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# TREATMENT OF DAIRY WASTEWATER BY ELECTROCOAGULATION AND ULTRASONIC-ASSISTED ELECTROCOAGULATION METHODS

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## Abstract

The application of electrocoagulation and ultrasonic-assisted electrocoagulation technologies for the treatment of dairy wastewater was addressed in the current work. The study highlights the effect of the key parameters such as applied current, ultrasound power amplitude, electrolyte concentration of NaCl, and wastewater concentration on the process removal efficiency. The energy consumption and the consequent operation cost were evaluated in order to assess the economic feasibility of the process. The results demonstrated that the increase of applied current from 0.1 to 0.5 A decreases the electrolysis time that is required to obtain the maximum removal efficiency of 99 % by a factor of 6.2. Consequently, the operation cost of the process was duplicated. The results cast a new light on the ultrasonic-assisted electrocoagulation of dairy wastewater. It is indicated that the removal efficiency of ultrasonic-assisted electrocoagulation was higher than that obtained with electrocoagulation for the first 20 minutes of the process, thereafter the removal efficiency of electrocoagulation was superior. Utilizing of the ultrasonic waves added a massive charge to the process operation cost. Furthermore, increasing the level of solution salinity has a positive effect on both of removal efficiency and cost of operation. This was demonstrated by increasing the electrolyte concentration of NaCl from 0.06 to 4 g/L which resulted in the redaction of the electrolysis time required to reach the maximum removal efficiency to the half. This reduced cost of operation by around 73%. The rate of removal was distinctly decreased when the concentration of the artificially made dairy wastewater increased.

*Keywords:* dairy wastewater, electrocoagulation, ultrasound waves

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## 1. Introduction

Dairy industries have experienced a globally substantial growth in the last few years in many countries around the world, that due to the increasing of demand for milk and its sub-products such as cheese, butter, yogurt, ayran, cream, ice cream, milk powder, and other milk-based food productions. According to the Food and Agriculture Organization of the United Nations (FAO) report in 2019, the production rate of milk was around 843 million tons and it is higher than the production rate in 2017 by approximately 2.2% (FAO, 2019).

Water is an essential material in the dairy industry, as it is involved in all stages of manufacturing even in packing process. Consequently, it is also expected that the aqueous waste from this industry will also be significant. Dairy wastewater contains different pollutants. Draining this water without actually treating it into rivers poses serious problems (Aitbara et al., 2016).

Typically, physico-chemical and biological methods were employed in order to treat the dairy wastewater. Several methods can be listed as biological treatments included activated sludge process, sequencing batch reactor, trickling filters,

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anaerobic filters, aerated lagoons and anaerobic sludge blanket reactors, beside other aerobic and anaerobic processes (Charalambous et al., 2020; Daverey and Pakshirajan, 2016; Karadag et al., 2015; Matsumoto et al., 2012; Suman et al., 2018). The most common concern with using the biological processes is the requirement for a large working area, long duration time of treatment, generating of excessive amounts of sludge, and may require additional treatments for the produced effluent (Bazrafshan et al., 2013; Kushwaha et al., 2010). The physicochemical methods on the other hand, comprise different approaches such as adsorption, coagulation-flocculation, adsorption, coagulation-flocculation, member filtration process which involved nanofiltration and reverse osmosis techniques (Ayeche, 2012; Kumar et al., 2016; Kushwaha et al., 2010; Murcia et al., 2018).

Recently, electrocoagulation technology (EC) has been successfully applied to treat wide ranges of wastewater from several industries including textile wastewater (Ayeche, 2012; Bener et al., 2019; Liu et al., 2019), soluble oils (Liu et al., 2019), heavy metal ions (Kim et al., 2020) and paper wastewater (Wagle et al., 2020).

