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

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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 future [5]. 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 region [6]. 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 environments [7]. Among the different types of antibiotics, the beta-lactam family is the most widely used drug [8]. Deaths of aquatic organisms such as fish [9], algae [10], and the development of drug resistance of microorganisms are some of the adverse effects of uncontrolled discharge of wastewater containing antibiotics to the environment [11]. 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.

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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 compounds [14]. Produced OH˚ can non-selectively oxidize and mineralize the persistent organic pollutants [4]. 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 environments [15]. Most of electrochemical processes for wastewater treatment are divided into two categories: electrocoagulation and electrolysis [16]. 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 process [17]. 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 1 [18]. Surface-produced hydroxyl radicals M(OH˚) are highly non-selective oxidizing agents which are capable to destroy organic pollutant completely and mineralized them [19].

(1) $M + H_2O \rightarrow M(^{\circ}OH) + H^+ + e^-$

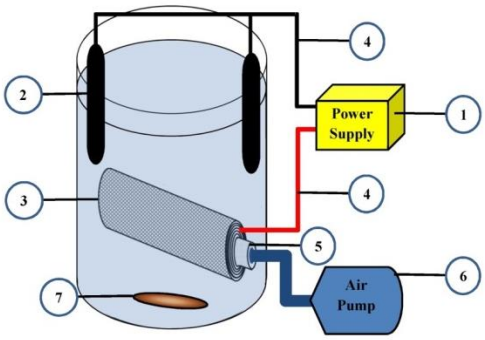
Considering AOP procedures, the Fenton-based process because of high efficiency, ease of application, and low cost is one of the most commonly used procedures [20]. This process is one of the advanced oxidation processes which can effectively destroy persistent aromatic compounds through the production of potent free radicals. Fenton process is accomplished by the reaction between iron II and hydrogen peroxide-based on formula 2 and 3 [21]:

(2) $Fe(II) + H_2O_2 \rightarrow Fe(III) + OH^- + {}^{\circ}OH$

(3) $Fe(II) + {}^{\circ}OH \rightarrow Fe(III) + OH^-$

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 biodegradation [19]. The efficiency of this process relays 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 production [19]. 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 conditions[23].



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]

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The application of porous carbon electrodes (e.g. graphite and carbon felts [24], carbon sponges [25], activated carbon fibers [12] and carbon nanotubes [26] 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.

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