Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2013.800268

52 (2014) 3047–3053
April

Taylor & Francis
Taylor & Francis Group

The effect of initial pH on treatment of poultry slaughterhouse wastewater by electrocoagulation method

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Received 18 November 2012; Accepted 18 April 2013

ABSTRACT

In this work, treatment of poultry slaughterhouse wastewater by electrocoagulation method with aluminum plate electrodes has been investigated. System was operated in batch and galvonostatic mode. Effect of initial pH of the wastewater was predominantly investigated on chemical oxygen demand (COD) removal efficiency but turbidity and oil–grease removal efficiencies were also evaluated. Operational parameters were kept constant at 150 rpm of stirring speed, 1 mA/cm^2 of current density, and 293 K of solution temperature. On the other hand, initial pH of the wastewater was in the range of 2–8. Initial pH of the wastewater was an effective parameter on COD removal efficiency as it is expected. When initial pH of the wastewater was in the range from 3 to 4, it was possible to obtain higher COD removal rates. In this study, when the initial pH of wastewater equals to 3 and current density is 1 mA/cm^2, system gives COD removal efficiency of 85% in 20 min. Meanwhile, system consumes 2.14 kWh/m^3 of energy under above conditions.

Keywords: Poultry wastewater; Electrocoagulation; Aluminum plate electrode

1. Introduction

Turkish poultry industry has grown substantially over the last two decades. The consumption of poultry products in Turkey, which constitutes a significant part of meat consumption, has steadily increased, reaching about 10 kg/capita in 2003 [1]. Poultry slaughterhouses produce significant volumes of wastewater during the slaughtering process and periodic washing of residual particles. Even though slaughterhouse wastewaters have different composition according to the industrial process and water demand for a slaughtered poultry, these wastewaters contain high levels of organics such as biochemical oxygen demand (BOD) and chemical oxygen demand (COD), nitrogen, and phosphorus due to the presence of organic materials such as blood, fat, grease, and proteins [2]. Environmental preservation efforts and developments in the technology have resulted in stringent discharge standards. With environmental regulations becoming more stringent, regulatory compliance has also become a matter of increasing concern to the poultry industries. To meet strict laws on environmental protection, pollutant loads discharged from the poultry industries should be first reduced to a significant extent, and a proper post-

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treatment (polishing) step should be further applied to improve the quality of the final discharge in terms of residual pollutant content. Several treatment methods have been reported for the treatment of poultry slaughterhouse wastewater (PSW) [3]. Biological processes such as aerobic or anaerobic systems are widely used for the PSW treatment [2,4–6]. Even though biological processes are effective and economical, long hydraulic retention time and large area requirements (i.e. large volume of the bioreactor) make sometimes these processes less attractive than physicochemical treatments, which require shorter retention time [7].

Electrocoagulation (EC) is an electrolytic process consisting of the dissolution of sacrificial anodes out of iron or aluminum upon the application of a current between two electrodes for the treatment of liquid wastewater containing inorganic or organic matter. The generated metallic ions, i.e. $Al^{3+}_{(aq)}$ and $Fe^{3+}_{(aq)}$, will undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides. These hydroxide/polyhydroxide/polyhydroxymetallic compounds have a strong affinity with dispersed/dissolved molecules to cause coagulation/adsorption [8].

Electrochemical methods for the treatment of wastewater have recently attracted great attention. These methods were found to be effective in the treatment of wastewater from dye production [9–11], chromium containing wastewater [12], iron containing drinking water [13], phosphate containing wastewater [14], electroplating wastewater [15], metal finishing effluents [16], textile wastewater [17], and boron wastewater [18,19]. Electrode materials most widely used in EC process are aluminum and iron. In the case of aluminum, main reactions are as follows;

$$\text{Anode: } Al_{(S)} \rightarrow Al^{3+}_{(aq)} + 3e^- \quad (1)$$

$$\text{Cathode: } 3H_2O + 3e^- \rightarrow 3/2H_{2(g)} + 3OH^-_{(aq)} \quad (2)$$

