



Electro-Fenton process efficiency for decolourization of aqueous solutions: study of reaction kinetics

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ABSTRACT

Electro-Fenton is one of the advanced oxidation processes, which have been applied effectively to degrade recalcitrant organics such as dyes in aqueous solutions. In the present research, the efficiency of electro-Fenton process in the removal of reactive yellow dyes of 15 and 42 were investigated. In this regard, the effects of various parameters such as pH, H₂O₂ concentration, current density, contact time and initial concentration of dyes were investigated. Colorimetry along with spectrophotometry was used to measure the concentration of the residual dye. The highest removal efficiencies (95%) for both reactive yellow dyes of 42 and 15 occurred at pH = 3, H₂O₂ concentration = 5 mg/L, contact time = 20 min, current density = 0.3 A and initial concentration = 80 mg/L. Also, the results of the kinetic study showed that data in this research follow the pseudo-first-order kinetic model.

Keywords: Electro-Fenton; Reactive dyes; Electrochemical oxidation; Reaction kinetics; Degradation

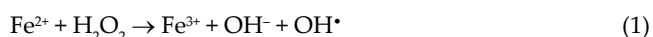
1. Introduction

The increasing development of industries and consequently the production of industrial wastewater and discharge of this sewage into the environment causes great concerns about contamination of surface and groundwater resources and environmental pollution [1]. Because of the high consumption of chemicals during the industrial processes, the effluent of these industries has high fluctuations in quality and quantity [2–5]. Synthetic dyes according to their application are one of the common pollutants found in effluent stream of textile industries. Generally, dyes used in industrial objectives include reactive, acidic, alkaline and dispersible [6]. At present, about 100,000 different types of dyes are produced worldwide whose annual production is about 700,000 tons [7–9]. Textile and dyeing industries are the most important consumers of synthetic and chemicals dyes

for dyeing processes. These dyes may be also used in other industries such as leather, paper, cosmetics, medicine, ink, printing, tanning [10–13]. In these industries, effluent wastewater is coloured due to the use of a variety of dyes at different stages of production and processing [14,15]. Previous studies show that 15%–20% of dyes used in these industries are discharged into the effluent wastewaters. In another study, it was found that the amount of dye discharged into the effluent varies from 2% for base colours and 50% for reactive dyes in the textile industry and effluent produced has concentrations between 10 and 100 mg/L [7–9]. Some types of dyes may be carcinogenic and mutagenic. They may also be toxic due to the presence of chromium, zinc, cadmium, copper and nickel salts in the structure of these dyes [16,17]. Therefore these coloured wastewaters must be treated before being discharged into the environment.

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Advanced oxidation processes are one of the new and effective methods for reducing organic compounds. The basis of these processes is the production and use of free radicals, and the final products of these processes are water and CO₂ [18–21]. Fenton process is the most common advanced oxidation method, which, under acidic conditions, will produce species with high oxidation power by reacting hydrogen peroxide with Fe²⁺ ions and these active species break down organic matter [22,23]. The following equation shows the reaction of the Fenton process:



