



Influences of Solution Chemistry on Phenol Removal From Aqueous Environments by Electrocoagulation Process Using Aluminum Electrodes

Mohammad Ali Zazouli ¹, Mahmoud Taghavi ^{1*}, Edris Bazrafshan ²

¹ Department of Environmental Health Engineering, Health Sciences Research Center and Faculty of Health, Mazandaran University of Medical Sciences, Sari, IR Iran

² Health Promotion Research Center, Zahedan University of Medical Sciences, Zahedan, IR Iran

ARTICLE INFO

Article type:
Original Article

Article history:
Received: 24 Jun 2012
Revised: 07 Jul 2012
Accepted: 07 Jul 2012

Keywords:
Fungi
Arsenic
Tolerance
Accumulation
Bioremediation

ABSTRACT

Background: Facultative marine fungi could potentially be arsenic tolerant and may be able to remove this highly poisonous metal from the environment.

Objectives: The objective of this work was to explore the degree of tolerance and removal efficiency of two facultative marine fungi.

Materials and Methods: Facultative marine fungi *Aspergillus flavus* and *Rhizopus* spp. were exposed to 25 mg/L and 50 mg/L sodium arsenite (As (III)). Tolerance of these species to the test concentrations was assessed by studying their biomass accumulation. Accumulation of arsenic by the fungal biomass was also evaluated.

Results: Our study revealed that both *A. flavus* and *Rhizopus* sp. exhibited tolerance towards the test concentrations of arsenic. Both of the test fungi also exhibited arsenic accumulation. *Rhizopus* sp. was found to be a slightly better potential accumulator.

Conclusions: This study reveals that the test fungi can be harnessed as bioremediation agents for arsenic contaminated sites.

► Implication for health policy/practice/research/medical education:

The Result of this article will be helpful for researchers who studying about water and waste water treatment by Electrocoagulation processes or effect of NOM on water and waste water treatment processes.

► Please cite this paper as:

Zazouli MA, Taghavi M, Bazrafshan E. Influences of Solution Chemistry on Phenol Removal From Aqueous Environments by Electrocoagulation Process Using Aluminum Electrodes. *J Health Scope*. 2012; 1 (2): 66-70.

1. Background

Industrial activities can contaminate aquatic environments via industrial wastes and wastewater discharges (1). Industrial wastewater includes some contaminants, therefore treatment of industrial wastewaters seems to be essential (2). Phenol and phenolic compounds are the most prevalent forms of organic chemical pollutants in industrial wastewaters (3). High concentrations of phenol and phenolic compounds exists in effluents

of oil refineries, petrochemical plants, ceramic plants, coking plants, resin and dye manufactures, steel plants, coal conversion processes, phenolic resin industries and pharmaceutical industries (3-6). Phenol is a potential carcinogen of human, which raises considerable health concerns, even at low concentrations. Its toxic effects include permeabilisation of cellular membranes and cytoplasmic coagulation (3, 6). Chlorophenol, which is formed by phenols in the presence of chlorine in drink-

* Corresponding author: Mahmoud Taghavi, Department of Environmental Health Engineering, Health Sciences Research Center and Faculty of Health, Mazandaran University of Medical Sciences, Sari, IR Iran. Tel/ Fax: +98-1513543080-8, E-mail: taghavam66@gmail.com

