# A comprehensive review on catalytic ozonation: emerging trends and future perspectives

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#### **ABSTRACT**

The push for advanced water treatment techniques has led to significant growth in research focusing on ozonation catalysis, aiming to enhance pollutant degradation efficiency and energy conservation. Conventional ozonation methods, although practical, face limitations in specific pollutant degradation and require excessive energy inputs. Introducing catalysis to ozonation can potentially overcome these challenges, driving the research frontier toward innovative catalyst materials and optimized procedures. This review delves deep into the fundamentals of catalytic ozonation, emphasizing its advantages and critical role in advanced water treatment solutions. To examine emerging trends, spotlighting state-of-the-art catalysts and their influence on improving ozonation outcomes. The future perspectives of catalytic ozonation are projected, highlighting its potential to revolutionize water treatment paradigms, harnessing the balance of energy efficiency and robust pollutant removal.

*Keywords:* Catalytic ozonation; Water treatment; Pollutant degradation; Catalyst materials; Advanced ozonation techniques; Energy efficiency

## **1. Introduction**

The surge in global industrial activities and rapid urbanization are significant contributors to the deterioration of water quality, leaving many ecosystems in peril. A pivotal concern in the 21st century has been the presence of emerging pollutants in water systems, many of which are resistant to conventional treatment methods [1–3]. The advancements in environmental monitoring technology have revealed that trace amounts of numerous organic pollutants can persist in treated waters, leading to the presence of these contaminants in aquatic systems. The World Health Organization and several environmental agencies globally

have expressed concern over the long-term health impacts of these pollutants, emphasizing the need for more efficient water treatment strategies [4–7].

Amidst the plethora of water treatment methodologies, ozonation stands out due to its potential to degrade a wide array of organic pollutants. However, standard ozonation processes have limitations regarding energy consumption and the complete mineralization of pollutants. Catalytic ozonation, where ozonation is paired with a catalyst, promises enhanced degradation rates and improved energy efficiencies [8–11]. Such processes involve generating potent oxidative species, making it possible to target even the most persistent contaminants. The effectiveness of this process

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largely depends on the choice of catalyst and its interaction with ozone  $(O_3)$ , emphasizing the need for extensive research in this domain.

Recent years have seen a substantial increase in the investigation of various materials as potential catalysts for ozonation, including metal oxides, carbon-based materials, and hybrid composites [12–15]. Notably, the efficiency of catalytic ozonation processes hinges on the catalyst's surface properties and ability to produce hydroxyl radicals (•OH). As research advances, there is a growing trend towards designing catalytic ozonation for specific pollutant degradation, paving the way for targeted water treatment solutions. Fig. 1 showcases the escalating number of publications focused on catalytic ozonation and wastewater treatment, indicating the growing research interest and its potential for real-world applications.

While a substantial amount of literature discusses various aspects of catalytic ozonation, this work distinguishes itself by providing a holistic and up-to-date analysis of emerging trends and future perspectives. The meticulously collate and synthesize information from recent studies, offering readers a panoramic view of the latest advancements, innovative materials, and cutting-edge applications in catalytic ozonation.

The review stands out by critically evaluating the performance of different catalysts, shedding light on their mechanisms of action, and pinpointing the factors that influence their efficacy. This paves by weaving together threads of knowledge from diverse sources to present a narrative that encapsulates the current state of affairs and charts a course for future explorations in this vibrant and ever-evolving domain.

### **2. Basic principles of catalytic ozonation**

The fundamental understanding of catalytic ozonation revolves around the combined effect of ozone decomposition and the catalytic reactions that facilitate the generation of reactive oxidative species.

#### *2.1. Fundamental reactions involved in ozonation*

At its core, catalytic ozonation enhances the traditional ozonation process by exploiting the surface of a catalyst to produce ozone decomposition and reactive oxidative species, most notably hydroxyl radicals [16–18]. Unlike conventional ozonation, where ozone  $(O_3)$  primarily acts as the oxidizing agent, the presence of a catalyst in catalytic ozonation promotes the quick decomposition of ozone into more potent oxidants [19,20].

When ozone comes into contact with water, it can undergo decomposition, resulting in the generation of hydroxyl radicals:

$$
O_3 + H_2O \rightarrow 2^\circ OH + O_2 \tag{1}
$$

However, this reaction is considerably enhanced in the presence of a catalyst, which can provide active sites to promote ozone decomposition [17,21,22]. The following reaction can describe the formation of hydroxyl radicals on a catalyst's surface:

$$
O_3 + \text{Catalyst} \rightarrow {}^{\shortmid} \text{OH} + O_2 + \text{Catalyst}
$$
 (2)

Hydroxyl radicals, due to their high oxidation potential, can indiscriminately oxidize a broad range of organic compounds present in water. The efficiency of the catalytic ozonation process, therefore, depends not only on the activity of the catalyst but also on the interactions between the catalyst and ozone, the catalyst and pollutants, and the availability of hydroxyl radicals [23–27].

Furthermore, the physicochemical properties of the catalyst play a pivotal role in the efficacy of the process [23,28–30]. Factors like the catalyst's surface area, pore size, and the presence of active metal sites significantly impact the production of hydroxyl radicals. It is essential to ensure the catalyst possesses optimized properties for effective ozone decomposition and subsequent pollutant degradation [31–33].



Fig. 1. Yearly rise in global wastewater treatment from 2000–2022 showcases the increasing need for efficient treatment methods. (*Source*: www.sciencedirect.com).

The catalytic ozonation operates on the principle of enhanced ozone decomposition on a catalyst's surface, generating highly reactive species that can efficiently oxidize a wide range of pollutants in water.

## *2.2. Role and significance of catalysts in ozonation*

The effective degradation of pollutants during the ozonation process requires the efficient generation and utilization of reactive oxidative species, particularly hydroxyl radicals. Catalysts play an instrumental role in catalytic ozonation by aiding in the decomposition of ozone to produce these reactive species, enhancing the overall oxidative strength of the process [31,34,35]. Different types of catalysts, depending on their mode of action and nature, have been employed to maximize the efficacy of the ozonation process.

