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*Full Length Research Paper*

# Expulsion of polyaromatic hydrocarbons from waste water by electrocoagulation

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**In the present study, two important groups of factors were studied; the first one corresponds to the Electrocoagulation (EC) process parameters such as current density, pH, electrolysis time and electrolyte concentration that were investigated in terms of their effects on the removal efficiencies of polyaromatic hydrocarbons, specially β-naphthols. The second group of factors analyses the effect of three different anode geometries on the percentage removal of β-naphthols. The present study achieved the following results: The optimum conditions for the removal of β-naphthols for cell No. 1 and 2 were achieved at current density 20 mA/cm$^2$, initial pH 7 treated volumes 1.5 L, NaCl concentration 1 g/L and temperature 25°C. Meanwhile, the optimum conditions for cell No. 3 achieved at 106 mA/cm$^2$, initial pH = 7, NaCl concentration = 1 g/L and temperature 25°C, treated volume = 0.5 L. The maximum separation efficiency was achieved for cell No. 1 and the lowest for cell No. 2.**

**Key words:** PolyaromaticHydrocarbons, β-naphthols, electrocoagulation, waste water treatment.

## INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are composed of “wo or more fused aromatic (benzene) rings”. However, “fused aromatic rings” is probably the best definition (Do and Chen, 1994). PAHs are to a certain degree resistant to biodegradation and are sometimes included in a class of persistent organic pollutants (POPs) (Sengil et al., 2004). PAHs can be present in both particulate and gaseous phases, depending upon their volatility. Light molecular weight PAHs (LMW PAHs) that have two or three aromatic rings are emitted in the gaseous phase, while high molecular weight PAHs (HMW PAHs), with five or more rings are emitted in the particulate phase (Aakinson et al., 1990), - and -naphtols can be considered as the derivatives of naphthalene which is the simplest polycyclic aromatic hydrocarbon (PAH).

PAHs can be formed from both natural and anthropogenic sources, though the anthropogenic sources contribute most to the hazards associated with PAHs (Marian, 2010; Baek et al., 1991). They are usually found as a mixture containing two or more of these compounds and commercially available pure PAHs are usually colourless, white or pale yellow - green solids which are odorless or have a faintly pleasant odor (Busetti et al., 2006). Some PAHs are used in medicines, dyes, plastics and pesticides. Others are contained in

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*Corresponding Authors. Email: prof.halima@mwtr.edu.fr asphalt used in road construction as well as found in substances such as crude oil, coal, coal tar pitch, creosote and roofing tar (ATSDR, 1995). Due to the widespread release of PAHs which consequently results in considerable health and environmental hazards (Nazimek and Ćwikła, 2004), the European Union as well as the United States Environmental Protection Agency (USEPA) have specified permissible limits for the 16 priority PAHs (Fang et al., 2002).

The major sources of PAH emissions may be divided into four classes: stationary sources including domestic (Chen et al., 2007; Ravindra and Sokh, 2008; Fabbri and Vassura, 2006) and industrial sources (Gerhard et al., 2009; Fan et al., 1995), mobile emission (Guo et al., 2003), agriculture activities (Biswas and Lazarescu, 1991; Lin and Peng, 1994), and natural sources (Pakpahan et al., 2009; Lui et al., 2001). There are different methods for the separation of PAHS: the thermal treatment of polycyclic aromatic hydrocarbons under controlled conditions is one of the more effective methods of degradation (Eriksson et al., 2003; Summers and Snoeyink, 1999; Rajeshwar et al., 1994; Golder et al., 2007), adsorption or sorption (Arlette et al., 2012; Drogui et al., 2007; Ebubekir et al., 2013), direct photolysis (Khandegar and Anil, 2013), and electrocoagulation (Pons et al., 2005; Modirshahla et al., 2008; Emamjomeh and Sivakumar, 2009; Mohd Salleh et al., 2011; Kliaugaitė et al., 2013).

Electrocoagulation has the potential to extensively eliminate the disadvantages of the classical treatment techniques. Moreover, the mechanisms of EC are yet to be clearly understood and there has been very little consideration of the factors that influence the effective removal of ionic species, particularly metal ions from wastewater by this technique. The mechanism of coagulation has been the subject of continual review (Modirshahla et al., 2008; El-Ashtoukhy et al., 2013). The theory of EC has been discussed by many authors (Daneshvar et al., 2006).

## ELECTROCOAGULATION THEORY

Electrocoagulation (EC) is a technology in which the coagulant is generated *in situ* by oxidation of a metal anode material when applying electrical current (Golder et al., 1994). As well known, $Al^{3+}$ ions dissolve and combine with hydroxyl ions in water (Daneshvar et al., 2003), when direct current passes through the Al anodes, they form metal hydroxides which are partly soluble in the water under definite pH values.

Additionally, electrolytic reactions evolve gas (usually as hydrogen bubbles) at the cathode that can enhance the process; this effect is known as electroflotation which results in better removal of contaminants (Linares-Hernández et al., 2009; Sengil, 2008). Electrocoagulation has occurred in three steps. In first step, coagulant has formed because of oxidation of anode. In second step, pollutants have destabilized. In last step, destabilized matters have united (Kobya and Delipinar, 2008). The most common electrode materials for electrocoagulation are aluminum and iron. They are cheap, readily available, and proven effective (Un and Aytac, 2013). When aluminum is used as electrode material, the reactions are as follows:

At the anode: $Al \rightarrow Al^{3+} + 3e$
(Equation 1)

At the cathode: $2H_2O + 2e^- \rightarrow H_{2\,(g)} + 2OH^-$
(Equation 2)

When the anode potential is sufficiently high, secondary reactions may occur, especially oxygen evolution (Equation 3):

$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$
(Equation 3)