ing water, has a medicinal taste, which is quite pronounced and objectionable (7). Phenol is classified by the USEPA as a priority pollutant and represents as one of the most challenging classes of pollutants requiring urgent removal from waste streams (1, 7). Many technologies such as adsorption, enzymatic treatments, electrochemical methods, advanced oxidation and combined techniques have been applied in order to removing and degradation of phenolic compounds from wastewaters (2, 3, 8-10). However these methods have some problems such as high costs, low efficiency, and generation of toxic by-products (2). Whereas, Electrochemical processes has little or no harmful effects on the environment. Additionally, electrochemical reactions are independent from conditions of wastewaters (2). Electrochemical methods are frequently used for treating wastewaters containing organic and inorganic compounds (8). According to previous studies, electrocoagulation has been known as an effective method to decontaminate various wastewaters such as landfill leachate, restaurant wastewaters, saline wastewaters, tar sand and oil shale wastewaters, urban wastewaters, laundry waste waters, nitrate and arsenic bearing wastewaters and chemical mechanical polishing waste waters (9). Electrocoagulation is based on the in situ formation of the coagulants as the sacrificial anode corrodes due to an applied current, while the simultaneous evolution of hydrogen at the cathode allows for pollutant removal by flotation. In this process, the flocculating agent is generated by electro oxidation of a sacrificial anode, generally made of iron or aluminum (5, 9). Many studies have been performed on phenol removal by the electrocoagulation process using aluminum & Iron electrodes (2, 8, 10-12). Despite the fact, those researches have investigated the effects of some parameters such as pH, voltage, concentration, but there are not enough data about the effects of natural organic matters such as humic and aliginic acid on the removal efficiency. Natural organic matter (NOM) is one of the components that are usually present in many aqueous feed streams. NOM can be generally divided into two fractions, i. e. , hydrophobic (humic) and hydrophilic (non-humic) divisions. NOM typically comprises a wide range of compounds, often measured as dissolved organic carbon (DOC), ranging from low to high molecular weight compounds. Among these, polysaccharides and humic substances are some of the most hydrophilic and hydrophobic macromolecules, respectively. In addition to these, monomeric species such as simple sugars and amino acids, proteins, and other classes of biopolymers are also contributed to NOM and are present in water sources. Alginate is an acidic polysaccharide produced by bacteria, microalgae, and macroalgae, and is typically made up of repetitive manuronic and guluronic acids. This may influence the removal efficiency of phenol by electrocoagulation.

2. Objectives

The main objective of this work is to focus on the removal of phenol by the electrocoagulation process using aluminum electrodes. In addition, the effects of solution chemistry such as pH, reaction time, current density, initial concentration of phenol and presence of polymeric natural organic matter on removal performance are described and discussed. Furthermore, it is expected that the results of this research will complement and reconcile the findings of previous publications.

3. Materials and Methods

3.1. Materials

All chemicals, including sodium hydroxide pellets (NaOH), concentrated sulfuric acid (H_2SO_4), sodium chloride (NaCl), Ammonium hydroxide (NH_4OH), Dipotassium phosphate (K_2HPO_4), Monopotassium phosphate (KH_2PO_4), Potassium ferricyanide ($K_3Fe(CN)_6$) and Phenol were purchased from Merck (Germany), 4-Aminoantipyrine from Sigma-Aldrich (America) and Alginate acid sodium salt from Fluka Biochemical (America). Analytical reagent chemicals and distilled water were used to prepare all stock solutions. Desired concentrations of Phenol solution were prepared by mixing proper amounts of Phenol with distilled water.

3.2. Electrocoagulation Experiments

In this study, synthetic wastewater was prepared by dissolving 1000 mg L⁻¹ of phenol into distilled water and adjusting pH with H_2SO_4 to 2 ± 0.1 for stabilization of the solution. There are six monopolar electrodes, three anodes and three cathodes of the same dimensions. Both aluminum cathodes and anodes were made from plates with dimensions of $10 \times 12 \times 0.2$ cm. the distance between electrodes was 2 cm and the electrodes were connected to a digital DC power supply (MPS-3005D, 30V, 5A). The Plexiglas cell as an electrolytic reactor having dimensions of $12 \text{ cm} \times 12 \text{ cm} \times 16 \text{ cm}$ with a working volume of about 1.5 L was used to conduct the experiments, and a constant stirring speed of 200 rpm was applied during all experiments. Before each run, the aluminum electrodes were washed with tap water, dipped in HCl solution (5% v/v) for at least 15 min and then rinsed again with Distilled water. In each run, 1.5 L of synthetic wastewater was placed into the electrolytic cell and the pH was adjusted to the desire value. In order to increase the conductivity of the solution, sodium chloride was added to the solution. The current density was adjusted to a desired value and then the experimental run was started. At different time intervals, the samples were taken from the reactor and centrifuged for 30 min at 4000 rpm before analysis. Analyses were performed for examination of water and wastewater according to the standard methods (13). Briefly, concentrations of phenol were determined by spectrophotometer (Hach DR2800 spectrophotometer)

according to the analysis of the color resulted from the reaction of phenol with 4-aminoantipyrine at 500 nm (13).

