

Article

Catalytic Ozonation of Environmentally Relevant Pharmaceuticals Using Oxygen- and Nitrogen-Doped Activated Carbons

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Abstract

Catalytic ozonation is one of the advanced oxidation processes (AOPs) that have yielded promising results in the treatment of emerging pollutants, in which the catalyst plays a fundamental role. In this study, surface oxygen and nitrogen groups were introduced onto two commercial activated carbons, and their catalytic activity in the ozonation of lamotrigine, carbamazepine, and ibuprofen was evaluated. The modified activated carbons were characterized by N₂ adsorption at −196 °C, elemental analysis, thermogravimetric analysis (TGA) and determination of the pH at the point of zero charge (pH_{pzc}). The by-products formed in the catalytic ozonation of the compounds with each catalyst were identified, and the mineralization of the pollutants was determined. Finally, the catalysts that showed the best mineralization of the individual compounds were used for testing with the mixture of pharmaceutical contaminants. These catalysts reduced the total organic carbon (TOC) of the mixture by over 74% after 180 min of reaction. The mineralization of pollutants was improved by catalysts with higher nitrogen content and fewer surface oxygenated acid groups.

Keywords: emerging contaminants; catalytic ozonation; modified activated carbons; lamotrigine; carbamazepine; ibuprofen

1. Introduction

Emerging contaminants (ECs) are resistant compounds that are present in various environmental matrices and are difficult to remove by conventional processes. ECs include compounds such as pesticides, fertilizers, pharmaceuticals, and natural and synthetic hormones. Although these ECs are not regulated by environmental laws in most countries, their adverse effects on the environment and human health are well known [1,2]. For instance, Mathias et al. (2018) [3] found that ibuprofen detected in environmental matrices at concentrations ranging between 0.1 and 10 µg L^{−1} caused nephrotoxicity in the fish species *Rhamdia quelen*. It was also demonstrated that at the highest concentration, it increased glutathione levels, whereas at low and intermediate concentrations, it exerted an immunosuppressive effect [3]. Silva et al. (2024) identified several genetic, biochemical and behavioural effects in early-life-stage zebrafish after exposure to environmental concentrations of carbamazepine [4]. Yang et al. (2023) found that lamotrigine, carbamazepine and oxcarbazepine can cause oxidative stress and neurotoxicity in zebrafish when tested at concentrations of 100 µg L^{−1} of each drug [5].



Academic Editors: Catherine N. Mulligan and Chengyun Zhou

Received: 20 May 2026

Revised: 6 July 2026

Accepted: 31 July 2026

Published: 3 August 2026

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Several studies show the presence of different ECs in various water sources [6,7], where wastewater treatment plant (WWTP) effluents are the main source of contamination. The negative impact of wastewater treatment plant discharge into rivers is high; for example, in the Jizera River (Czech Republic), a concentration increase of more than 50% was recorded for iomeprol, diclofenac, oxypurinol, celiprolol, telmisartan, lamotrigine, tramadol, carbamazepine, sulfamethoxazole and metoprolol as a consequence of WWTP discharge [8].

Advanced oxidation processes (AOPs) are emerging as an alternative for quaternary treatment of WWTPs due to their ability to degrade organic pollutants. In fact, the Directive (EU) 2024/3019 of the European Parliament and of the Council (2024), which entered into force in early 2025, mandates quaternary treatment for removing at least 80% of a list of micropollutants, among them pharmaceuticals, pesticides, and microplastics in big WWTPs [9]. Ozonation has been studied and employed in WWTPs for this purpose, given the high oxidative power of ozone (2.07 V), which enables the efficient removal of micropollutants and microbial loads [10]. Including a catalyst in the process (catalytic ozonation) can enhance the mineralization of organic compounds; this process promotes the formation of free radical species such as hydroxyl radicals ($\cdot\text{OH}$), which can react non-selectively with organic compounds. Carbon-based materials such as activated carbon, carbon nanotubes, biochar, and graphene have been shown to be efficient ozone catalysts. Table S1 [10–16] summarizes some studies employing these carbon materials in catalytic ozonation for pollutant removal.

The use of activated carbons as catalysts offers several advantages due to their large surface area and tunable surface chemistry. These textural properties and surface chemistry influence the acceleration of ozone decomposition. Some studies highlight the critical influence of the surface chemistry and porous structure of activated carbon on ozonation processes. For example, Sánchez-Polo et al. (2005) found that the most efficient activated carbons for decomposing O_3 into $\cdot\text{OH}$ are those with high basicity and a large surface area, demonstrating that the interaction between O_3 and the pyrrole groups on the activated carbon surface plays a key role [17]. Similarly, Álvarez et al. (2006) attributed the promotion of this decomposition to basic and hydroxyl surface oxygen groups on granular carbon, suggesting that they act as active sites for the conversion of O_3 to H_2O_2 [18]. Recently, Graça et al. (2026) confirmed the importance of these basic groups and structural defects when evaluating coals derived from cork waste for the catalytic ozonation of oxalic acid [19]. Furthermore, regarding textural properties, Messele et al. (2021) demonstrated that mesoporous catalysts with a high total pore volume favour the reaction, as the presence of larger pores mitigates the diffusion limitations of both O_3 and 1,3-adamantanedicarboxylic acid [20]. Similarly, Yang et al. (2022) [21] note that the proportion of different pore sizes in activated carbons influences mass transfer. They emphasize, in particular, the importance of mesopores, as an insufficient number of them restricts the diffusion of O_3 molecules from the solution to the material's internal surface [21].

Activated carbon can act as an initiator of ozone decomposition by generating free radicals, or by adsorbing and reacting with ozone on its surface, leading to the formation of surface free radicals [22]. This activity of activated carbons towards ozone can be further enhanced by modifying their surface with functional groups that increase their basicity [23].

One way to modify the surface of activated carbon is by introducing oxygenated groups to the surface, which affects the interactions between the carbon surface, organic pollutants and O_3 . Oxidation of activated carbon with HNO_3 is known to generate a large amount of surface acidic oxygen groups on the activated carbon, mainly carboxylic acids and, to a lesser extent, lactones, anhydrides and phenolic groups [24]. Thermal treatments under a N_2 atmosphere at high temperatures are used to selectively remove certain oxygen

groups, generating more basic materials [10,25]. Doping carbon materials with nitrogen is another way to modify the surface of activated carbon to increase catalytic activity, as this treatment induces a significant redistribution of charge density on the carbon and a considerable influence on electron mobility due to the large electronegativity difference between N and C [26]. Some studies show that carbonaceous materials modified with oxygen and nitrogen functional groups exhibit high catalytic activity [10,27,28]. Furthermore, these materials have the advantage of not presenting a risk of secondary contamination and are generally more economical than other catalysts [29].

In this study, commercial activated carbons were functionalized with oxygen groups using HNO_3 or with nitrogen groups using melamine as precursor, followed by thermal treatment at different temperatures. Then, these were tested as catalysts in the ozonation of lamotrigine (LTG), carbamazepine (CBZ) and ibuprofen (IBF), drugs detected in wastewater effluents at alarming concentrations. Although the functionalization of activated carbons with metal-free heteroatoms has a background in catalytic ozonation, its application in the degradation of pharmaceutical pollutant mixtures remains largely unexplored. To the best of our knowledge, this is the first study to directly correlate the chemical and textural properties of various modified activated carbons with the degradation and mineralization of LTG, whilst also evaluating the synergistic effect between the catalyst and ozone. Furthermore, this work provides an unprecedented comparison of the efficacy of carbons functionalized with oxygen and nitrogen groups for the simultaneous removal of LTG, CBZ and IBF in a mixed solution. This is important because it allows the identification of the relevant characteristics of activated carbon for use as a catalyst in catalytic ozonation to remove emerging pollutants. Therefore, in this study, the degradation of each pollutant was measured, along with total organic carbon (TOC) and the by-products generated. The two best carbons were analyzed to determine the degradation and mineralization of the mixture of the three contaminants.

2. Materials and Methods

2.1. Materials

Two commercial activated carbons were used as starting materials for the functionalization. The first was supplied by Degussa (Frankfurt am Main, Germany) (Hydrarffin as 12/450) and the second by Cabot (Boston, MA, USA) (Norit GAC 1240).