Recently, advanced electrochemical oxidation processes, which are carried out based on the Fenton process are used as a clean technology without side effects to destroy organic pollutants. One of these processes is the electro-Fenton (EF) process. The EF process is one of the indirect electrochemical methods. In other words; this process combines two processes of Fenton and electrocoagulation to achieve more efficiency that hydroxyl radicals are generated by electrical energy in a mildly acidic environment [24–26]. Radicals have a non-coupled electron they have a shorter life and they tend to react with organic matter [27–29]. Advantages of the EF method include environmental compatibility, rapidity, cost-effectiveness, versatility, higher degradation of organic pollutants due to the continuous production of Fe²⁺ in the cathode, and reduced sludge production volumes compared with the Fenton process [30,31]. Electrochemical processes in terms of guidance consist of four groups, in the first type, Fe²⁺ and H₂O₂ ions are produced electrically at the anode electrode surface and cathode electrode, respectively [26]. In the second kind, hydrogen peroxide is added manually and uses iron electrode as an anode and electrical source of Fe²⁺[11]. In the third kind, iron ion is added manually and hydrogen peroxide is produced by diffusion oxygen (electrolysis water) at the cathode [23,32] and in the fourth method, Fenton's reagent is used [33]. In this study, the second method was used. Mechanism of the process is that by creating an electric current at the cathode, oxidation–reduction reactions occur and Fe²⁺ ions are produced. Thereafter in the presence of hydrogen peroxide, Fe²⁺ ions react with hydrogen peroxide. The final product of these reactions is hydroxyl radical (OH[•]) and ferric ions (Eq. (1)). In the anode, the ferric ions revived to the ferrous by taking electrons, which this way resolves the problem of forming sludge (Eq. (6)). Ultimately, hydroxyl radicals with high reactivity lead to degradation of the organic matter [34].

In the study conducted by Sánchez-Sánchez et al. [35], EF process was applied to remove organic pollutants and the results showed that the removal efficiency of 95% was obtained [35]. In another research performed by Panizza and Cerisola [28], EF process was used for removal of organic pollutants from industrial wastewater and the results showed that the elimination of dye and COD was 89% and 87%, respectively. In a similar study conducted by Kurt et al. [11], the treatment of tannery wastewater was investigated by EF process and the result showed that COD removal was 95%, obtained at pH = 3 and electric current = 15 W [11]. In another study conducted by Hammami

et al. [20], the EF process was used to remove direct Orange 61 dye and the results showed that in the optimal conditions, 98% removal efficiency was obtained. In the study conducted by Rêgo et al. [36], the efficiency of EF process was determined in removal of Blue Novacron dye and the result showed that high performance was achieved in the optimal conditions of 400 mA, 1 μMFe²⁺ ion and pH = 3. In another study conducted by Cruz-González et al. [37], the EF process was used to remove acid yellow 36 dye and according to the results of this study, it was found that the maximal decolourization efficiency of 98% was achieved in the optimal conditions [37]. Therefore, this study aimed to investigate the EF process in the removal of reactive yellow 15 and 42 dyes from aqueous solutions.

2. Materials and methods

2.1. Chemicals and apparatuses

In this work, two reactive dyes (reactive yellow 42 and reactive yellow 15 [RY15 and RY42]) were used, which were purchased from Sigma-Aldrich Company (USA). The chemical structures of these dyes have been shown in Fig. 1. Other chemicals required to perform the experiments were purchased from Merck Company (USA). Stock solution of 1,000 mg/L of reactive dyes was prepared for the implementation of the experiments. Other specific concentrations of the required samples were prepared by dilution of this stock solution. pH of the samples was adjusted to the desired value by addition of H₂SO₄ or NaOH. For all experiments, the residual dye concentrations of RY15 and RY42 were measured using a spectrophotometer (DR 5000) in the wavelengths of 420 and 430 nm, respectively.

2.2. EF activity

The EF reactor consisted of a 1,000 mL glass cylinder and two iron electrodes with dimensions of 17 × 3 cm. The electrodes are connected by interface wires alternately to the electric current generator with different currents in which one of the electrodes was used as a cathode and other as anode (Fig. 2). The variables examined in this study included pH (3, 5, 7, 9, 11), concentration of hydrogen peroxide (0, 5, 10, 15, 20, 30, 50, 100 mg/L), concentration of dyes (10, 20, 30, 50, 80, 100, 150, 200 mg/L), applied current density (0.05, 0.2, 0.3, 0.5, 1 A) and reaction times (5, 10, 15, 30, 45, 60, 120 min). At all stages of the process, the removal efficiency was calculated by Eq. (2):

$$R = \frac{(C_0 - C)}{C_0} \times 100 \quad (2)$$

where *R* is the efficiency of the process for the removal of dyes. *C*₀ and *C* are the initial and final concentrations after a certain time, respectively.

