

Review

Pushing the Operational Barriers for g-C₃N₄: A Comprehensive Review of Cutting-Edge Immobilization Strategies

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Abstract: This comprehensive review explores recent advancements in immobilization strategies for graphitic carbon nitride (g-C₃N₄), a metal-free photocatalyst that has gained significant attention for its optical and physicochemical properties comparable to traditional photocatalysts like TiO₂. However, a critical challenge regarding their application has emerged from the difficulty of its recovery due to its powdery nature. Therefore, several alternatives are being explored to immobilize this material, facilitating its recovery and reuse. This review systematically categorizes various physical and chemical immobilization techniques, providing an in-depth analysis of their advantages, drawbacks, and applications. Techniques such as encapsulation, electrospinning, casting, and coating, along with their adaptations for g-C₃N₄, are thoroughly examined. Additionally, the impact of these strategies on enhancing the photocatalytic efficiency and operational stability of g-C₃N₄, particularly in environmental applications, is also assessed. Thus, this review aims to provide valuable insights and guide future research in the realms of photocatalysis and environmental remediation. The review contributes to the understanding of how immobilization strategies can optimize the performance of g-C₃N₄, furthering its potential applications in sustainable and efficient environmental solutions.

Keywords: 3D-printing; casting; coating; encapsulation; electrospinning; graphitic carbon nitride; immobilization; pollutant removal; sensors; water-splitting



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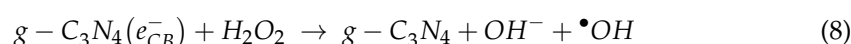
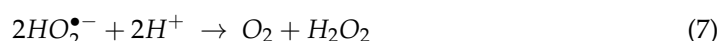
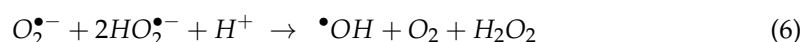
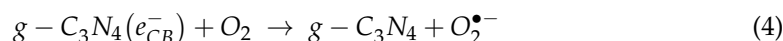
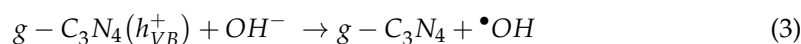
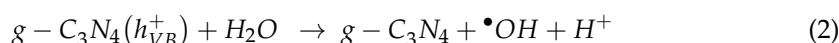
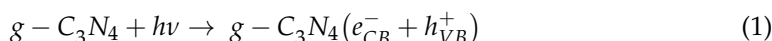
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1. Introduction

In recent years, the global water crisis has emphasized the urgent need for innovative and efficient methods to enhance water quality. Moreover, increasing pollutants and decreasing clean water availability have aggravated the situation, making sustainable solutions necessary. Among the treatments, advanced oxidation processes (AOPs) have emerged as powerful tools for degrading a wide variety of pollutants, with photocatalysis standing out prominently [1]. This process is essentially based on the use of photocatalysts, which are semiconductor materials capable of generating pairs of electrons and holes when exposed to light. Because there is no need to add any oxidizing agent and high efficiency, this process has gained importance compared to other AOP methods [2].

However, to harness all these advantages offered by the photocatalysis process, the development of catalysts that enhance this process is crucial. Among these catalysts, graphitic carbon nitride (g-C₃N₄) has gained considerable attention due to its unique properties such as visible light absorption, chemical stability, and adjustable bandgap, making it highly desirable for photocatalytic applications [3]. These mentioned properties arise from its unique structure, organization, and molecular composition. It is composed of regularly arranged heptazine rings formed by the sp² hybridization of C and N atoms [4]. Furthermore, its band gap is approximately 2.7 eV, substantially lower than that of TiO₂, which is around 3.2 eV. This makes it ideal for photodegradation under visible light (460 nm) [5]. Below

are the chemical reactions that occur during photocatalysis (Equations (1)–(8)) under acidic conditions, which result in the production of reactive oxygen species (ROS) [6,7].



These reactions enable the complete mineralization with breakdown of recalcitrant contaminants into CO_2 and H_2O until these ROS (hydroxyl ($\bullet OH$) or superoxide ($O_2^{\bullet -}$) radicals) are present in the water, as shown in Figure 1a [8]. Another removal process that has been on the rise in recent years is the use of $g-C_3N_4$ in photoelectrocatalysis processes, whose mechanism is simplified in Figure 1b. Meanwhile, the reactions to produce H_2 , crucial in the water-splitting process, with the $g-C_3N_4$ are the same, so it is involved only in the water oxidation reaction (Equation (9)) and the reduction of H^+ (Equation (10)) in this process, as well as the $g-C_3N_4$ catalyzing this production [9]. Furthermore, just like in the case of contaminant removal, water-splitting can also be enhanced through radiation.

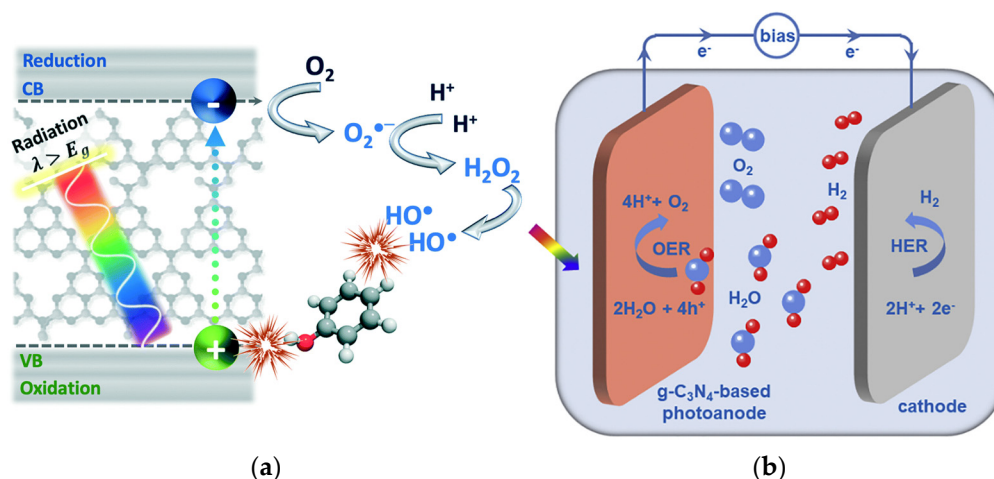


Figure 1. General reaction mechanism of $g-C_3N_4$ in the (a) photocatalytic (reproduced from [10] with permission from the Royal Society of Chemistry) and (b) photoelectrocatalysis processes under slightly acidic pH (adapted from [11] Copyright 2019 with permission from Elsevier (Amsterdam, The Netherlands)).

Another advantage of the photocatalyst is its simple and relatively economical and sustainable synthesis process, as it involves the thermal treatment of inexpensive nitrogen-rich precursors such as cyanamide, dicyanamide, melamine, thiourea, and urea [12], as seen in Figure 2. Although there are slight differences in the polymerization process based on the precursor used (as reported in the literature), the formation of $g-C_3N_4$ remains essentially the same. Figure 2 schematically illustrates the polymerization for one of the precursors commonly employed in the synthesis of $g-C_3N_4$. Initially, a melamine molecule

forms at a temperature of 335 °C, followed by melem at 390 °C, and finally, the melon monomer. As the temperature increases, the tri-s-triazine polymerization process occurs, heating to 550 °C to yield g-C₃N₄ [13]. It is worth noting that this described process occurs under a scarce air atmosphere. If conducted under an N₂ atmosphere, as reported by Chang et al. [14], defects in the structure may occur during the rearrangement of C, N, and H.

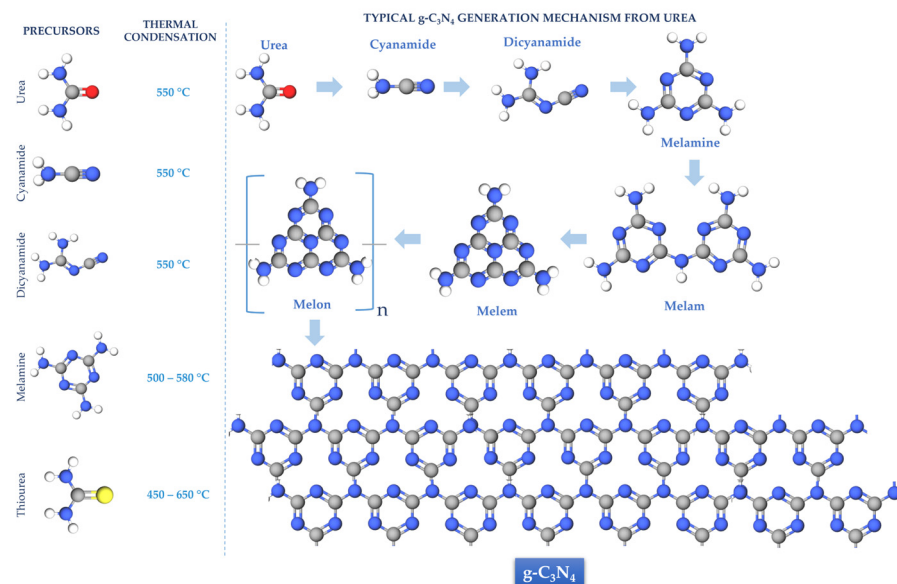


Figure 2. Scheme of the polymerization process for the formation of g-C₃N₄ using the urea precursor when operating under an air atmosphere. This figure also displays the other precursors along with their condensation temperature to obtain the polymer. Note: The color of each sphere represents a different chemical element: blue for nitrogen, gray for carbon, white for hydrogen, red for oxygen, and yellow for sulfur.

Furthermore, the g-C₃N₄ layer forms triangular nanopores with numerous edges, evenly distributing throughout the material [15]. This structure facilitates the modification (vacancy, doping) of g-C₃N₄. For example, other catalysts or elements such as F, S, and transition metals (Ni, Fe, etc.) are often incorporated as dopants to enhance the surface area of this photocatalyst [16]. Given all these advantages offered by g-C₃N₄, it finds numerous applications, with a fundamental emphasis on sensors [17], water splitting [18], pollutant degradation [19], and energy storage [20]. As depicted in Figure 3, there is a discernible evolution in the utilization of g-C₃N₄. Notably, there is a significant surge in applications related to the removal of pollutants and energy storage including H₂ generation by water splitting, experiencing an increase of approximately 8% and 2.3%, respectively. Meanwhile, there is a slight decline in the application of water splitting, with a decrease of around 11%.

However, a significant challenge is faced by the use of these photocatalysts, including g-C₃N₄, due to their small particle size [8]. These challenges can be delineated as follows: (i) limitation in the separation and recovery by the utilization of powdered photocatalysts, which often require complex and expensive filtration or centrifugation methods; (ii) material loss and operational impact during these separation processes, which directly impacts both operational cost and efficiency; (iii) tendency to agglomerate diminish the effective surface area of these powdered materials that can reduce their photocatalytic properties; and (iv) scaling up processes for industrial applications due to the variability in particle size and shape, which can lead to inconsistent performance and hinder reproducibility in laboratory-scale operations [21]. To circumvent these drawbacks, alternative approaches are being explored such as immobilization, which is a process that fixes the photocatalyst to a solid support [22]. This strategy enhances the efficiency and sustainability of the process by allowing for the recovery and reuse of the material [23,24]. A clear indication of the effectiveness of this strategy is evident in Figure 3b, which shows an increase of almost

54.5% in the last five years. However, the total number of articles in the last 10 years indicates that there is still much ground to cover within the field of g-C₃N₄ immobilization.

