INTERNATIONAL JOURNAL OF INNOVATIVE RESEARCH & DEVELOPMENT

ISSN: 2278 – 0211 (Online)

Effect Ofantibiotics In Wastewater Treatment

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Abstract:

Antibiotics are emerging contaminants in the aquatic environment because they have adverse effects on aquatic life and humans. The problem created by the presence of antibiotics at low concentrations in the environment is the development of antibiotic resistant bacteria. This paper aims at giving a review of antibiotics wastewater treatment. Different advanced oxidation processes (AOPs), such as Fenton, photo-Fenton and ozonation have been applied as pretreatment of antibiotics wastewater. AOP pretreatment can reduce organic concentration and improve biodegradability. Biological treatment, both aerobic and anaerobic systems, have also been used for treatment of antibiotics wastewater. The treatment of antibiotic wastewater by biological systems is influenced by type and concentration of the antibiotic (active substance) in wastewater. Application of other AOPs pretreatment and biological treatment for different types of antibiotics wastewater is a challenging area of research.

Keywords: *Advanced oxidation processes (AOPs), antibiotics wastewater, biological treatment.*

1.Introduction

From an environmental engineering point of view, pharmaceuticals, including antibiotics, are a new group of man-made chemicals of concern entering our environment at concentrations such that the health effects are unknown. Research has begun to determine the concentrations of antibiotics in the environment, and from this information, the health effects to humans and animals may be estimated by toxicologists. An additional problem that may be created by the presence of antibiotics at low concentrations in the environment is the development of antibiotic resistant bacteria. In recent years, the incidence of antibiotic resistant bacteria has increased and many people believe the increase is due to the use of antibiotics (Walter et al. 1985). The presence of antibiotics can result in selective pressure that favors organisms that possess genes coding for antibiotic resistance. This may pose a serious threat to public health in that more and more infections may no longer be treatable with known antibiotics (Hirsch et al. 1999). In the event that antibiotic resistance is spread from nonpathogenic to pathogenic bacteria, epidemics may result. In fact, bacteria have been observed to transfer their resistance in laboratory settings as well as in the natural environment (Kanay,1983). Pharmaceutical compounds are emerging contaminants in aquatic environments because they are introduced in larger amount and they are bioactive, polar and persistent which may cause adverse effects in aquatic life and humans. It is important to give economical technical solution for treatment of antibiotics wastewater which contains non-biodegradable organics. For these reasons, the search for solutions of this problem is an interesting research topic. The Advanced Oxidation Processes (AOPs) appear as interesting tools, in comparison with the well-established techniques like activated carbon adsorption, air stripping, reverse osmosis, combustion, etc. Indeed, many of these only transfer the pollutants from one phase to another without destroying them.

2.Background

2.1.Pharmaceutical Wastewater

In the 1970s, for the first time, pharmaceuticals in the environment (namely hormones) were the subject of scientific interest and public awareness (Tabak et al. 1970; Norpoth et al. 1973). The conclusion most often reached was that the hormones are not easily biodegraded. During the 1980s, there was only little interest in this topic. Other substances of environmental relevance such as heavy metals, polycyclic aromatic hydrocarbons, or chlorinated dioxins and furans as well as pesticides or detergents were investigated very extensively during this period. Since the middle of the 1990s, awareness of pharmaceuticals in the environment is growing. Parallel to this, discussion of endocrine disrupting (or endocrine modulating) substances (EDS) and non-hormone pharmaceuticals, such as lipid lowering agents and others, came into focus (Stan et al. 1992).EDS have been detected also in sewage, the effluent of sewage treatment plants, surface water, manure and soil since the 1980s. Most of those investigated so far were not easily biodegradable in test systems and are only slowly eliminated in the environment.

2.2.Antibiotics

Humans and animals serve as hosts to disease-causing organisms, such as bacteria, viruses, fungi, protozoa, and helminthes (worms). Before the development of antibiotics, which are chemical agents that fight and destroy these pathogens, simple infections often resulted in death. Over time, compounds were observed to posses antimicrobial properties and cure infections. Compounds that have been historically used to fight infections include mercury, silver, and cyanide. In the past, sore throat lozenges contained mercury. Silver was used to treat syphilis. Despite their effectiveness, the problem of using these compounds to treat infections is that they are toxic to the host. Silver and mercury are considered heavy metals, which are toxic to humans at low concentrations. Thus, it became desirable to find compounds with antimicrobial properties that were not lethal to the host. Alexander Fleming made the paramount discovery of antibiotics in 1928. He was trying to isolate Staphylococcus aureus and one dish had become contaminated. Fleming noticed that bacteria did not grow near the invading substance that he later identified as a common mold of the Penicilliumgenus. After culturing the mold and obtaining a tiny quantity of excreted material, Fleming demonstrated the antimicrobial properties of the product now known as penicillin (Shuler et al. 2002). From Fleming's discovery, a number of natural products have been discovered that are suitable for therapy. For example, secretions from soil bacteria and fungi have demonstrated antimicrobial activity. To improve the efficacy of production, reduce the side effects, prevent development of resistance to the antibiotic by the targeted bacteria, and expand the range of bacteria susceptible to the antibiotic, alterations have been made to the natural compounds isolated from bacteria and fungi

(Kimball, 2001). For example, broad-spectrum antibiotics are modified natural antibiotics that target both Gram-negative and Gram-positive bacteria.