Usually, aluminum or iron serves as a sacrificial electrode in the electrocoagulation process. Applying of a particular potential from a power source on the electrocoagulation cell, resulted in directly dissolving of the anode material by the action of oxidation reactions, thereafter, $Al^{3+}(aq)$ ions are generated when aluminum is employed as an anode material in the process. The generated ions spontaneously react to produce the corresponding hydroxides and/or polyhydroxides according to the following reactions Eqs.(1-4) (Alyaqoobi, 2010; Gao et al., 2010):

Anode:

$$Al \rightarrow Al^{3+} + 3e^{-} \quad (1)$$

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

In solution:

$$Al^{3+} + 3H_2O \leftrightarrow Al(OH)_3 + 3H^{+} \quad (3)$$

Cathode:

$$2H_2O + 2e^{-} \rightarrow H_2 + 2OH^{-} \quad (4)$$

The $Al(OH)_3$ constitutes by the reaction of $Al^{3+}$ and $OH^{-}$ ions, has a sufficient surface area that facilitates rapid adsorption of pollutant compounds and considered as an efficient and effective coagulant. The generated flocs can be easily removed from the aqueous medium by floatation induced by $H_2$ and $O_2$ microbubbles generated on the electrodes, and by sedimentation due to the density difference (Abdelwahab et al., 2009; Emamjomeh and Sivakumar, 2009; Gao et al., 2010; Mollah et al., 2001).

Furthermore, an additional technology like ultrasound waves could be introduced to the electrochemical systems to enhance the performance of the process. The modification mechanism occurs via constant regeneration of new electrode surfaces due to the effect of cavitation and/or microstreaming. The mass transfer of species can be also improved by the present of ultrasound waves (Esclapez et al., 2010; Wang et al., 2009; 2015). In addition, high oxidation power species ($OH^{\bullet}$) is produced through the process due to the pyrolysis of water vapor at the surface of cavitation bubbles.

The present work was conducted to investigate the efficiency of the dairy wastewater treatment by the electrocoagulation process and the effect of ultrasound waves in improving the performance of this process. To our knowledge, no prior studies have examined the sono-electrocoagulation method for the treatment of dairy wastewater. For this purpose, artificial solutions were prepared by dissolution of powder milk. The present study elucidates the effects of key parameters such as ultrasonic power, applied current, concentration of electrolyte, and milk concentration on the removal efficiency. The energy consumptions and process operation cost are evaluated to underline the economic feasibility of the process.

## 2. Material and methods

The experiments were carried out using laboratory set-up as presented in Fig. 1.

**Fig. 1.** The experimental setup

A cylindrical reactor made from Plexiglas was used to conduct the experimental work. The diameter of the reactor was 9 cm and its height was 10.5 cm. A plate of aluminum with dimensions of 9.5 cm × 6 cm × 1 cm was used as anode and a spiral aluminum electrode was used as a cathode for entire experiments. The cathode was coiled around the anode and the distance between them was fixed as 15 mm.

The spiral electrode was designed to improve the efficiency of the flotation process, and to decrease energy consumption (Baierle et al., 2015). The anode was connected to a positive pole, while the cathode was connected to the negative pole of the DC power supply (Smart power system, model EMA-01-32-15-P). All the experiments were carried out at a constant current mode, and the applied current was altered in a range of 0.1 – 0.5 A.

All experiments were performed with 500 ml of milk solution with different concentration. The synthetic wastewater was prepared by mixing different concentrations of milk powder of 600, 1200, and 2400 mg/L and with different concentration of NaCl between 0.06 and 4 g/L.

The reactor was constantly stirred at speed of 100 rpm by a magnetic stirrer (Corning, model PC-410). The ultrasound waves were introduced to the system by using digital sonifier (Branson, model-450), and the maximum power 450 watts, with an adjustable amplitude. The tip of the ultrasonic probe was immersed in the solution for 7 cm from the liquid surface.

The samples were periodically withdrawn at a distance of 5 cm from the liquid level. The turbidity of the suspension was measured using a turbidity unit (Lovibond, Turbidirect). The efficiency turbidity removal was calculated according to the following formula (Eq. 5).

$$Removal\ efficiency\ (\%) = (T_o - T_t/T_o)\ \times 100 \quad (5)$$

where: $T_o$ is the initial turbidity and $T_t$ is the turbidity at time (t).

