

The Effect of Different Parameters on Electrochemical Removal of Ampicillin Using New Generation Sn/Sb/Ni-Ti Anodes

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Abstract

In this study, it was aimed to investigate the feasibility of new generation and stable Sn/Sb/Ni-Ti anodes for treatment of wastewater containing Ampicillin (AMP) antibiotic by electrochemical oxidation processes. Ampicillin concentration was 0.05 g L⁻¹ in the aqueous solution. Residual ampicillin conc. was measured with Ultra-Performance Liquid Chromatography (UPLC) -Photodiode Array Detector (PDA). While, several evaluated such salt type and parameters were concentration, pH, current density, and anode-cathode distance; salt type and concentration was found as the most effective parameter for the removal of COD, TOC and AMP. Two different type of salts were used for this purpose; sodium chloride (NaCl) and potassium chloride (KCl). However, the removal efficiencies were found more higher in the presence of potassium chloride (KCl) when compared to sodium chloride (NaCl), generally. In the presence of KCl, ≥99% removal of COD and AMP after 60 min, pH 8, 50 mA/cm², 750 mg/L and 1cm distance between anodes. However, in the presence of sodium chloride (NaCl), complete removal of COD and AMP after 90 min, pH 8, 50 mA cm⁻², 2500 mg/L and 1cm distance between anodes.

Keywords: Sn/Sb/Ni-Ti anodes, electrochemical oxidation, ampicillin, wastewater treatment, anode production

1. Introduction

In the last decades, the attention for the novel emerging contaminants such as drugs, pesticides, and endocrine

disrupting chemicals (EDC) has been increasing in the environment (1-2). Pharmaceuticals (analgesic, antidepressants, antibiotics, antidiabetics, contraceptives, growth regulators, painkillers, impotence drugs, and tranquilizers) are widely used to treat or prevent human and animal diseases; however, over the last 20 years, they have been defined as micro-contaminants in soil and water ecosystem (3-4).

Among all these pharmaceuticals, antibiotics possess significant/influential place because of its elevated consumption rates in both veterinary and human medicine (5). The development of antibiotic-resistant bacteria is the most important problem that (may) occur due to unconscious/increased use of antibiotics (6). Antibiotics are either naturally occurring or synthetic complex molecular chemicals. They can be categorized into various classes such as tetracyclines, ß-lactams, macrolides, quinolones, and sulfonamides according to their action mechanism: cell membrane modification, protein synthesis inhibition, inhibition of cell wall synthesis, synthesis of nucleic acids inhibition and metabolic or anti-competitive antagonism (7).

Antibiotic-resistant bacteria are one of the critical environmental problems that can be developed even if antibiotics present at low concentration in the environment. Ampicillin ($\underline{C_{16}H_{19}N_3O_4S}$) is a beta-lactam group of antibiotics which is broadly used in medicine because of its destructive potential of the cell wall of both gram-positive and gram-negative bacteria (8). The main

sources of these antibiotics in the environment are wastewater from hospitals, pharmaceutical industries and veterinary medicine (9).

Introduction of antibiotics to the environment depends on several factors such as; dose and amount used, the frequency of excretion from the body, and the tendency to retain the solids and the metabolic ability of microorganisms in the wastewater treatment plant (10). Antibiotics have the ability only to be metabolized partially in a human body, therefore, a great amount is subtracted/ejected unchanged or as active metabolites by urine and feces: (11-12-13) as a result, human antibiotics reach wastewater treatment plant.

Conventional treatment methods (physical, chemical, biological) are inefficient in the removal of these endocrine disrupting antibiotics. (14-15-16). Conventional disinfection processes, such as chlorination and UV applications, are also not effective in the controlling of resistance developing bacteria. For these reasons, various advanced treatment methods such as carbon adsorption, advanced oxidation processes (AOP),

References

1. L. T. Benjamin, D. W. Hawkera, J. F. Muller, L. A. Tremblay. H. F. Chapman, Water Research 42 (2008) 404.

2. S. Imai, A. Shiraishi, K. Gamo, I. Watanabe, H. Okuhata, H. Miyasaka, K. Ikeda, T. Bamba, K. Hirata, J. Bioscience and Bioengineering 103 (2007) 420.

