PHENOL DEGRADATION: NUMERICAL SIMULATION IN A STIRRED TANK ELECTROCHEMICAL REACTOR

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ABSTRACT

In this work, phenol degradation of wastewater in a stirred tank chemical reactor was numerically simulated. The effect of the applied current density (I/A), changes in the concentration of dissolved O_2 *and initial concentration of* Fe^{2+} *ions were studied. The operation conditions were 25°C, pH = 2.5, with 1000 mM phenol feed concentration at 1 atm of pressure. The results show that the phenol removal achieved was 78.97%, 84.2%, 97.0% and 99.8% for values of I/A of 0.3, 0.32, 0.37 and 0.4 mA* cm^{-2}*, respectively. For initial dissolved* O_2 *concentration values of 0.007, 0.01, 0.03, and 0.4 mM, degradation was achieved at 74.7%, 77.4%, 81.9% and 84.2%, respectively. Finally for values of initial* Fe^{2+} *ions concentration of 40, 9 and 2 mM, the phenol removal was achieved at 76.0%, 80.7% and 86.3%, respectively. After several simulations times, it was found* $[C_f]_0 = 1000$ *mM were the optimum operating conditions in which* $[O_2]_0 = 0.4$ *mM,* $[Fe^{2+}]_0 = 2$ *mM and I/A = 0.4 mA* cm^{-2}*. With these values the phenol removal reached 99%, at dilution rate of 0.15* min^{-1}*. A viable model for phenol degradations proposed.*

KEYWORDS

Electrochemical treatment, Phenol degradation, Stirred tank chemical reactors, and Wastewater.

1. INTRODUCTION

Phenol is an important organic pollutant and is considered primary contaminant in wastewater due to high toxicity, high oxygen demand and slow biodegradability [13]. This compound is obtained from coal tar and is widely used as disinfectant for industrial and medical applications. This compound is present in the atmosphere as emission of fuel consumption (factory and vehicles). For this reason most of the phenol waste products is released into the environment [3]. Phenol concentrations higher of 2 mg l^{-1} in water can be toxic for living system, specially for fish, in fact the range between 10 to 100 mg l^{-1} can kill aquatic life in 96 h [9][13]. In many countries wastewater contaminated with phenol has been considered a health problem, stating the maximum permissible limit of 4 mg l^{-1} for potable water and 3.5 mg l^{-1} in superficial waters (lakes and brooks) to consume contaminated fishes [2]. As a consequence scientists are leading to use of technology to destroy this compound and decrease the environmental damage.

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Electrochemical methods (EM) for destruction of organic pollutant in wastewater has been attracted the attention, since this is a powerful clean method which environmentaly-frienly [19]. EM has the advantage over the traditionally biological treatment since this is not suitable for the degradation of chlorinated compounds, in the case of phenol, it can be toxic for the microorganism [8]. The electrochemical treatment can be set up easily and show high efficiency. In the EM, the pollutants are destroyed by either the direct or indirect oxidation process. In a direct anodic oxidation process, the pollutants are first adsorbed on the anode surface and then destroyed by the anodic electron transfer reaction [15]

From engineer point view the scientists have paid attention in order to develop of electrochemical reactor to remove pollutant from wastewater and heavy metal recovery. For that it is compulsory to star from pre-industrial phase (fundamental study) to the scaling processes. And to reach this latter is necessary to have a complete control of the process which implies time consumption and cost. For this reason a computing and simulation processes is good tool to know the characteristics and predict the functionality of any electrochemical reactor and chemical process.

Several scientific work have been carried in the phenol remove from water using electrochemical methods such as the work of Körbahti and Tanyolac (2003), in this they used a continuous tubular reactor of stainless steel with a cylindrical carbon anode at the centre for organic wastewater treatment in a fundamental study. They investigated the influence of residence time on phenol removal at 25°C, the electrolyte concentration of 120 g l^{-1}, 450-3100 mg l^{-1} phenol feed concentrations and an applied current density of 61.4 and 54.7 mA cm^{-2}, respectively were studied. These authors found that for 10 and 20 min of resident time, phenol degradation was 56% and 78%, respectively, when 450 mg l^{-1} phenol feed concentration and above 40 min of residence time all phenol was consumed within the reactor. Finally, when 3100 mg l^{-1} phenol feed concentration was considered and 1, 1.5, 2 and 3 h of residence time a phenol degradation of 42%, 71%, 81%, and 98%, respectively was observed. It was also found that more than 95% of the initial phenol was degraded into non toxic compounds, the latter show that this treatment can be environmental safe.

