

Malathion removal by electrocoagulation process: iron and stainless-steel electrodes, direct and alternating current and determining energy and electrode consumption and kinetic study

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ABSTRACT

The aim of this study was to investigate the effect of electrocoagulation (EC) process for malathion removal from aqueous solution. Iron and stainless-steel rod electrodes were used in the electrochemical cell and effective operation parameters on the electrochemical process were studied. The results showed that with increasing reaction time, current density, removal efficiency of the malathion increased, but the initial concentration of malathion had a reverse effect on the malathion

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removal. The maximum removal efficiency was acquired at pH: 6 (94.6%). In the inter-electrode distance of 2 cm, the removal efficiency of malathion was 95.6% and 95.8% for iron and 92.7% and 88.1% for stainless steel rod electrodes using alternating and direct current, respectively. The maximum energy consumption was 0.91 and 3.12 kWh/m³ using iron rod electrodes, and 0.99 and 3.18 kWh/m³ by stainless steel rod electrodes for the alternating and direct current, respectively. Sludge produced in direct current mode is higher than alternative current mode. The pseudo-second-order model was better fitted for the removal of malathion by the EC process than the pseudo-first-order model (R^2 : 0.99). According to the results, this process is suggested as an effective and efficient method for removing malathion pesticide.

Keywords: Electrochemical process; Environmental pollution; Pesticide separation; Sludge production; Energy consumption

1. Introduction

One of the most important issues related to public concern is water pollution by organic and inorganic compounds [1]. Water contaminants can be created by industrialization, urbanization as well as agricultural activities [2]. Pesticides, that are chemicals or biological substances, are used to control fungi, microorganisms, insects, unwanted plants (weeds) and other pests [3]. The variety and quantities of the pesticides that contaminate water through the extensive use of them and the intensive development of new chemicals in agriculture, domestic and industrial activities and also as well as chemical warfare agents like nerve agents, have dramatically increased [1,4]. They are in different environments (i.e. soil, air, and water) and may remain in the soil or may leach into streams, run-off and ground waters [5–7]. The most applied group of insecticides has been used for the last two decades in the world are organo-phosphorus pesticides (OPs) and are frequently detected in surface and ground waters. In 2001, the OPs insecticides were 70% of the total usage of insecticide in the United States and malathion (32%-44% of total OPs insecticide) was the most widely used [3,8]. In addition, lindane and malathion have been widely used in India [1] [25-30B]. Malathion is highly toxic for aquatic organisms [9,10], amphibians [11,12], vertebrate, and even human beings [6,13]. In small amounts, malathion is acutely toxic and at higher concentrations may create serious body disorders resulting in death by the inability of the respiratory system [5]. In addition, malathion is genotoxic by the mechanism of DNA damage [14,15]. Additionally, according to the experiments of embryo development, malathion indicates an endocrine disruptor and finally inhibited reproductive functions [16,17]. The toxicity of OPs is attributed to the binding of these compounds to the acetylcholinesterase and the concluding deactivation of the enzyme [4,18]. According to the European Economic Community directive pesticide concentration in drinking water should be below 0.1 µg/L [19] and also maximum permissible limit has been recommended 0.1 µg/L for individual pesticides and 0.5 µg/L for total pesticides in drinking water and 1–3 μ g/L for surface waters [5].

Some of the conventional physicochemical treatment processes that have been used for pesticide removal from aqueous solutions include activated carbon, steam-stripping, coagulation/flocculation, resin adsorption, photocatalysis (UV photolysis and TiO₂ photocatalysis), advanced chemical oxidation, membrane processes and hydrolysis [5,19– 22]. Each method has its own merits and limitations. For example, carbon adsorption becomes saturated rapidly, and their efficiency decreases with the presence of competitive material contained in the matrix and coagulation/ flocculation and sedimentation are used as a pretreatment process before the advanced treatment [3,23–26].

In recent years, researchers have attracted to focus on electrocoagulation (EC) for the treatments of various types of water as well as wastewaters [27]. EC is an environmentally friendly, simple technology and does not generate secondary pollutants and including the sacrificial metal (usually aluminum or iron) and the power of electricity [28,29]. Some advantages of this process are short reaction time, simple operation and require simple equipment, easy to operate, compact size, absence of adding chemicals, rapid sedimentation of the produced flocs, less sludge production, low capital and operating costs [27,30]. The disadvantage of EC is that high conductivity water is needed (especially for drinking water treatment) [28]. The EC process includes applying an electric current (both alternating and direct current) to sacrifice electrodes and then the generation of coagulants in situ. When iron or aluminum electrodes use as anodes, trivalent iron or aluminum produces [31]. EC reactors could be operated by monopolar and dipolar connection [30,32]. The EC technology has been widely studied for the treatment of effluents from tannery [33], slaughterhouse [29], organic material, textile [34], drug manufacturing industry as well as for the removal of heavy metals [35]. The ability of malathion for creating toxicity and carcinogenicity has led to concern about environmental contamination of this pesticide. On the other hand, malathion is a very toxic pesticide that exists in water resources in some countries like Iran, India and other countries. Therefore, the removal of this pesticide is very important in the view of environmental contamination and human health [7].

