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Comparative experimental study on the COD removal in aqueous solutions of pesticides by the electrocoagulation process using monopolar iron electrodes

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ABSTRACT

The aim of this work is a comparative experimental study on the COD removal in aqueous solutions of the following pesticides, Chlorpyrifos-Ethyl48EC, Fenitrothion 3% and Acetamiprid20% SP by electrocoagulation process. The effect of various parameters of electrocoagulation on removal efficiency was studied and optimized. The COD removal using nonpolar iron electrodes was affected by current density, contact time, initial pH and initial concentration of pesticides. The optimum conditions for the electrocoagulation process were identified as contact time (4, 5 and 10 min), for a maximum abatement of 100 mg/L pesticide listed above respectively. The results of this investigation provide important data for the development process to remove significant concentrations of COD using moderate iron consumption, and thereby lowering the cost of treatment. Moreover, our results show that the pesticide Chlorpyrifos-Ethyl was removal quickly with a maximum rate of 87% at contact time of 4 min.

Capsule Summary: The EC process is effective in COD removal of pesticide and not costly. Aqueous solutions of organophosphorus pesticides are rapidly eliminated with a low optimum current density. The optimum pH of the solutions is without addition of acid or base

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INTRODUCTION

Pesticides are being ever more used to control the loss of agricultural crops and improve yield. In developing countries like Algeria, the use of pesticides is dispossessed to be so imperative that they are associated with growth of human benefit (Younes et Galal-Gorchev, 2000). Extensive usage of pesticides is accountable for water contamination because of their leaching and runoff losses. Inappropriate discarding of the empty pesticide bottles, washing of spray instruments and unfettered discharge from manufacturing units are further sources of water resources contamination The removal mechanisms reported in the electrolysis process generally include oxidation, reduction, decomposition, so that the mechanisms of EC processes include coagulation, adsorption, precipitation and flotation (Rajeshwar et al., 1994; Vlyssides et al., 1999). EC is a low-cost process and effective method for treating water and wastewater. It was tested successfully to treat drinking water (Mameri et al., 1998; Vik et al., 1984), aqua cultural wastewater (Lin et Wu, 1996), textile wastewater (Lin et Chen, 1997; Lin et Peng, 1996), industrial wastewater (Lin et al., 1998), and landfill leachate (Lin et Chang, 2000). It was also used to remove phenol (Awad et Abuzaid, 2000) and surfactants (Ciorba et al., 2000) from industrial wastewaters. Although electrochemical coagulation has been utilized for over a century, the available literature reveals little studies on the removal of herbicides by electrochemical coagulation such as Malathion (Pitulice et al. 2013), methyl parathion, atrazine and triazophos (Babu et al. 2011), Malathion, imidacloprid and Chlorpyrifos (Nasser et Nader, 2015a) and abamectin (Nasser et Nader, 2015b).The feedback on these electrodes produces metal hydroxides:

Anode:

$4Fe_{(S)} \rightarrow 4Fe^{2+}_{(aq)} + 8e^{-}$ (1)

$Fe^{2+}_{(aq)} + 2OH^{-}_{(aq)} \rightarrow Fe(OH)_{2(S)}$ (2)

The hydrogen thus generated is therefore involved in the flotation of the flocks and thereby promotes both elimination of suspended solids that the removal of dissolved organic compounds adsorbed partly on the flocks. Iron hydroxides formed Fe $(OH)_{n(S)}$ where n is 2 or 3 (equations 2 and 4) remain in the aqueous solution as a gelatinous suspension which can remove pollutants from the waste water (Ibanez et al., 1998; Xinhua et Xiangfeng, 2004).Other hydrated forms of the ion Fe3 +, pH dependent, have been suggested (Kobya et al., 2003):

$Fe(H_2O)_2^{3+}$, $Fe(H_2O)_5OH^{2+}$, $Fe(OH)^{2+}$, $Fe(OH)_2^{+}$, $Fe_2(OH)_2^{4+}$, $Fe(OH)_4^{-}$, $Fe(OH)_6^{3-}$, $Fe(H_2O)_4(OH)_2^{+}$, $Fe_2(H_2O)_8(OH)_2^{4+}$,$Fe_2(H_2O)_6(OH)_4^{2+}$.

