

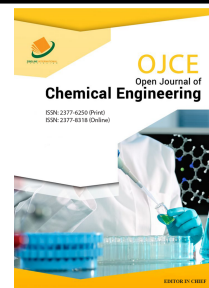


ISSN: 2377-6250 (Print)
ISSN: 2377-8318 (Online)

CODEN : OJCEB4

Open Journal of Chemical Engineering (OJCE)

DOI : <http://doi.org/10.26480/ojce.01.2018.01.03>



DEVELOPMENT OF ELECTRO-FENTON TECHNIQUE AND THE STUDY ON BACTERIAL INACTIVATION IN SEAWATER BY Nb-Ta/ACF BIPOLAR ELECTRO-FENTON

Jin Zhang*, Daming Xue, Xuelu Xu

School of Environmental Science and Engineering, Dalian Maritime University, Dalian 116000, China

*Corresponding Author E-mail: zhangjin7986@163.com

This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

ARTICLE DETAILS

Article History:

Received 12 November 2017
Accepted 10 December 2017
Available online 06 January 2018

ABSTRACT

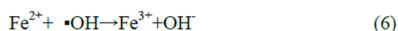
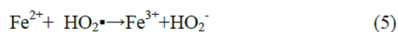
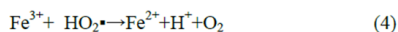
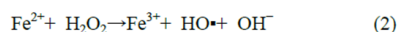
Electro-Fenton technology is a new type of electrochemical advanced oxidation technology in water treatment in recent years. The treatment for microorganisms, such as bacteria, has been the main problem of discharge of ballast water. In this paper, a new type of bipolar electro-Fenton of Nb-Ta/ACF was proposed, and the inactivation for bacteria by the bipolar electro-Fenton technology was studied. The results showed that the Nb-Ta/ACF bipolar electro-Fenton technique is effective for the inactivation for bacteria, and the sterilization efficiency could reach up 99%. Electrodes materials, operation voltage, pH and electrolytic time have influence the sterilization efficiency. In experimental range, the optimum pH was 8.17, which is the pH of raw water without adjustment. High voltage is helpful to improve the sterilization efficiency. Electrodes materials affect the sterilization efficiency by the bipolar electro-Fenton process. Anode of Nb-Ta (Niobium Tantalum) is much better than the effect of anode of Fe.

KEYWORDS

Bipolar electro-Fenton process, hydroxyl radical, sterilization, wastewater treatment, seawater

1. INTRODUCTION

In recent studies, advanced oxidation technologies have been described as efficient procedures for obtaining high oxidation yields from several kinds of organic compounds [1-3]. According to a scholar, as one of advanced oxidation technology, electro-Fenton technology has been researched for treatment for refractory organic wastewater and has achieved preferable development [4-7]. Based on a study, Electro-Fenton is mainly relied on an in situ and catalytic production of Fenton's reagent – a mixture of hydrogen peroxide (H₂O₂) and ferrous iron (Fe²⁺) to produce hydroxyl radicals to react with organic pollutants in water and finally leading to their destruction [8-10].



Compared with the traditional Fenton's reagent, there have many advantages in electro-Fenton technology. Firstly, electro Fenton technology can realize the in-situ production of H₂O₂, which effectively avoid risks in the transportation, storage or reagent treatment. Secondly, due to the in reagent treatment. Secondly, due to the in situ production of H₂O₂ and Fe²⁺ and ensure the reaction, the Fenton reaction does not need adding of chemical reagents, which greatly reduce the cost. Thirdly, in the process of electrolysis, Fe³⁺ could be reduced to Fe²⁺, which reduced the sludge production. Fourthly, in addition to the oxidation of hydroxyl radicals, there has anodic oxidation and electro adsorption and so on in process of the electro-Fenton, the collaboration of these roles improves the removal efficiency for pollutants from water [11].

The reagent of Fe²⁺ and H₂O₂ were produced in-situ in anodes and cathodes in electro-Fenton process, the materials of electrodes are important. According to a scholar, the cathode materials usually contain mercury electrode, graphite, carbon- polytetrafluoroethylene (PTFE) gas diffusion electrode, three-dimensional porous electrode and so on [12-14]. Recently, because of its no toxicity, higher hydrogen evolution potential, low catalytic activity for decomposition of H₂O, good chemical stability, conductivity and resistant to corrosion et al., activated carbon materials has been used as alternative reduction cathode material. Because the solubility of O₂ in aqueous solution is low, gas diffusion electrode and the porous electrode become the best choice for cathode. The anode's material was metal, and the metal ions were produced by anode electrolysis. So, the materials affect the kinds of ion, and also affect the catalytic effect. So, different materials of anode would affect the efficiency of the electro-Fenton.