2.3.Strategies For The Choice Of Appropriate Treatment Processes

1. Possibility of recycling and reuse treated wastewater (pollution prevention).
2. If recycling of wastewater constituents is not suitable for any reason, biological processes are preferred because of low costs compared to other processes.
3. If the wastewater contains non-biodegradable organic pollutants, microorganisms cannot degrade the main part of the organics, biological processes are not suitable.
4. Advanced oxidation processes (AOPs) are suitable alternatives for the treatment of the wastewater containing toxic or non-degradable pollutants.

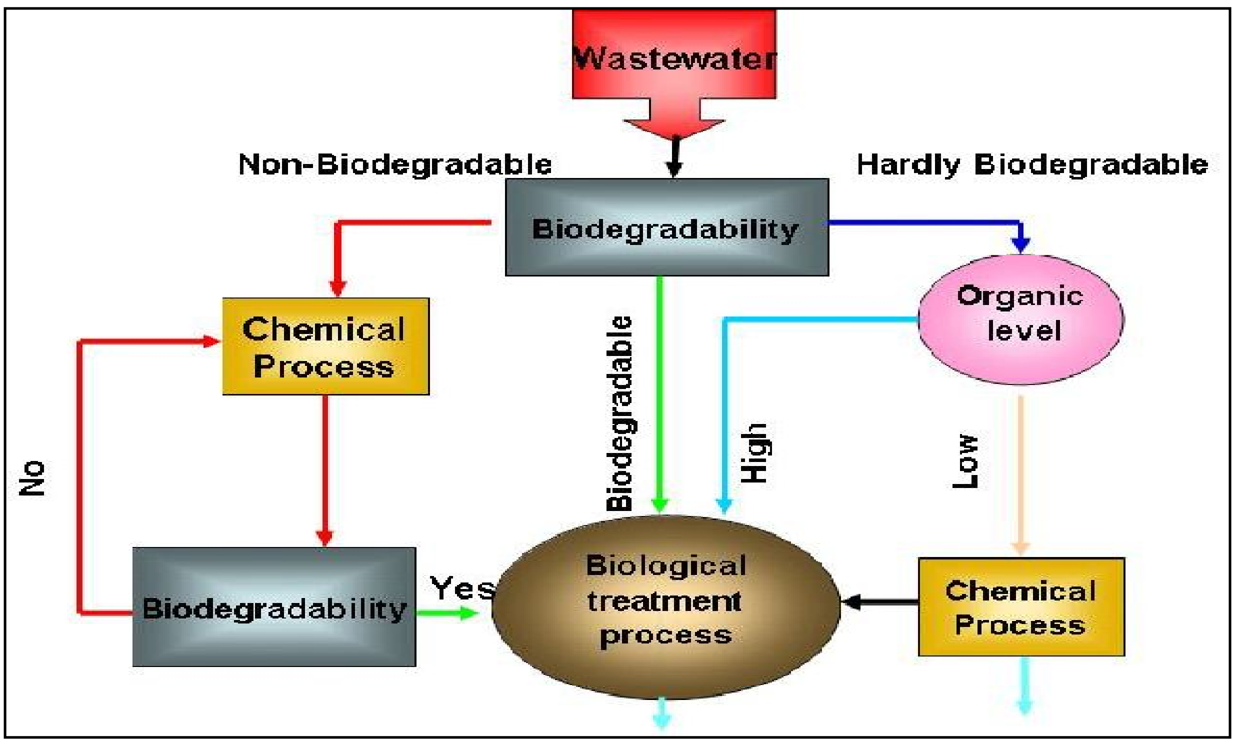

Figure 1: Strategies for the choice of appropriate treatment processes

2.4.Treatment of antibiotics wastewater(Experimental)

Advanced oxidation processes (AOPs)The Fenton's reagent pre-oxidation of an extremely polluted pharmaceutical wastewater (chemical oxygen demand (COD) value of 362,000 mg/l) was studied. The parameters influencing the COD removal of the wastewater, namely temperature, ferrous ion and hydrogen peroxide concentrations were optimized to achieve a COD global reduction of 56.4%. The optimal values of hydrogen peroxide and ferrous ion concentration were 3 and 0.3 M, respectively, whereas temperature only showed a mild positive effect on COD removal. In addition, during the first 10 min of Fenton's reaction, more than 90% of the total COD removal was achieved. Fenton's reaction proved to be a feasible technique for the pre-oxidation of the wastewater under study, and can be considered a suitable pre-treatment for this type of wastewaters (Martinez et al. 2003).