These complexes act as coagulant. They are adsorbed on the particles, and so cancel the colloidal fillers.

The aim of this work is a comparative experimental study on

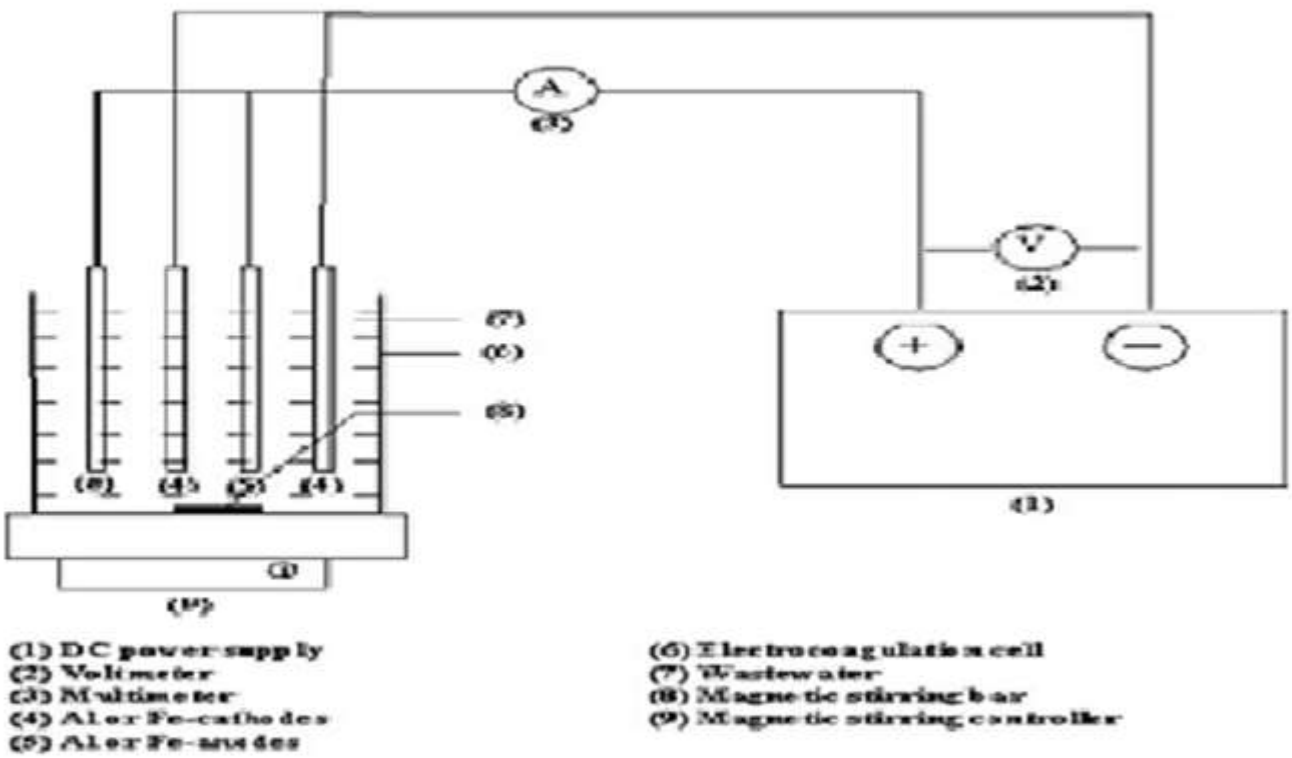


Fig. 1: Electrocoagulation system with Fe electrodes connected in parallel monopolar mode

the COD removal in aqueous solutions of the following pesticides, Chlorpyrifos-Ethyl48EC, Fenitrothion 3% and Acetamiprid20% SP by electrocoagulation process using sacrificial anodes Iron.