The energy consumption per unit volume was obtained from the relation shown in (Eq. 6) (Brillas and Martínez-Huitle, 2015). The amount of the dissolved aluminum in the electrocoagulation process was calculated using Faraday's Law which can be expressed by the relationship as illustrated in (Eq. 7). The estimations for the energy consumption and amount of aluminum dissolved were calculated based on a removal efficiency of 99%.

$$Energy\ consumption\ (KWh/m^3)\ = E\ I\ t/V_r \quad (6)$$

where: $E$ is the cell voltage (V), $I$ is the applied current (A), $t$ is the electrolysis time (h) and $V_r$ is the solution volume ($m^3$).

$$w = tIM/nFV_r \quad (7)$$

where: $w$ is the amount of the dissolved aluminum ($g/m^3$), $t$ is the time (s), $M$ is the molecular weight of Al (27 g/mol), $n$ is the number of electrons that involve in the oxidation reaction ($n$ = 3), $F$ is Faraday's constant (96500 C/mol).

## 3. Results and discussion

### *3.1. Effect of applied current on process efficiency*

The applied current plays an important role in an electrochemical process. In the current study, the effect of applied current was explored in a range of 0.1 – 0.5 A. The NaCl electrolyte was selected as a minimum concentration of 0.06 g/L gave an electrolytic conductivity of 123 µS. The variation of the removal efficiency with time for each applied current is as presented in Fig. 2.

The results of Fig. 2 demonstrate that the removal efficiency increases with the applied current. At a low current value of 0.1 A, the removal efficiency does not considerably change for the first 20 minutes of operation. Thereafter, the removal efficiency increases sharply until it reaches its maximum value of approximately 99% in 10 minutes. However, using high currents of 0.2, 0.3 and 0.5 stimulates the augmentation of the efficiency and reached to the maximum efficiency after less than 15, 11 and 5 minutes respectively.

According to Faraday's law, increasing of the applied current results in producing more $Al^{3+}$ ions in the aqueous solution that is generated from the oxidation reaction of the anode material. The $Al^{3+}$ ions transform through the bulk into soluble monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al(OH)_3$ and $Al(OH)_4^-$ in an acidic medium and $Al(OH)_4^-$ constituting in an alkaline medium.

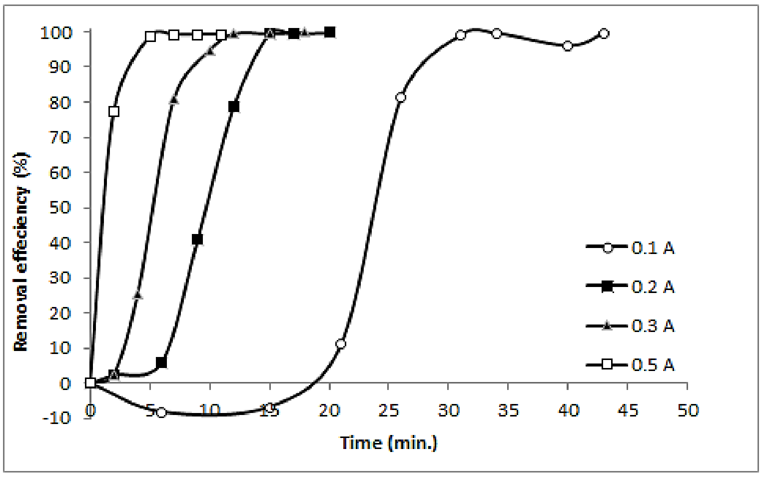


**Fig. 2.** Effect of applied current on removal efficiency

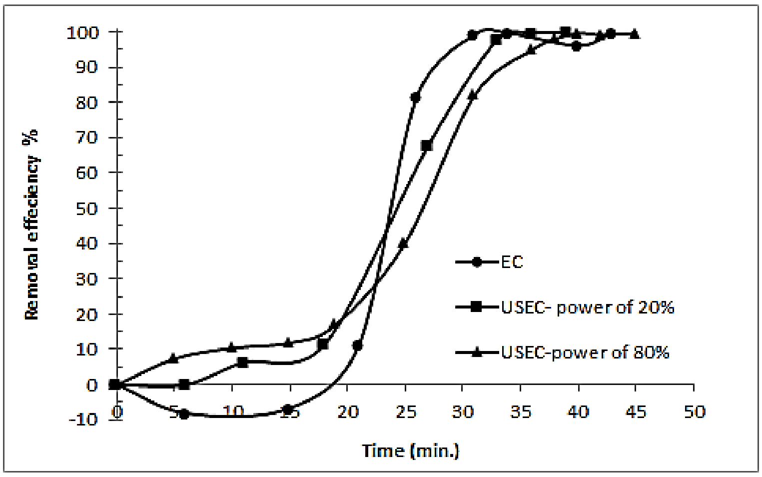


**Fig. 3.** The electrocoagulation process with and without the sonication effect

In addition, polymeric species can be developed by a different configurations such as $Al_2(OH)_2^{4+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$ and $Al_{13}(OH)_{34}^{5+}$ (Brillas and Martínez-Huitle, 2015).