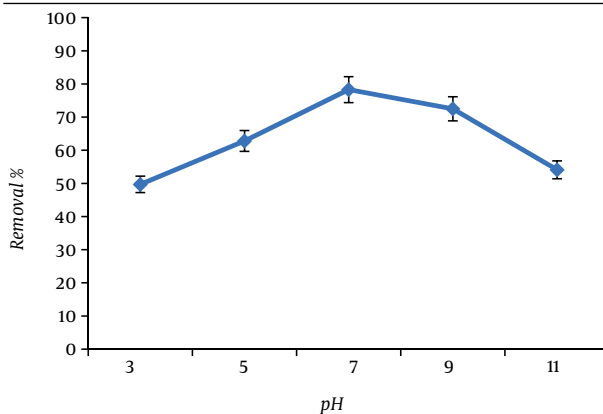
4. Results

In the present study the effects of some parameters such as phenol concentration, current density, concentration of electrolyte solution, initial pH and Alginic acid concentration have been investigated on the final phenol removal efficiency. Percentage of phenol removal has been calculated using the formula $[(1 - (C/Co)) \times 100]$, where, Co and C are concentrations of phenol before and after treatment, respectively.

4.1 Effect of pH

An important operating factor that influences the performance of EC process is pH (14, 15). EC process would change the pH of the solution during the process, which depends on the type of electrodes material and also the initial pH. Meanwhile, this process exhibits some buffering capacity, especially in alkaline solutions, which prevents high changes in pH (6). The effect of initial pH on phenol removal efficiencies is presented in Figure 1. As shown, very low ranges of pH are not desirable. The highest removal efficiency has been achieved near neutral pH. At this point the phenol removal was as high as 78%, while it has dropped dramatically above pH 7. These results are in accordance with other researchers who have reported the maximum performance for the ECP at pHs between 7 to 8 when using Al as the sacrificial electrode (2, 14, 15).

Figure 1. The Effect of pH on Phenol Removal by Electrocoagulation



c. d = 10 mA/cm², NaCl = 2 g/L, phenol = 100 mg/l, T = 20 °C, time = 60 min

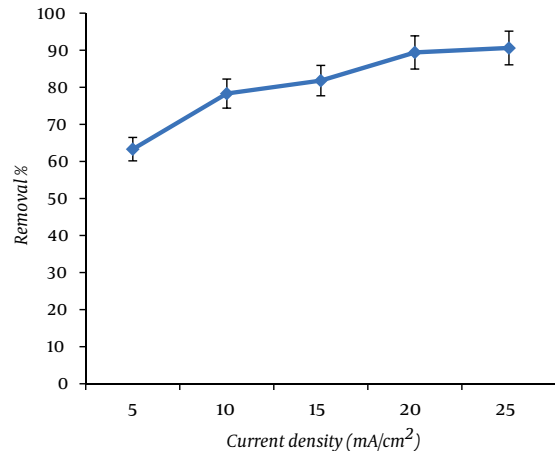
ficinal electrode (2, 14, 15). This behavior was attributed to the amphoteric character of aluminum hydroxide which does not precipitate at very low pHs. On the other hand, high pHs lead to formation of $Al(OH)_4^-$, which is soluble and impractical for adsorption of Phenol (2, 11).

4.2 Effect of Current Density

Figure 2 depicts the effect of current density on phenol

removal efficiency with a constant time of 60 min. In this study, selected range of applied current density was from 5 to 25 mA/cm² and selected optimum value of current density was 25 mA/cm² for which 91% removal efficiency has been achieved. This result is due to the fact that at high current