#### *2.2.1. Homogeneous catalysts*

Homogeneous catalysts in catalytic ozonation are typically in the same phase as the pollutants and ozone. This group primarily consists of metal salt solutions such as ferrous or ferric salts, copper ions, and other transition metal ions [36,37]. Ferrous ions (Fe<sup>2+</sup>), in particular, have gained attention because they can react with ozone, forming hydroxyl radicals through the Fenton reaction:

$$
Fe^{2+} + H_2O_2 \to Fe^{3+} + OH^- + {}^{*}OH
$$
 (3)

The main advantages of homogeneous catalysts include rapid reaction rates due to the molecular-level dispersion of the catalyst in the solution. However, one significant challenge associated with homogeneous catalysts is their separation and recovery after ozonation. This limitation often necessitates additional post-treatment steps, potentially escalating costs and complexity.

Moreover, the effectiveness of homogeneous catalysts can vary depending on the conditions, such as pH, temperature, and concentration of the catalyst. For instance, the Fenton reaction is highly pH-dependent, with optimal activity typically observed in slightly acidic conditions [38,39]. While using metal ions as homogeneous catalysts can lead to increased generation of hydroxyl radicals, there is also a risk of secondary pollution due to the residual metal ions left in the treated water.

Recently, research has been directed towards developing advanced homogeneous catalytic systems that are both efficient in ozone decomposition and easy to recover. Incorporating ligands, chelating agents, and other enhancers can maximize the benefits of homogeneous catalysts while minimizing their drawbacks [40,41].

While homogeneous catalysis offers an intriguing avenue for enhancing catalytic ozonation, future developments must ensure sustainable and eco-friendly practices considering this approach's challenges and potential risks.

### *2.2.2. Heterogeneous catalysts*

#### *2.2.2.1. Metal oxide-based catalysts*

Metal oxides are an integral class of heterogeneous catalysts in catalytic ozonation processes, primarily due to their ability to generate hydroxyl radicals upon interaction with ozone. Their inherent characteristics, such as high surface area, thermal stability, and various oxidation states, make them viable candidates. The fundamental mechanism underlying metal oxide-based catalysts in ozonation is the transfer of electrons between the ozone molecules and the metal sites on the oxide surfaces. This interaction fosters ozone decomposition into more reactive species that can facilitate the degradation of various pollutants. However, while metal oxides like manganese dioxide ( $\text{MnO}_2$ ),  $\text{Fe}_2\text{O}_3$ , and copper(II) oxide (CuO) show promising activity, challenges like leaching of metal ions into the treated water and deactivation over extended cycles can hamper their broader application. Recent advancements have explored developing mixed metal or doping oxides with other elements to enhance stability and improve their catalytic performance [42–47].

## *2.2.2.2. Carbon-based catalysts*

Carbon materials mainly activated carbon (AC), carbon nanotubes (CNT), and graphene, have garnered significant interest as heterogeneous catalysts for catalytic ozonation. Given their expansive surface areas and modifiable functional groups, these materials can adsorb ozone and pollutants, positioning them close and accelerating the reaction rate. The primary mechanism here is the adsorption and subsequent decomposition of ozone on the carbon surface, leading to the generation of reactive oxygen species, particularly hydroxyl radicals. To further augment their catalytic activities, modifications such as doping carbon materials with other elements (e.g., boron, nitrogen) or metal nanoparticles have been explored [48–51]. While carbon-based catalysts are cost-effective and feature high stability, potential challenges include pore blockage due to the accumulation of intermediates and decreased activity over time. Recent research is directed towards designing hierarchical porous structures and surface functionalization to overcome these limitations [52–54].

#### **3. Catalysts used in ozonation**

Since the early days of research in environmental engineering and water treatment, the role of catalysts in ozonation has been recognized and studied. Catalysts, when used with ozone, amplify the degradation rate of pollutants and reduce the amount of ozone required. These catalysts can be broadly categorized based on their origin and characteristics, and they encompass a wide range of materials, including metal oxides, carbonaceous substances, and organic polymers [55–58].

Catalysts in ozonation serve primarily as platforms where ozone decomposes to generate highly reactive radicals, especially hydroxyl radicals, which are instrumental in the degradation of various organic and inorganic contaminants. These catalysts' physical and chemical properties, such as porosity, surface area, and active sites, are critical in determining their efficiency [59–61]. Additionally, these materials have found applications beyond water treatment, like air purification and industrial waste treatment, given their potential to accelerate reactions and improve the selectivity of specific processes [62].

Metal oxide-based catalysts, such as  $\text{Fe}_2\text{O}_3$  and  $\text{MnO}_{2'}$ have emerged as a prominent choice due to their high activity and stability in ozonation processes. The structure and electronic properties of these metal oxides play a pivotal role in ozone decomposition and the subsequent generation of reactive species [25,63–65]. On the other hand, carbon-based catalysts, with their extensive surface areas and modifiable functional groups, offer an economical and robust solution, particularly in the treatment of water laden with pharmaceutical residues and dyes [13,49,66].

Furthermore, a recent study highlighted the capabilities of composite materials, which combine the beneficial properties of two or more materials, for catalytic ozonation [67–69]. Such composites often synergize the strengths of the individual components, thereby boosting the overall efficiency of the ozonation process.

Fig. 2 displays the removal efficiencies of different contaminants when treated using various catalysts. The catalysts are categorized into three types: oxide-based, carbon-based, and hybrid. Each bar represents the percentage of a specific contaminant successfully removed using a particular catalyst in catalytic ozonation processes. For instance, carbon-based catalysts seem particularly effective in removing atrazine, while oxide-based catalysts have high efficiency in removing semi-coke cooling wastewater. The hybrid catalysts, especially Fe/ozone-Ag, demonstrate versatility in treating multiple contaminants.

#### **4. Emerging catalysts and materials for catalytic ozonation**

#### *4.1. Metal oxides*

Metal oxides have recently emerged as one of the most promising materials for catalytic ozonation, primarily due to their stability, abundant nature, and significant reactive sites. Metal oxides like  $Fe<sub>2</sub>O<sub>3</sub>$ , MnO<sub>2</sub>, and CuO have exhibited remarkable performance in accelerating the decomposition of ozone to produce reactive species, facilitating the degradation of contaminants [25,63,64,70–73].

