



Diazinon Removal from Aqueous Environments by Electrocoagulation Process

Edris Bazrafshan, Davoud Balarak*

Department of Environmental Health, Health Promotion Research Center, Zahedan University of Medical Sciences, Zahedan, Iran.

*Corresponding Author: dbalarak2@gmail.com

Abstract Electrocoagulation (EC) process is an electrochemical method of treating polluted water whereby sacrificial anodes corrode to release active coagulant precursors into solution. At present study, EC was investigated for removal of diazinon pesticide from aqueous environments under different operating conditions such as initial pH (3-10), applied voltage (20-40 V), electrolysis time (10-60 min), and initial concentration of diazinon (10-100 mg/L) by using aluminum anode in batch mode. The results of this study showed that diazinon concentration can effect on removal efficiency and for higher concentration of diazinon, more applied voltage or higher reaction time is needed. Maximum removal efficiency of diazinon (>99%) was obtained after 60 min at pH 3. According to results of this study it can be concluded that Electrocoagulation process using aluminum electrode is an efficient method for diazinon removal from aqueous environments.

Keywords Pesticide, Diazinon, Electrocoagulation, Aluminum electrodes

Introduction

Chemical pesticides are well recognized as an economic approach to controlling pests, at the same time such chemicals are highly toxic to other species in the environment. Now there is growing concern worldwide over the indiscriminate use of such chemicals that results in environmental pollution and toxicity risk to nontarget organisms [1].

The contamination of surface and ground water resources by pesticides has become a serious environmental problem in recent years due to the wide application of these agrochemicals in crop farms, orchards, fields and forest lands. This contamination arises from surface runoff, leaching, wind erosion, deposition from aerial applications, industrial discharges and various other sources. As a result, pesticides have frequently been detected in water resources in different countries of the world [2-3].

Organophosphorus pesticides (OPPs) such as diazinon are neurotoxic chemical agents that inhibit acetyl cholin esterase activity. OPPs and carbamates are the most widely used acetylcholinesterase activity inhibitors as insecticides [4]. These agents inhibit acetylcholinesterase activity, not only in insects but can also affect the nervous system of humans. In addition, these compounds cause accumulation of acetylcholine at cholinergic synapses leading to increased activation of nicotinic and muscarinic receptors [5].

Diazinon ($C_{12}H_{21}N_2O_3PS$) is one of the most common used organophosphate pesticides in the world [6] and is an insecticide which acts as a contact stomach and respiratory poison, and has been identified to be potential chemical mutagens [7]. World Health Organization (WHO) classified Diazinon as "moderately hazardous" class II [8]. This pesticide released to environment is subject to various processes including volatilization, photolysis, hydrolysis, and biodegradation [9].

In general, pesticides can be removed through various methods, such as photocatalytic degradation [10], combined photo-Fenton and biological oxidation [11-14] and nanofiltration [15]. In addition, some techniques have been used for degradation of diazinon in aqueous solution, such as chemical oxidation using ozonation, aqueous chlorine and Fenton treatment, irradiation degradation through X-ray and gamma-ray, biodegradation and ultrasonic technique [16].

In recent years, electrochemical techniques such as electrocoagulation, electroflotation and electrooxidation have drawn the attention of many researchers since the main reagent is the electron which is a "clean reagent".



Furthermore secondary contamination could be avoided by electrocoagulation. A probable technique for removal of diazinon pesticide from aqueous solutions is electrocoagulation. In this procedure, the treatment process is done without adding any chemical coagulant or flocculants, thus reducing the amount of sludge which must be disposed [17].

The principal objectives of this study are to provide direct evidence of diazinon adsorption onto electrochemically generated aluminum hydroxide flocs under different experimental conditions and obtain clues to associated uptake mechanisms.

Materials and methods

Chemicals

At present study all chemicals (Diazinon, acetone, hexane, sodium hydroxide pellets, concentrated sulfuric acid and sodium chloride) were of standard analytical grade and used without further purification unless otherwise noted. Predetermined concentrations of diazinon were prepared by mixing proper amount of diazinon (60%) with deionized water. The conductivity of solutions was raised up and adjusted by adding NaCl using a conductivity meter (Jenway, Model 4200). The pH of the solutions was measured using a pH meter (E520, Metrohm Herisau, Switzerland) and adjusted by adding 0.1 N NaOH or H₂SO₄ solution.