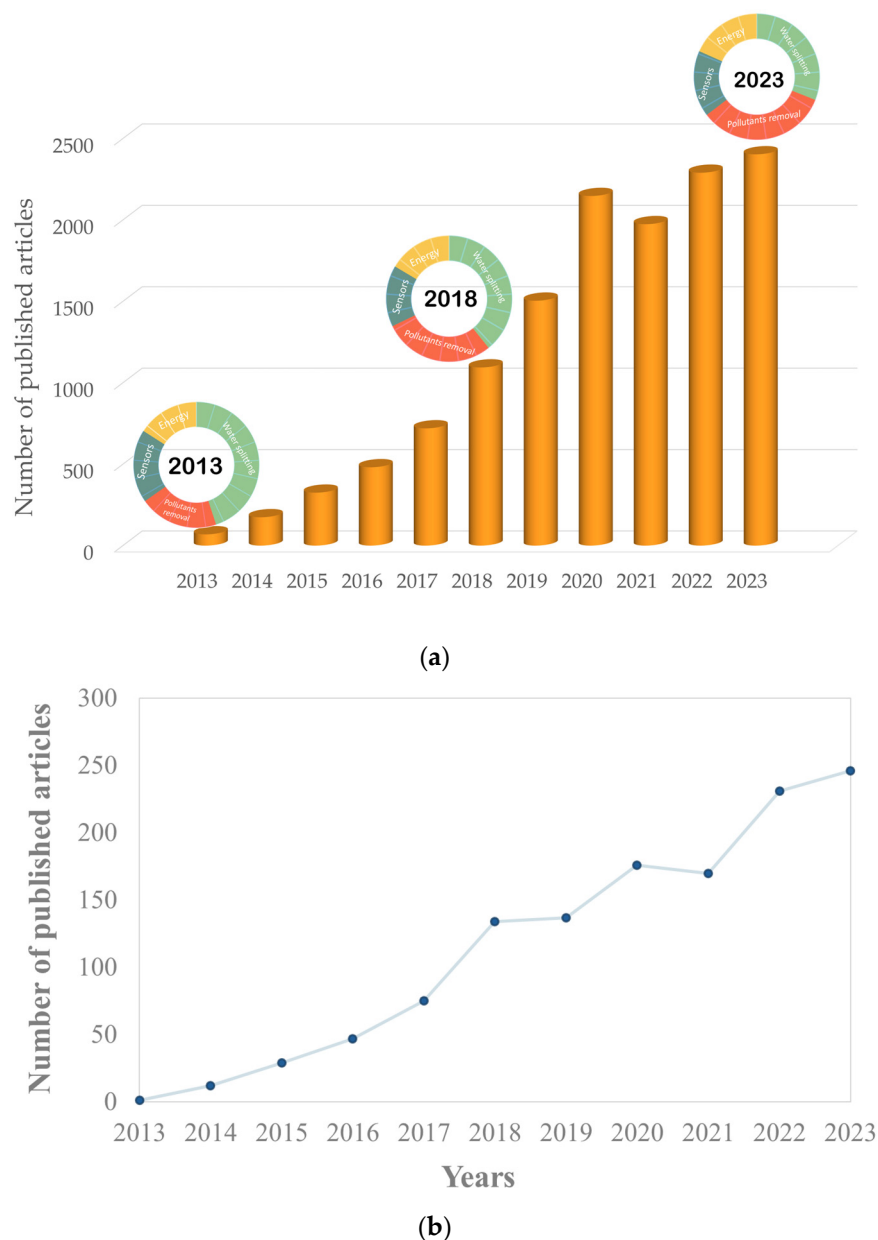


Figure 3. Illustration of the evolution of the number of published articles using (a) g-C₃N₄ for different applications and (b) its immobilization in the Scopus® database over the last 10 years. The distribution of articles published on the application of g-C₃N₄ is shown for the years 2013, 2018, and 2023. The keywords employed include ‘graphitic carbon nitride’ with a specific focus on each application (sensor, pollutant removal, and energy including water-splitting).

Nevertheless, the immobilization techniques employed in g-C₃N₄ are similar to those used with other catalysts and enzymes. The principle behind these immobilization techniques lies in fixing the photocatalyst onto solid supports, such as porous materials or a matrix. However, it is necessary to explore various immobilization techniques to enhance specific properties, such as catalytic performance for pollutant removal or H₂ and O₂ generation [25,26]. Among the highlighted strategies are the formation of photocatalytic membranes, incorporation of materials with magnetic properties, and immobilization in biopolymers, structured inorganic materials, etc. Additionally, within these strategies, two

distinct approaches have emerged. The first involves the application of more conventional techniques such as encapsulation or coating, with modifications aimed at enhancing the material properties for application. For example, improving the floating of g-C₃N₄-retaining materials allows for the maximizing of the utilization of incoming radiation. The other approach, which holds less weight in innovation strategies, involves the development of innovative materials containing g-C₃N₄.

Due to the complexity of these strategies, the aim of this review is to comprehensively explore and categorize the various immobilization strategies most commonly employed for g-C₃N₄, thus providing the state of the art in the last 5 years. These techniques will be discussed in detail, including their respective advantages, limitations, and applications.

2. Encapsulation

The encapsulation is the first innovative technique highlighted in this review to enhance the performance and durability of photocatalytic materials. This involves enclosing the photocatalyst within a protective shell or matrix, which can be composed of various materials such as polymers, silica, zeolites, or other suitable compounds for encapsulation. Also, this immobilization technique can allow for improving the stability, efficiency, and selectivity by restricting access to the active sites of the photocatalyst.

For all these reasons, the literature has reported the global interest in research focused on photocatalyst integration with hydrogel frameworks to create innovative 3D porous hydrogel photocatalysts [27]. This approach has been found to improve the properties of the material notably by boosting its adsorption capacity, stability, and separability. Furthermore, it augments the quantity of active sites and establishes an internal conductive pathway, facilitating efficient charge transfer [28].

Hydrogels are polymeric materials that can retain large amounts of water within their 3D structure. These materials are useful for immobilizing biological or chemical substances due to their capacity to maintain a moist environment. Hydrogels find widespread use in biomedical applications, including controlled drug releases and tissue engineering. Due to their nature, their application for environmental applications is recommended. Gelation of hydrogels is a promising strategy for incorporating the photocatalyst as the starting material to build 3D porous architectures.

In addition to harnessing light energy effectively and demonstrating self-control capabilities, hydrogel composite photocatalysts have the unique ability to float on the surface and in shallow water. This characteristic enhances their visible-driven photocatalytic performance, making them particularly practical for environmental applications, such as algae pollution control [29,30].

2.1. Alginate

Sodium alginate is a biodegradable and biocompatible substance which can electrostatically interact with many metal cations as a result of its carboxylate and hydroxyl functional groups [31]. By using cross-linking reactions, hydrogels that are suitable for supporting or embedding catalysts can be formed. Its application in the immobilization of photocatalysts has been reported in the literature. Thus, Rodriguez-Couto et al. [32] reported the immobilization of ZnO in alginate gel beads to operate in a fluidized bed reactor, showing a good performance at different residence times and arising high degradation levels. Dalponte et al. [33] evaluated the potential of TiO₂ confined in alginate gel beads which presented low density and an easy separation from the reacting medium, remaining available to be reused in another batch.

Thus, the immobilization of photocatalysts within hydrogel matrices, such as alginate, has emerged as a pivotal advancement. In this context, the application of this technique to g-C₃N₄ immobilization has received particular attention due to the 3D porous architectures allowing for the preservation of their unique sheet-like sp² structure [27]. By a facile cross-linking polymerization method, alginate modified g-C₃N₄ composite hydrogels could

be produced with the ability to remove several pollutants such as inorganic and organic pollutants, as is shown in Table 1.

Table 1. Summary of several articles covering various g-C₃N₄ immobilization techniques and their application in wastewater treatment.

Material	Immobilization Technique	Target Pollutant	Removal (%)	Reuses	Ref.
Alginate	Encapsulation	Diclofenac Rhodamine B Isoproturon	99 99 90	5 cycles, Rhodamine B 92%	[22]
Alginate	Encapsulation	Methylene Blue Rhodamine B	80 50	5 cycles, Methylene Blue 70%	[34]
3D kaolinite/alginate	Encapsulation	Brilliant Green	97	10 cycles, 82%	[35]
Alginate	Encapsulation	Pb(II), Ni(II) Cu(II)	383.4 mg g ⁻¹ 306.3 mg g ⁻¹ 168.2 mg g ⁻¹	5 cycles, capacity loss: 15.2%, 16.5%, 15.5%	[36]
Alginate	Encapsulation	Rhodamine B	99	5 cycles, 99%	[37]
Alginate	Encapsulation-3D printing	Methylene Blue	95	3 cycles, 95%	[38]
Chitosan	Encapsulation	Methylene Blue	99	5 cycles, 97%	[39]
Cellulose/graphene oxide hybrid aerogels	Encapsulation	Methylene Blue	99.9	5 cycles, 83%	[40]
Graphene aerogel	Encapsulation	Methylene Blue	83	4 cycles, 78%	[41]
Cellulose	Encapsulation	Methylene Blue	99.8	4 cycles, 95%	[42]
Hydroxyethyl cellulose	Cross-linking and gelation	Bisphenol A	98	-	[43]
Polyacrylonitrile (PAN)/polyaniline	Electrospinning	Methylene Blue Methyl Violet Ciprofloxacin Acetamiprid	97.0 94.3 88.9 87.6	- 5 cycles, 94% 5 cycles, 88% 5 cycles, 87%	[44]
PAN	Electrospinning	As(III) As(V)	97 98	5 cycles, As(III) 80% 5 cycles, As(V) 85%	[45]
Polyvinyl alcohol (PVA)/poly(dopamine)	Electrospinning	Methylene Blue <i>Escherichia coli</i>	98.7 93.1	5 cycles, 90% -	[46]
Silica nanofiber	Electrospinning	Tetracycline	90.0	5 cycles, 83.7%	[47]
Polycaprolactone	Electrospinning	Aflatoxin B ₁	96.9	5 cycles, 96%	[48]
Cellulose nanofibers	Casting	Rhodamine B	98	-	[49]
Nb ₂ O ₅ embedded polyethersulfone	Casting	Tetracycline	88	-	[50]
PAN nanofibers	Electrospinning and casting	Congo red Methyl blue	92 87	-	[51]
Polyurethane foam immobilized with Reduced Graphene Oxide (rGO)/TiO ₂ /ultrathin-g- C ₃ N ₄ (PRTCN)	Dip-coating and UV-light ageing process	Norfloxacin	95.4	6 cycles, 95.4%	[52]
Aluminum-plastic supported	3D printing and coating	Tetracycline hydrochloride	93.6	-	[53]

Table 1. Cont.