3. Results and discussion

3.1. Effect of different systems of H₂O₂, electrolysis and EF

This study was first carried out to investigate the effects of various systems such as H₂O₂ (pH = 3, concentration of

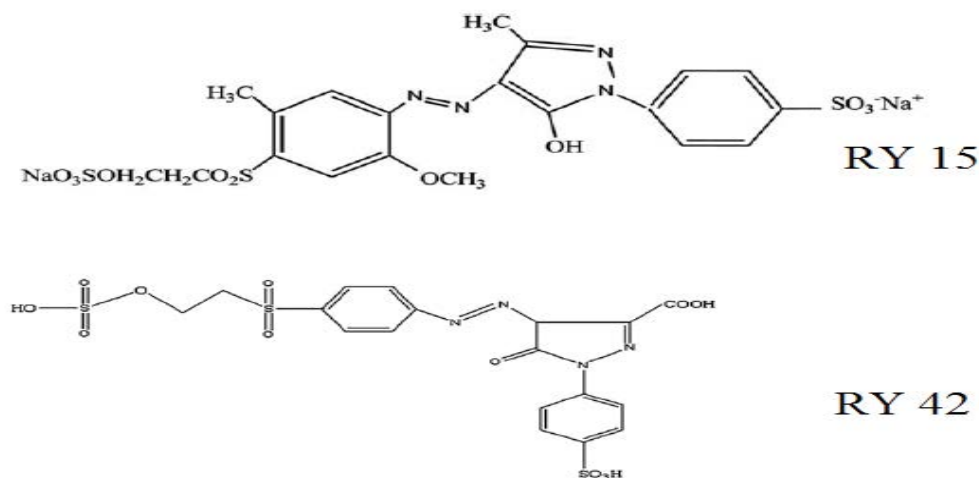


Fig. 1. Chemical structure of reactive yellow 15 and reactive yellow 42 dyes.

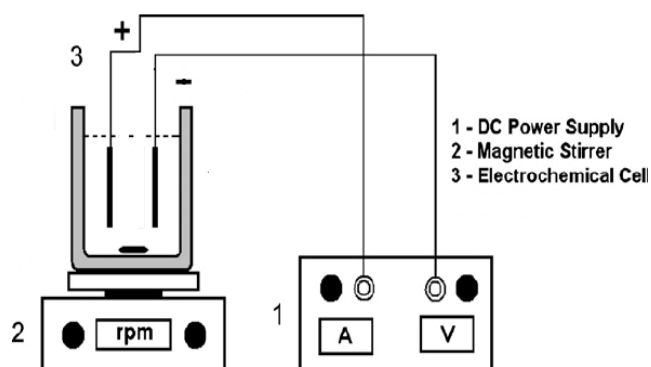
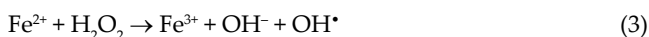


Fig. 2. Schematic of the EF reactor.

$\text{H}_2\text{O}_2 = 5 \text{ mg/L}$, initial concentration of dyes = 20 mg/L), electrolysis (pH = 3, current density = 0.4 A, initial concentration of dyes = 20 mg/L) and EF (pH = 3, current density = 0.4 A, concentration of $\text{H}_2\text{O}_2 = 5 \text{ mg/L}$, initial concentration of dyes = 20 mg/L) on the removal of reactive yellow 42 and 15. The results of the various systems impact and their synergistic effect on the removal of reactive yellow 15 and 42 have been shown in Figs. 3a and b, respectively. The results show that hydrogen peroxide has a poor performance as an oxidizing agent singly and the highest removal efficiency was about 20% in this process. The reason maybe that the H_2O_2 is stable in environmental conditions. According to the results, the removal efficiency was low in the electrolysis process so that the removal efficiency for both colours was about 40%, and the reason is that because catalyst iron and hydrogen peroxide were not added to electrolyte solution and thus the production of radical hydroxyl and degradation of dyes were low in the electrolysis process. In the EF system, the following general reaction (Eq. (3)) is performed by adding hydrogen peroxide and providing an electric current.



