Combined System of Ozonation and Biodegradation in Continuous Mode with Internal Recirculation for Mineralization of Phenols Mixture

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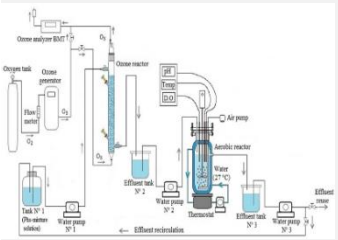

In the present work, it was studied the mineralization of three phenols: phenol (Ph), 4-chlorophenol (4-CPh), and 2,4-dichlorophenol (2,4-CDPh) and their mixture through the combination of ozonation and biodegradation in a continuous multistage system, evaluating the recirculation of the treated water, to emulate its potential reuse. In the continuous ozonation, the three phenols and most of the ozonation intermediates were decomposed (≈ 99%), employing a hydraulic retention time (HRT) of 60 min. Under these operation conditions, the Total Organic Carbon (TOC) reduction was 36.7%. The pre-ozonized effluent was fed to a bioreactor operated in continuous mode, where a global mineralization of 97% was achieved. Finally, a part of the bioreactor's effluent was recirculated to the ozonation column, achieving similar TOC reduction.

Introduction

The application of ozone-based advanced oxidation processes (AOPs) for eliminating chlorophenols (CPhs) in water has been widely studied; however, the total mineralization of these pollutants requires high amounts of oxidants and energy. The coupling of processes such as ozonation and biodegradation is a good alternative to improve the efficiency of individual treatments, reducing the toxicity of contaminants and enhancing the degree of mineralization in the coupled process [1,2], which facilitates the reuse of treated water. Most previous studies focus on the individual degradation of phenols and chlorophenols operating in semi-batch systems [2-4]; then, this study investigates the treatment of a mixture of CPhs through a coupled process of ozonation-biodegradation in a continuous system. The main objectives were to study (i) the degradation efficiency of a mixture of three phenols during the continuous ozonation, (ii) the impact of ozone dose over the pre-ozonated phenols mixture toxicity for biodegradation, (iii) the mineralization effectiveness of the coupled process: ozonation-biodegradation, and (iv) the effect of recirculation of treated effluent on ozonation performance.

Material and Methods

Ozonation procedure

The ozonation of Ph, 4-CPh and 2,4-DCPh and their mixture was conducted in a bubble column (800 mL) (pH0 = 5.0-5.1). The initial concentration of the individual phenols was 120 mg L-1 and 40 mg L−1 of each in the mixture. The concentration of phenols and ozonation by-products was monitored by HPLC. The ozonation kinetics was monitored through the ozone concentration in the gas phase at the reactor outlet (ozonogram).

Before the study of the continuous system, the effect of the ozone dose (mgO3 per mgC) over the mineralization (in the coupled process) was studied (in semi-batch ozonation and batch biodegradation) to establish the experimental conditions employed in the continuous system (ozonation-biodegradation).

In the continuous system, the CPhs solution was pumped into the reactor at a flow of 13 mL min-1 (hydraulic retention time of 60 min); the pre-ozonated water was accumulated in tank 2 and fed into the bioreactor (see Graphical Illustration). Biomass growth, residual phenolic compounds, and ozonation intermediates concentration, as well as the TOC values, were monitored through the bioprocess.

Internal recirculation of treated water

The bioreactor's effluent was collected in tank 3 (Graphical Illustration). Then, a part of this effluent was mixed with the non-ozonized phenolic solution (1:12 ratio) and fed to the ozonation column under the same experimental conditions.

Results and Discussion

Effect of the ozone dose over the global mineralization degree

According to our previous studies [2,5], the extent of pre-ozonation is critical to reducing the toxic compounds that could inhibit the bioprocess. Thus, several experiments were conducted varying the ozone dose from 0 to 17.2 mgO3 per mg C. A clear effect of the ozone dose over the mineralization and the biodegradation time was observed; this was related to the decomposition of the intermediates and the corresponding reduction of the ozonation

effluent's toxicity. With the highest studied dose, 97% of mineralization was achieved. These results were considered to establish the hydraulic retention time for continuous ozonation and replication of these conditions in the continuous mode of operation (ozonation-biodegradation).

Phenols ozonation in the continuous mode

Figure 1 shows the variation of the ozone concentration in the gas phase (ozonograms) for the individual phenols and their mixture.

The decomposition kinetics turned out to be different, depending on the chemical structure of phenols and their reactivity with ozone.

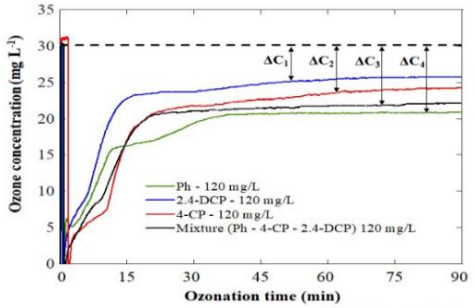

Figure 1. Ozonograms of individual phenols and their mixture in the continuous system.

Despite the high phenols removal during continuous ozonation (99%), only around 40% of the TOC removal was achieved. Therefore, the recalcitrant ozonation products remained in the ozonized water, constituting the substrate for microorganisms in the subsequent biodegradation step.

Biodegradation of the ozonation products of the phenols mixture

The pre-ozonized mixture (TOC = 44.8 mg L-1) was treated in a 3.5 L bioreactor, employing a microbial consortium previously adapted to the ozonation products (Figure 2).

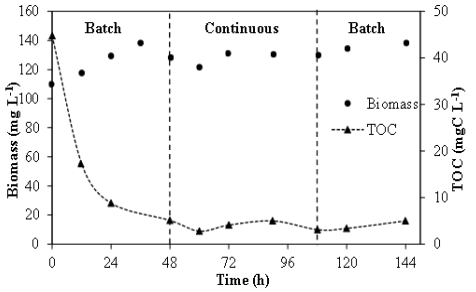

Figure 2. Evolution of biomass and TOC removal during the continuous biodegradation of pre-ozonized CPhs-mixture.

Through the bioprocess, an active reduction of the TOC was observed, from 80.3% in the first 24 hours to 93.0% during 108 hours (97% of global mineralization, considering the coupled process).

The results of Figure 2 suggest that the selected ozone dose reduced the toxicity of the phenols' initial solution, and the remaining ozonation products were efficiently eliminated in the biodegradation step. Controlling the ozone dose before the bioprocess ensures the high quality of treated water, allowing its reuse or safe discharge into the environment.

Internal recirculation of treated water after biodegradation

In the case of the internal recirculation of a part of the treated water to the continuous ozonation step, a TOC removal in this step of 47.7% was obtained, suggesting good stability of the ozonation in continuous mode.

Conclusions

The proposed treatment, combining ozonation-biodegradation processes operating in continuous mode, allowed the achievement of a high mineralization degree (97%). As a pre-treatment stage, ozonation degraded the bio-recalcitrant organic fraction, reducing the ozonized effluent's toxicity and improving the performance of the biodegradation step, where the previously adapted microorganisms were able to consume the remaining ozonation products. This treatment scheme could facilitate the reuse and recirculation of the treatment water to enhance the proper management of the water resources.

Acknowledgments

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