Material	Immobilization Technique	Target Pollutant	Removal (%)	Reuses	Ref.
Exfoliated g-C ₃ N ₄	Electrophoretic deposition	Acid Orange 7 4-chlorophenol	60 40	-	[54]
Ni-Fe LDH Modified Sulphur Doping	Layer-by-layer assembly	2,4-dinitrophenol	98	5 cycles, 90%	[55]
Fe ₂ O ₃ /Ni-Fe LDH	Layer-by-layer assembly	Rhodamine B Methylene Blue	88.5 91.2	5 cycles, <30% 5 cycles, <40%	[56]
Mg/Al-LDH	Layer-by-layer assembly	Pyrene	79	-	[57]
ZnCr LDH	Layer-by-layer assembly	Rhodamine B	99.8	3 cycles, 84.5%	[58]

Thus, Falletta et al. [22] prepared g-C₃N₄ alginate beads and examined their ability to photodegrade different pollutants such as dyes, drugs, and herbicides under solar light irradiation using two water matrixes (ultrapure and simulated drinking water). Exceptionally high photodegradation results were observed (99% for diclofenac and rhodamine B and 90% for isoproturon), which is higher than the previously reported in the literature. It has also been demonstrated that g-C₃N₄ alginate microspheres are easily synthesized and have excellent photocatalytic activity for the degradation of different dyes (rhodamine B and methylene blue). A significant benefit of calcium alginate is the presence of the hydroxyl group, which enhances the adsorption of organic pollutants as well as the transfer and separation of photogenerated charge carriers [34]. Furthermore, the hydrogel microspheres are stable and can be easily removed from water for reuse. For this reason, it is possible to operate in successive cycles, showing that after five cycles, the hydrogel microspheres remained stable. In addition, they determined that the g-C₃N₄ alginate microspheres could be biodegraded anaerobically to produce methane, which could be utilized for energy recovery and recycling [34]. In another study, to enhance its photocatalytic activity, g-C₃N₄ was modified (g-C₃N₄-TiO₂ P25) and encapsulated into calcium alginate beads, nearly achieving the complete removal of dye reactive brilliant red X-3B in 1 h of light exposition. Similarly, the reusability of kaolinite/g-C₃N₄ proved to be quite inadequate, with its recovery process being notably cumbersome. Consequently, kaolinite/g-C₃N₄ alginate beads were synthesized using a cross-linking strategy. It is noteworthy that the introduction of alginate into the kaolinite/g-C₃N₄ composite resulted in a reduction of the bandgap and a recombination of electron-hole pairs within the beads [35].

Recently, floating g-C₃N₄ alginate beads were prepared by the dispersion of photocatalysts in sodium alginate solution and calcium carbonate, and the solution was dropped into a solution of calcium chloride and acetic acid. Floating beads of calcium alginate with g-C₃N₄ (25%) showed good photocatalytic activity, with a methylene blue degradation of around 81% being achieved after 42 h [34]. These floating materials are recognized for their superior surface irradiation and oxygenation, along with the convenience of recovery and potential for repeated use. Previously, Fu et al. [37] developed a new strategy based on the interconnection of macroporous photocatalyst-immobilized gels. In their study, the mass transfer of contaminants was enhanced by the interconnectivity of the macropores in the g-C₃N₄ nanosheet-immobilized gels. To do this, g-C₃N₄ nanosheets were dispersed into the sodium alginate solution in the presence of an anionic surfactant (sodium dodecyl sulphate) and sodium bicarbonate. This viscous solution was stirred with air bubbles that act as pore templates to increase its foamy characteristics. Finally, the solution containing microbubbles was dripped into a solution of calcium chloride in acetate acid. In this stage, two reactions take place: (i) the crosslink of Ca²⁺ with alginate, generating the gelation of the drops; and (ii) the reaction between acetate acid and sodium bicarbonate with the accumulation of CO₂ gas. In addition, by vacuuming the pores on the surface of

beads, they were openly facilitating the movement of contaminants on the immobilized photocatalyst. As shown in Figure 4, the cross-sectional morphologies of the structure and porosity are clearly different to the conventional gelation without including bubbles as pore templates. Thus, these beads display macropores with diameters until 300 μm providing high surface area and channels for the transfer of contaminants, where they are degraded by $\text{O}_2^{\bullet-}$ radicals obtained by the reduction of oxygen (O_2), which occurs as a result of the photogenerated electrons. In order to optimize the photocatalyst, several factors (such as sodium alginate content or bead sizes) should be considered in order to provide physical stability with a network density without affecting the photocatalytic performance.

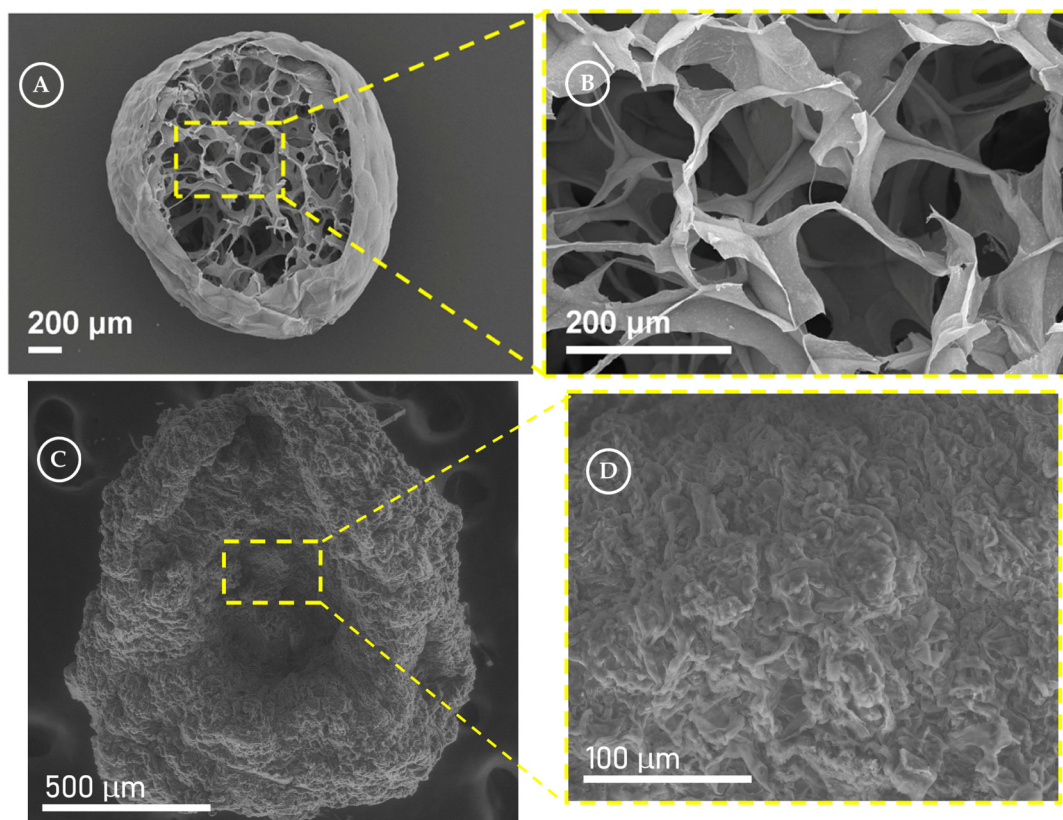


Figure 4. SEM images of (A,B) floating g-C₃N₄ alginate beads (adapted with permission from [37]. Copyright 2021 American Chemical Society, Washington, DC, USA) and (C,D) conventional g-C₃N₄ alginate beads.

Leveraging the cutting-edge advancements in 3D printing technology, a novel and striking approach involves the use of alginate to produce hybrid aerogels that also has macroscopic architectures of intricate patterns. In their study, He et al. [38] explored the use of g-C₃N₄-based ink and printing by three routes shown in Figure 5: (i) direct printing in ambient air; (ii) within a supportive reservoir containing a fluidic matrix made up of calcium chloride and glycerol solution, or (iii) using Pluronic®F127, also known as poloxamer 407, as non-toxic and non-ionic copolymer. In 3D printing, the rheological characteristics of the ink emerge as pivotal determinants, significantly influencing the performance of the resultant aerogel. Consequently, the g-C₃N₄ nanostructures undergo a transformative process, evolving into 3D aerogel networks with customizable geometries adaptable to a spectrum of specific applications [38,59].

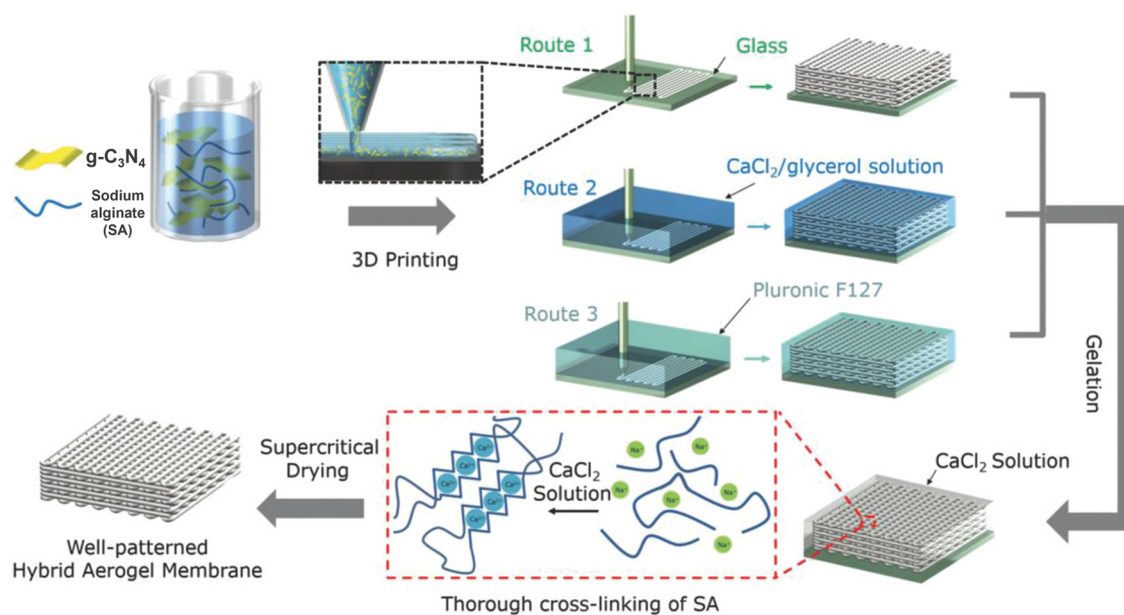


Figure 5. Graphical depiction of the manufacturing technique of hybrid aerogels using 3D printing and $g-C_3N_4$ -based ink. Route (1) ink extruded and directly applied onto either a glass base coated with a Vaseline layer; Route (2) deposited the ink into a bath with $CaCl_2$ and glycerol mixture; or Route (3) Pluronic F127. Reproduced from [38] with permission from the WILEY-VCH Verlag GmbH and Co. KGaA, Weinheim (Weinheim, Germany).

2.2. Chitosan

Chitosan is a biopolymer derived from chitin, which is not only biodegradable and biocompatible but also possesses functional groups, with a high capability of immobilizing $g-C_3N_4$, which has advanced the field of material science and environmental remediation. The integration of chitosan with $g-C_3N_4$ has been shown to improve the photocatalytic degradation of pollutants, attributed to the increased surface area and enhanced adsorptive properties of the composite material (Figure 6). This innovative approach not only leverages the unique characteristics of chitosan but also contributes to the development of sustainable and efficient environmental cleanup solutions [23].

Zhao et al. [39] synthesized $g-C_3N_4$ chitosan beads via the blend cross-linking method in which the structural characterization demonstrated that $g-C_3N_4$ was distributed on the surface of the chitosan matrix with a definite morphology. For the synthesis, the chitosan must be dissolved in acetic acid in which an adequate amount of $g-C_3N_4$ powder is added. To form the beads, epichlorohydrin was added and the solution was dropped into a basic media. The spherical beads showed improved pollutant adsorption and photocatalytic activity and can be regenerated by the addition of sodium hydroxide and hydrogen peroxide without negative effects on the beads, which kept their activity after four cycles (Table 1). It was found that the ratio of $g-C_3N_4$ /chitosan plays a critical role in determining the photocatalytic efficiency, particularly in the adsorption and decomposition of dye methylene blue under visible light. Thus, at the optimal ratio, the beads showcased superior photocatalytic performance (a factor of 1.8), outperforming pure $g-C_3N_4$ chitosan. This enhancement underscores the significance of precise components proportioning in the synthesis of photocatalytic materials.

