Review Article

Advanced Materials Science Research

Graphitic Carbon Nitride is used as a Visible-Light Photocatalyst to Boost Ozonation in Water Treatment

Abstract

By either directly photolyzing ozone with UV light or employing a photocatalyst that can be excited by UV-Vis or solar light, advanced oxidation processes (AOPs) can boost ozone's efficiency.The literature on the use of ozone and the g-C3N4 photocatalyst to break down probe molecules in water, including oxalic, p-hydroxybenzoic, and oxamic acids, ciprofloxacin, and parabens, is compiled in this review.g-C3N4 is an abundant element-based metal-free visible-light photocatalyst that creates a synergistic effect with ozone.The combination of photocatalysis and ozonation is more effective than either of the two methods on their own.This synergy is thought to originate in the effective electron quenching of photogenerated conduction band electrons by O3 in the g-C3N4 photocatalyst, as demonstrated by the available data.Due to the widespread use of ozonizers in water treatment, it is suggested that using ozonizers in conjunction with g-C3N4 photocatalysis could also increase the effectiveness of ozone in the AOPs of actual waste waters.

Keywords: Carbon nitride • Organic pollutants • Ozone • Photocatalysis • Visible light

Ezeh E M*

Department of Material Science and Nano Material, Nigeria

*Author for correspondence:

mbamalu116@gmail.com

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Introduction

Advanced oxidation processes (AOPs) are well-known treatments for wastewater remediation. Ozone (O3) is used as a precursor for ROS in one of the most effective AOP treatments, and light is used to encourage O3 conversion. Oxygen or oxidizing agents in the AOP produce reactive oxygen species (ROS) by any means, including chemical, photophysical, electrochemical, or other methods.Because of O3's UV absorbency, direct irradiation necessitates artificial light from lamps, which raises overall costs.This O3-based AOP can use visible light and even natural sunlight because it uses a photocatalyst [1].

Even though the structure of g-C3N4 is similar to that of graphite, it is made up of stacked, two-dimensional sheets of s-tris triazine units that are condensed by tertiary nitrogen atoms. The interaction of these layers via van der Waals forces is the reason for the material's high chemical and thermal stability [2].The electronic structure of this material has a band gap of approximately Eg 2.7 eV, making it suitable for photodegradation of organic pollutants and heterogeneous catalysis.With energy values of 1.1 and +1.6 eV, respectively, the conduction band and the valence band are suitable for a variety of redox reactions.g-C3N4 differs from other conventional photocatalysts in a number of interesting ways, such as the fact that it is a metal-free solid made only of carbon and nitrogen, two elements that are extremely abundant in nature;It is economical [3]. It is good for the environment;It is very secure;It has a suitable alignment of the band's energy;and it responds to light that is visible.These intriguing factors sparked the creation of transition metal-free photocatalysts based on g-C3N4 for organic pollution degradation [4].

g-C3N4 preparation is always challenging, and numerous synthetic approaches have been described in the literature.A variety of g-C3N4 structures have been made using liquid-based methods by shaping and casting to get a large solid with a large surface area; however, toxic chemicals are used in these methods.In recent years, high-quality, low-defect density g-C3N4 material has also been produced through the thermal decomposition of single precursors like cyanamide, dicyanamide, melamine, thiourea, or urea. One common method for preparing g-C3N4 is the solid-state reaction of cyanuric chloride and/or calcium cyanide, lithium azide, or melamine [5].

Pure g-C3N4 has significant morphological variation because its structure contains both an acidic bond and a C-N bond.Thicker, structurally distinct g-C3N4 nanosheets can also be produced through exfoliation and treatment with inorganic acids.Additionally, transition metal carbides can be used as an active site in the photocatalytic reaction to produce thin g-C3N4 nanosheets [6].Although the bubble effect has been used to try to introduce porosity into g-C3N4, controlling the size of the pores is still difficult.g-C3N4's morphology can be easily designed and controlled for any application thanks to its photocatalytic performance, high mechanical resistance, and simple recovery [7].

