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COD REDUCTION OF BAKER'S YEAST WASTEWATER USING BATCH ELECTROCOAGULATION

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Abstract

The aim of the present work was to investigate experimentally the reduction of Chemical Oxygen Demand (*COD*) levels from baker's yeast wastewater (BYW) using batch electrocoagulation (EC) treatment process. An electrocoagulation unit with six iron electrodes was designed to achieve this treatment task. The effect of several operational parameters including pH, direct current (DC) density (I_ρ), mixing speed (MS), and temperature (T) on the performance of EC process was studied. The results indicated that the maximum removal efficiency of the COD was up to 85%. The visual test showed that the electrocoagulation process was able to remove completely the dark brown color of baker's yeast wastewater. The corresponding electrical energy and electrode consumption per kg *COD* removed was around 0.493 kWh and 2.956 kg iron, respectively. In addition, it was demonstrated that the reduction rate of *COD* follows the first-order model whose kinetic parameter, *k*, depends strongly on the DC current density according to a power law relation.

Key words: Baker's yeast wastewater, COD kinetics, electrocoagulation, iron electrodes, wastewater treatment

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1. Introduction

Food and fermentation industries usually generate highly polluted wastewater containing high level of different organic pollutants, quantified as Chemical Oxygen Demand (*COD*), including biodegradable organic pollutants expressed as Biochemical Oxygen Demand (*BOD*), and sulfates (Zub et al., 2008). In particular, the production of baker's yeast from molasses fermentation processes generates high amounts of wastewater. The resulted baker's yeast wastewater (BYW) is characterized by high COD and BOD values that reach 100 and 50 g/L, respectively (Liang et al., 2009). The baker's yeast wastewater is strongly nitrogenous with total nitrogen content of 1.5 g/L, while sulfate content could reach 10 g/L (Kalyuzhnyi et al., 2005). Moreover, BYW is acidic (pH from 4 to 5), has a strong odor, and a dark brown color (Gengec et al., 2012). The dark brown color is attributed to the

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presence of melanoidins pigments. Melanoidins are high molecular weight heterogeneous polymers that are formed when sugars and amino acids combine through the Maillard reaction at high temperatures and low water activity. These compounds have antioxidant properties that make them resistant to biodegradation (Wedzicha and Kaputo, 1992). The presence of these colored compounds could also impede light penetration in the biological treatment pans, thereby upsetting the biological treatment processes. Therefore, the removal of melanoidins is urgently required prior the effective treatment of baker's yeast industry wastewater (Liang et al., 2009).

Several treatment processes were investigated to treat the dark brown BYW in the last two decades. These processes include aerobic and anaerobic biological treatment processes (Satyawali and Balakrishnan, 2008), adsorption (Tahar et al., 2004), membrane processes (Mutlu et al., 2002), advanced oxidation processes (AOPs) using Fenton's reagents (Pala and Erden, 2005), H_2O_2/UV (Catalkaya and Sengul, 2006), ozone (Pena et al., 2007), and coagulation (Kalyuzhnyi et al., 2005). Unfortunately, all the above treatment processes suffer from serious drawbacks and were not able to effectively treat BYW.

In biological treatment processes as an example, the *BOD* is reduced to acceptable values, but the dark brown color and the corresponding *COD* persist and thus limiting the reuse of the treated water. On the other hand, AOPs could decolorize the wastewater but the *COD* level does not change and this could lead to the formation of new hazardous pollutants in the treated effluents (Pala and Erden, 2005). Consequently, baker's yeast wastewater requires more efficient treatment processes in terms of color and *COD* removal before its disposal or possible reuse (Gengec, 2012; Satyawali and Balakrishnan, 2008).

Electrocoagulation (EC) process has been proven as a feasible, economical, and ecological alternative in the treatment of various types of water and wastewater with promising results. Kuokkanen et al. (2013) have published a comprehensive review about the applications of electrocoagulation in treatment of water and wastewater. They have divided EC applications into the following main categories: tannery, textile and colored wastewater; pulp and paper industry wastewater; oily wastewater; food industry wastewater; surface water as well as model water and wastewater containing heavy metals, nutrients, cyanide and other elements and ions.