The results showed that the EF process causes the RY42 and RY15 to be removed about 85% and 80%, respectively.

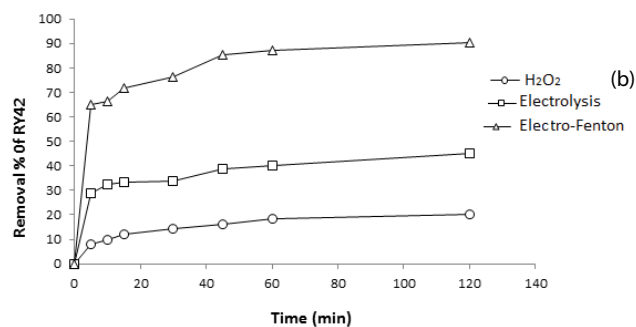
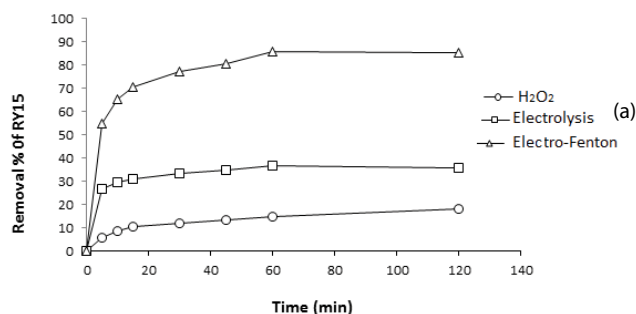


Fig. 3. Effect of different systems of H_2O_2 , electrolysis and electro-Fenton in degradation of RY 15 (a) and RY 42 (b).

When the electric current is established in the cathode and anode, the Fe^{2+} ions enter into the solution and produces hydroxyl radicals in reaction with hydrogen peroxide. Therefore, hydroxyl radical is the main factor in increasing the efficiency of this system with the decomposition of organic dyes [11].

3.2. Effect of pH

The pH of the solution is one of the most important parameters affecting the chemical reactions. This parameter affects both, directly and indirectly, oxidation of organic

compounds and production of hydroxyl radicals. To evaluate the effect of pH on the EF process in dye removal, solutions with initial concentrations of 50 mg/L for reactive yellow 15 and 100 mg/L for reactive yellow 42, pH adjusted between 3 and 11, H_2O_2 concentration of 5 mg/L, the voltage of 0.05 A and contact time of 30 min were investigated, which the results were shown in Fig. 4a. According to the results, the highest removal efficiencies of 93.44% and 97.55% for RY 15 and RY 42 dyes were observed in pH = 3. Which this value was considered as optimum pH. At the pH ranges 3–5 with applying the electric current, Fe^{2+} ions are generated and these ions act as catalysts and cause decomposition of hydrogen peroxide. The result of these reactions is the production of hydroxyl radical and decolorization of reactive dyes has occurred as a result of the performance of these radicals. However when pH of the solution is less than 3, $Fe(OH)^{2+}$ species are formed, which the reaction of this compound gently performed with hydrogen peroxide and will lead to reduce the hydroxyl radicals production and thereafter reduce the efficiency of the process. In these pHs, the hydrogen peroxide takes a proton and converts to the oxonium ion ($H_3O_2^+$). This compound is more stable than hydrogen peroxide and it has decreased the reaction with Fe^{2+} ions which reduce the production of hydroxyl radicals and reduce the efficiency of EF process [38]. However, when the pH of the solution is alkaline, Fe^{2+} ions get converted to Fe^{3+} ions and get precipitated as $Fe(OH)_3$ thus these ions get out of the catalytic cycle of the process. On the other hand, H_2O_2 is unstable at higher pH and may be converted to water and oxygen, which in this state will lose their oxidation ability and the coagulation process be replaced with EF. According to the results of this study

as well as other studies, pH is effective in the production species of ferrous iron catalytic and radical hydroxyl [3,38,39].

