

Removal of Abamectin Pesticide by Electrocoagulation Process Using Stainless Steel and Iron Electrodes

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Abstract

In this work the removal efficiency of abamectin and chemical oxygen demand (COD) from aqueous solution using the electrocoagulation process were investigated. The effects of operational parameters such as initial pH, initial abamectin concentration, current density, type electrolyte, salt concentration, and temperature on the pesticide and COD removal efficiency have been studied. With an initial pH of 3.7, an initial pesticide concentration of 150 mg/L, (current density 87.5 mA/cm² and 50 mA/cm² for stainless steel (SS) and iron (Fe) electrodes respectively), salt concentration of 1 g/L and temperature of 30°C, the results showed that abamectin and COD removal were 94% and 76.9% by using SS and were 64.5% and 50%, by using Fe electrodes. Pesticide removal kinetic followed pseudo first and second order kinetics using SS and Fe electrodes respectively. It can be concluded that electrocoagulation process by SS electrode is very efficient and clean process for abamectin removal and COD from wastewater.

Keywords: Electrocoagulation; Stainless steel; Iron; Abamectin; Pesticide; Aqueous solution

Introduction

Pesticides are encountered as pollutants in wastewater effluents of pesticide industry and agricultural activities. Water pollution by pesticides is considered as a pervasive problem, because these compounds usually have direct adverse effect on the living organisms. These compounds are toxic and carcinogenic in nature even at low concentration [1]. Abamectin belongs to the family avermectins which are macrocyclic lactones. It is a mixture of two homologues containing about 80% avermectin B1a and about 20% avermectin B1b show Table 1 [2]. These two components, B1a and B1b, have similar biological and toxicological properties [3,4]. The oral LD50 for abamectin in rats is 10 mg/kg, and in mice ranges from 14 mg/kg to greater than 80 mg/kg. Rats given 0.40 mg/kg/day of abamectin had increased stillbirths, decreased pup viability, decreased lactation, and decreased pup weights. Abamectin is highly toxic to insects and may be highly toxic to mammals as well emulsifiable concentrate formulations may cause slight to moderate eye irritation and mild skin irritation, Juliana et al. 2012. In soils with pH 5–9, avermectin half-life ranged between 20 and 47 days where it degraded into at least 13 different products [5].

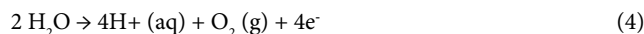
There are several methods to remove the abamectin from water, such as treatment by oxidation [2], Photo-fenton [6], and adsorption [7,8]. In recent years, novel methods for water purification have been developed including chemical, electrochemical and photochemical processes [9,10]. Electrochemical processes (electrolysis and electrocoagulation) have been successfully demonstrated for removing pollutants in various industrial wastewaters [11,12]. Removal mechanisms reported in the electrolysis process generally include oxidation, reduction, decomposition, whereas the mechanisms in the electrocoagulation process include coagulation, adsorption, precipitation and flotation [13,14].

Electrocoagulation (EC) has been applied successfully to treating various wastewater contaminants, such as reactive dyes [15], azo dyes [16], oily bilge water [17], industrial wastewater [18], petroleum refinery wastewater [19], fluoride [20], pulp and paper mill wastewater [21], and phosphate and zinc [22]. EC has several advantages that make it attractive for treating various pesticides such as Malathion [23], methyl parathion, atrazine and triazophos [24], malathion, imidacloprid and chlorpyrifos [25], and monochrotophos [26]. Electrocoagulation (EC) is an electrochemical method to treat polluted wastewaters and aqueous solutions. The removal of diazinon was studied by EC on aluminum

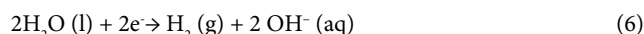
electrode. The effect of several parameters such as initial concentration of diazinon, current density, solution conductivity, effect of pH, and electrolysis time were investigated. The obtained results showed that the removal efficiency of EC depends on all previous parameters except conductivity [27].