The characteristics of popular metal oxides and their applications in catalytic ozonation. The synthesis of metal oxides typically involves calcination processes and sol–gel methods [74,75]. For instance,  $Fe<sub>2</sub>O<sub>3</sub>$  can be produced by thermally treating iron-based precursors such as iron nitrate or chloride at elevated temperatures. The produced metal oxides possess varied morphologies, which can significantly impact their performance in catalytic ozonation. An example includes the synthesis of nano-structured  $TiO<sub>2</sub>$  using hydrothermal treatment, leading to higher surface areas and enhanced ozone degradation rates [76].

Table 1 provides a comprehensive overview of various catalyst types, the target contaminants they are designed to treat, and the key findings associated with their use. Spanning from metal-based catalysts to more complex compounds, the list reflects the versatility and potential of these substances in environmental remediation. The target contaminants vary from common pharmaceutical products to specific organic compounds, illustrating the broad spectrum of applications. Notably, many of these catalysts enhance the production of hydroxyl radicals, which play a crucial role in the breakdown of organic contaminants.

The performance of metal oxide catalysts in ozonation processes is closely related to their physico-chemical properties. For instance,  $MnO<sub>2</sub>$ , with its inherent redox potential, is more active in ozone decomposition than some other metal oxides, leading to higher hydroxyl radical yields [64,97–100]. Recent advancement in this domain has been the doping or co-doping of metal oxides with other metals or non-metals to tailor their electronic and structural properties, thereby enhancing their catalytic performance [68,101,102]. For example, Fe-doped  $TiO<sub>2</sub>$  showcased a higher degradation rate for organic contaminants than pure  $\rm TiO_2$  due to increased reactive sites and better electron–hole pair separation.

Adding to the growing body of literature, researchers demonstrated that nano-MgO, when impregnated with CNT and graphite, forms a composite catalyst that exhibits enhanced catalytic performance, specifically in the



Fig. 2. Comparative efficacy of catalytic ozonation methods for contaminant removal.

## Table 1





degradation of micropollutants [103]. Furthermore, catalyst design and synthesis advancements have paved the way for creating hierarchical structures integrating micro and mesoporous domains, which enhance mass transfer and provide additional active sites for ozonation.

Moreover, insights from computational and simulation studies have shed light on the adsorption and reaction mechanisms of contaminants on metal oxide surfaces during ozonation [104]. For example, molecular simulations on the interaction of ozone with  $Fe<sub>2</sub>O<sub>3</sub>$  surfaces revealed the critical role of surface hydroxyl groups in promoting the formation of reactive species.

While the application of metal-based catalysts in catalytic ozonation has showcased promising results in the effective decomposition of pollutants, it is imperative to consider the broader environmental implications of these pollutants. The target pollutants, often from industrial discharges and agricultural runoff, threaten aquatic ecosystems and human health. They can disrupt hormonal balances,

lead to the bioaccumulation of toxic substances, and degrade the quality of water resources.

In this scenario, the employment of heterogeneous catalysts represents a pivotal breakthrough. Recent advancements highlight the efficiency of heterogeneous catalysis in pollutant eradication, offering substantial benefits in catalyst stability, selectivity, and reusability [105]. Such innovations contribute to water treatment practices' enhanced sustainability and cost-effectiveness, emphasizing the necessity for further research and implementation in this field.

Due to their versatility and adaptability, metal oxides continually evolve with research directed towards optimizing their structures, improving their performance, and integrating them with other materials to address water treatment and air purification challenges. Future directions should consider the sustainable synthesis of these materials, their scalability for real-world applications, and the exploration of novel metal oxides or hybrid systems that could offer superior performance in catalytic ozonation [55,106,107].

## *4.1.1. Practical challenges and considerations in catalytic ozonation*

While catalytic ozonation has shown significant promise in laboratory and field demonstrations, it is imperative to consider the practical challenges associated with scaling up and real-world applications.

- *Energy consumption:* One of the primary concerns in catalytic ozonation is the energy demand. Efficient ozonation processes require optimized conditions to ensure that ozone generation and consumption are balanced. This optimization directly affects energy consumption. Studies such as those on the removal of 1,3-adamantane dicarboxylic acid using carbon xerogels [108] and atrazine with MnO*<sup>x</sup>* /biochar and FeO*<sup>x</sup>* /biochar [109] have shown that while catalytic ozonation can be more energy-efficient than conventional ozonation in specific settings, energy requirements can vary based on the nature of the wastewater, the type of catalyst used, and operational parameters.
- *Reactor design*: Robust reactor design is pivotal for the effectiveness of the catalytic ozonation process. The design must ensure effective contact between ozone, wastewater, and the catalyst while minimizing ozone off-gassing. Factors such as reactor geometry, mixing, and flow regime play a crucial role in determining the efficiency of ozone utilization and overall treatment performance. For example, biologically pretreated semi-coking wastewater using spinel-type MnFe<sub>2</sub>O<sub>4</sub> magnetic

nanoparticles [110] or the treatment of nitrobenzene using Mn-Fe/ZSM-5 [111] both emphasize the importance of reactor design.

*Nanoparticle recovery*: The use of nanoparticles as catalysts in ozonation brings forth challenges related to their recovery and reuse. Nanoparticles can be lost during the process or undergo agglomeration, affecting their catalytic efficiency. Technologies such as membrane filtration, magnetic separation, or flocculation can be employed for nanoparticle recovery, but each has advantages and limitations. Recent studies on dibutyl phthalate (DBP) degradation using the  $O_{3}/ZnO/ACs$  system [112] and sulfamethoxazole degradation with  $Fe_{3}O_{4}/$  $Co<sub>3</sub>O<sub>4</sub>$  composites [113] highlight these challenges.

Understanding and addressing these challenges is crucial for successfully implementing and scaling up catalytic ozonation processes. As the field moves forward, ongoing research and development efforts aim to optimize these aspects, ensuring that catalytic ozonation remains an effective, efficient, and sustainable option for water and wastewater treatment (Table 2).

#### *4.2. Carbon-based catalysts*

The advent of carbon materials has consistently attracted researchers due to their distinct characteristics, including high surface area, tunable pore structures, excellent electrical conductivity, and remarkable stability, making them intriguing candidates for catalytic ozonation processes.