Aluminum ions ($Al^{3+}$) produced by electrolytic dissolution of the anode (Equation 1) immediately undergoes spontaneous hydrolysis reactions which generate various monomeric species according to the following sequence (Pulkka et al., 2014):

$Al^{3+} + H_2O \rightarrow Al(OH)_2^+ + H^+$
(Equation 4)

$Al(OH)_2^+ + H_2O \rightarrow Al(OH)_2^+ + H^+$
(Equation 5)

$Al(OH)_2^+ + H_2O \rightarrow Al(OH)_3 + H^+$
(Equation 6)

The overall reaction in the solution:

$Al^{3+}_{(aq)} + 3H_2O \rightarrow Al(OH)_3 + 3H^+_{(aq)}$
(Equation 7)

EC process like other treatment methods has some advantages and disadvantages. Main disadvantages of this method are lack of a systematic approach to EC reactor design and operation, replacement of electrodes at regular intervals, high cost of electricity and anode passivation (Behbahani et al., 2011).

On the other hand, main advantages of EC are: the simplicity of the equipment (Linares-Hernandez et al., 2009), no need of additional chemical matter after or before treatment, relatively low area demand, also sludge from this process is intensive and has low water (El-Naas et al., 2009).

For these reasons the present work is focused on the EC

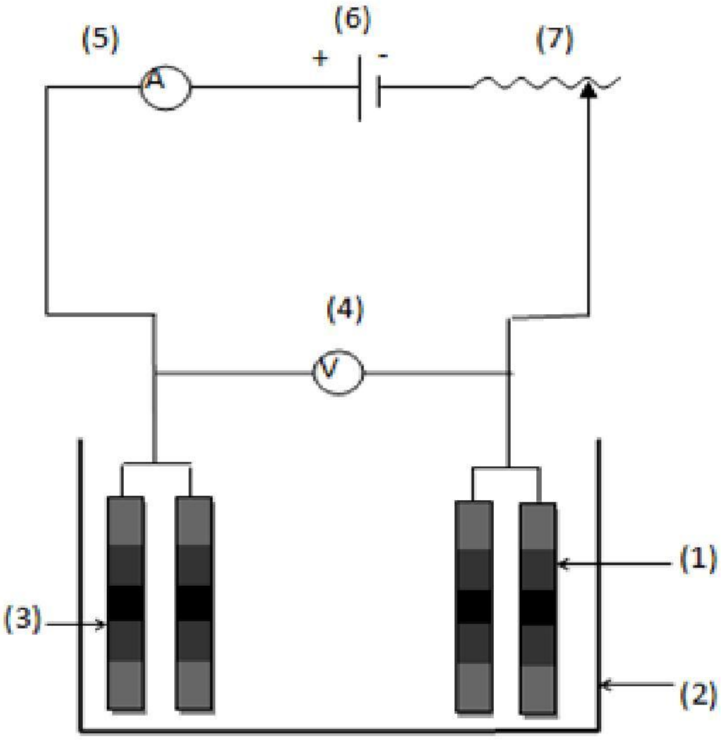

**Figure 1.** A schematically. Diagram for Cell No. 1 (1) Al anode, (2) Plexiglas stank, (3) Al cathode, (4) Voltammeter, (5) Ampere meter, (6) Power supply and (7) Resistance.

processes using a sacrificial aluminum anode to remove PAH from waste water by studying variables like current density, initial concentration, time, temperature, pH, and electrolyte concentration (NaCL). The objective of this study is to remove β-naphthol which is very dangerous to people and environmental. The removal technique done in this work is electrocoagulation method. The main objective is to examine the different parameters which affect the PAH removal. These parameters are the initial concentration of the solution, the pH of the solution, the contact time and the current density.

## EXPERIMENTAL PART

### Experimental set-up

The three experimental set-ups used in the present work are schematically shown in Figures 1, 2 and 3. The EC Cell No. 1 consisted mainly of a rectangular vessel made of plexi glass with dimensions 10 × 10 $cm^2$ base and a height of 20 cm (Figure 1). A two perforated aluminum sheets (10 × 20 cm) which placed vertically in the vessel and operate as an anode, the anode-cathode distance was kept at 4 cm. The other cell (No. 2) made of the same rectangular vessel with the same dimensions, but with different anode geometry which consists of two smooth rods of aluminum sheets (10 × 20 cm). The anode-cathode distance was kept also at 4 cm as shown in (Figure 2). Meanwhile, the third cell consists of a cylinder vessel made of plexi glass with diameter 6 cm and a height of 10 cm (Figure 3). A perforated aluminum sheet with diameter (5.5 cm) was isolated with epoxy from its back and then placed in plexi glass cylinder and acts as the cathode, while the anode in the bottom consists of a perforated cylindrical connected together with a thin wire of aluminum to make sure that the layer of anode was fed with the electrical current. The anode-cathode distance was kept at 2 cm. The electrical circuit of each cell consists of power supply (10V, 5A) with a voltage regulator and multi range ammeter, all connected in series with the cell; a voltmeter was connected in parallel with the cell to measure its voltage.

### Reagents and analytical procedures

Stock solution of -naphthols compound was prepared by using an analytical grade of chemicals and dissolving it in distilled water. Experimental solutions of desired concentrations were obtained by successive dilution with distilled water. The pH of the solution was adjusted by means of HCl and/or NaCl solutions. A digital calibrated pH-meter (Hanna, Model pH 211) was used to measure the pH of waste solutions. The analytical determination of -naphthols was carried out with the standard spectrophotometric procedure using U. V. spectrophotometer (UNICO, Model U.V 2100) and measured at wave length 320 µm. At the end of each experiment, the treated solutions were filtered by using Whitman No. 40 filter paper and centrifuged to remove any traces before analysis.