The overall objective of this research was to investigate malathion removal from water using EC to obtain optimum conditions.

2. Material and methods

2.1. Material and equipment

All chemicals and materials used in this study were of analytical grade and were obtained from Merck, (Germany) and Sigma Chemical Co., (USA). Stock solutions of malathion were prepared in *n*-hexane. In order to achieve the desired pH, HCl (1 N) or NaOH (1 N) were used. pH was measured by pH meter (Hach, USA) and the concentrations of malathion were determined by high-performance liquid chromatography (HPLC) using UV-Vis detection with an autosampler fitted with a fused octadecyl silica gel (C18) column (length: 25 mm and diameter: 4.6 mm) (Water Company, USA) for separating of these compounds. The mobile phase was methanol, acetonitrile, and water with a ratio 15:45:40 and a velocity of 1 mm/min. The wavelength used for the detection of these compounds was ascertained at 140 nm. The separating column used in the HPLC set was the DB5.6.25 capillary column. The application vertex for malathion was obtained at 10.23 min. The temperature program was initially held at 160°C for 1 min, then raised at 25°C/min to 188°C and hold at 188°C for 4 min and again raised to 190°C at 20°C/min for 1 min and finally maintained at 244°C for 6.21 min. Helium was used as a carrier gas with pureness >99% and a velocity rate of 0.7 cm/s.

A stock solution of malathion was prepared according to standard methods. For the preparation of working samples, the stock solution was added to the predetermined quantity of distilled water. The experiments were done with a stock solution of 1,000 mg/L.

The experimental set-up used in this study has been shown in Fig. 1. The electrochemical cell was made of plexiglass with a dimension of 12 cm × 10 cm × 12 cm (efficient volume: 1 L). Acetone was used for fat removal from the surface of the electrodes. A magnetic stirrer has been used for mixing at the rate of 300 rpm in the whole stages of the study. Voltmeter and ohmmeter that installed on the circus, measured the electrical potential differences and current intensity, respectively. Iron rods were used with 50 mm length and 5 mm diameter and 15 mm apart from each other. Rod electrodes were connected as monopolar. The alternating and direct current was provided by a power source (model GW GPC-3060D).

2.2. Experiment

In this study pH, current densities, initial malathion concentration, reaction time, amount of energy used and kind of electrode, current type (alternative or direct current), distances between the electrodes and the amount of sludge produced were studied. To evaluate each stage, the synthetic wastewater inside the reactor was agitated by a magnetic mixer and samples were taken from the reactor in order to observe the treatment progress in time intervals of 10, 20, 30 and 40 min. Then the samples were passed through 0.45 μ m filters and the filtered samples were stored at 4°C and eventually, samples were analyzed for malathion residual. By the end of each stage of the study, the pH of the solution after the EC process and the amount of producing sludge were measured in order to find the number of flocs and the fluctuates in pH during the process.

2.3. Calculations

One of the most important economic parameters for the application of the EC process is the amount of electrical energy consumed. This parameter is determined by the following equation:

$$E = \frac{UIt}{V} \tag{1}$$

where *E* is the electrical energy consumption (kWh/m³), *U* is the potential difference (V), *I* is the current rate (A), *t* is the reaction time (h) and *V* is the volume of the reactor (L). The amount of sacrifice an anode is determined by measuring the initial and final weight of the anodes.

The amount of electrochemical adsorbent (aluminum or iron hydroxide) formed is determined by the Faraday Law:

$$C = \frac{ItM}{ZFV}$$
(2)

where *C* (kg electrode/m³ potable water treated) is the amount of ion produced by applied current (A) passed for a duration of time *t* (s), *Z* is the number of electrons involved in the oxidation/reduction reaction (chemical equivalent) (2 for Fe), *M* is the atomic weight of anode (Fe = 0.05585 kg/mol), *F* is the Faraday's constant rate (96,485 C/mol) and *V* is the volume (m³) of the potable water in the EC reactor.



Fig. 1. Electrocoagulation reactor schematic used in this study.

3. Results

3.1. Effect of current density and time on malathion removal

Current density plays an important role in the EC process. Table 1 showed the effect of current density and time on the malathion removal in the EC process using two types of current (alternating and direct) and two types of rod electrode (iron and stainless steel). Totally, as current density increases, the removal of pesticides also increases. According to these results, the highest pesticide removal was obtained in current density 10 mA/cm² for both current and electrode. In addition, 40 min was found the best time for removing malathion by the EC process. The scarification of anode and dosage of coagulant produced as well as generation of hydrogen gas at cathode could be affected by current density. The mass transfer between bubbles generated, contaminants and coagulants might be determined with the current density [36].