2. FACTORS AFFECT ELECTRO FENTON PROCESS

Many factors affect the process of electro Fenton, such as the electrolytic voltage, solution conductivity, pH, current density, aeration rate and so on.

2.1 Effect of Electrolytic Voltage

The applied voltage is the driving force for electro-Fenton reaction. Increase the voltage, the removal efficiency of organic matters increased. But, when the voltage is high, there will be a large number of energy consumed in side reactions. So, the applied voltage should not very high, in general, 5-25 V is appropriate.

2.2 Effect of Solution Conductivity

Solution conductivity is important in electro Fenton process. Under low

conductivity, the current efficiency is low, and the electrolysis process would be suppressed. Usually, the solution conductivity of organic wastewater is very poor, so supporting electrolyte should be added before.

2.3 Effect of pH

The pH of pollution influences the reaction of oxygen. In alkaline condition, O₂ would react to produce OH⁻ (seen as formula (7)), while under acidic condition, O₂ react to produce H₂O₂ (seen as formula (8)). To generate more ·OH, the pH of water was usually controlled in 1.0-3.5.



2.4 Effect of Current Density

In theory, large current density is favorable for the reaction of electrolysis. But, on the other hand, large current density would increase the polarization of electrode, which leads to the side reactions in the electrodes, and is unfavorable for the produce of OH. So, in actually, the current density should be controlled within a proper range.

2.5 Effect of Aeration Rate

In electro Fenton process, the flow of air supplies the O₂ needed, and mainly influences the mass transfer process. In the absence of air supply, the mass transfer rate of O₂ is slow, and the O₂ take part in the electro-Fenton reaction comes from dissolve oxygen and electrolysis, which is unfavorable for the reaction of electro-Fenton process. According to a scholar, when the air flow increased to a certain value, the O₂ increased, and the electro-Fenton reaction rate was controlled by reaction process, and then the effect of aeration on the reaction is very small [15].

2.6 Effect of Electrode Materials

Study on electrode materials is always the hot issues of electrochemical oxidation. In general, the anode materials contain Pt and Diamond carbon coating of borax (BDD) and other inert materials with high oxygen evolution potential. The cathode materials usually include porous graphite, carbon-PTFE, activated carbon fiber (ACF) etc. On the other hand, nanostructure electrodes have been applied in EF systems. The discharge of ballast water would cause biological invasion and threat the coastal safety seriously. The treatment for microorganisms, such as bacteria, has been the main problem of discharge of ballast water. In this paper, a bipolar electro-Fenton process was put forward to treat simulated ballast water, and bacteria were used as subjects of microorganism. Through systematic analysis, the feasibility of the technology has been discussed.

3. EXPERIMENTAL SECTION

Electro-Fenton process was performed in batch mode in an undivided glass cell under vigorous stirring performed by a magnetic stirrer with 1000 mL of solution. The anode was a sheet of iron and Nb-Ta of size of 10 cm*8 cm*0.5 cm, and cathode was an activated carbon fiber of size of 10 cm*8 cm. Compressed air was fed to the cathode by an air pump (seen as Figure 1). Electrolysis was performed with a DC power supply.

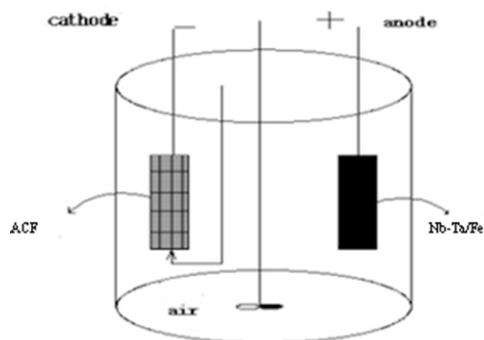


Figure 1: Schematic diagram of the bipolar electro-Fenton system.

Electrolysis was performed at room temperature, and pH of water was adjusted by addition of solution of 0.1 mol/L sulfuric acid and sodium hydroxide. The number of bacteria was analysis by microscopic counting method.

4. RESULTS AND DISCUSSION

In this study, the effects of pH, electrolysis time and operational voltage were investigated. The results of this study are given below.

