



Application of a new sandwich of granular activated and fiber carbon as cathode in the electrochemical advanced oxidation treatment of pharmaceutical effluents



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ABSTRACT

Society technological advances and environmental requirements claim for the development of highly effective, low cost and environmental friendly technologies able to degrade emerging organic pollutants widely used such as pharmaceuticals. The development of electrochemical advanced oxidation processes may increase the efficiency of the traditional treatments used on the remediation of polluted water. Therefore, this work has been devoted to the design of a cathode that permits to enhance the generation of hydroxyl radicals on the system. The best results were obtained when a cathode made with granular activated carbon and carbon fiber (SGAFC) was used. After determining the efficiency of this cathode on the decoloration of two model dyes, its application to the chloramphenicol antibiotic reduction was evaluated, obtaining near complete removal after 120 min. In addition, the treatment on successive batches and different continuous electrochemical cell configurations has emphasized the ability of this new cathode to be used on different water treatment facilities.

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1. Introduction

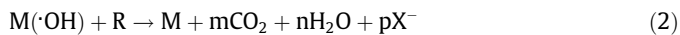
Human activities release large volumes of polluted effluents daily in the aquatic environments. Given the potential toxicity of the pollutants and their metabolites even at low concentrations, it becomes necessary to develop a proper and efficient treatment to avoid environmental risks and potential hazards for living beings. During the last years, a particular concern about the presence in the water of “emerging organic contaminants”, including pharmaceuticals, personal care products, “life-style” and selected industrial compounds have emerged. Although the effect of these pollutants is still unknown, it has been documented that they have adverse effects on human health and aquatic ecosystems. The presence of different pollutants and their metabolites in rivers, ground-water and drinking water represents a potential hazard. After administration, humans and animals excrete a high percentage of non-metabolized antibiotics. As consequence an active fraction of these pollutants persists in the aquatic system, ending up in the conventional sewage treatment plants [1,2]. In addition to their harmful effect on biota, the recalcitrant characteristic of these pollutants is an impediment to their elimination by conventional methods [3].

Several researchers have focused on the development of new technologies capable to attain the complete destruction of these contaminants [4]. Advanced oxidation processes (AOPs) are technologies based on the *in situ* generation of hydroxyl radicals ($\cdot\text{OH}$), a highly oxidant and low selective agent able to react with persistent organic pollutants and oxidize them until their complete mineralization [5]. The use of different AOPs involving Fenton processes, photocatalysis, $\text{UV-H}_2\text{O}_2$, ozone, ozone-UV, plasma based processes and sonolysis as promising technologies to deal with some environmental problems has been intensely investigated worldwide by numerous researchers [6–10]. Over the past years, Electrochemical Advanced Oxidation Processes (EAOPs) have reached an excellent state of development [11,12]. They employ the electrical current to generate $\cdot\text{OH}$, avoiding the use of chemicals and thus, reducing the potential hazards related to their transport, storage and handling during the process. Among the different EAOPs, electro-oxidation or anodic oxidation (AO) is the most popular electrochemical technology used for the treatment of wastewater [13]. On this process, the degradation mechanism is based on the oxidation of the organics by the $\cdot\text{OH}$, directly generated at the anode surface (M) by the oxidation of water (Eq. (1)) [14]. In addition, an interesting study performed by Marselli et al. [15] proposed the degradation mechanism of the organic pollutants, which is helped by the heterogeneous formation of hydroxyl

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radicals by electrolysis of water and the oxidation of the organic matter (Eq. (2)).



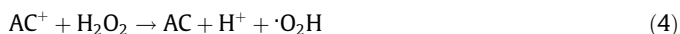
where M represents the anode material, M(\cdot OH) are the adsorbed radicals on the anode, R is the organic matter and X are the inorganic ions [16].

On this way, traditional anodes, such as Pt, IrO₂, RuO₂, allow the generation of low amounts of oxidant species. Conversely, high oxygen overvoltage anodes such as Boron-Doped Diamond (BDD), recently appeared as promising anode materials. Indeed, this BDD anode has demonstrated to significantly increase the generation of hydroxyl radicals, enhancing the removal rate of the pollutants achieved by anodic oxidation [13,15–18]. Besides, several research studies have demonstrated the high oxidizing power of \cdot OH physisorbed on BDD, which leads to a great oxidation/mineralization efficiency. For this reason, BDD is a commonly used anode material [8,15,18,19].

Since the generation of \cdot OH is highly dependent on the electrode materials, their optimization and configuration are decisive to increase the concentration of this reagent on solution and thus the effectiveness of the oxidative treatment [20].

Thus, in order to improve the efficiency of the AO process it is necessary to increase the \cdot OH generated at the bulk. For this proposal, in this study the use of a manufactured cathode with sandwich configuration formed by the combination of carbon fiber and activated carbon was proposed.