Cathode:

$2H_2O_{(l)} + 2e^{-} \rightarrow H_{2(g)} + 2OH^{-}_{(aq)}$ (3)

Fe^{2+} ions are oxidized to Fe^{3+} ions by dissolved oxygen and there is the formation of ferric hydroxide Fe $(OH)_{3(S)}$ color rust, according to the reaction:

$4Fe^{2+}_{(aq)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(S)} + 8H^{+}$ (4)

MATERIAL AND METHODS

Experiments

It is preferable to use a monopolar configuration since the consumption of the electrical energy is much higher in a

Table 1: Initial characteristics of pesticides

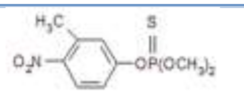
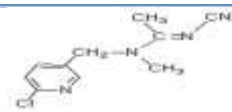
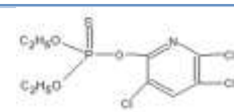
Trade name	Asmidion	Aceplan	Alphychlore
Chemical Name	Fenitrothion 3%	Acetamiprid 20%SP	Chlorpyrifos48EC
pH	9.72	6 -7	6.7
COD (mg/L O_2)	388	125	187
Solubility in Water	<3mg/L	4.2g/L	<3mg/L
Appearance	Green Powder	Blue Powder	Orange-Honey Liquid
Brute formula	$C_9H_{12}NO_5PS$	C_{10}-H_{11}-ClN_4	$C_9H_{11}Cl_3NO_3PS$
molar mass(g)	*277*	223	350
Formula developed			

Table 2. Pseudo-kinetic rate constants with second-order models for COD removal of pesticides

Pesticides	t/C_t	k_2	R^2
Fenitrothion	0.02575 + 0.01924.t	0.01728	0.993
Acetamiprid	0.02061 + 0.01041.t	0.00673	0.992
Chlorpyrifos	0.00032 + 0.01143.t	0.40956	0.999

Table 3: Estimated EC process to optimal conditions.

Pesticides	Fénitrothion3%	Chlorpyrifos	Acetamiprid
E (KWh/ Kg of COD Removal)	0.273	0.552	5.503
Cost($)	0.012 $	0.025 $	0.25 $

bipolar electrochemical cell for the same imposed current intensity and while maintaining the same spacing between the two electrodes (Yu et al., 2005). The pesticides solutions were prepared with deionized water at 100 mg/L. The electrocoagulation system that was used is shown in Fig. 1. For more precision in reading the current and voltage values, a multimeter and a voltmeter were connected, with a direct current power supply (Leybold Didactic CMBH, Hürth, Germany [30 V and 2.5 A]). The arrangement of a single cell with many electrodes is electrically similar to monopolar electrodes with cells set in parallel (Yousuf et al., 2001). Experiments were carried out in Plexiglas cell with the dimensions (11cm x 8cm x 8cm), equipped with four iron electrodes (99.40%) connected in monopolar parallel mode. Two cathodes and two anodes with the dimensions (10cm x 5cm x 0.1cm) and a total active area of 85 cm^2 were used. The net spacing between two electrodes was 1 cm. A volume of 300 mL of an aqueous pesticides solution was introduced to the cell of electrocoagulation and then stirred at 400 rpm using a magnetic stirrer. The current density was fixed at the desired value at the start of each reaction. All the experiments were conducted at a constant temperature of 25°C. At the end of each electrochemical reaction, the solutions were decanted for 12 hours and then filtered. Before each electrocoagulation, the electrodes were washed with acetone, then immersed in a solution of hydrochloric acid (5%) for 5 minutes, rinsed with distilled water and dried. Conductivities and pH values of solutions were adjusted by 0.3g potassium chloride (KCl) and sulfuric acid H_2SO_4or sodium hydroxide NaOH solutions0.1M respectively, the chemicals used above are provided by (Merck, Darmstadt, Germany). The particulates of colloidal ferric oxyhydroxides gave yellow-brown color into the solution after electrocoagulation and electrolytic flotation. The sludge was separated by filtration with filter paper, analyzed by FTIR (Vertex 80, Bruker, France). Then the liquid was analyzed for COD determination, which carried out according to Standard Methods for examination of water (APHA). The COD removal efficiency and the electrical energy consumed in the experiments are:

$$COD\ Removal\ (\%) = \{(COD_0 - COD_{final}).100\}/COD_0 \quad (5)$$

Where, COD (initial) and COD final of the pesticide solutions are calculated in mg/L.

$$E = U.I.t/ 3.6.10^6 \quad (6)$$

Where, E (kWh), U (Volts), I (Ampere), t (Seconds).