In the study by Zhou et al. [39], EF process was used to remove Methyl Red and also in the study of Chiou et al. [40], EF process was used to remove Reactive Black 5 dye and it was found that increasing the pH value reduced the process efficiency and pH = 3 was considered as optimum. In another study conducted by Cruz-González et al. [37] elimination of Acid Yellow 36 by EF was investigated and the results showed that the EF process had higher efficiencies in acidic pH.

3.3. Effect of H_2O_2 concentration

Another important parameter in EF process is H_2O_2 concentration because it is the only source of hydroxyl radical production in Fenton and EF processes. H_2O_2 is either manually added to the reactor or produced by diffusion of oxygen at the cathode [41], which in this study H_2O_2 was added manually. Therefore, determination of optimal hydrogen peroxide concentration is important in EF and Fenton-based reactions due to related costs of hydrogen peroxide impact on the process efficiency as well as the creation of chemical oxygen demand conditions due to residual hydrogen peroxide in water [42]. In this study, the effect of H_2O_2 concentration in the range 0–100 mg/L was investigated at pH = 3, applied current = 0.05 and contact time = 30 min. The results are expressed in Fig. 4b. The diagram shows that at first, by increasing the H_2O_2 concentration, dye removal efficiency was increased but at high H_2O_2 concentrations, the removal efficiency was decreased.