In comparison with the classical chemical coagulation process, it has been reported in literature that electrocoagulation is a simple, economic, and efficient method that potentially eliminates most of the problems encountered in the applications of the previous treatment processes (Al-Shannag et al., 2013; Chen, 2004; Kannan et al., 2006; Mollah et al., 2001; Zhu et al., 2005).

Electrocoagulation (EC) involves several complicated physical/chemical phenomena that can be simplified by the following steps (Daneshvar et al., 2012; Holt et al., 2005): (i) electrolytic reactions at sacrificial electrode surfaces, (ii) in situ formation and migration of coagulation agents that initiate the coagulation process of the pollutants present in the wastewater, (iii) adsorption of pollutants on coagulants, and (iv) flotation or sedimentation of aggregates.

The detailed oxidation-reduction reactions and the other corresponding chemical reactions are given in the study of Al-Shannag et al. (2012) and Daneshvar et al. (2012). When EC uses iron electrodes, as in the present work, the ferric hydroxides, $Fe(OH)_2$ and $Fe(OH)_3$, flocs that serve as coagulant agents are generated according to the following chemical reactions (1, 2) (Al-Shannag et al., 2012):

$$Fe_{(s)} + 2H_2O_{(l)} \leftrightarrow Fe(OH)_{2(s)} + H_{2(g)} \qquad (1)$$

$$4\,Fe_{(s)} + 10H_2O_{(l)} + O_{2\,(g)} \leftrightarrow 4Fe(OH)_{3(s)} + 4H_{2\,(g)} \qquad (2)$$

Notably, very limited information is available in the literature concerning the treatment of BYW by electrocoagulation (Gengec, 2012; Gengec et al., 2012; Kobya and Delipinar, 2008). Such studies considered the effect of various parameters like pH, current density, electrolysis time on the treatment efficiency. In addition, very few researches have considered kinetic modeling of EC processes treating various industrial wastewaters. Hu et al. (2007) used a variable order kinetic (VOK) model derived from the Langmuir equation to specify the kinetics of the fluoride removal reaction for electrocoagulation (EC).

Subsequently, Canizares et al. (2008) developed a mathematical model for the electrocoagulation treatment of kaolin suspensions. Their model, which was tested with experimental data, determines the concentration of the different aluminum species as a function of the total aluminum concentration and of the pH, and then it classifies these actual species into several model species as a function of their reactivity with the pollutant contained in the wastewater. Espinoza-Quinones et al. (2009) used the second-order kinetic model to explain the kinetics of electrocoagulation of tannery effluent using aluminum electrodes. Recently, Rajabi et al. (2011) used the pseudo first-order, pseudo second-order and intraparticle diffusion models to analyze the kinetic data of Remazol Yellow G removal by electrocoagulation. More recently, Sangal et al. (2013) investigated the de-emulsification of soluble-oil-water emulsion using electrocoagulation with aluminum electrodes. They reported that simple first-order kinetic fits the experimental data at a constant value of current density. However, the previous studies did not investigate the kinetic modeling and the effect of temperature on the *COD* removal of BYW emulsions using electrocoagulation. For this reason, the present work was devoted to investigate such important issues. In addition, the effect of other parameters such as pH, current density, and EC time on the EC treatment efficiency was also taken into account. In the absence of clear understanding of the kinetic mechanism for BYW emulsion break-up and *COD* removal by electrocoagulation, the experimental data were used to explore the *COD* reduction rate equation.

2. Materials and methods

In this experimental work, the baker's yeast wastewater (BYW) solution was collected from the yeast production company, ASTRICO, Al-Rusaifah, Zarqa, Jordan. Values of *COD* concentration, total solid concentration, electrical conductivity, and pH and color of this BYW are given in Table 1. The baker's yeast wastewater solution was filtered using filter paper to remove any suspended solids. The filtered solution was subjected to dilution in order to get stock solution with *COD* of 12 g/L for further use. The *COD* of treated and untreated wastewater was determined according to the standard methods (APHA, 1998).

