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Sustainability-driven photocatalysis: oxygen-doped g-C_3N_4 for organic contaminant degradation

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Due to its distinct electrical structure and environmental compatibility, graphitic carbon nitride (g-C_3N_4) has become a viable photocatalyst for various applications. Significant initiatives are currently being implemented to enhance the photocatalytic activity of g-C_3N_4 by adding oxygen dopants to its structure. The unique characteristics of oxygen-doped g-C_3N_4 (O@g-C_3N_4), including enhanced charge carrier mobility and changed electronic structure, make it especially appealing for photocatalytic applications. The synthetic techniques used to create O@g-C_3N_4 are thoroughly examined in this paper, along with the structural changes brought on by oxygen doping and the processes underpinning its increased photocatalytic activity. The methods for adding oxygen atoms to the g-C_3N_4 lattice are covered in the synthesis section, including solid-state processes, chemical vapor deposition, hydrothermal synthesis, co-precipitation, and post-treatment procedures. Using these techniques, the type and density of oxygen functional groups may be precisely controlled, allowing O@g-C_3N_4's photocatalytic characteristics to be tailored. O@g-C_3N_4's characteristics are discussed, emphasizing its modified electronic band structure, better surface reactivity, and enhanced light absorption abilities. Recent developments in the field are also presented, exhibiting cutting-edge techniques, including heteroatom doping, nanostructuring, and co-catalyst integration that further enhance O@g-C_3N_4's photocatalytic capabilities. As a versatile photocatalyst, O@g-C_3N_4 has been extensively reviewed in this study, emphasizing its synthesis processes, structural characteristics, and current developments in improving its photocatalytic activity. Our understanding of O@g-C_3N_4 is deepened by this review's insights, which also open the door for future research into using the compound in environmentally friendly and sustainable technology.

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Sustainability spotlight

The mounting challenges caused by the rapid development of urbanization and industry drive the urgent demand for wastewater treatment. As long as these trends continue, the volume of wastewater generated will rise, bringing with it additional toxins, heavy metals, and organic contaminants. The release of untreated or inadequately treated wastewater endangers ecosystems owing to pollution, eutrophication, and public health concerns. This paper demonstrates a dedication to sustainability in materials science and environmental remediation. The focus of the investigation on oxygen-doped g-C_3N_4 (O@g-C_3N_4) yields intriguing advances with evident implications for material sustainability and ecological preservation. The importance of O@g-C_3N_4 in the development of sustainable solutions cannot be overstated. This material demonstrates its potential to reduce environmental pollution and dependency on energy-intensive, non-renewable resources by harnessing sunlight for photocatalytic breakdown of organic pollutants. Its catalytic characteristics provide an environmentally benign alternative to traditional pollution removal technologies, leading to a greener, more sustainable future. Furthermore, the synthesis strategies mentioned in this article underline the significance of resource efficiency. Researchers are leveraging on the ubiquity of carbon and nitrogen supplies by altering g-C_3N_4 with oxygen, decreasing the requirement for rare or expensive elements. This strategy is consistent with sustainability goals, emphasizing resource stewardship while providing efficient pollution removal. The structural features of O@g-C_3N_4 improve its sustainability credentials even further. The material's stability and durability provide long-term efficacy in environmental cleanup, decreasing the need for regular replacements and minimizing waste. Its chemical stability under varied climatic circumstances promotes the concept of sustainability by allowing it to perform over long periods of time. In summary, the study described in this paper on O@g-C_3N_4 constitutes a significant step toward sustainability in materials science. This material provides an eco-friendly solution for the elimination of organic pollutants by utilizing renewable energy sources, optimizing resource utilization, and boosting long-term performance. Its ability to reduce environmental harm while also contributing to a more sustainable future emphasizes its critical role in tackling global ecological concerns.