3.2. Effect of initial pH

pH is an important parameter and has a significant effect on the formation of hydroxide metal types [37]. The effect of pH on the EC process with alternating and direct current and iron and stainless-steel rod electrodes for removing malathion pesticide are shown in Fig. 2. The removal efficiency of malathion was increased with increasing the pH and the maximum removal efficiency of malathion was obtained at pH 6 (94.6% and 92.2% using an iron electrode and 83.6% and 80.9% using stainless steel electrode for alternative current (AC) and direct current (DC) current, respectively. The malathion removal efficiencies decreased at low and high pH values. The solution pH as a key factor in EC has a significant effect on the zeta potential, the solution conductivity and the dissolution of the electrode [38]. When solution pH increases from 4 to 7, the amount of insoluble hydroxide significantly also increases and there are no hydroxide ions of metals in this pH range [36].

3.3. Effect of the initial concentration of malathion

Table 2 discloses the effect of the initial malathion concentration on the removal efficiency of malathion for various times (from 5 to 60 min). It was expected that the malathion removal attained 97.2% after 60 min for the initial malathion concentrations of 15 mg/L in alternating current and iron rod electrodes. The malathion removal efficiency decreased when the initial concentration of malathion was increased.

3.4. Distance between electrodes

The effect of the distance between electrodes is presented in Fig. 3. It has been cleared that maximum removal efficiency with both electrodes (iron and stainless steel) and electrical current mode (AC and DC) was reached at a distance of electrodes 2 cm. The removal efficiency of malathion increases with increasing the inter-electrode distances up to 2 cm and then decreases.

In this inter-electrode distances (2 cm), the removal efficiency of malathion was 95.6% and 95.8% for iron rod electrodes and 92.7% and 88.1% for stainless steel rod electrodes using alternating and direct current, respectively.

Table 1

The efficiency of malathion removal (%) using various electrical current density and electrodes

Iron										
Time (min)			DC							
	Current density (mA/m ²)					Current density (mA/m ²)				
	2	4	6	8	10	2	4	6	8	10
5	15.6	21.2	32.5	55.3	82.5	13.8	19.1	25.1	53.2	80.3
10	30.2	55.5	74.6	83.2	92.5	27.6	52.8	71.3	80.2	90.7
20	60.23	75.8	82.1	90.2	95.6	57.6	73.6	79.9	88.5	90.7
30	71.7	77.6	91.3	94.3	97.1	68.9	70.3	89.9	92.7	95.5
40	75.6	85.9	94.2	95.6	97.9	73.4	82.9	92.2	94.8	96.1
50	77.8	87.3	95.1	96.7	98.1	74.2	83.2	92.6	95.1	96.8
60	78.1	88.2	95.9	96.8	97.8	74.9	83.5	93.7	95.6	96.5
Stainless	steel									
5	12.3	16.7	21.9	45.8	68.2	10.6	14.2	19.3	43.6	66.1
10	21.7	42.3	59.1	72.3	74.5	19.3	39.2	56.9	68.9	71.6
20	48.7	65.7	72.5	84.8	81.9	45.9	61.9	70.2	81.6	78.9
30	62.5	72.4	81.6	87.9	88.9	60.1	70.2	78.9	85.3	87.1
40	64.5	76.7	83.6	90.6	90.9	61.3	73.6	80.9	88.4	88.8
50	65.1	76.8	84.9	90.9	91.6	61.5	73.8	81.9	89.3	90.8
60	64.9	77.8	87.9	92.6	93.8	63.6	75.6	84.9	91.8	91.8



Fig. 2. Effect of pH on removing malathion using iron and stainless steel electrodes with direct and alternating current (current density 6 mA/cm²; contact time 40 min; the distance between electrodes 2 cm; mixing rate 200 rpm).

Table 2

Effect of the initial concentration of malathion on the removal efficiency of malathion using iron and stainless steel electrodes with direct and alternating current (current density 6 mA/cm², the distance between electrodes 2 cm, mixing rate 200 rpm, pH: 6)

Iron										
	AC					DC				
Time (min)	C (mg/L)					C (mg/L)				
(mm)	15	40	50	75	100	15	40	50	75	100
5	68.8	57.5	31.5	13.5	10	66.2	55.2	28.9	11.5	8.5
10	81.2	68.9	58.6	33.3	20	78.3	66.2	56.3	31.8	18.3
20	93.6	83.5	72.1	60.2	45	90.3	81.3	64.6	56.9	42.8
30	96.2	94.2	92.1	70.67	57	92.8	90.5	68.9	61.3	55.1
40	96.8	94.8	92.8	81.2	70	94.2	91.8	89.9	66.9	67.9
50	96.9	95.1	93.1	84.5	83	94.8	92.9	90.3	72.9	70.1
60	97.2	95.6	93.6	87.6	86.8	95.1	93.1	90.8	73.9	72.6
Stainless ste	eel									
5	61.3	51.6	24.9	9.9	8.2	58.2	46.9	22.6	8.8	7.2
10	74.9	61.9	51.9	27.5	14.9	70.6	58.7	49.3	24.9	16.9
20	86.9	74.6	64.9	52.9	36.4	81.9	72.9	55.8	48.2	38.9
30	87.3	85.9	79.6	62.8	48.9	82.6	78.6	67.6	54.6	49.1
40	88.9	86.8	81.6	76.8	61.8	83.9	79.3	78.9	61.3	61.9
50	90.3	87.2	82.9	78.5	74.9	85.7	80.6	79.3	67.2	83.6
60	90.8	87.9	85.9	81.6	79.3	86.2	81.1	80.2	68.8	84.2