Experimental apparatus and procedure

The batch electrocoagulation cell (bipolar mode) used in the experimental study was constructed of Plexiglas with the dimensions of 10 cm × 13 cm × 12 cm. The total volume of sample in each experiment was approximately 1000 mL. In each batch, six aluminum plates were constructed in the electrochemical reactor and the distance between plates was fixed at approximately 15 mm. The current was maintained constant by means of a precision DC power supply characterized by the ranges 0–5 A for current and 0–40 V for voltage. The solution was agitated with a magnetic stirrer during electrocoagulation at 150 rpm. Electrodes were sanded and washed with dilute HCl between the experiments.

To follow the progress of the treatment, samples were periodically (10-60 min) taken from the electrocoagulation cell then filtered (0.45 μm) to eliminate flocs formed during electrolysis.

Electrical energy consumption is a very important economical parameter in the electrocoagulation process. The electrical energy consumption was calculated using the following equation:

$$E = \left[\frac{UIt}{1000V} \right]$$

where E is the energy consumption (kWh/m³), U is the applied voltage (V), I is the current intensity (A), t is the electrocoagulation time (h), and V is the volume of the treated wastewater (m³). The amount of electrode consumption was calculated by weighting of plates before and after of electrocoagulation process in each step. For a particular electrical current flow in an electrolytic cell, the mass of aluminum theoretically dissolved from the sacrificial anode is quantified by Faraday's law:

$$w = \left[\frac{ItM}{ZF} \right]$$

where "w" is the amount of anode material dissolved (g), I the current (A), the electrolysis time (t), M the specific molecular weight of electrode (g/mol), Z the number of electrons involved in the reaction and F is the Faraday's constant (96485.34 C/mol).

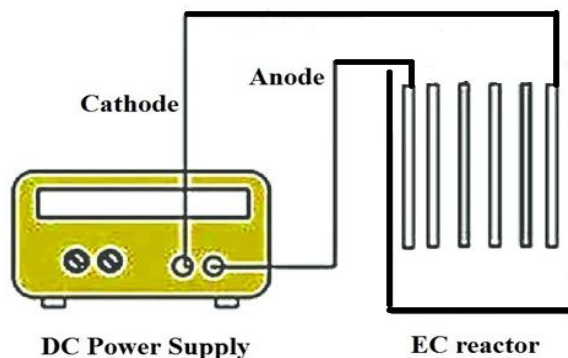


Figure 1: Schematic diagram of electrochemical reactor



Results and Discussion

At present study, application of electrocoagulation process as a treatment method using aluminum electrodes was used for diazinon removal from aqueous solutions. It has been established that pH is an important parameter influencing the performance of the electrochemical process [18-19]. Hence, at current study diazinon removal efficiency was determined in the pH range from 3 to 10. It can be seen from Fig 2-6 that the removal efficiency of diazinon decreased with the increase of pH in the range 3-10 and maximum removal efficiency (more than 90%) was observed at acidic condition (pH=3). The lowest diazinon removal efficiency was observed at an initial pH of 7.0.

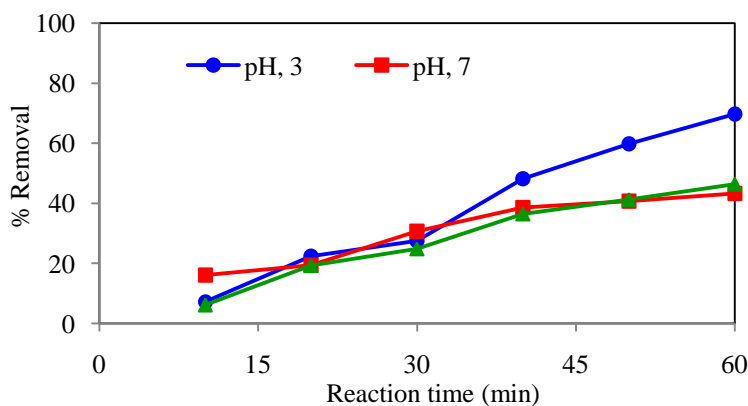


Figure 2: Diazinon removal efficiency (Initial concentration = 10 mg/L, applied voltage = 20 V)