1. Introduction

The urgent need for wastewater cleanup is motivated by the growing difficulties brought on by the fast expansion of

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urbanization and industrialization.[1] As long as these trends continue, there will be an increase in the amount of wastewater produced, full of new toxins, heavy metals, and organic pollutants.[2] Ecosystems are seriously threatened by the discharge of untreated or insufficiently treated wastewater due to pollution, eutrophication, and public health issues. Adequate wastewater treatment is crucial to solving these urgent problems. Innovative technologies are being created to address the increasing complexity of wastewater compositions and to support sustainable water resource management.[3] These technologies include enhanced oxidation processes, membrane filtration, and biological treatments.[4] In addition to removing pollutants, these methods seek to conserve energy, recover priceless resources, and lessen the environmental effects of wastewater management. Researchers are making critical efforts to secure freshwater supplies, preserve ecosystems, and assure the welfare of populations worldwide by advancing wastewater cleanup.[5]

As a critical area of environmental research and technology, photodegradation is essential for reducing the harmful effects of organic pollutants and toxins on ecosystems and human health.[6,7] Advanced photocatalytic materials have drawn much attention because they provide effective and sustainable methods for removing resistant organic molecules from various environmental matrices.[8,9] As a competitor among these materials, oxygen-doped graphitic carbon nitride (O@g-C_3N_4) has shown great promise in photodegradation.[10] With a thorough examination of its synthesis, characteristics, current developments, and applications in the photodegradation of organic pollutants, this review paper digs into the complex world of O@g-C_3N_4. It gives a thorough overview of the crucial role O@g-C_3N_4 plays in tackling today's environmental concerns by illuminating the structural complexities and functional improvements made possible by oxygen doping. Additionally, this review thoroughly analyzes the most recent developments in the industry, providing insightful information on the material's advancing capabilities and its transformative potential to create a cleaner, more sustainable future.

2. Synthesis techniques for fabrication of O@g-C_3N_4

The promising material O@g-C_3N_4 has prospective uses in various industries, including photocatalysis and energy storage.[11] Oxygen doping can improve an object's electrical characteristics and performance in multiple tasks. The following are some typical techniques for producing O@g-C_3N_4:

2.1. Solid state reaction

This process includes combining an oxygen source, such as oxalic acid dihydrate, with a precursor containing carbon and nitrogen, often melamine, and heating the mixture to high temperatures in a controlled environment.[12] During the heating step, *i.e.*, up to 550 °C for 4 hours at a ramp rate of 20 °C min^{-1}, the oxygen source's oxygen is integrated into the g-C_3N_4 lattice to form O@g-C_3N_4. The fabricated O@g-C_3N_4 by Mishra *et al.*[12] was used to photodegrade methylene blue, rhodamine B, and methyl orange under visible light irradiation. Moreover, using ascorbic acid as the source, Zhang *et al.*[13] manufactured O@g-C_3N_4 and used the photocatalyst to remove chromium. As can be seen in Fig. 1a,[13] the concentration of oxygen rises in O@g-C_3N_4, and new C–O and N–C–O bond was observed compared to pristine g-C_3N_4 (Fig. 1b–d).[13]

2.2. Chemical vapor deposition (CVD)

In CVD, as carried out by Chubenko *et al.*,[14] a suitable substrate and carbon, nitrogen, and oxygen precursor gas are injected into a reaction chamber. The gas-phase precursors break down and deposit O@g-C_3N_4 onto the substrate at a regulated temperature and pressure. The oxygen doping resulted in the compression of the g-C_3N_4 lattice, which could be ascribed to the slightly nonstoichiometric composition of the material.

2.3. Hydrothermal synthesis

In this process, a precursor solution, including the carbon, nitrogen, and oxygen sources, is enclosed in a high-pressure autoclave and cooked to a high temperature. The oxygen source in this approach is often hydrogen peroxide or urea peroxide. This technique facilitates the development of O@g-C_3N_4 nanoparticles. According to Chen *et al.*,[15] the obtained g-C_3N_4 powder (1 g) was combined with 100 mL of a 30 vol% H_2O_2 solution using ultrasonic dispersion for 20 min. The mixture was then put into a Teflon-sealed autoclave and kept at 120 °C for 6 h. The finished product underwent centrifugation, washing, and 60 °C drying overnight. Fig. 1e (ref. 15) shows that the (002) diffraction peak of O@g-C_3N_4 moved from 27.3° to 27.6° and was significantly sharper, demonstrating the relationship among the adjacent layers was bolstered, and the interplanar layered distance was reduced due to oxygen atoms doping. As further evidence that the O_2 atoms are successfully incorporated into the g-C_3N_4 framework, a fresh faint peak at 1208 cm^{-1} originating from the tensile vibration of C–O is seen in the O@g-C_3N_4 sample (Fig. 1f).[15]

2.4. Co-precipitation method

This process entails the co-precipitation of metal ions with a source of carbon, nitrogen, and oxygen. After that, the precipitate is thermally processed to create oxygen-doped g-C_3N_4.[16] Solgi *et al.*[16] fabricated O@g-C_3N_4 using the same process and used oxalic acid as the oxygen source.