LTG, CBZ and IBF, at 98% each, were supplied by Thermo Scientific (Waltham, MA, USA), Alfa Aesar (Ward Hill, MA, USA), and Sigma-Aldrich (St. Louis, MO, USA), respectively. Nitric acid (65 wt.%) was obtained from VWR Chemicals (Radnor, PA, USA).

2.2. Activated Carbon Functionalization

2.2.1. Oxidation and Thermal Treatment

The activated carbons were oxidized with HNO_3 following the methodology described in [25]. Briefly, 9 g of the activated carbon was placed in a Soxhlet extraction apparatus connected to a boiling flask containing 300 mL of HNO_3 (6 M) and a condenser. The HNO_3 solution was heated to boiling temperature with a heating mantle for 3 h. Activated carbon was washed with distilled water until neutral pH and then dried in an oven at 100 °C.

The oxidized activated carbon was subjected to thermal treatment at 400, 600 and 900 °C under a nitrogen flow for 1 h. The modified -activated carbons were used as catalysts in the ozonation of the pollutants. After oxidation and thermal treatment, the activated carbon supplied by Degussa (AC) was called ACH-400, ACH-600, and ACH-900, respectively. When supplied by Cabot (GAC), it was designated as GACH-400, GACH-600, and GACH-900.

2.2.2. Nitrogen Doping

A 0.06 M solution of melamine (99.0%, Sigma-Aldrich) was prepared in 350 mL of ethanol (96.0%, VWR Chemicals). Then, 4.0 g of activated carbon was added and stirred for 24 h. Subsequently, melamine-impregnated activated carbon was filtered and dried at 100 °C. The material was then thermally treated at 600 °C under a nitrogen flow for 1 h. Previous studies show that treatments at this temperature have a higher nitrogen content. For example, Öztürk et al. (2021) identified that melamine-doped carbon samples tend to decrease in nitrogen content at temperatures above 600 °C [30]. Perazzolo et al. (2015) show that melamine-doped carbon begins to decompose into N₂ at 500 °C and exhibits a pronounced weight loss at 609 °C [31]. The AC and GAC treated with melamine were designated ACM and GACM, respectively.

2.3. Catalyst Characterization

The modified -activated carbons were characterized by N₂ adsorption at −196 °C, elemental analysis, thermogravimetric analysis (TGA) and determination of pH at the point of zero charge (pH_{pzc}).

The Quantachrome Nova 4200e (Quantachrome, Boynton Beach, FL, USA) was used for the textural characterization of modified -activated carbons based on the N₂ adsorption isotherm, determined at −196 °C. The samples were degassed at 150 °C for 3 h under vacuum. The Brunauer—Emmett—Teller equation (BET) was used to calculate the surface area (S_{BET}). The surface area of the meso-pores (S_{meso}) and the volumes of the micropores (V_{micro}) were calculated by the t-method. The total specific pore volume (V_p) was determined from the adsorbed amount at P/P₀ = 0.95.

The carbon, hydrogen, nitrogen and sulphur content was determined by elemental analysis using the Elementar-vario MACRO cube (Elementar, Langenselbold Germany). The oxygen content was measured in the Elementar-rapid OXY cube (Elementar, Langenselbold Germany).

The surface chemical states and chemical composition of the catalysts were determined using X-ray Photoelectron Spectroscopy (XPS). A Kratos AXIS Ultra HAS spectrometer (Kratos Analytical, Manchester, UK) was used for this purpose, with VISION software (<https://www.visionsoftwareonline.com/>) for data acquisition and CASAXPS software (<https://www.casaxps.com/>) for analysis.

By thermogravimetric analysis (TGA), the content of fixed carbon, volatiles, and ash was determined using the Netzsch STA 409 PC (NETZSCH, Selb, Germany). The sample was heated under nitrogen flow from 50 to 900 °C at a heating rate of 10 °C min^{−1}, with two isothermal steps: 7 min under nitrogen and 20 min under air.

The pH at the point of zero (pH_{pzc}) was determined by preparing solutions of NaCl (0.01 M) at different pHs (between 2 and 10); the pHs were adjusted with NaOH (0.01 M) and HCl (0.01 M). A total of 20 mL of each solution was taken, and 0.02 g of activated carbon was added and stirred for 24 h. Then, the pH was measured. The pH_{pzc} value was determined when the final pH vs. initial pH curve crossed the line pH_{final} = pH_{initial}.

Energy-dispersive scanning electron microscopy (SEM-EDS, Phenom ProX Desktop SEM) was used to characterize the surface morphology and composition of some samples.

2.4. Ozonation and Adsorption Experiments

Individual solutions of LTG, CBZ and IBF were prepared in ultrapure water (UW) at a concentration of 10 mg L^{−1} and subjected to catalytic ozonation tests with the commercial catalysts (AC and GAC) and modified catalysts (ACH, ACH 400, ACH 600, ACH 900, ACM, GACH, GACH 400, GACH 600, GACH 900 and GACM). The experiments were conducted under controlled conditions in a stirred reactor. These conditions include stir-ring at

400 rpm, an ozone concentration of 50 g Nm^{-3} , an ozone flow rate of $150 \text{ Ncm}^3 \text{ min}^{-1}$, a catalyst dose of 0.5 g L^{-1} , a solution volume of 700 mL, and a reaction time of 180 min. The catalyst dose (0.5 g L^{-1}) was selected based on previous studies and also to ensure a high density of functional groups and surface area, thereby ensuring mass transfer and ozone decomposition. Furthermore, although the concentration of 10 mg L^{-1} is above typical environmental levels, this value was selected to ensure rigorous monitoring of both the emerging contaminant and its transformation by-products. Moreover, this initial concentration allowed the degree of mineralization to be assessed with greater precision and analytical reproducibility.

Aliquots were taken at selected times throughout the reaction to determine the contaminants' concentration and the formation of by-products. A test was performed in the absence of a catalyst under the same conditions for control purposes. The catalysts with the best results in mineralizing the individual solutions of pharmaceuticals were used for the catalytic ozonation test of the mixture of the three pharmaceutical compounds, at a concentration of 10 mg L^{-1} each.

The same conditions were used for the adsorption tests as for the ozonation tests, where oxygen was injected instead of ozone. In this way, an attempt was made to reproduce the hydrodynamics of the ozonation system.

To determine the contribution of reactive oxygen species (ROS) to the catalytic ozonation mechanism, experiments were conducted using specific inhibitors. Tert-butyl alcohol (TBA, 1 mM) was used as a hydroxyl radical ($\cdot\text{OH}$) scavenger and sodium azide (NaN_3 , 1 mM) was used for singlet oxygen ($^1\text{O}_2$).

2.5. Analytical Methods

The concentrations of LTG, CBZ and IBF were measured by high-performance liquid chromatography coupled to a diode array detector (HPLC DAD, Hitachi Elite LaChrom, Mumbai, India). A Purospher STAR RP-18 column ($5 \mu\text{m}$, $250 \times 4.6 \text{ mm}^2$) was used as the stationary phase. As mobile phases, 70% (*v/v*) methanol and 30% (*v/v*) dihydrogen phosphate solution (0.065 M) were used at a flow rate of 1 mL/min. The injection volume was 50 μL ; the compounds were detected at 234 nm for LTG, 246 nm for CBZ, and 220 nm for IBF. The detection and quantification of benzoquinone (BZQ) as a by-product of the reaction were performed using a mobile phase of 70% water and 30% methanol at 245 nm.

HPLC with a UV detector (HPLC UV, Elite LaChrome, Mumbai, India) was used on a Hichrom OA-1000 column ($300 \times 6.5 \text{ mm}^2$) to quantify the acidic by-products: oxalic acid (OXL), oxamic acid (OXM), formic acid (FA), pyruvic acid (PYR) and fumaric acid (FuA). These by-products were selected since they are acknowledged as common final degradation products of the ozonation of organic pollutants [32]. Their identification was performed by comparing the chromatographic profiles of the treated samples with those obtained from the by-products' analytical standards. H_2SO_4 5 mM was used as the mobile phase with a flow rate of 0.5 mL/min and an injection volume of 30 μL .

Total organic carbon was determined in a TOC-L analyser (Shimadzu TOC-5000A, Kyoto, Japan).

