



Electro-Oxidation as an Effective Process for Removing Antibiotics and Persistent Organic Compounds Resistant to Biodegradation

Reza Ali Fallahzadeh^{1,2}, Fariborz Omid^{3*}

¹ *Environmental Science and Technology Research Center, Department of Environmental Health Engineering, Shahid Sadoughi University of Medical Sciences, Yazd, Iran.*

² *Genetic and Environmental Adventures Research Center, School of Abarkouh Paramedicine, Member of Student Research Committee, Shahid Sadoughi University of Medical Sciences, Yazd, Iran.*

³ *Research Center for Environmental Determinants of Health (RCEDH), Health Institute, Kermanshah University of Medical Sciences, Kermanshah, Iran.*

ARTICLE INFO

LETTER TO EDITOR

Article History:

Received: 09 August 2019

Accepted: 20 October 2019

***Corresponding Author:**

Fariborz Omid

Email:

omidifariborz@yahoo.com

Tel:

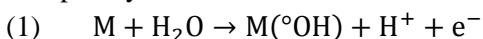
+989128360892

Citation: Fallahzadeh RA, Omid F. *Electro-Oxidation as an Effective Process for Removing Antibiotics and Persistent Organic Compounds Resistant to Biodegradation*. J Environ Health Sustain Dev. 2019; 4(3): 862-5.

Releasing a large amount of hazardous synthetic chemicals to the environment is one of the results of human daily activities; some of which could enter the water cycle and contaminate it and threaten the health of consumers. Treatment of the wastewaters containing chemicals is one of the routine ways to reduce the chemical-induced risks as well as environmental pollution^{1, 2}. Over the past 15 years, drugs have been widely considered as potentially bioactive chemicals in the environment^{3, 4}. Having high persistence in the environment, these compounds affect the quality of drinking water resources and, as a potential hazard to the ecosystem, could affect human and animal welfare in the future⁵. Antibiotics are high consuming drugs in which, worldwide use of antibiotics have been increased by 36 percent between 2000 and 2010. According to the WHO, people in the European Region had a median consumption of 17.9 defined daily doses (DDD)/1000 inhabitants per day. This had a 4-fold

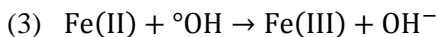
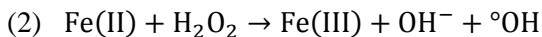
difference between the lowest- and the highest-consuming country in the region⁶. Annually, between 100,000 to 200,000 tons of antibiotics are consumed worldwide; therefore, large amounts of antibiotics are continuously introduced into the aquatic and soil environments⁷. Among the different types of antibiotics, the beta-lactam family is the most widely used drug⁸. Deaths of aquatic organisms such as fish⁹, algae¹⁰, and the development of drug resistance of microorganisms are some of the adverse effects of uncontrolled discharge of wastewater containing antibiotics to the environment¹¹. Due to presence of organic pollutants which are resistant to biodegradation and also the sensitivity of microorganism against toxic substances, biological methods are not suitable for the treatment of these kinds of wastewaters^{12, 13}. In recent years, many efforts have been made to develop new treatment processes able to overcome the problem of antibiotic resistance to biological purification.

Moreover, various treatment methods based on advanced oxidation processes (AOPs) were conducted to remove the toxic compounds and resistant organic pollutants. Such processes can produce active OH° radical species that is able to oxidize resistant organic compounds¹⁴. Produced OH° can non-selectively oxidize and mineralize the persistent organic pollutants⁴. Electrochemical processes are among the advanced oxidation processes. Recently, in the various studies, electrochemical processes are extensively used as simple, economical, safe and environmental friendly technologies to remove versatile pollutants from aquatic environments¹⁵. Most of electrochemical processes for wastewater treatment are divided into two categories: electrocoagulation and electrolysis¹⁶. The electrocoagulation process is based on the production of coagulants through the reduction of the electrode in anode; the produced coagulant reacts with the pollutant and settles. In electrolysis, the anodic oxidation process of the contaminant destroys and removes the contaminant from the solution; the produced sludge in the electrolysis process is much less in comparison to electrocoagulation process¹⁷. Electro-Oxidation (EO) processes degrade organic compounds in two ways, one being "direct oxidation" on the anode surface, and the other "indirect oxidation" by the products produced on the anode surface. Anodic oxidation is one of the electrochemical advanced oxidation processes (EAOPs) that is capable of effectively oxidizing organic compounds present in aquatic environments. This process is operated by the hydroxide radicals produced at the anode surface (M) and the oxidation of water according to equation 1¹⁸. Surface-produced hydroxyl radicals $\text{M}(\text{OH}^\circ)$ are highly non-selective oxidizing agents which are capable to destroy organic pollutant completely and mineralized them¹⁹.