Table 2

Contaminant type	Catalyst material	Operational parameters	Removal efficiency	References
1,3-Adamantane dicarboxylic acid	Carbon xerogels prepared at pH 5.5 (CX5.5)	$O3$ dosage: 30 mg/L; pH: 8; contact time: 15 min	65%	[108]
Atrazine	MnO <sub>v</sub> /biochar (20 mg/L), FeO <sub>y</sub> /biochar (20 mg/L)	$O_3$ dosage: 2.5 mg/L; pH: 7; contact time: 30 min	83% (MnO,/biochar), 100% (FeO /biochar)	[109]
Atrazine	MnCe oxide	O <sub>3</sub> dosage: 0.8 mg/min; pH: 7; contact time: 40 min	99.99%	$[110]$
Biologically pretreated semi-coking wastewater	Spinel-type MnFe <sub>2</sub> O <sub>4</sub> mag- netic nanoparticles	O <sub>3</sub> dosage: 1.2 mg/min; pH: 7; contact time: 70 min	85.2% (COD), 94.1% (VP)	[111]
Dibutyl phthalate (DBP)	O <sub>2</sub> /ZnO/ACs system	$O3$ dosage: 15 mg/L; pH: 4; contact time: 80 min	99.04% DBP degradation	[112]
Enrofloxacin (EFC), tylosin tartrate (TT), COD, BOD <sub>5</sub> , turbidity	Ni-Co-zeolite 5 Å catalyst, catalyst dose = $10 g/L$	$O_3$ dosage: 1.1 mg/min; $pH: 7.1 \pm 0.2$ ; contact time: $30 \text{ min}$	EFC: 97%, TT: 98%, COD: 90%, BOD <sub>5</sub> : 86%, turbidity: 93%	$[82]$
Fischer-Tropsch synthesis wastewater	Fe/Mn@CH (50 g/300 mL)	$O_3$ dosage: 6 g/h; pH: 9; contact time: 60 min	64.37% (COD)	$[113]$
Nitrobenzene (NB)	Mn-Fe/ZSM-5	O <sub>3</sub> dosage: 25 mg/L initial; pH: 6; contact time: 40 min	72% Removal rate of TOC, 33.5% higher than that in the bubbling reactor (BR)	[114]
p-CBA	$Co(II)$ , Fe(II); 1 mg/L	$O_3$ dosage: 2 mg/L; pH: 7-8; contact time: 3 min	95.5% (O <sub>2</sub> /Co(II)), 92.5% $(O_2/Fe(II))$	[115]
Sulfamethoxazole	$\text{Fe}_3\text{O}_4/\text{Co}_3\text{O}_4$ composites, $0.10$ g/L	O <sub>3</sub> dosage: 6.0 mg/min; pH: 5.1; contact time: 60 min	60% TOC removal	$[116]$

Removal efficiencies during catalytic ozonation

Unlike traditional catalysts, carbon-based materials, specifically AC, graphene, and CNT, boast easy synthesis and offer unique physicochemical properties [117–124].

The sulfur-doped graphene catalyst enhance ozonation [121]. Its functionalized oxygenated groups and vast surface area were deployed to accelerate ozone decomposition and generate hydroxyl radicals. A systematic study suggested that oxygen functionalities facilitated the generation of active sites, which in turn showed superior ozonation rates compared to bare graphene.

Fig. 2 depicts a structural representation of sulfur-doped graphene, emphasizing its unique oxygen functionalities. The sulfur-doped graphene structure is illustrated on the left, with sulfur atoms represented in yellow. Interaction with  $O_3$  generates HO<sup>•</sup>, which, in turn, contributes to the transformation of carbamazepine (CBZ) into its breakdown products.

Due to hollow cylindrical nanostructures and enhanced surface area, CNTs showcased a proclivity to activate ozone more effectively than other carbon materials. The study concluded that intrinsic metallic impurities in CNTs might act as active sites for ozone decomposition, thus accentuating the production of hydroxyl radicals [122].

Recent developments in the field of carbon-based heterogeneous catalysts for environmental remediation have highlighted the effectiveness of mesoporous graphitic carbon nitride (mpg- $C_3N_4$ ) and its composites. In particular, the sonocatalytic removal of methylene blue from water has been achieved using cobalt ferrite/mesoporous graphitic carbon nitride ( $\text{CoFe}_2\text{O}_4/\text{mpg-C}_3\text{N}_4$ ) nanocomposites, with the process parameters optimized through a response surface methodology approach [125]. A novel mpg- $C_{3}N_{4}/$ Ag/ZnO nanowire/Zn photocatalyst plate was developed, demonstrating enhanced photocatalytic activity for dye pollutant degradation through a facile dip-coating process [126]. The potential of agricultural waste as a source for wastewater treatment is also gaining traction, with biochar

and green nanoparticles derived from such waste showing promise in removing refractory pollutants from water and wastewater [127]. Together, these studies underscore the versatility and efficiency of carbon-based catalysts in wastewater treatment, presenting innovative approaches for pollutant degradation.

AC derived from sustainable resources was tailored for catalytic ozonation [128]. The tailored AC exhibited superior ozone adsorption due to its high porosity and vast surface area. The mechanism suggested the involvement of carbonyl and carboxyl groups present on the surface of AC in ozone activation. The results inferred that the tailored AC showed high ozonation rates and remarkable stability and reusability in successive cycles.

In conclusion, carbon-based catalysts offer a sustainable and efficient avenue for catalytic ozonation, with each type contributing unique attributes that facilitate the generation of reactive species crucial for enhanced water treatment. Future endeavors should focus on developing carbon-based composites that amalgamate the beneficial properties of each type, ensuring maximum efficiency and economic viability in real-world applications.

#### *4.3. Zeolite-based catalysts*

Zeolite catalysts, renowned for their structured microand mesoporous architectures, have recently seen an influx in research interest for their application in catalytic ozonation processes. These catalysts offer remarkable potential for pollutant degradation due to their defined porous structures, high surface areas, and inherent acidic sites, all contributing to enhanced ozone decomposition and the production of oxidative radicals.

Fig. 3 offers detailed microscopic images of two distinct zeolite-based catalysts. Image (a) captures the morphology of ZSM-5, characterized by elongated, prism-like structures. Conversely, image (b) presents the structure of hollow zeolite,



Fig. 3. Structural representation of sulfur-doped graphene with highlighted oxygen functionalities. Reproduced with permission by the study of Asghar et al. [121].

which, as the name suggests, displays numerous uniformly sized hollow spherical formations.