### Electrocoagulation procedure

For each run, a known volume of synthetic waste solution was mixed with the appropriate amount of sodium chloride which was used as a supporting electrolyte. The solutions were placed into the

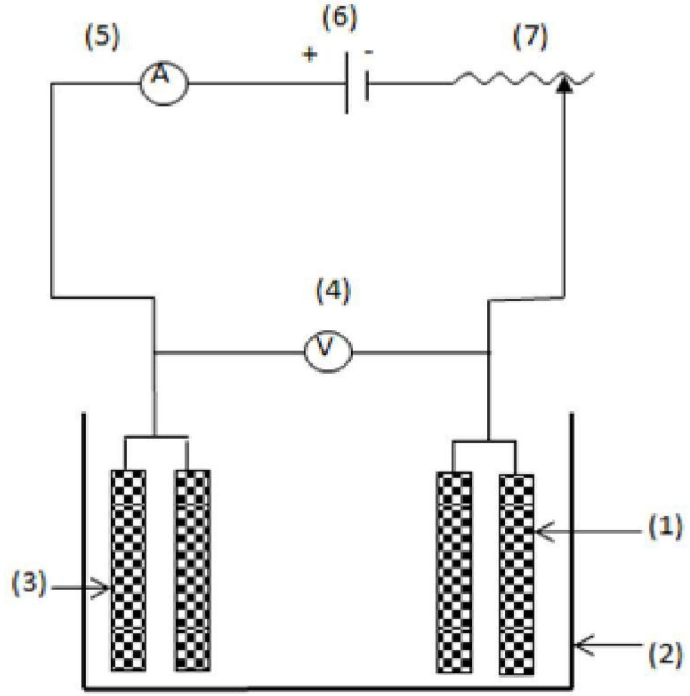

**Figure 2.** A schematically. Diagram for Cell No. 2 (1) Al anode, (2) Plexiglas stank, (3) Al cathode, (4) Voltammeter, (5) Ampere meter, (6) Power supply and (7) Resistance.

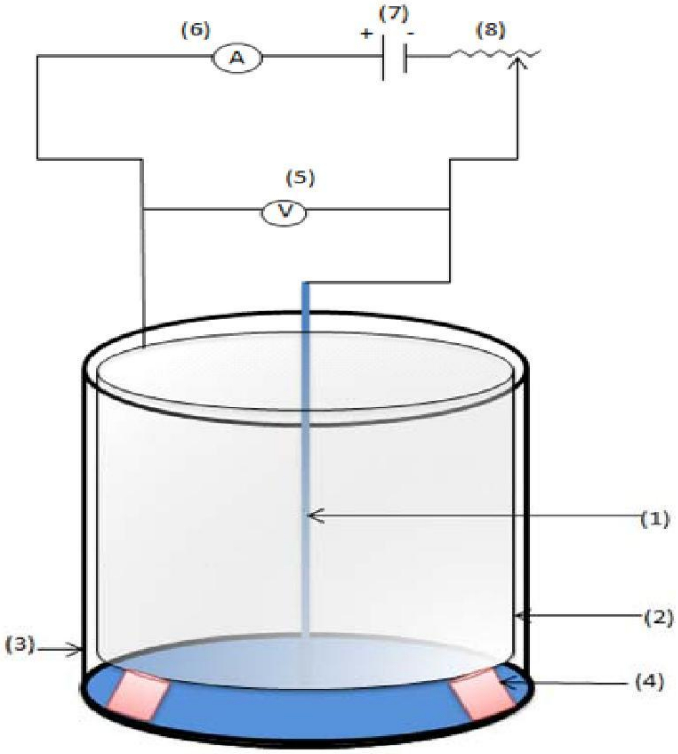

**Figure 3.** A schematically. Diagram for Cell No. 3 (1) Al anode, (2) Aluminum Cathode, (3) Plexi-glass tank,
(4) Epoxy from plexi-glass, (5) Voltammeter, (6) Ammeter and (7) Power supply (8) Resistance.

electrolytic cell. The pH was adjusted by the addition of NaOH and/or HCl solutions. Direct current from the D. C. power supply was passed through the solution via the two electrodes during 120 min electrolysis; 10 ml of the solution was drawn at specified interval time during the experiment. The location of the drawn samples was kept constant for each run. Samples were filtered and centrifuged, and then taken for absorbance measurements at an appropriate wavelength of the maximum absorption for -naphthols. The measured absorbance was then converted to the residual concentration of the compound using a calibration curve obtained from a plot between the absorbance versus the concentration. The electrodes were washed with HCl solution (15% w/v) before each run in order to remove any adhering scales or oxides. Following each run, the electrodes were washed with distilled water, dried until the second use. The efficiency of -naphthol removal (% Removal) was calculated as:

$$\% \qquad \frac{\quad}{\quad} * 100 \qquad (1)$$

Where $C_i$ is the initial -naphthols concentration (mg/l) and $C_f$ is the final -naphthol concentration (mg/l).

## RESULTS AND DISCUSSION

### Effect of electrolysis time

Figures 4 to 6 show the effect of electrolysis time on the removal efficiency of -naphthols at a current density of 15 mA/cm$^2$ for cells No. 1 and 2), and current density of 106 mA/cm$^2$ for cell No. 3. As the duration of the electrolysis treatment increased, a comparable increase in the removal efficiency of -naphthols was observed. It was also observed that as the reaction time increased from 30 to 180 min, the removal efficiency increased from 27 to 77%, from 20.8 to 58.3% and from 25.0 to 72.9% for Cells 1, 2 and 3, respectively. This is due to that, as the time of electrolysis increased, the time of mixing and reaction also increased (Golder et al., 1994). This is ascribed also to the fact as the time increased, more hydrogen bubbles were generated at the cathode; these bubbles improved the degree of mixing and enhanced the flotation ability of the cell with a consequent increase in the percentage removal (Mouedhen et al., 2008; Zaroual et al., 2009).

