



Removal of high concentration phenol from aqueous solutions by electrochemical technique

Shaimaa T. Alnasrawy ^{a*}, Ghayda Y. Alkindi ^b, Talib M. Albayati ^c

^a Department of Civil Engineering, University of Technology, Baghdad, Iraq, 40124@uotechnology.edu.iq

^b Department of Civil Engineering, University of Technology, Baghdad, Iraq, 40126@uotechnology.edu.iq

^c Department of Chemical Engineering, University of Technology, Baghdad, Iraq, Talib.M.Naieff@uotechnology.edu.iq

*Corresponding author.

Submitted: 10/05/2020

Accepted: 02/11/2020

Published: 25/02/2021

KEY WORDS

Aluminum electrodes, current density, electrochemical, phenol, wastewater treatment.

ABSTRACT

In this study, the ability of the electrochemical process to remove aqueous high concentration phenol using an electrochemical cell with aluminum anode and cathode was examined. The removal rate of phenol was monitored using different parameters phenol concentration, pH, electrolysis time, current density, and electrode distance. Obtained results indicated that the low removal rates of phenol were observed at both low and high pH. However, the removal rate of phenol increased with an increase in the current density, each electrochemical process conditions need a certain electrodes distance. removal rate of phenol decreased with the increase in the initial phenol concentration. The maximum removal rate of phenol obtained from this study was 82%.

How to cite this article: S. T. Alnasrawy, G.Y. Alkindi and T. M. Albayati, "Removal of high concentration phenol from aqueous solutions by electrochemical technique," Engineering and Technology Journal, Vol. 39, Part A, No. 02, pp. 189-195, 2021.

DOI: <https://doi.org/10.30684/etj.v39i2A.1705>

This is an open access article under the CC BY 4.0 license <http://creativecommons.org/licenses/by/4.0>

1. INTRODUCTION

Phenols were classified as a priority pollutant, due to their high toxicity to human, aquatic life, and ecosystem even at tinny concentration. Phenol and its derivatives exist in the wastewater of many industries, such as phenol manufacturing, pharmaceuticals, refineries, and petrochemical industries [1]. The high concentration of phenol could be a serious obstruction for wastewater treatment processes.

Several traditional and advanced technics have been utilized for the removal of aqueous phenol such as chemical oxidation [2,3], microbial degradation [4,5], membrane separation [6], photocatalytic degradation [7], solvent extraction [8], ultrasonic degradation [9], enzymatic polymerization [10], Fenton-Like [11], adsorption [12; 13], and electrochemical oxidation [14, 15].

Electrocoagulation is the technique using a sacrificial anode to generate an effective coagulant which is utilized to remove pollutants by physical processes. The electrocoagulation process has

several features like the ability to remove the smallest colloidal pollutant, reduction of a sludge production problem, minimum space requirement, no need for addition or store chemicals, and simple and easy to work [16].

In this study, an electrochemical cell with aluminum electrodes and Na₂SO₄ electrolyte was used to investigate the possibility of the removal of the aqueous high initial phenol concentration. The effect of several parameters such as pH, electrolysis time, current density (CD), electrodes distance, and initial phenol concentration on the removal rate of phenol was examined as well as the consumption of electrical energy was calculated

2. ELECTROCHEMICAL (ELECTROCOAGULATION) MECHANISM

Electrochemical process consists of the following sequence of steps: (1) electrolysis at electrodes which generates aluminum ions at the anode and OH ions at the cathode, (2) oxidation of aluminum ions followed by precipitation of Al(OH)₃, and (3) soluble or colloidal pollutants adsorb onto coagulants which are disposed of by physical processes. The major reactions consist of [17]:



Reaction of aluminum and hydroxyl ions formed by equations (1) and (2) generate different monomeric and polymeric species and convert lastly into Al(OH)₃.

The overall reaction is:



Amorphous Al(OH)₃ with a high specific surface area is useful as an efficient adsorbent for soluble organics and a trap for colloidal pollutants. Lastly, coagulant flocs are disposed of by physical processes [18].