3.5. Amount of energy and electrode consumption

The most important economic parameter in the EC process is the amount of electrical energy used in this process. According to obtained results, when the current density increases from 2 to 6 mA/cm², the energy consumption slightly increased in iron and stainless-steel rod electrodes with alternating and direct current (Fig. 4). Then in the current density 6 to 10 mA/cm², energy

consumption in alternating current slightly increased, but it increased significantly in direct current. The maximum energy consumption was 0.91 and 3.12 kWh/m³ using iron rod electrodes, and 0.99 and 3.18 kWh/m³ stainless steel rod electrodes with alternating and direct current, respectively.

The amount of electrode consumption in removing malathion using alternating and direct current is shown in Fig. 5. The highest amount of iron rod electrodes used for malathion removal was 1.06 and 1.82 kg/m³ and for stainless



Fig. 3. Effect of inter-electrode distances on the removal efficiency of malathion using iron and stainless steel electrodes with direct and alternating current (current density 6 mA/cm²; initial concentration of malathion: 50 mg/L; mixing rate 200 rpm; pH: 6).



BEEREEN Iron, AC III Iron, DC --- Stainless Steel, AC --- Stainless Steel, DC

Fig. 4. The amount of energy consumption in removing malathion using alternating and direct current (current density: 6 mA/cm²; pH: 5; contact time: 40 min; the distance between electrodes: 2 cm; mixing rate: 200 rpm).

steel, rod electrode was 1.05 and 1.58 0.9 kg/m³ by using alternating and direct current, respectively.

3.6. Amount of sludge produced from EC

Sludge production creates some problems related to disposal and transfer of solid waste from this process. The amount of sludge production in removing malathion using alternating and direct current is illustrated in Fig. 6. The highest amount of sludge was produced 0.093 and 0.095 kg/m³ and in iron rod electrodes and 0.105 and 0.112 kg/m³ in steel electrodes using alternating and direct current, respectively.

3.7. Kinetic study

Fig. 7 illustrates the plot and the coefficient of kinetic model study for the malathion removal. According to these results, the pseudo-second-order kinetic model presents a good agreement with experimental data for different removal rates. The correlation coefficient (R^2) for this model was 0.99.

4. Discussion

4.1. Effect of current density and time on malathion removal

Current density has a significant impact on the removal efficiency of the malathion. The coagulant dosage rate



Fig. 5. The amount of electrode consumption in removing malathion using alternating and direct current (current density: 6 mA/cm²; pH: 5; contact time: 40 min; the distance between electrodes: 2 cm; mixing rate: 200 rpm).



Iron, AC VIIII Iron, DC - Stainless steel, AC - Stainless steel, DC

Fig. 6. The amount of sludge produced in alternating and direct current using iron and stainless steel electrodes (current density: 6 mA/cm²; pH:6; contact time: 40 min; distance between electrodes: 2 cm; mixing rate: 200 rpm).

and controlling the reaction rate inside the electrochemical reactor can be determined by the current density. The results showed that the removal efficiency of the malathion increased with the increasing current density. At a high current density, the extent of anodic dissolution increases, resulting in a greater amount of $Fe(OH)_{n(s)}$ flocs and the rate of floc growth. Higher amounts of the dissolved coagulant allowed higher coagulation efficiency and promoting the removal of the pollutants. In addition, more effective alternating current and iron rod electrodes probably are due to the uniform decomposition of the electrodes in the alternating current. These results are in line with other researches [39]. Reaction time also affects the removal efficiency of the EC process. This factor specifies the production rate of Fe^{2+} or Fe^{3+} ions. According to the results, the removal efficiency increases with the increase of reaction time up to 40 min and then no significant improvement in the removal efficiency is observed. These results are observed by other researchers [29].

4.2. Effect of initial pH

pH plays an important role in chemical or electrochemical segregation processes and has a significant effect as an important operating factor on the mechanism of pollutant



Fig. 7. Kinetic study of malathion removal by electrocoagulation with alternative current and iron rod electrodes (current density: 6 mA/cm²; pH: 6; the distance between electrodes: 2 cm; mixing rate: 200 rpm).

removal and formation of hydroxide metal types. The maximum removal of malathion pesticide was found at pH: 6 and this is in line with many previous works related to the EC process [40].