PVA strengthens the chitosan matrix by immobilizing it, forming H-bonds with the amino acids of chitosan in addition to providing more active sites for adsorption [60]. In addition, several composites were synthesized by a combination of chitosan and PVA [61]. In this context, oxygenated $g-C_3N_4$ nanosheets were immobilized into a solution of PVA/chitosan by electrospinning technology, which will be explained in Section 3.2, to fabricate nanofiber membranes [62]. The immobilization of $g-C_3N_4$ by this method permits

its application in other uses such as food packaging [61] or antibacterial activity [62]. Table 2 summarizes several examples of g-C₃N₄ immobilization, highlighting uses different to the degradation of pollutants.

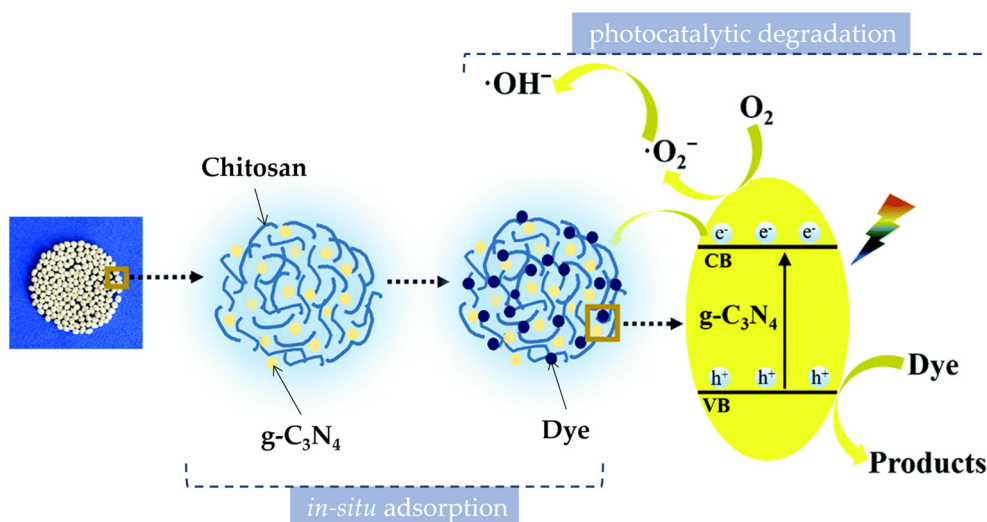


Figure 6. General representation of the photodegradation process, in this case, using beads made of g-C₃N₄ and chitosan. In this process, a synergistic and chained effect occurs, beginning with the adsorption of pollutants (in this example, a dye, followed by its subsequent photodegradation under visible light). Adapted from [39] with permission from the Royal Society of Chemistry (London, UK).

Floating chitosan-based composite with g-C₃N₄ was synthesized by the cross-linking reaction of chitosan and PVA, in which to decrease the hydrogel density, air bubbles were added [63]. It was found that this 3D hydrogel with a lightweight and highly porous structure exhibited good properties and was capable of being suspended in aqueous solutions, while the Ag₂O/g-C₃N₄ photocatalysts were tightly attached to the pores. As was mentioned above, the floating immobilized Ag₂O/g-C₃N₄ 3D hydrogel demonstrated high adsorption and photocatalytic inactivation of *Microcystis aeruginosa* under visible light with limited reusability in successive runs. This fact could be explained by the hydrogel swelling property decreased over time and the presence of algae cells and metabolites that filled the pores of the hydrogel, reducing both active sites and effective contact with photocatalysts. Thus, 3D hydrogel should be regenerated using solvent exchange and ultrasonic treatment to restore these active sites [64].

Table 2. Overview of several immobilization techniques for g-C₃N₄ and their applications in different fields of wastewater treatment.

Material	Immobilization Technique	Observation and Application	Ref.
Chitosan and PVA	Encapsulation	Preparation of film for food packaging	[61]
Chitosan and PVA	Electrospinning	Nanofiber membranes with light catalytic antibacterial activity	[62]
Natural latex foam	Brush coating	PVA/MXene and protonated-g-C ₃ N ₄ . Solar steam generation. Good structural stability over 6 cycles	[65]
rGO/indium tin oxide (ITO)	Drop-casting	The MoS ₂ -g-C ₃ N ₄ immobilized on the surface of the rGO/ITO electrode enables dopamine detection with a linear response ranging from 0.005 to 1271.93 µM and a detection limit of 1.6 nM.	[66]

Table 2. Cont.

Material	Immobilization Technique	Observation and Application	Ref.
Glassy carbon	Coating	Nitrobenzene contaminant detection ranges from 10 μM to 1 mM, with a detection limit of 1.3 μM using g-C ₃ N ₄ immobilized on glassy carbon	[67]
Amorphous Ni-imidazole framework	Ultrasonication	The amorphous Ni-imidazole framework in g-C ₃ N ₄ enhances photocatalytic H ₂ production by around 2272.6 $\mu\text{mol/g/h}$	[68]
CdS quantum dots with Ni decorated	Ultrasonication and chemical deposition	A dual functionalization of g-C ₃ N ₄ was carried out with Ni atoms and CdS quantum dots to enhance H ₂ production, achieving an evolution rate of 9.5 mmol/g/h	[69]
Boron and graphene quantum dots co-doping in ITO	Impregnation and coating	A highly sensitive photoelectrochemical sensor for dopamine detection, with a broad linear range (0.001–800 μM) and low detection limit (0.96 nM), was achieved by co-doping boron and graphene quantum dots into g-C ₃ N ₄ in an ITO electrode	[70]
Screen-printed electrodes	Drop-casting	Incorporating 2D-carbonylated g-C ₃ N ₄ into the screen-printed electrode improved glucose determination with a linear range of 0 to 5 mmol L ^{−1} and a detection limit of 0.43 mmol L ^{−1} .	[71]
NiFe LDH */ sulphur-doping	Layer-by-layer assembly	Designed a Ni-Fe LDH using a deep eutectic solvent-based fabrication method for the detection of dimetridazole, an antiprotozoal drug. The assay exhibits a linear range from 0.0008 to 110.77 μM with a detection limit of 1.6 nM	[72]
CoAl-LDH *	Layer-by-layer assembly	This material is added to the cement to generate a photocatalytic cement mortar for NO _x degradation, achieving an efficiency of around 42%	[73]
Carbon nanotubes and lignin	3D-printing solid support	Vertical 3D printing creates an eco-friendly electrode with a H ₂ yield of up to 4.36 $\mu\text{mol/cm}^2$ h, surpassing g-C ₃ N ₄ films 41.6 times.	[74]
Nafion	Drop-casting	This electrocatalysts for water splitting show impressive O ₂ evolution reaction performance with a low overpotential of 355 mV and a Tafel slope of 46.8 mV dec ^{−1} .	[75]

* LDH: layered double hydroxide.

2.3. Cellulose-Based Materials

Cellulose, known for its cost-effectiveness and eco-friendliness, can act as a sustainable electron mediator, enhancing photocatalytic hydrogen production synthesis via hydrogen bonding [76].

As a non-toxic biomass material, carboxymethyl cellulose boasts outstanding water solubility, high viscosity, and film-forming properties. Consequently, it finds wide applications in food, pharmaceutical, and textile industries. The structure of this compound allows for the formation of hydrogen bonds of varying strengths with the amino group of g-C₃N₄ [77].

Cellulose/graphene oxide hybrid aerogels exhibited the potential to overcome the poor recyclability of g-C₃N₄ due to their porous structure and enhanced electronic conductivity. Cai et al. [40] fabricated a 3D recyclable g-C₃N₄ by encapsulation into cellulose/graphene oxide hybrid aerogels using the facile freeze-dried method. This 3D aerogel showed dual

properties related to the porous structure and light absorption capacity that make it an excellent material with high efficiency and reusability for pollutant degradation (Table 1).

Hydroxyethyl cellulose is a polymer with a high content of hydroxyl group with a tendency to establish hydrogen bonds. In light of previous studies (in which the g-C₃N₄ was bonded by intermolecular electrostatic attraction with polyethyleneimine by the hydrogen peroxide production [78]), recent research was focused on linking g-C₃N₄ with hydroxyethyl cellulose via hydrogen bonds and used as potential proton donor for the generation of hydrogen peroxide. This fact increases the feasibility for the application of this material in the degradation of pollutants. In addition, it highlighted that this method under mild conditions, via non-covalent bonds, avoids the introduction of impurities in the generated hydrogen peroxide reducing its production operation cost [79].

g-C₃N₄ powders were encapsulated by their mixture with hydroxyethyl cellulose in a simple and mild one-pot polymerization by the cross-linking action of epichlorohydrin [43]. This fact allows the transformation of this solution into a 3D hydrogel network. The synergistic effect of dynamic adsorption and photodegradation, described in Figure 6, was also detected achieving higher bisphenol A removal than the g-C₃N₄ powders (around 3.4 times). The stability of the composite was tested in successive cycles exhibiting the same behavior after four cycles. XRD patterns showed that the overall structure of the 3D photocatalyst is similar, and only several impurity peaks were observed [43].

Smykalov et al. [80] synthesized nanocomposites of g-C₃N₄ and char, active under visible irradiation, from melamine and hydroxyethyl cellulose/glucose. Thus, a char from hydroxyethyl cellulose and glucose was obtained by thermal synthesis and integrated into the g-C₃N₄ structure, where the carbon acted as an electron collector. This facilitated the separation of photoinduced electrons and holes, thus increasing the photocatalytic efficiency of g-C₃N₄. Consequently, this process led to a significant degradation of ofloxacin (96%) in only 120 min, with an efficiency nearly constant in four cycles.

A simple method for the fabrication of ZnO-g-C₃N₄ photocatalyst nanocomposite film by encapsulation in polyacrylic acid-grafted hydroxyethyl cellulose and PVA by free radical polymerization using ammonium persulfate as an initiator and ferric chloride as a cross-linker was described by Sultan et al. [81]. Another alternative is the use of hydroxyethyl cellulose/silica/g-C₃N₄ to obtain solid foams with hierarchical porous structure by use of gas bubbles template combination with the freeze-drying method [82]. As is shown in Figure 7, sodium dodecyl sulphate, an anionic surfactant, was added to the synthesis procedure as a foaming agent and bubbles stabilizer. It was determined that the air bubble template improved the synergic effect of adsorption and photocatalytic activity due to the higher porosity and specific area. In this case, the solidification of air bubbles into hydrogel took place before their rupture, generating a material of high porosity. In this process, the foaming step is one of the most important factors. Two foaming systems to generate sufficient air bubbles within the solution were tested: mechanical and magnetic stirring. Mechanically foaming led to a foam with a pore diameter ranging from 200 µm to a few millimeters, resulting in an open-cell structure that is beneficial for the diffusion and transport of pollutants. However, in the hydrogel obtained by magnetic stirring, only fewer bubbles and irregular pore structures were detected with diameters around 20–100 µm. For this reason, the porosities of the hydrogel fabricated by mechanical foaming significantly enhanced from 92% to 96%, exhibiting a more favorable structure with higher porosity, larger surface area, and greater pore volume. These characteristics contribute to a symbiotic development on pollutant adsorption and photocatalytic activity, as shown in Figure 6.