Table 1. Some chemical/physical characteristics of baker's yeast wastewater used in the present study before treatment

Parameter	***Unit***	***Value***
pH	-	5.1-6.2
COD	g/L	50-60
Total Solids (TS)	g/L	12.9-21.9
Electrical conductivity EC	mS/cm	17.24
Color	-	Dark brown

Fig. 1 shows the laboratory scale electrocoagulation unit designed to have working volume of 600 mL of wastewater solution. Six rectangular iron plates with the dimensions of 45 mm×53 mm×3 mm were aligned vertically inside electrocoagulation unit such that they are parallel to each other. The total effective surface area of these iron electrodes immersed in BYW solution was around $A_{eff} = 247.5$ cm^2. A uniform spacing of 15 mm was fixed between the electrodes. The electrodes were connected to a terminal of an adjustable voltage and direct current (DC) power supply which delivers current of 0-1 A and voltage of 0-30 V. The applied voltage and the direct current were tuned to have current densities in the range, $I_\rho = I/A_{eff} = 8\text{-}40$ A/m^2; where I is the applied DC current in ampere. As shown in Fig. 1, the electrocoagulation unit was immersed in water bath with analogue thermostatic controller to adjust the operating temperature.

During each electrocoagulation experiments, the solution was mixed continuously using mechanical agitator (Stuart Scientific, UK) with mixing speed in the range of MS = 100-600 rpm. In order to study the effect of solution acidity on the *COD* reduction, the pH was regulated using either 0.1 M NaOH or 0.1 M H_2SO_4. The pH was measured using 3151 MWT pH meter. In the parametric study of current density, pH, EC time, and mixing speed, the operating temperature was kept at 40 ± 1 ^{o}C. While in investigating the effect of temperature on the EC performance, the temperature was varied in the range between 20 and 60^{o}C. Dilute HCl was used to wash the electrocoagulation unit between the experimental runs, followed by washing using distilled water. After terminating each run, the treated solution was filtrated using filtration papers (0.45 µm, Millipore, USA). Samples of the filtrate solution were used to measure residual *COD*.

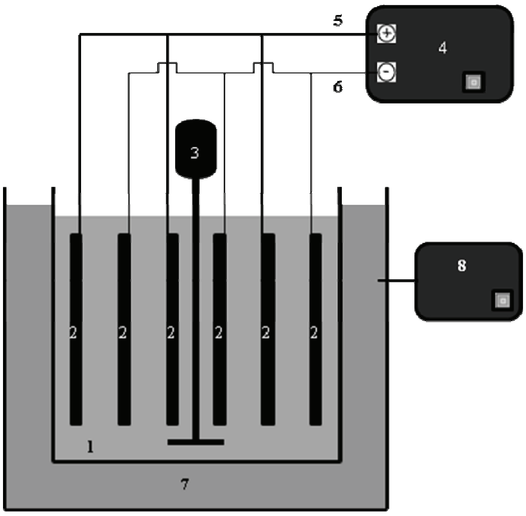


Fig. 1. Experimental setup: (1) Electrocoagulation unit, (2) Iron electrodes, (3) Mechanical stirrer, (4) DC power supply, (5) Anode, (6) Cathode, (7) Water bath, (8) Analogue thermostatic controller

3. Results and discussion

3.1 Effect of pH on the COD reduction of BYW

The solution acidity is considered to be an important parameter that influences the whole performance of electrocoagulation processes (Al-Shannag et al., 2012; Mahesh et al., 2006). In this part of experiments, various values of pH = 3, 4, 5, 6, 7, 7.5, 8 and 9 were tested to assess the effect of solution acidity on the electrocoagulation performance. The applied DC current density, operating temperature, and mixing speed had values of $I_\rho = 32$ A/m^2, T = 40 ^{o}C, MS = 300 rpm, respectively. Fig. 2 depicts the time course of *COD* residual concentration during electro-coagulation of BYW for three values of pH = 3, 7 and 9. The percentage removal of *COD* from BYW by electrocoagulation was estimated using Eq. (3):

$$\%Removal = 100\,\frac{COD_0 - COD}{COD_0} \tag{3}$$

where COD_0 is the initial concentration (g/L) in the BYW stock solution before electrocoagulation process and *COD* is the corresponding concentration (g/L) after terminating the electrocoagulation experiment.