Coagulant is generated as a result of oxidation of the anode material by passing the electrical current. Electrocoagulation of pesticide solution using stainless steel (SS) and iron (Fe) electrodes takes place according to the following mechanisms [28,29].

At the anode:



At the cathode:



Where M is the material used as electrode and n is the number of electrons. During the electrocoagulation process metal hydroxides, polyhydroxides and/or polyhydroxymetallic compounds of the electrode material will be generated. These materials contain strong affinity for dispersed particles and counter ions, which results in coagulation [28].

The purpose of the present work is to study the performance of EC process on the removal of abamectin and COD in aqueous solution using stainless steel and iron electrodes. Moreover, the effects of initial pH, current density, type electrolyte, initial abamectin concentration, concentration electrolyte and the temperature on the removal efficiency were measured and investigated.

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Materials and Methods

Chemicals

The pesticide used in the present work was abamectin, pesticide solutions. Abamectin solutions were prepared from the commercially available pesticide, in distilled water at a concentration of 18 g/L. This concentration is the same as that used by farmers during strawberry cultivation. The property of the abamectin is given in Table 1. Sodium chloride, calcium chloride, potassium chloride, potassium iodide, sodium hydroxide, sulfuric acid, potassium dichromate, were of analytical grade and purchased from Merck. Distilled water was used for the preparation of solutions. Standard solutions of potassium dichromate ($K_2Cr_2O_7$), sulfuric acid (H_2SO_4) reagent with silver sulfate (Ag_2SO_4), mercury sulfate ($HgSO_4$) and were prepared to measure the COD. A stock solution of pesticide (500 mg/L) was prepared by dissolving an accurate quantity of the pesticide in distilled water and suitably diluted to the required initial concentrations. Different standard solutions of pesticide with concentration from 50–200 mg/L were prepared to measure its removal at different conditions. The pH of the working solution was adjusted to the desired values with 0.1N HCl or 0.1N NaOH.

Equipments and procedures

The electrocoagulation unit consisted of a 100 mL electrochemical reactor with stainless steel and iron electrodes (anode and cathode) with an effective surface area of 4 cm². The electrodes were 20×10 mm and inter electrodes distance was 1 cm. The electrodes were positioned vertically and parallel to each other. The current density was maintained constant by means of a precision DC power supply; model (DZ040019) EZ Digital CO. Ltd. (Korea). The pesticide concentration was determined using a double - beam UV-Vis spectrophotometer, model UV 1601 is from Shimadzu (Japan) at 515 nm. Hot Plate, model (HB502), Bibby Sterilin Ltd (UK). A pH meter model AC28, TOA electronics Ltd., (Japan). Water bath model SB-650, Tokyo Kikakai CO. Ltd. (Japan). A closed reflux titrimetric unit was used for the COD determination. Chemical Oxygen Demand (COD), HANNA instruments, Thermo reactor, model C9800 reactor in Hungary – Europe.

Analysis

Two main parameters were measured to evaluate the electrochemical

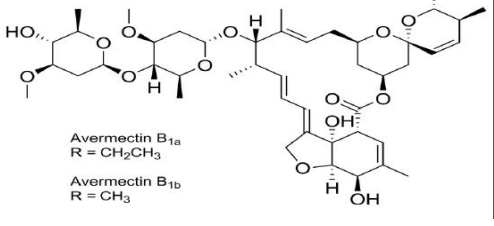
Pesticide	Abamectin
Chemical structure	 <p>Avermectin B_{1a} R = CH₂CH₃ Avermectin B_{1b} R = CH₃</p>
Chemical formula	<p>avermectin B_{1a} C₄₈H₇₂O₁₄ avermectin B_{1b} C₄₇H₇₀O₁₄</p>
Molecular weight (g/mol)	<p>avermectin B_{1a} M (873,07 g.mol⁻¹) avermectin B_{1b} M (859,05 g.mol⁻¹)</p>
λ_{max}	275 nm

Table 1: Properties of abamectin.

treatment efficiency, the remaining pollutant concentration and the COD. Remaining pollutants (abamectin) concentration was measured with the double-beam UV-visible spectrophotometer at $\lambda_{max} = 275$ nm using calibration curve with standard error $\pm 0.5\%$. The COD was determined using a closed reflux colorimetric method [30]. The equation used to calculate the pesticide removal efficiency in the treatment experiments was:

$$\%E = [(A_0 - A) / A_0] \times 100 \quad (1)$$

Where A_0 and A are absorbance values of pesticide solutions before and after treatment with respect to their λ_{max} [31].