It is well-known that granular activated carbon has adsorption properties such as a large porous volume, and a vast surface area [21,22]. Furthermore, according to Lücking et al. [23], H₂O₂ in presence of activated carbon was able to oxidize 4-chlorophenol. More recently, these results were confirmed by Santos et al. [24], who reported similar results on the removal of dyes by the synergistic effect between H₂O₂ and a modified activated carbon. Bansal et al. [25] reported the catalytic decomposition of H₂O₂ by activated carbon, which involves directly the exchange of the hydroxyl group on activated carbon surface with a hydrogen peroxide anion (\cdot OOH). Additionally, oxygen is produced and regenerated on the activated carbon surface when the peroxide anion latter generated decomposes another H₂O₂ molecule. Besides, regarding the Haber–Weiss mechanism, the activated carbon (AC) can act as an efficient electron-transfer catalyst thus, involving the reduction (AC) and oxidation (AC⁺) of the catalyst (Eqs. (3) and (4)).



In this sense, the presence of granular activated carbon in an EAOP treatment enhances the removal of the pollutants, given the combined action between oxidation and activated carbon adsorption. Recently, Kim et al. [26] determined the high efficiency of a combined adsorption–Fenton oxidation on granular activated carbon showed effective removal of bisphenol A, demonstrating that granular activated carbon is an excellent catalyst/adsorbent.

During EAOP, H₂O₂ is *in situ* generated on the cathode surface in presence of oxygen (Eq. (5)). Thus, the continuous aeration and the use of a proper cathode electrode provide to the system the H₂O₂ required to react with activated carbon.



Recently, several authors have focused their studies in the evaluation of different carbonaceous materials used as electrodes to produce H₂O₂, for example graphite [27], vitreous carbon [28],

carbon fiber [29], carbon sponge [30] or gas-diffusion [31]. One of the main limitations to generate hydrogen peroxide is the poor oxygen solubility, which restrict the mass transfer. On the other hand, the small surface area of gas-diffusion cathodes, together with their high cost prevent their application to large volumes of wastewater [31]. These drawbacks can be effectively avoided using inexpensive carbon fiber with higher surface area which increases the hydrogen peroxide production.

The aim of this study was to develop a sandwich cathode formed by the combination of carbon fiber and granular activated carbon (SGAFC) and determine its ability to enhance the degradation of recalcitrant compounds in the electrochemical advanced oxidation of several pollutants. Initially, SGAFC cathode configuration was optimized by decoloration of two dyes (Reactive Black 5 and Rhodamine B) and after that the removal of an antibiotic such as chloramphenicol was carried out. Then, the performance of different electrochemical cell configurations using SGAFC as cathode and the possibility to operate in continuous mode was evaluated and discussed.

2. Materials and methods

2.1. Materials

Three model pollutants, Reactive Black 5 (RB5), Rhodamine B (RhB) and chloramphenicol (CAP) were provided by Sigma–Aldrich (Barcelona, Spain) and used without further purification. All the solutions and standards for analytical measurements were prepared in distilled water. Their chemical structure and properties are listed in Table 1.

2.2. Sandwich of granular activated and fiber carbon (SGAFC) cathode

The sandwich electrode used as cathode was composed by two sheets of carbon fiber (Advanced Composites Group UK) linked with a nylon thread to entrap granular activated carbon inside (Granulated n°2 QP Panreac Spain). The different geometric characteristics of the electrodes, based on sandwich electrode concept, are described in the following sections.

2.3. Electrochemical cell configurations

2.3.1. Electrochemical stirred cell (ESC)

The electrochemical process was carried out in a cylindrical glass cell with a working volume of 0.15 L (Fig. 1a). The electric field was applied by a SGAFC (11 cm² of area containing 2.65 g of granular activated carbon) and a BDD anode of 11 cm² (DIACHEM®, Germany). The electrodes were placed opposite to each other at 1 cm above the bottom of the cell with an electrode gap of 6 cm. A constant potential drop of 5 V was applied with a power supply (HP model 3662). Current intensity was continuously monitored along the process with a multimeter (Fluke 175). Continuous saturation of air at atmospheric pressure was ensured bubbling 1 L/min of compressed air near the cathode.

For each experiment, 0.15 L of the wastewater sample at the desired concentration of organic pollutant and Na₂SO₄ (used as electrolyte 0.01 M), was continuously homogenized by means of a magnetic stirrer in order to avoid concentration gradients. The pH was stabilized at 2 by the addition of H₂SO₄ or NaOH provided by Sigma–Aldrich (Barcelona, Spain). The temperature of the solution was kept constant at ambient temperature throughout all the experiments.

2.3.2. Electrochemical plug flow cell (EPFC)

A cylindrical glass cell (internal diameter of 4 cm) with a working volume of 0.162 L was used to design an EPFC to operate in

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