2.5. Ion exchange method

This process involves soaking pre-synthesized g-C_3N_4 in an oxygen-ion-containing solution, such as one containing nitrate or sulfate ions. The oxygen ions swap out some nitrogen particles in the g-C_3N_4 lattice through ion exchange, leading to oxygen doping. Researchers carried out similar work, as illustrated in Fig. 1g.[17] HNO_3 was used as the oxygen-ion-containing solution, and g-C_3N_4 was submerged and hydrothermally treated at 80 °C to form oxygen-doped g-C_3N_4.

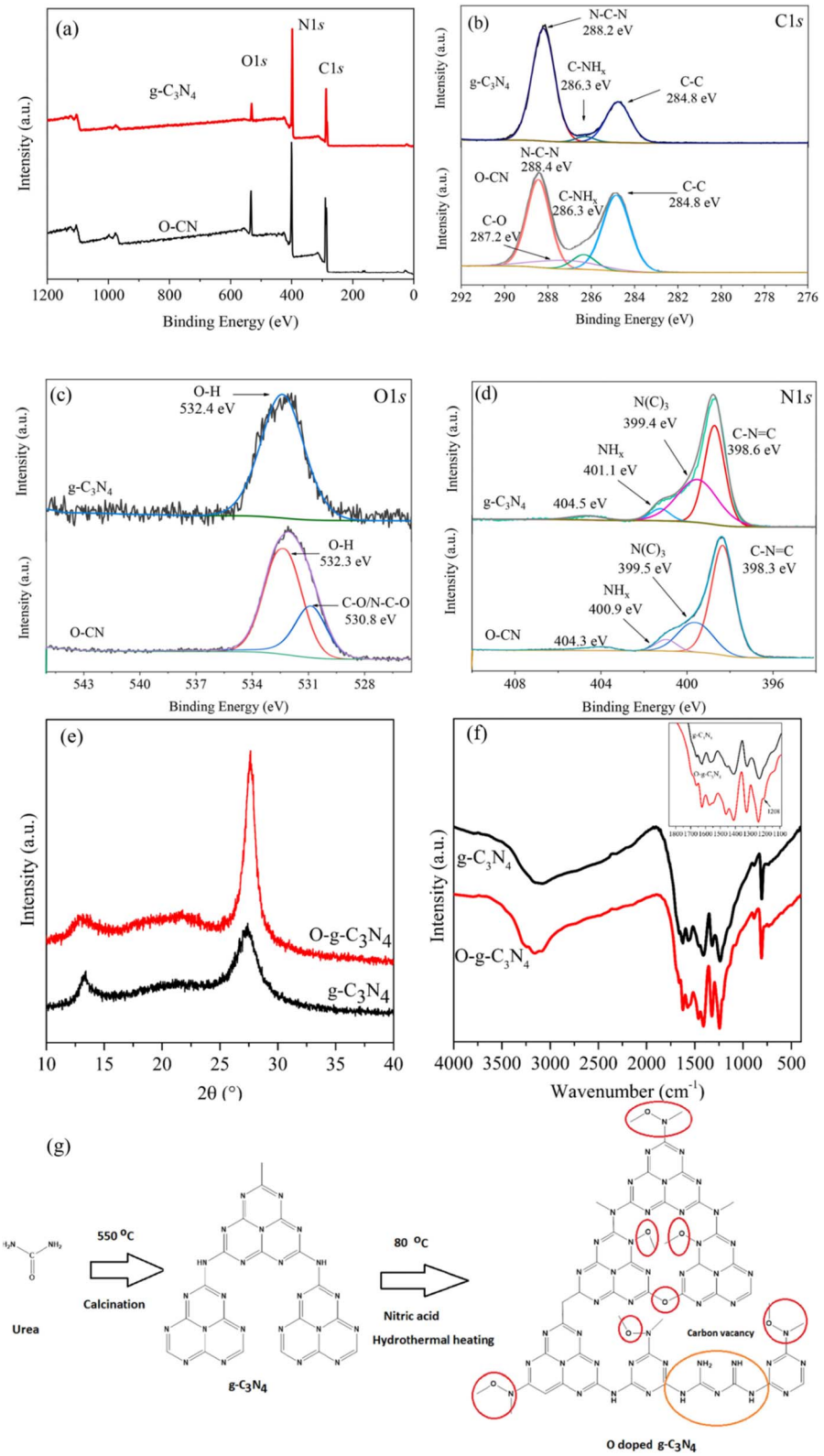

Fig. 1 (a) XPS survey spectra and short scan spectra of oxygen-doped g-C_3N_4 with elements (b) C 1s, (c) O 1s, and (d) N 1s[13]; (e) XRD and (f) FTIR spectra of fabricated oxygen-doped g-C_3N_4 (ref. 15); (g) ion exchange fabrication of oxygen-doped g-C_3N_4.[17]