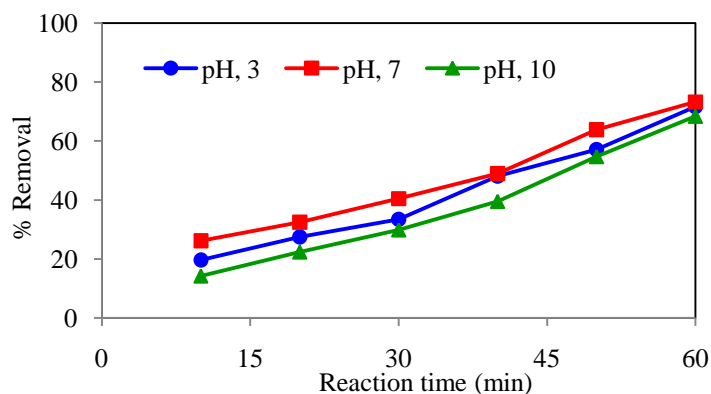


Figure 3: Diazinon removal efficiency (Initial concentration = 10 mg/L, applied voltage = 30 V)

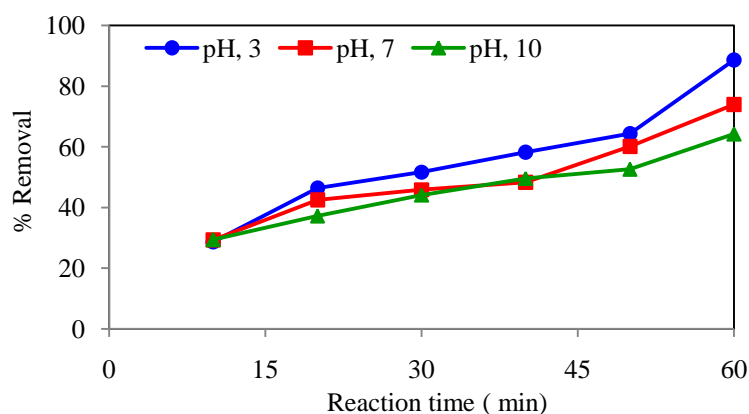


Figure 4: Diazinon removal efficiency (Initial concentration = 10 mg/L, applied voltage = 40 V)

As observed by other investigators [20-21], a pH increase occurs when the initial pH is low (less than 7). Shafaei et al. [22] ascribed this increase to hydrogen evolution at cathodes. However, this was contested by Chen et al. [23] who explained this increase by the release of CO_2 from wastewater owing to H_2 bubble



disturbance. This result is in agreement with previously published works and suggests that electrocoagulation can act as pH buffer [24].

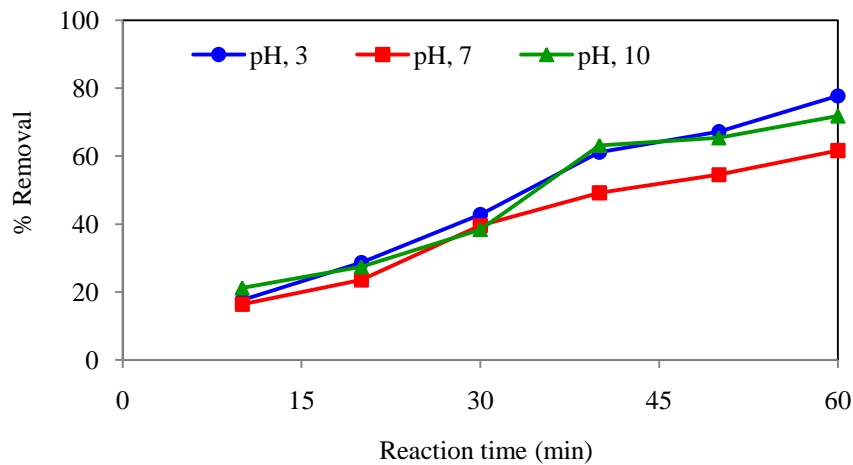


Figure 5: Diazinon removal efficiency (Initial concentration = 50 mg/L, applied voltage = 20 V)