Figure 2. The Effect of Current Density on Phenol Removal by Electrocoagulation



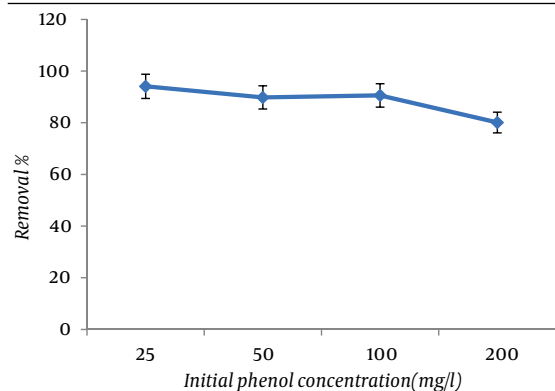
pH = 7, NaCl = 2 g/L, phenol = 100 mg/l, T = 20 °C, time = 60 min

densities, the extent of anodic dissolution of aluminum increases, resulting in a greater amount of precipitate for the removal of pollutants. Moreover, bubble generation rate increases and the bubble size decreases with increasing current density. These effects are both beneficial for high pollutant removal by H₂ flotation (16-19).

4.3 Effect of Initial Phenol Concentration

Effect of phenol concentration from 25 to 200 mg/L on the removal efficiency has been shown in Figure 3. It can be observed that an increase in concentration for same electrocoagulation time and current density results in a decrease in removal efficiency. The results of this study

Figure 3. The Effect of Initial Phenol Concentration on Phenol Removal by Electrocoagulation



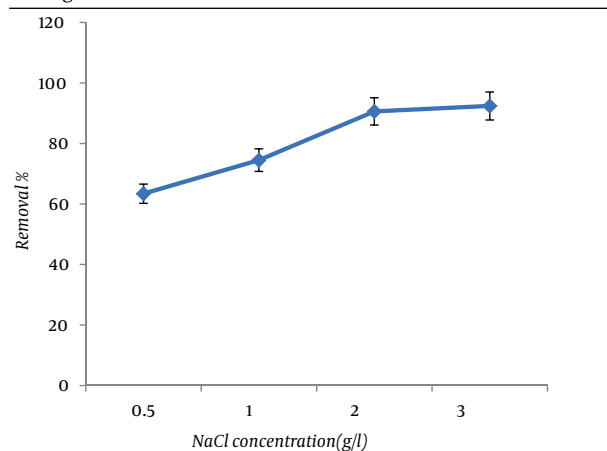
pH = 7, c. d = 25 mA/cm², NaCl = 2 g/L, T = 20 °C, time = 60 min

are consistent with the result of the research conducted by Yusuf Yavuz and Savas Koparal on electrochemical oxidation of phenol in a parallel plate reactor using ruthenium mixed metal oxide electrode (12). This can be attributed to the fact that at a constant current density, a same number of aluminum ions passes to the solution at different phenol concentrations. Consequently, the formed amount of complex aluminum hydroxide was insufficient to coagulate the greater number of phenol molecules at higher phenol concentrations (2).

4.4 Effect of Ionic Strength

It is well known that increasing the electrical conductivity causes an increase in the current density at constant cell voltages, or a decrease in the cell voltage at constant current densities (14). To study the effect of ionic strength on efficiency of phenol removal, the salt content of the feed solution was adjusted with NaCl (0.5, 1, 2 and 3 mg/l). Figure 4 shows the effect of ion strength on the performance of the EC process. As seen, with increasing

Figure 4. The Effect of NaCl Concentrations on Phenol Removal by Electrocoagulation



pH = 7, c. d = 25mA/cm², phenol = 100mg/l, T = 20 °C, time = 60 min

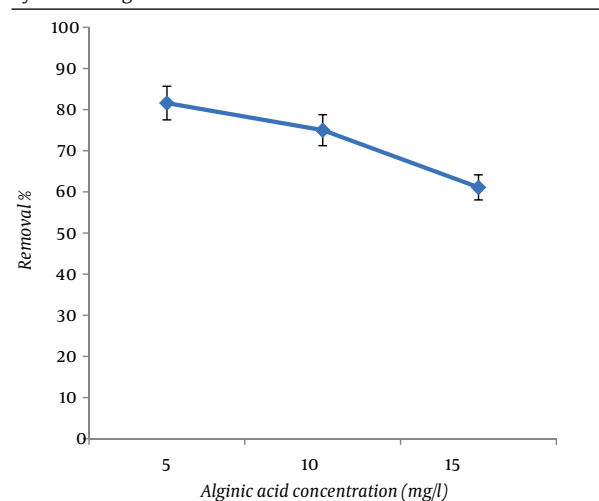
ion strength, the phenol removal efficiency is considerably enhanced. Conductivity of the solution depends on the number of ions present. Increase in the concentration of electrolytes results in an increase in the number of ions. Thus, ultimately solution becomes more conductive, which results in passage of more electrons per a unit time. Hence, we can have more removal efficiency (15).