The calculation of COD removal efficiencies after electrocoagulation treatment were performed using the following formula [32].

$$C_r(\%) = [(C_0 - C) / C_0] \times 100 \quad (2)$$

Where C_0 and C are concentrations of wastewater before and after electrocoagulation.

Result and Discussion

Effect of electrolyte concentration

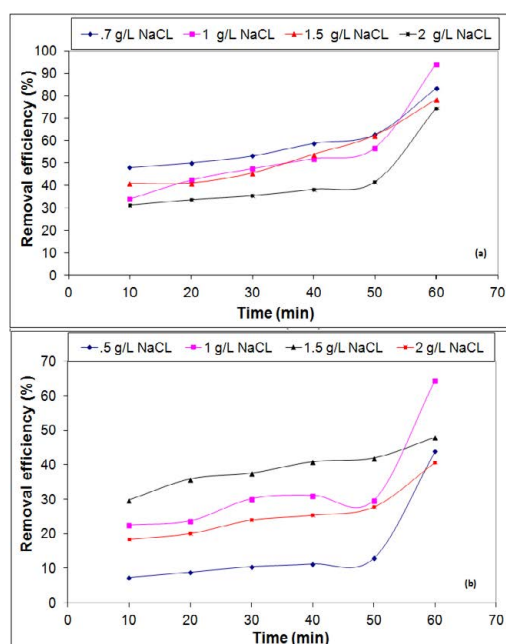
It is important to investigate the effect of electrolyte concentration since actual wastewater usually contains certain amount of salts [33]. Figure 1 and Table 2 represent the effect of electrolyte concentrations on pesticide removal and COD at 60 min using (SS) and (Fe) electrodes respectively at initial concentration of 150 mg/L, (a current density of 87.5 mA /cm² at (SS) and 50 mA/cm² at (Fe) electrodes), inter electrode distance of 1 cm, a temperature of 30°C and at pH of 3.7. Figure 1 and Table 2 show that, as the electrolyte concentration increased, the removal efficiency increased due to the increment of the electrical conductivity reaching the maximum value at 1 g/L NaCl. However, with the increase in NaCl concentration >1 g/L the removal efficiency decreased. It can be attributed that at a constant voltage with increasing of electrolyte concentration, conductivity of pesticide solution increases and resistance decreases, so the passed current increases and the produced amount of metallic hydroxide and pesticide removal increases [34].

Effect of current density

The effect of current density on the removal of abamectin and COD was studied at four different current densities. The reactions were carried out for 60 min using (SS) and (Fe) electrodes under the following conditions: the initial concentration of 150 mg/L, pH of 3.7, inter electrode distance of 1 cm, a temperature of 30°C and NaCl concentration of 1 g/L. Figure 2 and Table 2 show the effect of current density for the removal of pesticide and COD from aqueous solutions. The removal efficiency of abamectin and COD increased up to [94% and 76.9% at SS and 64.5% and 50% at Fe respectively] by increasing the current density. The current density determines the coagulant production rate and the size of the bubble production and hence affects their growth [35,36]. Upon increasing current density, the amount of oxidized stainless steel and iron increased and amounts of metal hydroxide compounds for precipitation and adsorption of the matrix were also increased [37].