At high and low pH, removal efficiency is low and it was attributed to an amphoteric behavior which leads to soluble of Fe(OH)₂ and production of proton reduced in the cathode that leads to inhibit of hydroxide ion production (at acidic pH) and hydroxide ions are oxidized at the anode to monomeric anions $Fe(OH)_{6}^{3-}$ and $Fe(OH)_{4}^{-}$ (at alkaline pH) and/or interaction the hydroxyl metal ion complexes formed at these pH values with the hydronium ion (H_2O^+) . It is obvious that these soluble types of flocs are not appropriate for the removal of pollutants [41,42]. Most of the metal ions formed at the anode produced polymeric species and precipitated as three valent metal ions hydroxide leading to further removal efficiency. This result was similar to the results from the Şengil and Özacar study in the decolorization of C.I. reactive black 5 in aqueous solution by EC using sacrificial iron electrodes and Al-Shannag study in chemical oxygen demand reduction of baker's yeast wastewater using batch EC [42].

4.3. Effect of the initial concentration of malathion

According to obtained results, the malathion removal efficiency decreased, as the initial concentration was increased. According to Faraday's law, the anode electrode releases a constant amount of metal hydroxides for the same current density and reaction time for all malathion concentrations. So, the amount of metal hydroxides produced in the solution is similar. Consequently, at a constant current density, the malathion removal efficiency was decreased with increasing initial concentration.

4.4. Distance between electrodes

An increase in removal efficiency was observed while increasing the inter-electrode distance (up to 2 cm) and then removal efficiency decreases. The distance between electrodes impacts on the amount of energy consumption and production of metal ion hydroxide. The decrease of the distance between electrodes led to a slight decrease in removal efficiency of malathion pesticide. These results depend on the electrostatic field change. Electrostatic field decreases with increasing the distance between electrodes and decreasing the electrostatic field may cause to decrease the movement of produced ions during the reaction time and then the opportunity of aggregation of metal ions and floc production is improved. Moreover, the ability of malathion adsorption and binding of malathion in solution by a metal hydroxide floc is enhanced [43,44]. In addition, during the EC process, a very fine film of metal hydroxide would be formed on the anode electrodes and this film creates the resistance that reduces the rate of anode oxidation and production of metal hydroxide flocs. In the current density constant, this resistance increases with increasing the inter-electrode distances and followed by the rate of anode oxidation and production of metal hydroxide flocs as well as adsorption of malathion on flocs would be decreased. So, removal efficiency also decreases with increasing the inter-electrode distances. Similar behavior has already been reported by previous researchers [43].

4.5. Amount of energy and electrode consumption

According to the electrical energy consumed equation, the amount of required energy depends on potential differences, current and reaction time. Energy consumption increased when reaction time increased. This parameter is the major operating cost of the EC process. The energy consumptions for iron electrodes were a little lower than that for stainless steel electrodes. The potential required to attain a certain current density for the stainless-steel electrode rod is probably higher than the iron electrode. In addition, according to Faraday rule, electrode consumption depends on current, time, the chemical equivalent of electrode and volume. So, the high current density and time also lead to more increase in electrode consumption or a decrease in the weight of the electrodes. These results are in agreement with some previous studies [45]. The total cost of operation activity in the EC process at full-scale plant depends on the electrode type, water/wastewater characteristics, energy consumption, sludge management and operation and maintenance cost. Based on the previous study on the EC process with the monopolar operation, iron electrode, the retention time of 15 min, pH: 7, the current density of 30 A/m² was obtained about 0.25 \$/m³. In addition, the cost of the EC process was calculated to be 3.2 times lower than conventional coagulation [38].

4.6. Amount of sludge produced from EC

One of the most important factors in the EC process is problems of sludge handling (sludge dewatering) and its disposal. Sludge produced in DC mode is higher than AC mode. Increase in the sacrifice of the anode, the floc production and the hydrogen generated at the cathode which increased the flotation efficiency and the produced sludge are as results of an increase in current density and reaction time. The amount of sludge production is attributed to increasing the operating cost. This is confirmed by many previous studies related to the EC process for removing many pollutants [46].

4.7. Kinetic study

Studying the kinetics of the reaction is essential to selecting the optimum conditions, predicting the reaction rate, understand the dynamics of the reactions [47,48]. In this study, the kinetics data for malathion removal by the EC process were analyzed by the MS Excel Solver function spreadsheet to find out higher fitness of the models and predict the curve fitted with experimental values. According to Fig. 7, the pseudo-second-order model was slightly higher than the pseudo-first-order model which illustrated that the pseudo-second-order model was better fitted for the removal of malathion by the EC process than pseudo-first-order model (R^2 : 0.99) [49–53].

5. Conclusion

EC process is an electrical current based method that forms the floc through the scarification of anode electrodes and generates the gas at the cathode electrodes. This method is an eco-friendly process that removes contaminants such as organic toxins. In the present study, the EC process was employed for malathions removal as a phosphorous pesticide. Based on these results, the following conclusion can be taken:

- Removal efficiency of the malathion increased with the increasing current density.
- Maximum removal of malathion pesticide was found at pH: 6.
- Increase in removal efficiency was observed while increasing the inter-electrode distance (up to 2 cm) and then removal efficiency decreases.