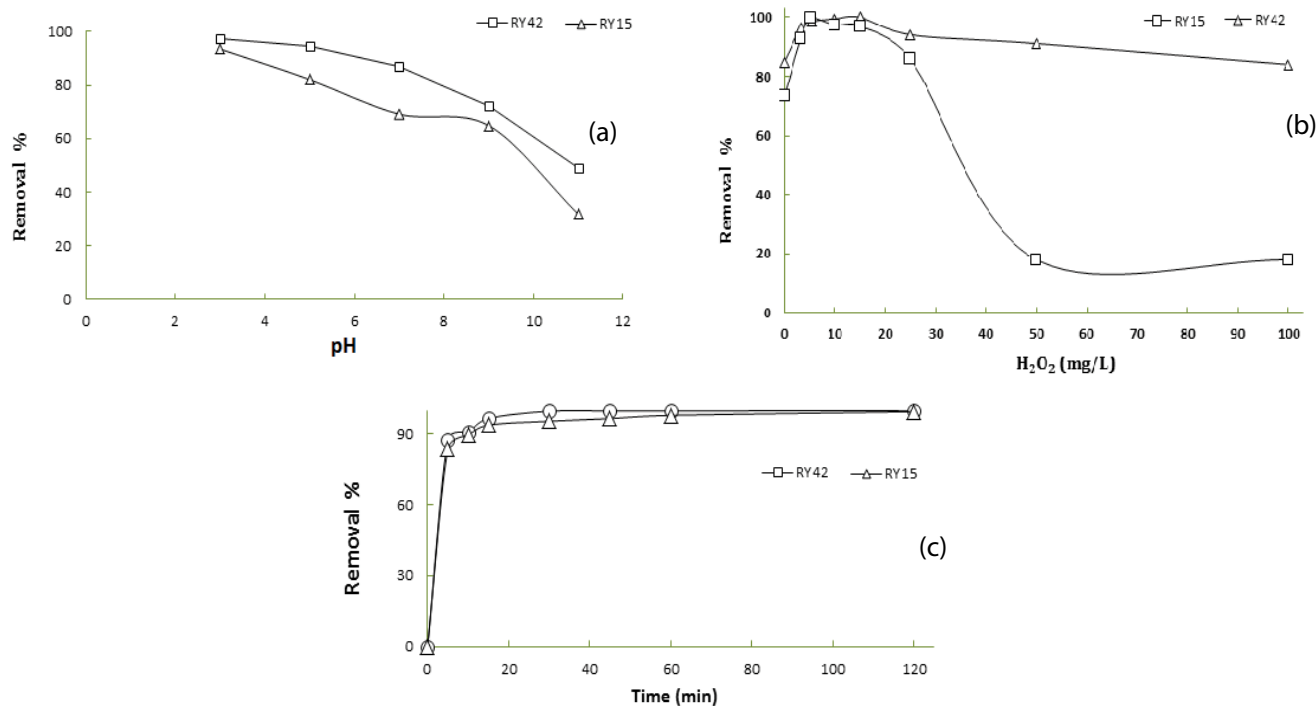


Fig. 4. Effect of pH (a), H_2O_2 concentration (b) and contact time (c) on the efficiency of electro-Fenton process in degradation of RY 15 and RY 42 dyes.

Regarding to Fig. 3, by increasing the H_2O_2 concentration from 0 to 15 mg/L, the removal of reactive yellow 42 has been increasing but at concentrations higher than 15 mg/L, dye removal has decreased because H_2O_2 reacts with Fe^{2+} ions in the solution and hydroxyl radical is produced which these radicals remove organic dye [41]. But in high H_2O_2 concentrations, it has a scavenger role and it consumes the generated OH^\bullet radicals. The scavenger process takes place in two ways, first H_2O_2 reacts directly with hydroxyl radical and produced (HO_2^\bullet) radical has weak oxidation power unlike radical hydroxyl (Eq. (4)). Second, the produced (HO_2^\bullet) radical will be indirectly consumer of hydroxyl radical (Eq. (5)). The results of the study conducted by Sun et al. [43] on the removal of direct Blue 15 with Fenton process showed that increase in the concentration of hydrogen peroxide from 50 mg/L resulted in a reduction of process efficiency. Ertugay and Acar [44] studied the removal of direct Blue 71 with Fenton process and it was found that excessive concentration of H_2O_2 reduced the process efficiency, the researchers stated that the process has been reduced due to the consumption of hydroxyl radicals by hydrogen peroxide and converted to HO_2^\bullet radical.



3.4. Effect of contact time

In this study, the effect of contact time in the range 5–120 min, pH of 3, H_2O_2 concentration of 5 mg/L, respectively, for both reactive yellow dyes 15 and 42 and current intensity of 0.05 A was investigated.

In the chemical processes, contact time is one of the main factors affecting the process efficiency. During the relevant processes including EF and Fenton processes, over the time, hydrogen peroxide decomposes and instead the byproducts increased. On the other hand, mixing in the reactor increases the probability of ion contact with the intermediate products resulting from the decomposition of hydrogen peroxide, and finally, the amount of hydroxyl radical production in the reaction is increased and the efficiency of the process improves. Fig. 3 shows the effect of reaction time on the efficiency of EF process in the removal of RY15 and RY42. According to Fig. 4c, at the first 10 min, 89.7% of RY42 and 87% of RY15 were removed by EF process. Higher efficiencies were achieved in the early stages of reaction and after the removal efficiency increased with lower rate by increasing time up to 120 min. So that in 120 min, the removal efficiency was 97.9% and 99.7% for RY15 and RY42, respectively. The reason is that by increasing the reaction time the intermediate compounds are produced and these compounds have been acted as rivals of pollutants which react with hydroxyl radical [45]. In Fenton-based processes, the highest amount of hydroxyl radical is produced in the first 10 min of the process, so the highest removal efficiencies occurred at this time [46–48].