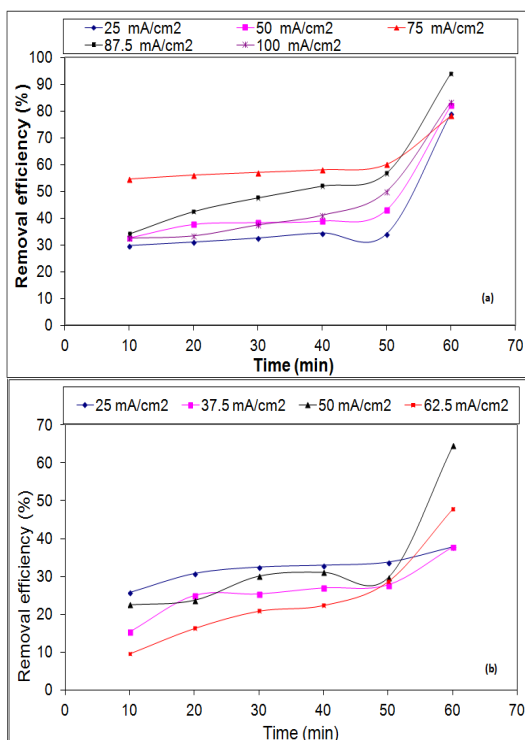
Effect of pH

pH is an important operating factor influencing the performance of the electrocoagulation process [38,39]. A series of experiments was carried out to evaluate this effect, using solutions containing a sample with an initial pH varying in the range (2.3–10.2), at initial concentration of 150 mg/L, (a current density of 87.5 mA/cm² at



Initial concentration of the pesticide =150 mg/L, volume of the solution=100 ml, (a current density of 87.5 mA/cm² at (SS) and 50 mA/cm² at (Fe) electrodes), pH=3.7, inter electrode distance=1 cm, dimension of the electrodes=20 mm × 10 mm and temperature=300°C.

Figure1: Effect of electrolyte concentration on the efficiency of abamectin removal using SS (a) and Fe (b) electrode.



Initial concentration of the pesticide=150 mg/L, volume of the solution=100 ml, [NaCl] concentration=1 g/L, pH=3.7, inter electrode distance=1 cm, dimension of the electrodes=20 mm × 10 mm and temperature=300°C.

Figure 2: Effect of current density on the efficiency of abamectin removal using SS (a) and Fe (b) electrodes

(SS) and 50 mA/cm² at (Fe) electrodes), inter electrode distance of 1 cm, and at NaCl concentration of 1 g/L. From Figure 3 and Table 2, the removal efficiency of the pesticide and COD is low in acidic (pH 2.3) electrolyte, meanwhile, in neutral (pH 3.7 in normal) and alkaline medium, the removal efficiency is much higher using (SS) and Fe electrodes. Also, as pH increases the dissolved metal during the electrocoagulation process increases due to the formation of metal hydroxide species which absorb the pesticide molecules and causes the increase of the removal efficiency [36,40,41].

Effect of initial pesticide concentration

To determine influence of initial pesticide concentration on pesticide and COD removals efficiencies during electrocoagulation, different initial concentrations in the range of 50–200 mg/L were treated at 60 min using (SS) and (Fe) electrodes in the optimum condition (a current density of 87.5 mA/cm² at (SS) and 50 mA/cm² at (Fe) electrodes), NaCl concentration of 1 g/L a temperature of 30°C, pH of 3.7 and inter electrode distance of 1 cm. Results showed that when pesticide concentration increased from 50 to 200 mg/L, removals efficiencies decreased (Figure 4 and Table 2). One of the most important pathways of pesticide removal by electrocoagulation is adsorption of pesticide molecules on metallic hydroxide flocs. The adsorption capacity of flocs is limited and specific amount of flocs is able to adsorb specific amount of pesticide molecules [42]. So, with increasing of pesticide concentration, amount of produced flocs is insufficient to adsorb all pesticide molecules, therefore pesticide and COD removal decreases.

