].

Large production of ferric hydroxide sludge is considered to be one of the main disadvantages of most of Fenton. To overcome this problem the electro-Fenton (EF) process was developed by many researchers to minimize the generation of ferric hydroxide during the reaction [13,14]. In addition, it enhances the degradation efficiency of target compounds [15,16]. In EF, the reduction of ferric hydrogen sludge to form ferrous ions on the cathode is induced by applied current intensity [17]. The negative potential near the cathode in the EF cell reduce the dissolved oxygen to form  $\text{H}_2\text{O}_2$  in aqueous solutions. The addition of  $\text{Fe}^{2+}$  salts leads to start a Fenton reaction with the electrically produced  $\text{H}_2\text{O}_2$  and ferric ions are continually formed. During the process, Ferric ions are reduced back to ferrous ions near the cathode which enhance the oxidation of contaminants and reduce the amount of sludge [18]. The major advantages of this method is the in situ production of  $\text{H}_2\text{O}_2$  which minimize the transport and storage. Besides, it utilizes a clean energy source to improve the performance of contamination removal

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[19].

Several parameters could influence the of EF process such as initial contaminant concentration, pH, current intensity and reagent dosage. The obtained experimental results could be analyzed by linear and nonlinear regression to describe the correlation of these parameters to the process efficiency [20]. Artificial neural network (ANN) is another method to describe the relation between the operating parameters and the process performance [21]. The advantage of ANN is linking the effect of all inputs to each other and to the outputs in three steps including training, validation, and test [22].

The aim of this study is to investigate the performance of EF process for the degradation of phenol as a recalcitrant organic pollutant. Operational conditions such as initial phenol concentration, current intensity,  $\text{Fe}^{2+}$  dose, pH, and anode material were studied. Moreover, simple predictive models are developed to describe phenol removal at different conditions, using statistical regression analysis and the artificial neural networks (ANNs).

## 2. Materials and method

### 2.1. Materials

Methanol, phenol ( $\text{C}_6\text{H}_5\text{OH}$ , 99.5% pure), Ferrous sulfate heptahydrate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ , 99.75% pure), hydrochloric acid (HCl 98.9% pure), and sodium hydroxide (98% pure) were purchased from Merck. Sodium sulfate (99.5% pure) and all other reagents were obtained from Sigma-Aldrich. All chemicals were used without further purification. The solutions were prepared by distilled water with different concentrations of chemicals. Stock solution of phenol (5 g/L) was prepared to obtain the required solutions with different initial concentrations. Hydrochloric acid was used for pH adjustment.

### 2.2. Experimental procedure

Electro-Fenton reaction was carried out in 1000 mL cylindrical beaker. The reactor was equipped with two electrodes totally immersed into the solution and filled with 600 mL phenol solution of certain concentration. A piece of carbon felt was used as a cathode ( $30\text{cm} \times 13.5\text{cm} \times 0.5\text{cm}$ ) and fixed on the inner wall of the cylindrical glass. Two materials were tested as an anode including sacrificial stainless steel grade 304 (Plate  $5\text{cm} \times 5\text{cm} \times 0.5\text{cm}$ ), and nickel (Plate  $4.2\text{cm} \times 4.2\text{cm} \times 0.5\text{cm}$ ). The anode was placed in the center of the cylindrical glass. A magnetic stirrer was used to provide a continuous vigorous stirring of the solution. The solution was aerated for 10 min prior to the reaction and continued to the end of experiment time to increase the dissolved  $\text{O}_2$ . HCl acid with specific quantities was added to obtain the required pH before starting the electro-oxidation reaction. A certain quantity of  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  was added to the solution with the desired concentration.  $\text{Na}_2\text{SO}_4$  was added to the solution with concentration of 1000 mg/L to maintain the ionic strength constant and improve the conductivity of the medium. A DC power supply was connected to the electrodes and worked with a constant current. An ammeter (PHYWE-07036-00) was used to measure the current intensity. The voltage difference was measured by a voltmeter (Digital Voltmeter (G-1002-500)). An initial Sample (5 mL of phenol) was taken before the pH adjustment. Periodic samples (5 mL of phenol solution) were taken every 10 min and filtered using ( $0.2\ \mu\text{m}$ ) filters to remove any suspended materials before analysis.