The application's particular needs, the equipment and knowledge available, and the choice of synthesis process are all factors that can enhance the properties of a nanomaterial. Each technique has benefits and drawbacks, and depending on the synthesis path chosen, the characteristics of the O@g-C_3N_4 material can change. Thus, the ideal approach for the study or industrial demands should be carefully selected.

3. Structure and properties of O@g-C_3N_4

3.1. Structure of g-C_3N_4 and O@g-C_3N_4

A modified form of graphitic carbon nitride (g-C_3N_4), oxygen-doped graphitic carbon nitride (O@g-C_3N_4), is distinguished by adding oxygen atoms to the g-C_3N_4 lattice. It is crucial first to be familiar with the fundamental structure of pure g-C_3N_4 to comprehend the structure of O@g-C_3N_4. The essential structure of g-C_3N_4 characterizes it as a two-dimensional, layered substance made only of carbon (C) and nitrogen (N) atoms.[18] This structure is frequently compared to a polymer created when heptazine (C_6N_7) units are linked together by nitrogen atoms, resulting in a planar, honeycomb-like lattice resembling graphene.[19] The extraordinary lack of oxygen atoms in the structure of g-C_3N_4's pure state highlights the substance's inherent purity. The short version of its chemical formula is $(C_3N_4)_n$, where "*n*" stands for the number of repeating units that make up this polymeric structure, as can be illustrated in Fig. 2a.[20] Notably, the nitrogen atoms in g-C_3N_4 adopt a sp^2-hybridized configuration, which contributes to the lattice's planarity and gives it characteristic graphitic features.[21] As a result, g-C_3N_4 is a material of significant attention in various scientific and technical applications.[22]

The intentional inclusion of oxygen atoms into the g-C_3N_4 framework results in the structural configuration of O@g-C_3N_4. These inserted oxygen atoms take the form of oxygen-containing functional groups, such as hydroxyl (–OH), carbonyl (C=O), and carboxyl (–COOH) groups, among others. The two main methods for this oxygen doping are functional group attachment and substitutional incorporation. In the former, oxygen atoms take the place of a portion of the nitrogen atoms in the g-C_3N_4 lattice, constituting a crucial part of the material's atomic structure, as described in Fig. 2b.[23] In the latter, oxygen atoms form oxygen-rich functional groups by adhering to the edges and flaws in the g-C_3N_4 layers. Notably, the presence of these oxygen dopants significantly alters the electrical and molecular characteristics of O@g-C_3N_4. It promotes increased catalytic activity, allowing for greater reactivity in several chemical reactions.[15] Additionally, adding oxygen enhances the material's charge carrier mobility, a crucial component for its effectiveness in photocatalytic and electrical applications. Because of this, oxygen doping's structural changes play a vital role in enhancing O@g-C_3N_4's functional capabilities and transforming it into a versatile substance of significant scientific and technological value.[24]

3.2. Structural properties of O@g-C_3N_4

Several distinctive characteristics of the structural properties of O@g-C_3N_4 substantially impact its functional capabilities.

Oxygen functional groups are one of O@g-C_3N_4's most notable structural features. These functional groups, which include carboxyl (–COOH), carbonyl (C=O), and hydroxyl (–OH) groups, are thoughtfully incorporated into the material's structure as well as its surface.[25] These functional moieties are crucial in shaping O@g-C_3N_4's chemical reactivity since they make interacting with other molecules easier and enable various chemical activities.