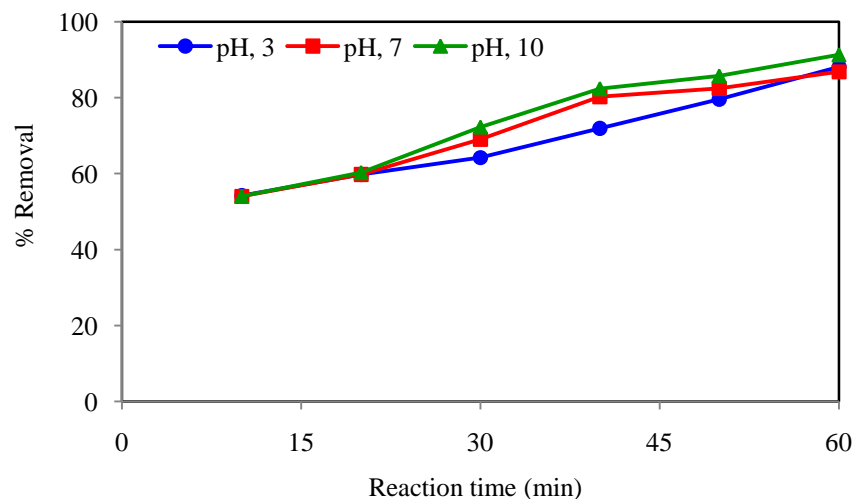


Figure 6: Diazinon removal efficiency (Initial concentration = 100 mg/L, applied voltage = 20 V)

It can be seen from Figures 2-6 that the removal efficiency of diazinon increased with the increase of applied voltage in the range 20-40 V. Further increase of the applied voltage results only in a small increase of the diazinon removal efficiency. Moreover increasing increases energy and electrode consumption. It is well known that applied voltage determines the coagulant dosage rate, bubble production rate, and flocs growth, which can influence the treatment efficiency of the electrocoagulation. Additionally, it was demonstrated that bubbles density increases and their size decreases with increasing voltage, resulting in a greater upwards flux and a quicker removal of pollutants and also sludge flotation [25-26].

As previously mentioned, the efficiency of electrocoagulation process depends directly on the concentration of aluminum ions, which is produced by the electrode dissolution. In addition, the production rate of Al^{3+} ions is related to the time and current flow in the electrocoagulation reactor, according to Faraday's law. Consequently, at present study a set of experiments were performed with different initial concentrations of diazinon (10, 50 and 100 mg/L) to determine the time required for diazinon removal under various conditions. It can be seen from figures 4-6 that when the initial concentration of diazinon increased from 10 to 100 mg/L, removal efficiency decreased that is because of the decrease in aluminum ions in high concentration of pollutant. However, in higher concentration, 100 mg/L the adsorption capacity did not reduce and the pesticide removal rate was relatively constant. Furthermore, it is clear from Figures 4-6, that for higher concentration of diazinon (100 mg/L), the greater time needed, but that higher initial concentrations of diazinon were reduced significantly in relatively less time than lower concentrations. Similar findings were reported by other researchers (27-29).



Table 1: The amount of electrode consumption during electrocoagulation process (g/L)

Voltage (V)	Diazinon concentration, (mg/L)		
	10	50	100
20	3.0	2.2	2.23
30	4.47	4.3	3.43
40	7.5	6.67	5.77

Table 2: Energy consumption during electrocoagulation process (kwh/g) at applied voltage = 40 V

Initial pH	Diazinon concentration, (mg/L)		
	10	50	100
3	14.32	2.78	1.16
7	14.28	2.81	1.41
10	14.24	2.90	1.39