3. Results and Discussion

3.1. Characterization of Activated Carbon

Degussa and Cabot, designated AC and GAC, respectively, supplied the commercial activated carbons used. Their oxidized versions were designated ACH and GACH, whilst the thermally treated samples were identified by adding the treatment temperature to their respective names (ACH-400/600/900 and GACH-400/600/900). Activated carbons were oxidized with HNO_3 in a Soxhlet system for 3 h. They were then washed with

distilled water until the pH reached neutrality and dried in an oven at 100 °C. Finally, the oxidized material was subjected to thermal treatment at 400, 600 and 900 °C under a nitrogen flow for 1 h. The melamine-doped carbons were identified as ACM and GACM. The melamine-impregnated activated carbon was filtered and dried at 100 °C. The material was heat-treated at 600 °C under a nitrogen flow for 1 h.

Oxidation treatment with HNO₃ generates a diverse range of oxygenated groups on the carbon surface, including carboxylic acid groups, lactones, carboxylic anhydrides, phenolic groups and carbonyl/quinone groups [25,33]. The acid groups decrease significantly with thermal treatments above 400 °C, and the phenol and carbonyl/quinone groups decrease in treatments above 700 °C [25]. This agrees with the elemental analysis results (Table S2), in which the oxygen percentage increases after oxidation and then decreases as thermal treatment temperature increases. This is because the oxygenated groups on the carbon surface undergo cleavage at elevated temperatures, releasing gases. AC has a higher oxygen content than GAC and is also more susceptible to gaining oxygen groups after oxidation with HNO₃; this may be due to the original raw material used to manufacture the activated carbons or to the manufacturing method.

The addition of oxygen-containing groups on the surface of the activated carbons by oxidation with HNO₃ (ACH and GACH) also results in a substantial increase in the volatile content, which decreases with the thermal treatments performed (Table S3). The HNO₃ treatment also decreased the pH_{pzc} in the ACH and GACH compared to the original activated carbon (AC and GAC), as expected, since more oxygen-containing functional groups can enhance the acidic character of the surface [34].

The melamine treatment also achieved its objective of introducing nitrogen groups on the surface of the ACM and GACM samples (Table S2). Thermal treatment at 600 °C introduces N-pyrrolic and N-pyridinic groups [35]. Pels et al. (1995) mention that under pyrolysis conditions, all the nitrogen present as N-pyrrolic is released into volatiles, but most of it is converted into N-pyridinic and N-quaternary [36]. TGA results (Table S3) indicate that this treatment increased the volatiles in both samples (ACM and GACM), confirming that the findings align with the existing literature.

Table 1 shows the specific surface area (S_{BET}) and pore volume obtained from N₂ adsorption isotherms at −196 °C (the adsorption/desorption isotherms are graphically represented in Figure S1). AC has a larger BET surface area than GAC, and GAC exhibits a higher degree of mesoporosity. When AC is oxidized, it shows a slight reduction in BET surface area, possibly due to the large number of surface oxygen groups introduced, which may block the entry of N₂ molecules into the small pores [37]. On the other hand, the thermally treated samples (ACH-400, ACH-600, and ACH-900) exhibit a higher BET surface area than ACH, and this surface area increases with thermal treatment temperature; some of the oxygen groups on the activated carbon decompose, unlocking the carbon's porosity. It has been reported that heating to 600 °C induces the decomposition of carboxylic anhydrides and most lactones, whilst at 750 °C, the total elimination of lactones, phenols and carbonyl fractions is achieved [24].

ACM suffered a 24% reduction in BET surface area compared to AC due to the introduction of nitrogen surface groups, which, like oxygen surface groups, can block the entry of N₂ into the small pores. This result is consistent with the findings of Graça et al. (2023), who reported a 19.7% and 14.3% reduction in BET surface area for two types of activated carbons treated with melamine [10].

Unlike ACH, GACH did not exhibit a reduction in BET surface area compared to the original activated carbon (GAC). This could be because the oxidation of GAC did not significantly increase the oxygen percentage. On the other hand, melamine treatment reduced

the BET surface area of the GACM by 43.9%, which may be related to the introduction of nitrogen groups that could clog the small pores.

Table 1. Textural properties of the activated carbons.

Sample	S_{BET} ($\text{m}^2 \text{g}^{-1}$)	S_{meso} ($\text{m}^2 \text{g}^{-1}$)	V_{micro} ($\text{cm}^3 \text{g}^{-1}$)	$V_{\text{P/P0}} = 0.95$ ($\text{cm}^3 \text{g}^{-1}$)
AC	1294	69	0.508	0.580
ACH	1176	56	0.478	0.541
ACH-400	1186	55	0.468	0.528
ACH-600	1335	74	0.523	0.600
ACH-900	1303	79	0.511	0.595
ACM	983	52	0.386	0.440
GAC	972	94	0.381	0.475
GACH	972	95	0.378	0.475
GACH-400	925	84	0.360	0.445
GACH-600	924	92	0.349	0.442
GACH-900	880	72	0.346	0.420
GACM	545	68	0.201	0.271

In terms of pore volume, the two oxidized activated carbons (ACH and GACH) underwent a slight reduction compared to the original activated carbons. This was due to the fixation of oxygen complexes on the carbon surface, which increased micropore constriction [38]. ACM and GACM showed the largest pore volume reduction due to nitrogen group incorporation on their surfaces. ACM underwent a 24% reduction in micropore volume and a 25% reduction in mesopore volume. GACM experienced a 47% reduction in micropore volume and a 26% reduction in mesopore volume.

Table 2 shows the XPS analysis of ACH-600 and ACM. The oxygen content of ACH-600, determined by XPS, was 9.7%, similar to the volatile amounts determined by TGA (see Table S3), while the elemental analysis shown in Table S2 reveals a lower percentage of oxygen, indicating that oxygen-containing functional groups are predominantly distributed on the surface of the activated carbon. The surface concentration of these active sites is crucial for promoting ozone decomposition and enhancing the efficiency of the catalytic ozonation reaction. XPS analysis shows that ACM contains a slightly higher amount of nitrogen (8.6%) than reported by elemental analysis, indicating that N is distributed mainly on the surface of ACM. The existing forms of nitrogen in ACM were identified based on their bond energies. ACM showed three peaks at 397.7, 400.3, and 402.8 eV, corresponding to N-pyridinic (3.0 wt.%), N-pyrrolic (4.4 wt.%), and N-quaternary (1.2 wt.%) sites, respectively. N-Pyrrolic was found to be the predominant species on the surface of the ACM, accounting for more than 50% of the total nitrogen content. These results are consistent with the findings of Pels et al. (1995), who demonstrated that pyrrole structures are stable up to 600 °C, undergoing a gradual transition to pyridine and quaternary configurations at higher temperatures [36]. Similarly, Quan et al. (2020) reported that pyrrole and pyridine functional groups are abundant in melamine-doped activated carbons following heat treatment at 700 °C [39]. Finally, it has been found that the incorporation of nitrogen into the carbon surface increases its basicity, which has a significant impact on oxidation reactions [40].

Table 2. XPS analysis of ACH-600 and ACM.

	ACH-600	ACM
C (wt.%)	89.8	90.9
N (wt.%)	0.5	8.6
O (wt.%)	9.7	0.5

The morphology and chemical composition of ACH-400, ACH-600 and ACM were studied by SEM-EDS analysis at different magnifications, as shown in Figure S2. The low-magnification images (left panels) include particle size measurements, showing that the activated carbons are millimetre-sized pellets, with the particles analyzed ranging from 1.49 to 3.35 mm in diameter. For comparison, the corresponding SEM images (right panels) were acquired at the same magnification (300 $\times$), and EDS maps (Figures S3–S5) were acquired over the imaged areas. Regardless of the functionalization treatment, all samples exhibited a similar macroscopic morphology, consisting of compact pellets formed by the agglomeration of carbon granules. No noticeable differences in particle shape, surface roughness, or structural integrity were observed among ACH-400, ACH-600, and ACM at this magnification, suggesting that the different functionalization procedures did not significantly alter the overall morphology of the activated carbon pellets.

Comparison of the EDS elemental maps of ACH-400 (Figure S3) and ACH-600 (Figure S4) reveals a decrease in the relative oxygen and nitrogen contents as thermal treatment temperature increases. This trend is consistent with the progressive removal of oxygen- and nitrogen-containing surface functional groups during thermal treatment, resulting in a more carbon-rich surface at higher activation temperatures. While the carbon and oxygen contents determined by XPS and EDS are in reasonable agreement for ACH-600, a larger discrepancy is observed for nitrogen. This difference can be attributed to the inherent characteristics of the two techniques. ACM exhibited a substantially higher nitrogen content than ACH-400 and ACH-600, as determined by both EDS (Figure S5) and XPS analyses. The good agreement between the two techniques confirms the successful incorporation of nitrogen through melamine doping.