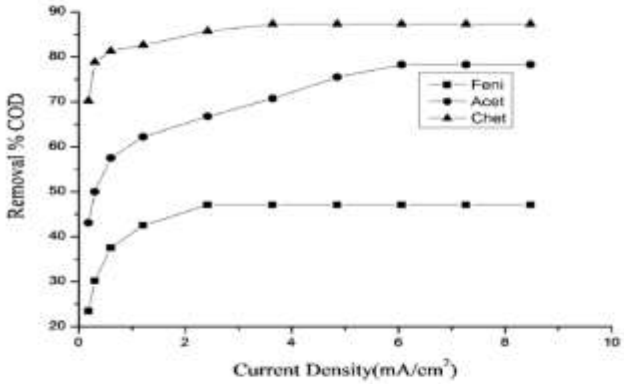


Fig. 2: Efficiencies of COD removal from Pesticides solutions as a function of current density: **Feni**: Temps=5min, pH =9.72. **Acet**: Temps=5min, pH =6-7. **ChEt**: Temps=5min, pH =6.7.

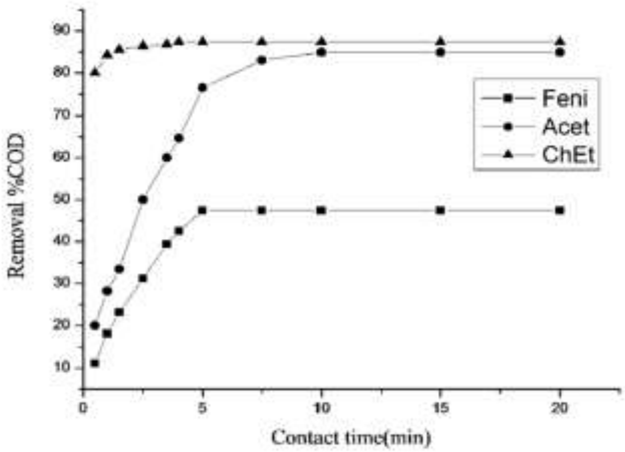


Fig. 3: Efficiencies of COD removal from Pesticides solutions as a function operating time: **Feni**: J (mA/cm^2) = 2. 42, pH= 9. 72. **Acet**: J (mA/cm^2) = 6. 1, pH= 6-7. **ChEt:** J (mA/cm^2) = 3.64, pH= 6. 7.

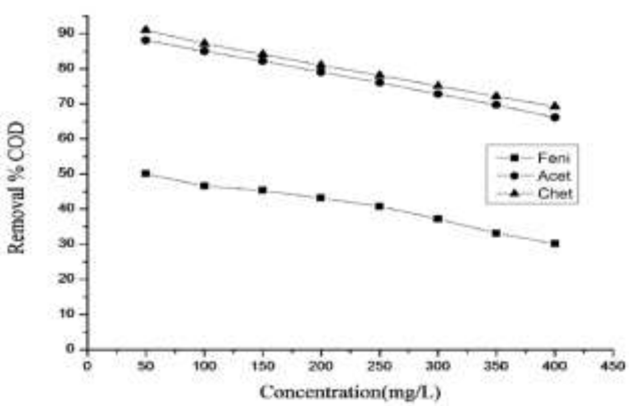


Fig. 4: Efficiences of COD removal from Pesticides solutions as a function initial concentration: **Feni:** J (mA/cm^2)= 2.42, Temps= 5min, pH= 9.72.**Acet**: J (mA/cm^2) = 6.1mA/cm^2, Temps= 10min, pH= 6-7.**ChEt:** J (mA/cm^2) = 3.64, Temps= 4min, pH= 6.7.

Chemicals

The pesticides are provided by Rivale France. Acetamiprid20%SP: (E)-N'-[(6-chloro-3-pyridyl) methyl]-N2-cyano-N'-methyl acetamidine. Chlorpyrifos48EC: *O*, *O*-diethyl *O*-3, 5, 6-trichloropyridin-2-yl phosphorothioate. Fénitrothion3%: *O*, *O*-Dimethyl *O*-(3-methyl-4-nitrophenyl) phosphorothioate).