The applicability of Fenton's oxidation to improve the biodegradability of a pharmaceutical wastewater to be treated biologically was studied. Optimum pH was determined as 3.5 and 7.0 for the first (oxidation) and second stage (coagulation) of the Fenton process, respectively. For all chemicals, COD removal efficiency was highest when the molar ratio of H2O2/Fe2+ was 150–250 and at H2O2/Fe2+ ratio of 155, 0.3 M H2O2 and 0.002 M Fe2+, provided 45–65% COD removal (Tekin et al. 2006). Amoxicillin antibiotic wastewater which has high TOC, low biodegradability, presence of organic solvents and dissolved salts was treated using a combined process of extraction (EX), Fenton oxidation (FO) and a two-stage reverse osmosis (RO) (Zhang et al. 2006). EX and FO were performed in a serial approach to function as the pretreatments to remove the solvents and other organics before RO treatment. Under the appropriate conditions, TOC was reduced by 50.6% and 37.8% in the EX and FO units, respectively. TOC can be further reduced by 10.1% and 1.2% in the first and second RO 4processes, respectively. There was consequently an overall TOC removal efficiency of 99.7% by the whole process of EX–FO–RO–RO. Dissolved salts were also greatly removed during the two-stage RO process. The pre-treatment of penicillin wastewater using different advanced oxidation processes (AOPs) O3/OH−, H2O2/UV, Fe2+/H2O2 ,Fe3+/H2O2 ,Fe2+/H2O2/UV and Fe3+/H2O2/UV)was studied (Arslan-Alaton et al., 2004). the oxidative formulation effluent. (average COD0 = 1395 mg/L, TOC = 920 mg/L, BOD5 ≈ 0 mg/L). Alkaline ozonation and the photo-Fenton's reagents both appeared to be the most promising AOPs in terms of COD (49–66%) and TOC (42–52%) abatement rates, whereas the BOD5 of the originally non-biodegradable effluent could only be improved to a value of 100 mg/L with O3 at pH 3 treatment (BOD5/COD 0.08). Degradation of Procaine Penicillin G (PPG) formulation effluent (COD = 600 mg/l; BOD5 =53 mg/l; TOC = 450 mg/l) by Fenton-like (Fe3+ /H2O2) and UV-A light assisted Fenton-like (Fe^{3+} /H_2O_2 /UV-A) processes was investigated (Arslan-Alaton et al., 2004). At optimum reaction conditions ([Fe^{3+}] = 1.5 mM and [H_2O_2] = 25 mM, pH = 3), 56% COD and 42% TOC removal were achieved by the photo-Fenton-like process after 30 min treatment time, whereas the removal efficiencies of the dark Fenton-like process were limited to 44% COD and 35% TOC removal for the same treatment period. The BOD_5/COD ratio increased from 0.10 to 0.45 and 0.10 to only 0.24 after application of the photo-Fenton-like and Fenton-like processes, respectively. The degredation of diclofenac by advanced oxidation processes, ozonation and H_2O_2/UV was investigated (Vogna et al., 2004). Both ozonation and H_2O_2/UV systems proved to be effective in inducing diclofenac degradation, ensuring a complete conversion of the chlorine into chloride ions and degrees of mineralization of 32% for ozonation and 39% for H_2O_2/UV after a 90 min treatment. The abatement of 200 mg/L sulfamethoxazole (SMX) solution by means of photo-Fenton process was studied (Gonz´alez. 2007). Biodegradability of the treated solutions was evaluated by (BOD_5/COD) ratio and toxicity by Microtox and inhibition tests. Experiments with different initial concentration of H_2O_2 were carried out. The initial amount of Fe^{2+} and pH of the solution were set at 10 mg/L and 2.8, respectively. The temperature of the reactor was kept constant in all the experiments (25±0.8 ◦C). Photo-Fenton process is thought to be a successful treatment step to improve the biodegradability of wastewater containing SMX. The complete antibiotic removal was achieved for a H_2O_2 dose of over 300 mg/L.