The *COD* concentration and the estimated maximum removal efficiency resulted after 60 minutes EC treatment were plotted against the corresponding pH values as shown in Fig. 2. It is evident from Fig. 2 that the minimum *COD* concentration and the corresponding maximum *COD* removal occur at pH=7. At this pH value, the *COD* value and the % removal were 2.20 g/L and 81.7%, respectively. For strongly acidic (pH=3) and alkaline medium (pH=9), the *COD* values were 5.7 and 4.8 g/L and the corresponding percentage removal efficiencies were 52.5% and 60%, respectively. This indicates that the electrocoagulation of BYW effluent is highly pH-dependent. This result agrees with those of Liang et al. (2009) for the *COD* removal from molasses wastewater and Al-Shannag et al. (2012) for the removal of *COD* from paper industry wastewater. The effect of pH on the EC processes using iron electrode could be attributed to the interaction of the hydronium ion (H_3O^+) with the hydroxyl iron (III) complexes formed at these pH conditions.

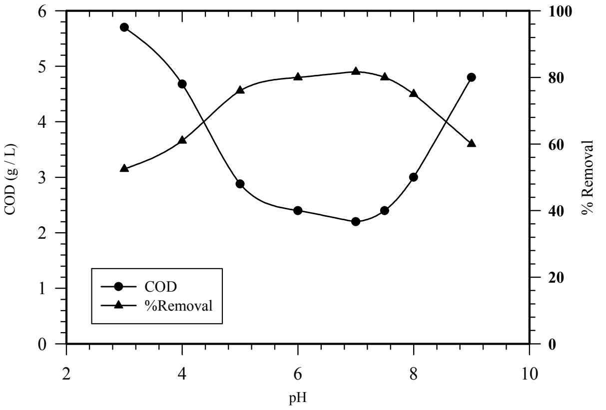


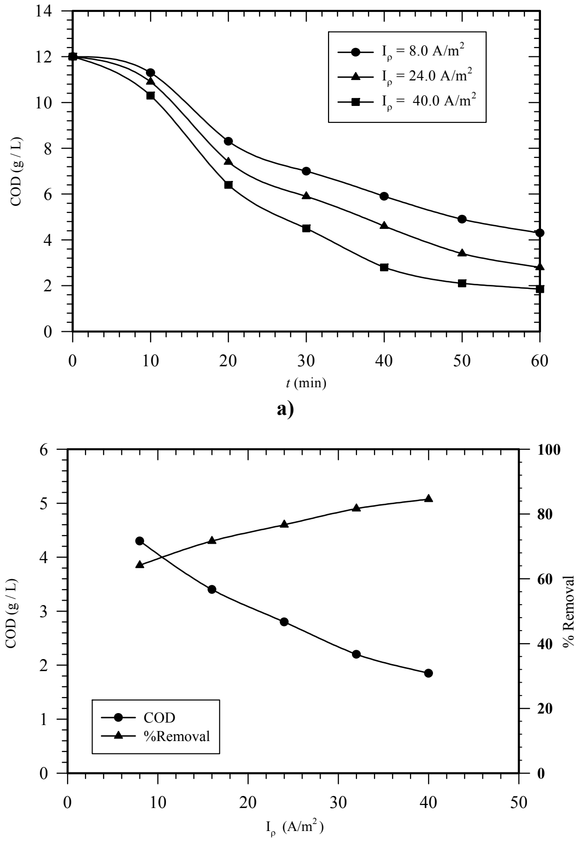
Fig. 2. Effect of pH on the *COD* concentration and its removal percentage after 60 minutes EC treatment of baker's yeast wastewater (COD_0 = 12 g/L, MS= 300 rpm, I_ρ = 32.0 A/m^2, T = 40 ^{o}C)