RESULTS AND DISCUSSION

Effect current density

It has already been reported by several authors that the applied current density has significant influence on the efficiency of the EC process (Mahesh et al., 2006; Holt et al., 2001).The current density determines the coagulant dosage rate. This parameter should have a significant impact on the removal efficiency of the COD. To examine the effect of current density on COD removal efficiency, a series of experiments were carried out with the current density ranging from (0.18 to 8.48 mA / cm^2) at a contact time of 5 minutes. Fig. 2 shows the optimum current densities were (2.42, 6.1 and 3.64 mA / cm^2) and COD removal efficiencies were (47%, 78% and 87%) for pesticide solutions (Feni, Acet and ChEt) respectively. The increase of coagulant and bubbles generation rate lead to the increase number of H_2 bubbles and decreases their size with increasing current density resulting in a faster removal of COD (Mollah et al., 2004; Holt et al., 2002). Further increase in current density above optimal condition did not lead to an increase in COD removal efficiencies. But the sufficient amount of flocks needed to coagulate the pesticide might be available at optimal current density and further formation of flocks which did not change COD removal efficiency (Khandegar et Saroha, 2013). The optimal current density of the pesticide (Acet) is higher compared to pesticides (ChEt) and (Feni). This is due to the stronger solubility of (Acet) in water (4.2 g / L) and also to differences in chemical structures. In fact, ChEt and Feni are organophosphorus insecticides with water solubility less than 3 mg /L (Barbash et Resek, 1996). It is noted that the removal efficiency pesticide (Feni) is the lowest; it's partly due to the initial COD concentration which is about double compared to the other two pesticides because the organic matter from pesticides themselves.

Effect contact time

In the present study, contact time also influenced the treatment efficiency of COD removal. With an increase in contact time, the anodic electrode dissolution led to release of metal ions and the cathode released OH^- which formed their hydroxides into pesticides solutions (Benefield et al., 1982; Babu et al., 2007). Fig. 3 depicts that the removal of COD increased progressively with an increase in the contact time from 0.5 to 20 min. with the operating conditions of pesticides solutions, Feni, Acet and ChEt (2.42, 6.1 and 3.64

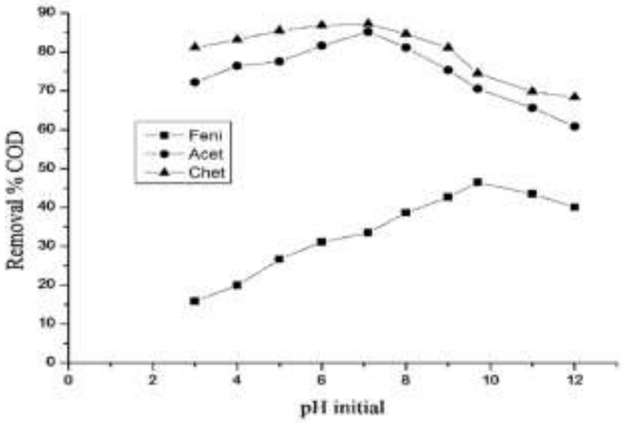


Fig. 5: Efficiences of COD removal from Pesticides solutions as a function initial pH: **Feni:** J (mA/cm^2)= 2.42, Temps= 5min, **Acet:** J (mA/cm^2) = 6.1, Temps= 10min. **ChEt:** J (mA/cm^2) = 3.64, Temps= 4min.

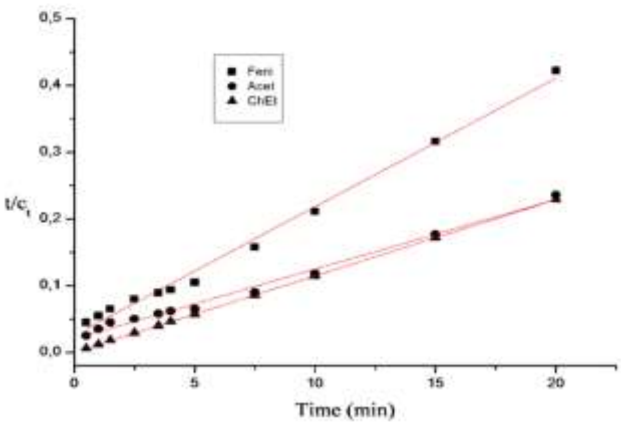


Fig. 6: t/Ct= f(Time) pesticides: Feni, Acet, ChEt

mA/cm^2), the maximum removal of COD (47, 85 and 87%) at optimal contact time (5, 10 and 4 min) respectively. Beyond which there was no significant removal. This may be due to the fact that the dissolved metal ions and their hydroxides in the pesticides solutions achieved the saturation stage for the flock formation. In this process, EC involves two stages which are destabilization and aggregation. The first stage is usually short, whereas the second stage is relatively long (Holt et al., 2005). As shown in Fig. 3 the pesticide (Chet) was quickly removal, for a time of 1min removal efficiency was 84%. This is probably due to its chemical formula and the size of the molecule.