### Effect of current density

Another important parameter influencing the performance and the economy of the electrocoagulation process is the density of current applied at the electrodes (Lacasa et al., 2011). To study the effect of current density on the efficiency of electrocoagulation in removal of β-naphthols, the experiments carried out from 2.5 to 20 mA/cm$^2$. From

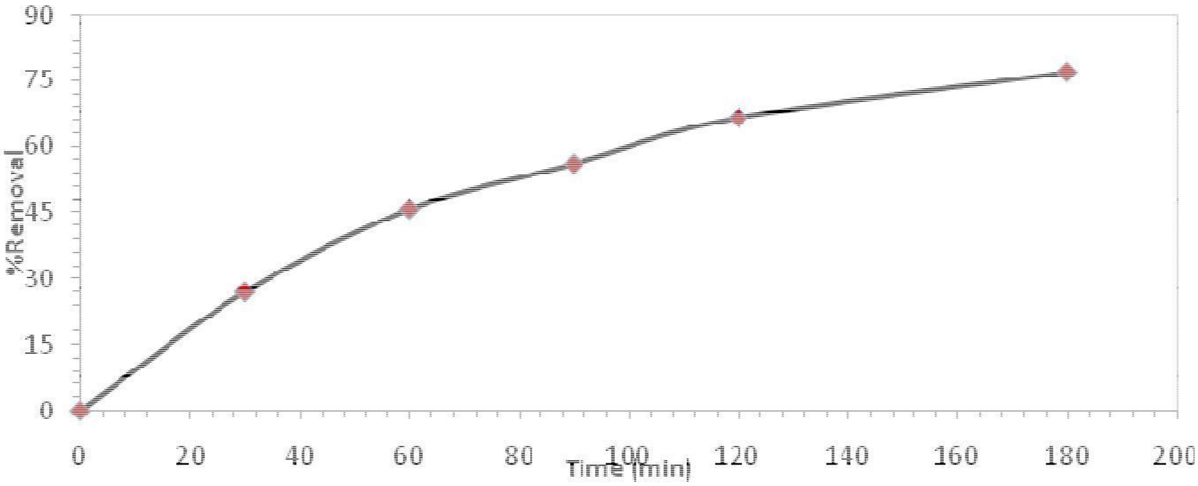

**Figure 4.** Effect of electrolysis time on the removal efficiency (Current density 20 mA/cm$^2$, Initial pH7, initial concentration 40 ppm, NaCl concentration = 1 g/l, temperature 25°C, Cell No. 1).

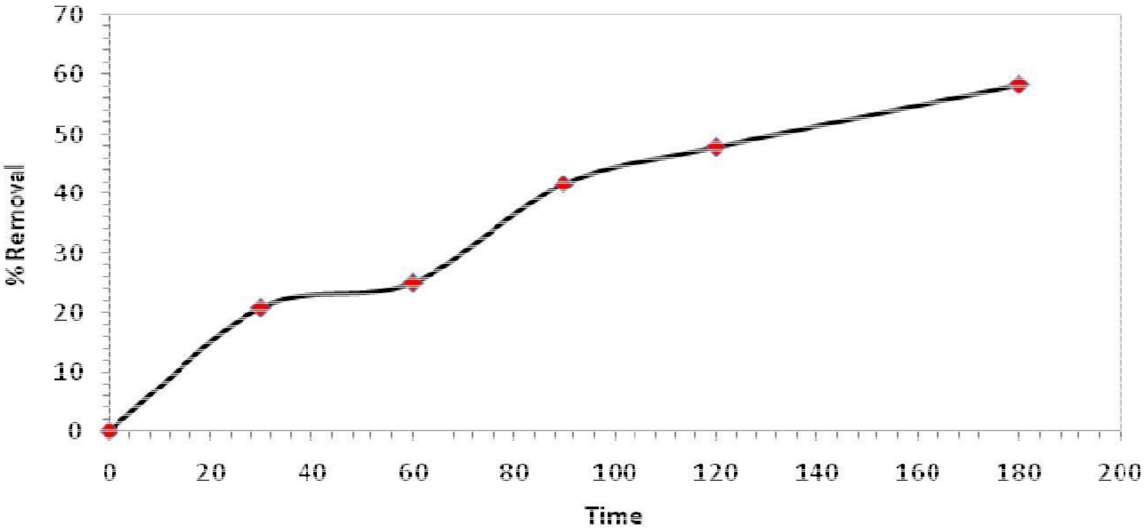

**Figure 5.** Effect of electrolysis time on the removal efficiency (At Current density 20 mA/cm$^2$, initial pH7, Initial concentration 40 ppm, NaCl Concentration = 1 g/l, temperature 25°C, cell No. 2).

Figures 7 and 8, it was observed that as the current density increased from 2.5 to 20 mA/cm$^2$, the efficiency of electrocoagulation increased from 25 to 79% and from 16.7 to 66.6% for Cells 1 and 2, respectively. But for Cell No. 3, the same trend was observed, but with higher values of current densities due to the difference in the area of the cathode as shown in Figure 9. This trend was observed for the three cells due to the fact that as the current density increased, the formation of flocs of aluminum hydroxide at the anode of the cell increased and this enhanced the rate of adsorption of β-naphthols on the flocs of aluminum hydroxide. Also, by increasing the current density of the cell, the amount of hydrogen bubbles at the cathode increased, resulting in a greater upward flux and a faster removal of β-naphthols and sludge flotation (Kurt et al., 2008; Khandegar et al., 2013). At high current density, the quantity of $Al^{3+}$ from anode dissolution increases according to Faraday's low. An increase in current density above the optimum current density does not result in an increase in the pollutant removal efficiency as sufficient number of metal hydroxide flocs are available for the sedimentation of the pollutant.