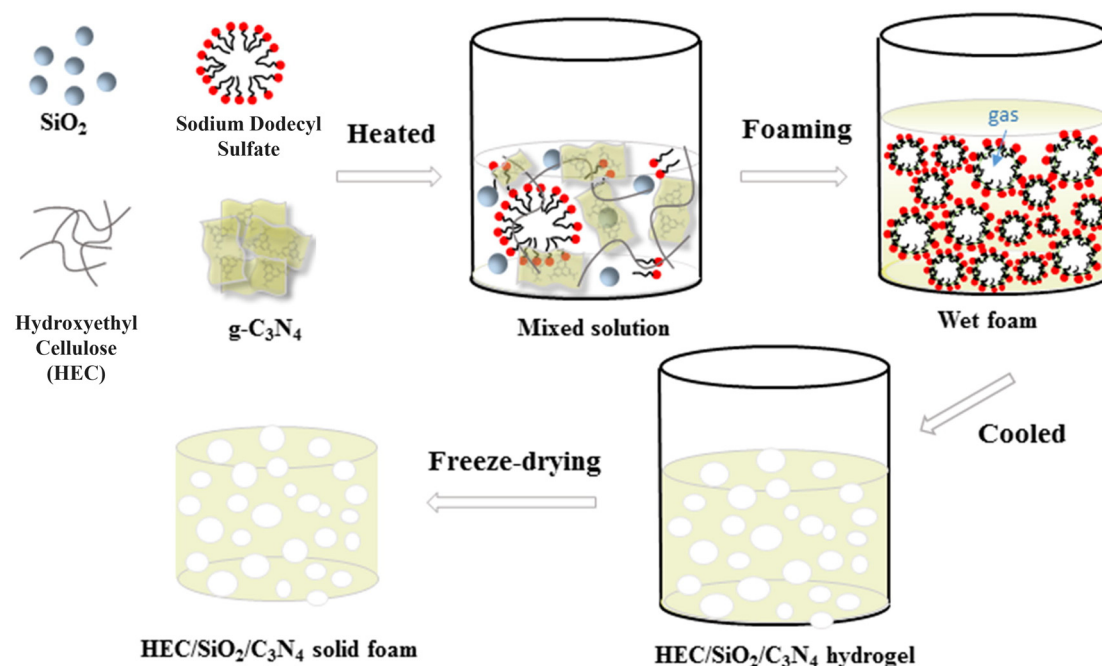


Figure 7. Schematic illustration of the preparation of solid foams with hierarchical porous structure using HEC/SiO₂/g-C₃N₄ and gas bubbles template in combination with freeze-drying method. Reproduced from [82]. Copyright 2022 with permission from Elsevier (Amsterdam, The Netherlands).

3. Electrospinning

In recent years, there has been a notable increase in the use of this technique to create nanoscale materials known as nanofibers using various polymers [83], with the electrospinning method considered the most advanced and robust for this purpose [84]. This technique is applied in various areas, from generating micro/nanofibers for human health applications to environment and energy generation [85]. The process of producing these fibers essentially involves applying a high voltage to a solution (typically polymeric) between the tip and the collector—where the fibers are deposited. This directional movement occurs as the solution is guided towards the collector due to the difference in charge between them [86]. Therefore, it is not surprising that this technique is widely used to create membranes and fibers that serve as physical support for retaining catalysts such as g-C₃N₄.

It should be noted that the process of fiber synthesis is inherently complex, as can be seen in Figure 8, as various parameters (such as concentration, molecular weight, pH, applied voltage, surface tension in the droplet, viscosity of the mixture, among other factors) affect this operation [87,88]. Despite these complexities, the inherent advantages of these materials have been noted, in recent years, by the generation of the polymeric materials together with g-C₃N₄ for their application in micro/nano-extraction and photocatalytic membranes. Among these, the polymers most used are PAN and PVA, due to their properties.

3.1. Membrane Based on PAN Fibers

PAN allows the generation of nanometric fiber diameters for loading photoactive material with macroscopic porous structures that favor mass transfer during reactions [89]. Another property of these PAN fibers is that they enable floating photocatalysis due to their flexible self-supporting structures, which makes them a highly porous material [90]. The reason why the floating fibrous materials are attractive for research is related to the loss of UV radiation and solar radiation which are 1% and 20% lower, respectively, at a depth of 0.5 m under water [91].

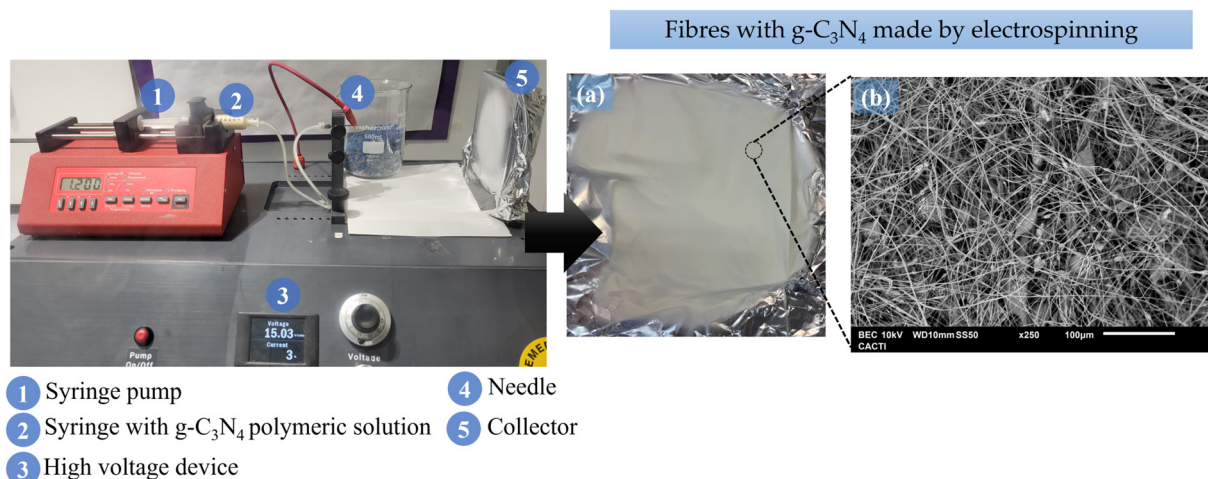


Figure 8. Representation of the formation of (a) PAN fiber with g-C₃N₄ on aluminum foil, (b) as detected through scanning electron microscope.

Examples of this are found in several studies performed by Tripathy et al. [45], Mao et al. [44], and Chai et al. [92], who generate PAN fibers (both unmodified and modified) to retain modified g-C₃N₄. Additionally, the three studies successfully address four different water treatment issues: heavy metals, organic contaminants (dyes and pharmaceuticals), pathogenic bacteria, and oil/water separation, which shows the versatility of this technique.

Tripathy et al. [45] focused on synthesizing an effective adsorbent for removing As(III/V) from water (Table 1). To achieve this, they supported an amine-functionalized graphitic carbon nitride/magnetite material on the PAN membrane. Notably, the photocatalyst was modified with diethylenetriamine to incorporate NH₂- groups, and magnetite was added through a solvothermal process. The incorporation of these amino groups and magnetite enables the generation of electrostatic attractions and surface complexation during the adsorption of As(III/V), resulting in 97.0% removal for As(III) and 99.0% for As(V) using 0.4 g/L of the material within 60 min of contact time. Consistent with this, the study presents pseudo-second-order kinetics and Langmuir isotherm models, indicating that the adsorption process is endothermic and spontaneous. In fact, it was demonstrated that this material is efficient since As(III/V) removal of over 85% was achieved for real waters from four zones of India (Harail gram-panchayat of Mohiuddinagar Block in Samastipur District of Bihar State).

The other two studies are centered on the synthesis of materials serving as membranes for separation, incorporating solar radiation to eliminate water pollutants. Thus, Mao et al. [44] synthesized a membrane using the coaxial electrospinning method, combining PAN with polyaniline (PANI) on LaFeO₃ cable fiber to retain g-C₃N₄, obtaining a Z-scheme g-C₃N₄/PAN/PANI@LaFeO₃ cable nanofiber membranes. PANI is included for its conductive properties as a polymer, attributed to its extensive conjugated P electrons, which confer exceptional h⁺ transporting capability, a slow charge recombination rate through electron transfer processes, and outstanding stability. Moreover, it consumes electrons and holes from LaFeO₃ and g-C₃N₄, efficiently promoting the separation of photo-generated charge carriers and enhancing catalytic performance. Consequently, in situ generation of H₂O₂ is facilitated, favoring the treatment by an advanced oxidation process as photo-Fenton process and enabling the removal of contaminants such as methylene blue, methyl violet, ciprofloxacin, and acetamiprid in 75 min and with virtually no loss of activity after five cycles (Table 1).

Regarding the Chai et al. [92] study, the synthesis of their material was inspired by the natural structure of *Euplectella aspergillum*. To achieve this structure, a combination of PAN, polyvinylpyrrolidone (PVP), and g-C₃N₄ was carried out. The primary advantage of this structure lies in the fact that g-C₃N₄ particles in the membrane are responsible for

the photocatalytic degradation of contaminants, made accessible through the properties provided by PAN. Simultaneously, PVP acts as a pore-forming agent, facilitating efficient large-scale wastewater purification. This dual polymer combination makes the material an excellent separator of oil from water, along with conferring an outstanding multicomponent purification capability. The efficiency in degrading contaminants such as rhodamine B, methylene blue, malachite green, tetracycline, and Cr(VI) was 99.64%, 99.38%, 99.99%, 74.33%, and 91.30%, respectively. Additionally, the antibacterial efficiency exceeded 87%.

3.2. Enhanced Applications and Properties of PVA Membranes

PVA is one of the most commonly used polymers, which, like PAN polymer, possesses highly hydrophilic, biodegradable, and biocompatible attributes, along with excellent properties such as strength, water solubility, and thermal characteristics [93]. Due to these properties, this material is employed in various applications, ranging from environmental to biomedical and drug delivery, as exemplified by PVA with zinc acetate nanowebs [94]. However, the goal in these applications is to retain the desired characteristic material content inside the fiber or on its surface. In the past year, numerous studies have sought to modify fibers using non-metal components for antibacterial performance. Examples of this trend and application to immobilize g-C₃N₄ are the studies of Luan et al. [46] and Bai et al. [62]. In the research of Luan et al. [46], polydopamine (PDA) has been added to improve the dispersion of g-C₃N₄ nanosheets in the PVA polymer solution, providing an enhancement in photocatalytic bacterial inhibition activity. This approach has achieved a good inhibition effect on *Escherichia coli* and *Staphylococcus aureus* under visible light irradiation, with a maximum inhibition zone diameter of 10.8 mm and 11.6 mm, respectively. Due to these positive results, they analyzed the photocatalytic property of the material with the methylene blue dye and found that almost all of it was degraded (98.1%). However, reusability led to a loss of efficacy, with elimination reduced to 90% in the fifth use.

Bai et al. [62] used a PVA fiber with chitosan and modified g-C₃N₄, specifically oxygenated and termed O-g-C₃N₄ with 12% O₂ content. In contrast to the study of Luan et al. [46], using a fiber containing 17% O-g-C₃N₄ detected a higher inhibition effect on *Escherichia coli* and *Staphylococcus aureus* with inhibition zone diameters of 26 mm and 16 mm, respectively. This fact was attributed to the synergistic effect between chitosan and O-g-C₃N₄. Hence, this immobilization technique represents a significant step towards improved photocatalytic bacteriostasis.

3.3. Membrane with Incorporation of Carbon Quantum Dots

Another trend found in recent years is the incorporation of carbon quantum dots with g-C₃N₄ in the membrane, as it allows for a considerable increase in photocatalytic activity due to the increase of active sites, excellent electron transfer ability, easier adsorption of pollutants, and up-converting photoluminescence of the g-C₃N₄.

Huang et al. [47] synthesized silica nanofiber membranes on which they grew in situ quantum dots/g-C₃N₄. With this material, the contaminant (tetracycline) was practically eliminated (90%) after 150 min, and additionally, the removal of tetracycline was almost doubled compared to the material without carbon quantum dots. Moreover, this new material demonstrated effectiveness over a wide pH range and in the presence of different inorganic anions.