3.2. Degradation and Mineralization

The degradation of the studied pharmaceutical compounds was tested using commercial and modified activated carbons, as shown in Figure 1. The decomposition of LTG, CBZ and IBF in the ozonation process followed a pseudo-first-order reaction in all tests performed, as confirmed by the linear relationship between $\ln([\text{Pollutant}]/[\text{Pollutant}_0])$ and reaction time; the slope of this line corresponds to the apparent degradation rate constant (k' , min^{-1}) (Table S4).

The k' (min^{-1}) values for LTG, CBZ and IBF without catalyst, i.e., single ozonation, were 0.214 min^{-1} , 1.074 min^{-1} and 0.233 min^{-1} , respectively. This indicates that CBZ is the least resistant pharmaceutical among the three tested to ozone oxidation. Both molecular structure and pKa can explain this. CBZ contains electron-rich aromatic rings and a conjugated amide group (pKa $\sim$ 13 for the amide N) [41], which provides sites highly susceptible to direct electrophilic attack by ozone. In contrast, IBF possesses a carboxylic acid group (pKa $\sim$ 4.9) that is deprotonated at neutral pH [42], which slightly reduces the electron density on the aromatic ring and thereby decreases its reactivity toward ozone. Similarly, LTG contains amino groups (pKa $\sim$ 5.7–7) that are partially protonated under neutral conditions [43], which also lowers the electron density on the triazine ring and limits direct ozonation. These factors explain the higher degradation rate observed for CBZ under single ozonation conditions.

Although ozonation alone is limited by ozone selectivity, adding a catalyst to the process promotes O_3 decomposition at the solid–liquid interface, increasing the generation of $^*\text{OH}$ that reacts non-selectively with organic compounds and has greater oxidizing power than ozone. According to Keen et al. (2024), the transformation pathway of LTG upon reaction with $^*\text{OH}$ was the hydroxylation of the chlorinated benzene ring ($\text{C}_9\text{H}_8\text{Cl}_2\text{N}_5\text{O}^+$) [44]. For CBZ, Mohammed-Amine et al. (2025) propose that $^*\text{OH}$ directly attacks the carbon atom within the aromatic ring of CBZ, forming monohydroxylated isomers ($\text{C}_{15}\text{H}_{14}\text{N}_2\text{O}_4$)

that differ in the position of the carbon atom that has been attacked [45]. Xue et al. (2023) reported that IBF degradation occurs mainly at the propanoic acid substituent and proposed hydroxylation and decarboxylation as the main degradation pathways [46].

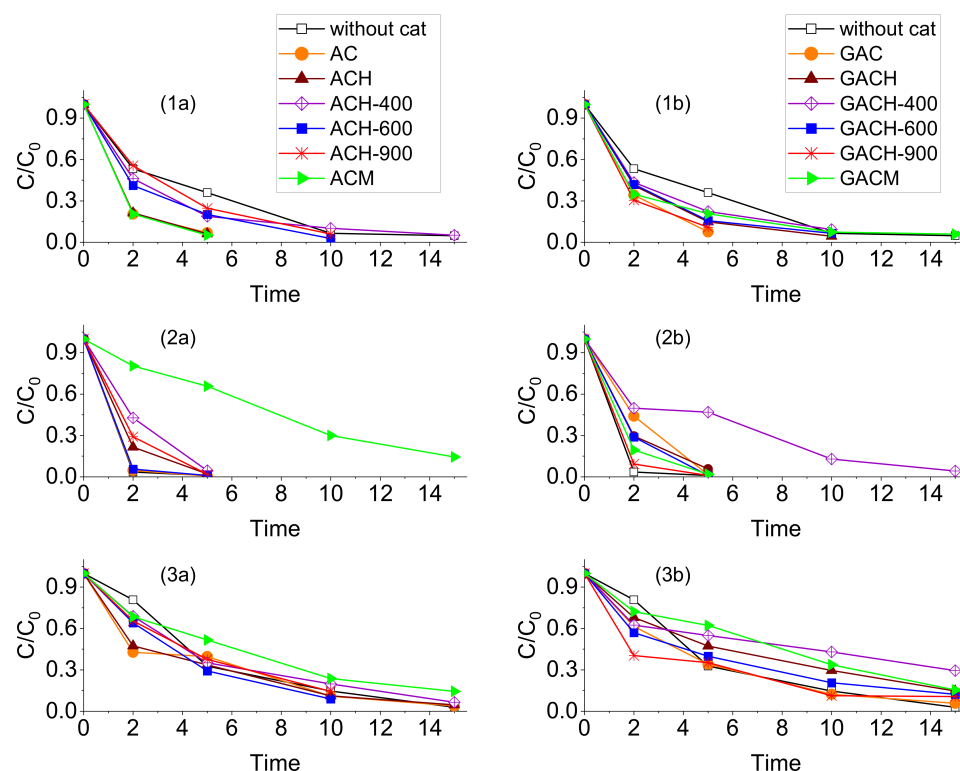


Figure 1. Ozonation of (1) LTG, (2) CBZ and (3) IBF, in the presence of: (a) modified activated carbon AC and (b) modified activated carbon GAC. Experimental conditions: $[O_3] = 50 \text{ g Nm}^3$, $[Pollutant] = 10 \text{ mg L}^{-1}$, catalyst dose = 0.5 g L^{-1} , O_3 flow rate = $150 \text{ cm}^3 \text{ min}^{-1}$, stirring = 400 rpm, reaction time = 180 min.

The evaluation of the degradation of the original activated carbons revealed that AC performed slightly better than GAC in the removal of the three contaminants (Table S4). In contrast, in the adsorption tests (using oxygen instead of ozone), GAC showed a greater capacity to remove LTG, CBZ and IBF (Table S5), even though AC has a higher S_{BET} . This behaviour can be attributed to the fact that GAC exhibits a greater mesoporosity (S_{meso}), which facilitates the internal diffusion of these drugs. These results indicate that ozonation is the main route for contaminant removal. However, adsorption played a notable role when GAC was used, whilst with AC, degradation contributed significantly more to the process.

When catalysts were used, CBZ degradation was hindered, particularly with ACM ($k = 0.130 \text{ min}^{-1}$) and GACH-400 ($k = 0.204 \text{ min}^{-1}$). These results are consistent with those obtained by Asghar et al. (2023) and Aguilar et al. (2020), who suggest CBZ decomposition is faster through a direct ozone attack rather than the hydroxyl radical [15,47].

GACH-400 was also the catalyst that most hindered IBF degradation (0.061 min^{-1}), whereas ACH-600 was the only catalyst that improved IBF degradation over ozonation alone, with a first-order rate constant of 0.244 min^{-1} . The most prominent catalysts in LTG degradation were AC, ACH and GAC, with first-order rate constants of 0.520 min^{-1} , 0.548 min^{-1} and 0.517 min^{-1} , respectively.

Figure 2 correlates the %O in the catalyst with the TOC reduction normalized by the S_{BET} promoted by AC-derived materials, thereby linking oxygen functional groups to the area actually active for mineralization. It is observed that higher TOC reduction/ S_{BET} is

achieved for all three pollutants when the %O is between 3.8% and 5.6%, corresponding to the ACH-600 and ACM catalysts. Meanwhile, the catalyst with the lowest %O has the lowest TOC reduction/ S_{BET} (ACH-900).