The initial pH of the solution was (6.9 ± 0.1) and it was observed that the pH of the solution increased gradually with the increased of applied current and electrolysis time. In practice, the applied current of 0.1A augments the final pH magnitude to around 7.8, while the final pH value of the solution was around 8.3 when the applied current altered to 0.3 A. This could be attributed to the accumulation of $OH^-$ in the solution. This is consistent with the above of oxidation and reduction equations (Eqs. 1 and 4), and it is agreed with the stoichiometric molar ratio of $Al^{3+}$ and $OH^-$ of 1/3. This behavior was also observed in many studies of electrocoagulation processes (Can et al., 2003; Gao et al., 2010; Koparal et al., 2008).

In addition, amplifying the applied current increases the generation rates of $H_2$ and $O_2$ bubbles. This enhances phase separation by flotation due which enhances phase separation by flotation where the potential of the collision between the rising bubble and particles-coagulant floc increases. According to many authors (Holt et al., 2002; Khosla et al., 1991), the size of bubbles that are formed on both electrodes reduces with the elevation of the applied current. Generating of small bubbles leads to high surface area to volume ratio, together with a long resident time and speed of mixing, this stimulates the ability of bubbles in improving the efficiency of the separation (Kim et al., 2013; Zimmerman et al., 2011).

*3.2. Effect of sonication*

The combination effect of the ultrasonic and electrocoagulation was investigated by using the same electrochemical cell as constructed in Fig. 3. The experimental conditions of our investigation to study these effects were constant as 0.1 A for the applied current, the mixture concentration of 1200 mg /l, and electrolyte (NaCl) concentration of 0.06 g/L. Two amplitudes of 20% and 80% of the maximum ultrasonic power were supplied to the process. The dependency of the removal efficiency on time for both electrocoagulation (EC) and the ultrasonic-assisted electrocoagulation (USEC) modes are presented in Fig. 3.

The results of Fig. 3 depict that the removal efficiencies of USEC for both ultrasonic supplied power amplitudes (i.e., 20% and 80%) exhibited two distinct stages. For the first 22 minutes of operation, the removal efficiencies of (USEC) were higher than that achieved with the only (EC) for both ultrasonic supplied power amplitudes. The inflection point is located in the electrolysis time between 21-22 minutes. After this period, the removal efficiencies of USEC is obviously lower than that of EC. It can be observed that at electrolyte time of 26 minutes, the removal efficiency of the USEC of 20% power amplitude reduces by about 17% from that obtained with using only EC. Increasing the ultrasonic power amplitude to 80% results in reducing the removal efficiency by around 50% of that achieved with EC.

The remarkable difference between the removal efficiencies of USEC and EC could be interpreted to cleaning of the electrode surface by cavitation/micro-streaming effect which occurs via applying ultrasound. The microbubbles that generated due to the sonication effect improve the flotation of floc and can destroyed the restart layer build upon or near the electrode. This eventually increase the removal efficiency of the (USEC) process. However, the decrease of the removal efficiency in the USEC system after 21 minutes is probably due to an adverse function of the ultrasound. This effect appears through a dramatic elevation in the temperature of the mixture. The final temperature of the mixture reaches to around 80 ºC when the ultrasonic power amplitude is 80% which is due to the high temperature of the core and the surface of cavitation bubbles (Asgharian et al., 2017). It has been reported that the rising of temperature may alter the chemical and physical properties of milk (Hlaváč and Božiková, 2011; Šestan et al., 2016). Accordingly, the turbidity of the solution is highly expected to change as well. Another effect of increasing the temperature is that the rate of formation of the dissolved electrode ions increases while the probabilities of formation aluminum hydroxide reduced (Aitbara et al., 2016; Kabdaşlı et al., 2012). Furthermore, it has been reported that the ultrasound has some negative impact on the aluminum hydroxide colloids where some of the pollutants that are adsorbed on the aluminum hydroxide can easily desorb and return to the solution (Wang et al., 2009). This unfavorable phenomena may be due to the effect of acoustic waves on colloidal hydroxides and damage the adsorption layer at the surface of the colloidal particle (Al-Rubaiey and Al-Barazanjy, 2018).