Effect of temperature

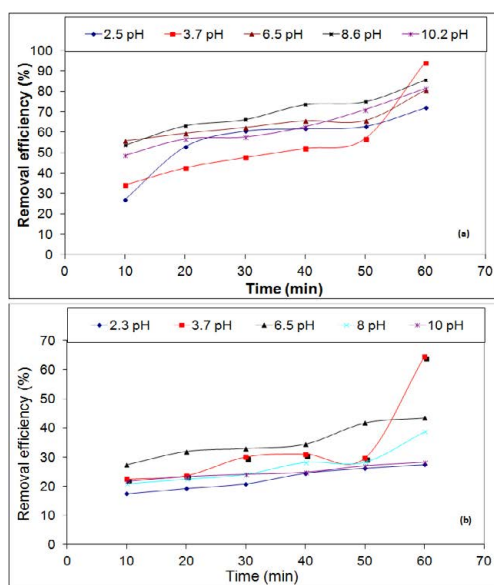
Figure 5 and Table 2 show the effect of temperature on abamectin removal and COD at 60 min using (SS) and (Fe) electrodes at initial concentration of 150 mg/L, (a current density of 87.5 mA/cm² at (SS) and 50 mA/cm² at (Fe) electrodes), inter electrode distance of 1 cm, pH of 3.7 and at NaCl concentration of 1 g/L. The results from Figure 5 and Table 2 indicate that increasing temperature has a negative effect on removal efficiency of pesticide and COD, where at 30°C the pesticide removal and COD percentage reached to 94 and 76.9 using (SS) electrode and reach to 64.5 and 50 using Fe electrode respectively. While at higher temperature value (40°C) the pesticide removal and COD percentage dropped to 78.6 and 63.9 using (SS) electrode and dropped to 53.8 and 42.2 using Fe electrode respectively. More dropping occur at 50°C using the two electrodes. However, it should be noted that the operation of electrocoagulation process at higher temperature significantly reduced electrical energy consumption [43]. So, the production of hydroxide species increases rapidly.

Effect of type of electrolyte

Figure 6 and Table 2 explain the effect of electrolyte type on the removal efficiency of abamectin and COD at 60 min using SS and Fe electrodes in the presence of different supporting electrolytes including NaCl, KCl, CaCl₂, KI was studied at initial concentration of 150 mg/L, (a current density of 87.5 mA /cm² at (SS) and 50 mA/cm² at (Fe) electrodes), inter electrode distance of 1 cm, a temperature of 30°C, pH of 3.7 and at NaCl concentration of 1 g/L. It can be seen from Figure 6(a) and Table 2(a) that in the presence of chloride ions of NaCl, KCl, CaCl₂ electrolytes the removal efficiency of pesticide were [94, 73.7 and 58.3%] and COD [76.95, 67, 52.5%] using SS electrode. But in another electrolytes which not contain chloride ions such as KI, the removal pesticide and COD efficiency dropped to 73% and 52.5% respectively at the same time. Also Figure 6(b) and Table 2(b) explain that in the presence of chloride ions of NaCl, KCl, CaCl₂ electrolytes, the removal efficiency of pesticide were [64.5, 33.6 and 33%] and COD [50, 30.5,

SS (a)					
Current density (mA/cm ²)	25	50	75	87.5	100
COD (%)	38.8	42.1	58.8	76.9	55.1
pH	2.3	3.7	6.5	8.6	10.2
COD (%)	61.6	76.9	64.9	67	65.6
Electrolyte	KCl	CaCl ₂	NaCl	KI	-
COD (%)	67	52.5	76.9	61.2	84.6
[NaCl] (g/L)	0.7	1	1.5	2	-
COD (%)	43.2	76.9	46.6	44.9	-
[PESTICIDE] (mg/L)	50	100	150	200	-
COD (%)	87	76	76.9	35.4	-
Temperature (°C)	10	20	30	40	50
COD (%)	26.2	45.2	76.9	63.9	51.9
Fe (b)					
Current density (mA/cm ²)	25	37.5	50	62.5	
COD (%)	35.6	36	50	43.7	
pH	2.3	3.7	6.5	8.6	10
COD (%)	21.7	50	48.5	46	36
Electrolyte	KCl	CaCl ₂	NaCl	KI	-
COD (%)	30.5	29	50	30.1	-
[NaCl] (g/L)	0.5	1	1.5	2	-
COD (%)	23.4	50	48.3	42.3	-
[PESTICIDE] (mg/L)	50	100	150	200	-
COD (%)	87.3	77	50	42.3	-
Temperature (°C)	10	20	30	40	50
COD (%)	28.3	40	50	42	40.3