Biodegradability (BOD_5/COD) rose from zero (SMX solution) to values higher than 0.3 (treated solutions). Toxicity and inhibition tests pointed out in the same direction: oxidized intermediates for initial H_2O_2 dose of over 300 mg/L showed no toxicity effects on pure bacteria and no inhibition on activated sludge activity. Biological Treatment of antibiotics wastewater The anaerobic and aerobic treatment of high-strength pharmaceutical wastewater including antibiotics was evaluated (Zhou et al. 2006). The antibiotics ampicillin and aureomycin, with influent concentrations of 3.2 and 1.0 mg/L, respectively, could be partially degraded in the anaerobic baffled reactor: ampicillin and aureomycin removal efficiencies were 16.4 and 25.9% with an HRT of 1.25 day, and 42.1 and 31.3% with HRT of 2.5 day, respectively. Although effective in COD removal, the biofilm airlift suspension reactor did not display significant antibiotic removal, and the removal efficiencies of the two antibiotics were less than 10%.The performance of an up-flow anaerobic stage reactor (UASR) treating pharmaceutical wastewater containing macrolide antibiotics was investigated (Chelliapanel al. 2006)).

UASR was developed with an active reactor volume of 11 L being divided into four 2.75 L stages. Each stage of the reactor was an up-flow sludge blanket reactor and had a 3-phase separator baffle to retain biomass. The reactor was fed with real pharmaceutical wastewater containing Tylosin and Avilamycin antibiotics and operated with step-wise increase in the 5reactor organic loading rate (OLR) from 0.43 to 3.73 kg (COD) m_3 d_1, and then reduced to 1.86, over 279 days. At a total hydraulic retention time of 4 d and OLR of 1.86 kg CODm-3 d-1, COD reduction was 70–75%. Average 95% Tylosin reduction was achieved in the UASR, indicating that this antibiotic could be degraded efficiently in the anaerobic reactor system. The influence of elevated Tylosin concentrations on the UASR process performance was studied using additions of Tylosin phosphate concentrate. Results showed similar efficiency for COD removal when Tylosin was present at concentrations ranging from 0 to 400 mg/L (mean removal over this range was 93%), however, at Tylosin concentrations of 600 and 800 mg/L there was a slight decline in treatment efficiency at 85% and 75% removal, respectively. The treatability of antibiotic sulfamerazine was studied using the lab-scale continuously feed upflow anaerobic sludge blanket (UASB) reactor andcontinuously stirred tank reactor (CSTR) system (Sponza et al. 2007). The BOD5/COD ratio was used to assess the approximate biodegradability. The initial sulfamerazine concentration was 10 mg/l the methane percentage reached around 76% while it was measured at 60% at a sulfamerazine concentration of 90 mg/l. The total (overall) COD removal efficiency of the sequential UASB + STR treatment system was determined as 97%. The results of this study indicated that the system exhibited a good removal performance for sulfamerazine. Coupled chemical-biological treatment of antibiotics wastewater Coupled chemical-biological treatment of antibiotics wastewater was studied (Arslan-Alaton et al., 2004). According to the experimental results, the overall Chemical Oxygen Demand (COD) removal efficiency varied between 10 and 56% for ozonation and 30% (no H2O2) and 83% (20 mM H2O2) for the O3+H2O2 process. The addition of H2O2 improved the COD removal rates considerably even at the lowest studied H2O2 concentration. An optimum H2O2 concentration of 20 mM existed at which the highest COD removal efficiency and abatement kinetics were obtained. An ozone input of 800 mg/l in 20 min was sufficient to achieve the highest BOD5/COD (biodegradability) ratio (0.45) and BOD5 value (109 mg/l) for the pre-treated penicillin formulation effluent. COD removal efficiencies of the activated sludge process were 71, 81 and 72% for pharmaceutical wastewater containing synthetic domestic wastewater mixed with either raw, ozonated or perozonated formulation effluent, respectively. The ultimate COD value obtained after 24-h biotreatment of the synthetic domestic wastewater + pre-ozonated formulation effluent mixture was around 100 mg/l instead of 180 mg/l which was the final COD obtained for the wastewater mixture containing raw formulation effluent, indicating that pre-ozonation at least partially removed the non-biodegradable COD fraction of the formulation effluent.

2.5.Conclusion

This paper gives a review of antibiotics wastewater treatment. Various advanced oxidation processes (AOPs) were applied as pretreatment for antibiotics wastewater. AOP pretreatment reduced the organic content and improved biodegradability. Ozonation pretreatment resulted in TOC removal of 49% for penicillin wastewater and 66% for diclofenac wastewater. Fenton pretreatment of amoxicillin wastewater resulted in 37.8% TOC removal and photo-Fenton pretreatment of sulfamethoxazole wastewater increased the BOD5/COD ratio from 0 to 0.30. Biological treatment can be used as sole treatment if the concentration of antibiotics in the wastewater is low. Biological treatment degraded only 16.4% and 25.9% of ampicillin and aureomycine, respectively in anaerobic reactor and less than 10% for both the antibiotics in 6 aerobic reactor. Degradation of antibiotics by biological treatment is influenced by type and concentration of the antibiotic (active substance) in wastewater. Application of other AOPs pretreatment and biological treatment for different types of antibiotics wastewater is a challenging area of research.

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