Some researchers stated that the technique of ultrasonic waves improve the efficiency of electrocoagulation process when it applies for treatment of car-washing wastewater (Chu et al., 2012), removal of dyes (He et al., 2016), and humic acid from wastewater (Asgharian et al., 2017). However, the combined of ultrasonic and electrocoagulation has a negative effect on removal efficiency for bentonite suspended wastewater and oily wastewater (Al-Rubaiey and Al-Barazanjy, 2018).

### *3.3. Effect of NaCl electrolyte concentration*

To study the effect of NaCl concentration on the removal efficiency, we examined different concentrations of 0.06, 2, 3 and 4 g/L. Fig. 4 shows the variation of the removal efficiency with time with adding different concentration of NaCl electrolyte to the milk solution. The results indicate that the increase of NaCl concentration improves the removal efficiency. With electrolyte concentration of 4 g/L, the required electrolysis time to reach the maximum removal efficiency of 99% is about 15 minutes, while using a concentration of 2 g/L takes more time of around 26 minutes of electrolysis time to accomplish the same removal efficiency.

The presence of $Cl^-$ ions in the electrolysis system play effective roles in enhancement of the electrocoagulation process. Chlorides ions hinder the reaction of calcium ions that are contained in wastewater with $HCO_3^-$. The main drawback of this reaction is the formation of an insulating layer that precipitates on the electrode surface. This layer increases the overall resistance of the electrochemical cell and inhibits the formation of $Al^{3+}$ (Chen, 2004). Furthermore, the presence of $Cl^-$ ions in the solution excites the de-passivating action of the oxide layer that is formed on the electrode surface, consequently increases the dissolved aluminum hydroxide in the solution (Golder et al., 2007; Mohammed, 2008).

In addition, the molecular chlorine could be hydrolysed to form hypochlorous acid and hypochlorite ions. These chlorine derivatives are powerful situ-formed oxidizing agents (Kabdaşlı et al., 2012). On top of that, the chloride ions can be oxidized at the anode surface forming chlorine gas ($Cl_2$) bubbles. This could eventually accelerate the flotation process and improve the separation of the flocs.

### *3.4. Concentration of milk*

It's well known that the dairy industry result in various concentration and composition of wastewater streams depends the type of products and the manufacturing processes. Thus, different milk concentration was used to investigate how this affects the removal efficiency. The effect of the variation of milk concentrations were studied in range of 600, 1200 and 2400 mg/L. The NaCl concentration was constant as 0.006 g/L and the applied current was fixed at 0.1 A for all experiments. Fig. 5 presents the alteration of the removal efficiency with time for solutions of different milk concentrations.

From Fig. 5, it is clearly to note that the increase in milk concentration decreases the removal efficiency. The removal efficiency was only around 45% after 47 minutes of electrolysis with milk concentration of 2400 mg/L, while it needs only 25 minutes for the removal efficiency to reach around 99% when the concentration decreases to 600 mg/L.