## Effect of pH

Solution pH is one of main factors affecting electrochemical

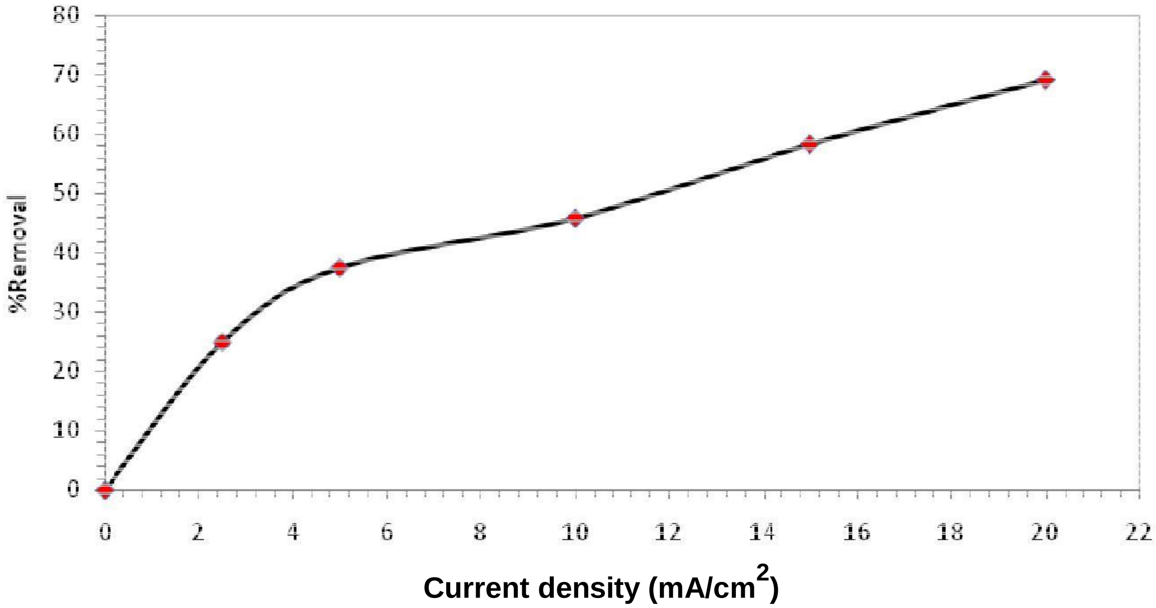

**Figure 7.** Effect of current density on the removal efficiency (Initial pH7, initial concentration 40 ppm, NaCl concentration 1 g/l, temperature 25°C, time 180 min, cell No. 1).

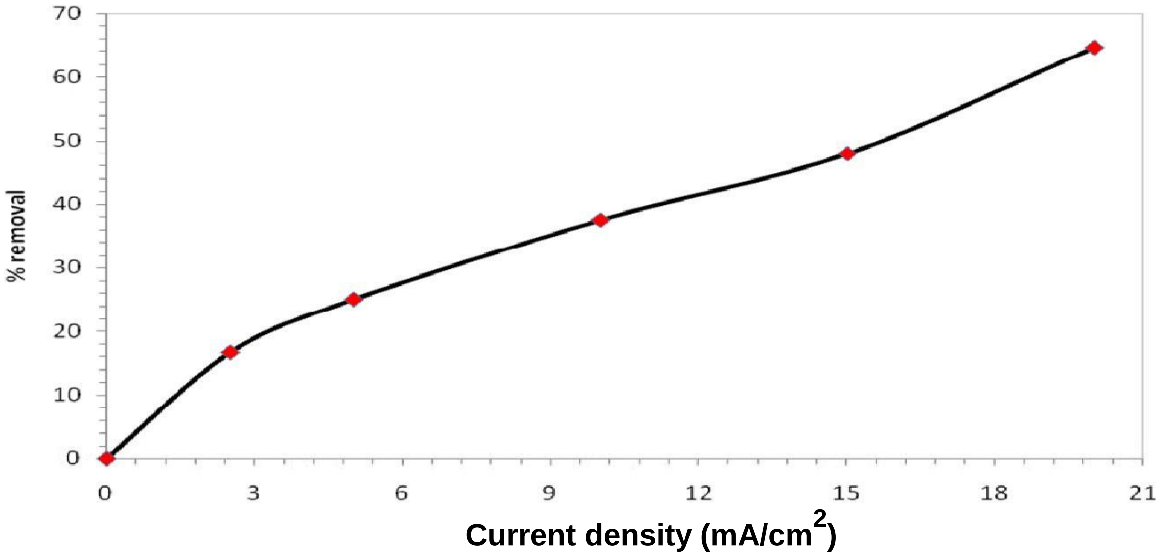

**Figure 8.** Effect of current density on the removal efficiency (Initial pH7, Initial concentration 40 ppm, NaCl concentration 1 g/l, temperature 25°C, time 180 min, cell No. 2).

processes. Therefore, pH (in the range of 1 to 11) was examined as one of the main variables affecting electrocoagulation removal of β-naphthols from waste water. The results of all experiments of the three cells showed the same trend according to the change in pH as was shown in Figure (10). The trend was that, when the pH was increased from 1 to 7, the percentage removal increased and then decreased for a pH between 9 and 11. This finding supports that electrocoagulation efficiency is a function of pH and these results are in accordance with other researchers who have reported the maximum performance for the ECP at pH between 7 and 8 when using Fe or St.St as the sacrificial electrode (Pulkka et al., 2014; Behbahani et al., 2011). The lower percentage of removal at higer acidic and alkaline pH can be explained by amphoteric behavior of $AL(OH)_3$ which