3. MATERIALS AND METHODS

I. Materials

The simulated wastewater was made utilizing phenol crystal (C₆H₅OH) with a purity of 99.5% and deionized water. For pH adjustment, 0.01 M sodium hydroxide (NaOH), 0.01 M sulfuric acid (H₂SO₄) were used and employing pH meter (Crison MM40). Sodium sulfate (Na₂SO₄) was used to enhance the conductivity and minimize the electrode passivation [19]. All chemicals were purchased from Thomas Baker (India).

II. Electrochemical cell

The electrochemical cell utilized in this study is demonstrated in Figure 1, in which a glass reaction tank with dimensions 10 × 6 × 5 cm, two parallel aluminum plates (2.5 x 3 x 0.1 cm) with good electrochemical stability were used as anode and cathode electrodes. Also, a 60V 5A DC power supply (Maisheng MS-605D, China) and magnetic stirrer were used.

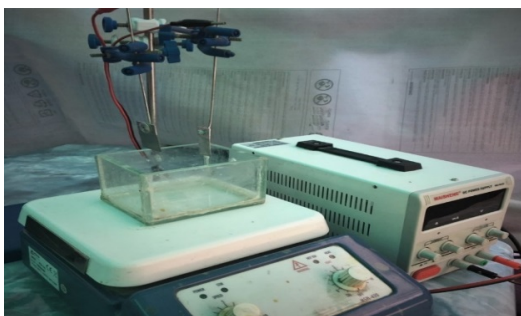


Figure 1: The apparatus used in this study

III. Experimental work

All experiments were performed with solutions of 250ml of deionized water, a specific concentration of phenol, and 0.25 g/l of the electrolyte (Na_2SO_4). The pH adjustment was made using 0.1 N NaOH or 0.1 N H_2SO_4 solutions. Samples were taken at certain intervals to measure phenol concentration. Spectrometric measurements were done by UV 9200 Biotech Engineering, with a maximum λ of 268 nm. Depending on the measured concentration, the removal rate of phenol was calculated.

$$\text{Removal rate \%} = \frac{C_0 - C_t}{C_0} \times 100 \quad (4)$$

where C_0 and C_t are the concentration of phenol (mg/L) at times zero and t , respectively

4. RESULTS AND DISCUSSION

I. Effect of pH

The initial pH value has a significant influence on the electrochemical technique [17] and therefore on the removal rate of phenol. The pH effect on the removal rate of phenol was investigated in the range from 2-5, CD = 40 mA/cm², plate distance = 4 cm, phenol concentration = 0.5 g/L, (Na_2SO_4) = 0.25 g/L, and temperature at 45°C. Figure 2 illustrates the effect of the initial pH of the simulated phenol wastewater on the removal rate of phenol. Figure 2 reveals that the lower values of the removal rates were at high or low pH, and the maximum removal rate was at pH of 4. The possible explanation is that at strong acidic conditions, Al (OH)₃ does not precipitate due to its amphoteric property [20]. Similarly, at the higher pH value, Al (OH)₃ or Al (OH)₄⁻ forms, and this compound is ineffective for phenol adsorption [21].

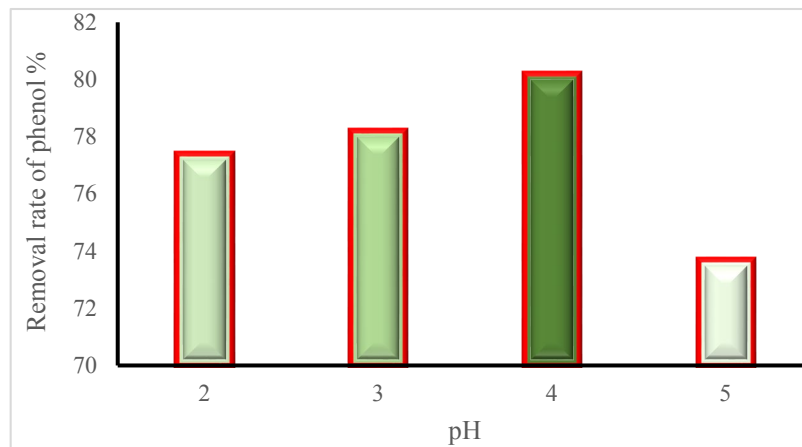


Figure 2: pH effect on removal rate of phenol

II. Effect of electrolysis time

Batch electrochemical experiment was carried out over 90 min and at an initial phenol concentration of 0.5 g/L and 0.25 g/L (Na_2SO_4), the temperature of 45°C, pH of 4, CD of 40 mA/cm², and plate distance of 4 cm as initial conditions. As illustrated in Figure 3, the increase in the processing time was accompanied by a rise in the removal rate of phenol until it reached 30 min. After this time, there was no considerable increase in the removal rate of phenol. The probable causes of these results are the decrease in the chance of contact of the pollutants with the electrode due to the reduction in the concentration of pollutants after a specific time within the electrochemical process and the second potential cause is the formation of intermediate recalcitrant compounds due to the insufficient oxidation of organics [22].