Zeolite Y, known for its supercage structure, to enhance the catalytic ozonation process for removing toluene pollutants [131]. The high aluminum content and unique structure enhanced the generation of hydroxyl radicals, leading to efficient pollutant degradation. Moreover, the stability of zeolite Y in ozone-rich environments was noteworthy, with minimal leaching of aluminum ions [132].

Another zeolite catalyst, ZSM-5, was demonstrated possess intriguing characteristics that favored ozone decomposition [129]. The spatial arrangement and concentration of aluminum sites played a pivotal role in determining the catalytic activity of ZSM-5. Intriguingly, ZSM-5 with higher silicon-to-aluminum ratios showed more excellent resistance to ozone degradation, rendering them more durable during the ozonation process.

Recent advancements have looked at integrating zeolite catalysts with other materials to capitalize on synergistic effects. For instance, the composite of ZSM-5 and nanometer cerium oxide (CeO<sub>2</sub>) revealed an enhanced pollutant removal rate, where the zeolite provided active sites for ozone decomposition, and nanometer  $CeO<sub>2</sub>$  enhanced the degradation of sulfamethoxazole compounds [133].

Given their diverse structures and physicochemical properties, zeolites have emerged as potent catalysts in catalytic ozonation. Their capability to augment the generation of oxidative radicals while maintaining structural stability makes them promising candidates for sustainable water treatment technologies. Future endeavors may focus on optimizing the structural properties of zeolites and exploring novel composite materials to elevate their catalytic performances.

### *4.4. Metal–organic frameworks*

Progressive research into the potential of metal–organic frameworks (MOF) as catalysts in ozonation processes has unveiled distinct advantages over traditional materials, especially in pollutant degradation [134–136]. MOFs' inherent porosity and tunability allow for the strategic placement of metal nodes to promote interaction with ozone molecules. The MOF, Ce-MOF enhancing the decomposition of ozone to form highly reactive radicals [134]. The presence of cerium nodes in Ce-MOF significantly boosted the generation of hydroxyl radicals, furthering the catalytic ozonation process.

Recent breakthroughs with the Fe-based MOF suggest intriguing revelations. The MOF showcases an admirable performance in the ozonation of water, mainly due to its iron clusters that enhance the formation of reactive radicals [137–139]. A comparative study observed that the ozonation rate with Fe-MOF exceeded that of other common catalysts, making it an optimal choice for water treatment applications.

To summarize, MOFs' diverse structural attributes and adjustable nature have propelled them to the forefront of catalytic ozonation research. With promising preliminary results, further research into the fine-tuning of MOF structures can undoubtedly lead to more efficient and sustainable ozonation processes in the future.

#### *4.5. Nano-structured catalysts*

The dynamic evolution of nanotechnology has ushered in a transformative era in the field of catalytic ozonation, with nano-structured catalysts demonstrating superior capabilities over traditional bulk catalysts. At the forefront of this innovation is the nanoparticulate realm, which optimizes catalytic performance through a significant increase in surface area and the potential for multifunctional synergism [124,140–143].

Nano-sized  $\text{CeO}_2$  catalyst enhanced ozonation capabilities [144]. The nanostructure, attributed to the increased density of reactive sites and unique redox properties of Ce<sup>3+</sup>/ Ce4+, facilitated faster ozone decomposition and generation of reactive oxygen species, thereby heightening pollutant degradation efficiency.

In another paradigm-shifting work developed iron oxide  $(Fe<sub>3</sub>O<sub>4</sub>)$  nanoparticles coated with AC for ozonation processes [145]. The hybrid nano-catalyst showcased efficient pollutant removal due to enhanced surface interactions and offered easy magnetic separation post ozonation, underscoring the significance of nanotechnological integration.

Transitioning from metal oxides, noble metal-based nano-catalysts, particularly palladium (Pd), have come into focus. The synthesis of Pd nanoparticles supported on ceria, revealing a synergistic effect that magnified ozone decomposition rates, setting a new benchmark in the catalytic ozonation domain [146]. The nano-architecture was conducive to electron transfer, accelerating the formation of hydroxyl radicals.

Titanium dioxide  $(TiO_2)$  nanotubes surfaced as an excellent catalyst for ozonation processes [147]. The tubular structure, offering an increased surface area and enhanced light absorption, played a pivotal role in the rapid degradation of organic pollutants. The effectiveness of the tubular structure was further accentuated when doped with elements like ZnO or Ag and Pt, offering a more comprehensive range of photocatalytic activation [148,149].

Developing nano-structured MOF and covalent organic frameworks offers exciting avenues. Their porous structure and tunable functionality render them ideal candidates for ozonation catalysts, spotlighting the Ce-based nano-MOFs exhibiting rapid ozone decomposition and pollutant removal [150].

In essence, the infusion of nanotechnology in catalytic ozonation has precipitated a paradigm shift, with nano-structured catalysts signposting the future trajectory. Their superior capabilities, underpinned by the unique physicochemical properties and synergetic effects at the nanoscale, ensure they are at the vanguard of future research and applications in ozone-based water treatment systems.

#### *4.6. Bio-based catalysts*

Bio-based catalysts have begun to command attention in exploring sustainable solutions for catalytic ozonation, particularly for their eco-friendly nature, sustainability, and potential to tap into nature's inherent catalytic capabilities. These catalysts, primarily derived from organic biomasses, showcase a promising alternative to conventional catalysts in ozonation processes, making them critical candidates for advancing environmental remediation practices.

Biosynthesis protocol has the potential for bio-synthesized multi-metal oxides with varying Fe/Mn ratios as a catalyst for ozonation [151]. The porous nature of this bio-based catalyst, combined with the high presence of functional groups, manifested in an enhanced generation of hydroxyl radicals, making it a competent medium for pollutant degradation. Such a natural derivation, originating from abundant lignocellulosic biomass, places it as a front-runner in sustainability.

Biochar-supported catalysts, which complement bio-synthesized materials, have been spotlighted for their unique physicochemical properties in ozonation using piggery biogas residue. Biochar significantly increased the ozonation rate of organic pollutants [152]. The bio-polymer matrix facilitates pollutant adsorption, while the metal sites act as active centers for ozone decomposition.

Furthermore, residues from agricultural by-products have been harnessed for catalytic applications [153,154]. When carbonized and activated, coconut shells, almond shells, olive pits, and rice husks exhibit a commendable capability for catalytic ozonation. Their high surface area and intrinsic functionalities support enhanced pollutant degradation rates, thus repurposing agricultural waste into a valuable environmental tool.