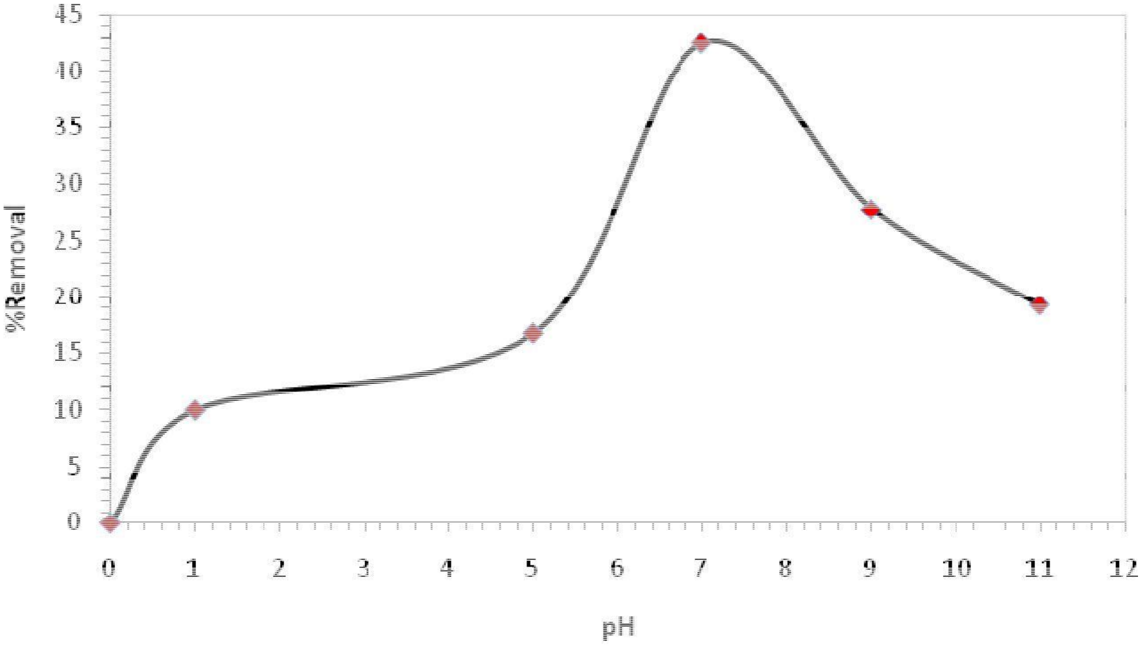

**Figure 10.** Effect of pH on the removal efficiency (Current density 20 mA/cm$^2$, initial concentration 40 ppm, NaCl concentration 1 g/L, temperature 25°C, time 180 min, Cell No. 2).

does not precipitate at very low pH. The figure show the effluent pH after EC treatment would increase for acidic influent but decrease for alkaline influent.

### Effect of sodium chloride concentration

Table salt is usually employed to increase the conductivity of the water or wastewater to be treated. Besides its ionic contribution in carrying the electric charge, it was found that chloride ions could significantly reduce the adverse effect of other anions such as $HCO_3$, $SO_4^{2-}$. The existence of the carbonate or sulfate ions would lead to the precipitation of $Ca^{2+}$ or $Mg^{2+}$ ions that form an insulating layer on the surface of the electrodes. This insulating layer would sharply increase the potential between electrodes and result in a significant decrease in the current efficiency. It is therefore recommended that among the anions present, there should be 20% $Cl^-$ to ensure a normal operation of electrocoagulation in water treatment. The addition of NaCl would also lead to the decrease in power consumption because of the increase in conductivity (Linares-Hernandez et al., 2009). The same trend was also observed for all cells as shown for one of them in Figure 11). From this figure, it was observed that with increasing initial concentration of Nacl from 0.5 to 5 g, the percentage removal increased. But it was also observed that; there is a slight change in results beyond 1 g/L of NaCl; so, the optimum amount of the electrolyte was taken to be 1 g/L. Previous works shows a similar results (El-Naas et al., 2009; Mouedhen et al., 2008) variety of electrolytes like NaCl, KCl, $NaNO_3$, $NaNO_2$, $NaSO_4$, etc., are available. But, due to low cost and easy availability, NaCl has been selected as the best electrolyte (Zaroual et al., 2009).

### Comparison between the anode geometry of three types of cells

Figures 12 to 14 shows the comparison of the three anodes geometry of cells was studied in case of the effect of electrolysis time, pH, and NaCl concentration on the percentage removal of the β-naphthols. As indicated from the previous Figures, the anode geometry of Cell No. 2 which has a cylindrical perforated aluminum anode produced the lowest percentage removal of 55%. While the anode geometry of Cell No. 1 which has a perforated rectangular aluminum sheet as an anode with larger area produced the highest percentage removal of 88% at similar experimental conditions .The geometry of the reactor affects operational parameters including bubble path, flotation effectiveness, floc formation, fluid flow regime and mixing/settling characteristics. From the literature, the most common approach involves plate electrodes (aluminum or iron) and continuous operation (El-Naas et al., 2009). From previous literature (Mouedhen et al., 2008; Zaroual et al., 2009; Lacasa et al., 2011; Kurt et al., 2008), it was found that the perforated anode sheet with larger surface area achieved the maximum percentage removal than the non-perforated anode; this is the same as results obtained

