

Advanced catalytic and electrochemical ozonation for sustainable pharmaceuticals degradation

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Highlights

- Ni-Sb-SnO₂ anodes enhance electrochemical ozone evolution reaction efficiency.
- Fe-H-Beta-25-EIM zeolite boosts metoprolol mineralization and total organic carbon removal.
- Elevated stirring speeds optimize reactive oxygen species mass transfer.
- A significant reduction in toxic pharmaceutical transformation products was achieved through the combined application of a Ni-Sb-SnO₂ (NSS/SiOx) electrocatalytic system and Fe-H-Beta-25-EIM zeolite.

1. Introduction

Pharmaceuticals represent an emergent category of persistent environmental toxicants that largely escape conventional wastewater treatment processes, accumulating in aquatic ecosystems and posing acute ecological risks.[1] Metoprolol, a widely prescribed β -blocker, is frequently detected in surface waters, where it and its transformation products, particularly metoprolol acid exhibit pseudo-persistence and toxicity toward aquatic organisms [2]. Recent assessments emphasize that traditional biological and physicochemical water treatments are inadequate for completely mineralizing such complex pharmaceutical structures [3]. Consequently, advanced oxidation processes (AOPs) have gained prominence. While traditional ozonation degrades parent compounds, it often generates partially oxidized intermediates with undetermined or increased toxicity [4]. To address this, the integration of electrocatalytic ROS (Reactive Oxygen Species) production using novel heterojunction anodes with heterogeneous catalytic ozonation presents a highly efficient, sustainable pathway for the complete mineralization of persistent pharmaceuticals [5].

2. Methods

The experimental framework utilized a double-jacketed 250 mL batch reactor to evaluate the degradation of metoprolol tartrate (18 μ M/L). Ozone was generated *in situ* using an advanced electrocatalytic ROS method. The anode consisted of a heterojunction Ni-Sb-SnO₂ NSS/SiOx electrocatalyst. A heterogeneous catalyst, Fe-H-Beta-25-EIM zeolite, was deployed within a SpinChem rotating bed apparatus to maximize mass transfer. Process parameters, including stirring speed (60 vs. 400 rpm), temperature (5 °C, 20 °C, 35 °C), and applied current (50 mA vs. 100 mA), were systematically varied. Liquid chromatography-mass spectrometry (LC-MS) and Total Organic Carbon (TOC) analysis were employed to quantify metoprolol degradation, identify transformation products, and measure the extent of carbon mineralization. As shown in Figure 1, the experimental setup used for metoprolol degradation equipped with a Ni-Sb-SnO₂ anode as well as a heterogeneous catalyst (Fe-H-Beta-25-EIM) inside SpinChem rotating bed.

3. Results and Discussion

The combination of the Ni-Sb-SnO₂ ROS system and the Fe-H-Beta-25-EIM zeolite catalyst yielded a synergistic enhancement in both metoprolol degradation kinetics and TOC removal. Under optimized conditions (100 mA, 35 °C, 400 rpm), the system facilitated rapid oxidation. The Ni-Sb-SnO₂ anode shifted selectivity toward the ozone evolution reaction (OZER), generating high local concentrations of molecular ozone and hydroxyl radicals. Concurrently, the Fe-modified zeolite accelerated ozone decomposition into secondary radical species. Kinetic evaluations demonstrated that elevating the stirring speed to 400 rpm overcame mass transfer limitations, while an increase in temperature provided the requisite activation energy for accelerated degradation. LC-MS analysis confirmed that the integrated catalytic process minimized the accumulation of recalcitrant intermediates, driving the system toward complete mineralization. The application of the Fe-H-Beta-EIM catalyst combined by

electrocatalytic ozone generation (ROS) yielded a significant decrease in TOC to 1.23 mg/L (Figure 2), confirming that mineralization took place over the course of the experiment.