3.2 Effect of EC time and current density on the COD reduction of BYW

The *COD* removal depends on the concentration of ferrous ions produced at the anode, which in turn depends upon the current density and EC time as described by Faraday's law. Thus, current density (I_ρ) and EC time (t) represent very significant parameters in EC processes. To investigate the current density influence upon the performance of EC, several EC experiments were carried out at different values of current density ranging from I_ρ= 8.0 to 40 A/m^2 , pH=7, MS=300 rpm, and T = 40 ^{o}C. In all EC experiments that considered EC time effect, the current density was fixed at I_ρ= 32 A/m^2, pH = 7, MS = 300 and the EC time was varied from *t* = 10 to 60 min.

The experimental results are shown in Fig. 3. It can be observed in Fig 3(a) that *COD* concentration decreases as EC time increases until approaching around steady state value after about 60 minutes. This is attributed to the fact that the EC time current affects the amount of ferrous ions produced at the anode.

As the duration of EC treatment increases, more iron dissolves from the anode and consequently the available amount of particulate flocs increases to remove more *COD*. Fig. 3(b) shows plots of the *COD* concentration and its percentage removal against the current density resulted from 60 minutes EC treatment. It can be seen in the figure that residual *COD* concentration decreases and the corresponding percentage removal increases as the current density increases.



a)

b)

Fig. 3. Effect of DC current density, I_ρ on the *COD* reduction of baker's yeast wastewater (COD_0 = 12 g/L, MS = 300 rpm, pH = 7, and T = 40 ^{o}C). a) Variations of COD concentrations with EC time at different DC current density; b) Variations of *COD* concentrations and their percentage removal with DC current density after 60 minutes EC treatment

For instance and when the current density increases from I_ρ = 8 to 40A/m^2, the *COD* concentration decreases from 4.3 to 1.85g/L and the corresponding percentage removal efficiency increases from 64.2 to 84.6%. Obviously and similar to the influence of EC time, increasing the DC current increases the release of ferrous ions at the anode that are necessary to produce flocculating agents which lead to more *COD* removal.

However, increasing the DC current may result in an energy waste for heating up the solution inside electrocoagulation unit (Mahesh et al., 2006). For this reason a compromise of EC time and current density should be considered.

3.3 Effect of mixing speed on the COD reduction of BYW

Mixing is a vital parameter in EC process in order to homogenize the coagulants throughout the aqueous phase so that it will destabilize colloidal impurities found in the wastewater. To investigate the effect of mixing speed (MS) on the *COD* reduction of BYW, several experiments were performed at I_ρ= 32.0 A/m^2, pH=7, and T = 40 ^{o}C. The mixing speed was varied from MS = 100-600 rpm. Fig. 4 shows plots of the *COD* concentration and the corresponding percentage removal against mixing speed. At MS = 100 rpm, the measured *COD* concentration was 4.3 g/L whereas the corresponding percentage removal was 64.2%.

Increasing the mixing speed up to MS = 300 rpm, decreases the *COD* concentration and consequently increases the removal efficiency. At this mixing speed, MS = 300, the value of *COD* concentration was 2.20 g/L and its corresponding percentage removal was 81.7%. When the mixing speed increased above a value of MS = 300 rpm, the measured *COD* concentration started to increase. When the MS reached a value of 600 rpm, the *COD* concentration became 3.4 g/L and its corresponding percentage removal was 71.7%. Accordingly, the optimal mixing speed to achieve maximum performance was found to be 300 rpm.