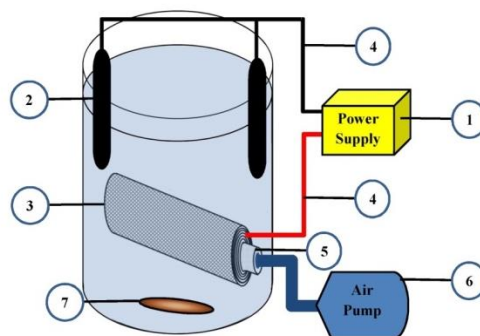
Considering AOP procedures, the Fenton-based process because of high efficiency, ease of application, and low cost is one of the most commonly used procedures²⁰. This process is one of the advanced oxidation processes which can

effectively destroy persistent aromatic compounds through the production of potent free radicals. Fenton process is accomplished by the reaction between iron II and hydrogen peroxide-based on formula 2 and 3²¹:



Because of the high potential of hydroxyl radicals (2.8 V) created by the Fenton reaction, this reaction is utilized to destroy organic compounds that are resistant to biodegradation¹⁹. The efficiency of this process relies on some variables such as temperature, pH, and the concentration of hydrogen peroxide and Fe^{2+} ²². Producing a high amount of sludge in electrocoagulation process is the most disadvantage, but using new methods has reduced its production¹⁹. In recent years, the use of porous electrodes as the cathode electrode, which increases the contact surface of the electrode with the sample solution, has significantly been used as an efficient technique for pollutant removal.

Figure 1 illustrates an Electro-Oxidation reactor to produce H_2O_2 using porous cathode. In this reactor, H_2O_2 is produced by reducing 2 electrons from oxygen (either purely or by injecting air into the solution) at the cathode surface under acidic or normal conditions²³.



1. Power supply
2. Graphite electrodes (anode)
3. Steel porous electrode (cathode)
4. Interface cable
5. Porous air injection tube
6. Blower pump
7. Magnetic stirrer

Figure 1: An Electro-Oxidation reactor using porous cathode to produce H_2O_2 ²³

The application of porous carbon electrodes (e.g. graphite and carbon felts²⁴, carbon sponges²⁵, activated carbon fibers¹² and carbon nanotubes²⁶ have been significantly increased because of their characteristics such as transmission of current density, stability, and cost. Porous electrodes can be utilized in electro-oxidation and photoelectro-Fenton processes. Using porous electrode as the cathode electrode in the electro-oxidation processes, the contact surface between the electrode, air and wastewater will retain at its maximum value; therefore, the production of oxidants in solution will be as high as possible, causing the destruction of toxic and persistent compounds suitably conducted. Considering the technology development and production of degradation-resistant pollutants that enter the environment, the use of processes that can quickly and non-selectively remove these compounds is great of importance. Therefore, the development of electrooxidation-based processes can effectively increase the speed and efficiency of degradation of these pollutants and also protect the environment against these pollutants.

This is an Open Access article distributed in accordance with the terms of the Creative Commons Attribution (CC BY 4.0) license, which permits others to distribute, remix, adapt and build upon this work for commercial use.