Similarly, Yao et al. [48] synthesized a polycaprolactone membrane modified with PDA, like the study by Luan et al. [46], and containing g-C₃N₄/carbon quantum dots. As in the previously reported study, the results are promising, achieving a 96.88% removal of aflatoxin B₁, which is the most toxic biological toxin in food, in 30 min under visible light irradiation. After five cycles of use, it presents practically the same removal capacity.

Recently, several studies have focused on the improvement of electrospinning PAN nanofiber membranes for membrane bioreactor processes by incorporating g-C₃N₄ nanotubes/carbon dots (g-C₃N₄ NT/CDs) and applying thermal post-treatments. This modifi-

cation significantly improves the water flux and anti-fouling properties of the membrane. The optimum composition found was 0.5% by weight of g-C₃N₄ NT/CDs, which, when hot pressed at 90 °C for 10 min, achieved a remarkable flux recovery coefficient of 70%. These advances position modified PAN nanofiber membranes as promising candidates for wastewater treatment due to their enhanced performance and resistance to fouling [95].

4. Casting

In the casting process, a liquid or molten material is poured into a mold and is allowed to solidify to achieve the desired shape. These molds or supports are carbon-based, and once the mixture is poured, it is left to deposit onto the surface [96]. The use of this support type is driven by the non-covalent hybridization interaction between the material surface (catalyst or photocatalyst) in the support, which is a relatively weak bond, and the integration of carbon supports aims to overcome this limitation and boost H₂ evolution reaction activity [97]. In Figure 9, a brief schematic of the simplest and most used casting method is shown, using a polymeric solution as a base, wherein the g-C₃N₄ is dispersed. However, it is worth mentioning that within this casting process there are various ways to perform this immobilization technique, but in recent years the use of drop-casting (Figure 9) has become quite common in the literature. Essentially, the liquid mixture is dripped onto the material surface in a low and constant flow. This type of technique has been widely employed in electrode fabrication for applications in water splitting or sensors, as seen in studies by Salgaonkar et al. [98], Xiaowei et al. [99], and Velmurugan and Yang [66] (Table 2). However, this technique can generate composite foil, as exemplified in the study of Hegedűs et al. [100] that investigated the generation of this foil through the casting of TiO₂ and PVA, which was subsequently subjected to heat treatment to enhance its stability in water for removing Triton X-100, that is a commonly used non-ionic detergent. In this context, in recent years, the immobilization of g-C₃N₄ for these applications has also been explored.

A clear example of this immobilization through casting is explored by Zhou et al. [101], who prepared a casting solution containing 15% polyvinylidene fluoride, 2% PVP, 82% N-methyl-2-pyrrolidone (NMP) solvent, and 1% of the combination of g-C₃N₄ with nitrogen deficiency and doped with dopamine, to self-polymerize and generate a PDA coating. It is worth noting that this self-polymerization process carried out in this research allows PDA to adhere to g-C₃N₄ with nitrogen deficiency through strong covalent and non-covalent interactions, and π - π^* interaction between them. In the end, this new material, with a pure water flux of 390 L/(m²·h), achieved a bovine serum albumin rejection of 95.9%, excellent self-cleaning performance, and a flux recovery rate of 84.5%.

Regarding immobilization through drop-casting, a clear example is found in the study by Torres-Pinto et al. [75]. In their study, they employed drop-casting to immobilize g-C₃N₄ in a Nafion and ethanol solution, integrating it into a nickel sponge. Throughout the water-splitting process, alongside optimizing the g-C₃N₄ synthesis, their focus extended to enhancing the efficiency of the O₂ evolution reaction. The most favorable O₂ evolution reaction properties were observed for a catalyst load of 1.00 mg/cm², as an elevated g-C₃N₄ content could potentially obstruct active sites. Additionally, the resulting electrode demonstrated remarkable stability, facilitating continuous operation for over 45 h, achieving an overpotential of 355 mV at 10 mA/cm², and showcasing a Tafel slope of 46.8 mV/dec. This research underscored the significance of attaining lower Tafel slope values without resorting to supplementary treatments or co-catalysts, a feat that has not been realized in other studies utilizing g-C₃N₄ under the same conditions.

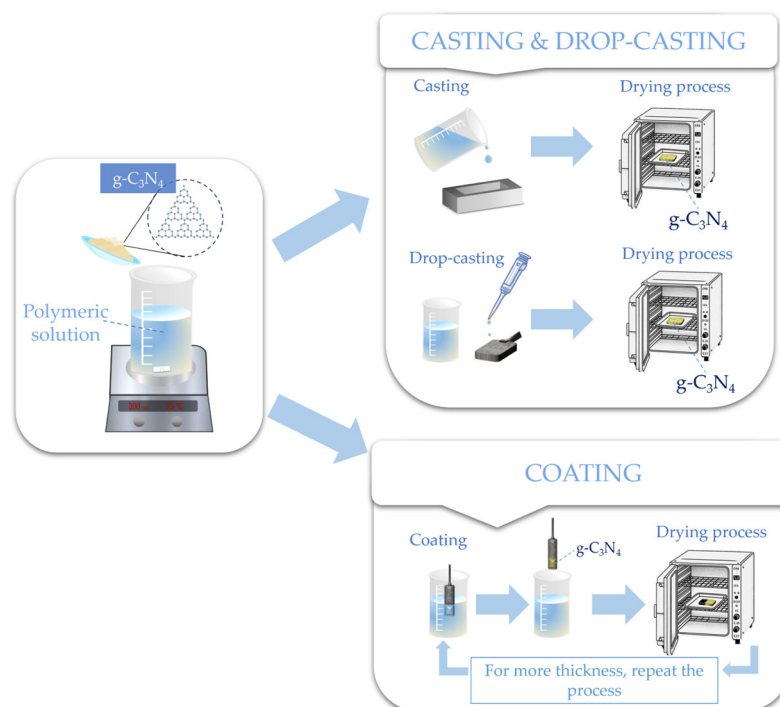


Figure 9. Descriptive diagram of casting processes (and drop-casting) to generate polymeric films with $g-C_3N_4$ and coating to retain $g-C_3N_4$ on a porous surface.

Gowri et al. [102] conducted a comparative study of different immobilization techniques to retain $g-C_3N_4$ nanosheets on a glassy carbon electrode using adsorption, drop casting, and potentiodynamic, which is another casting method involving electrodeposition by applying an electric current to secure the material onto the electrode. According to a study by Gowri et al. [102], this electrodeposition of $g-C_3N_4$ nanosheets directly onto the glassy carbon electrode can be achieved because of adsorption via amine groups and π stacking. Therefore, upon successfully performing these three immobilizations, their electrocatalytic activity was evaluated, and among the three, $g-C_3N_4$ nanosheets made by the potentiodynamic method exhibited an improved electroactive surface area and less charge transfer resistance. Indeed, by achieving these properties, the material demonstrated an enhanced electrochemical activity for substances such as ascorbic acid, dopamine, and hydrogen peroxide.

In a study by Anusuyadevi et al. [49], immobilization was carried out using the casting technique to obtain foams formed by polysaccharide-based buoyant carriers, such as eco-friendly cellulose nanofiber-based buoyant foams, to which $g-C_3N_4$ is added. The result of these foams is a material that floats. Due to the components that form the $g-C_3N_4$ -cellulose nanofiber foam, they validated its stability, and the results were promising, as it maintained its properties and flotation capacity for four weeks under radiation. This is believed to be a consequence of the cell wall materials and the Pickering stabilization provided by the cellulose nanofiber surfactants at the interface of the encapsulated air bubbles. As a result of this, the photocatalytic activity of the new material was assessed through the removal of rhodamine B, revealing an elimination efficiency more than twice that of $g-C_3N_4$ (50% removal compared to 11% within a 360 min period). Despite the environmentally sustainable fabrication of $g-C_3N_4$ -cellulose nanofiber foam, it exhibits lower efficacy compared to other immobilization techniques, since to eliminate the dye, a minimum of 14 h of experimentation under radiation was required. Therefore, with this research, Anusuyadevi et al. [49] take the initial step in the synthesis pathway of wet-stable nanocomposite foam.

For embedding $g-C_3N_4$ porous nanoribbons pillared and MXene into PVA, a heterostructure was created using a simple method of solution casting, which incorporates

different weight percentages of filler into the dielectric composite films in order to achieve the desired consistency and properties [103]. The composite films demonstrated superior mechanical properties compared to the MXene–PVA nanocomposite, displaying significant enhancements in tensile strength and Young’s modulus, while also preserving their elasticity, confirming the possible use of these films as functional energy storage dielectric.

Ultrafiltration (UF) membranes are typically manufactured using the non-solvent induced phase separation technique, in which a polymer is dissolved in a casting solution and solidified by immersion in a non-solvent. These membranes typically have a finger-like structure and a top selective layer that is prone to damage by external factors. The properties and structure of the membrane are adjusted by varying the composition of the casting solution, including the concentration of polymer and additives such as pore formers and nanoparticles, which can improve functionalities such as photocatalytic and antibacterial properties, and overall performance [104]. Thus, photocatalytic membranes with g-C₃N₄ were fabricated via the preparation of casting solutions. Thus, Letswalo et al. [50] prepared flat sheet membranes with g-C₃N₄/Nb₂O₅ nanocomposite that were previously synthesized in an in situ hydrothermal treatment [105] by its dispersion into NMP and polyethersulfone (PES) solutions with the addition of hyperbranched polyethyleneimine (HPEI) to the casting solution. To remove air bubbles, the solution was degassed for one day and then cast onto a glass plate and immersed in deionized water, leading to mixing and polymer precipitation. This formed a thin polymeric membrane, which was subsequently stored in distilled water. This membrane showed high efficiency in the degradation of tetracycline under specific conditions, favored by the generation of radicals on the catalyst surface and achieving removals around 88% after 3 h of irradiation at pH 10. The analysis of degradation products identified the fragmentation of tetracycline into seven innocuous by-products. It is important to highlight that the membrane also demonstrated self-cleaning properties, maintaining a constant flux by avoiding pore clogging [50]. Zhou et al. [51] introduced a UF membrane by using a PAN-C₃N₄ nanofiber layer prepared by electrospinning. These nanofibers were treated by immersion into a solution of NaOH and ethanolamine to become hydrophilic. After that, the nanofibers were included in casting solutions and slowly cooled down to room temperature to degasification. The study revealed that membrane exhibited superior dye rejection and photoactivity compared to other membranes, despite the fact that C₃N₄ reduced photoactivity due to the PAN coverage effect. Figure 10 shows the membrane design. The submerged selective layer acts as a barrier to dye transport by electrostatic repulsion, which hinders dye permeation through the membrane. In contrast, in typical UF membranes, only the surface layer exhibits this positive repulsion toward charged dye molecules. Finally, self-cleaning of membrane by photoirradiation was confirmed by exposition to an Xe lamp for 1 h.