- High current density and time also lead to more increase in electrode consumption or a decrease in the weight of the electrodes.
- Sludge produced in DC mode is higher than the AC mode.
- The pseudo-second-order model was better fitted for the removal of malathion by the EC process than the pseudo-first-order model.

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References

- V.K. Gupta, C.K. Jain, I. Ali, S. Chandra, S. Agarwal, Removal of lindane and malathion from wastewater using bagasse fly ash—a sugar industry waste, Water Res., 36 (2002) 2483–2490.
- [2] I. Ali, M. Asim, T.A. Khan, Low cost adsorbents for the removal of organic pollutants from wastewater, J. Environ. Manage., 113 (2012) 170–183.
- [3] Y.M. Zhang, K. Pagilla, Treatment of malathion pesticide wastewater with nanofiltration and photo-Fenton oxidation, Desalination, 263 (2010) 36–44.
- [4] A.K. Janeczko, E.B. Walters, S.J. Schuldt, M.L. Magnuson, S.A. Willison, L.M. Brown, O.N. Ruiz, D.L. Felker, L.A. Racz, Fate of malathion and a phosphonic acid in activated sludge with varying solids retention times, Water Res., 57 (2014) 127–139.
- [5] M. Akhtar, S.M. Hasany, M.I. Bhanger, S. Iqbal, Low cost sorbents for the removal of methyl parathion pesticide from aqueous solutions, Chemosphere, 66 (2007) 1829–1838.
- [6] R.R. Kalantary, A. Azari, A. Esrafili, K. Yaghmaeian, M. Moradi, K. Sharafi, The survey of malathion removal using magnetic graphene oxide nanocomposite as a novel adsorbent: thermodynamics, isotherms, and kinetic study, Desal. Water Treat., 57 (2016) 28460–28473.
- [7] A. Kumar, A. Kumar, G. Sharma, A.H. Al-Muhtaseb, M. Naushad, A.A. Ghfar, F.J. Stadler, Quaternary magnetic BiOCl/g-C₃N₄/Cu₂O/Fe₃O₄ nano-junction for visible light and solar powered degradation of sulfamethoxazole from aqueous environment, Chem. Eng. J., 334 (2018) 462–478.
- [8] G.Z. Kyzas, K.A. Matis, Methods of arsenic wastes recycling: focus on flotation, J. Mol. Liq., 214 (2016) 37–45.
 [9] H.B. Yu, X.H. Wang, H.W. Sun, M.X. Huo, Photocatalytic
- [9] H.B. Yu, X.H. Wang, H.W. Sun, M.X. Huo, Photocatalytic degradation of malathion in aqueous solution using an Au-Pd-TiO, nanotube film, J. Hazard. Mater., 184 (2010) 753–758.
- [10] A.M. Donia, A.A. Atia, R.A. Hussien, R.T. Rashad, Comparative study on the adsorption of malathion pesticide by different adsorbents from aqueous solution, Desal. Water Treat., 47 (2012) 300–309.
- [11] H.P. Gurushankara, S.V. Krishnamurthy, V. Vasudev, Effect of malathion on survival, growth, and food consumption of Indian cricket frog (*Limnonectus limnocharis*) tadpoles, Arch. Environ. Contam. Toxicol., 52 (2007) 251–256.
- [12] A. Kumar, Shalini, G. Sharma, M. Naushad, A. Kumar, S. Kalia, C.S. Guo, G.T. Mola, Facile hetero-assembly of superparamagnetic Fe₃O₄/BiVO₄ stacked on biochar for solar photo-degradation of methyl paraben and pesticide removal from soil, J. Photochem. Photobiol., A, 337 (2017) 118–131.
- [13] M. Ahmed, J.B.T. Rocha, C.M. Mazzanti, A.L.B. Morsch, D. Cargnelutti, M. Corrêa, V. Loro, V.M. Morsch, M.R.C. Schetinger, Malathion, carbofuran and paraquat inhibit *Bungarus sindanus* (krait) venom acetylcholinesterase and human

serum butyrylcholinesterase in vitro, Ecotoxicology, 16 (2007) 363–369.