Table 2: Effect of current density, pH, Type of electrolyte, Concentration electrolyte, pesticide concentration, inter electrode distance and temperature on the efficiency of COD removal for abamectin using SS (a) and Fe (b) electrodes. Initial concentration of the pesticide =150 mg/L, volume of the solution =100 ml, [NaCl] concentration =1 g/L, (a current density of 87.5 mA /cm² at (SS) and 50 mA/cm² at (Fe) electrodes), pH =3.7, inter electrode distance =1 cm, dimension of the electrodes =20 mm × 10 mm and temperature =30°C.



Initial concentration of the pesticide=150 mg/L, volume of the solution=100 ml, [NaCl] concentration=1 g/L, (a current density of 87.5 mA /cm² at (SS) and 50 mA/cm² at (Fe) electrodes), inter electrode distance=1 cm, dimension of the electrodes=20 mm × 10 mm and temperature=30°C.

Figure 3: Effect of pH on the efficiency of abamectin removal using SS (a) and Fe (b) electrodes

29%] at the same time using Fe electrode respectively. Later experiments were done using NaCl because it is cheap and the solution contains it has high conductivity thus it need low voltage for electrocoagulation and so it is economical in industrial scale.

Energy consumption

In an electrochemical process, the most important economical parameter is energy consumption E (kWh/m³) [44, 45]. This parameter is calculated from the following expression:

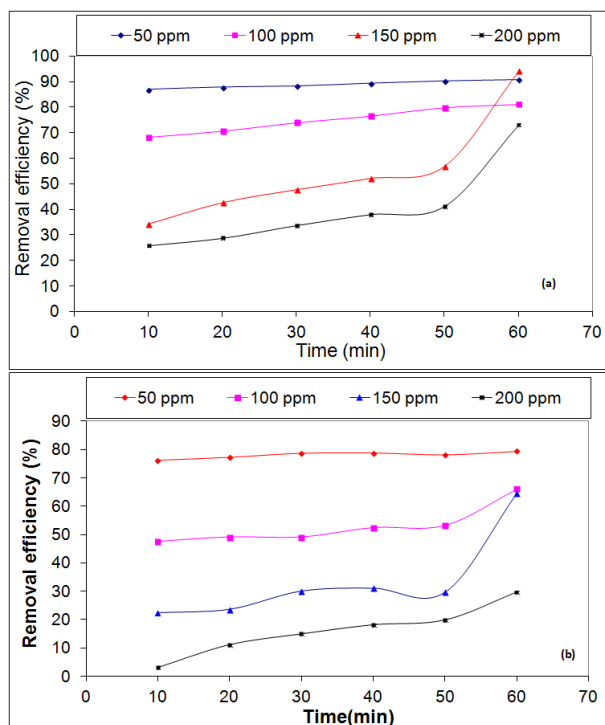
$$E_c = \frac{V.I.t}{Volume.1000}$$

where V, I and t stand for average voltage of the EC system (V), electrical current intensity (A) and reaction time (h), respectively.