4.5 Effect of Alginic Acid Concentration

In this study, alginate or Alginic acid was selected as a model of natural organic matter (NOM). Effect of NOM was investigated by introducing alginate at three different concentrations (5, 10 and 15 mg/L) into the feed solutions. To examine the effect of Alginic acid on the phenol removal efficiency, while other parameters were kept

constant, a nominal value of Alginic acid concentration ranging from 5 to 15 mg/l was added to synthetic aqueous solutions. Figure 5 illustrates the influence of Alginic acid concentrations on phenol removal. It was observed that increasing Alginic acid concentrations decreased the removal efficiency of phenol. It seems that one reason can be negative charge of the Alginic acid. pHzpc is defined as the pH at which the total surface charge becomes zero. It means that at a pH range upper than pHzpc, the surface of biomass shows a negative charge. Jeon *et al.* have determined the pHzpc of Alginic acid as about 2.83 (20). While, in this step of experiment, the pH of solution was 7. So, the surface charge of the Alginic acid will be negative, and therefore it would attract small parts of the metal ions produced. Furthermore, Alginic acid has been shown to be effective at removing many cationic met-

Figure 5. The Effect of Alginic Acid Concentrations on Phenol Removal by Electrocoagulation



pH = 7, c. d = 25mA/cm², NaCl = 2g/L, phenol = 100 mg/l, T = 20 °C, time = 60min

als from some solutions (21). As a result, increasing the amount of Alginic acid will decrease efficiency of phenol removal by electrocoagulation. Electrocoagulation processes remove the organic matters as the researchers have reported that these processes have the potential to be utilized for a cost-effective removal of HA from aqueous environments (22). Thus when PNOM is precipitating, it may remove the phenol.

5. Discussions

In the present work, phenol removal from synthetic aqueous solutions containing different concentrations of phenol up to 200 mg/L by electrocoagulation, with aluminum electrodes, has been investigated. According to the results, an increase in current density and NaCl concentration can increase the efficiency of phenol removal. Conversely, increasing initial phenol and Alginic acid concentrations result in decreasing phenol removal

by electrocoagulation. It occurs because of the negative surface charge of the Alginate acid in neutral and alkaline pH which attracts small parts of the metal ions produced and will consequently compete with phenol. The optimum value of current density, allowing high removal rates (91%) of phenol, was found to be 25 mA/cm². The highest removal efficiency was achieved near neutral pH (78%). High conductivity is in favor of high process performances. Based on the results of this study, the phenol removal by electrocoagulation using aluminum electrodes seems to be a very efficient method for decontamination of wastewaters containing high concentrations of phenol.

Acknowledgements

The authors would like to express their thanks to the laboratory staff of the Department of Environmental Health Engineering, Faculty of Health, for their collaboration and also to Health Sciences Research Center and the Research Deputy of Mazandaran University of Medical Sciences for the financial support of this study.

Authors' Contribution

The overall implementation of this study including design, experiments and data analysis, and manuscript preparation were the results of joint efforts by individuals who are listed as co-authors of this paper. All authors have made extensive contribution into the review and finalization of this manuscript.

Financial Disclosure

None declared.

Funding/Support

The project has been supported financially by Health Sciences Research Center and the Research Deputy of Mazandaran University of Medical Sciences (Project No: 89-140).