The electrical band structure of g-C_3N_4 undergoes significant alterations due to oxygen doping. The optical and electrical characteristics of the material are critically affected by this change in the electronic environment.[12] It affects its ability to absorb light in various parts of the electromagnetic spectrum by causing absorption and emission spectra shifts. The improved photocatalytic and electronic performance of O@g-C_3N_4 is primarily due to these modifications in the electronic structure, which make it easier for effective charge separation and migration to occur during photocatalytic processes and electronic device applications, as can be observed in Fig. 2c.[15]

Another essential structural property imparted by oxygen dopants is the development of active spots on the material's surface. Centres for catalytic and chemical processes are located at these active locations. Such sites make O@g-C_3N_4 more reactive on its surface, increasing its potency for various chemical reactions.[26] Eliminating harmful organic pollutants from water and air sources is one application where this increased surface reactivity is very useful. O@g-C_3N_4 has great promise for environmental remediation by offering many sites for pollutant adsorption and degradation.[15]

The adsorption of reagents and, as a result, oxygen doping can dramatically influence the photocatalytic reactivity of g-C_3N_4. Incorporating oxygen atoms into the g-C_3N_4 lattice can result in the forming of oxygen-containing functional groups, which alters the surface characteristics and electronic structure.[27] Based on the nature of the oxygen dopants, this change can either increase or reduce reagent adsorption. For example, oxygen-containing groups may offer extra active sites for reagent adsorption, increasing total photocatalytic activity by allowing for improved interaction between the reagents and the photocatalyst.[28] Excessive oxygen doping, conversely, may produce non-reactive surface sites or modify the electronic structure so that reagent adsorption is hampered, resulting in a drop in photocatalytic reactivity. The effect of oxygen doping on reagent adsorption is critical in modifying g-C_3N_4 surface chemistry for optimum photocatalytic performance in various applications.

In general, the structural characteristics of O@g-C_3N_4 are closely related to its capabilities. The material's distinctive structural properties, which are the basis for its multifaceted utility in various scientific, technological, and environmental applications, are collectively defined by the existence of oxygen functional groups, changes in electronic structure, and the introduction of active surface sites.[29]