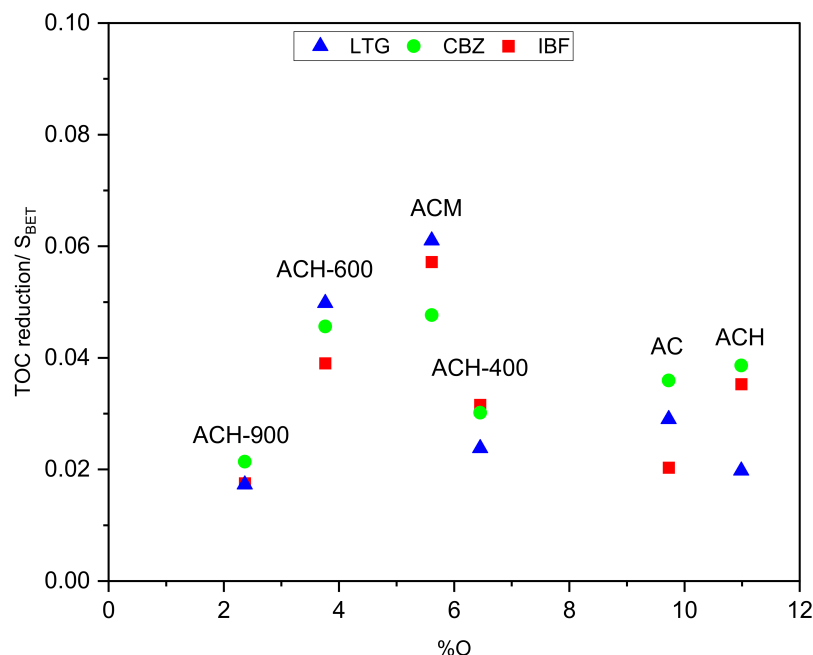


Figure 2. Variation in %O of modified activated carbons (ACs) with respect to TOC reduction/ S_{BET} .

Figure 3 correlates the %O in the catalyst with the TOC reduction/ S_{BET} promoted by GAC-derived materials. No single GAC-modified activated carbon exhibited a consistent TOC reduction for all three pharmaceuticals simultaneously. However, GACH-900, GACM, GACH-600 and GAC excel in mineralization, particularly in CBZ mineralization (Figure 3).

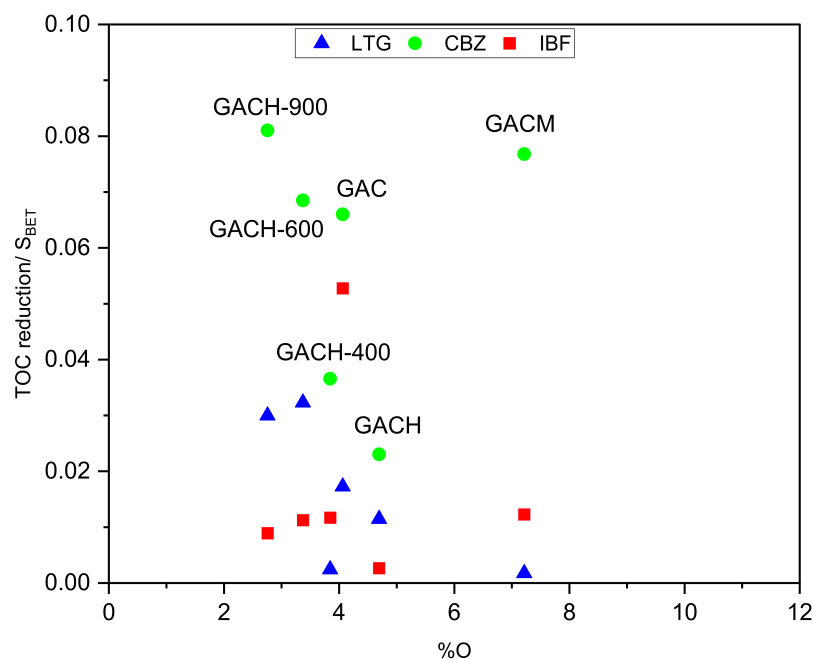


Figure 3. Variation in %O of modified activated carbons (GACs) with respect to TOC reduction/ S_{BET} .

CBZ is the compound that presented the highest degree of mineralization among all modified ACs, except for ACH-600 and ACM, for which a higher degree of mineralization was observed for LTG.

Although CBZ degradation was faster without using a catalyst (i.e., by direct mechanism with ozone), CBZ mineralization was favoured when all activated carbons were tested (reduction of TOC with catalyst > 22%, reduction of TOC without catalyst = 17%), suggesting that the catalysts developed produce reactive oxygen species (ROS) capable of converting CBZ into inorganic species [15], while attack by ozone preferably converts CBZ into other organic by-products. The TOC reduction data for the three contaminants in the individual tests are shown in Table S6.

The role of ROS was assessed using chemical trapping assays with the ACH-600/LTG system. Interestingly, the inhibitory effects of both tert-butyl alcohol (TBA) and sodium azide (NaN_3) were more pronounced during the initial stages of the reaction than at later reaction times. This observation suggests that the interactions among ozone, the catalyst surface, and the target pollutant evolve throughout the reaction, likely altering the relative contributions of the reactive oxygen species involved in the degradation process. Nevertheless, considering the initial stage of the reaction, when the inhibitory effects were most evident, NaN_3 caused a greater reduction in catalytic performance than TBA, suggesting that singlet oxygen ($^1\text{O}_2$) plays a more significant role than hydroxyl radicals ($^{\bullet}\text{OH}$) in the degradation process under the conditions studied. This behaviour is in agreement with previous work by [48].

3.3. By-Product Formation

During LTG degradation, mainly oxalic acid (OXL) and oxamic acid (OXM) were identified as reaction by-products, as shown in Figure S6. For the catalysts GAC and GACH, fumaric acid (FuA) was also identified at low concentrations ($<1.1 \text{ mg L}^{-1}$) as a degradation by-product. The formation of FuA indicates the oxidative degradation of the dichlorobenzene aromatic ring in LTG, accompanied by the release of chloride ions (Cl^-) into the environment. Furthermore, the detection of OXM indicates the effective fragmentation of the 1,2,4-triazine ring. Both processes ultimately converge in the production of OXL, identified as a common refractory by-product.

All catalysts evaluated showed OXL formation from 5 min to 180 min. The modified ACs had higher OXL formation than the modified GACs. GAC-400 was the only catalyst where OXM was not identified. This catalyst also exhibited low OXL formation, which is consistent with the low mineralization of LTG.

ACM is one of the prominent catalysts in the mineralization of LTG. When using this catalyst, a high concentration of OXL was formed in the first minutes of the reaction (compared to the other catalysts), which subsequently decreased. After 180 min of reaction, OXL is the main contributor to the final TOC (around 70%). The formation of OXM was different, as its concentration increased slightly over time.

ACH-600 is the other catalyst that outperformed LTG mineralization. In the first minutes of reaction, a high concentration of OXL was detected, which decreased by 60 min, followed by a slight increase. This indicates that OXL is being formed and simultaneously degraded. OXL is the main reaction by-product after 180 min, accounting for approximately 52% of the final TOC. On the other hand, OXM was not detected until min 30, and its concentration varied very little until min 120; at the end of the reaction, it was no longer detectable. The degradation of OXM can benefit from the solution pH (pH of the LTG solution = 5.8) since at basic and neutral pH, OXM is highly resistant to ozone and hydroxyl radical attack [22].

In the degradation of CBZ, OXL and OXM were also identified as main reaction by-products, as shown in Figure S7. In the case of ACM, GAC and GACH-600 catalysts, formic acid (FA) was also detected; however, it was no longer detected after 120 min. Aguilar et al. (2020) had previously detected the formation of OXL and FA in CBZ

degradation [47], while Orge et al. (2024) identified the formation of OXL and OXM as by-products of catalytic ozonation with activated carbon [14]. The detection of intermediate FA suggests that the azepine ring is initially cleaved via electrophilic attack by ozone and/or ROS on its reactive double bond, releasing single-carbon fragments. Subsequently, the opening and advanced fragmentation of the side aromatic rings result in OXL as a refractory final by-product. Likewise, the minor presence of OXM indicates the oxidative stabilization of the carbonaceous residues that still retain the terminal amide group of the original molecule.

The formation of OXL in CBZ degradation is much higher than in LTG degradation, which may be related to the rapid degradation of CBZ and, consequently, conversion into small-chain carboxylic acids. OXL formation was identified in reaction with all catalysts used.

The most prominent catalysts in CBZ mineralization were GACH-900, GAC, GACH-600 and ACH-600. GACH-900 and GAC catalysts formed a high concentration of OXL in the first minutes of the reaction, which gradually decreased. While the GACH-600 and ACH-600 showed a tendency to reduce the concentration, at certain times, the concentration increased slightly. OXL accounts for approximately 27% of total TOC when using ACH-600 after 180 min of reaction.

GAC was the only catalyst where no OXM formation was detected. GACH-900 and ACH-600 exhibited OXM formation from the first minutes of the reaction, which steadily decreased over time. GACH-600 also showed a tendency to reduce the OXM concentration after the first hour of reaction.

