Enhancing Carbamazepine Degradation in Aqueous Solutions Using Hydrodynamic Cavitation with Ozone and UV Radiation

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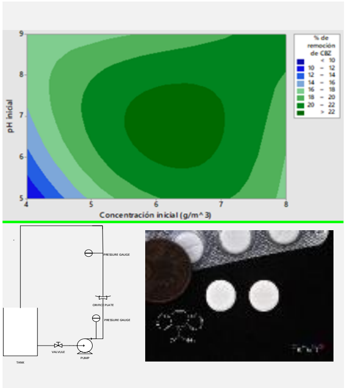

Carbamazepine (CBZ) is a pharmaceutical product used for the treatment of mood disorders. Due to its stable structure, CBZ is not completely absorbed in the human body and is consequently discarded into water resources, generating a negative environmental impact. Therefore, the researchers conducted experiments using hydrodynamic cavitation (HC) with UV and ozone, which resulted in a significant reduction in the concentration of CBZ in aqueous solutions. The effects of different pH levels (5, 7, and 9) and initial CBZ concentrations (4 g/m³, 6 g/m³, and 8 g/m³) were analyzed. The results indicate a significant reduction in CBZ concentration. However, despite a considerable reduction in the final CBZ concentration, the process did not achieve complete mineralization, as indicated by the lack of TOC (total organic carbon) removal.

Introduction

Pharmaceuticals and personal care products (PPCPs) are a broad group of chemical compounds, also known as emerging contaminants. In recent years, the consumption of PPCPs has gradually increased, including carbamazepine (CBZ) [1]. CBZ is an antiepileptic drug used to control seizures and treat conditions such as trigeminal neuralgia, depression, and other disorders. Due to its high annual production and very stable molecular structure [2], carbamazepine (CBZ) is frequently found in bodies of water. Considering that 13% of the CBZ dosed into the body is excreted unchanged, it generates direct effects on aquatic ecosystems. These effects include histopathological changes in various organs of fish and impacts on the growth and adaptability of species such as Artemia parthenogenetica [3]. Consequently, it is important to research alternative wastewater treatment technologies that efficiently remove this emerging pollutant. Advanced oxidation processes (AOPs) are techniques that have shown effective results in reducing the concentration of the compounds [4]. Hydrodynamic cavitation (HC), in particular, has demonstrated excellent performance and efficiency due to the generation of highly reactive free radicals and intense mixing. This phenomenon is caused by a pressure drop that generates the formation of bubbles, which, upon implosion, create hot spots with high pressures and temperatures, producing OH radicals that facilitate the dissociation of water [5]

The simultaneous use of different advanced oxidation processes is known as intensification. In this project, the HC was intensified with ozone and UV radiation to maximize the production of OH radicals and thereby achieve a decrease in the concentration of CBZ in aqueous solutions [6][7]. The use of these two oxidizing agents improves the process because neither leaves traces of unwanted ions in the water. They can directly attack contaminants and promote the formation of OH radicals. Previous studies indicate that UV radiation alone is not sufficient to reduce the concentration of CBZ in water within 2 hours [8]. On the other hand, ozone can reduce the concentration of CBZ in water by up to 49.1% within 2 hours of reaction, while cavitation alone can achieve approximately 2.5% removal [9]. Therefore, it is of interest to analyze the results of applying these three treatment techniques simultaneously.

Material and Methods

The aqueous solutions were prepared from CBZ with 99% purity, and supplied by a local pharmaceutical company. Ultrapure water was supplied from an ELGA Pure Lab water purifier. The initial pH of the aqueous solutions was adjusted with 0,1 N NaOH and 0,1 N HCl, reagents of the brand Merck Titripur. The ozone was supplied with an ozone generator with a power consumption of 10 W and a dose of 0.5 g/h, and the UV radiation was applied with a submersible lamp with a light source of 254 nm and a power of 10 W. The cavitation reactor consists of a tank with a 0.1 m^3 capacity, connected from the bottom to a 745,7 W pump. The discharge pressure of the pump is 449988,56 Pa. The perforated plate has 5 holes of 2 mm in diameter. Located 0,2 m from the pump discharge. After the plate, the water travels 0.6 m vertically and finally returns to the bottom of the tank. The volume used for each test was 0,017 m^3. A factorial experimental design was implemented, with two factors and three levels of each factor; the factors were the initial pH of the solution (5, 7, and 9) and the initial concentration of CBZ (4 g/m^3, 6 g/m^3, and 8 g/m^3). All the experiments were carried out in triplicate.

Results and Discussion

The average results of the tests are presented in Table 1. It can be observed that the maximum removal was achieved at an initial pH of 7 and an initial CBZ concentration of 6 g/m^3. However, there was no decrease in TOC concentration in any of the tests, indicating that the evaluated process has the capacity to transform CBZ but does not achieve complete mineralization. Furthermore, at an acidic pH, the rate of recombination of OH radicals to form water is higher than at an alkaline pH [11].

Conversely, for concentrations equal to or greater than 6 g/m^3, the removal of CBZ tends to exceed 14%. At pH levels greater than or equal to 6, the removal increases. This can be attributed to the fact that at pH levels between 0 and 5, CBZ is found mainly in molecular and positively charged ionic forms [12]. At higher pH levels, the ionization of CBZ allows the radicals to act more effectively, resulting in a greater decrease in the concentration of CBZ. Despite the greatest effectiveness being observed at pH 7, the difference between the values obtained at pH 9 is less than 4%. This difference becomes even smaller at pH 9 with an initial concentration of 8 g/m^3, confirming the role played by ozone in the treatment of organic pollutants through the formation of radicals, which occurs more effectively at alkaline pH. The maximum removal percentage with only HC was 4,76%. For the test with UV radiation alone, there was an increase in degradation of 1%, and the tests carried out with ozone alone achieved a 15% removal of CBZ. In none of these tests was there a decrease in TOC.

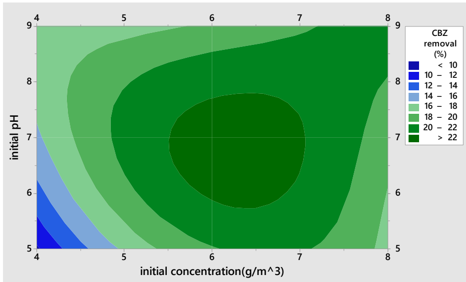

Figure 1. Contour plot for percentage removal of CBZ.

Table 1. Results of CBZ degradation.

pH	Initital concentration	% CBZ	Standard deviation	pH	Initital concentration	% CBZ	Standard deviation
5	4	10,11	0,38	9	6	18,31	0,83
7	4	17,34	1,53	5	8	17,33	0,50
9	4	16,46	2,38	7	8	18,72	1,88
5	6	19,42	0,33	9	8	21,29	0,31
7	6	22,03	0,75	9	6	18,31	0,83

Conclusions

The hydrodynamic cavitation (HC) process, when intensified with UV radiation and ozone, effectively reduces the concentration of CBZ but does not achieve complete mineralization. The tests indicate that the individual effectiveness of each process is lower compared to the combined intensified process. Both the pH and the initial concentration of CBZ in the aqueous solution significantly influence the treatment's effectiveness. Furthermore, the interaction between these two variables also affects the removal efficiency of CBZ.

Acknowledgments

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