3.5. Effect of current density

In the EF reaction, for providing the Fe^{2+} ions, the chemicals are not added into the solution. The only source of Fe^{2+}

ions in the reactor is the anode electrode for this process. The voltage applied to make electrical current and production of iron ions is one of the most important parameters in the process [39]. Iron ions have an important role in electro-Fenton and Fenton processes and act as a catalyst improving the production of hydroxyl radicals by H_2O_2 . Importance of iron ions is so high that H_2O_2 alone is not able to oxidize molecules of dye [49,50]. In this process, the main factor in the production of iron ions is the voltage variation applied at anode [39]. In the EF process, concentration of iron ions from an iron electrode was determined (Eq. (6)), and Fe^{2+} ion regeneration process through continuous resuscitation of Fe^{3+} ions at the cathode was performed (Eq. (7)) and therefore the production of radical hydroxyl was controlled by applied current density [51].



By applying current density, iron ions are produced and reacted with hydrogen peroxide in the solution and hydroxyl radicals are produced which performs the destruction of dyes. Fig. 5a shows the effect of the electric current in the voltage range of 0.05 to 1 A, pH = 3 and contact time = 15 min in removal of RY15 and RY42 using EF process. Regarding this figure, increasing the electrical current up to 0.5 A has increased the removal efficiency of dye, because the increase in current density leads to an increase in production of Fe^{2+} ions and hydroxyl radical. But an increase in the current density more than 0.5 A led to a loss in removal efficiency. The reason is that the increase in voltage leads to the diffusion of oxygen from the cathode (Eq. (8)) and therefore at high voltages, decomposition of hydrogen peroxide into the water reduces the removal efficiency of dyes [52].



3.6. Effect of initial concentration of dye

The initial concentration of pollutant is one of the factors which influence the chemical and electrochemical processes. Fig. 5b shows the effect of initial dye concentration changes (10–200 mg/L) on the EF process efficiency in removal of RY15 and RY42 at pH = 3, contact time = 15 min and current density = 0.3 A. According to the results, it was found that the removal of contaminant decreases by increasing the initial concentration. So that under the same conditions at the concentration of 10 and 100 mg/L, the removal efficiency of RY15 and RY42 were about 97% and 70%, respectively. The reason for this trend is that by increasing the initial concentration, the consumption of hydroxyl radical increases. The result produces more intermediate materials and this intermediate material reduces the removal efficiency of dye by consumption of some of the hydroxyl radicals [53]. In the study conducted by Zhou et al. [39] that investigated the removal of methyl red by EF process, it was found that increasing the initial concentration of dye reduced the process efficiency and the researchers stated that the reason for this trend is the

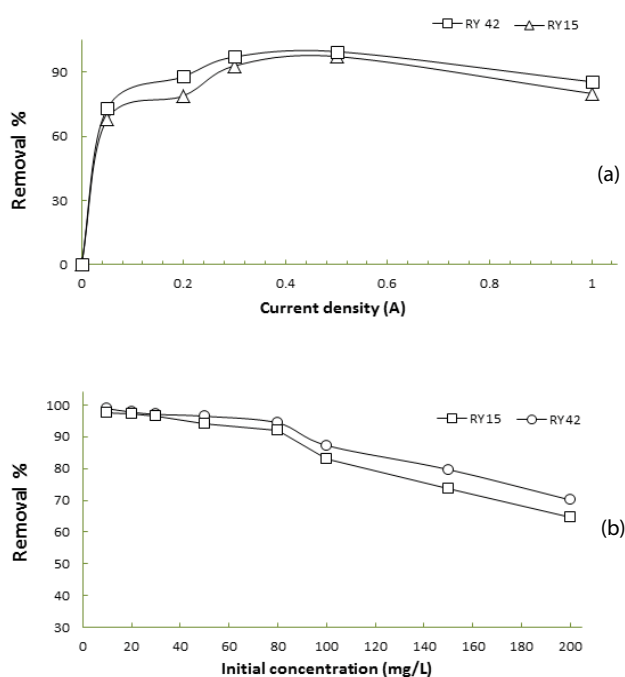


Fig. 5. Effect of current density (a) and initial concentration (b) on the efficiency of electro-Fenton process in degradation of RY 15 and RY 42 dyes.

consumption of hydroxyl radical by intermediate materials. In another study conducted by Panizza and Oturan [54] which investigated the removal of Alizarin Red dye with the EF process, the efficiency of EF process decreased with increasing initial concentration of dye. The researchers also concluded that with increasing initial concentration, to achieve the high efficiency, the required time should be increased [54].