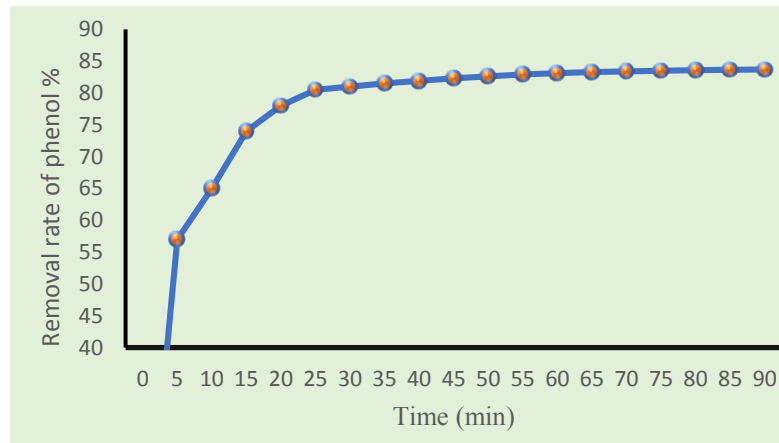


Figure 3: Effect of the electrolysis time on the removal rate of phenol

III. Effect of current density (CD)

The influence of CD on the removal rate of phenol was carried out with an initial phenol concentration of 0.5 g/L and 0.25 g/L Na_2SO_4 , temperature of 45°C , pH of 4, electrolysis time of 30 min, and plate distance of 4 cm, and the current density was changed from 20 to 60 mA/cm^2 . As depicted in Figure 4, the removal rate of phenol increased with the rise in CD. The increase in the current density meant an increase in the transmission of electrons, which promoted the redox reactions of the pollutants on the electrodes [23]. Nevertheless, when the current density surpassed 40 mA/cm^2 , the removal rate of phenol revealed a slight and inconsiderable increase. The explanation of these results is the generation of additional oxygen and hydrogen bubbles, as a result of the increase in CD, limit the contact of the pollutants with the electrodes [24]. For maximal energy savings and an appropriate removal rate of phenol, the optimum CD was 50 mA/cm^2 . The consumption of electrical energy is represented in the following:

$$EC = \frac{VIt}{v} \quad (5)$$

where EC is the energy consumption (kWh/m^3), V is the voltage (V), I is the applied current (A), t is the electrolysis time (min), and v is the volume of wastewater (m^3). For a CD of 50 mA/cm^2 and an electrolysis time of 30 min, the energy consumption was 6.34 $\text{KW.h}/\text{m}^3$.