On the other hand, at high pH values, both cathode and anode may be chemically attacked by OH^- ions:

$$2Al_{(S)} + 6H_2O + 2OH^-_{(aq)} \rightarrow 2Al(OH)_4^- + 3H_{2(g)} \quad (3)$$

$Al^{3+}$and OH^- ions are generated by electrode reactions (1) and (2) react to form various monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al(OH)_2^{2+}$, $Al(OH)_4^-$, and polymeric species such as $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$, and $Al_{13}(OH)_{34}^{5+}$, which transform finally into $Al(OH)_{3(s)}$ according to complex precipitation kinetics [20].

$$Al^{3+}_{(aq)} + 3H_2O \rightarrow Al(OH)_{3(S)} + 3H^+_{(aq)} \quad (4)$$

Freshly formed amorphous $Al(OH)_{3(s)}$ "sweep flocs" have large surface areas, which are beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. Finally, these flocs are removed easily from aqueous medium by sedimentation or H_2 flotation.

2. Materials and methods

2.1. PSW effluent

Wastewater used in this work was obtained from a local poultry slaughterhouse plant with a daily processing capacity of 20,000 chickens, located in the city of Erzincan (Turkey), producing approximately 450 tons of wastewater daily. The wastewater emerging from various operations such as chicken cutting, scalding, defeathering, eviscerating, chilling, packing, and plant clean-up is filtered using a screen filter to remove hair and solids and then collected in an equalization tank. The chemical analysis of PSW is given in Table 1.

The pH of the wastewater was adjusted to the required value with concentrated nitric acid and sodium hydroxide. All the chemicals were of analytical grade and supplied by Merck. The oil–grease analysis was carried out by infrared determination (Wilksir HATRT-2) according to EPA Method 1664. Analysis of the COD was conducted by closed reflux method according to Standard Methods for Water and Wastewater Analyses [21].Turbidity was measured by turbidity meter (Orbeco-Helliege).

2.2. Experimental setup

A laboratory-scale reactor, made of Plexiglas, was used in all experiments. The experimental setup is shown in Fig. 1. Two groups of alternating electrodes being cathodes and anodes (by five plates of each type) made of aluminum were arranged vertically.

Table 1
The characterization of PSW

Parameters	Values
Specific conductivity (μs/cm)	2,858
BOD_5 (mg/L)	1,123
Turbidity (NTU)	176.6
pH	6.73
COD (mg/L)	2,171
PO_4-P (mg/L)	9.65
Oil–grease (mg/L)	143.1
TSS (mg/L)	750
TKN (mgN/L)	148
The flow of wastewater (tons/day)	450

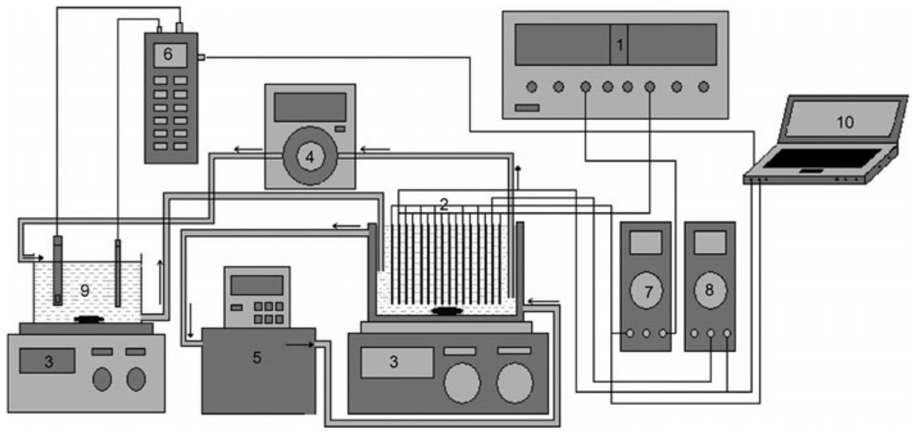

Fig. 1. Schematic view of the experimental system ((1) DC power supply, (2) EC cell, (3) Magnetic stirrer, (4) Pump, (5) Circulator, (6) pH and conductivity meter, (7) Ampermeter, (8) Voltmeter and (9) pH control unit).