3. A.B.A. Boxall, The environmental side effects of medication, EMBO Rep. 5 (2004) 1110–1116.

4. I.T. Carvalho, L. Santos, Antibiotics in the aquatic environments: a review of the European scenario, Environ. Int. 94 (2016) 736–757.

5. M.V. Walter, J.W. Vennes, Appl. Environ. Microbiol. 50 (1985) 930–933.

6. H. Kanay, Jpn. J. Vet. Sci. 45 (1983) 171-178.

7. K. Kümmerer, Antibiotics in the aquatic environ-ment - a review - part I. Chemosphere 75 (2009) 417–434.

8. O. Rozas, D.M. Contreras, M.A. Mondacab, M. Pérez-Moyac, H.D. Mansilla, Experimental design of Fenton and photo-Fenton reactions for the treatment of ampicillin solutions. J. Hazard Mater. 177 (2010) 1025–1030.

9. T. Kosjek, E. Heath, and B. Kompare, Removal of pharmaceutical residues in a pilot wastewater treatment plant. Anal. Bioanal. Chem. 387 (2007) 1379-1387.

10. C.G. Daughton, T.A Ternes. Pharmaceuticals and personal care products in the environment: agents of subtle change. Environ Health Perspect. 107(6) (1999) 907–938.

11. Joint Interagency Antimicrobial Consumption and Resistance Analysis (JIACRA) Report. (2015) 114 pp. http://www.efsa.europa.eu/sites/default/files/scientific_output/ files/main_documents/4006.pdf (accessed January 2017).

12. N. Kemper, Veterinary antibiotics in the aquatic and terrestrial environment, Ecol. Indic. 8 (2008) 1–13.

13. J.N. Louvet, C. Giammarino, O. Potier, M.N. Pons, Adverse effects of erythromycin on the structure and chemistry of activated sludge, Environ. Pollut. 158 (2010) 688–693.

membrane processes are proposed as an alternative and innovative technological solution in the removal of antibiotics (17).

2. Material and Methods

2.1. Chemicals, Solvents and materials

The main material was 2.5 cm x 2.5 cm sized titanium screen substrate ((3Ti7-077FA mesh, Dexmet, USA). Dexmet, USA). As a cathode, a 5x5 cm platinized titanium electrode (NRK Electrochem) was provided and used (DuPont Corp., USA). NaCl (Merc), KCl (Merck), SnCl₄.5H₂O (Merck), Sb₂O₃ (Emsure), NiO (Alfa Aeser), HCl (Merck), H₂SO₄ (Merck), ethanol (Merck), methanol (Merck), formic acid (Merck), oxalic acid (Merck) were used directly without any extra purification. All solutions were prepared with Millipore Milli-Q (18MQ cm) ultrapure water. Ampicillin antibiotic was provided from Sigma-Aldrich. All experiments were performed with synthetic antibiotic solution which has been prepared at room temperature, 20-25°C. pH measurements were performed with laboratory pH meter Cyber Scan 500 (Eutech instruments). A power supply DC-Power Supply.

14. T.A. Ternes, Occurrence of Drugs in German Sevage Treatment Plants and Rivers. Water Res. 32 (11) (1998) 3245-3260.

15. K. Kümmerer, Pharmaceuticals in the Environ-ment: Sources, Fate, Effects and Risks, Second Ed. (2000) Springer, Germany.

16. T. Heberer, Occurrence, fate and Removal of Pharmaceuticals Residues on The Aquatic Environment: A Review of Recent Research Data. Toxicol. Lett. 131 (1/2) (2002) 5-17.

17. K. Kosutic, D. Dolar, D. Asperger, B. Kunst, Removal of Antibiotics from a model wastewaters by RO/NF membranes. Sep. Purif.Technol. 53 (2007) 244-249.