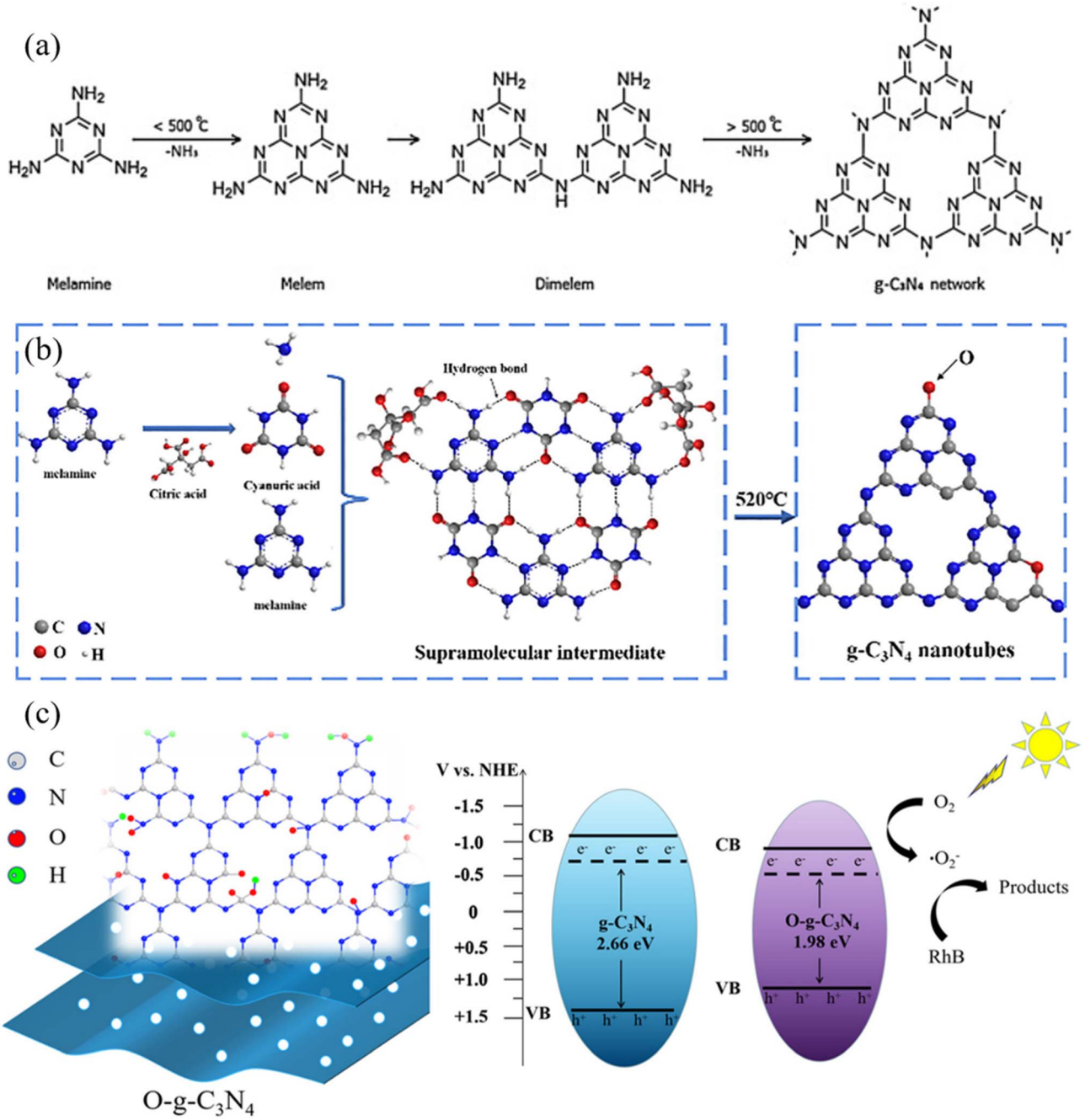

Fig. 2 Schematic illustration of structural changes during the synthesis of (a) g-C_3N_4 (ref. 20) and (b) O@g-C_3N_4 (ref. 23); (c) decrease in bandgap and increase in light absorption capacity due to oxygen doping onto g-C_3N_4.[15]

4. Recent advancements in the photodegradation performance of O@g-C_3N_4

In the field of photodegradation, O@g-C_3N_4 has recently become a compelling and adaptable photocatalyst.[30] These developments have considerably improved the material's ability to break down harmful organic contaminants and pollutants, revolutionizing the area. The sorts of oxygen functional groups included in the lattice and the oxygen doping levels can now be precisely controlled thanks to advancements in synthesis techniques. O@g-C_3N_4 now exhibits increased catalytic activity, expanded spectrum responsiveness, and better charge carrier separation, all of which have improved photocatalytic performance.[31] To further enhance O@g-C_3N_4's photodegradation abilities, synergistic techniques have been investigated, including coupling it with co-catalysts or heteroatom doping. These recent developments highlight O@g-C_3N_4 as a critical component in the search for environmentally friendly and