The net spacing between the aluminum electrodes was 5 mm. Aluminum electrodes had 1,400 cm^2 of effective surface. They were connected to terminals of a direct current power supply characterized by the ranges 0–5 A for current and 0–30 V for voltage. At the beginning of each run, the PSW of the certain concentration and pH fed into the reactor. Each run was timed starting with the DC power supply switching on. Two digital multimeters (Brymen Bm 201) as ampermeter and voltmeter were used to measure the current passing through the circuit and the applied potential, respectively. The EC unit has been stirred at 150 rpm by a magnetic stirrer (Heidolp MR 3,004 S). The thermostat electrocoagulator is made of Plexiglas with the volume of 850 mL. During the experiments, temperature, conductivity, and pH of the wastewaters were measured by a multiparameter (WTW Multiline P-4 F-Set-3). The experimental system ran under current densities of 0.5, 1.0, 1.5, and 2.0 mA/cm^2. Treated wastewater was collected over a desired period of time (0, 5, 10, 15, 20, 25, 30, 40, 50, and 60 min) from the reactor and collected samples were filtered by the cellulose acetate membrane filter with the pore diameter of 0.45 μm before the analysis. Reactor was operated in batch and galvanostatic modes. In order to provide the constant pH conditions, system has been operated in recirculation mode. In these test runs, experimental setup shown in Fig. 1 was modified by attaching a peristaltic pump, magnetic stirrer (Heidolp MR 3,004 S), and tank. The pH of the system was kept constant by adding acid or base to the tank.

3. Results and discussion

3.1. The effect of initial pH on COD removal

The effect of initial pH on COD removal from wastewater using aluminum plate electrodes by EC was investigated. The initial pH is one of the important factors affecting the performance of electrochemical process. It has been established that the pH has a considerable influence on the performance of EC process. In this study, initial pH values were in the range of 2–8, while the effect of initial pH on COD removal was investigated. During the experiments, current density, stirring speed, reaction time, and solution temperature were kept constant at 1 mA/cm^2, 150 rpm, 60 min, and 293 K, respectively. The authors also examined operating parameters such as current density and stirring speed in a previous study [22]. Optimum values obtained in that study for current density and stirring speed were 1 mA/cm^2 and 150 rpm, respectively, as in the present study. Relationship between initial and final pH values can be seen in Fig. 2. Before the experiment, turbidity, COD, temperature, electrical conductivity, and pH values of wastewater were recorded. As can be seen in Fig. 2, the final pH values always increased at all initial pH values. While system pH rapidly rose in low initial pH, this increase was slower in higher initial pH values. This tendency is accordance with the literature [11,14,17,23,24]. Increase in the wastewater pH was derived from hydrogen evolution at the cathode (see reaction (2)).

Fig. 3 presents the effect of initial pH on COD removal efficiency. When considering Fig. 3, it can be seen that the highest removal efficiencies were obtained when initial pH values were equal to 3, 4, and 6. The pH values giving the highest removal efficiency rates are important for the determination of the applicability of the system. However, whether this pH value is optimum or not cannot be determined. Similar results were obtained by Chen et al. [25] and Kobya et al. [26].

The reason for this is the fact that optimum pH must also provide the lowest energy consumption

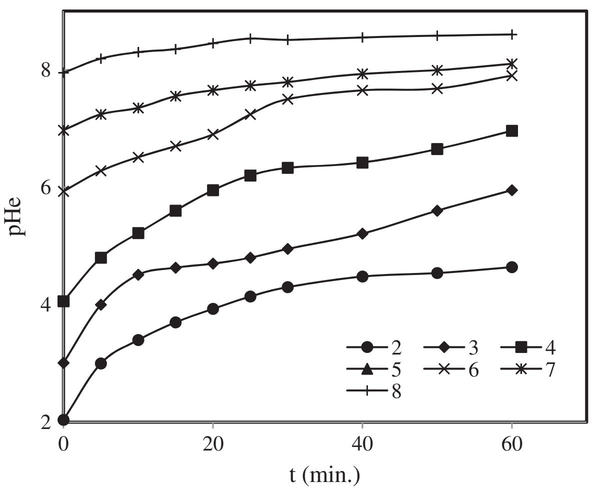

Fig. 2. The change of different initial pH during reaction time.