- [14] S. Agarwal, I. Tyagi, V.K. Gupta, M.H. Dehghani, A. Bagheri, K. Yetilmezsoy, A. Amrane, B. Heibati, S. Rodriguez-Couto, Degradation of azinphos-methyl and chlorpyrifos from aqueous solutions by ultrasound treatment, J. Mol. Liq., 221 (2016) 1237–1242.
- [15] A.A. Atia, A.M. Donia, R.A. Hussien, R.T. Rashad, Efficient adsorption of malathion from different media using thermally treated kaolinite, Desal. Water Treat., 30 (2011) 178–185.
- [16] Y. Ducolomb, E. Casas, A. Valdez, G. González, M. Altamirano-Lozano, M. Betancourt, In vitro effect of malathion and diazinon on oocytes fertilization and embryo development in porcine, Cell Biol. Toxicol., 25 (2009) 623–633.
- [17] A.A. El-Kady, H.H. Abdel Ghafar, M.B.M. Ibrahim, M.A. Abdel-Wahhab, Utilization of activated carbon prepared from agricultural waste for the removal of organophosphorous pesticide from aqueous media, Desal. Water Treat., 51 (2013) 7276–7285.
- [18] K. Gangadhara Reddy, G. Madhavi, B.E. Kumara Swamy, Mobilized lipase enzymatic biosensor for the determination of Chlorfenvinphos and malathion in contaminated water samples: a voltammetric study, J. Mol. Liq., 198 (2014) 181–186.
- [19] S. Chatterjee, S.K. Das, R. Chakravarty, A. Chakrabarti, S. Ghosh, A.K. Guha, Interaction of malathion, an organophosphorus pesticide with *Rhizopus oryzae* biomass, J. Hazard. Mater., 174 (2010) 47–53.
- [20] Y. Zhou, L. Zhang, Z.J. Cheng, Removal of organic pollutants from aqueous solution using agricultural wastes: a review, J. Mol. Liq., 212 (2015) 739–762.
- [21] G.Z. Kyzas, K.A. Matis, Electroflotation process: a review, J. Mol. Liq., 220 (2016) 657–664.
- [22] A. Mittal, M. Teotia, R.K. Soni, J. Mittal, Applications of egg shell and egg shell membrane as adsorbents: a review, J. Mol. Liq., 223 (2016) 376–387.
- [23] K.G. Reddy, G. Madhavi, B.E.K. Swamy, S. Reddy, A.V.B. Reddy, V. Madhavi, Electrochemical investigations of lipase enzyme activity inhibition by methyl parathion pesticide: voltammetric studies, J. Mol. Liq., 180 (2013) 26–30.
- [24] I. Mironyuk, T. Tatarchuk, M. Naushad, H. Vasylyeva, I. Mykytyn, Highly efficient adsorption of strontium ions by carbonated mesoporous TiO₂, J. Mol. Liq., 285 (2019) 742–753.
- [25] A.A. Alqadami, M. Naushad, Z.A. Alothman, A.A. Ghfar, Novel metal–organic framework (MOF) based composite material for the sequestration of U(VI) and Th(IV) metal ions from aqueous environment, ACS Appl. Mater. Interfaces, 9 (2017) 36026–36037.
- [26] A. Bagheri, A.H. Mahvi, R. Nabizadeh, M.H. Dehghani, B. Mahmoudi, B. Akbari-Adergani, K. Yaghmaeian, Rapid destruction of the non-steroidal anti-inflammatory drug diclofenac using advanced nano-Fenton process in aqueous solution, Acta Medica Mediterr., 33 (2017) 879–883.
- [27] S. Zhao, G.H. Huang, G.H. Cheng, Y.F. Wang, H.Y. Fu, Hardness, COD and turbidity removals from produced water by electrocoagulation pretreatment prior to reverse osmosis membranes, Desalination, 344 (2014) 454–462.
- [28] K.L. Dubrawski, M. Mohseni, Standardizing electrocoagulation reactor design: iron electrodes for NOM removal, Chemosphere, 91 (2013) 55–60.
- [29] M. Ahmadian, N. Yousefi, S.W. Van Ginkel, M.R. Zare, S. Rahimi, A. Fatehizadeh, Kinetic study of slaughterhouse wastewater treatment by electrocoagulation using Fe electrodes, Water Sci. Technol., 66 (2012) 754–760.
- [30] A. Akyol, O.T. Can, E. Demirbas, M. Kobya, A comparative study of electrocoagulation and electro-Fenton for treatment of wastewater from liquid organic fertilizer plant, Sep. Purif. Technol., 112 (2013) 11–19.
- [31] M. Malakootian, N. Yousefi, A. Fatehizadeh, Survey efficiency of electrocoagulation on nitrate removal from aqueous solution, Int. J. Environ. Sci. Technol., 8 (2011) 107–114.
- [32] M. Malakootian, N. Yousefi, The efficiency of electrocoagulation process using aluminum electrodes in removal of hardness from water, Iran. J. Environ. Health Sci. Eng., 6 (2009) 131–136.