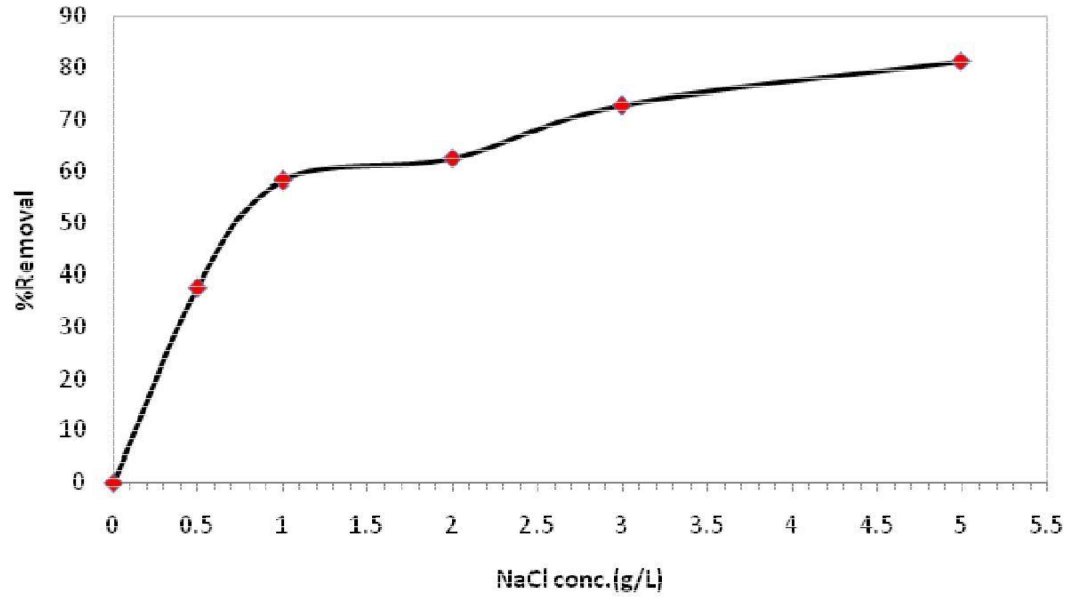

**Figure 11.** Effect of NaCl Conc. on the removal efficiency (Current density 20 mA/$cm^2$, initial pH7, initial concentration 40 ppm, temperature 25°C, time 180 min., Cell No. 1).

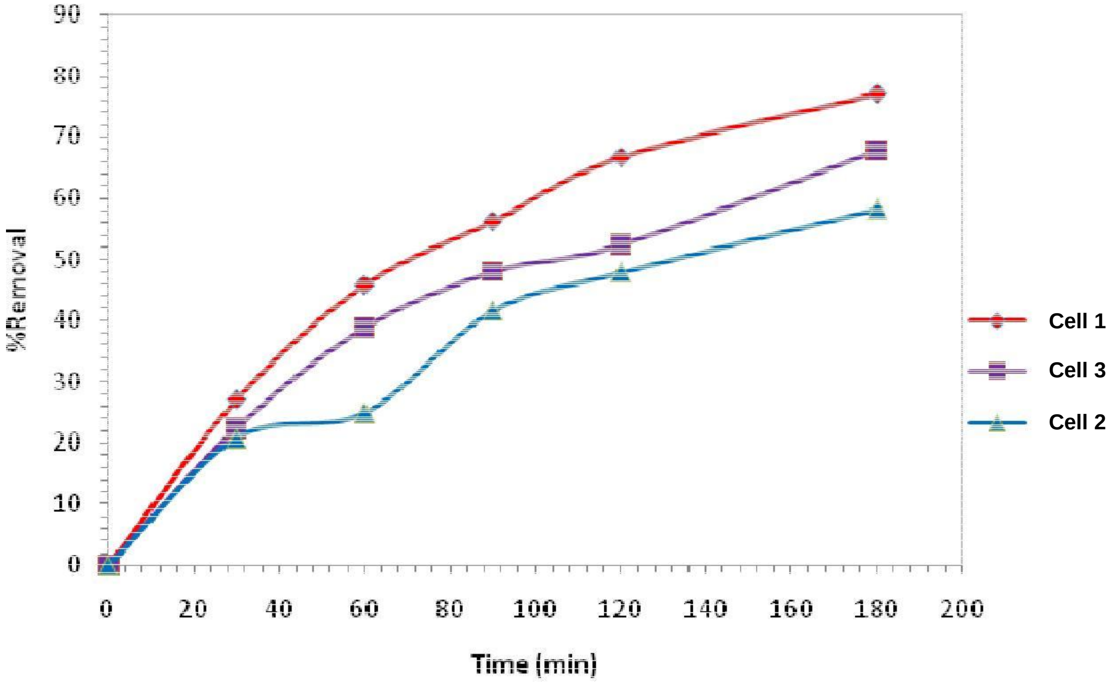

**Figure 12.** Comparison between the 3 cells in electrolysis time (Current 4A, initial pH7, initial concentration 40 ppm, NaCl concentration 1 g/l, temperature 25°C).

from this study. The shape of the electrodes affects the pollutant removal efficiency in the electrocoagulation process. It is expected that the punched holes type electrodes will result in higher removal efficiency compared to the plane electrodesn (Khandegar et al., 2013) as indicated in this thesis. They have reported higher discharge current for the electrode with punched holes than for plane electrode resulting in higher collection efficiency with punched electrode compared with plane electrode. The electric field intensity at the