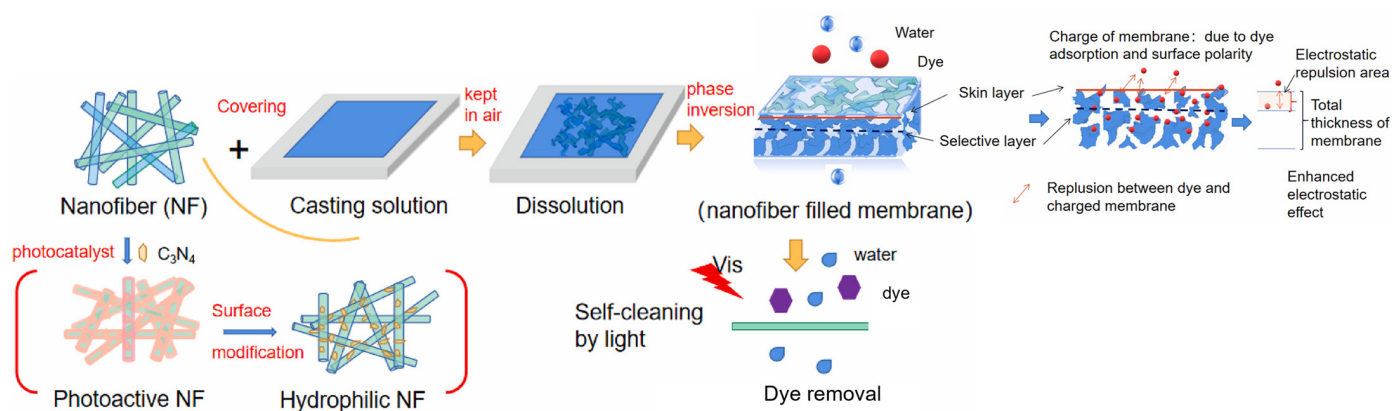


Figure 10. Graphical description of the synthesis procedure and dye removal for the UF membrane of PAN-C₃N₄. Adapted with permission from [51]. Copyright 2023 Elsevier (Amsterdam, The Netherlands).

5. Coating

Coating involves applying a thin layer of material onto a surface to provide properties such as protection and adhesion, or for the controlled release of substances. Normally, the procedure is dipped into for a certain period (which allows the material in the liquid mixture to deposit onto the surface), and then pulled out [21], as is shown in Figure 9. This entire process has a predetermined speed to allow sufficient time for the deposition on the surface. In fact, as the number of times this process occurs increases, the layers on the surface also increase [106].

The immobilization of g-C₃N₄ by coating has been explored in various applications. Dong et al. [107] demonstrated the immobilization of g-C₃N₄ on structured Al₂O₃ ceramic foam for efficient visible light photocatalytic air purification. The immobilization was performed by an in situ thermal approach in which the precursor in powder (dicyandiamide) was mixed with distilled water in an alumina crucible and stirred. A pretreated Al₂O₃ foam was immersed in this mixture and dried in an oven to recrystallize the dicyandiamide. Finally, the covered crucible was heated in a muffle furnace at temperatures from 450 to 600 °C for 2 h. It should be noted that the adhesion of g-C₃N₄ to the Al₂O₃ ceramic foam was robust enough to withstand continuous airflow, which can be attributed to the unique chemical interaction between g-C₃N₄ and Al₂O₃. Furthermore, with optimal support, g-C₃N₄ demonstrated exceptionally high NO removal efficiency, reaching 77.1%, under low-power lamp illumination. The material was compared with other photocatalysts immobilized on Al₂O₃ foam by a similar method. This fact can be explained due to the enhanced crystallinity, high porosity and surface area with more active sites promoting the charge separation and enlarged band gap.

Using a one-step chemical vapor deposition method, tubular g-C₃N₄ was deposited on a support carrier as a porous carbonized grapefruit peel. Initially, the carbon framework was prepared from a freeze-dried grapefruit peel by calcination at 750 °C. After that, the obtained carbon framework was placed in urea powder and heated at 550 °C for 2 h. During this process, a large amount of g-C₃N₄ was deposited on the carbonized grapefruit peel [108]. In addition, the photocatalytic degradation of methylene blue was studied in comparison with bulk g-C₃N₄. It was concluded that several factors have contributed to the efficient photocatalytic degradation efficiency: (i) synthesis procedure reduces the size of g-C₃N₄ to form nano-sized tubes, which increases its photocatalytic efficiency; (ii) owing to its excellent adsorption capabilities, the carbonized framework continuously enriches pollutants on its surface, enhancing its photocatalytic efficiency; and (iii) since carbonized frameworks possess excellent electron transport capabilities, they can effectively separate electrons generated by photosynthesis and enhance light absorption range and catalyst activity by separating electrons effectively.

Rusek et al. [54] furthered this study by immobilizing exfoliated g-C₃N₄ particles on a glass support. This coating was performed using electrophoretic deposition from a suspension in ethanol that had been treated with ultrasound and aged for up to 84 days. The stable part of the split suspension contained particles of a fairly consistent size, which facilitated the creation of stable films of g-C₃N₄ in stirred aqueous solutions. This photocatalyst was tested in the removal of organic pollutants (4-chlorophenol and acid orange 7 dye) from water, showing limited activity in the degradation of 4-chlorophenol.

Coating was used to synthesize a floating material by immobilization on the surface of natural latex foam of a photothermal solar absorber as a mixture of MXene and protonated g-C₃N₄ dissolved in PVA [65]. This solution was brush-coated on the surface of natural latex foam and dried overnight, obtaining hydrophilicity and stronger light absorption material. It was detected a uniform PVA layer on the surface of the convex-shaped latex structure connected with granular structures of MXene/g-C₃N₄. Similarly, PRTC� was anchored onto polyurethane foam by a simple dip-coating and UV-light ageing process. PRTC� demonstrated a high photodegradation and mineralization efficiency towards excellent reusability and anti-interference capability. The removal efficiency of the optimal PRTC�-15% on norfloxacin exceeds 95.4%, achieving a mineralization rate of 70.6%. Moreover,

when exposed to natural sunlight, the PRTC demonstrates a remarkably norfloxacin removal efficiency of 77.5% in the river water background [52].

A 3D aluminum plastic substrate was successfully coated with g-C₃N₄ by coating technology obtaining a self-suspending photocatalyst. Its photocatalytic activities were tested using rhodamine B dye as a target pollutant and compared with the same g-C₃N₄. Under visible light irradiation, the degradation efficiency of g-C₃N₄ adhered to aluminum plastic exhibited the best activity [53]. The purpose of this study is not only to enhance the photocatalytic efficiency of powdered photocatalysts in solutions but also to recover them. In addition, the use of aluminum plastic substrates in the fabrication of self-suspending photocatalyst devices offers a novel approach to provide a new idea for the recovery of aluminum plastic retrieval, suggesting significant prospects for further developments in the future.

6. Layer-by-Layer Assembly

The layer-by-layer assembly technique involves the sequential deposition of alternating layers of materials onto a substrate. This technique is used to construct 2D/2D heterostructure photocatalysts with specific properties. In this context, these structures can be achieved by combining layered double hydroxides (LDH) with g-C₃N₄, enhancing their photocatalytic properties [109]. In addition, Song et al. [109] reviewed the advantages of the use of the combination of g-C₃N₄ and LDHs, describing graphically the main reasons for the improvement of photocatalytic applications (Figure 11). This LDH has a general formula that is $[M^{2+}_{1-x}M^{3+}_x(OH)_2] A^{-}_{x/n} \cdot mH_2O$, where M²⁺ and M³⁺ denote divalent and trivalent metal cations, respectively. Normally the metals used are Zn, Cu, Fe, etc. [110], and the A[−] interlayer anion. Also, the LDH surface has OH groups that facilitate the generation of highly active radicals [111]. Due to these properties, this type of immobilization is primarily used in water splitting and sensor applications. Therefore, in this section, the most recent studies within these applications are highlighted.

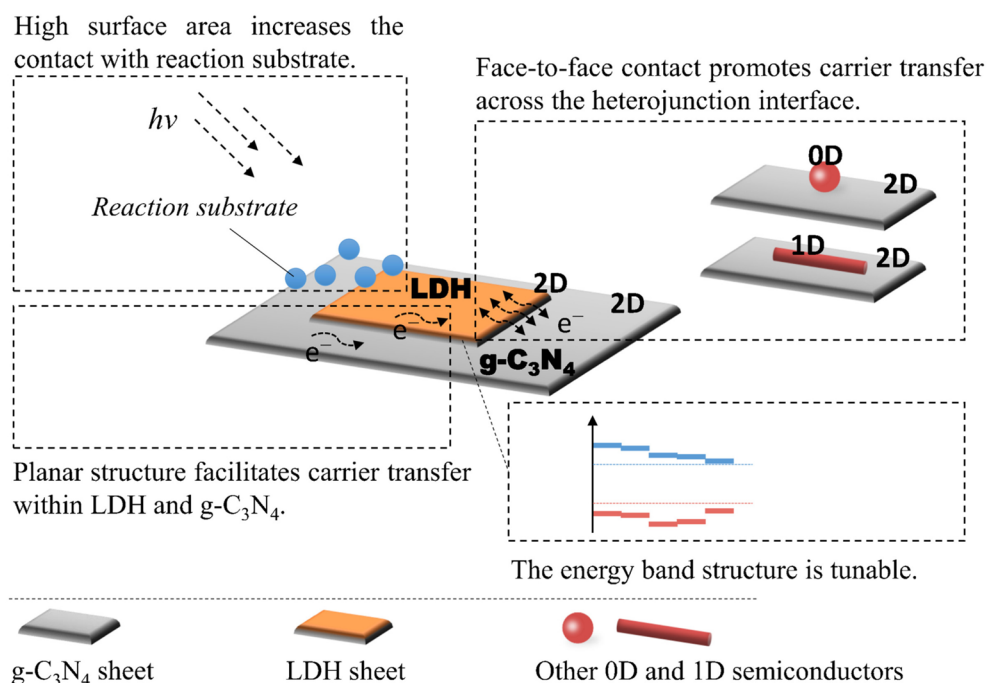


Figure 11. Graphical description of the main advantages of the combination of g-C₃N₄ and LDH for their photocatalytic applications. Reproduced with permission from [109]. Copyright 2018 Elsevier (Amsterdam, The Netherlands).

Regarding to water-splitting application, Boumeriame et al. [112] synthesized CuAl-LDH combined with g-C₃N₄ to enhance the separation and transport of charge carriers,

extending the photoresponse to longer wavelengths. Thus, the goal was to improve the photoresponse for the generation of both O_2 and H_2 . In this case, when working with 0.2% CuAl-LDH combined with $g-C_3N_4$, there is a substantial increase in the amount of evolved H_2 and O_2 exceeding 30 and 1.5 times more, respectively, compared to the production obtained with $g-C_3N_4$ alone. Thus, it confirmed an improvement in the efficient separation of charge carriers at the heterojunction interface via an S-scheme and an increase in the surface area, leading to a significant enhancement in the reactions involved in the processes of H_2 and O_2 generation.

Regarding the application of LDH and $g-C_3N_4$ for the detection of vanillin, the research of Gopi et al. [113] deserves attention. In this case, the combination is with ZnCr-LDH. The reason why this combination can be used as sensors is that there is an effect that enhances electrochemical performance. The properties offered by this material, synthesized through a one-pot hydrothermal method, include a low detection limit, a wide linear range, reproducibility (RSD = 4.40%), and repeatability (RSD = 4.46%). Furthermore, this sensor was also capable of detecting vanillin in real and complex matrices such as ice cream, chocolate, and water.

Nevertheless, Khamesan et al. [58] slightly modified the synthesis of ZnCr-LDH conducted by Gopi et al. [113]. While Gopi et al. [113] carried out the synthesis of bimetallic LDH in the presence of $g-C_3N_4$ in the hydrothermal reactor, Khamesan et al. [58] synthesized them separately and subsequently combined both materials through ultrasonic irradiation. This change in the synthesis procedure implies a significant difference in the application of ZnCr-LDH since Khamesan et al. [58] focused on the production of H_2O_2 and the degradation of rhodamine B. The synthesized material under xenon lamp radiation is capable of generating in situ up to 4.75 mM H_2O_2 after 90 min, making it more effective in eliminating the contaminant rhodamine B. This is because, in the presence of H_2O_2 and electrons, a breakdown occurs to produce $\bullet OH$. Under these conditions, the near-complete removal of the pollutant is achieved (99.8%). Nonetheless, a notable challenge arises with the reuse of this material, as there is a noticeable loss of efficiency. In its third reuse, the elimination efficiency decreases to 84.5% (Table 1).