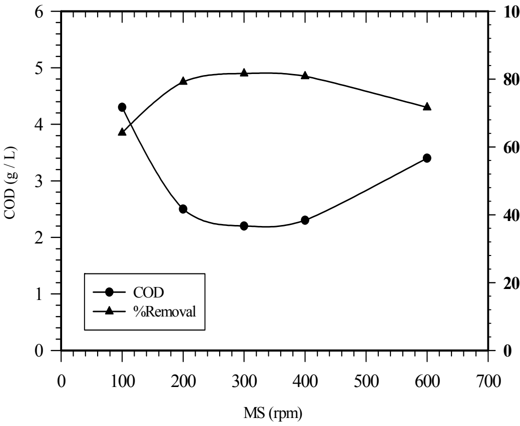


Fig. 4. Effect of mixing speed on the *COD* concentration and its percentage removal for 60 minutes EC treatment of baker's yeast wastewater (COD_0 = 12 g/L, I_ρ = 32.0 A/m^2, pH = 7, and T = 40 ^{o}C)

To explain this behavior, it should be noted that at low mixing speeds below 300 rpm, a non homogenous mixture will result where ferrous ions are over dosed near the anode, causing charge reversal and destabilization of the organics, whereas these ions will be under dosed far away from the anode, resulting in incomplete charge neutralization (Liang et al., 2009). On the other hand, at high mixing speeds like MS = 600 rpm, shear forces will break the flocs which stay in the solution rather than to separate.

3.4. Effect of temperature on COD removal of BYW

In baker's yeast industries, the yeast fermentation production process is conducted at a temperature around 40^{o}C, then the effluents wastewater is normally at the same range of this temperature. In colloidal solutions, an increase in temperature causes a decrease in viscosity, an increase in surface potential and surface charge density, and a decrease in electrical double layer thickness. The surface potential of the particles does not decrease with a corresponding decrease of surface charge (Tari et al., 2000). A decrease in electrical double layer thickness and an increase in surface charge density promote agglomeration of destabilized colloids, while an increase in surface potential increases repulsion between the particles (Sangal et al., 2013).

Fig. 5 shows plots of the *COD* concentration and its percentage removal against temperature for 60 minutes EC treatment at current density, I_ρ = 32.0 A/m^2, pH = 7, and MS = 300 rpm. As can be observed in the Figure, the dependency between *COD* reduction and temperature is not significant which contradicts the more pronounced effect reported by Sangal et al. (2013). This may be due to the fact that electrocoagulation is very complicated process in which the temperature can affect the EC performance in many ways like the kinetics of chemical reactions and the thermodynamics of hydrogen gas bubbles released, in addition to solution physiochemical properties such as the electrical conductivity, the size of colloidal particles and the solubility of metal hydroxides (Phalakornkule et al., 2010).

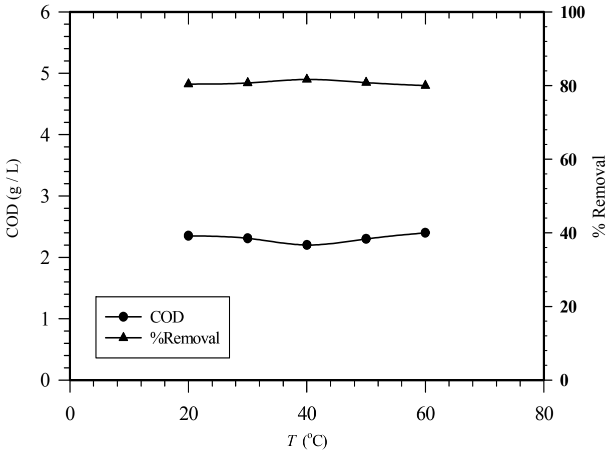


Fig. 5. Effect of operating temperature on the *COD* concentration and its percentage removal for 60 minutes EC treatment of baker's yeast wastewater (COD_0 = 12 g/L, I_ρ = 32.0 A/m^2, pH = 7, and MS = 300 rpm)

3.5. Kinetic modeling of COD reduction of BYW using electrocoagulation

As mentioned above, none of previous studies modeled the kinetic mechanism for the *COD* reduction from BYW. In this context, it should be remembered that the kinetic mechanism for pollutants removal using EC process is quite complicated. In addition, the nature of pollutants and their characteristics are quite different.