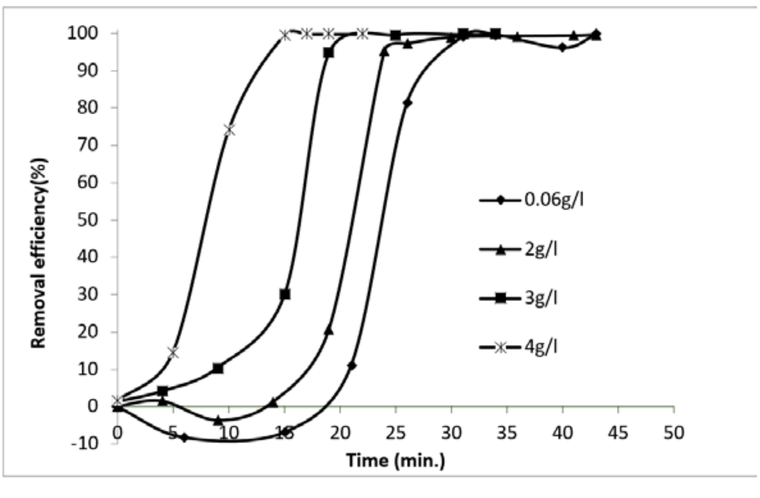


**Fig. 4.** The effect of electrolytes concentration on the removal efficiency

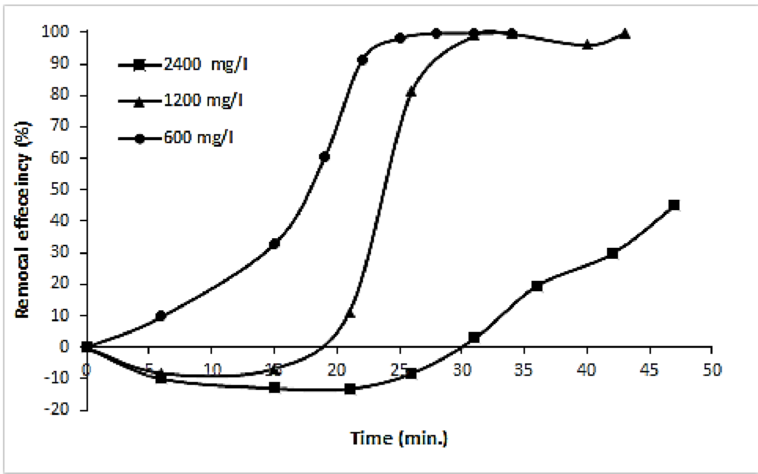


**Fig. 5.** Effect of milk concentration on the removal efficiency

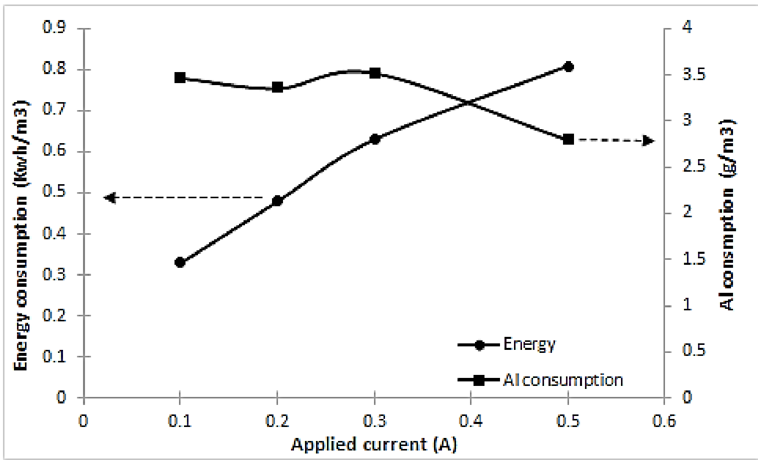


**Fig. 6.** Energy consumption and aluminum consumption with different applied current at maximum removal efficiency

According to Faraday's law, the amount of aluminum oxidized that is presented in the solution increases with the increase of electrolysis time. A relatively high concentration of milk in the solution such as 2400 mg/L requires high electrolysis time to produced aluminum oxidized that is adequate to generate the coagulant floc. In the same prospect, the increase the milk concentration to high values could impede the electrocoagulation process even when a large amount of floc presents in the solution (Tchamango et al., 2010).

### *3.4. Operating cost analysis*

It is essential to assist the economic feasibility of the process in order to obtain a comprehensive comparison between the electrocoagulation approach and other technologies. The operation cost of the process can be estimated based on the most effected parameters which are the cost of electrode material consumed, the required electrical energy, and chemicals consumed during the process.