In summary, the infusion of bio-based materials in catalytic ozonation is emblematic of a broader shift towards eco-friendly and sustainable solutions. Bio-based catalysts promise enhanced ozonation efficiency and underscore the importance of harnessing nature's resources judiciously. As research continues to evolve in this domain, these catalysts stand to redefine the standards of sustainable environmental remediation.

## *4.6.1. Utilization of biomass-derived adsorbents for pollutant removal*

Degradation of pollutant include catalytic ozonation is adsorption catalytic. Using a catalyst with adsorption capacity will enhance pollutant removal and sustainability when using biomass-derived adsorbents. Recent advancements in the field have demonstrated the potential of mesoporous-activated carbons and composites derived from waste biomass for the adsorption of various pollutants [155–157].

Biomass-derived catalysts and adsorbents have attracted significant attention due to their sustainable and ecofriendly nature. The mesoporous zeolite–activated carbon composite prepared from oil palm ash [158] and the mesoporous-activated carbon prepared from chitosan flakes [159] are two exemplary biomass-derived adsorbents that have shown promising results for methylene blue removal. These materials are used for agricultural and industrial wastes and exhibit superior adsorption capacities compared to similar adsorbents. The composite outperforms many existing adsorbents and provides a low-cost alternative for dye removal [158].

When comparing these biomass-derived adsorbents with other studies, it is apparent that they have advantages in terms of cost-effectiveness and efficiency. The activated carbon prepared from rattan furniture wastes [160] and Karanja fruit hulls [161] *via* NaOH activation also exhibited

impressive adsorption abilities towards methylene blue, showcasing the potential of biomass wastes as precursors for high-performance adsorbents [160,161]. However, it must be noted that the performance of these materials can be influenced by operational conditions, such as temperature, pH, and initial dye concentration, necessitating a comprehensive understanding of their adsorption mechanisms for optimization.

Despite the promising results, there are limitations and drawbacks associated with these materials. Preparing these adsorbents often involves chemical activation processes, which may pose environmental and safety concerns. These materials' long-term stability and reusability need further investigation to ascertain their practical applicability. In this regard, the work on single-step pyrolysis of phosphoric acid-activated chitin for efficient adsorption of cephalexin antibiotic provides an alternative pathway, showcasing the feasibility of using less hazardous activating agents for the preparation of high-performance adsorbents [162].

Biomass-derived catalysts and adsorbents offer a sustainable and cost-effective solution for pollution remediation. Future research could address the limitations associated with their preparation processes, explore greener activation methods, and investigate their long-term stability and reusability to promote their practical implementation in environmental remediation further.

## *4.7. Transition metal catalysts*

Transition metal catalysts, leveraging transition metals' versatile and unique electronic configurations, have emerged as pivotal components in catalytic ozonation. These metals (e.g., Mn, Co, Cu, and Fe) stand out for their diverse oxidation states and the ability to mediate redox reactions efficiently, which are instrumental in promoting ozone decomposition and generating radical species [154,163–166]. The potential to modulate their electronic and structural properties offers myriad possibilities for tuning catalytic performance [167,168].

Prominent among these, the MnO<sub>x</sub>-based catalysts have shown to possess an enhanced ozone decomposition capacity (Fig. 4). Studies revealed the efficacy of MnO<sub>x</sub>-CeO<sub>2</sub> hybrid systems where the synergistic interplay enhanced ozone conversion, leading to an augmented generation of hydroxyl radicals [169]. Furthermore, the stability of these catalysts, even at varying pH levels, provides them an edge in real-world wastewater treatment scenarios.

A composite of Fe incorporated into AC [170] showcased an increased ozone decomposition rate and effective dye degradation. The surface chemistry of these catalysts, especially the availability of active Fe sites, facilitated efficient pollutant breakdown. The catalyst's structure played a vital role, with more significant surface areas correlating with higher pollutant removal rates.

Exploring the potential of multi-metal systems, Ru-Cu bi-metallic catalyst exhibits a synergistic effect that enhances ozone decomposition and pollutant degradation [171]. This synergistic enhancement, as observed, arises from the spatial proximity of Ru and Cu active sites, promoting intra-catalyst electron transfer, thus speeding up radical generation.



Fig. 4. Microscopic images of zeolite-based catalysts: (a) zeolite Socony Mobil-5 (ZSM-5) and (b) hollow zeolite. Reproduced with permission by the study of Jiang et al. [129] and Shao et al. [132].

Another groundbreaking development is the introduction of transition metal-doped porous structures. Synthesized Mn-Cu-doped porous zeolite has remarkable ozone decomposition capabilities [172]. The porous structure facilitated more extensive pollutant access, while the embedded Mn-Cu sites acted as hotspots for radical generation. Interestingly, these catalysts' high surface area and intricate pore network could be modulated by adjusting the Mn-Cu content, resulting in varied performance characteristics.

Fig. 4 illustrates the sequential steps involved in ozone decomposition over MnO<sub>x</sub>-CeO<sub>2</sub> catalyst surfaces. The process begins with ozone adsorption onto the catalyst, influenced by electrostatic forces and hydrogen bonding (Steps 1 and 2). This is followed by ozone decomposition, releasing hydroxyl radicals and oxygen molecules (Step 3). Surface reactions yield various products, including gaseous nitrogen oxides (Step 4). The latter stages (Steps 5 and 6) highlight the regeneration of the catalyst surface and the electron transfer process, preparing the catalyst for subsequent ozone decomposition cycles.

Conclusively, transition metal catalysts, backed by their distinct electronic properties and diverse redox capabilities, are emerging as indispensable tools in catalytic ozonation. By tailoring these catalysts at the atomic and macroscopic scales, advancements are pushing the envelope of what is achievable regarding pollutant degradation efficiency. As the quest for cleaner waterways persists, these catalysts hold the promise of reshaping the trajectory of sustainable wastewater treatment strategies.

#### *4.8. Noble metal catalysts*

Noble metal catalysts, predominantly consisting of metals such as gold (Au), platinum (Pt), palladium (Pd), and silver (Ag), have garnered significant attention in the domain of catalytic ozonation due to their unparalleled electronic and structural characteristics [173–177]. Their innate ability to function as excellent electron mediators promotes swift ozone decomposition, generating reactive oxygen species crucial for pollutant degradation.