It is well know that organic pollutant degradation in aqueous media depends from other parameters and conditions, for example; the electrochemical reactions of phenol destructions depend of hydrogen peroxide (H_2O_2) concentration, ferrous ions (Fe^{2+}) concentration, dissolved oxygen and applied current density. Liu *et al.* (2003) described and proposed a kinetic model for phenol degradation in aqueous solution. The model considers the rates of H_2O_2 generation and consumption in the reaction solution. The authors also studied other parameter that affects the organic degradation such as the applied current density, dissolved oxygen concentration and initial ferrous ion concentration. All the parameters mentioned above have not been studied for the electrochemical degradation of phenol in a stirred tank reactor.

In this work, were proposed the model of phenol degradation in a stirred tank electrochemical reactor as a treatment for phenolic wastewater. The effect of the applied current density, the dissolved O_2 concentration and initial concentration of Fe^{2+} ions were studied. Other parameters such as the dilution rate and phenol feed concentration to obtain the bifurcation map were also considered.

2. PROBLEM DESCRIPTION

It has been reported that the degradation of organic contaminants by biological processes is limited, because at concentrations higher than 50 mg l^{-1} [18] the reaction is inhibited, requiring a costly physical or physiochemical pre-treatment. Electrochemistry actually plays an important role in a vast number of fundamental studies and applied areas [6] [21].

In the last two decades, electrosynthesis of hydrogen peroxide has received considerable attention, and several papers have demonstrated that in situ electrogenerated H_2O_2 may also be successfully used for the treatment of aqueous effluents containing organic pollutants [4] [5]. Recent research about Electro-Fenton reaction by generation H_2O_2 from dissolved oxygen in aqueous solution provides fresh H_2O_2 in a continuous mode, which is more efficient and cost-effective than conventional chemical dosing methods [13]. The Electro-Fenton kinetic reaction involves three key reactions: (1) the generation of H_2O_2 from dissolved oxygen on the surface cathode (Reaction 1), (2) the generation of hydroxyl radicals (·OH) between H_2O_2 and Fe^{2+} (Reaction 2), and (3) the degradation of organic substance by ·OH (Reaction 5). In the meantime, some reversed reactions and side reactions (Reactions 3, 4, 6 and 7) coexist along with the key reactions as summarized below [13]:

$$O_2 + 2H^+ + 2e^- \xrightarrow{k_1} H_2O_2 \qquad \text{(Reaction 1)}$$

$$Fe^{2+} + H_2O_2 \xrightarrow{k_2} Fe^{3+} + \cdot OH + OH^- \qquad \text{(Reaction 2)}$$

$$Fe^{2+} + \cdot OH \xrightarrow{k_3} Fe^{3+} + OH^- \qquad \text{(Reaction 3)}$$

$$Fe^{3+} + H_2O_2 \xrightarrow{k_4} Fe^{2+} + HO_2^{\cdot} + H^+ \qquad \text{(Reaction 4)}$$

$$\cdot OH + \text{organic} \xrightarrow{k_5} \text{products} \qquad \text{(Reaction 5)}$$

$$H_2O_2 + \text{organic} \xrightarrow{k_6} \text{products} \qquad \text{(Reaction 6)}$$

$$\cdot OH + H_2O_2 \xrightarrow{k_7} HO_2^{\cdot} + H_2O \qquad \text{(Reaction 7)}$$

2.2. System description

The concept of the ideal continuous stirred tank reactor (**CSTR**) is well known in engineering and is used as chemical synthesis [7] [12] [20]. The ideal continuous stirred tank reactor is a device in which can be a complete mixing of the contents. It operates isothermally, with the composition and temperature of the outlet solution matching the reactor contents [17].

The design equation for the **CSTR**, comes directly from the material balance. It describes the concentration change over the reactor caused by reactions. Referring to Figure 1, which depicts a unit of volume [15]. The flowrates into and out of the reactor are assumed equal and given by v and the inlet and outlet concentrations of species are C_{inlet} and C_{outlet}, respectively [7] [12].