Electrical energy and electrode consumption are very important economical parameters in the electrocoagulation process. Hence, for the same operating conditions, after 60 min of electrocoagulation, consumption of energy and aluminum electrode at applied voltage 40V was measured which these results presented in Tables 1 and 2. As it can be seen from Table 1, electrode consumption increase with increasing of applied voltage and maximum electrode consumption was observed at 40 V. On the other hand, it can be concluded that the higher the applied voltage, the higher the weight of the electrode consumed. Also, higher initial diazinon concentrations in the solution result in lower electrode consumption weights. The amount of consumed energy at applied voltage 40 V and for different diazinon concentrations calculated and presented in Table 2 and with increasing the applied voltage from 20 V to 40 V, energy consumption decreased considerably, from 14.32 to 1.16 kwh/g. As presented in Table 2, electrical energy consumption was found to decrease with increasing the initial concentration of diazinon, because the flocs formation will help the adsorption of the diazinon from the solution. Similar findings were reported by other researchers [18-20].

Conclusion

At present study experiments were carried to remove diazinon pesticide by electrocoagulation process covering wide range in operating conditions. Diazinon removal by electrocoagulation process was affected by applied voltage, initial pH and initial concentration of diazinon in batch process. The results showed that for an initial diazinon concentration of up to 50 mg/L with an initial pH adjusted in the range 3-10, the use of an applied voltage of 40 V gave access to a diazinon removal efficiency between 90 and >99.9%. From the present study, it transpires that electrocoagulation process can very successfully purify diazinon-contaminated samples.

Acknowledgments

The authors would like to thank the Center for Environmental Research, Tehran University of Medical Sciences for financially supporting this study under contract number 2711.

References

- [1]. Rao JV. Sublethal effects of an organo phosphorus insecticide (RPR-II) on biochemical parameters of tilapia, *Oreochromis mossambicus*. *Comparative Biochemistry and Physiology, Part C* 2006; 143: 492-98.
- [2]. Akcay G, Akcay M, Yurdakoc K. Removal of 2,4-dichloro phenoxyacetic acid from aqueous solutions by partially characterized organo philicsepiolite: thermodynamic and kinetic calculations. *Journal of Colloid And Interface Science*. 2005; 281; 27-32.
- [3]. Rebich RA, Coupe RH, Thurman EM. Herbicide concentrations in the Mississippi River Basin-the importance of chloro acetanilide herbicide degradates. *Science of the Total Environment*. 2004; 321;189-199.
- [4]. 10. Delfino RT, Ribeiro TS, Figueroa-Villar JD. Organophosphorus compounds as chemical warfare agents: a review. *Journal Of The Brazilian Chemical Society*. 2009; 20: 407-28.
- [5]. Timcheh Hariri A, Moallem SA, Mahmoudi M, Memar B, Hosseinzadeh H. Sub-acute effects of diazinon on biochemical indices and specific biomarkers in rats: Protective effects of crocin and safranal. *Food and Chemical Toxicology*. 2010; 48:2803-8.
- [6]. Gokcimen A, Gulle K, Demirin H, Bayram D, Kocak A, Altuntas I. Effects of diazinon at different doses on rat liver and pancreas tissues. *Pesticide Biochemistry and Physiology* 2007;87:103-8.
- [7]. Bolognesi C, Morasso G. Genotoxicity of pesticides: potential risk for consumers. *Trends in Food Science and Technology*. 2000;11:182-87.