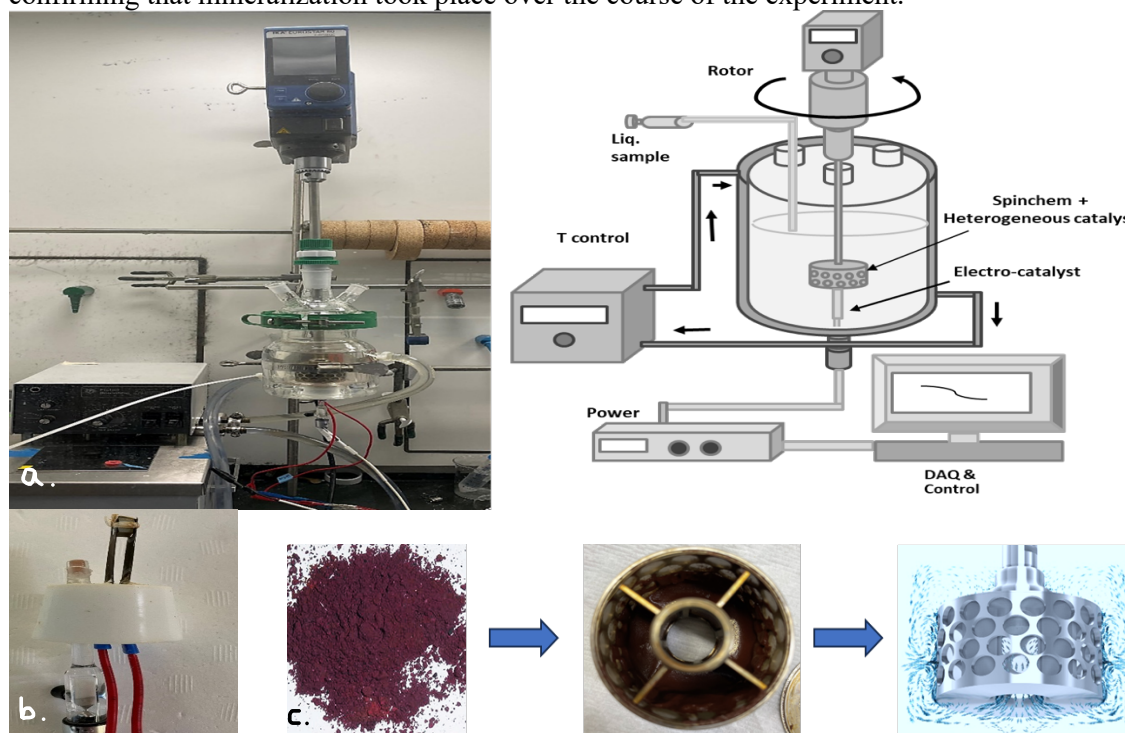


Fig. 1. a. Schematic of the Batch Reactor Used for Metoprolol Degradation. b. ROS: Electro-catalyst anode Ni-Sb-SnO₂ (NSS/SiO_x) and reference electrode and cathode c. Heterogeneous catalyst (Fe-H-Beta-25-EIM) and SpinChem rotating bed.

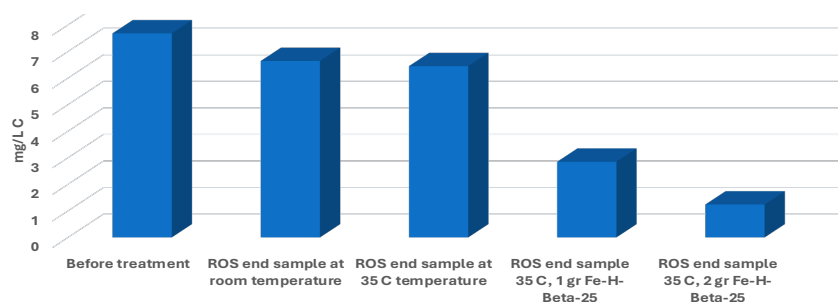


Figure 2. Total Organic Carbon (TOC) concentration during the experiment.

4. Conclusions

The integration of electrochemical ozone production with heterogeneous zeolite catalysis constitutes a highly effective, scalable approach for pharmaceutical wastewater remediation. The Ni-Sb-SnO₂ NSS/SiO_x anode demonstrated superior efficiency for in situ ozone generation, which, when coupled with the high-surface-area Fe-H-Beta-25-EIM catalyst, achieved profound reductions in metoprolol concentrations and TOC. This synergistic methodology mitigates the risk of toxic byproduct formation, operating efficiently at low energy inputs. Future optimizations of this integrated AOP hold significant promise for deployment in sustainable water treatment infrastructures to meet increasingly stringent environmental regulations.

References

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Keywords

Ozone; Electrocatalyst; Heterogenous catalyst; Metoprolol