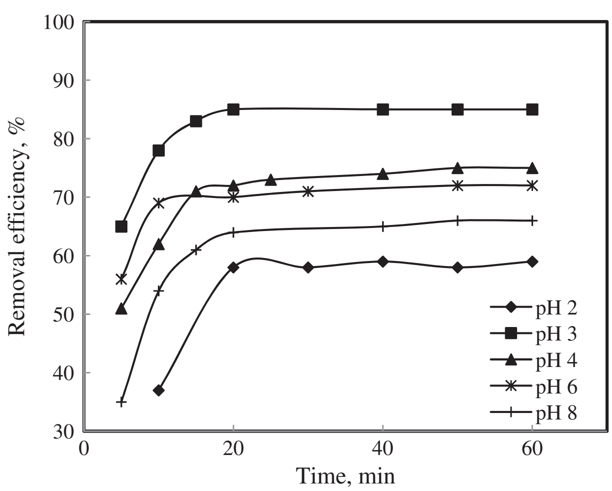

Fig. 3. The effect of initial pH on COD removal as function of reaction time (pH 2–8) (CD = 1 mA/cm^2, 150 rpm, 60 min and 293 K).

which is important for operational cost. At the beginning of the experiments, removal efficiencies increased rapidly and then it did not behave so in spite of the gradual increase in treatment time and energy consumption.

This is a general tendency and actually this period should be considered when the system is operated, i.e. the time interval when the plateau begins to form. One of the most important parameters (perhaps the most important one), which can determine the applicability of treatment technique is the operational cost of the system. Therefore, while providing the cost effectiveness it should be reminded that system should be operated at the critical points in order for the system to provide cost effectiveness. From this point of view, critical thresholds are 15th, 15th, and 10th minutes for the pH values of 3, 4, and 6, respectively. At these retention times, the system provided the COD removal efficiencies of 83, 71 and 69%, respectively.

Fig. 4 represents the effects of the initial pH on turbidity and oil–grease removal. As can be seen from the figure, initial pH levels between 3 and 5 are the most effective in the turbidity removal of the system. The results obtained are compatible with the literature [26].

From Fig. 4, which represents the oil–grease removal of the system based on initial pH values, it can be stated that lower pH values are more effective in the removal of oil–grease. Floating effect of the hydrogen gas released form cathode is the basis of oil–grease removal. However, effects of other mechanism on this process should be investigated. Based on the mentioned fact, energy consumption levels of the system under the conditions given in Fig. 3 are given in Fig. 5.

It can be seen from Fig. 5 that the highest energy consumption is obtained at the initial pH of 2 followed by 4. As mentioned above, in order to provide cost effectiveness, the initial conditions which offer high removal efficiency and low energy consumption should be preferred. However, the initial conditions providing both of these requirements are not prevalent. In such situations, different parameters should also be considered in order to make a decision and this decision should be based on whether the targeted removal efficiency or cost effectiveness is more important.

Although the most advantageous initial pH seems to be 8 according to Fig. 5, initial pH of 3 is more suitable if removal efficiency is considered. However, in order to make this evaluation, parameters of charge loading (Q) and space–time efficiency should also be considered. Figs. 6 and 7 represent the trend in removal efficiency depending on the charge loading

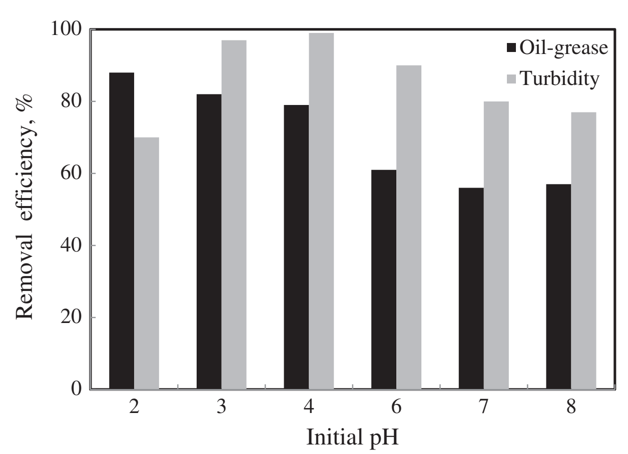

Fig. 4. The effect of initial pH on turbidity and oil–grease removal efficiency (CD = 1.0 mA/cm^2, n = 150 rpm, 30 min).