Accordingly, the removal steps and consequently the mechanism may differ from pollutant to another. For this reason, the present study used a simplified integral approach in which certain kinetic model is assumed and its ability to resolve the experimental *COD* concentrations will be examined. In order to describe the reduction rate of *COD* from BYW using EC process, the following first-order kinetic model was tested (Eq. 4):

$$-d(COD)/dt = k(COD) \tag{4}$$

where *COD* is the residual *COD* concentration in the wastewater, g/L, *k*, is the first-order rate constant. Integrating Eq. (4) gives the linearized equation (5):

$$ln\left(COD_0/COD\right) = kt \tag{5}$$

In order to check the validity of the first-order model to describe the *COD* reduction by EC process, plots of $ln(COD_0/COD)$ versus EC time at various current densities are plotted according to Fig. 6. The experimental *COD* concentration at each EC time was obtained at temperature, T = 40 °C, mixing speed, MS = 300 rpm, pH = 7 and COD_0 = 12 g/L. It is clear from Fig. 6 that the residual *COD* concentration data fit a straight line for all current densities considered. The corresponding values of kinetic parameter, *k*, obtained from the slope of each plot, and the squared correlation coefficient, R^2, are shown in Table 2.

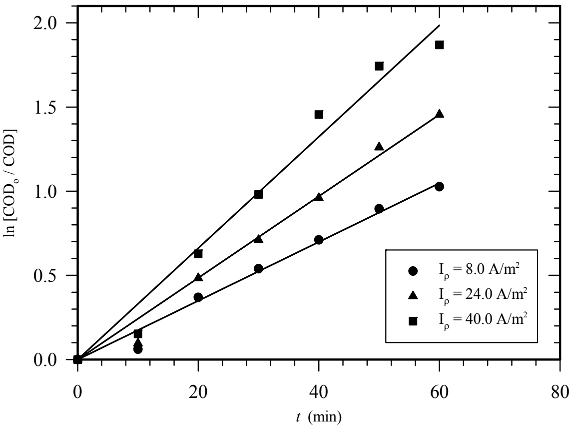


Fig. 6. Linearized first-order kinetic model at different DC current density, $I_ρ$, for EC treatment of baker's yeast wastewater (COD_0 = 12 g/L, T = 40.0 °C, pH = 7, and MS= 300 rpm)

It is clear that all values of R^2 are closed to one. This indicates the goodness of the first-order kinetics to describe the *COD* reduction adequately. In addition, Table 2 shows that the kinetic parameter, *k*, increases as the current density increases.

Table 2. Values of first-order constant, *k*, and squared correlation coefficient, R^2, at various values of current density, $I_ρ$, for EC treatment of baker's yeast wastewater (COD_0 = 12 g/L, T = 40.0 °C, pH = 7, and MS = 300 rpm)

$I_ρ$ (A/m²)	*k* (min⁻¹)	R^2 (-)
8.0	0.017	0.9842
16.0	0.022	0.9872
24.0	0.024	0.9869
32.0	0.029	0.9910
40.0	0.033	0.9788

To explore the relation between the kinetic parameter and DC current density, the *k* values were plotted against $I_ρ$ according to Fig. 7. As can be demonstrated in the Fig. 7, the power law relation fits the *k* parameter as function of current density parameter in excellent manner. This is also confirmed by the value of corresponding squared correlation coefficient which is around unity.

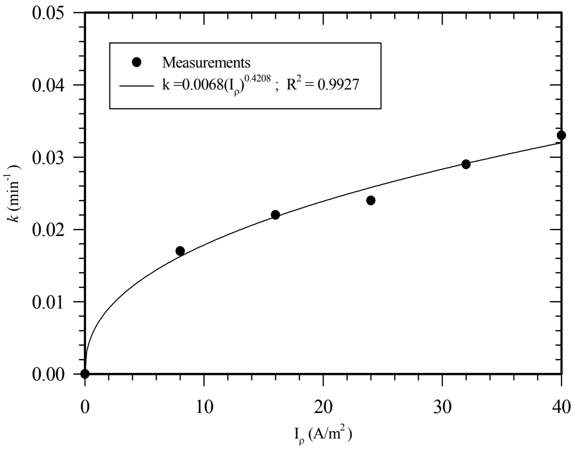


Fig. 7. Variation of first-order kinetic constant, *k*, with DC current density, $I_ρ$, for EC treatment of baker's yeast wastewater (COD_0 = 12 g/L, T = 40.0 °C, pH = 7, and MS = 300 rpm)