4.1 Sterilization Effect by Nb-Ta-ACF Bipolar Electro Fenton Process

In this part, the anode used was a sheet of Nb-Ta, and the cathode was a sheet of Activated Carbon Fiber. The initial content of bacteria was 2380 cfu/ml. The sterilization results were seen as in Table 1 and Figure 2.

Table 1: Inactivation effect for bacteria (cfu/ml).

Voltage (v)	pH	Electrolysis time (min)			
		5	10	15	20
2	8.17	1575	700	675	235
6	81.7	275	100	50	25
4	8.79	1200	110	50	40
4	5.09	1460	555	335	315

From the results of Table 1 and Figure 2, it could be seen that the voltage and pH have effect on the sterilization in process of the electro-Fenton. When the voltage increased, the sterilization efficiency increased. For example, when voltage was 2 V, the sterilization efficiency was 44.44%-99.11% in 5-20 min at pH of 8.17, while when voltage increased to 4 V, the sterilization efficiency increased to 90.29%-99.71% in 5-20 min at the same pH value. On the other hand, when voltage is stable, the sterilization efficiency increased with the increase of pH. For example, when voltage was 4 V, the sterilization efficiency increased from 48.5%-88.88% at pH of 5.09 to 57.67%-98.58% at pH of 8.79 in 5-20 min.

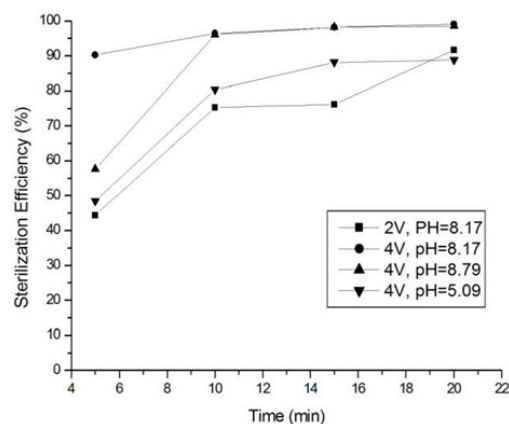


Figure 2: Sterilization by Nb-Ta/ACF bipolar electro-Fenton.

From the results of the same pH value in Figure 2, it could be seen that the pH nears neutral is favorable for the removal of bacteria. When the voltage is the same, the sterilization efficiency at pH of 8.17 is high than that at pH of 8.79 and 5.09.

4.2 Sterilization Effect by Fe/ACF Bipolar Electro Fenton Process

In this part, the cathode used was a sheet of Activated Carbon Fiber, but the anode used was a sheet of iron. The initial content of bacteria was 2256 cfu/ml. The sterilization results were seen as in Figure 3.

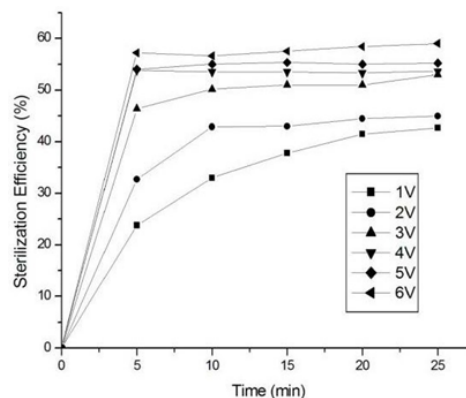


Figure 3: Sterilization by Fe/ACF bipolar electro-Fenton.

From the results in Figure 3, it could be seen that the sterilization rate increased with the process of the electrolysis. At the same time, the sterilization efficiency increased significantly in the first 5 min, and then increased slowly. When the voltage was 1 V~6 V, the sterilization rate for bacteria in the first 5min was 23.8, 32.7, 46.4, 53.8, 54.0 and 57.2%, and the sterilization rate in 25 min was 42.7, 45.0, 53.0, 53.6, 55.2 and 59.0 respectively. It could be seen that when voltage is low, the electrolysis time would influence the sterilization obviously, while, when the voltage is high, the influence of electrolysis time on the sterilization was very small, and the sterilization could be reach to a higher level in short time, and then the voltage would become the main factor influence the sterilization rate. So, in actually, the voltage and electrolysis time should be adjusted according to actual situation.