An interesting observation is that GAC and GACH-600 produced similar TOC reduction results but different by-products (no OXM detected with the former), suggesting that these catalysts may produce different ROS that could induce different CBZ degradation routes.

The main by-product identified from IBF degradation was OXL, as shown in Figure S8. Other by-products detected were FA and pyruvic acid (PYR). FA was detected in all the catalysts evaluated, while PYR was detected in low concentrations ($<0.55 \text{ mg L}^{-1}$) in the catalysts AC, ACH-400, ACH-600, ACH-900, ACM, GACH-400, GACH-600 and GACH-900. These results are in agreement with those reported by Almeida et al. (2022), which show the formation of OXL and PYR as by-products of IBF degradation [49]. Orge et al. (2024) also reported the formation of OXL as a by-product of catalytic ozonation with activated carbon [14]. The intermediate detection of PYR suggests the initial cleavage of the propionic acid side chain. Meanwhile, the formation of FA is attributed to the progressive demethylation of the isobutyl chain, as well as to the decarboxylation of PYR. The breakdown of the central benzene ring, together with the advanced oxidation of the aliphatic fragments, leads to the production of OXL.

OXL formation in the first minutes was low compared to OXL formation in CBZ degradation and LTG degradation. However, 120 min into the reaction, the OXL concentration reached very high levels for specific catalysts (ACH-600 and ACM). However, this concentration is reduced at the end of the reaction, which is more evident for ACH-600. The high OXL formation by ACH-600 and ACM coincides with the high IBF mineralization achieved by these two catalysts. When ACH-600 is used, OXL accounts for approximately 50% of the final TOC, whereas with ACM, it accounts for approximately 81%.

The most outstanding catalysts in the mineralization of the three pollutants in the individual tests were ACH-600 and ACM. ACH-600 obtained a TOC reduction of 66.5%, 60.9% and 52.1% for LTG, CBZ and LTG, respectively. Using the ACM, TOC reductions were 60.0%, 46.9%, and 56.2% for the same pollutants. In the case of ACM, the good performance could be attributed to N doping, which stimulates electron transfer from

adjacent C atoms due to the higher electronegativity of N compared to C (3.04 vs. 2.55) [50]. On the other hand, the basicity of ACM ($\text{pH}_{\text{pzc}} = 8.3$) could also favour the reaction with ozone [51].

3.4. Adsorption and Removal of Organic Compounds

Figure 4 shows the adsorption capacity of ACH-600 and ACM for the three pharmaceutical compounds for 15 min. Although both catalysts adsorb pollutants, the major contribution to their removal is through reaction with ozone. ACH-600 has the highest adsorption capacity for all three compounds, which may be related to its higher S_{BET} and S_{meso} .

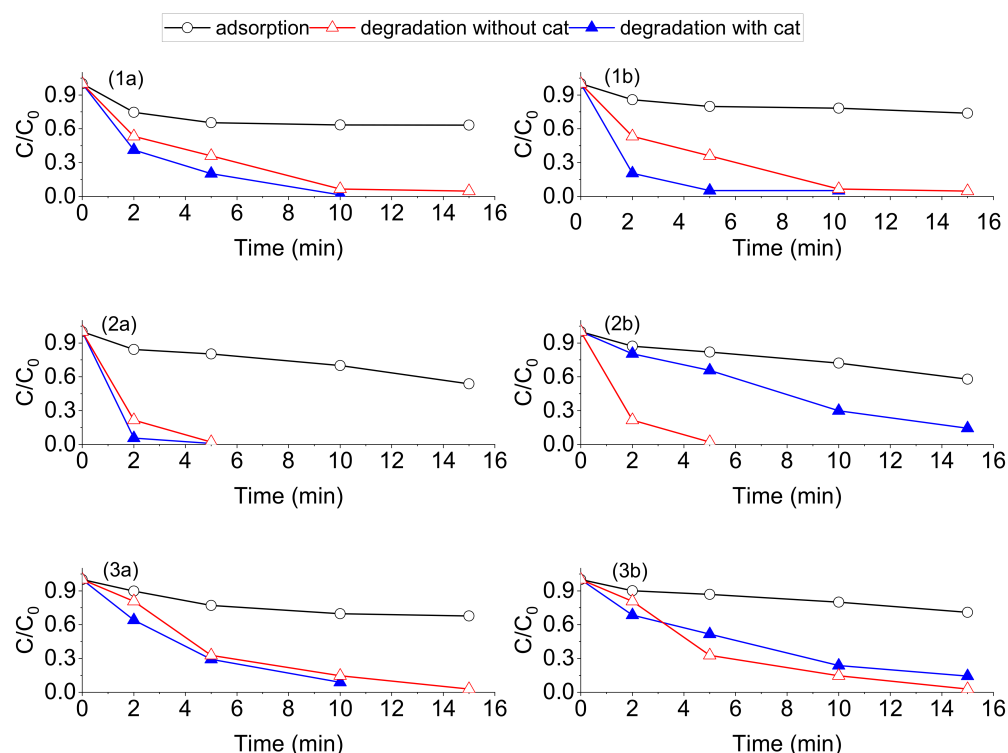


Figure 4. Adsorption and degradation of (1) LTG, (2) CBZ and (3) IBF using (a) ACH-600 and (b) ACM. Experimental conditions: $[\text{O}_3] = 50 \text{ g Nm}^3$, $[\text{Pollutant}] = 10 \text{ mg L}^{-1}$, catalyst dose = 0.5 g L^{-1} , O_3 flow rate = $150 \text{ cm}^3 \text{ min}^{-1}$, stirring = 400 rpm , reaction time = 15 min. For the adsorption tests, oxygen (O_2) was used instead of O_3 under the same conditions.

CBZ is the compound that is most adsorbed on activated carbons (46.3% for ACH-600 and 42.2% for ACM). These results align with a study by Dittmann et al. (2022), where different activated carbons removed over 80% of CBZ even under competitive conditions [52]. Delgado et al. (2019) also observed that 90% of CBZ was adsorbed onto powdered activated carbon after 10 h, and that higher CBZ adsorption was associated with a higher surface area [53].

The adsorption of organic compounds is influenced by the polarity of the compounds, the surface properties of the catalyst, and the charge of the compound, which depends on the pH of the solution [54]. At the start of the reaction, the initial solutions of LTG, CBZ, and IBF had pH values of 5.8, 5.2, and 5.6, respectively. CBZ in this solution remains neutral, and the adsorbents acquire a positive charge because $\text{pH} < \text{pH}_{\text{pzc}}$ (pH_{pzc} of ACH-600 = 7.0 and pH_{pzc} of ACM = 8.3), i.e., there is no high electrostatic attraction. Therefore, the adsorption of CBZ can be explained by π - π interactions and the hydrophobic effect. The adsorption of IBF could be due to electrostatic forces, since IBF obtains a negative charge because $\text{pH} > \text{pKa}$ and the ACs would get a positive charge because $\text{pH} < \text{pH}_{\text{pzc}}$. LTG is

a weak base that remains predominantly neutral under these pH conditions, while ACs are positively charged; like CBZ, the adsorption of this molecule is explained by π - π interactions and the hydrophobic effect.

Figure 5 compares the destruction efficiency of the compounds versus the removal of contaminants by adsorption and simple ozonation at $t = 2$ min. The destruction efficiency (%) was calculated using Equation (1) [55].

$$\text{Destruction efficiency (\%)} = \left[\frac{Mb - (Ma + Md)}{(Mb - Ma)} \right] \times 100 \quad (1)$$

where Md represents the amount of contaminant adsorbed, Mb is the amount of contaminant at the start of the catalytic reaction, and Ma is the amount of contaminant at the end of the catalytic reaction.

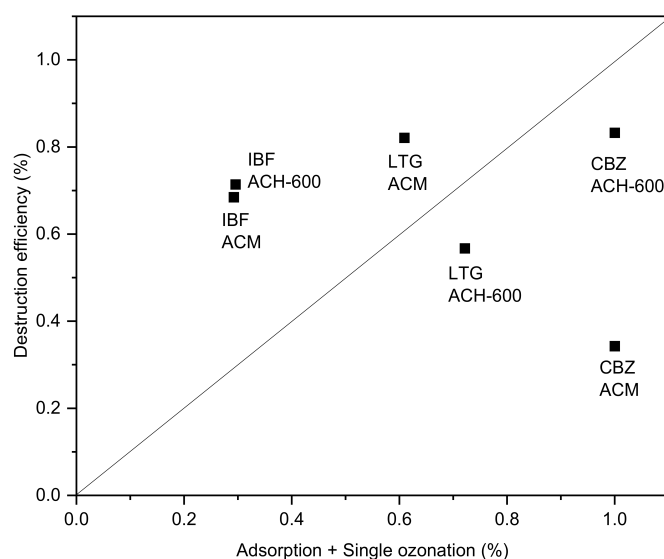


Figure 5. Comparison between elimination in 2 min by synergistic effect (adsorption + single ozonation) versus destruction efficiency.