### 2.3. Analytical methods

The HPLC system (Agilent 1200 series) was used to measure the phenol concentrations. The system includes a photodiode array detector, and a multi mobile phase pump. An auto-sampler was used to inject samples (25  $\mu\text{L}$ ) into a  $150 \times 4.6\text{mm}$  Apollo C18 column. Peaks were monitored at wavelength ( $\lambda = 270\text{nm}$ ) under these conditions:

column temperature,  $40^\circ\text{C}$ ; mobile phase flow rate,  $0.9\text{mL}\cdot\text{min}^{-1}$ . The composition of the mobile phase was 30% distilled water and 70% Methanol. Total organic carbon (TOC) analyzer (Analytik Jena AG Co., Germany) was used to measure the TOC of samples. Remained  $\text{Fe}^{2+}$  was quantified by atomic absorption spectrometer (Varian; AA2280FS)

### 2.4. Modeling of experimental results

The experimental work has been extended to cover different operational conditions for the phenol degradation efficiency by electro-Fenton. For the experiments done by nickel anode, the results were combined in one database. The database was used to develop a model for the prediction of the degradation efficiency. Regression analysis by the least square method was used for model development. The developed model for phenol degradation was used to test the sensitivity of predicted values to each input parameter. In addition, an artificial neural network model was developed based on Tanch axon function to describe the experimental results.

## 3. Results and discussion

The electrochemical degradation of phenol was assessed at different conditions of initial concentration, pH, current intensity,  $\text{Fe}^{2+}$  dosage, and anode material. These factors were chosen due to their significant influence on the performance of electro-Fenton process. In order to introduce the performance of electro-Fenton process, curves for different operating conditions were plotted portraying the remaining phenol amount ( $C/C_0$ ) versus the reaction time.

### 3.1. Effect of initial phenol concentration

The degradation of phenol at different initial concentrations is shown in Fig. 1a, and Fig. 1b. The obtained results indicated that the removal efficiency decreases with the increase in the initial concentration of phenol and more reaction time is required for complete degradation. Fig. 1a states that the removal efficiencies of phenol by nickel anode were 72%, 69.2%, 65%, 64.7%, and 51.42% at initial concentration of 50, 75, 100, 150, and 200 mg/L respectively in 120 min of electro-Fenton reaction. Fig. 1b demonstrates that the removal efficiencies of phenol by stainless steel electrode were 95.2%, 92.5%, 90.6%, and 72.4% at initial concentration of 50, 75, 100, and 200 mg/L respectively in 90 min of reaction time. The degradation efficiency of phenol at higher initial concentration decreased because the amount of hydroxyl radicals remained constant regardless of the initial phenol concentration. Additional time and Fenton reagent may be needed to attain more efficient degradation at higher initial concentration [23]. Mineralization of phenol was observed by measuring the TOC removal during electro-Fenton reaction as shown in Fig. 1c. The removal of TOC was about 41% in 120 min, while phenol degradation was 65%. At the beginning of the electro-Fenton reaction, the mineralization rate was effective, then became slower, but faster than phenol decay. It may be due to the fact that phenol forms aromatic and aliphatic intermediates which need prolonged time and more hydroxyl radicals for complete mineralization. Benzoquinone, catechol and hydroquinone are the most prevalent intermediates that generated by oxidation of phenol [24]. These intermediates are observed in water as TOC during the reaction and they are oxidized by hydroxyl radicals to more benign organics [25]. At the beginning of reaction, maleic, fumaric, succinic and glyoxylic are predominantly formed, while oxalic and formic acids are produced later. The end products after the complete degradation of intermediates are mainly  $\text{CO}_2$  and  $\text{H}_2\text{O}$  [26]. Adding supporting electrolyte such as sodium sulfate with dose of 1 g/L improved degradation of phenol from 23% to 88.3% in 60 min of reaction as shown in Fig. 1d. Adding  $\text{Na}_2\text{SO}_4$  improves the conductivity of the solution which led to higher current density that resulted in improved production of  $\text{H}_2\text{O}_2$ . Higher concentration of  $\text{H}_2\text{O}_2$  enhanced

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