As will be shown in a number of examples, g-C3N4 can absorb photons in the Vis/O3/g-C3N4 photocatalyst under visible-light irritation to produce holes (h+) on the VB and electrons (e-) on the CB.Due to the synergistic effect, visible light irradiation of g-C3N4 in the presence of O3 produces a significant amount of •OH.At pH 7, the CB potential of g-C3N4 can be as low as 0.78 V, which makes it much simpler for O3 to capture CB electrons than it is for SCE.As a direct consequence of this, an ozonide radical (•O3) is produced, which rapidly protonates into a HO3• radical in the medium. This trioxide radical easily breaks down into •OH. g-C3N4 and O3 must be combined for these equations to work.Nothing would occur in the absence of one of these reagents [8].

In light of recent advances in wastewater treatment using the AOP, the current review focuses on the AOP using graphitic carbon nitride (g-C3N4) as the photocatalyst and oxygen as the oxidizing reagent. However, readers should refer to the catalytic activity of Ag-ZnO photocatalysts or metal-doped TiO2 for the degradation of pollutants.The purpose of this review is to demonstrate that, in the absence of any precious metal or even another transition metal, g-C3N4 is a highly effective visible-light photocatalyst for activating ROS that produce O3. The most potent and destructive species of the various potential ROS are hydroxyl radicals (•OH) due to their high oxidation potential. They are also a species with strong electrophilicity and can extract hydrogen from almost any C-H bond, resulting in carbon-centered radicals.This review will show that O3 and a photocatalyst like g-C3N4 can usually be used to make high fluxes of •OH, which makes it a very good AOP treatment [9].

In order to evaluate and demonstrate the advantages of combining the g-C3N4 photocatalyst with O3 and visible light, model molecules have frequently been utilized.This review focuses on the probe that was used to show that the g-C3N4 photocatalyst and O3 worked well together.Oxylic acid (OA) and other organic pollutants that are selectively degraded by •OH are the preferred probes for demonstrating the synergy between the g-C3N4 photocatalyst and O3. The concluding section provides a summary of the primary results that have been achieved with the g-C3N4 photocatalyst and ozonation, as well as our hopes for future developments in this field [10].

Oxalic Acid (OA)

In one of the studies demonstrating the activity to generate •OH radicals, bulk g-C3N4 was produced from either thiourea (GCN-T) or dicyandiamide (GCN-D). The SEM images of the two GCN samples are provided.The photocatalytic potential of these solids was evaluated through the mineralization of OA and p-hydroxybenzoic acid (PHBA) under UV and visible light irradiation.Under visible light, ozonation and photocatalysis with g-C3N4 were found to work together.When the reaction conditions were optimized, Vis/O3/GCN-D's rate constant for OA removal was 20.6 times higher than that of ozonation and Vis/GCN-D.Vis/O3/GCN-D, on the other hand, removed 98% of the TOC from PHBA, which is about 39.3% more than the combined values of Vis/GCN-D and ozonation.Interestingly, the UV/O3/GCN-D system had a stronger oxidizing capacity than the Vis/O3/GCN-D photocatalytic system for the same intensity of OA degradation.The lower activity of UV/O3/GCN-D was hypothesized to be due to the competition between the ozone quenching of the photoinduced electrons on GCN, the sole operating process under visible-light irradiation. As a result, UV had a smaller effect on the amount of •OH produced than visible light did [11].

Ciprofloxacin

In another study, nanosheets of g-C3N4 were used as catalysts to photocatalyze the degradation of a ciprofloxacin (CIP) antibiotic in water under visible light irradiation with white light LEDs. Under ozonation conditions, the degradation of CIP was approximately 90% in 60 minutes using g-C3N4 and visible light irradiation.Other objectives of this work included determining the sequential pathway for CIP degradation with possible experimental evidence from Q-TOF-coupled high-resolution mass spectrometry and liquid chromatography and determining the intermediate byproducts produced during degradation.There were seven possible intermediates, three of which had never been reported before.Kinetic studies demonstrated that CIP degradation follows a pseudo-first order kinetics with a rate constant of 0.035 min-1.The fact that the rate constant decreased significantly to 0.00072 min-1 when triethanolamine was added indicates that CIP degradation is initiated by the catalyst's holes.Additionally, the primary pathway for CIP degradation was the attack of the piperazine ring by •OH radicals.The heterocyclic ring rupture and a series of subsequent reactions, including the loss of two carbon atoms as CO2, defluorination, oxidation, and cleavage of this intermediate's cycles, summarize the proposed CIP degradation sequence [12].