Therefore, the energy consumption and the amount of dissolved aluminum were determined with different operation conditions. Fig. 6 shows that energy consumption increases from about 0.33 kWh/m$^3$ to about 0.8 kWh/m$^3$ when the applied current increases from 0.1 A to 0.5 A. Furthermore, rising of the applied current results in increasing of the dissolved aluminum. Bear in mind that the time used to estimate the consumed aluminum in (Eq. 7) is the time required to achieve 99% removal efficiency which can be recognized from Fig. 2. The applied current of 0.1 A, requires about 31 minutes to produces 3.46 g/m$^3$ of the dissolved aluminum to achieved 99% removal efficiency. Increasing of the applied current to 0.5 A makes the time of 5 minutes enough to produce 2.79 g/m$^3$ of the dissolved aluminum to achieving the same efficiency. This reflects a liner relation between the consumption of electrodes material and the applied current.

In order to estimate the operation cost of the process, the aluminum local price of 1.93$/Kg and the electrical energy cost of 0.05 $/kWh are considered. Fig. 7 shows the effect of the change in applied current on the operation cost. The results demonstrated that the operation cost increases with the increase of applied current and it is considerably affected by the small changes of applied current. The maximum operation cost of around 0.045 US$/m$^3$ is obtained at applied current of 0.5 A, and it drops to 0.023 US$/m$^3$ by applying a lower current of 0.1 A.