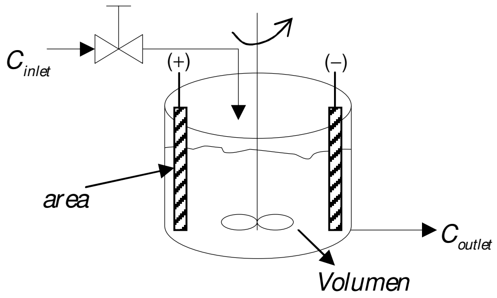

Figure 1. Continuous stirred tank reactor model [17] [1].

The dynamics of **CSTR's** are important due to considerations of stability, control and operating procedures. Two approaches to modelling are generally considered, analytical models of linear or linearized systems and dynamic simulation based on numerical methods. Both aspects are considered briefly here in the context of electrochemical reactions [17].

The dynamics model of **CSTR** in which m reactions take place involving n (n > m) chemicals species can be described by Eq. (1). Non-linearity in models of Eq. (1) is introduced by the reaction kinetics r (C) [16].

$$\frac{dC}{dt} = \theta\left(C_{in} - C\right) + Er\left(C\right) \tag{1}$$

Table 1. Model variables.

Nomenclature	
$C \in \mathbf{R}^n$	Vector of concentrations of chemical species
$C_{in} \in \mathbf{R}^n$	Vector non-negative and constant feed concentrations
$r\left(C\right) \in \mathbf{R}^m$	Smooth, non-negative, bounded vector of reaction kinetics, with r (C) = 0 $\forall \quad t \leq 0$
$E \in \mathbf{R}^n \times \mathbf{R}^m$	Stoichiometric matrix
Greek symbol	
θ	Dilution rate (i.e. flow rate/volume)

2.1. Case study

The dynamic model of **CSTR** was obtained from mass balance for phenol solution (C_f), which is represented by Eq. (2). The kinetic reaction is given by Liu *et al.* (2003) and the kinetic parameters are shown in Table 2. The model considers that reaction volume is constant, electrochemical reaction is homogenous, pH constant with isothermic process and atmospheric pressure.

$$\begin{aligned} r_f &= \frac{k_5}{k_3} \frac{1}{\left\{0.2\left[Fe^{+2}\right]_0 + \frac{k_5}{k_3}\left(C_f\right)\right\}} \frac{I}{A} \frac{k_1 K_{ad}\left[O_2\right]}{\left(1 + K_{ad}\left[O_2\right]\right)} \left(1 - e^{-\left(k_2(0.2)\left[Fe^{+2}\right]_0\right)t}\right) C_f \\ \frac{dC_f}{dt} &= \theta\left(C_{f_{inlet}} - C_f\right) - r_f \end{aligned} \tag{2}$$

Table 2. Kinetic and dynamic parameters.

Nomenclature	
$C_{f,\ inlet}$	Phenol feed concentration (mM)
I/A	Current density (mA cm^{-2})
$[Fe^{2+}]_0$	Initial ferrous ion concentration (mM)
K_{ad}	Adsorption equilibrium constant (1.1 l mM^{-1})
k_1, k_2, k_5/k_3,	Kinetic constants (3.96 mM^{-1} min^{-1}, 4.86 mM^{-1} min^{-1}, 0.31 dimensionless)
$[O_2]$	Oxygen concentration (mM)
r_j	Rate reaction (mM min^{-1})
t	Time (min)
Greek symbol	
θ	Dilution rate (min^{-1})

3. DISCUSSIONS AND RESULTS

The numerical simulations were done to validate proposed mathematical models at different operating conditions. Figure 2 shows the concentration profiles of phenol degradation at different dilution rates with initial phenol feed of 1000 mM. The phenol concentration at equilibrium increases as dilution rate increases. At dilution rate of 0.9, 0.4, 0.2 and 0.15 min^{-1} the phenol degradation is 14%, 31.6%, 63.19% and 84.2%, respectively. The phenol degradation reaches 100%, at a dilution rate of 0.05 min^{-1}. This means that batch regimen operation must be selected, which reduces highly the benefits of the continuous operation in large production, especially for industrial applications [1].