Kinetic studies

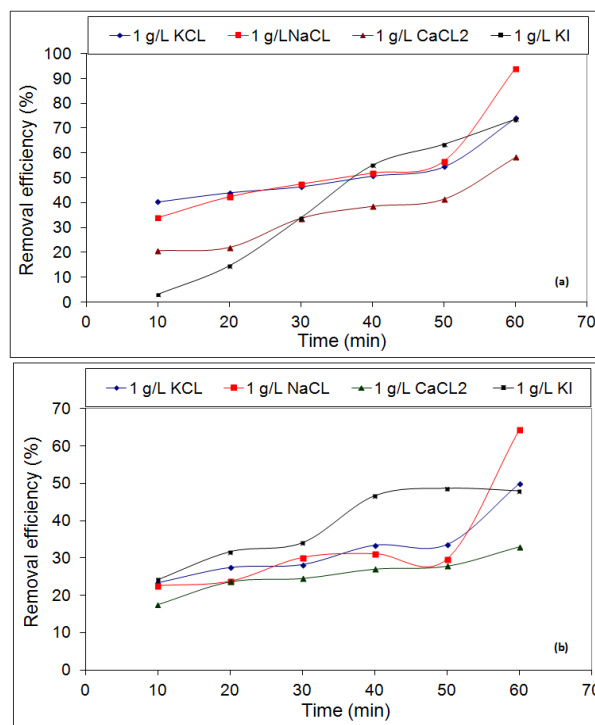
Kinetics studies have important role in determining the rate constant and the order of reaction of this treatment removal [46]. So, rate constant is very significant in the design of wastewater treatment units. It is very essential to know the type of reaction rates for design a wastewater treatment unit [47-50]. Rate of reaction describes the rates of change in concentration of reactant per unit time. Figure 7 represents the removal of pesticide exhibited pseudo first order with good correlation coefficients (0.9989 for SS electrodes) according to following equation:

$$\ln A/A_0 = -kt \quad (10)$$



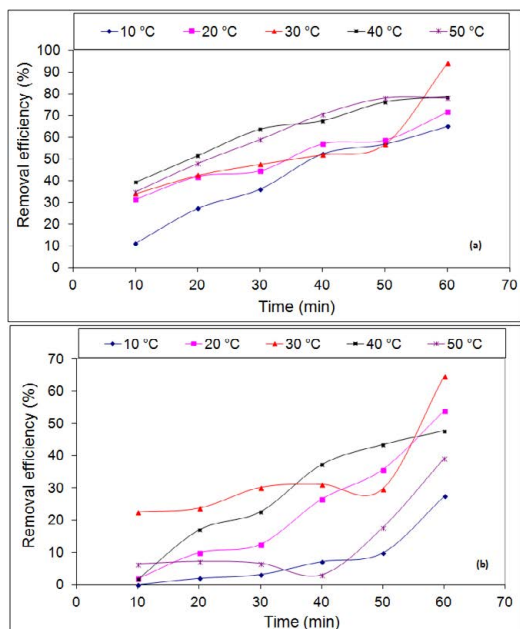
[NaCl] concentration=1 g/L, volume of the solution=100 ml, (a current density of 87.5 mA/cm² at (SS) and 50 mA/cm² at (Fe) electrodes), pH=3.7, inter electrode distance=1 cm, dimension of the electrodes=20 mm × 10 mm and temperature =300° C.

Figure 4: Effect of initial concentration on the efficiency of abamectin removal using SS (a) and Fe (b) electrodes.



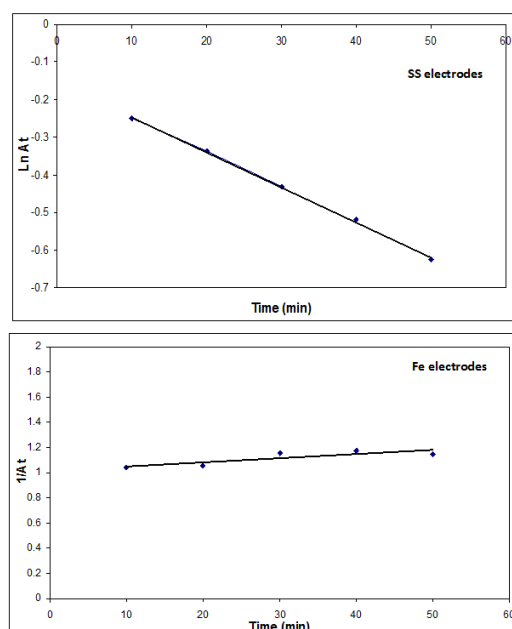
Initial concentration of the pesticide=150 mg/L, volume of the solution=100 ml, (a current density of 87.5 mA /cm² at (SS) and 50 mA/cm² at (Fe) electrodes), pH=3.7, inter electrode distance=1 cm, dimension of the electrodes=20 mm × 10 mm and temperature=300° C.