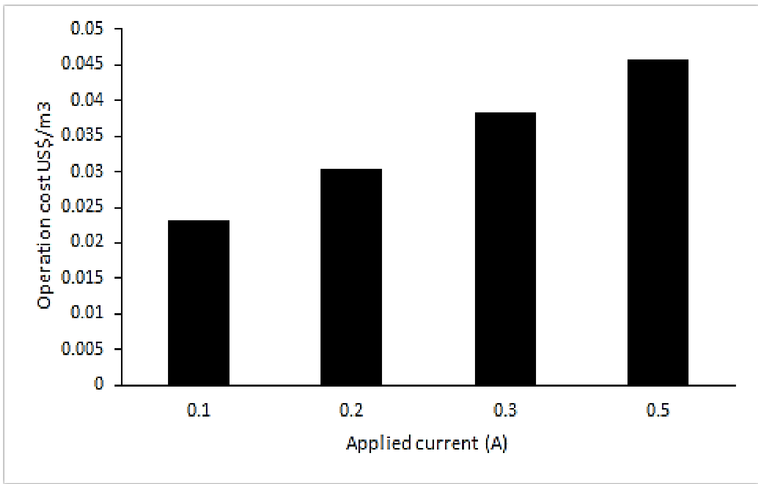


**Fig. 7.** Operation cost with different applied current

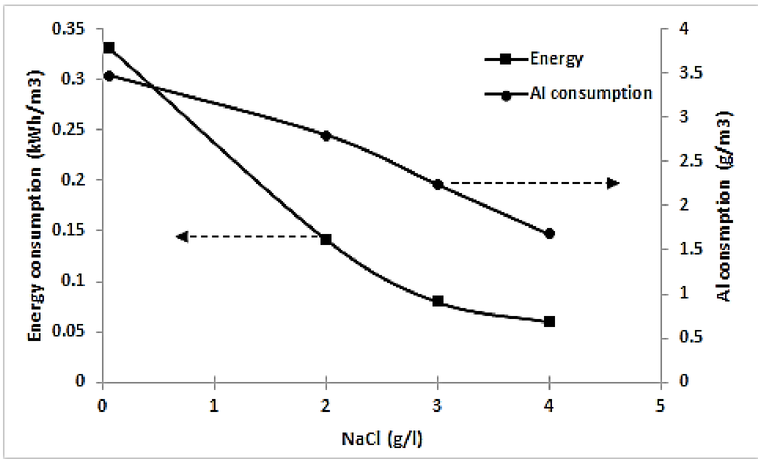


**Fig. 8.** Energy consumption and dissolved aluminum at different NaCl concentration

Further, economic evaluation of the process was achieved by quantifying the consumed of energy and aluminum electrode as functions of NaCl concentration. Fig. 8 illustrates the effect of electrolyte concentration of NaCl on these two parameters. It can be observed that the energy consumption dramatically decreases with the increase of NaCl concentration in the solution. It is about 0.33 kWh/m$^3$ when the NaCl concentration is 0.06 g/L, and it sharply decreases to 0.14 kWh/m$^3$ as the NaCl concentration rises to 2 g/L. Thereafter it gradually decreases until it reached to 0.06 kWh/m$^3$ at the highest NaCl concentration of 4 g/L. This can be attributed to the presence of NaCl in the solution which improves the conductivity of the solution and consequently reduces the voltage of the electrochemical cell. The trend of aluminum consumption was similar to that of energy consumption. The results elucidate that aluminum consumption reduces through increasing of the electrolyte concentration. By using NaCl concentration of 0.06 g/L, about 3.46 g/m$^3$ of the aluminum anode is required to be dissolved which needs around 31 minutes to achieve the maximum removal efficiency of 99%. In order to achieve the same efficiency by utilizing NaCl concentration of 4 g/L, the aluminum consumption becomes 1.67 g/m$^3$ within 15 minutes.

The operation cost in salinity dairy effluents is also estimated as a function of electrolyte concentration as shown in Fig. 9.

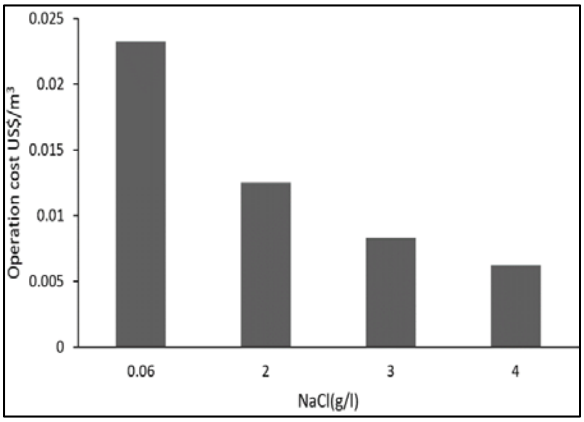


**Fig. 9.** Operation cost with different electrolyte concentration

The increase of electrolyte concentration from 0.06 to 4 g/L results in the decrease of the operation cost by around 73%. The calculated values of the operation cost in Fig. 9 is included the costs of energy and the consumed aluminum. The level of salinity of industrial dairy effluents is depended on the type of the product and manufacturing process. The inorganic salts such as NaCl and KCl, deliberately add during the manufacturing process which rise the concentration of $Cl^-$ ions in in the dairies wastewater to around 0.8 – 1 g/L (Kolev Slavov, 2017). Even higher ranges of salinity can be found in dairy wastewater comparable to that found in seawater desalination and other industries effluents where a large quantity of brine is produced (Chen et al., 2019).

Otherwise, the cost of adding NaCl to the electrocoagulation process should be considered in the cost estimations if the NaCl existing in the wastewater stream isn't taken into account.

Other factors can add more expenses to the cost of the process. Introducing the ultrasonic waves to the electrocoagulation process add an extra charge to the cost of the process, where with an ultrasonic power of 20% increases the operation cost to 9.01 US$/m$^3$. This cost dramatically raises to 22.8 US$/m$^3$ when we increase the ultrasonic power to 80%. That cost of operation for USEC process was the evaluation for 99% removal efficiency, which is significantly higher than that of EC process when both are evaluated in the same conditions.

## 4. Conclusions

The treatment of artificial dairy wastewater by the electrocoagulation process (EC) and ultrasonic assistance electrocoagulation process (USEC) was investigated in the current study. The removal efficiency was evaluated based on the effect of applied current, electrolyte concentration of NaCl dairy concentration and ultrasonic power amplitudes. The energy consumption and operation cost were evaluated to demonstrate the economic feasibility of the process. The results showed that the increase of applied current resulted in improving the removal efficiency which is combined with a dramatic increase in the energy consumption and ultimately the cost of production. The lowest cost of operation was 0.023 US$/m$^3$ at applied current of 0.1A, which increased to 0.045US$/m$^3$ with the increase of applied current to 0.4A. The effect of the ultrasound wave was fluctuated for the low and high ultrasonic power amplitudes, and the cost of operation was remarkably higher than that obtained only by (EC) process. Furthermore, increasing the level of salinity leads to improve both removal efficiency and cost of operation. The operation cost minimized from 0.023 US$/m$^3$ to around 0.006 US$/m$^3$ when the electrolyte concentration of NaCl augmented from 0.06 g/L to 0.4 g/L. Increasing the milk concentration resulted in an increase of electrolysis time required to reach the highest removal efficiency of 99 %.

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