Efficiency of the Catalytic Ozonation with Fe Foam Catalyst for Naproxen degradation

ORAL
Ph.D. Student: Y
Journal: CEJ, ESPR

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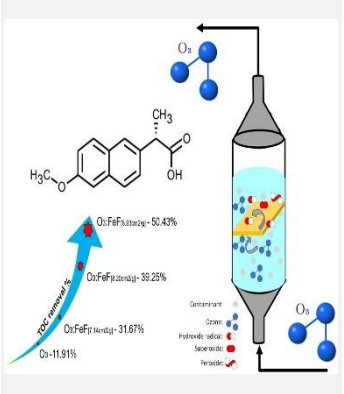

In the present study, the degradation of naproxen (NP- 20 mg L^{-1}) was carried out by ozone (11 mg L^{-1}) in the presence of the iron foam (FeF) catalyst of the different transversal seccion area and massa (10.0cm^2/1.1232g, 5.0 cm^2/0.6088g, 2.5 cm^2/0.3325g) during 60 and 120 min. On the base of the HPLC result, the lower concentrations of the NP by-products was observed in the presence of the catalyst with the higher size (10.0cm^2/1.1232g) during 2 hrs of treatment, marking the effect of catalyst size. The incease of the mineralization degree to 50.43% for catalytic ozonation compared with conventional ozonation (11.91%) confirmed catalytic activity of the FeF.

Introduction

The most common sites for pharmaceutical waste are hospitals, pharmaceutical industries, and some domestic areas, which secrete drugs and other wastes into wastewater, groundwater, and surface water [1]. The progressive accumulation of these organic compounds, among which are emerging compounds such as pharmaceuticals, hormones, personal care, and hygiene products, lack regulations and final disposal applications for several years. In the case of drugs, they are persistent, especially antibiotics, which are very difficult to biodegrade using conventional treatment methods [2]. Their presence has been quantified in effluents of wastewater plants from different hospitals, principally with a high concentration of pharmaceutical compounds. In the countries of Latin America for example Brazil, Venezuela, Argentina, Colombia, and Ecuador have been detected the presence of the naproxen was higher than the Predicted No Effect Concentration for health determined a few years ago through the United States Environmental Protection Agency and Committee for Medicinal Products for Human Use (0.15ppm).

The heterogeneous catalytic ozonation in the presence of the catalists in the form of films or foam is the preferred method, because it is simple to separate the catalyst after the reaction finish, prevents secondary pollution by not adding metal ions, and the possible reuse of the catalysts. The objective of the present study was the determination of the efficiency of the catalytic ozonation in the presence of the iron foam (FeF) catalyst of three different transversal seccion area and massa (10.0cm^2/1.1232g, 5.0 cm^2/0.6088g, 2.5 cm^2/0.3325g) for the naproxen (NP) degradation in water.

Material and Methods

Catalysts pretreatment

Before the use of the FeF catalyst, the cleaning was carried out, which include the consecutive use of different solution and disolvent. First, the foam put in 1.0 mol/L HCl solution, then in ethanol, acetone, and distilled water. After each etape an ultrasonic tretament during 15 min. Then, the foam was dried at 60 °C for 12 h [3].

Ozonation procedure

The ozonation system consists a glass reactor (640 mL), which is loaded with a model solution of the NP with the concentration of 20 mg L^{-1}. Ozone (O_3) was produced from extra dry oxygen employing a corona discharge type generator (Longevity Resources), and then it was introduced into the reactor through a plate diffuser located in the lower part of it. Residual ozone in the output of the reactor was mesaured by an ozone sensor (BMT 964 BT) to monitor its concentration in the gas phase. The O_3/O_2 gas flow of 0.5L min^{-1}, with an ozone concentration of 11 mg L^{-1} for both processes conventional and catalytic. The monitoring of the NP decomposition, as well as the behavior of ozonation intermadiates were carried out by means the high-performance liquid chromatography (HPLC), using Flexar Series 200-Perkin Elmery, with a diode array detector, Platinum C18 column (150 x4.6 mm), mobile phase: ACN: acidic H2O (40:60), flow: 0.4 µL/min, and injection volume of 20 µL.