Effect of the initial concentration

Fig. 4 shows the evolution of COD removal efficiency as a function of the initial pesticides concentration, using the optimum conditions obtained previously for current density, reaction time and pH of solution. In this Figure, pesticides solutions with different concentrations in the range 50–400 mg/L were treated by electrocoagulation process. As expected, the rate of COD removal decreases with the increase in initial pesticide concentration. On the other hand, the percentage COD removal was gradually decreased from 50 to 30% (Feni), 88 to 66 % (Acet) and 91 to 69% (Chet) as the initial pesticide concentration increased from 50 to 400 mg/L. This is may be attributed to the fact that at a constant current density the same amount of iron ions passes to the solution at different pesticides concentrations. Consequently, the formed amount of iron hydroxide complexes were insufficient to coagulate the greater number of pesticide molecules at higher pesticide concentrations (Daneshvar et al., 2004; Modirshahla et al., 2008). On the other hand, the decrease in removal efficiency with increasing initial pesticide concentration may be attributed to requiring more coagulant when increasing levels of pollutant. Therefore, it is quite clear that under the present experimental conditions, the lower is the pesticide concentration the better is the COD removal efficiency.

Effect of initial pH

The pH is an important operating factor influencing the performance of the electrocoagulation process Chen et al. (2000).A series of experiments were carried out to evaluate effect of initial pH using solutions containing a sample with an initial pH varying in the range (3-12) at current density of (2.42, 6.1 and 3.64 mA / cm^2) and at times (5, 10 and 4 min), for pesticide solutions (Feni, Acet and Chet) respectively. Fig. 5 display that the removal efficiencies of COD were low in acidic medium, meanwhile, in neutral and alkaline medium the removal efficiencies were much higher using all working electrodes due to the formation of metal hydroxide species which adsorb the pesticides molecules and causes the increase of the removal efficiency (Daneshvar et al., 2004). Fig. 5 shows that the maximum COD removal (47, 85 and 87%) was at pH (9.7, 7.1 and 7.1) for pesticides (Feni, Acet and ChEt) respectively. We observe that the optimal pH was the pH values of the initial pesticides solutions (Table 1). This fact is an advantage and avoids the addition of acid or base to adjust the pH. It was observed that above pH (9.7, 7.1 and 7.1) for pesticides (Feni, Acet and Chet) respectively, there was decreasing trend in adsorption, this may be due to the oxidation of ferrous iron Fe (II) to ferric iron Fe (III) diminishes, resulting decreased removal efficiency in acidic pH values. Neutral and slightly alkaline pH, however, tends to favor Fe (II) to Fe (III) oxidation as well as complex polymerization. Finally, hydroxylated colloidal polymers and an insoluble precipitate of hydrated ferric oxide were formed and the removal efficiency was increased. The decrease of removal efficiency when the pH is higher than 10, and more acidic was observed by many investigators (Vasudevan et Lakshmi, 2012) and was attributed to an amphoteric behavior of $M(OH)_3$ which leads to soluble metal cations (at acidic pH) and to monomeric anions (at alkaline pH).We note that the optimal pH values proved conveying the removal

