Enhancement of Niobium Nanocatalysts with MWCNTs for the Degradation of Emerging Contaminants via AOPs

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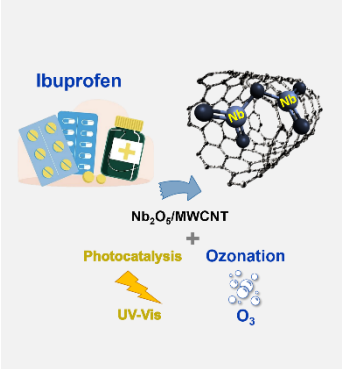
M.Z. Fidelis [1,2*] , *G.G. Lenzi* [3], *A.S.G.G. dos Santos* [2], *O.S.G.P. Soares* [2], *O.A.B. Andreo* [1]. *(1) Institution, Address, City, Country, email of the 1st author. (2) Institution, Address, City, Country*
(1) Universidade Estadual de Maringá (UEM), Av. Colombo, 5790 - Bloco D90 - Zona 7, Maringá, Brazil, michelmzzf@gmail.com. (2) Associate Laboratory in Chemical Engineering (ALiCE), Faculty of Engineering, University of Porto, Porto, Portugal. (3) Universidade Tecnológica Federal do Paraná (UTFPR), Rua Doutor Washington Subtil Chueire, 330, Ponta Grossa, Brazil



Concerns over the ineffective breakdown of persistent water pollutants like ibuprofen highlight the need for advanced treatment technologies. Nb_2O_5 emerges as a promising catalyst, resembling the properties of TiO_2. In this study, Nb_2O_5/MWCNT nanocomposites, synthesized through three sol-gel methods, demonstrate remarkable catalytic performance in ozonation and photocatalysis. These composites completely degrade ibuprofen within 30 minutes, outperforming traditional catalysts and methods due to their increased surface area and synergistic effects, providing an efficient and sustainable solution for water purification.

Introduction

The increasing presence of emerging contaminants in the environment has become a significant issue in recent years. One such pollutant, ibuprofen (IBP), a commonly used non-steroidal anti-inflammatory drug, is often found in wastewater treatment facilities [1, 2]. Ibuprofen's high lipophilicity and low biodegradability contribute to its environmental persistence and bioaccumulation. One of its by-products, 4-isobutylacetophenone (4-IBAP), has been noted for its harmful effects [3, 4]. Consequently, developing effective methods for its removal from wastewater is critical, especially since traditional treatment plants fail to eliminate it entirely. Advanced Oxidation Processes (AOPs) have shown potential in addressing these contaminants. In this context, niobium pentoxide (Nb2O5) has been identified as a viable alternative to more traditional oxides like TiO_2 and ZnO due to its impressive catalytic performance, chemical stability, non-toxic nature, and a band gap of 3.4 eV, closely matching that of TiO_2 [5]. Notably, Brazil is home to over 95% of the global niobium reserves. Furthermore, multi-walled carbon nanotubes (MWCNTs) are being explored for their unique electronic and structural qualities that enhance catalytic processes [6]. Our research aims to compare three different sol-gel methods for synthesizing niobium-based catalysts, with and without MWCNTs, assessing their efficacy in photocatalysis and catalytic ozonation.

Material and Methods

The photocatalytic and ozonation tests were conducted in a laboratory-scale reactor, as illustrated in 1a and 1b, respectively.

Figure 1. Batch photocatalytic system scheme (a) and Batch ozonation system scheme (b).

Catalyst Preparation

Three sol-gel-based methodologies were utilized to prepare the catalysts:
I) Sol-gel Method: Used niobium pentachloride ($NbCl_5$), isopropanol, Tween 20, and water to create fine, homogeneous particles.
II) Sol-gel-Pechini Method: Combined niobium ammonia oxalate with citric acid and ethylene glycol, known for producing high-purity, porous oxides.
III) Sol-gel Method: Employed $NbCl_5$, polyacrylonitrile, and N-N-dimethylformamide, enhancing structural properties like thermal stability.

Nb_2O_5/MWCNT nanocomposites

To create Nb_2O_5/*MWCNT* composites, a ball mill (Retsch MM200) with zirconia balls and jars was used. The milling was performed for 30 minutes at 10 Hz, maintaining an 80/20 mass ratio of Nb_2O_5 to MWCNTs [7].
The naming convention for the materials derived from each methodology incorporates both the method used and the calcination temperatures. For example, "CM1 NC" represents the non-calcined composites produced using Methodology 1 and “M1 NC” represents the non-calcined catalysts without MWCNTs.

Results and Discussion

Photocatalytic Tests

The effectiveness of adsorption, photolysis, and photocatalysis in the degradation of IBP was assessed using the composites with the best performance, as depicted in Fig. 2. For comparison, P25 and the M3 550 catalysts lacking CNTs (1 g L^{-1}) were also tested. The results showed that the CM3 550 composite performed comparably to the C P25, and both outperformed the catalysts without MWCNTs and the purely photolytic process. They managed to completely degrade the contaminant within 30 minutes. Additionally, it was noted that despite the inclusion of nanotubes, the adsorption process did not play a critical role in the degradation process.

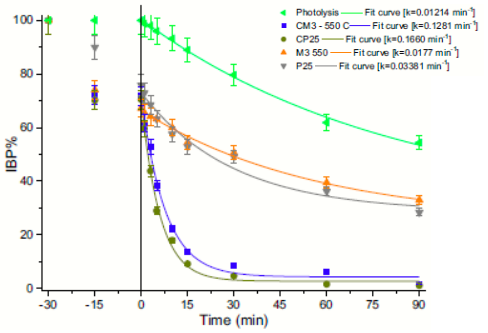


Figure 2. Ibuprofen adsorption, photolysis and photocatalytic tests using different catalysts.

Ozonation Tests

The outcomes of the IBP degradation using the catalytic ozonation process with the most effective composites are presented in Fig. 3. The results indicate that all catalysts demonstrated comparable performance, and the ozonation process alone was effective in removing the IBP molecule from the medium.

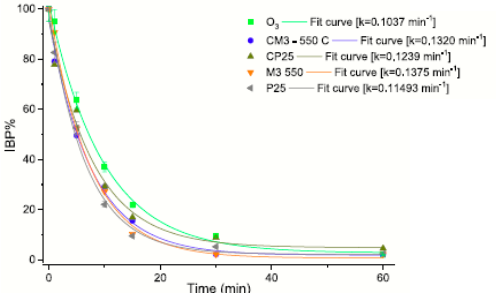


Figure 3. Ibuprofen ozonation tests using different catalysts.

However, while ozonation can quickly react with a broad array of organic compounds, transforming them into more biodegradable intermediates, the non-catalytic process typically fails to fully mineralize these compounds. This often results in the rapid formation of end-chain acids like oxalic acid.be of high quality.

It is also noticeable that the rates of ozonation/catalytic ozonation reactions are higher compared to photocatalytic reactions with catalysts without the presence of MWCNT and photolysis. However, the photocatalytic reactions with the composites exhibited higher reaction rates.

Conclusions

This study highlights the promising potential of Nb_2O_5/MWCNT nanocomposites as effective catalysts for advanced oxidation processes aimed at degrading persistent pollutants, such as ibuprofen. Among the different MWCNT/Nb_2O_5 nanocomposites synthesized, the CM3 variant treated at 550°C demonstrated superior performance in both photocatalysis and catalytic ozonation processes.

Acknowledgments

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