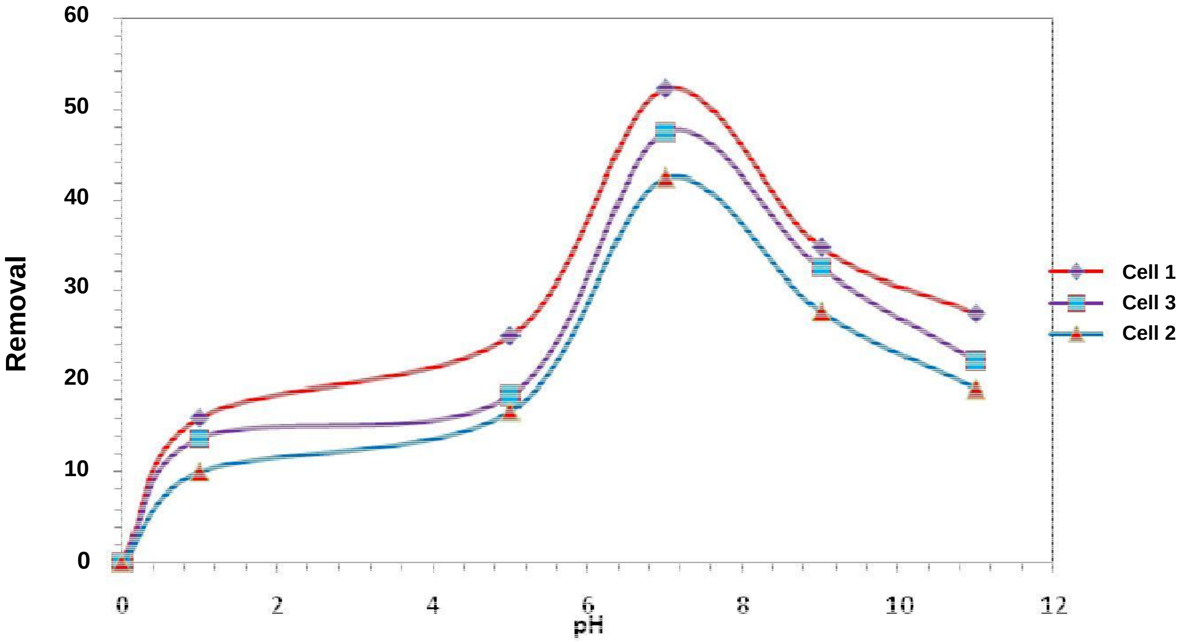

**Figure 13.** Comparison between the three cells (pH) (Current 4A, initial concentration 40 ppm NaCl concentration 1 g/L, temperature 25°C, time 180 min).

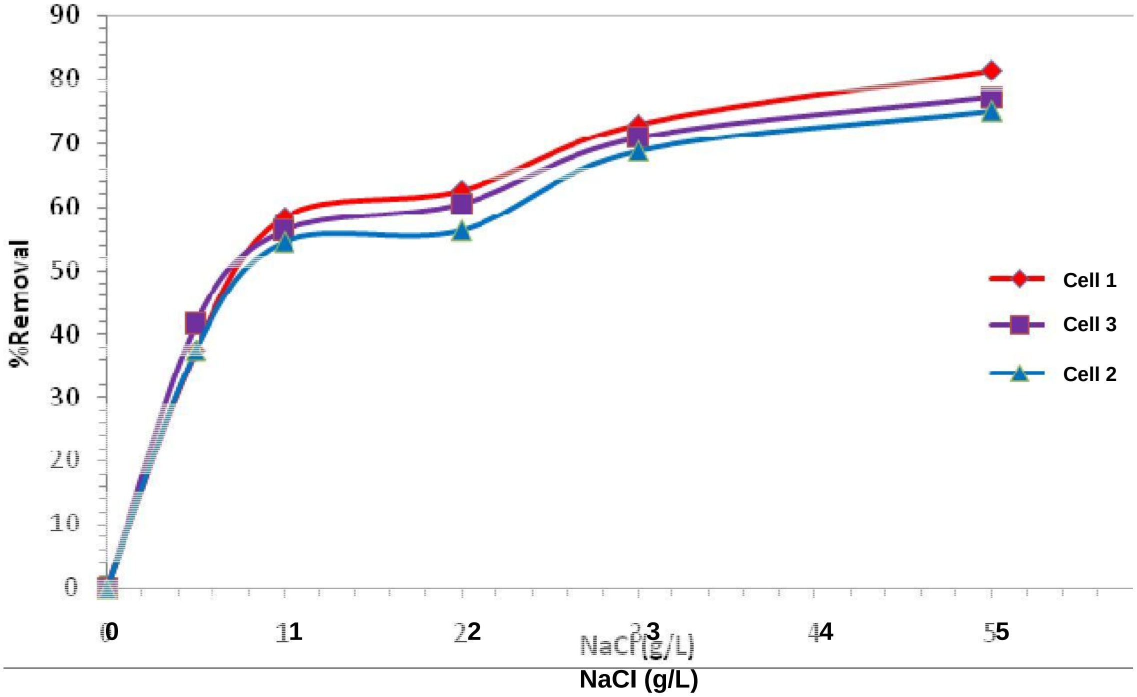

**Figure 14.** Comparison between the three cells (NaCl) (Current 4A, initial pH7, initial concentration 40 ppm, temperature 25°C, time 180 min).

edge of punched holes type electrodes is higher (1.2 times) than at plane type electrode resulting in an increase in the discharge current at punched type electrode.

## Conclusions

Electrocoagulation has successfully treated a wide range of waste streams. Previous research has focused on the application of electrocoagulation to a particular situation (plant and waste stream). The results of this study indicate that EC is a promising treatment for the removal of polyaromatic hydrocarbons (β-naphthols) from waste water. The following conclusions could be drawn from this experimental study:

i. Experimental parameters such as current density, electrolysis time, pH, and electrolyte concentration were investigated for β-naphthols compound removal in batch mode apparatus with three different anode geometries.
ii. The maximum removal of (β-naphthols) was attained at current density 20 mA/cm$^2$, initial pH7, treated volume 1.5 L, NaCl concentration 1 g/L and temperature of 25°C in Cell 1.
iii. Increasing the current density and NaCl concentration resulted in an increase in the β-naphthols removal.
iv. It was found that Cell No. 1 has a higher efficiency of removal than Cell No. 2 and 3 which has the perforated shape anode.

## Conflict of Interest

The authors have not declared any conflict of interest.

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