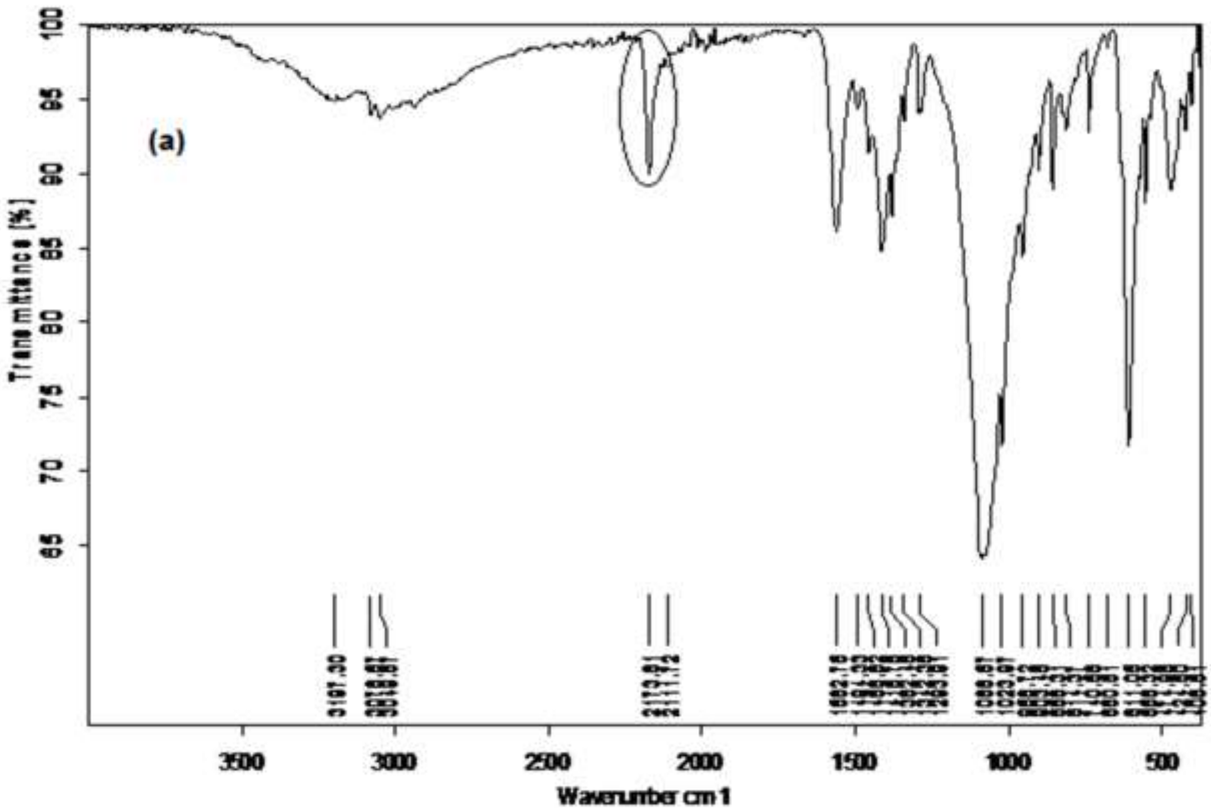


Fig. 7a: FT-IR spectra of initial pesticide Acetamiprid

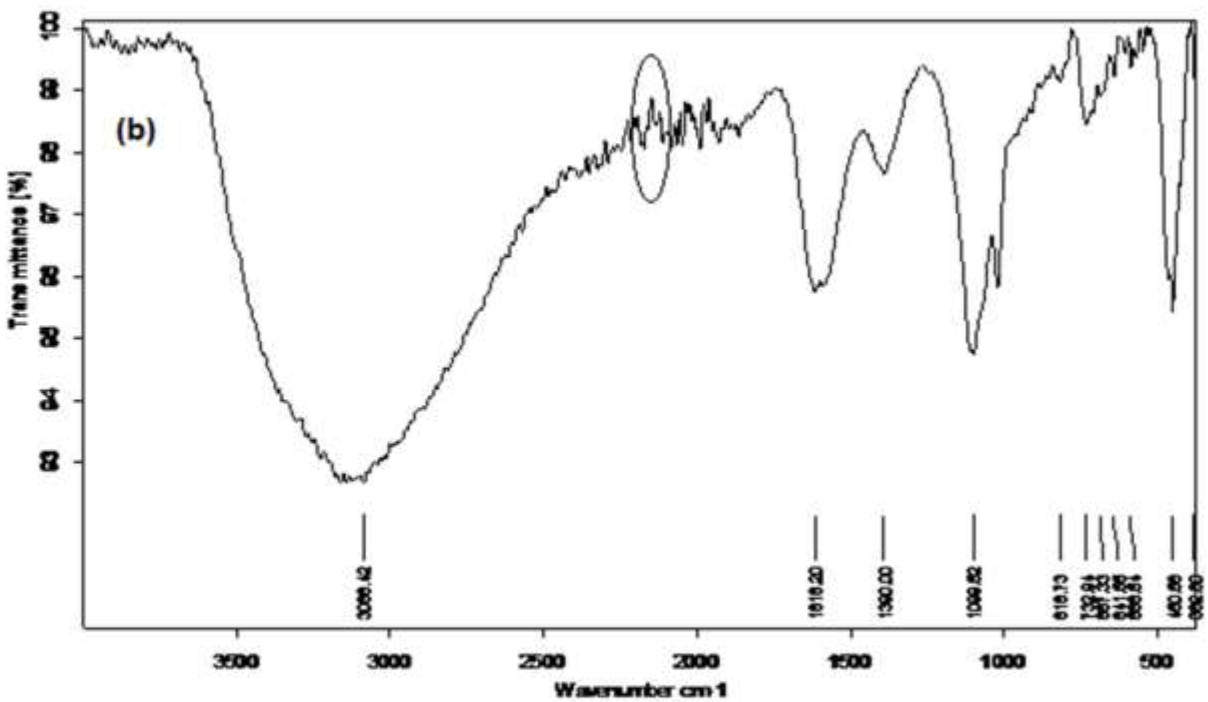


Fig. 7b: FT-IR spectra of Sludge Acetamiprid after EC

efficiencies are due to the presence of metal hydroxides zone are consistent with the following reference (Pourbaix, 1974).

Kinetic studies of the pesticides removal

The removal rate of pesticides can be represented by the following linear- pseudo - second – order equation:

$$t/C_t = 1/k_2C_e^2 + t/C_e \quad (7)$$

C_t: the removal rate % of the COD at t time.
C_e: maximum removal rate % of the COD.
k_2 (min-1): the reaction rate coefficient, t: contact time (min).
As seen in Fig. 6, a pseudo-second-order kinetic model provided a good fit to the experimental results for COD removal of pesticides. It can be ascertained that a higher value of regression coefficient of R^2 (~0.99) for the pseudo-second order kinetics confirms that the EC process of COD removal. The values of k_2 obtained from the slope are shown in Table 2.

FTIR spectral studies

After the experiment at the optimum conditions, the sludge is recovered above the reactor by filtration, was dried to 105 °C and analyzed by IR. Compared with the IR spectrum of the initial pesticide as shown in Fig. 7b:
Pesticide Acetamiprid: Enlargement of the bands, e.g. only one Peak due to stretching absorption of aromatic =C-H at 3086cm^{-1} instead of two, one Peak for aromatic C = C at 1616cm^{-1}. This is likely due to intermolecular bonds.
- No Peak due to stretching absorption of -CΞN to 2173cm-^{1}, this is probably due to a combination inductive effect or effect (-I) chlorine.
For the other two pesticides, there is no remarkable difference between the initial pesticide and the sludge.

Energy Cost