- [8]. Shemer H, Linden KG. Degradation and by-product formation of diazinon in water during UV and UV/H₂O₂ treatment. *Journal of Hazardous Materials*. 2006; B136: 553-9.
- [9]. Feigenbruge LV, Calve SL, Mirabel P. Temperature dependence of Henry's law constants of metolachlor and diazinon. *Chemosphere* 2004;57:319-327.
- [10]. Aungpradit T, Sutthivaiyakit P, Martens D, Sutthivaiyakit S, Kettrup AAF. Photocatalytic degradation of triazophos in aqueous titanium dioxide suspension: identification of intermediates and degradation pathways. *Journal of Hazardous Materials*. 2007;146: 204-13.
- [11]. Ballesteros Martín MM, Sánchez Pérez JA, García Sánchez JL, Montes de Oca L, Casas López JL, Oller I, Malato Rodríguez S. Degradation of alachlor and pyrimethanil by combined photo-Fenton and biological oxidation. *Journal of Hazardous Materials*. 2008; 155: 342-9.
- [12]. Dehghani M, Naseri S, Zamanian Z. Biodegradation of alachlor in liquid and soil cultures under variable carbon and nitrogen sources by bacterial consortium isolated from corn field soil. *Iranian Journal of Environmental Health Science and Engineering* 2013;10(1);10-21
- [13]. Dehghani M, Naseri S, Hashemi H. Study the bioremediation of atrazine under variable carbon and nitrogen sources by mixed bacterial consortium isolated from corn field soil in Fars province of Iran. *Journal of Environmental and Public Health*. 2013; 1; 1-7.
- [14]. Rajashekara Murthy HM, Manonmani HK. Aerobic degradation of technical hexachloro cyclohexane by a defined microbial consortium. *Journal of Hazardous Materials*. 2007;149:18-25.
- [15]. Ahmad AL, Tan LS, Shukor SRA. Dimethoate and atrazine retention from aqueous solution by nanofiltration membranes. *Journal of Hazardous Materials*. 2008;151:71-77.
- [16]. Zhang Y, Zhang W, Liao X, Zhang J, Hou Y, Xiao Z, Chen F, Hu X. Degradation of diazinon in apple juice by ultrasonic treatment. *Ultrasonics Sonochemistry*; 2010; 17: 662-8.
- [17]. Gurses A, Yalcin M, Dogan C. Electrocoagulation of some reactive dyes: a statistical investigation of some electrochemical variables. *Waste Management*; 2002; 22: 491-9.
- [18]. Bazrafshan E, Kord Mostafapour F, Soori MM, Mahvi AH. Application of combined chemical coagulation and electrocoagulation process to carwash wastewater treatment. *Fresenius Environmental Bulletin*. 2012; 21(9): 2694-1.
- [19]. Bazrafshan E, Moein H, Kord Mostafapour F, Nakhaie S. Application of electrocoagulation process for dairy wastewater treatment. *Journal of Chemistry*. 2013; 8;1-8.
- [20]. Bazrafshan E, Kord Mostafapour F, Farzadkia M, Ownagh KA, Mahvi AH. Slaughterhouse wastewater treatment by combined chemical coagulation and electrocoagulation process. *Plos One*. 2012;7 (6):1-8.
- [21]. Bazrafshan E, Mahvi AH, Naseri S, Mesdaghinia A R. Performance evaluation of electrocoagulation process for removal of chromium (VI) from synthetic chromium solutions using iron and aluminum electrodes. *Turkish Journal of Engineering and Environmental Science*. 2008;32 (2):59-66.
- [22]. Shafaei A, Rezayee M, Arami M, Nikazar M. Removal of Mn²⁺ ions from synthetic wastewater by electrocoagulation process. *Desalination*. 2010;260:23-8.
- [23]. Chen X, Chen G, PoLY. Separation of pollutants from restaurant wastewater by electrocoagulation. *Separation and Purification Technology*. 2000; 19: 65-76.
- [24]. Kobya M, Taner-Can O, Bayramoglu M. Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes. *Journal of Hazardous Materials*. 2003; B100:163-178.
- [25]. Daneshvar N, Oladegaragoze A, Djafarzadeh N. Decolorization of basic dye solutions by electrocoagulation: an investigation of the effect of operational parameters. *Journal of Hazardous Materials*. 2006;129(1-3): 116-22.
- [26]. Emamjomeh M, Sivakumar M. Fluoride removal by a continuous flow electro-coagulation reactor. *Journal of Environmental Management*. 2009;90:1204-1212.
- [27]. Bazrafshan E, Biglari H, Mahvi AH. Phenol removal by electrocoagulation process from aqueous solutions. *Fresenius Environmental Bulletin*. 2012;21(2): 364-71.
- [28]. Bazrafshan E, Ownagh KA, Mahvi AH. Application of electrocoagulation process using Iron and Aluminum electrodes for fluoride removal from aqueous environment. *E-Journal of Chemistry*. 2012; 9(4);2297-8.
- [29]. Bazrafshan E, Biglari H, Mahvi AH. Humic acid removal from aqueous environments by electrocoagulation process using iron electrodes. *E-Journal Chemistry*. 2012; 9(4):2453-61.