Pd and bimetallic (Pd-Cu, Cu-Pd) zeolite catalysts [178] showcased the incredible characteristic. The electronic

interactions between Pd-Cu nanoparticles and the zeolite support expedited ozone decomposition, producing hydroxyl radicals at an unprecedented rate, improved ozone conversion and increased the mineralization efficiency for complex organic contaminants.

Similarly, Pt-based catalysts, with their distinctive electronic configurations, have set new benchmarks in ozonation [179]. A comparative study, leveraging Pt's varied oxidation states, unraveled Pt's potential (IV) in enhancing the lifetime of hydroxyl radicals, consequently magnifying pollutant degradation rates.

On another front, Pd-supported zeolites, through meticulous engineering, demonstrated promising outcomes [178]. Incorporating Ag into the zeolitic structure fortified ozone decomposition pathways and introduced a multifaceted mechanism, utilizing both molecular ozone and generated radicals, thereby optimizing contaminant removal efficiencies.

In retrospect, the inclusion of noble metals into catalytic ozonation platforms has paved the way for optimizing efficiency and broadening the applicability scope. As these metals offer diverse electronic and steric modulations, their calibrated integration into tailored catalyst systems sets the stage for revolutionary advances in water treatment strategies. Notably, while their exemplary performance is undisputed, challenges tied to their economic viability and sustainable sourcing remain. Future endeavors must balance performance prowess with economic and ecological considerations.

## *4.9. Polymer-supported catalysts*

Polymer-supported catalysts have recently emerged as a potent arsenal in the toolbox of catalytic ozonation, paving the way for both versatility in application and enhanced pollutant degradation [180]. These catalysts, designed to leverage polymers' innate flexibility and adaptability, have provided avenues to address challenges like leaching and recovery issues associated with traditional catalyst systems.

Due to their design flexibility, polymer-supported catalysts can potentially overcome catalyst separation and reuse issues. Moreover, the ability to tailor the polymer

characteristics—varying its porosity, chemical functionality, and mechanical properties—offers a multifaceted approach to optimizing catalyst performance. Future trajectories should be aimed at developing stimuli-responsive polymer supports, which can modulate the catalytic activity based on real-time pollutant concentrations, pH, or other environmental parameters. Such advancements will undoubtedly elevate the realm of catalytic ozonation to new pinnacles of efficiency and adaptability.

#### *4.10. Magnetic catalysts*

The incorporation of magnetic properties into catalysts for ozone decomposition stands as a progressive stride in the domain of catalytic ozonation. Magnetic catalysts, primarily composed of ferromagnetic materials or materials exhibiting paramagnetic properties, provide the dual advantages of effective ozone degradation and hassle-free catalyst recovery using external magnetic fields [181]. The latter proves paramount in ensuring the environmental sustainability of the ozonation process by facilitating catalyst recycling and reuse.

Notably, synthesized magnetically responsive  $MnFe<sub>2</sub>O<sub>4</sub>$ nanoparticles and subsequently embedded them within a carbon microsphere matrix [182]. As elucidated in Fig. 5, this study introduces a comprehensive oxalic acid (OA) breakdown mechanism that includes OA adsorption. This mechanism offers valuable insights for further investigations into the catalytic degradation processes of small molecule acids and the quest for innovative, effective catalysts in wastewater management.

Fig. 5 showcases the proposed catalytic ozonation reaction mechanism on carbon microspheres (CMS) embedded with  $MnFe<sub>2</sub>O<sub>4</sub>$ . The process is delineated into two phases:

*Early reaction phase*: In the initial stage, ozone adsorption  $(O_3)$  takes precedence, followed by nucleophilic addition and eventual  $O_3$  decomposition. This results in forming various reactive species that partake in organic



Fig. 5. Ozone decomposition pathways over MnO<sub>x</sub>-CeO<sub>2</sub>. Reproduced with permission by the study of Meng et al. [130].

acid (OA) decomposition. The emphasis here is on a high OA concentration environment.

*Reaction's latter phase*: As the reaction progresses, the conditions shift, mirroring a lower pH and higher OA concentration. Adsorption patterns are affected by the presence of oxygen vacancies. A series of complex interactions involving nucleophilic bonding, ozone decomposition, electron transfer, and organic acid decomposition dominate this phase, releasing carbon dioxide  $(CO<sub>2</sub>)$ and other products.

Another noteworthy contribution is wherein cobalt ferrite (CoFe<sub>2</sub>O<sub>4</sub>) nanoparticles were employed as a magnetic catalyst for ozone decomposition [183]. The  $\text{CoFe}_2\text{O}_4$  catalyst exhibited superior catalytic ozonation efficiency and demonstrated a swift magnetic response, simplifying its recovery post-ozonation process.

The dawn of magnetic catalysts in catalytic ozonation promulgates a sustainable and efficient avenue to pollutant degradation. The amalgamation of high catalytic activity and magnetic recovery simplifies the ozonation protocol. Optimizing the magnetic properties with the catalyst's ozone decomposition proficiency could translate to a paradigm shift in ozone-based water and air purification methodologies.

## *4.11. Composite catalysts*

The emerging wave of composite catalysts in the field of catalytic ozonation amalgamates the best attributes of distinct catalytic materials, offering augmented catalytic performance and flexibility in reaction conditions. These composite catalysts, which capitalize on synergistic interactions, meld diverse functionalities to drive ozone decomposition effectively.

For instance, masterfully crafted a composite catalyst by integrating  $\text{CeO}_2$  with CNT for ozone decomposition [122]. The process involved impregnating AC with cerium nitrate, followed by calcination. This  $\text{CeO}_2\text{-CNT}$  composite catalyst showcased superior ozone decomposition rates compared to their counterparts. The proficiency of this composite stemmed from the enhanced redox properties and the extended surface area furnished by the symbiotic relationship between the cerium oxide particles and the porous structure of AC.

Delving further into this realm, there is potential for a bimetallic catalyst integrating Mn and Fe, Cu, Ru, and Ag onto a zeolite substrate [163]. The resultant Mn-Fe, Cu, Ru, Ag/zeolite composite was evaluated for its ozonation efficacy. The bimetallic facet of the catalyst accelerated electron transfers, facilitating rapid ozone decomposition. Moreover, the zeolite matrix offered many active sites, amplifying the overall catalytic activity.

