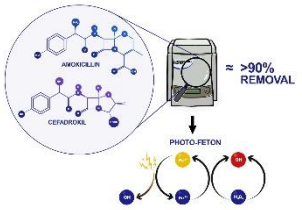
Degradation of amoxicillin and cefadroxil by the photo-Fenton process using UV-LEDs at near-neutral pH

POSTER
Ph.D. Student: N
Journal: NONE

H. Souza[1], *C. Xavier*[2], *V. Labriola*[2], *E. Azevedo*[2]. *(1) Federal University of Acre, Rod. BR 364, km 04 - Distrito Industrial, Rio Branco, Brazil, hialesouza@hotmail.com (2) University of São Paulo, Av. Trabalhador São-Carlense, 400, Pq. Arnold Schimidt, São Carlos, Brazil*



The simultaneous degradation of the antibiotics amoxicillin and cefadroxil in simulated hospital effluent was carried out using the photo-Fenton process, mediated by ferrioxalate, UV LEDs as the light source, and pH close to neutrality. Degradation experiments were performed according to a 2^3 full factorial design, with triplicate at the central point (factors: LED power, Fe^{3+} and H_2O_2 concentrations). The drugs were analyzed by high-performance liquid chromatography. The desirability function was used to consider the joint degradation of the two antibiotics, both with an initial concentration of 15 mg L^{-1}. The applied process was able to virtually 100% of the antibiotics in 10 min.

Introduction

Among the wide variety of pollutants, contaminants of emerging concern stand out, as they are potentially toxic substances, often difficult to be degraded under environmental conditions, and not regulated [1]. These compounds are usually present in the environment from ng L^{-1} to µg L^{-1}, being bioaccumulative and pseudo-persistent. Antibiotics are drugs that, when they enter ecosystems, can cause bacterial resistance, increase mortality, and morbidity [2]. Considering the deleterious effects of antibiotics in water resources, it is essential to improve treatment methodologies capable of removing this type of contaminant from those kinds of matrices. Advanced Oxidative Processes (AOPs) are an effective choice for the degradation of recalcitrant contaminants [3]. Those processes generate highly oxidizing species, such as hydroxyl radicals (•OH), which react with most organic compounds with low selectivity [4]. Therefore, the scope of this research was to degrade the antibiotics amoxicillin (AMO) and cefadroxil (CFX) were simultaneously degraded in simulated hospital effluent (SHE) by the photo-Fenton process mediated by ferrioxalate, using UV LEDs as the light source, at pH close to neutrality.

Material and Methods

The following reagents were used as received: hydrogen peroxide (H_2O_2, 30%), ferric nitrate nonahydrate ($Fe(NO_3)_3 \cdot 9H_2O$, 98%), concentrated sulfuric acid (H_2SO_4, P.A.), oxalic acid nonahydrate ($H_2C_2O_4 \cdot 9H_2O$, 98%), and sodium hydroxide (NaOH, 98%). The Simulated Hospital Effluent was prepared with the following substances (mg L^{-1}): cefadroxil (15), amoxicillin (15), sucrose (30), glucose (30), urea (20), citric acid (20), sodium acetate (20), magnesium carbonate (50), sodium carbonate (50), ammonium chloride (20), ammonium phosphate (20), potassium nitrate (10), and sodium sulfate (25). Experiments were performed according to a 2^3 full factorial design with a triplicate at the central point and three factors (Fe^{3+} and H_2O_2 concentrations and LEDs power).

Results and Discussion

The simultaneous degradation of the antibiotics in the SHE was optimized (Figure 1). Table 1 shows the performed experiments, the degradations obtained, the respective desirability functions, and the global desirability. The simultaneous degradation of AMO and CFX increased as the of Fe^{3+} concentration increased from 5 to 15 mg L^{-1}, H_2O_2 concentration from 20 to 40 mg L^{-1}, and LEDs power from 40 to 60% (in other words, from the – – – experiment to the + + + one). The global desirability reached 0.96, which virtually means complete removal of the AMO and CFX antibiotics.

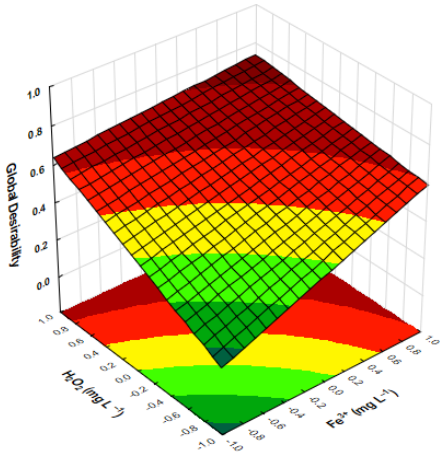


Figure 1. Response surface of the performed 2^3 factorial design (main factors).

Table 1. Experimental matrix and results of the performed 2^3 full factorial design with triplicate in the central point. Desirability limits: L_{AMO} = 60% and U_{AMO} = 100%; L_{CFX} = 50% and U_{CFX} = 100%;. Time: 10 min.

		−	+
Factors:	1: Fe^{3+} (mg L^{-1})	5	15
	2: H_2O_2 (mg L^{-1})	20	40
	3: LEDs power (%)	40	60

Run #	1	2	3	Degradation (%)		Desirability		Global Desirability
				AMO	CFX	AMO	CFX	
1	0	0	0	82.2	79.3	0.56	0.59	0.57
2	−	−	−	63.0	51.8	0.08	0.04	0.05
3	+	−	+	91.7	91.0	0.79	0.82	0.81
4	+	+	−	79.0	73.1	0.48	0.46	0.47
5	0	0	0	81.5	75.9	0.54	0.52	0.53
6	+	+	+	97.6	98.5	0.94	0.97	0.96
7	+	−	−	74.8	64.9	0.37	0.30	0.33
8	0	0	0	86.2	83.0	0.66	0.66	0.66
9	−	−	+	69.6	59.7	0.24	0.19	0.22
10	−	+	−	87.3	90.5	0.68	0.81	0.74
11	−	+	+	76.8	78.8	0.42	0.58	0.49

Conclusions

The performed 2^3 full factorial design was capable of optimizing the simultaneous degradation of amoxicillin and cefadroxil antibiotics using (LED)photo-Fenton process, mediated by ferrioxalate at a near-neutral pH. That condition is not possible in the conventional photo-Fenton process. The simultaneous removal of the antibiotics was close to 100% in 10 min of treatment. The use of UV LEDs in the photo-Fenton process is somewhat recent and presents the advantages of longer service life, lower energy consumption, and the feasibility of compacting the system. In addition, LEDs do not require heating to reach maximum emission.

Acknowledgments

This work was supported by the São Paulo Research Foundation (FAPESP), the Coordination for the Improvement of Higher Education Personnel – Brasil (CAPES) – Finance Code 001, and the National Council for Scientific and Technological Development – CNPq.

References

[1] J. Sim, W. Sima, J. Lee, E. Lee, S. Shin, S. Hwang, *Chemosphere*, 82 (2011) 179.
[2] R. Loureiro, F. Roque, A. Rodrigues, M. Herdeiro, E. Ramalheira, *Revista Portuguesa de Saúde Pública*, 34 (2016) 77.
[3] M. Marchetti, E. Azevedo, *Journal of Water Process Engineering*, 35 (2020) 101.
[4] K. Araújo, C. Arruda, R. Antonelli, B. Gaydecxka, A. Granato, G. Malpass, *Revista Ambiente & Água*, 11 (2016) 12.