Oxamic Acid (OMA)

g-C3N4 was recently discovered to be a heterogeneous photocatalyst for the photocatalytic ozonation of OMA in aqueous solution. The bulk g-C3N4 material was thermally post-treated at 500 °C to produce g-C3N4-500, which has a larger surface area than the bulk material.The high activity of C3N4-500 is due to the combination of photoinduced charge separation and ozonation to produce a higher number of •OH radicals. The experimental data demonstrate that the photocatalytic ozonation of C3N4 was highly effective in the removal of OMA, reaching complete OMA degradation with C3N4-500 after 120 minutes of irradiation.However, when scavengers are present, the slower rate of OMA removal can be attributed to photogenerated holes on the catalyst surface, which are more important for OMA degradation than •OH radicals.Changes in the exposed C3N4-500 structure that occurred during the photocatalytic reaction were blamed for a slight decrease in degradation performance.The physicochemical properties of the solid did not change much over the course of the three cycles [13].

Parabens

In addition to the examples with visible light irradiation that were previously mentioned, it was reported that g-C3N4 could be used as a photocatalyst for the photo-assisted degradation of a number of parabens in a way that was both economical and efficient.In control experiments, UV-A radiation was found to be more effective than visible light at activating g-C3N4.When compared to the dark ozonation method, the photocatalytic ozonation process utilized a lower dosage of ozone, resulting in higher paraben degradation rates [14].The medium with basic and neutral conditions (pH = 7–11) has the highest radical generation and a better interaction between contaminants and catalysts, according to optimization studies.Under ideal reaction conditions of 500 mg L1 catalyst concentration and 1 mg L1 paraben concentration, the three parabens—methyl, ethyl, and propylparaben—were eliminated by more than 95% in less than 15 minutes.Additionally, the luminescence inhibition of Allivibrio fischeri bacteria was significantly reduced under these conditions, resulting in a solution that was both non-toxic and disinfected [15].

Conclusions and Prospects

As demonstrated in the examples that came before it, the degradation of probe molecules occurs when photocatalysis, O3, and g-C3N4 are combined.According to the available mechanistic data, this synergy comes from the higher efficiency of •OH formation with the combined g-C3N4 photocatalysis/ozonation process because O3 acts as an electron acceptor and captures photogenerated electrons in the g-C3N4 semiconductor. Activity data have demonstrated a degradation level that is significantly higher than the sum of the degradation degrees achieved independently by any of the two components.•OH generated by photosynthesis attacks g-C3N4, releasing additional pollutants into the water. However, when competing organic molecules are present, self-degradation is a minor process.Due to the absence of any transition metal, the wastewater treatment process that employs the g-C3N4 photocatalyst and ozonation appears to be straightforward and practical.Their application is also possible due to the commercially available large-scale ozonizers that have already been installed in numerous wastewater treatment plants.Combining these ozonizers with natural sunlight photocatalysis will result in a significant increase in efficiency.The objective in this area will be to use probe molecules to only confirm laboratory data in actual wastewater treatment plants.

The presence of unreacted debris and surrounding response layers made it viable to observe the kinetics of elemental diffusion all through sintering. It changed into proven that similarly to the diffusion of B and C into the matrix, there may be a selection of Al and Cr withinside the contrary route (into the B4C particle). The response layer is composed in particular of Ti, B, and a small quantity of Mo and V, which inhibit in addition diffusion of B into the matrix. The C content material of the matrix is excessive, indicating that its diffusion is now no longer mainly inhibited via way of means of the alloying factors. Hardness measurements confirmed a boom in hardness because of the reinforcement. It changed into proven that the boom in hardness outcomes in most cases from in situ nucleated stages and from a feature microstructure such as colonies of α” segment lamellas. the observe confirmed that thru the in situ response all through sintering, it’s far viable to enhance the β-titanium alloy crafted from elemental powders and that the TiB and TiC synthesis is managed via way of means of the ok addition of alloying factors.

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