The energy cost (kWh/ Kg for removal of COD) was calculated by applying the equation (6) and results are shown in Table 3.

CONCLUSIONS

The electrocoagulation is a quick and efficient process. Effect of various parameters such as operating time, current density and pH was evaluated. It was found that 47 %(Feni), 85 %(Acet) and 87 %(Chet) removal in COD was achieved within (5, 10 and 4min) of EC treatment for the pesticides. Further increase in treatment time did not improve their removal efficiency. Change in pH value during EC treatment was also noted and maximum value of 9.7(Feni) and 7.1(Acet and Chet) was observed at the end of treatment which is within allowable limits. Applied current density has significant effect on the removal efficiency of EC process. It was found that the current density of (2.42, 6.1 and 3. 64 mA/cm^2) has the highest removal efficiency for studied pesticides (Feni, Acet and Chet) respectively. Further increase in current density showed insignificant improvement in removal efficiency. It has been found that the pesticide (Chet) was quickly removal and the most and the pesticide (Feni) has a low maximum rate, it's in initial concentration of COD is on about double the other two pesticides. Since the Acet pesticide has its current density and optimal contact time greater than the other two, the cost of electricity consumption is greater (0.25$ / Kg of COD removal). This strongly suggests that the pesticides removal rate is most appropriately represented by a pseudo - second – order process. Finally, according to findings of this study it can be concluded that electrocoagulation process can effectively remove pesticides from aqueous solutions and cost no expensive.

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