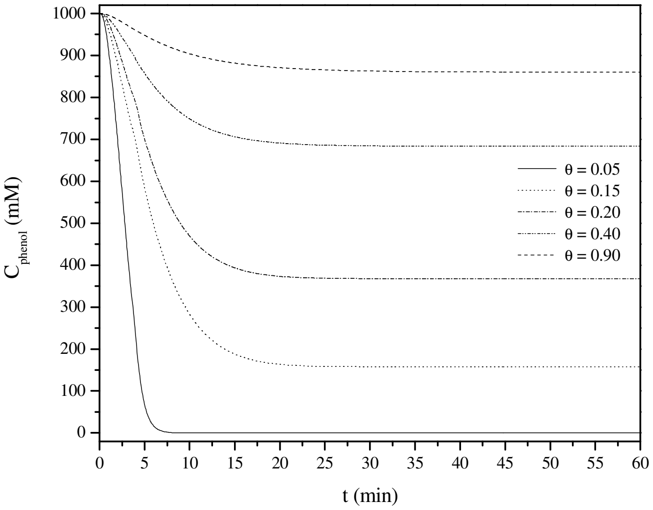

Figure 2. Phenol degradation at different dilution rate (θ), $C_{f,\,inlet}$ = 1000 mM, $[O_2]_0$ = 0.4 mM, $[Fe^{2+}]_0$ = 0.2 mM and I/A = 0.32 mA cm^{-2}.

The United States Health Department and Human Services report that maximum value of phenol concentration in wastewater is 4 mg l^{-1} or 0.048 mM. To accomplish this limit the dilution rate must be 0.05 min^{-1}. With the results shown in Figure 3, it is necessary implement a control strategy in order to maintain continuous operation [1].

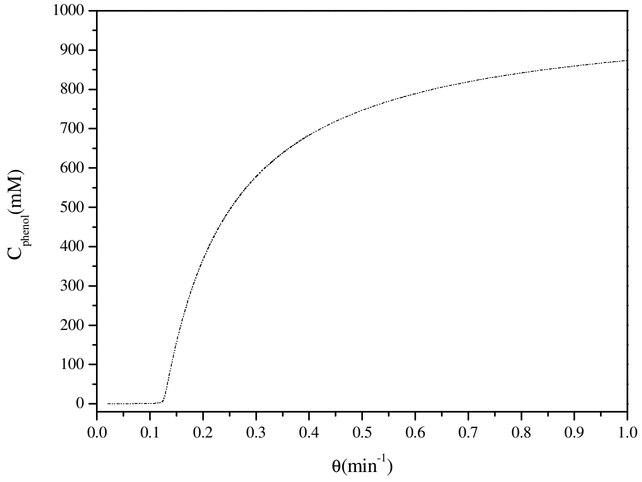

Figure 3. Bifurcation map of C_{phenol} vs. dilution rate at $C_{f,\,inlet}$ = 1000 mM, $[O_2]_0$ = 0.4 mM, $[Fe^{2+}]_0$ = 0.2 mM and I/A = 0.32 mA cm^{-2}.

Figure 4 shows the profiles of phenol degradation at differents $C_{f,\ inlet}$, $\theta = 0.15\ min^{-1}$. In this figure results show that the outlet phenol concentration increases when the phenol feed concentration increases. The phenol removal was 42.5%, 56.6%, 84.0% and 100% at phenol feed concentration of 2000, 1000, 1500 and 500 mM, respectively.

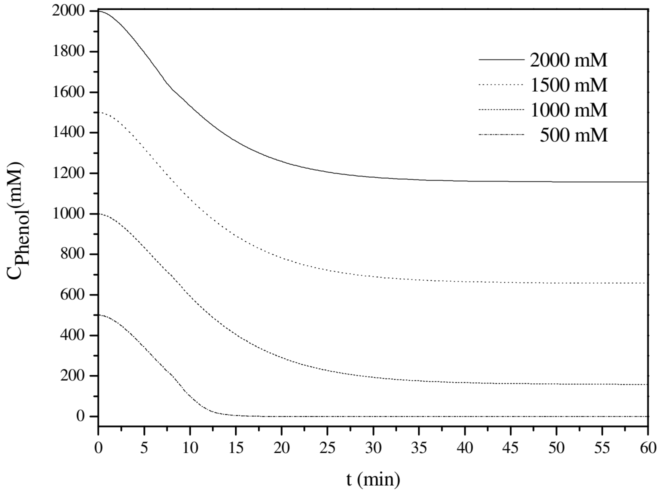

Figure 4. Profiles of phenol degradation at different $C_{f,\ inlet}$, $[O_2]_0 = 0.4$ mM, $[Fe^{2+}]_0 = 0.2$ mM, I/A = 0.32 mA cm^{-2} and $\theta = 0.15\ min^{-1}$.