3.7. Kinetic study of reactive dyes degradation using EF process

In electrochemical and chemical processes, the reaction kinetics can be applied to study the mechanisms of pollutant degradation. In several studies, it has been reported that the kinetics of electrochemical and chemical process of pollutants can be investigated by Langmuir–Hinshelwood (L–H) kinetic model (Eq. (9)) [55–59].

$$\ln\left(\frac{C_0}{C_t}\right) = -K_{\text{obs}}t \quad (9)$$

In this study, to evaluate the experimental kinetics by EF process, the equilibrium concentration of RY15 and RY42 dyes of 80 mg/L were applied at different reaction times (5, 10, 15, 30, 45, 60 and 120 min). In Eq. (9), C_0 is the initial concentration of dyes, C_t is the concentration of dyes at time t , K_{obs} is the pseudo-first-order reaction rate (min^{-1}) and t is the contact time (min). The result of reaction kinetics has been shown in Fig. 6. According to the values obtained for the regression parameter (R^2), it is known that the EF process in degradation of dyes in all the experiments well fitted to the using the pseudo-first-order kinetics with

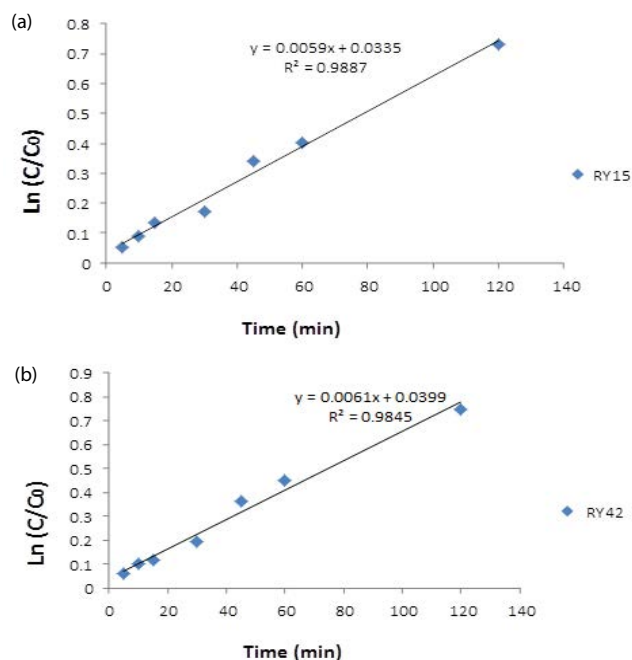


Fig. 6. Kinetic curve of pseudo-first-order for reactive yellow 42 and 15 dye degradation (pH = 3, hydrogen peroxide concentration = 5 mg/L, reacton time = 20 min, intensity of electric current = 0.3 A and initial concentration = 80 mg/L).

high correlation coefficients (R^2). On the other hand, the previous researchers in their studies reported data which follows the pseudo-first-order kinetics [55–59].

4. Conclusion

The results of this study showed that the EF process had a high efficiency in the removal of RY15 and RY42 using producing iron ion by applying the voltage and current density. The optimum conditions were pH = 3, concentration of hydrogen peroxide for reactive yellow 42 and 15 = 5 mg/L, contact time about 20 min, intensity of electric current = 0.3 A and initial concentration = 80 mg/L. In this situation, the removal efficiency of 95% was achieved to remove RY15 and RY42 dyes. On the other hand, the data follow the pseudo-first-order kinetics. As a result, the EF process can be used as an acceptable method for removal of reactive yellow 15 and 42 dyes from aqueous solution.

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Ethical issues

The authors hereby confirm that all data collected during the Research are as expressed in the manuscript, and no

data from the study has been or will be published separately elsewhere.

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