References

1. Azadi NA, Fallahzadeh RA, Sadeghi S. Dairy wastewater treatment plant in removal of organic pollution: a case study in Sanandaj, Iran. *Environmental Health Engineering and Management Journal*. 2015;2(2):73-7.
2. Rezaei M, Karimi F, Parviz M, et al. An empirical study on aflatoxin occurrence in nuts consumed in tehran, Iran. *Health*. 2014;6(8):649.
3. Ghaneian MT, Tabatabaee M, Ehrampoush MH, et al. Synthesis of Ag (I) and Cu (I) complexes with 4-Amino-5-Methyl-2h-1, 2, 4-Triazole-3 (4h)-thione ligand as thiocarbohydrazide derivatives and their antimicrobial activity. *Pharmaceutical Chemistry Journal*. 2015;49(3):210-2.
4. Ghaneian MT, Ebrahimi A, Salimi J, et al. Photocatalytic degradation of 2, 4-Dichlorophenoxyacetic acid from aqueous solutions using In₂O₃ nanoparticles. *Journal of Mazandaran University of Medical Sciences*. 2016; 26(137):159-70.
5. Klavarioti M, Mantzavinos D, Kassinos D. Removal of residual pharmaceuticals from aqueous systems by advanced oxidation processes. *Environment international*. 2009;35(2):402-17.
6. World Health Organization. Wide difference in antibiotic use between countries, according to new data from WHO. Available from: https://www.who.int/medicines/areas/rational_use/oms-amr-amc-report-2016-2018/en/ [cited July 30, 2019].
7. Fallahzadeh RA, Mahvi AH, Meybodi MN, et al. Application of photo-electro oxidation process for amoxicillin removal from aqueous solution: Modeling and toxicity evaluation. *Korean J Chem Eng*. 2019;36(5):713-21.
8. Van Boeckel TP, Gandra S, Ashok A, et al. Global antibiotic consumption 2000 to 2010: an analysis of national pharmaceutical sales data. *Lancet Infect Dis*. 2014;14(8):742-50.
9. Yu X, Zuo J, Li R, et al. A combined evaluation of the characteristics and acute toxicity of antibiotic wastewater. *Ecotoxicol Environ Saf*. 2014;106:40-5.
10. Trovo AG, Nogueira RFP, Agüera A, et al. Degradation of the antibiotic amoxicillin by photo-Fenton process—chemical and toxicological assessment. *Water Res*. 2011;45(3):1394-402.
11. Sirtori C, Zapata A, Oller I, et al. Decontamination industrial pharmaceutical wastewater by combining solar photo-Fenton and biological treatment. *Water Res*. 2009;43(3): 661-8.
12. Wang A, Li Y-Y, Estrada AL. Mineralization of antibiotic sulfamethoxazole by photoelectro-Fenton treatment using activated carbon fiber cathode and under UVA irradiation. *Appl Catal B*. 2011;102(3-4):378-86.
13. Eslami H, Sedighi Khavidak S, Salehi F, et al. Biodegradation of methylene blue from aqueous solution by bacteria isolated from contaminated soil. *J Environ Manage*. 2017;5(1):10-5.

14. Cotillas S, de Vidales MJM, Llanos J, et al. Electrolytic and electro-irradiated processes with diamond anodes for the oxidation of persistent pollutants and disinfection of urban treated wastewater. *J Hazard Mater.* 2016; 319:93-101.
15. Dalvand A, Gholami M, Joneidi A, et al. Dye removal, energy consumption and operating cost of electrocoagulation of textile wastewater as a clean process. *Clean (Weinh).* 2011;39(7): 665-72.
16. Alimohammadi M, Askari M, Dehghani M, et al. Elimination of natural organic matter by electrocoagulation using bipolar and monopolar arrangements of iron and aluminum electrodes. *Int J Environ Sci Technol (Tehran).* 2017;14(10):2125-34.
17. Shan L, Liu J, Ambuchi JJ, et al. Investigation on decolorization of biologically pretreated cellulosic ethanol wastewater by electrochemical method. *Chem Eng J.* 2017;323:455-64.
18. Trellu C, Oturan N, Pechaud Y, et al. Anodic oxidation of surfactants and organic compounds entrapped in micelles—Selective degradation mechanisms and soil washing solution reuse. *Water Res.* 2017;118:1-11.
19. Oturan MA, Aaron J-J. Advanced oxidation processes in water/wastewater treatment: principles and applications: A review. *Crit Rev Environ Sci Technol.* 2014; 44(23): 2577-641.
20. Ghaffari Y, Mahvi A, Alimohammadi M, et al. Evaluation of fenton process efficiency in removal of tetracycline from synthetic wastewater. *Journal of Mazandaran University of Medical Sciences.* 2017;27(147):291-305.
21. Bishop D, Stern G, Fleischman M, et al. Hydrogen peroxide catalytic oxidation of refractory organics in municipal wastewaters. *Ind Eng Chem Res.* 1968;7(1):110-7.
22. Homem V, Alves A, Santos L. Amoxicillin degradation at ppb levels by Fenton's oxidation using design of experiments. *Sci Total Environ.* 2010;408(24):6272-80.
23. Fallahzadeh RA, Ehrampoush MH, Mahvi AH, et al. Designing and modeling of a novel electrolysis reactor using porous cathode to produce H₂O₂ as an oxidant. *Methods X.* 2019;6:1305-12.
24. Zhou M, Tan Q, Wang Q, et al. Degradation of organics in reverse osmosis concentrate by electro-Fenton process. *Journal of hazardous materials.* 2012;215:287-93.
25. Özcan A, Şahin Y, Kopal AS, et al. Carbon sponge as a new cathode material for the electro-Fenton process: comparison with carbon felt cathode and application to degradation of synthetic dye basic blue 3 in aqueous medium. *J Electroanal Chem.* 2008;616(1-2):71-8.
26. Khataee AR, Zarei M, Khataee AR. Electrochemical treatment of dye solution by oxalate catalyzed photoelectro-fenton process using a carbon nanotube-ptfe cathode: optimization by central composite design. *Clean (Weinh).* 2011;39(5):482-90.