References

1. Yun-Hwei S. Removal of phenol from water by adsorption-flocculation using organobentonite. *Water Research*. 2002;**36** (5):1107-14.
2. Abdelwahab O, Amin NK, El-Ashtouky ES. Electrochemical removal of phenol from oil refinery wastewater. *J Hazard Mater*. 2009;**163** (2-3):711-6.
3. Bodalo A, Gomez JL, Gomez M, Leon G, Hidalgo AM, Ruiz MA. Phenol removal from water by hybrid processes: study of the membrane process step. *Desalination* 2008;**223** (1-3):323-9.
4. Stoilova I, Krastanov A, Stanchev V, Daniel D, Gerginova MAZ. Biodegradation of high amounts of phenol, catechol, 2,4-dichlorophenol and 2,6-dimethoxyphenol by *Aspergillus awamori* cells. *Enzyme Microb Tech*. 2006;**39** (5):1036-41.
5. Bazrafshan E, Mahvi AH, Nasseri S, Shaieghi M. Performance evaluation of electrocoagulation process for diazinon removal from aqueous environments by using iron electrodes. *Iran J Environ Health Sci Eng*. 2007;**4** (2):127-32.
6. Maleki A, Mahvi AH, Mesdaghinia A, Naddafi K. Degradation and toxicity reduction of Phenol by Ultrasound waves. *Bull Chem Soc Ethiop*. 2007;**21** (1):33-7.
7. Maleki A, Erfan MB, Mohammadi AS, Ebrahimi R. Application of commercial powdered activated carbon for adsorption of carbolic acid in aqueous solution. *Pak J Biol Sci*. 2007;**10** (14):2348-52.
8. Ugurlu M, Gurses A, Dogar C, Yalcin M. The removal of lignin and phenol from paper mill effluents by electrocoagulation. *J Environ Manage*. 2008;**87** (3):420-8.
9. Bazrafshan E, Mahvi AH, Zazouli MA. Removal of Zinc and Copper from Aqueous Solutions by Electrocoagulation Technology Using Iron Electrodes. *Asian J Chem*. 2011;**23** (12):5506-10.
10. Moreno-Casillas HA, Cocke DL, Gomes JAG, Morkovsky P, Parga JR, Peterson E. Electrocoagulation mechanism for COD removal. *Separ Purif Method* 2007;**56** (2):204-11.
11. Adhoum N, Monser L. Decolourization and removal of phenolic compounds from olive mill wastewater by electrocoagulation. *Chem Eng Process*. 2004;**43** (10):1281-7.
12. Yavuz Y, Koparal AS. Electrochemical oxidation of phenol in a parallel plate reactor using ruthenium mixed metal oxide electrode. *J Hazard Mater*. 2006;**136** (2):296-302.
13. APHA, AWWA, WEF. American Public Health Association, American Water Work Association. In: Clesceri L GA, Eaton A, editor. *Standard Methods for the Examination of Water and Wastewater*. 20th ed: Water Environment Federation; 1999.
14. Bayramoglu M, Kobya M, Can OT, Sozbir M. Operating cost analysis of electrocoagulation of textile dye wastewater. *Separ Purif Method* 2004;**37** (2):117-25.
15. Murthy ZVP, Parmar S. Removal of strontium by electrocoagulation using stainless steel and aluminum electrodes. *Desalination*. 2011;**282**:63-7.
16. Aouni A, Fersi C, Ben Sik Ali M, Dhahbi M. Treatment of textile wastewater by a hybrid electrocoagulation/nanofiltration process. *J Hazard Mater*. 2009;**168** (2-3):868-74.
17. Mahvi AH, Bazrafshan E. Removal of Cadmium from Industrial Effluents by Electrocoagulation Process Using Aluminum Electrodes. *World Apply Sci J*. 2007;**2** (1):34-9.
18. Bazrafshan E, Biglari H, Mahvi AH. Phenol removal by electrocoagulation process from aqueous solutions. *Fresen Environ Bull*. 2012;**21** (2):364-71.
19. Bazrafshan E, Biglari H, Mahvi AH. Application of electrocoagulation process using Iron and Aluminum electrodes for fluoride removal from aqueous environment. *E J Chem*. 2012;**9** (4):2297-308.
20. Jeon C, Park JY, Yoo YJ. Characteristics of metal removal using carboxylated alginate acid. *Water Res*. 2002;**36** (7):1814-24.
21. Min JH, Hering JG. Arsenate sorption by Fe (III)-Doped Alginate gels. *Water Res*. 1998;**32** (5):1544-52.
22. Bazrafshan E, Biglari H, Mahvi AH. Humic acid removal from aqueous environments by electrocoagulation process using iron electrodes. *E J Chem*. 2012;**9** (4):2453-61.