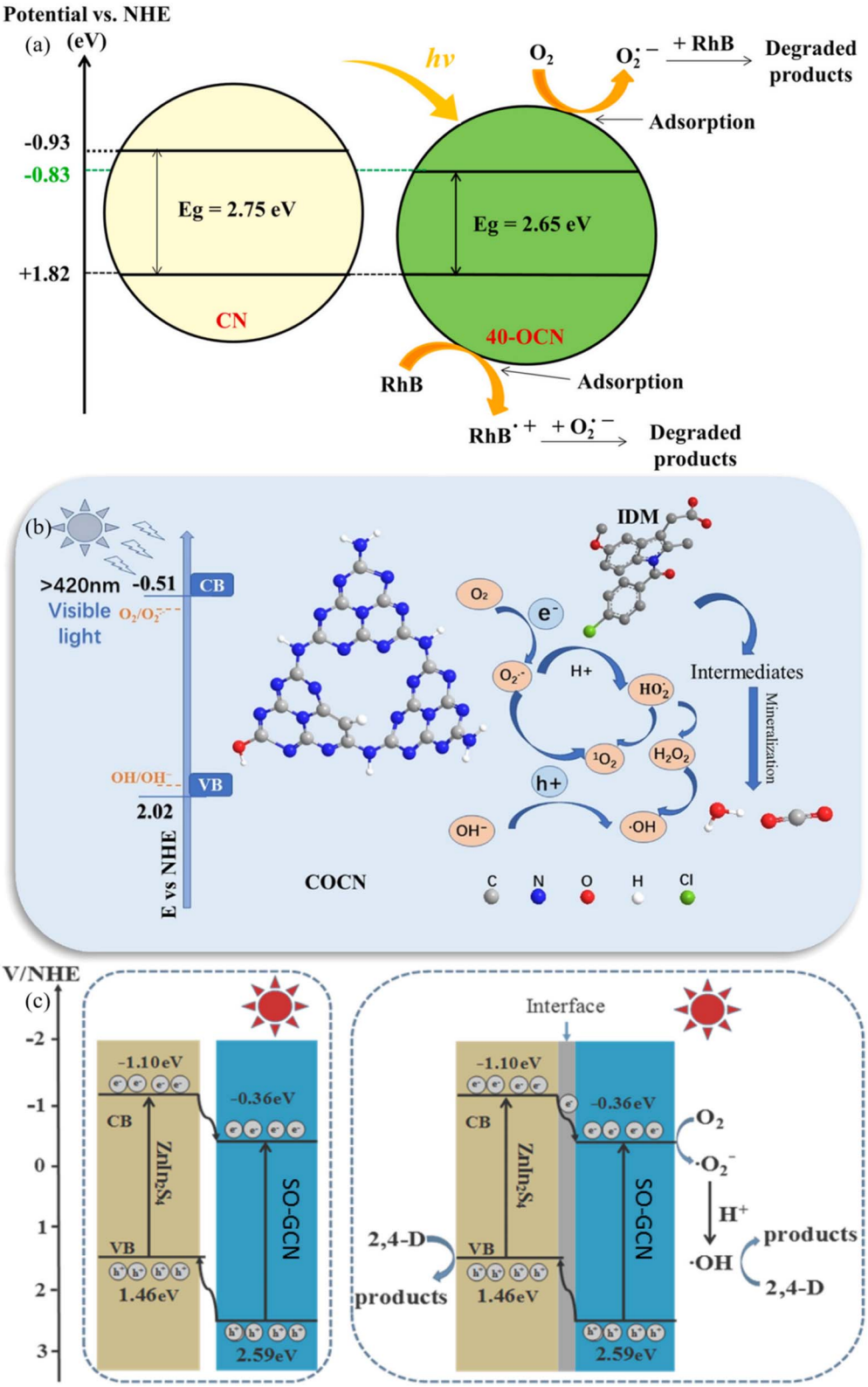

Fig. 3 (a) Degradation mechanism of pristine (a) O@g-C_3N_4 (ref. 32), (b) O@g-C_3N_4 developed on a 2D surface[33], and (c) O@g-C_3N_4-based nanocomposite.[34]

Table 1 Summary of the photocatalytic performance of O@g-C_3N_4 and O@g-C_3N_4-based nanocomposite

Photocatalyst	Pollutants	Catalyst dosage (g L^{-1})	Pollutant conc. (mg L^{-1})	Time (min)	Efficiency (%)	Ref.
O@g-C_3N_4	Methylene blue	0.2	20	30	95.05 ± 2.17	12
O@g-C_3N_4	Rhodamine B	0.2	20	30	97.79 ± 1.58	12
O@g-C_3N_4	Methyl orange	0.2	20	30	89.77 ± 2.19	12
O-g-C_3N_4	Rhodamine B	1	1.0×10^{-5} mol L^{-1}	70	90	15
O@g-C_3N_4	Tetracycline	0.4	30	120	87.2	29
O@g-C_3N_4	Rhodamine B	0.5	1.0×10^{-5} mol L^{-1}	15	98	35
O@g-C_3N_4	Rhodamine B	1	30	140	94	32
O@g-C_3N_4	2,4-Dinitrophenol	0.3	10	75	100	24
O@g-C_3N_4	Bisphenol A	0.4	10	180	63	36
O@g-C_3N_4	Methylene blue	—	—	180	81	25
Carbon doped O@g-C_3N_4	Indometacin	0.4	4	60	98.9	33
Carbon doped O@g-C_3N_4	Bisphenol A	1	10	240	97.85	37
OCN/Py	Tetracycline	0.16	10	60	97.37	38
Polyoxometalates/O@g-C_3N_4	Sulfosalicylic acid	1	40	35	99	39
Protonated g-C_3N_4/O-g-C_3N_4	Deoxynivalenol	—	10	150	86.6	40
O@g-C_3N_4/$NaBiS_2$	Methylene blue	1	5	90	81	41
O@g-C_3N_4/$NaBiS_2$	Rhodamine B	1	5	240	90	41
In_2O_3/O@g-C_3N_4	Bisphenol A	1	—	180	91	42
β-Bi_2O_3/O@g-C_3N_4	Bisphenol A	0.5	15	180	98.7	43
O@g-C_3N_4/CdS	2-Chlorophenol	—	10	60	100	44
O-doped CN-M/CN-U	Bisphenol A	—	5	90	75	45
POCN/anatase TiO_2	Enrofloxacin	1	10	60	98.5	26
$ZnIn_2S_4$/SO-GCN	2,4-Dichloro phenoxyacetic acid	0.4	—	180	92	34
O@g-C_3N_4/g-C_3N_4/TiO_2	Gatifloxacin	1	10	120	91.7	46

sustainable photodegradation solutions, holding great promise for addressing urgent environmental challenges like detoxifying industrial wastewater and removing persistent organic pollutants from water.