The degradation of IBF with both catalysts, and that of LTG with ACM, yielded better results than the sum of adsorption and simple ozonation. In contrast, CBZ with both catalysts and LTG with ACH-600 showed better results, reflecting the synergistic effects of adsorption and simple ozonation.

In the particular case of CBZ, single ozonation manages to eliminate 96% of the compound in that time ($t = 2$ min), which reinforces the suggestion by Asghar et al. (2023) and Aguilar et al. (2020) that decomposition of this particular pharmaceutical with ozone is faster than that with hydroxyl radicals [15,47].

Figure 6 shows S_{BET} as a function of the destruction efficiency (%) of the three compounds. CBZ is the compound most influenced by higher S_{BET} ; this suggests that its removal is governed by the availability of active sites and surface mass-transfer phenomena. The destruction efficiency of IBF (%) is slightly higher when ACH-600 is used, which may be related to the fact that its degradation is primarily controlled by radical reactions in the homogeneous phase; the higher reaction rate constant of ACH-600 is due to the removal of IBF by catalytic ozonation, and also because ACH-600 adsorbs more IBF than ACM. On the other hand, the destruction efficiency of LTG does not improve with higher S_{BET} . For LTG, ACM has the highest destruction efficiency (%), indicating that this catalyst contributes more to LTG removal via catalytic ozonation than ACH-600. This decrease in performance with ACH-600 suggests that, beyond the development of surface area, the

selectivity of surface functional groups is the determining factor in the decomposition of LTG.

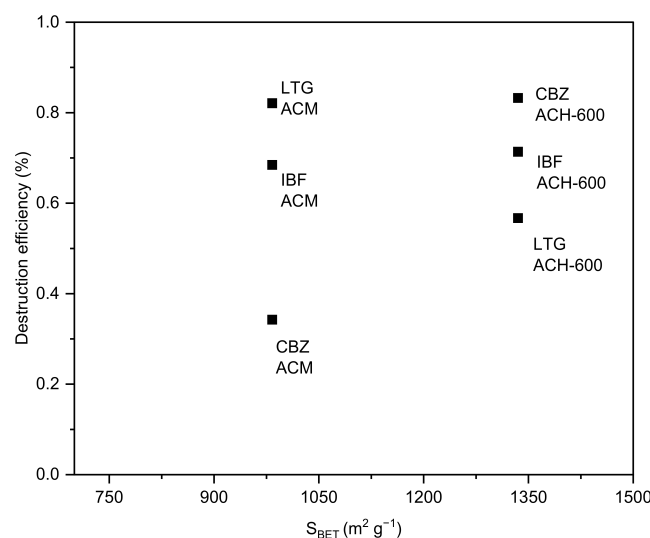


Figure 6. Influence of S_{BET} on the destruction efficiency (%) of LTG, CBZ and IBF.

3.5. Catalytic Ozonation of the Pollutant Mixture

The catalysts ACH-600 and ACM were chosen to evaluate their catalytic activity in the pharmaceutical mixture as these revealed superior performances in the individual mineralization of LTG, CBZ and IBF.

Figure 7 shows that LTG is the most recalcitrant pollutant, degrading after 15 min of reaction in tests with both catalysts. This is consistent with the results of Keen et al. (2014), who found that the reaction rate of LTG and $\cdot OH$ was among the lowest of 100 other common ECs [44]. On the other hand, CBZ is the pollutant that degrades faster in the pollutant mixture, regardless of the catalyst used (approximately 5 min). These results align with the CBZ degradation time observed in the individual degradation test using ACH-600, as shown in Figure 4(2a). However, the degradation of CBZ in the individual test using ACM took more than 15 min (see Figure 4(2b)), indicating that the mixture of pharmaceutical compounds enhanced CBZ degradation when ACM was used as the catalyst.

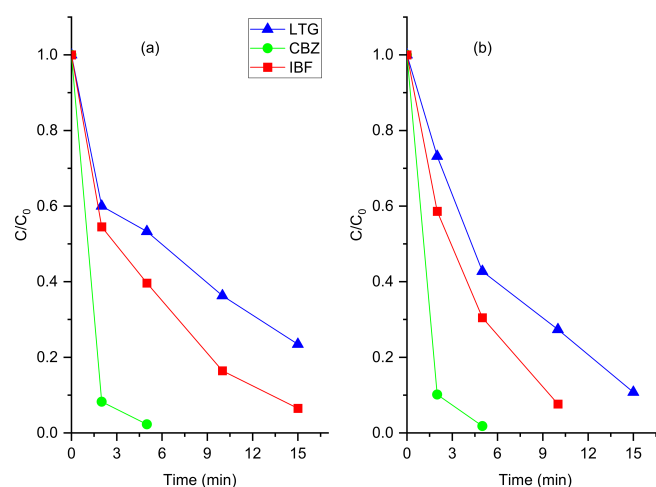


Figure 7. Degradation of pharmaceutical mixture using as catalyst: (a) ACH-600 and (b) ACM. Experimental conditions: $[O_3] = 50 \text{ g Nm}^3$, $[\text{Pollutants}] = 30 \text{ mg L}^{-1}$, catalyst dose = 0.5 g L^{-1} , O_3 flow rate = $150 \text{ cm}^3 \text{ min}^{-1}$, stirring = 400 rpm, reaction time = 180 min.

In the individual tests, ACH-600 exhibited faster degradation of IBF (see Figure 4(3a,3b)), with a first-order rate constant of 0.244 min^{-1} , whereas ACM yielded a rate constant of 0.129 min^{-1} . When tested on the mixture of the pharmaceutical compounds, IBF degradation was better with ACM, yielding a first-order rate constant of 0.255 min^{-1} , whereas with ACH-600, it was 0.175 min^{-1} . In the LTG degradation tests, the results were similar to those obtained with IBF. In the individual tests, the degradation of LTG with ACH-600 (0.419 min^{-1}) was faster than when using ACM (0.276 min^{-1}). Conversely, in the solution containing a mixture of pharmaceutical compounds, LTG degradation was faster with ACM (0.143 min^{-1}) than with ACH-600 (0.095 min^{-1}).

Since the pH of the compound mixture was 8.3, the ACM acquired a neutral charge ($\text{pH}_{\text{pzc}} = 8.3$) and the ACH-600 acquired a negative charge ($\text{pH}_{\text{pzc}} = 7.0$). The CBZ remained neutral, so adsorption could be driven mainly by hydrophobic forces and π - π interactions. Because IBF acquires a negative charge given that $\text{pH} > \text{pKa}$, the adsorption of IBF on ACH-600 is therefore hindered by electrostatic repulsion and adsorption could occur due to non-electrostatic forces. Studies conducted by Baccar et al. (2012) show that IBF adsorption is affected by more alkaline pH levels [56]. For its part, LTG in a solution with a pH of 8.3 is mainly found in its non-ionized form, so adsorption is mainly driven by hydrophobic forces and π - π interactions.

Table 3 shows the formation of nitrate (NO_3^-) and chloride (Cl^-) ions using ACH-600 and ACM as catalysts. Nitrate ions can be formed by the degradation of LTG and CBZ, because they contain N in their chemical structures. The formation of chlorides is only due to the degradation of LTG, as it is the only compound that contains chlorine (Cl) in its structure. It can be observed that, at the end of the reaction, the nitrate concentration is practically the same whether ACH-600 or ACM is used ($\sim 0.55 \text{ mg L}^{-1}$); the same is true for the chloride concentration after 180 min of reaction ($\sim 2.38 \text{ mg L}^{-1}$). The concentrations of these ions obtained after treatment with any of the catalysts comply with Portuguese and Colombian regulations for use as water intended for human consumption, aquaculture, and irrigation [57–59], as shown in Table S7. Furthermore, Montenegro-Apraez et al. (2025) [32] assessed the phytotoxicity of a mixture of pharmaceutical compounds (LTG, CBZ and IBF) before and after catalytic ozonation with ACH-600. The authors observed that the treated water improved the germination percentage (%G), seedling vigour index (SVI) and phytotoxicity index (PI) in seeds of *Lepidium sativum* and *Sorghum saccharatum*. However, in the case of *Sinapis alba*, the treated effluent negatively affected shoot and root growth [32], suggesting greater sensitivity of this species to the degradation by-products formed. This highlights the need for specific studies on the impact of degradation by-products on agricultural development, particularly regarding the reuse of treated wastewater for crop irrigation.