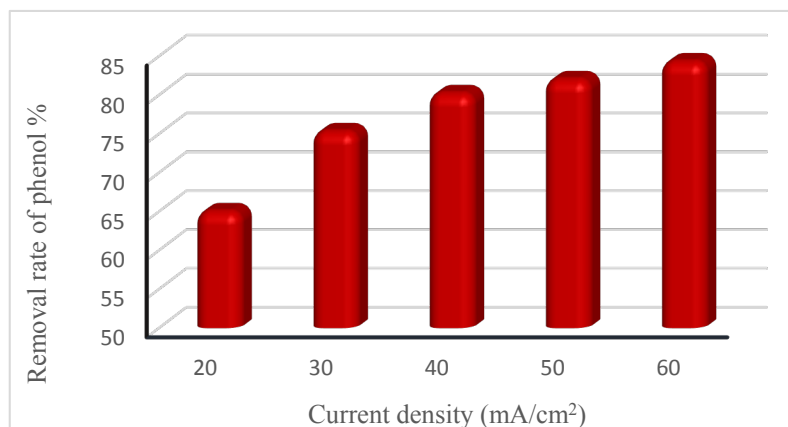


Figure 4: Effect of the current density on the removal rate of phenol

IV. Effect of electrodes distance

Electrochemical experiments were performed with four varying electrodes plates distances (3, 4, 5, and 6 cm), and with an initial phenol concentration of 0.5 g/L and 0.25 g/L Na_2SO_4 , temperature of 45°C , pH of 4, electrolysis time of 30 min, and current density of 50 mA/cm^2 to investigate the effect of electrodes plate distance on the removal rate of phenol. Figure 5 showed that at an electrode distance of 4 cm, the removal of phenol is at its highest level, and a slight decrease in the removal rate when the electrode distance exceeded 4 cm. This decrease was perhaps due to the decline in the formation of aluminum cations, which resulted from the increase in the ohmic potential. Hence, there

was a decrease in the effectiveness of the electrochemical process [25]. The maximum removal rate of phenol was 82%.

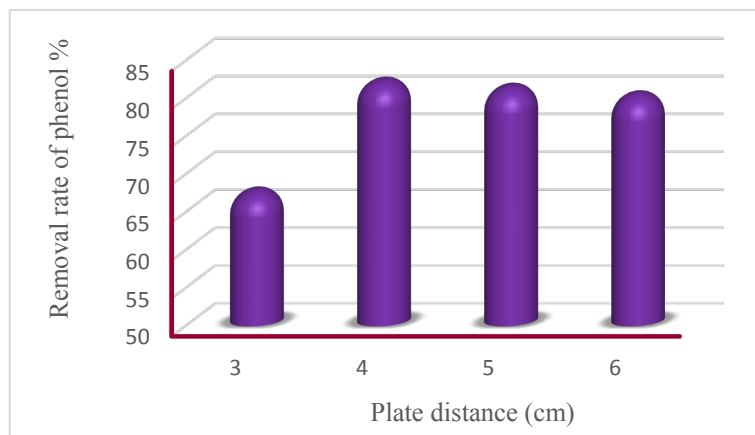


Figure 5: Effect of the electrode plate distance on phenol removal.

V. Influence of initial phenol concentration

The influence of the phenol concentration was studied using varying phenol concentrations from 0.5 to 1.5 g/L. Batch experiments were done with 0.25 g/L Na_2SO_4 , temperature of 45°C, pH of 4, electrolysis time of 30 min, CD of 50 mA / cm^2 , and electrodes plate distance of 4 cm. Figure 6, showed that the efficiency of the electrochemical cell to remove phenol is inversely related to the initial phenol concentration. This behavior can be explained by the fact that the amount of aluminum ions that pass to the electrolyte is the same when the density of the electric current is constant and the amount of complex aluminum hydroxide formed is the same and independent of phenol concentration and it stood short of coagulating the extra amount of phenol when the phenol concentrations were raised [17].

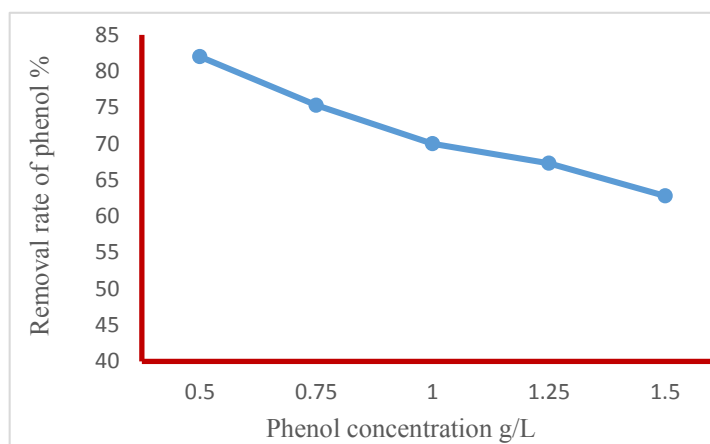


Figure 6: Effect of initial phenol concentration on phenol removal

VI. Optimum operating conditions

The parameters of optimal operating conditions for the maximum removal rate of phenol were established. The optimum operating conditions were pH = 4, electrolysis time = 30 min, current density = 50 mA/ cm^2 , electrode distance = 4 cm, and initial phenol concentration = 0.5 g/l. The maximum removal rate of phenol at the optimum operating conditions was 82 %.