Results and Discussion

The initial compound decomposed quite rapidly in both ozonations conventional and catalytic with a significant difference. However, the NP elimination from water may be realized by the three methods: an adsorption on the catalyst, and decomposition by conventional and catalytic ozonation. Figure 1 (a) presents the NP elimination by an adsorption on the catalyst, as well as in conventional and catalytic ozonation (Fig. 1b). As observed, the NP elimination was more rapedly by catalytic ozonation (1-2 min), compared with conventional process (4 min), and by an adsorpion only 35% during 30 min. The catalyst activity demonstrated by the different behaviors of the formation and decomposition of the recalcitrant ozonation intermediates (not presented). The NP decomposition in

the presence of the FeF catalyst of three different sizes (8.93 cm^2g^{-1}, 8.20 cm^2g^{-1}, 7.14 cm^2g^{-1}) demonstrated the significant influence of this parameter. In general, the catalytic ozonation showed better results in the reduction of the recalcitrant ozonation products concentration, the number of the total compounds formed (4), in comparison to the conventional ozonation (5) (data not presented).

The more representative catalytic effect of the FeF catalyst were confirmed by means of the global mineralization degree obtained by means the TOC measurement (Figure 2). As can observed, the presence of the catalysts increased the mineralization degree up to 37.85% compared with 9.65% for the conventional ozonation during 60 min, and up to 50.43% after 120 minutes of treatment in the comparison with simple ozonation (11.91%). In the same time, it may be see the positive effect of the catalyst size on the mineralization degree. Namely, the augment of catalyst size leads to the increase of the mineralization degree. So, the catalyst size variation from 7.14 cm^2g^{-1} up to 8.93 cm^2g^{-1} leads to the 1.5 times increase the mineralization degree during 60 minutes and to 1.6 times during 120 minutes. Furthermore, the treatment time increase from 60 minutes up to 120 minutes also augment the mineralization degree of 30%.

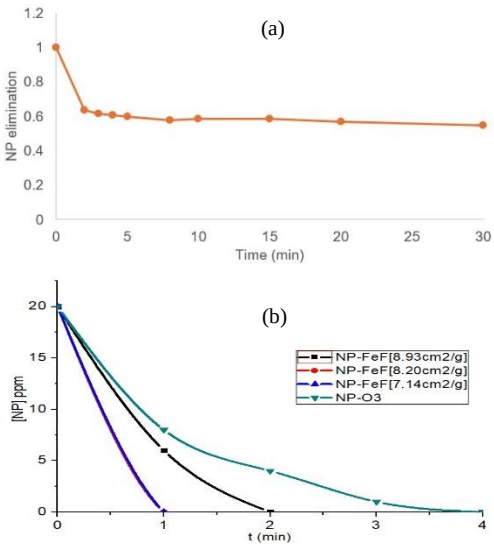

Figure 1. Elimination of the NP by adsorption on the catalyst (a), and by conventional and catalytic ozonation (b).

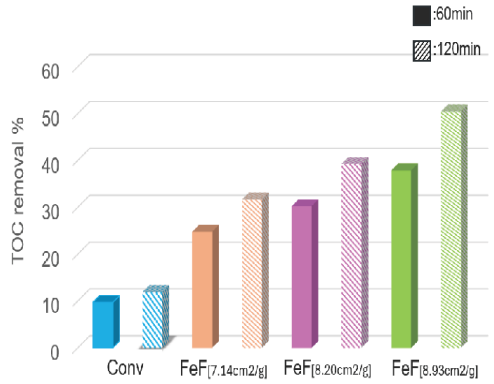

Figure 2. Mineralization degree of the NP in conventional and catalytic ozonation during 60 min and 120 min of treament.

Conclusions

The catalytic ozonation in the presense of the iron foam (FeF) catalyst film was effective in the decomposition as of the naproxen in an aqueous solution, as well as the recalcitrant ozonation intermediates. The better catalytic activity demonstrated the FeF catalyst with the higher volume and massa ($3cm^3$/1.1232g) during 2 hrs of treatment, which permited obtain the high mineralization degree (50.43%).

Acknowledgments

The authors acknowledge Consejo Nacional de Humanidades Ciencias y Tecnologías (Project No. A1-S-21608), Instituto Politécnico Nacional (Projects No. 20230273), that provided financial support.

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