Pristine O-doped g-C_3N_4 was fabricated by Tran *et al.*[32] for visible light photodegradation of rhodamine B. Compared to pure g-C_3N_4, the OCN compounds exhibit better photodegradation of rhodamine B. This is because the photocatalytic activity is improved due to a smaller bandgap energy and a slower recombination rate of generated electrons and holes, as illustrated in Fig. 3a.[32] Furthermore, O@g-C_3N_4 was developed on carbon surface to photodegrade indometacin (IDM) under visible light irradiation by Zheng *et al.*[33] The improved photocatalytic activity may have been accredited to effective charge separation and a wider visible light absorption zone, as seen in Fig. 3b.[33] The carbon and oxygen-doped g-C_3N_4 photocatalytic accomplishes excellent mineralization of IDM and offers a new approach to further strategize promising photocatalysts with exceptional wide spectral-responsive characteristics as a promising substitute for conventional water treatment technology. O@g-C_3N_4-based nanocomposites, as researched by Uddin *et al.*,[34] demonstrated a 3–5 times faster rate of 2,4-dichlorophenoxyacetic acid degradation under visible light irradiation (*i.e.*, 0.0112/min, by 30%-$ZnIn_2S_4$/SO-GCN), equated to pristine $ZnIn_2S_4$ and SO-GCN. This is ascribed to the nanocomposite's harmonious effect in an exclusive 3D/2D heterojunction, which speeds up the e^-/h^+ separation. The h^+ and $^{\bullet}OH$ species play a significant part in the breakdown of 2,4-dichlorophenoxyacetic acid over 30%-$ZnIn_2S_4$/SO-GCN, as illustrated in Fig. 3c.[34] Encouraging recent developments in O@g-C_3N_4 for photodegradation applications has, in summary, ushered in a new age of environmental remediation and sustainable pollution elimination. The material has evolved into a highly effective and promising photocatalyst, ready to solve the ever-growing ecological pollution and contamination difficulties. Table 1, provided below, summarises these cutting-edge advances in great detail.

5. Future perspective and conclusion

Future research and applications in oxygen-doped graphitic carbon nitride (O@g-C_3N_4) look promising. More research into precisely adjusting oxygen doping amounts and positions within the g-C_3N_4 lattice will yield a more excellent knowledge of structure–property correlations. Enhancing photocatalytic effectiveness and expanding the material's uses require developing nanostructuring methods and exploring synergistic co-doping with other heteroatoms. Further areas for investigation include O@g-C_3N_4's ability to solve new environmental problems and its incorporation into scalable, affordable technologies for pollutant dradation, energy conversion, and environmental purification. Innovation, practical application, and critical role in long-term solutions for a cleaner and greener world are the hallmarks of O@g-C_3N_4 research's future. Finally, O@g-C_3N_4 has become a highly promising photocatalyst with enormous promise for resolving today's ecological and energy concerns. This article has emphasized the crucial significance of oxygen doping in cultivating the photocatalytic activity of O@g-C_3N_4 by thoroughly studying synthesis techniques, structural alterations, and current developments. The g-C_3N_4 lattice

is given special features by adding oxygen functions, including changed electronic band structures and better surface reactivity, dramatically increasing photocatalytic activity. These developments have a lot of potential for uses, including the oxidation of organic pollutants, the production of renewable energy, and environmental cleanup. Future advancements in O@g-C_3N_4 research are expected to be interesting. It is anticipated that optimizing oxygen doping, investigating innovative co-doping procedures, and utilizing nanostructuring methods can further improve the material's performance. In addition, O@g-C_3N_4 must be incorporated into practical tools and techniques to convert laboratory findings into workable solutions, ultimately paving the way for a cleaner and more sustainable future. O@g-C_3N_4 is a tribute to the inventiveness of materials research and provides a look into the potential for functional materials to spur advancements in environmental rectification and energy conversion. Researchers and scientists are set to open new vistas in cutting-edge materials and sustainable technologies as the voyage of oxygen-doped g-C_3N_4 continues.

Conflicts of interest

There are no conflicts to declare.

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