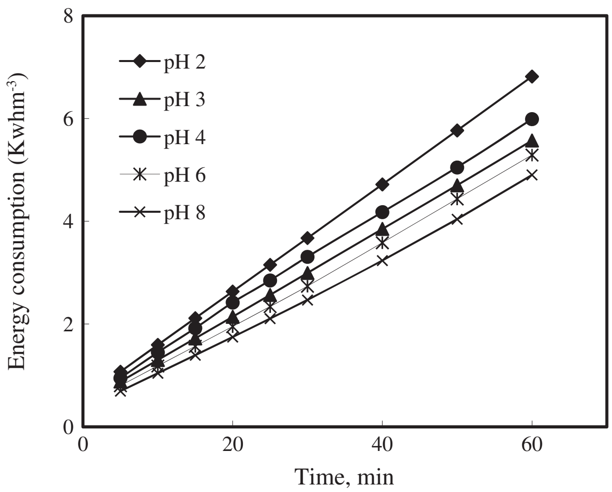

Fig. 5. The effect of initial pH on energy consumption ($CD = 1.0\,mA/cm^2$, $n = 150$ rpm, 60 min).

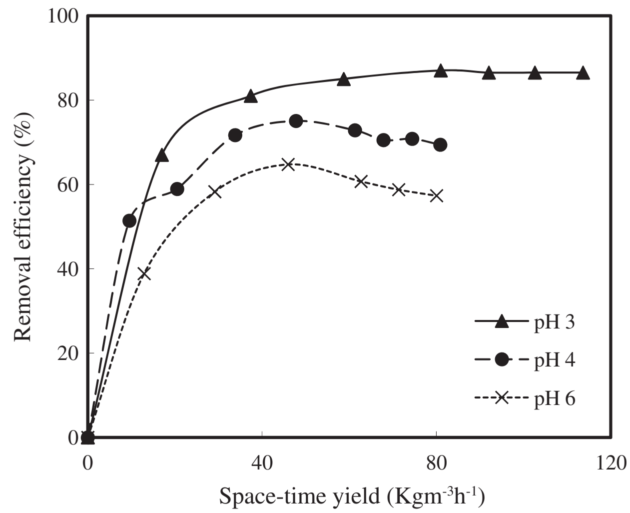

Fig. 7. Variation of removal efficiency vs. space–time efficiency ($CD = 1.0\,mA/cm^2$, $n = 150$ rpm).

and space–time efficiency at initial pH values of 3, 4 and 6, respectively.

When Figs. 6 and 7 are taken into consideration together, it can be stated that the best results can be obtained at the initial pH values of 3, 4, and 6. If Fig. 6 is evaluated in details, curves for charge loading are optimum at the value of $20\,Fm^{-3}$with the pH values of 3, 4. and 6. Similarly, in Fig. 7, space–time efficiency values are found to be 37.38, 33.81, and $29.14\,kgm^{-3}h^{-1}$, respectively, for the same pH values.

In the light of these evaluations, optimum initial pH may be said to be 3 based either on COD or energy consumption.

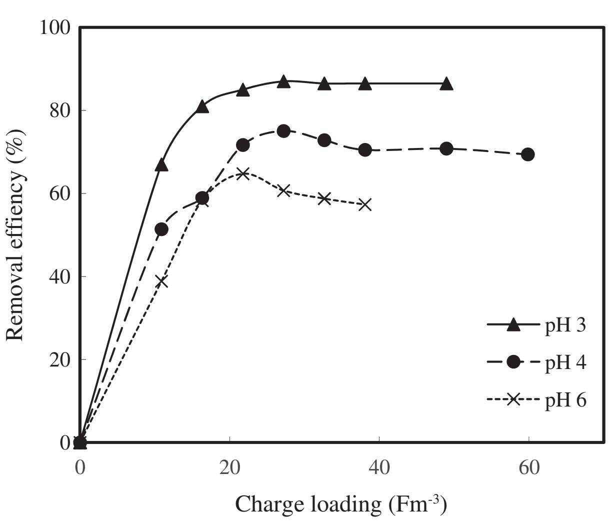

Fig. 6. Variation of removal efficiency vs. charge loading ($CD = 1.0\,mA/cm^2$, $n = 150$ rpm) 6.