Figure 5 show that outlet phenol concentration diminishes when the current density increases; this indicates that the phenol degradation increases. The phenol removal is 79.0%, 84.2%, 97.0% and 99.9%, at I/A values of 0.3, 0.32, 0.37 and 0.4 mA m^{-2}, respectively. The theoretical settling time can be calculated as 4 times the inverse of the dilution rate and mathematically as: $\tau = 4\theta^{-1}$ [16], which is 26. 7 min at I/A value of 0.4 mA m^{-2} and the settling time of simulation is approximately 32 min.

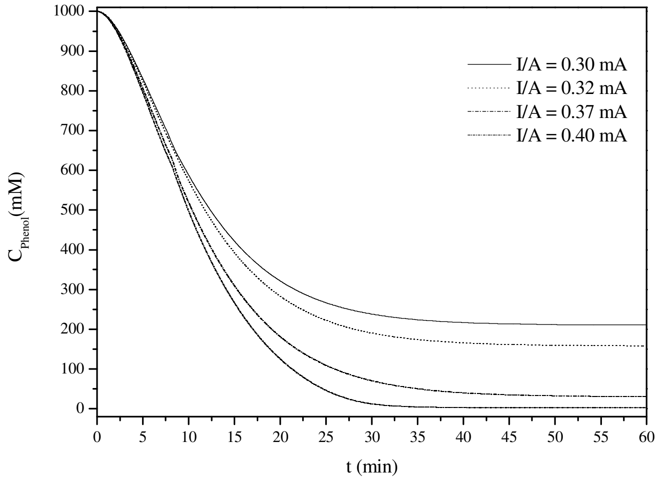

Figure 5. Profiles of phenol degradation at different I/A (mA cm^{-2}), $C_{f,\ inlet} = 1000$ mM, $[O_2]_0 =$ 0.4 mM, $[Fe^{2+}]_0 = 0.2$ mM, and $\theta = 0.15\ min^{-1}$.

If initial oxygen concentration increases, the outlet phenol concentration diminishes, as shown in Figure 6. In this figure the profiles of phenol degradation at different initial oxygen concentration is shown. The phenol removal was 74.7%, 77.4%, 81.9% and 84.2% with initial oxygen concentration of 0.007, 0.01, 0.03, and 0.4 mM, respectively. As shown, the phenol removal is not complete.

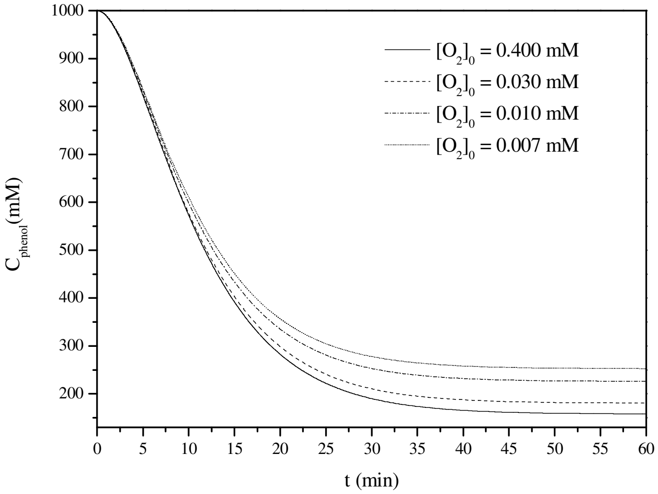

Figure 6. Profiles of phenol degradation at different $[O_2]_0$, $C_{f,\,inlet}$ = 1000 mM, $[Fe^{2+}]_0$ = 0.2 mM, I/A = 0.32 mA cm^{-2} and θ = 0.15 min^{-1}.

In Figure 7, the results shown indicate that when the initial ferrous ion concentration diminishes, the outlet phenol concentration diminishes, also indicating that the phenol degradation is not complete. Phenol removal is 76%, 80.7% and 83.6%, at $[Fe^{2+}]_0$ of 40, 9 and 2 mM, respectively.