Figure 6: Effect of type of electrolyte on the efficiency of abamectin removal using SS (a) and Fe (b) electrodes.



Initial concentration of the pesticide =150 mg/L, volume of the solution=100 ml, (a current density of 87.5 mA/cm² at (SS) and 50 mA/cm² at (Fe) electrodes), pH=3.7, [NaCl] concentration =1 g/L, dimension of the electrodes =20 mm × 10 mm and inter electrode distance=1 cm.

Figure 5: Effect of temperature on the efficiency of abamectin removal using SS (a) and Fe (b) electrodes.



Initial concentration of the pesticide =150 mg/L, volume of the solution =100 ml, (a current density =87.5 mA /cm² at (SS) and 50 mA/cm² at (Fe) electrodes), pH=3.7, NaCl concentration=1 g/L, inter electrode distance=1 cm, dimension of the electrodes=20 mm×10 mm and temperature=300° C.

Figure 7: Relation between Ln At and 1/At against the time for abamectin removal using SS (a) and Fe (b) electrodes

Type of degradation	Reference	% Degradation	pH	Time
Photo-fenton process	[6]	70 %	pH 2.5	60 min
UV Shielding	[7]	40%	Nutral	24 hours
Electrochemical oxidation	[2]	98%	PH 6.7	2.5 hours
Adsorption method	[8]	14.4% 9.1 %	PH 12 PH 7	150 min
Electrocoagulation method	This paper	94 % SS electrode	3.7	60 mim
Electrocoagulation method	This paper	64.5 Fe electrode	3.7	60 min

Table 3: Comparison between the Electrocoagulation method for removal of abamectin with other methods.

and exhibited pseudo second order with good correlation coefficients (0.97 for Fe electrodes) according to following equation:

$$1/A_t - 1/A_0 = Kt \quad (11)$$

Where, A_0 , A_t , t , and k are the pesticide absorbance at initial concentration, pesticide absorbance at each time, time of reaction (min), and reaction rate constant, respectively. The values of rate constants at optimum condition and reaction time were 0.0093 min^{-1} and $0.0033 \text{ mol}^{-1}\text{dm}^3\text{min}^{-1}$ using SS and Fe electrodes respectively. Results show that the removal rate using SS electrode was higher than the removal rate using Fe.

Comparison with other method :

The percentages of degradation for each method using in literature and the electrochemical method in this work were represented in the Table 3. It is clear that the electrochemical degradation is the best.

Conclusion

The removal efficiency of abamectin from aqueous solution was examined by electrocoagulation using stainless steel (SS) and iron (Fe) electrodes [51]. The effects of initial pH, initial abamectin concentration, current density, type electrolyte, salt concentration, and temperature were investigated on removal efficiency and COD.

It was observed that these variables significantly affected the abamectin pesticide removal efficiency. The optimum abamectin pesticide removal was obtained with typical operating conditions: an initial pH of 3.7, an initial pesticide concentration of 150 mg/L, (current density 87.5 mA/cm^2 of and 50 mA/cm^2 for stainless steel (SS) and iron (Fe) electrodes respectively), salt concentration of 1 g/L and temperature of 30°C , the results showed that abamectin and COD removal were 94% and 76.9% by using SS and were 64.5% and 50%, by using Fe electrodes. The removal of pesticide was exhibited pseudo first order with rate constant 0.0093 min^{-1} for SS electrode and pseudo second order with rate constant $0.0033 \text{ mol}^{-1}\text{dm}^3\text{min}^{-1}$ using Fe electrodes.

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