- [33] A.N. Módenes, F.R. Espinoza-Quiñones, F.H. Borba, D.R. Manenti, Performance evaluation of an integrated photo-Fenton – Electrocoagulation process applied to pollutant removal from tannery effluent in batch system, Chem. Eng. J., 197 (2012) 1–9.
- [34] S.M. Palácio, F.R. Espinoza-Quiñones, A.N. Módenes, C.C. Oliveira, F.H. Borba, F.G. Silva, Toxicity assessment from electro-coagulation treated-textile dye wastewaters by bioassays, J. Hazard. Mater., 172 (2009) 330–337.
- [35] A.K. Golder, A.N. Samanta, S. Ray, Removal of trivalent chromium by electrocoagulation, Sep. Purif. Technol., 53 (2007) 33–41.
- [36] J.N. Hakizimana, B. Gourich, M. Chafi, Y. Stiriba, C. Vial, P. Drogui, J. Naja, Electrocoagulation process in water treatment: a review of electrocoagulation modeling approaches, Desalination, 404 (2017) 1–21.
- [37] S. Vasudevan, J. Lakshmi, J. Jayaraj, G. Sozhan, Remediation of phosphate-contaminated water by electrocoagulation with aluminium, aluminium alloy and mild steel anodes, J. Hazard. Mater., 164 (2009) 1480–1486.
- [38] D.T. Moussa, M.H. El-Naas, M. Nasser, M.J. Al-Marri, A comprehensive review of electrocoagulation for water treatment: potentials and challenges, J. Environ. Manage., 186 (2017) 24–41.
- [39] H.J. Mansoorian, A.H. Mahvi, A.J. Jafari, Removal of lead and zinc from battery industry wastewater using electrocoagulation process: influence of direct and alternating current by using iron and stainless steel rod electrodes, Sep. Purif. Technol., 135 (2014) 165–175.
- [40] W. Lemlikchi, S. Khaldi, M.O. Mecherri, H. Lounici, N. Drouiche, Degradation of disperse red 167 azo dye by bipolar electrocoagulation, Sep. Sci. Technol., 47 (2012) 1682–1688.
- [41] P. Asaithambi, L. Garlanka, N. Anantharaman, M. Matheswaran, Influence of experimental parameters in the treatment of distillery effluent by electrochemical oxidation, Sep. Sci. Technol., 47 (2012) 470–481.
- [42] İ.A. Şengil, M. Özacar, The decolorization of C.I. Reactive Black 5 in aqueous solution by electrocoagulation using sacrificial iron electrodes, J. Hazard. Mater., 161 (2009) 1369–1376.
- [43] C.P. Nanseu-Njiki, S.R. Tchamango, P.C. Ngom, A. Darchen, E. Ngameni, Mercury(II) removal from water by electrocoagulation using aluminium and iron electrodes, J. Hazard. Mater., 168 (2009) 1430–1436.
- [44] N. Daneshvar, H. Ashassi Sorkhabi, M.B. Kasiri, Decolorization of dye solution containing Acid Red 14 by electrocoagulation with a comparative investigation of different electrode connections, J. Hazard. Mater., 112 (2004) 55–62.
- [45] L. Zaleschi, C. Teodosiu, I. Cretescu, M.A. Rodrigo, A comparative study of electrocoagulation and chemical coagulation processes applied for wastewater treatment, Environ. Eng. Manage. J., 11 (2012) 1517–1525.
- [46] O.J. Flores, J.L. Nava, G. Carreño, Arsenic removal from groundwater by electrocoagulation process in a filter-presstype FM01-LC reactor, Int. J. Electrochem. Sci., 9 (2014) 6658–6667.
- [47] M. Malakootian, A. Fatehizadeh, N. Yousefi, M. Ahmadian, M. Moosazadeh, Fluoride removal using regenerated spent bleaching earth (RSBE) from groundwater: case study on Kuhbonan water, Desalination, 277 (2011) 244–249.
- [48] M. Naushad, S. Vasudevan, G. Sharma, A. Kumar, Z.A. ALOthman, Adsorption kinetics, isotherms, and thermodynamic studies for Hg²⁺ adsorption from aqueous medium using alizarin red-S-loaded amberlite IRA-400 resin, Desal. Water Treat., 57 (2016) 18551–18559.
- [49] S. Vasudevan, J. Lakshmi, R. Kamaraj, G. Sozhan, A critical study on the removal of copper by an electrochemically assisted coagulation: equilibrium, kinetics, and thermodynamics, Asia-Pac. J. Chem. Eng., 8 (2013) 162–171.
- [50] M. Ahmadian, S. Reshadat, N. Yousefi, S.H. Mirhossieni, M.R. Zare, S.R. Ghasemi, N. Rajabi Gilan, R. Khamutian, A. Fatehizadeh, Municipal leachate treatment by Fenton process: effect of some variable and kinetics, J. Environ. Public Health, 2013 (2013), doi: 10.1155/2013/169682.

120

- [51] M. Naushad, Z.A. ALOthman, M.R. Khan, Removal of malathion from aqueous solution using De-Acidite FF-IP resin and determination by UPLC–MS/MS: equilibrium, kinetics and thermodynamics studies, Talanta, 115 (2013) 15–23.
 [52] M.A. Hossain, H.H. Ngo, W. Guo, Introductory of Microsoft
- [52] M.A. Hossain, H.H. Ngo, W. Guo, Introductory of Microsoft Excel SOLVER function-spreadsheet method for isotherm and kinetics modelling of metals biosorption in water and wastewater, J. Water Sustainability, 3 (2013) 223–237.
- [53] M. Naushad, Z.A. ALOthman, M. Rabiul Awual, S.M. Alfadul, T. Ahamad, Adsorption of rose Bengal dye from aqueous solution by amberlite Ira-938 resin: kinetics, isotherms, and thermodynamic studies, Desal. Water Treat., 57 (2016) 13527–13533.