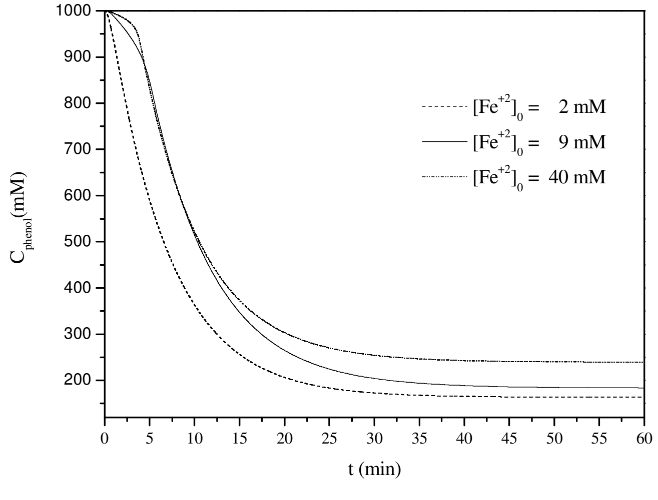

Figure 7. Profiles of phenol degradation at different $[Fe^{2+}]_0$ (mM), $[O_2]_0$ = 0.4 mM, $C_{f,\,inlet}$ = 1000 mM, I/A = 0.32 mA cm^{-2} and θ = 0.15 min^{-1}.

Figure 8 shows that there is a linear relationship in the range from 0.30 to 0.38 mA cm^{-2}, after which equilibrium is reached. Phenol equilibrium concentration diminishes if the current density increases to reach equilibrium at approximately at 0.44 mA cm^{-2} of I/A, therefore at which the phenol removal is 100%.

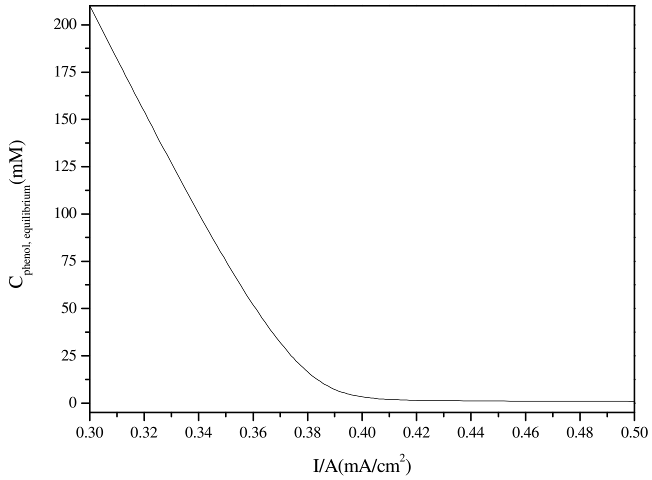

Figure 8. Profile of the phenol equilibrium concentration versus current density at $[Fe^{2+}]_0 = 0.2$ mM, $[O_2]_0 = 0.4$ mM, $C_{f,\ inlet} = 1000$ mM and $\theta = 0.15\ min^{-1}$.

The profile of the phenol equilibrium concentration versus initial dissolved oxygen concentration has an exponential decay form. If the initial dissolved oxygen increases the phenol removal increases to reach equilibrium at approximately $[O_2]_0 = 0.32$ mM as seen in Figure 9.

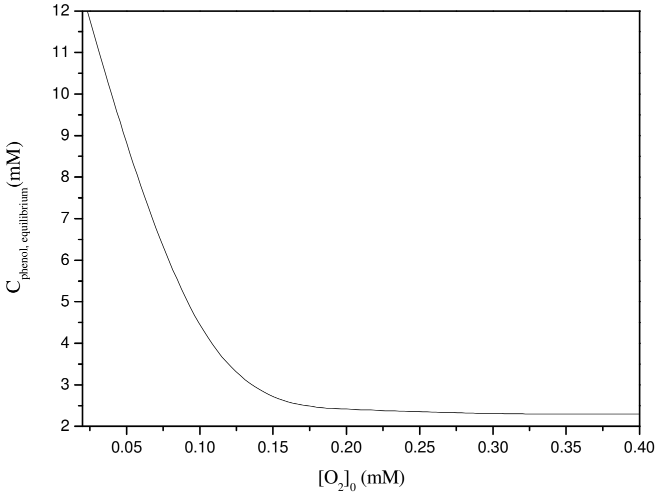

Figure 9. Profile of the phenol equilibrium concentration versus initial dissolved oxygen concentration at $[Fe^{2+}]_0 = 0.2$ mM, $C_{f,\ inlet} = 1000$ mM, I/A = 0.4 mA cm^{-2} and $\theta = 0.15\ min^{-1}$.

Liu *et al.* (2007) consider that at high $[Fe^{2+}]_0$ the phenol degradation is sluggish, because higher $[Fe^{2+}]_0$ could react with H_2O_2 and decompose it quite faster, as shown in Figure 10.