Composite catalysts, encapsulating a consortium of active materials, herald a new dawn in catalytic ozonation. The coalescence of disparate materials creates a mosaic of functionalities, culminating in higher catalytic efficiency and versatility. Future research endeavors could potentially tailor composite catalyst structures to address specific pollutants, thereby widening the scope of catalytic ozonation in diverse environmental matrices.

## *4.12. Natural mineral-based catalysts*

The planet's abundant natural mineral resources have caught the attention of catalytic ozonation researchers, showcasing a plethora of promising properties. These minerals, predominantly alumino-silicates and metal oxides, display inherent catalytic attributes that, when harnessed, can profoundly accelerate ozone decomposition and other vital redox reactions. Predominantly non-toxic, eco-friendly, and economically viable, these natural catalysts offer a green and sustainable option in ozonation catalysis.

One prime example is clinoptilolite; natural zeolite is proficient in enhancing the ozone decomposition rate in water [184]. After pretreatment procedures, clinoptilolite manifests its catalytic potential, attributed to its specific crystalline structure, which is replete with interconnected pores and cavities.

Manganese oxide ores, directly mined from the Earth's crust, were analyzed for their ozonation capabilities [185]. The studies underscored that these naturally occurring ores exhibit an innate ability to generate hydroxyl radicals during ozone decomposition. Their high surface area and an intricate matrix of mesoporous channels play a pivotal role in their observed efficiency.

In another innovative approach, pumice stone, a porous volcanic rock, was explored as a potential catalyst for ozonation [186]. Their findings revealed that pumice, significantly when modified with metal ions, exhibited commendable ozonation performance, outshining many synthetic catalysts.

In summary, utilizing natural mineral-based catalysts offers a refreshing direction for catalytic ozonation research. These earth-abundant materials, unique morphologies, and intrinsic catalytic properties can pave the way for sustainable, efficient, cost-effective water treatment solutions. Future exploration might delve into more uncharted natural resources, emphasizing the extraction of their latent catalytic properties for enhanced ozone-based remediation techniques.

#### *4.13. Layered double hydroxides*

In the expanding realm of catalytic ozonation, layered double hydroxides (LDHs) have marked their prominence due to their unique anion-exchange capacities and adjustable physicochemical properties. These lamellar compounds, also recognized as hydrotalcite-like compounds or anionic clays, typically comprise a combination of divalent and trivalent metal cations.

The LDHs are heterogeneous catalysts in the ozonation process [187]. The resultant LDH structures, rich in specific metal combinations, showcased heightened ozone decomposition rates and hydroxyl radical generation, crucial for efficient pollutant degradation.

Fig. 6 delves into the performance of LDHs in pollutant degradation: (a) Pollutant degradation efficiency with LDHs: This graph contrasts the efficacy of Ni-Fe LDH alone, ozone  $(O_3)$  alone, and their combined effect in the pollutant decomposition process over 120 min. The synergy between ozone and Ni-Fe LDH outperforms the individual effects, showcasing a more pronounced degradation of total organic carbon over time. (b) Decomposition rates by catalyst concentration: This graph emphasizes the decomposition



Fig. 6. Suggested oxalic acid catalytic ozonation reaction process using carbon microspheres (CMS)-MnFe<sub>2</sub>O<sub>4</sub>: (a) early reaction phase and (b) reaction's latter phase. Reproduced with permission by the study of Li et al. [143].

rates of contaminants when exposed to different concentrations of Ni-Fe LDH catalysts. As the catalyst concentration escalates, a noticeable increase in the degradation rate is observed, especially within the initial 30 min.

LDH modification by integrating metal nanoparticles will enhance the catalyst's efficiency [188]. They observed that incorporating noble metals into the LDH structure facilitated electron transfer processes, thereby amplifying the production of reactive oxidative species.

LDHs' ability to intercalate anionic species also plays a pivotal role in dictating their ozonation performance. Modulate the catalytic activity, allowing for tunable pollutant degradation profiles [189].

LDH are game-changers in catalytic ozonation, providing versatility and efficiency in pollutant remediation. Their structural flexibility and the potential for further modifications hint at a prosperous future for LDHs in water and wastewater treatment applications. Continued research in



Fig. 7. (a) Relationship between layered double hydroxides' metal combinations and pollutant degradation efficiency and (b) contaminant decomposition rates using various concentration Ni<sub>3</sub>-Fe layered double hydroxides catalysts. Reproduced with permission by the study of Yang et al. [148].

this domain is pivotal for optimizing their performance and making them economically viable for large-scale applications.

**5. Summary**

In the context of ever-increasing environmental concerns, the treatment of pollutants using innovative methods, such as catalytic ozonation, has garnered significant attention. Ozonation is a robust and versatile treatment method capable of decomposing various pollutants. However, standalone ozonation can sometimes be energy-intensive and less efficient. Thus, integrating catalysts to expedite the process and increase its efficiency has led to the evolution of catalytic ozonation.

Among the myriad catalysts explored, materials like LDH, zeolites, and metal-based nanoparticles have shown immense potential in enhancing the ozonation process. These catalysts amplify the generation of reactive oxidative species, such as hydroxyl radicals, and ensure the complete breakdown of pollutants into harmless by-products.

While tremendous progress has been made in this domain, challenges persist. Some catalysts exhibit rapid deactivation over time, while others might leach harmful components into the treated water. Most catalysts' optimal operating temperature range remains below the typical wastewater temperature, necessitating either a cooling process or further catalyst modifications. Cooling, much like in the case of  $CO<sub>2</sub>$  capture, is energy-intensive and might offset the environmental benefits of the treatment process. Therefore, Future research endeavors should focus on enhancing catalysts' stability and reusability, widening their effective temperature range, and exploring cost-effective catalyst synthesis methods.

Moreover, as navigate through an era marked by rapid industrialization, the diversity of pollutants continues to increase. Such a trend necessitates continuous research to develop catalysts capable of addressing a broader spectrum of contaminants. In conclusion, catalytic ozonation emerges as a beacon of hope in wastewater treatment. As research deepens and technology advances, it is anticipated that this

method will become an industry standard, ensuring cleaner water and a safer environment for all.

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