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## Degradation of a macrolide antibiotic through the ozone process catalyzed with $\text{H}_2\text{O}_2$ , $\text{Fe(II)}$ and magnetite.

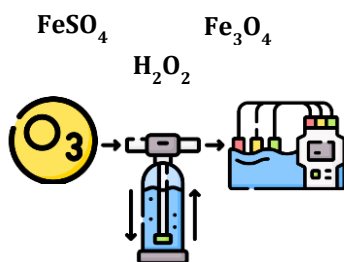
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Ph.D. Student: N

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The conventional application of chlorine for water disinfection has the disadvantage of producing trihalomethanes if their precursors are present, hence the recommendation for ozone application as a pretreatment, significantly reducing this possibility.

When ozone is catalyzed by other agents, its degradative capacity increases, becoming a very good alternative if this catalyst is low-cost or readily available. Commonly used catalysts include transition metal ions or oxides, hydrogen peroxide, and even radiation.

The application of catalytic ozonolysis is proposed in an aqueous solution of a macrolide antibiotic, comparing the dosage of  $\text{H}_2\text{O}_2$ ,  $\text{Fe(II)}$  and magnetite. The effect of a simulated hospital wastewater matrix on the degradation process under the best degradation conditions will also be assessed.

### Introduction

The consumption of antibiotics worldwide has dramatically increased due to COVID-19, especially fluoroquinolones and macrolides. Due to discharges from both manufacturing and consumption, quantities of these drugs have been detected in various water sources, including municipal wastewater treatment plant effluents [1].

The presence of antibiotics in drinking water increases the antimicrobial resistance of certain microorganisms and renders the antibiotics ineffective against these new variants, necessitating the use of stronger drugs, which may not be effective [2].

Since municipal wastewater treatment plants are unable to remove these compounds due to their antimicrobial properties, the implementation of a complementary system is necessary to ensure their destruction or at least their transformation into biodegradable molecules [3].

Advanced Oxidation Processes are recommended when it is necessary to eliminate recalcitrant compounds or when other conventional technologies fail to degrade them [4].

The purpose of this work was to determine an Advanced Oxidation Process based on catalyzed ozone treatment for a widely used antibiotic Azithromycin.

### Material and Methods

Azithromycin was provided by the pharmaceutical company Tecnoquímicas S.A., Hydrogen peroxide 30% p.a. (Carlo Erba), Ferrous sulfate p.a. (Merck) and Magnetite p.a. (Sigma-Aldrich) were used as

promoting agents for the formation of hydroxyl radicals. Dipotassium phosphate p.a. (Merck) and Methanol HPLC (Merck) were used in the elution process of drugs. Potassium indigotrisulfonate (Sigma-Aldrich), Phosphoric acid 85% (Merck) and Sodium dihydrogen phosphate p.a. (Merck) were utilized for residual ozone measurement (4500-O3B). Sodium metabisulfite (Panreac) was employed to stop the reaction. Hydroxy sodium (Merck) was used to a 0.01M solution for pH adjustment. The absorption spectrum of AZM was analyzed in a Jenway 7200 spectrophotometer. The degradation of drug was monitored by UHPLC Thermo Ultimate 3000 with DAD detector on a Restek Raptor C18 column (3 mm I.D. x 150 mm, particle size 2.7 $\mu$ m). The injection volume was 20  $\mu$ L. An isocratic elution mode was applied with a mixture of 1.0 mM phosphate buffer and Methanol at a ratio of 10:90, measured at 210 nm.

Solutions were prepared at a concentration of 100 ppm AZM and subjected to degradation by adding the oxidizing agent and exposing them to ozone. Catalyst concentrations were varied to 0.5 and 1.0 mM, pH levels were assessed at pH 9 and under unaltered conditions. Samples were collected every 5 minutes to assess degradation. All tests were performed in triplicate.

Two traps, each with a volume of 250 ml, were positioned in series following the reactor. They contained a 2% Potassium iodide solution, serving to capture any excess ozone.

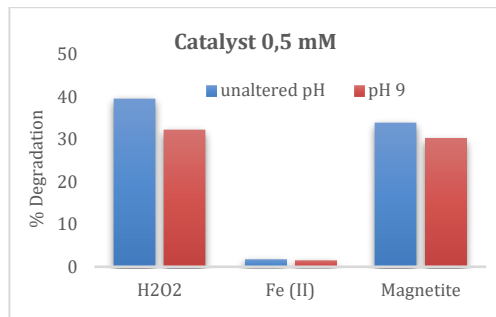
### Results and Discussion

The influence of catalyst addition on AZM

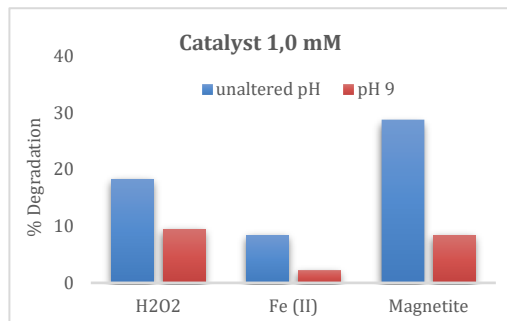
degradation via ozonation is depicted in Figs 1 and 2, correlating with the analyzed pH levels. Clearly, the combined use of catalysts and ozone significantly enhanced AZM degradation compared to the control treatment. At catalyst concentrations of 0.5 mM, the lowest level examined, degradation rates followed the sequence  $H_2O_2 > Fe_3O_4 > Fe(II)$ , while at 1.0 mM, the order shifted to  $Fe_3O_4 > H_2O_2 > Fe(II)$ . Notably, the most effective degradation rates were consistently observed under unchanged pH conditions, averaging approximately 7.4.

At elevated concentrations, pH exerts a pronounced influence on AZM degradation, particularly evident in treatments involving magnetite. Here, the impact is greater, with degradation that decreases from 28.6% at neutral pH to 8.4% at pH 9.

This underscores the efficacy of the application of catalytic ozonation in the removal of AZM, particularly with  $H_2O_2$  at a 0.5 mM concentration, achieving a notable 39.5% degradation rate under unchanged pH. However, when applying  $H_2O_2/O_3$  treatment to degrade AZM in hospital wastewater matrices, it's evident that present species interfere with the degradation mechanisms of the molecule, thereby decelerating bond-breaking reactions.



**Figure 1.** Percentage of AZM degradation, 0.5 mM catalyst.



**Figure 2.** Percentage of AZM degradation, 0.1 mM catalyst.

## Conclusions

The research has demonstrated that incorporating catalysts into the ozonation process enhances the degradation of AZM. However, the presence of other compounds can slow down this degradation. The pH of the solution also influences AZM degradation, with the impact varying depending on the catalyst type and concentration. Notably, the addition of magnetite to the ozonation process accentuates this pH-dependent effect, resulting in a decrease in degradation with a pH of 9.

## Acknowledgments

The authors would like to acknowledge the financial support of Institution University Colegio Mayor de Antioquia through project FAI67.

## References

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