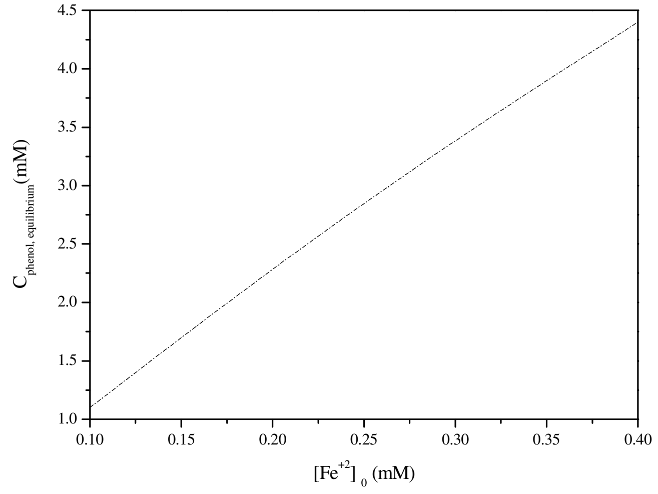

Figure 10. Profile of the phenol equilibrium concentration versus initial dissolved oxygen concentration at $[Fe^{2+}]_0 = 0.2$ mM, $C_{f,\,inlet} = 1000$ mM, I/A = 0.4 mA cm^{-2} and $\theta = 0.15$ min^{-1}.

After many simulations, it was found $[C_f]_0 = 1000$ mM to be the optimum operating conditions, which are $[O_2]_0 = 0.4$ mM, $[Fe^{2+}]_0 = 2$ mM, I/A = 0.4 mA cm^{-2}; with these values the phenol removal achieved is 99.9%, at a dilution rate of 0.15 min^{-1}. With the results shown in Figure 11 we can compare the theoretical settling time and simulation settling time as $\tau_{theoretical}/\tau_{simulation} = 0.8$, therefore the proposed model is feasible.

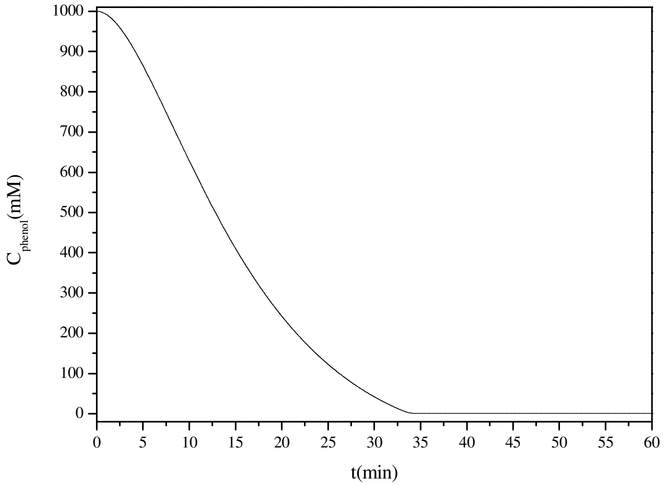

Figure 11. Profile of the phenol equilibrium concentration versus initial dissolved oxygen concentration at $[Fe^{2+}]_0 = 0.2$ mM, $C_{f,\,inlet} = 1000$ mM, I/A = 0.4 mA cm^{-2} and $\theta = 0.15$ min^{-1}.

4. CONCLUSIONS

The phenol degradation in a stirred tank electrochemical reactor depends on operating conditions, such as the dilution rate, current density, initial dissolved oxygen concentration and initial ferrous ion concentration.

When the current density and/or the initial dissolved oxygen concentration are high it is beneficial for phenol degradation, due to high levels of hydrogen peroxide that can be generated at the electrode surface, while when initial ferrous ion is high the phenol degradation is slow, due to the hydrogen peroxide decomposition.

This study found the optimum operating conditions of electrochemical reactor, as $[O_2]_0 = 0.4$ mM, $[Fe^{2+}]_0 = 2$ mM, I/A = 0.4 mA cm^{-2}. With these values the phenol removal achieved is 99%, at a dilution rate of 0.15 min^{-1}, thus the proposed model is feasible.

The results indicate that the stirred tank electrochemical reactor may be a feasible and safe alternative system for the electrochemical treatment of phenolic wastewater, being environmentally friendly, with little waste. Moreover, the treatment is low-cost.

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