Thus, the rate of *COD* reduction from BYW at pH=7 and mixing speed of 300 rpm, can be written as given by Eq. (6):

$$-d(COD)/dt = 0.0068(I_ρ)^{0.4208}\,COD \tag{6}$$

3.6 Energy and electrode consumptions for electrocoagulation treatment of BYW

The electrical energy consumption in kWh per kg *COD* (E) removed from BYW can be estimated using Eq. (7):

$$E = (V)(I)(t)/[60(v)(COD_0 - COD)] \tag{7}$$

where *V* is the applied voltage (Volt), I is the DC current (A), t is the EC operating time (min), and v is the volume (L) of the treated BYW.

On the other hand, the amount of iron electrode consumed per kg *COD* (M_{Fe}) removed from BYW can be assessed using Faraday's law:

$$M_{Fe} = \frac{60(I)(t)(M\ M)}{[(z)(F)\ \left(COD_0 - COD\left(v\right)\right)]} \qquad (8)$$

where *MM* is the molecular mass of iron (55.845 g/mol), z is the charge of Fe (z=2) and F is the Faraday's constant (96500 C/mol).

Table 3 summarizes the percentage *COD* removal and consumption amounts of electrical energy and iron electrode per kg of *COD* removed from baker's yeast wastewater at different current densities and electrocoagulation time. It is clear in the Table 3 that increasing EC time and/or current densities enhances the *COD* removal efficiency which is accompanied by higher consumption levels of both electrical energy and electrodes. The percentage *COD* removal reached 38.3% when the EC treatment conducted for 20 minutes at current density of 24 A/m^2.

Table 3. Amounts of consumed iron electrodes (M_{Fe}) and electrical energy (E) per kg *COD* removed and percentage *COD* removal at various EC time and current densities

EC time (min)	I_p ***(A/m^2)***	***E (kWh/kg COD removed)***	M_{Fe} ***(kg Fe consumed/ kg COD removed)***	***COD %Removal***
20	24	1.304	0.075	38.3
20	40	2.976	0.103	46.7
30	24	1.475	0.085	50.8
30	40	3.333	0.116	62.5
60	24	1.957	0.113	76.7
60	40	4.926	0.171	84.6

The corresponding consumptions of electrical energy and iron electrodes per kg *COD* removed were 1.304 kWh and 0.075 kg iron, respectively. Increasing the EC treatment time to 60 minutes and current density to 40 A/m^2 gave higher removal levels, percentage *COD* removal was 84.6%, which was accompanied by a consumption of 4.926 kWh and 0.171 kg iron per kg *COD* removed, for electrical energy and iron electrode, respectively.

4. Conclusions

The present study investigated batch electrocoagulation (EC) with iron electrodes as a simple process to treat baker's yeast wastewater containing high *COD* load. The experimental results showed that the *COD* removal efficiency increases significantly with increasing both EC time and DC current and the maximum *COD* reduction achieved at a mixing speed of around 300 rpm and pH of 7 (neutral medium).

Unfortunately, increasing EC time and/or DC current increases both the electrical energy and electrode consumption levels. However, electrocoagulation process can be operated effectively with EC time and current density of no more than 60 minutes and 40 A/m^2, respectively. Finally, the present work reported a first-order *COD* reduction rate with power-law dependency of its kinetic parameter upon the DC current density. These important kinetic results, together with the optimum operating conditions, are so helpful in the design of industrial scale batch or continuous electrocoagulation units to treat wastewater effluents resulted from baker's yeast industries.

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