5. CONCLUSION

The present work revealed that the electrochemical treatment of aqueous phenol using aluminum metal electrodes is an effective method. Experimental parameters like pH, electrolysis time, current density, electrode distance, and initial phenol concentration were studied for phenol removal. The optimal operation conditions which provided maximum removal rate of phenol of 82% were pH = 4, electrolysis time = 30 min, CD = 50 mA/ cm^2 , electrode distance = 4 cm, and initial phenol concentration = 0.5 g/l.

Acknowledgments

The authors gratefully acknowledge the scientific support and help of the Civil Engineering Department and Chemical Engineering Department, University of Technology, Baghdad, Iraq.

References

- [1] A.M. Khaksar, S. Nazif, A. Taebi, E. Shahghasemi, Treatment of phenol in petrochemical wastewater considering turbidity factor by backlight cascade photocatalytic reactor, *J. Photochem. Photobiol.*, 348 (2017) 161–167. doi.org/10.1016/j.jphotochem.2017.08.034
- [2] Y. Yavuz, S. Koparal, Electrochemical oxidation of phenol in a parallel plate reaction using ruthenium mixed metal oxide electrode, *J. Hazard. Mater.*, 136 (2006) 292-302. doi.org/10.1016/j.jhazmat.2005.12.018
- [3] Y. Wang, B. Gu, W. Xu, Electrocatalytic degradation of phenol on several metal-oxide anodes, *J. Hazard. Mater.*, 162 (2009) 1159-1164. doi.org/10.1016/j.jhazmat.2008.05.164
- [4] M. Shourian, K. Noghabi, H. Zahiri, T. Bagheri, G. Karaballaei, M. Mollaei, I. Rad, S. Ahadi, J. Raheb, H. Abbasi, Efficient phenol degradation by a newly characterized *Pseudomonas* sp. SA01 isolated from pharmaceutical wastewaters, *Desalination*, 246 (2009) 577-594. doi.org/10.1016/j.desal.2008.07.015
- [5] M.H. EL-Naas, S. Al-Zuhair, S. Makhlof, Batch degradation of phenol in a spouted bed bioreactor system, *J. Ind. Eng. Chem.*, 16 (2010) 267-272. doi.org/10.1016/j.jiec.2009.09.072
- [6] G. Busca, S. Berardinelli, C. Resini, L. Arrighi, Technologies for the removal of phenol from fluid streams, a short review of recent developments, *J. Hazard. Mater.*, 160 (2008) 265-288. doi.org/10.1016/j.jhazmat.2008.03.045
- [7] A. Agrios, K. Gray, E. Weitz, Photocatalytic transformation of 2,4,5- trichlorophenol on TiO₂ under sub-band-gap illumination, *Langmuir*, 19 (2003) 1402-1409. doi.org/10.1021/la026397x
- [8] C. Yang, Y. Qian, L. Zhang, J. Feng, Solvent extraction process development and on-site trial-plant for phenol removal from industrial coal-gasification wastewater, *Chem. Eng. J.*, 117 (2006) 179-185. doi.org/10.1016/j.ccej.2005.12.011
- [9] A.B. Pandit, P.R. Gogate, S. Mujumdar, Ultrasonic degradation of 2:4:6 trichlorophenol in presence of TiO₂ catalyst, *Ultrason. Sonochem.*, 8 (2001) 227-231. [doi.org/10.1016/S1350-4177\(01\)00081-5](https://doi.org/10.1016/S1350-4177(01)00081-5)
- [10] I.D. Buchanan, J.A. Micell, Model development for horseradish peroxidase catalyzed removal of aqueous phenol, *Biotechnol. Bioeng.*, 54 (1997) 251-261. [doi.org/10.1002/\(SICI\)1097-0290\(19970505\)54:3<251::AID-BIT6>3.0.CO;2-E](https://doi.org/10.1002/(SICI)1097-0290(19970505)54:3<251::AID-BIT6>3.0.CO;2-E)
- [11] MI. Pariente, R. Molina, JA. Melero, JÁ. Botas, F. Martínez, Intensified-Fenton process for the treatment of phenol aqueous solutions, *Water Sci Technol.*, 71 (2015) 359–65. doi.org/10.2166/wst.2014.515
- [12] A. Mohd Din, B.H. Hameed, A.L. Ahmed, Batch adsorption of phenol onto physicochemical-activated coconut shell, *J. Hazard. Mater.*, 161 (2009) 1522-1529. doi.org/10.1016/j.jhazmat.2008.05.009
- [13] J.M. Valente Nabais, J.A. Gomes Suhas, P.J.M. Carrott, C. Laginhas, S. Roman, Phenol removal onto novel activated carbons made from lignocellulosic precursors: influence of surface properties, *J. Hazard. Mater.*, 167 (2009) 904-910. doi.org/10.1016/j.jhazmat.2009.01.075
- [14] P. H. Britto-Costa, L. A. M. Ruotolo, Phenol removal from wastewaters by electrochemical oxidation using boron doped diamond (BDD) and Ti/Ti_{0.7}Ru_{0.3}O₂ DSA electrodes, *Braz. J. Chem. Eng.*, 29 (2012) 763 - 773. doi.org/10.1590/S0104-66322012000400008
- [15] IM Pillai, AK. Gupta, Batch and continuous flow anodic oxidation of 2,4-dinitrophenol: modeling, degradation pathway and toxicity, *J. Electroanal. Chem.*, 756 (2015) 108–117. doi.org/10.1016/j.jelechem.2015.08.020
- [16] M. Bharath, B M. Krishna, B. M. Kumar, A Review of Electrocoagulation Process for Wastewater Treatment, *Int. J. Chemtech Res.*, 11 (2018) 289-302.
- [17] O. Abdelwahab, N.K. Amin, E-S.Z. El-Ashtoukhy, Electrochemical removal of phenol from oil refinery wastewater, *J. Hazard. Mater.*, 163 (2009) 711-716. doi.org/10.1016/j.jhazmat.2008.07.016
- [18] M. Kobya, E. Demirbas, O.T. Can, M. Bayramoglu, Treatment of levafix orange textile dye solution by electrocoagulation, *J. Hazard. Mater.*, 132 (2006) 183–188. doi.org/10.1016/j.jhazmat.2005.07.084
- [19] G.V. Sleptsov, A.I. Gladkii, and E.Y. Sokol, Electrocoagulation treatment of oil emulsion wastewater of Industrial enterprises, *Surf. Eng. Appl. Electrochem*, 6 (1987) 96-99.