Table 3. Formation of NO_3^- and Cl^- in the catalytic ozonation of the mixture of pharmaceutical compounds.

	NO_3^-				Cl^-			
	t = 30	t = 60	t = 120	t = 180	t = 30	t = 60	t = 120	t = 180
ACH-600 (mg L^{-1})	0.27	0.27	0.27	0.55	1.64	1.44	1.43	2.38
ACM (mg L^{-1})	0.37	0.4	0.62	0.54	2.13	2.26	2.58	2.34

During the reactions with ACH-600 and ACM, the concentration of nitrate and chloride decreased at certain times, which could be explained by the adsorption of these ions on the surface of the activated carbons, which have a $\text{pH}_{\text{pzc}} > \text{pH}$, so that anionic species in solution have a high affinity to be adsorbed. Some studies have shown that nitrate [60–62] and chloride [63] can be efficiently adsorbed on activated carbons.

In terms of mineralization, both catalysts had TOC reduction rates greater than 76% after 180 min. Although ACM performed better during the first 60 min, ACH-600 showed a greater reduction in TOC after 180 min of reaction (see Figure S9). The apparent first-order rate constant k of ACM was higher than that of ACH-600 (see Table S8), which explains the higher mineralization rate in the first minutes of the reaction when using the ACM catalyst. The higher mineralization achieved by ACM during the first minutes also coincides with the high formation of nitrates and chlorides during this time.

The main degradation by-products of the contaminant mixture were OXL and OXM. For ACH-600, intermediate by-products such as FuA and BZQ were also detected. FuA was no longer detected after 30 min, and BZQ was not detected after 60 min of reaction. OXL accounts for 78% of the final TOC in tests conducted with ACM, whereas with ACH-600 it accounts for 39%. These results are similar to those obtained in individual tests with the three compounds. Both in individual tests and in the mixture of pharmaceutical compounds, OXL is formed and consumed as the reaction proceeds, confirming that the ACH-600 and ACM catalysts can degrade this acid. Graca et al. (2025) and López-Francés et al. (2024) identified that the degradation of OXL using activated carbon as a catalyst in a catalytic ozonation process was mainly promoted by the $^1\text{O}_2$ species [48,64].

Since the TOC removal reached values close to 80%, it can be inferred that a large fraction of the mixture was converted into CO_2 , H_2O , and inorganic species, whereas the remaining organic fraction corresponds to oxidation by-products (OXL, OXM and others). This indicates that the pharmaceutical mixture was effectively oxidized and transformed into a more innocuous solution.

Figure S9 shows the reduction in TOC over time. ACM has a higher mineralization rate in the first 60 min, which coincides with the faster degradation of pharmaceutical compounds by this catalyst compared to ACH-600. However, ACH-600 shows slightly better performance after 180 min (>80% mineralization). Previous studies have shown that ACH-600 has high stability even under real conditions, i.e., treating wastewater effluents and over consecutive reutilization cycles. For instance, after three cycles of use, this catalyst showed only a 14% loss [32].

When comparing the results with Table S1, the degradation of IBF in the pharmaceutical mixture using the ACH-600 and ACM catalysts (15 min) was similar to that reported by [11], despite the fact that a higher catalyst dose was used in our study, as was a higher organic load. On the other hand, Fe-Mn co-doped biochar [12] exhibited faster kinetics, achieving 95% IBF degradation in 9 min and 80.5% mineralization. Regarding CBZ removal, sulfur-doped graphene [15] achieved 60% mineralization in 60 min, equivalent to ACH-600 in the ozonation of the mixture over the same time interval, and was slightly outperformed by ACM at 65% (Figure S5). Although Orge et al. (2024) achieved total mineralization (~100%) of CBZ in 180 min using granular activated carbon, it is noteworthy that ACH-600 and ACM achieve mineralization of over 80% in a complex mixture of pharmaceuticals; this mixture includes LTG, which exhibits a lower reaction rate with $^{\bullet}\text{OH}$ groups compared to other common ECs [14].

4. Conclusions

Following the successful modification of commercial activated carbons through oxidation with HNO_3 , heat treatment and nitrogen doping, the ACH-600 and ACM catalysts outperformed in the mineralization of LTG, CBZ and IBF. In individual tests, the increase in the rate constant was influenced by the S_{BET} and the materials' ability to generate and adsorb ROS. The use of ACM led to high OXL formation at the end of the reaction for all three pollutants; OXL accounted for more than 70% of the final TOC. When ACH-600 is used, OXL accounts for 27–52% of the final TOC.

The results of degradation tests on individual pharmaceutical compounds differed from those obtained with the compound mixture, using ACH-600 and ACM as catalysts. In the individual tests, ACH-600 performed better. However, when the test was performed with the compound mixture, ACM degraded faster. The high mineralization of the mixture when using ACM in the first 60 min could be related to this rapid degradation.

The characteristics of activated carbons influence the degradation and mineralization of organic compounds. Activated carbons with higher N content and fewer surface oxygenated acid groups improve the mineralization of contaminants. Adsorption, meanwhile, is favoured by an increase in S_{BET} .

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/w18151892/s1>, Table S1: Results of some studies on carbon-based materials as catalysts; Table S2: Elemental analysis and pH_{pzc} of the activated carbons; Table S3: Thermogravimetric analysis—TGA; Table S4: First-order apparent rate constants for LTG, CBZ and IBF; Table S5: Adsorption tests of LTG, CBZ and IBF using AC, GAC, ACH-600 and ACM; Table S6: TOC reduction of LTG, CBZ, and IBF in individual tests; Table S7: Portuguese and Colombian regulations on NO_3^- and Cl^- ions present in water for different uses; Table S8: First-order apparent rate constant for the mixture of pharmaceutical compounds; Figure S1: N_2 adsorption/desorption isotherms at -196°C for (a) modified activated carbon AC and (b) modified activated carbon GAC; Figure S2: Scanning electron microscopy (SEM) images of ACH-400 (a,b), ACH-600 (c,d), and ACM (e,f); Figure S3: EDS analysis of ACH-400; Figure S4: EDS analysis of ACH-600; Figure S5: EDS analysis of ACM; Figure S6: Formation of (1) OXL and (2) OXM using (a) modified activated carbon AC and (b) modified activated carbon GAC during LTG degradation; Figure S7: Formation of (1) OXL and (2) OXM using (a) modified activated carbon AC and (b) modified activated carbon GAC during CBZ degradation; Figure S8: Formation of OXL using (a) modified activated carbon AC and (b) modified activated carbon GAC during IBF degradation; Figure S9: Evolution of TOC during catalytic processes of the pollutant's mixture, using ACM and ACH-600 as catalysts. Refs. [10–16] are cited in the Supplementary Materials.

Author Contributions: Investigation, D.M.-A.; validation, D.M.-A.; methodology, C.A.L.G. and O.S.G.P.S.; conceptualization, C.A.L.G. and O.S.G.P.S.; writing—original draft, D.M.-A.; writing—review and editing, D.M.-A., C.A.L.G., F.M.-M. and O.S.G.P.S.; supervision, F.M.-M. and O.S.G.P.S.; funding acquisition, F.M.-M. and O.S.G.P.S. All authors have read and agreed to the published version of the manuscript.

Funding: The authors gratefully acknowledge MINCIENCIAS COLOMBIA and Universidad del Valle (Colombia) for funding through the Proyecto FotoLIX 81829 CI 1252 and SGR (Colombia) Project NANOMAT BPIN 2024000100089. This work was financially supported by Fundação para a Ciência e a Tecnologia, I.P./MECI through national funds: LSRE-LCM, UID/50020/2025 (<https://doi.org/10.54499/UID/50020/2025>); and ALiCE, LA/P/0045/2020 (<https://doi.org/10.54499/LA/P/0045/2020>).

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding authors.

Conflicts of Interest: The authors declare no conflicts of interest.

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