Trend in pH profile is given in Fig. 2, which can show that pH of wastewater increases gradually even though the rate of increase in pH differs. The same trend of system was observed in previous studies and this situation was found to result from the following reactions according to Chen et al. [24].

$$Al \rightarrow Al^{3+} + 3e^- \quad (5)$$

$$Al^{3+}_{(aq)} + 3H_2O \rightarrow Al(OH)_3 + 3H^+_{(aq)} \quad (6)$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \quad (7)$$

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (8)$$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (9)$$

$$HOCl \rightarrow H^+ + OCl^- \quad (10)$$

If removal curves of these three values are remembered (Fig. 3), it may be stated that removal rate can become stable once nearly 20 min and then increases very slightly. At the end of the mentioned time interval, pH of the medium is nearly 4.70, 6.00, and 6.75, respectively.

It may be reasonable to obtain high removal efficiency at mentioned pH levels when considered the fact that $Al(OH)_3$ crystals form at an efficient rate beginning from the point where pH is 4 [27]. These crystals have minimum resolution in the pH range from 6.5 to 7.8 [28]. Removal of oil–grease in the system (Fig. 4) is considerably high and therefore constraining factor in the system is thought to be COD removal. In this respect, only the COD removal is taken into consideration as the performance criteria from this part onwards.

3.2. The effect of constant pH on COD removal

The effect of constant pH on COD removal efficiency is shown in Fig. 8. The experiments were conducted at a mixing rate of 150 rpm and current density of 1.0 mA/cm^2. It can be seen from Fig. 8 that the lowest removal efficiency is obtained at pH value of 3. When pH of the wastewater is lower than 4, reaction (4) does not form aluminum hydroxide crystals at a reasonable rate.

Therefore, Al^{3+} ions remain in hydrated forms in the environment even though coagulants are added to the medium and removal mechanism is the compression of electrical double layer. By considering this mechanism, COD removal is considered to remain lower at this pH.

Fig. 9 represents the tendency of system turbidity as a function of reaction time. It was seen that the main problem was at the turbidity correction when the system was operated at this constant pH mode. The reason for this problematic condition may be that the circulatory pump destroyed the flocks resulting in mud with weak subsiding characteristics, which adversely affected turbidity.

Events seen in Fig. 9 and a reactor combination with three zones occur in the experiments carried out over the typical EC cells. Stable flock layer formed in the upper part of the reactor by the floating effect plays an important role for the separation of solid and fluid. However, effect of circulatory pump delays the formation of this layer.

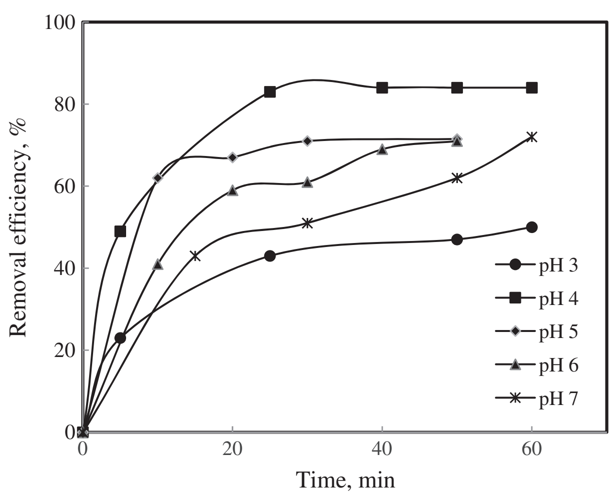

Fig. 8. The effect of initial pH on COD removal as function of reaction time (CD = 1.0 mA/cm^2, n = 150 rpm).