- [20] N. Adhoum, L. Monser, Decolorization and removal of phenolic compounds from olive mill wastewater by electrocoagulation, *Chem. Eng. Process.*, 43 (2004) 1281–1287. doi.org/10.1016/j.cep.2003.12.001
- [21] J. Zhu, H. Zhao, J. Ni, Fluoride distribution in electrocoagulation defluoridation process, *Sep. Purif. Technol.*, 56 (2007) 184–191. doi.org/10.1016/j.seppur.2007.01.030
- [22] Z. Chi, Z. Wang, Y. Liu, G. Yang, Preparation of organosolv lignin-stabilized nano zero-valent iron and its application as granular electrode in the tertiary treatment of pulp and paper wastewater, *J. Chem. Eng.*, 331(2018) 317-325. doi.org/10.1016/j.cej.2017.08.121
- [23] C. Zexu, Z. Wang, C. Huanqing, B. Pingping, and L. Lucia, Bentonite-supported nanoscale zero valent iron granulated electrodes for industrial wastewater remediation, *R. Soc. Chem. paperb.*, 7 (2017) 44605-44613. doi.org/10.1039/C7RA07584G
- [24] C. Wang, Y.K. Huang, Q. Zhao, M. Ji, Treatment of secondary effluent using a three-dimensional electrode system: COD removal, biotoxicity assessment and disinfection effects, *Chem. Eng. J.*, 243 (2014) 1–6. doi.org/10.1016/j.cej.2013.12.044
- [25] O. Sahu, Electro-oxidation and chemical oxidation treatment of sugar industry wastewater with ferrous material: An investigation of physicochemical characteristic of sludge, *S. Afr. J. Chem. Eng.*, 28 (2019) 26-38. doi.org/10.1016/j.sajce.2019.01.004