5. CONCLUSION

This work has shown that the bipolar electro-Fenton technique has good effect for sterilization in seawater. In this system, the alkaline condition was favorable for the inactivation of bacteria. Many factors, such as pH, anode materials, electrolysis time and operational voltage, affect the sterilization. At the experimental situation, the optimum pH was 8.17 (pH value of the raw water). In actually, the pH of seawater is usually in 8.0-8.5, and this is favorable for the treatment of bacteria with the process of bipolar electro-Fenton. Anode materials have effect on the sterilization by the bipolar electro-Fenton system. When Nb-Ta was used as anode, the sterilization could reach up to 99.11%, while when used with Fe, the sterilization efficiency was only nearly 60%. Under the same conditions, improve the operation voltage could improve the sterilization efficiency.

ACKNOWLEDGEMENTS

The authors thank to the supported of "Dalian construction planning project"; "Natural Science Foundation of Liaoning Province of Grant", No. 2011010158", "the Fundamental Research Funds for the Central Universities No. 3132013083".

REFERENCES

- [1] Bagal, M.V., Gogate, P.R. 2013. Degradation of 2,4-dinitrophenol using a combination of hydrodynamic cavitation, chemical and advanced oxidation processes, *Ultras on Sinochem*, 20, 1226-1235.
- [2] Paul, M.M.S., Aravind, U.K., Pramod, G., Aravindakumar, C.T. 2013. Oxidative degradation of fensulfothion by hydroxyl radical in aqueous medium, *Chemosphere*, 91, 295-301.
- [3] Chelliapan, S., Sallis, P.J. 2013. Removal of organic compound from pharmaceutical wastewater using advanced oxidation processes, *Journal of Scientific and Industrial Research*, 72, 248-254.
- [4] Nezamaddin, D., Soheil, A., Vahid, V., Mohammad, H.R. 2008. Electro-Fenton treatment of dye solution containing Orange II: Influence of operational parameters, *Journal of Electroanalytical Chemistry*, 615, 165-174.
- [5] Yusuf, Y., Koparal, A.S., Ülker, B.O. 2010. Treatment of petroleum refinery wastewater by electrochemical methods, *Desalination*, 258, 201-205.
- [6] Nidheesh, P.V., Gandhimathi, R. 2012. Trends in electro-Fenton process for water and wastewater treatment: An overview, *Desalination*, 299, 1-15.
- [7] Lunaa, M.D.G., Vecianab, M.L., Suc, C.C., Lu, M.C. 2012. Acetaminophen degradation by electro-Fenton and photo electro-Fenton using a double cathode electrochemical cell, *Journal of Hazardous Materials*, 217-218, 200-207.
- [8] Onofrio, S., Alessandro, G., Simona, S. 2013. Electro-generation of H₂O₂ and abatement of organic pollutant in water by an electro-Fenton process in a microfluidic reactor, *Electrochemistry Communications*, 26, 45-47.
- [9] Krzysztof, B. 2009. Fenton reaction controversy concerning the chemistry, *Ecological Chemistry and Engineering*, 16, 347-358.
- [10] Ignasi, S., Enric, B. 2012. Remediation of water pollution caused by pharmaceutical residues based on electrochemical separation and degradation technologies: A review, *Environment International*, 40, 212-229.
- [11] Nogueira, K.R.B., Teixeira, A.C.S.C., Nascimento, C.A.O., Guardani, R. 2008. Use of solar energy in the treatment of water contaminated with

phenol by photochemical process, *Brazilian Journal of Chemical Engineering*, 25, (4), 671-682.

[12] Tzedakis, T., Sayall, A., Clifton, M.J. 1989. The electrochemical regeneration of Fenton's reagent in the hydroxylation of aromatic substrates: batch and continuous processes, *Journal of Applied Electrochemistry*, 19, 911-921.

[13] Wang, C.T., Hu, J.L., Chou, W.L., Kuo, Y.M. 2008. Removal of color from real dyeing wastewater by Electro-Fenton technology using a three-dimensional graphite cathode, *Journal of Hazard Mater*, 152, (2), 601-606.

[14] Oturan, N., Oturan, M.A. 2005. Degradation of three pesticides used in viticulture by electrogenerated Fenton's reagent, *Agronomy for Sustainable Development*, 25, 267-270.

[15] El-Desoky, H.S., Ghoneim, M.M., El-sheikh, R., Zidan, N.M. 2010. Oxidation of Leyafix C A reactive azo-dyes in industrial wastewater of textile dyeing by electro-generated Fenton's reagent, *Journal of Hazard Mater*, 175, (1), 858-865.