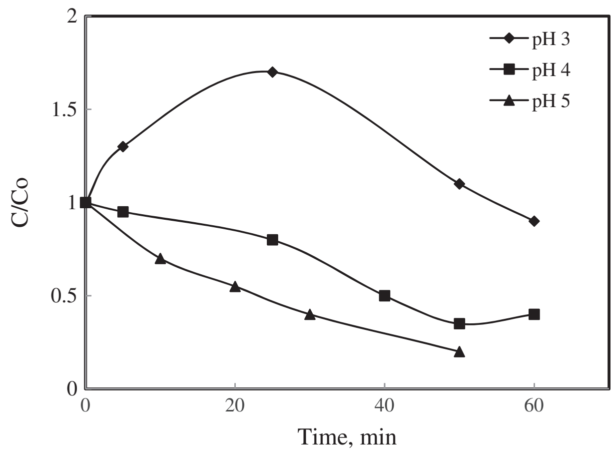

Fig. 9. The effect of controlled pH on turbidity removal efficiency (CD = 1.0 mA/cm^2, n = 150 rpm).

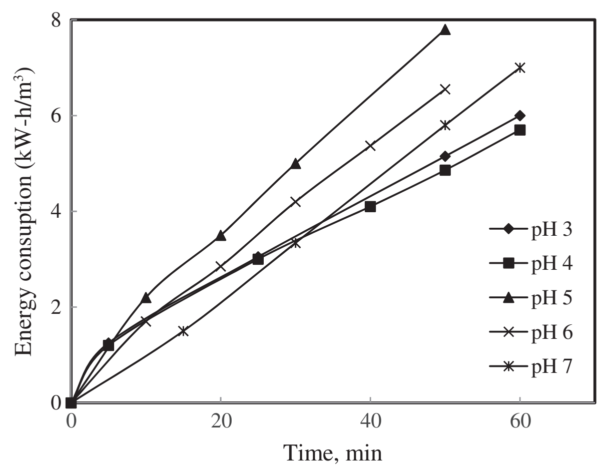

Fig. 10. The effect of controlled pH on energy consumption (CD = 1.0 mA/cm^2, n = 150 rpm).

Fig. 10 represents energy consumption data. As seen in Fig. 10, the lowest energy consumptions were obtained at pH 3 at the end of reaction time. However, when energy consumption values in Fig. 10 and removal efficiency in Fig. 8 are considered together, it can be stated that the condition where pH of wastewater is 4, is the most suitable condition.

4. Conclusion

As expected, the initial pH value is a considerable effective parameter in the treatment of wastewater from poultry processing facilities using EC. The optimum initial pH can be said to be 3 in the investigated range. However, when COD removal at the initial pH is considered, the range from 3 to 4 can produce suitable results. Therefore, the system has flexibility for pH parameter, since it can suitably work in a pH range rather than at only a value. On the other hand, suitable pH range is 3–5 for the turbidity while for the maximum oil and grease removal, initial pH is 2. At this pH level, either COD or turbidity showed a steep decrease. Therefore, when the system is evaluated for three mentioned removals, the most suitable range is between 3 and 4. In this range, the system consumed power nearly 3.0–3.5 kWh/m^3.

It was seen in constant pH studies that COD removal of the system did not increase significantly and turbidity was affected adversely. The reason for such behavior might have been caused from the fact that the peristaltic pump destroyed the flocks resulting in metal hydroxide sludge with weak settling characteristics. Based on these implications, it may be thought that a constant pH system requires additional devices such as a circulation pump and pH control unit and therefore, it may not provide enough benefits.

Evaluations mentioned above are based heavily on the COD removal efficiency of the system. However, it should be remembered that evaluation of the system considering its turbidity and oil–grease removal may reveal similar tendencies. Consequently, it can be stated that treatment of wastewater from poultry processing facilities using EC method is a suitable system at the initial stage.

Acknowledgment

Authors are grateful to the Scientific and Technological Research Council of Turkey for providing financial support with grant no. ÇAYDAG/105Y043.

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