Other pathways to enhance the removal of organic compounds from water involve modifying either $g-C_3N_4$ or the combination of $g-C_3N_4$ /LDH. One of the most employed modifications in recent years is the sulphur-doping of $g-C_3N_4$, as this addition to the structure alters the bandgap which leads to the development of photocatalysts with greater visible light activity, thereby improving its oxidizing potential [114]. Hence, Hasija et al. [55] combined sulphur-doped $g-C_3N_4$ with NiFe-LDH to delocalize the π -electrons of the graphitic structure, achieving chemical stability. This approach effectively removes nearly all of the 2,4-dinitrophenol under visible light irradiation after 120 min, which is nearly 40% more efficient than sulphur-doped $g-C_3N_4$ alone. Furthermore, they demonstrated that the material maintains a high stability, as even after five uses it still exhibits an efficiency of 90%. Regarding the modification of the $g-C_3N_4$ /LDH composite, as performed by Mohammadi et al. [56], it allows for imparting unique properties such as magnetic characteristics due to the incorporation of Fe_2O_3 . In this case, the material ($Fe_2O_3/g-C_3N_4/NiFe-LDH$) can be recovered using an applied magnetic field and exhibits high efficiency in the removal of cationic dyes such as methylene blue and rhodamine B, achieving 91.2% and 88.5%, respectively. However, despite this material being chemically stable and corrosion-resistant, the efficacy in dye removal is severely affected, as after five reuses its efficiency drops below 40%.

A recent innovation involves the development of $g-C_3N_4$ /LDH compounds that are rich in O_2 vacancies. To obtain this material, Zheng et al. [115] previously synthesized the ZnAl-LDH and then produced the LDH with O_2 vacancies through a treatment with ethylene glycol and sodium hydroxide. Once this LDH was obtained, it was combined with $g-C_3N_4$ through a mechanical grinding method. The purpose of this combination is to enable the LDH, due to the O_2 vacancies in its structure, to facilitate light absorption and promote the generation of radicals. In this way, with the synergistic effect between the

S-scheme heterojunctions and O₂ vacancies, the degradation of tetracycline reaches 95%, and its mineralization reaches 28% after 60 min of visible light irradiation. Additionally, the transformation products obtained in this degradation process are detoxified.

Despite the advantages of layer-by-layer assembly, a significant limitation of these approaches lies in the complexity of scaling up. Moreover, this material is susceptible to structural alterations when exposed to complex matrices, which can lead to instability.

7. Future Horizons in the Immobilization of g-C₃N₄

Briefly, this work has highlighted the numerous strategies being implemented for the immobilization of g-C₃N₄ in recent years. These techniques are highly diverse and allow for their use in multiple applications. However, further study is still needed to maximize the properties of this material when it is immobilized, as the number of works addressing g-C₃N₄ immobilization represents only 12% compared to the number of articles on powdered g-C₃N₄ for the main mentioned applications. Nevertheless, as evidenced in the works summarized in Tables 1 and 2, the efficacy and effectiveness of these new materials are high and heading in the right direction. Additionally, many of them also exhibit high physicochemical stability, allowing for their reuse on multiple occasions without losing much effectiveness.

As emphasized in various sections, the encapsulation technique is the most well-known among all the techniques mentioned in this work. However, in order to fully exploit the visible radiation reaching the aqueous medium and enable efficient degradation of contaminants, certain modifications are made during the synthesis process, such as generating more internal cavities so that the solid material has greater floating. In this case, floating and porosity are the parameters that can limit the degradation process the most when this technology is employed.

However, regarding materials synthesized through electrospinning, their focus is more on generating photocatalytic fibers or membranes. These membranes are capable of easily conducting the photodegradation process, as the g-C₃N₄ is deposited onto the threads forming the membrane, providing a high surface area in contact with water and allowing for the adsorption and subsequent removal of contaminants. The main challenge with these membranes lies in controlling their hydrophobicity and permeability, amongst other properties.

Despite the advantages offered by coating, such as ease of application and modularity for generating electrodes and sensors in water-splitting applications still faces significant challenges. One of the main challenges is ensuring durable adhesion of g-C₃N₄ to the substrate, which can be affected by factors such as coating composition and environmental conditions. Additionally, optimizing coating properties such as density, uniformity, and corrosion resistance remains an active area of research to enhance the performance and durability of g-C₃N₄-coated devices.

On the other hand, casting allows for material generation for both contaminant degradation applications, especially through the production of ultrafiltration membranes, and for water splitting and sensors. However, this technique has drawbacks, such as difficulty in controlling membrane morphology and uniformity, as well as limitations in the variety of materials that can be used.

Another immobilization technique applicable to all applications is LDH, which, due to its ability to enhance charge carrier separation and transport, allows for sensors with higher sensitivity and improvement in the photodegradation process and generation of O₂ and H₂. However, this immobilization strategy is costly when scaling up synthesis for large quantities. Furthermore, this work also highlighted studies that combine different immobilization strategies, demonstrating their versatility and the synergistic effect that combined use can have for the desired application, as reflected in the work of Zhou et al. [51].

Therefore, it is important to assess the intended application of g-C₃N₄ in order to determine the most suitable synthesis method. However, in recent years, g-C₃N₄ has been employed in other applications through photo-electrocatalysis, such as hydrogen gener-

ation and contaminant detection and removal [116–118]. This process exhibits enhanced catalytic capacity due to the combined effect of visible radiation and electric field application, leading to increased production of H_2O_2 . As illustrated in Figure 1a,b, and in the reactions depicted in Equations (1)–(10), H_2O_2 plays a crucial role in hydrogen generation and the generation of ROS for eliminating persistent water contaminants.

However, the majority of studies employ immobilized $\text{g-C}_3\text{N}_4$ on electrodes, such as the work by Yuan et al. [119], who immobilized the $\text{g-C}_3\text{N}_4/\text{Ti}_3\text{C}_2$ composite via drop-casting onto an L-type glassy carbon electrode. In this case, the electrode was used for the detection of ciprofloxacin, utilizing the photoelectrochemical process. This approach resulted in a sensor with long-term stability, good reproducibility, selectivity, and the capability to measure real samples, aspects unattainable without the combination of visible radiation and the electric field. Related to this line of electrode fabrication, Jang et al. [120] conducted the synthesis of graphitic carbon nitride-coated carbon hybrid nanofibers via one-dimensional electrospinning. The material demonstrated a robust photocurrent response under on-off cycling with UV-Vis light, credited to the heterojunction between $\text{g-C}_3\text{N}_4$ and carbonized PAN nanofibers. Also, H_2 evolution experiments revealed a significant enhancement in this generation ($437.4 \mu\text{mol/g}$ up to 4 h) and in maintaining stability across three recycling tests.

Moreover, alternative approaches for these processes should be explored, as demonstrated in the work by Fdez-Sanromán et al. [121]. In this study, a $\text{g-C}_3\text{N}_4@\text{PAN}$ fiber was synthesized via electrospinning. To maximize membrane surface exposure to radiation, the membrane was attached to the glass wall near the electrodes. This setup aimed to optimize radiation penetration through the membrane and its proximity to the electric field, resulting in the near-complete removal of the diclofenac contaminant within 90 min, with virtually no loss of efficacy after five reuses.

Another strategy different from immobilizing $\text{g-C}_3\text{N}_4$ on the electrode surface for photo-electrocatalysis processes is presented by Azhdeh et al. [122], who synthesized a wireless photo-electro cathode using electrophoretic deposition to immobilize MIL-53(Fe) and $\text{g-C}_3\text{N}_4$ particles on a titanium grid sheet, and a wireless photo-electro anode of $\text{TiO}_2/\text{graphite}$ for diazinon removal, all within a single cell. Under optimal conditions, up to 93% of total organic carbon and 97% of chemical oxygen demand were removed in just 35 min. Thus, in addition to its high process effectiveness, attributed to its innovative reactor configuration and design, this approach is considered a next-generation electrochemical advanced oxidation process strategy based on electric-field-driven technology.

Furthermore, recent years have witnessed the utilization of unconventional materials for immobilization such as paper fibers and the use of 3D printing materials as supports for $\text{g-C}_3\text{N}_4$, both within the printed structure and incorporated into subsequently printed materials.

An illustrative example of employing unconventional materials for $\text{g-C}_3\text{N}_4$ immobilization is demonstrated by Mašlana et al. [123], in which an innovative method to fabricate resembling paper by the addition of $\text{g-C}_3\text{N}_4$ with silver nanoparticles into pulp cellulose paper fibers is presented. These fibers were manufactured using the Rapid-Köthen automatic sheet forming machine [123]. Degradation tests with rhodamine B dye were conducted using the casting technique, and after 100 min, the pink color provided by the dye was virtually eliminated (with a 90% reduction), restoring the initial brown tone of the composite paper. These fibers are not only versatile in terms of dye degradation but also exhibit remarkable antibacterial and antiviral properties. They reported an effective elimination of *Escherichia coli* and *Staphylococcus aureus* bacteria, as well as *Pseudomonas virus phi6* ($\Phi 6$) (a bacteriophage of the virus family *Cystoviridae*) after 24 h of incubation using the absorption method. This time can be sharply reduced to 2 h operating at dynamic contact conditions.

The other trend that was found is the incorporation 3D technology. In here, there are diverse alternatives ranging from the incorporation of $\text{g-C}_3\text{N}_4$ into the polymeric matrix forming the 3D support, as demonstrated in the study by Jiang et al. [74], to the creation of 3D supports and 3D reactors in which $\text{g-C}_3\text{N}_4$ is incorporated to be linked to their surfaces,

as seen in the studies of Khezri et al. [124] and Phang et al. [125], respectively. These variations also play a significant role in its diverse applications, such as water splitting and contaminant removal.

For the production of H_2 application, the research of Jiang et al. [74] deserves attention. They utilize vertical 3D printing to create arrays using $g-C_3N_4$, carbon nanotubes, and lignin as a binder. This novel material, acting as a photocathode, enhances light absorption and facilitates efficient chemical reactions. In this way, sustainable energy production is achieved through environmentally friendly materials.

Another support alternative involves the creation of small structures or robots made from a composite material of polylactic acid, graphene, and Fe_3O_4 magnetic nanoparticles, as described by Khezri et al. [124]. In this approach, they coat the $g-C_3N_4$, which is present in a chitosan hydrogel, and fill its interior with an aluminum/gallium alloy. This alloy acts as a catalyst for achieving self-propulsion of the robot in water. Depending on the aluminum/gallium ratio contained inside, the velocity varies in the range of $70\text{--}250\ \mu\text{m s}^{-1}$. Furthermore, this material has an innovative feature, as it is capable of eliminating an explosive such as picric acid, with an efficiency of 38% in 10 min.

On the other hand, another way to utilize these 3D solid supports is by manufacturing reactors in which the material is incorporated and retained. For instance, Phang et al. [125] discuss the development of integrated photocatalytic nanomaterials and 3D printing technology to treat water contaminated with the dye rhodamine B. The printed reactor employs the digital light processing technique, incorporating $g-C_3N_4$. In this instance, a removal efficiency of 93.46% was attained through adsorption and photocatalysis within 8 h under the irradiation of two 50 W LED lights. Moreover, recyclability was examined, revealing that the photoreactors demonstrated exceptional recyclability, with a minimal decrease in photocatalytic efficiency after three consecutive experimental runs.

Therefore, $g-C_3N_4$ immobilization is being taken to new horizons, aiming to enhance the traditional applications that are currently familiar. However, the possibilities of immobilizing $g-C_3N_4$ are practically limitless in terms of reactor configuration and adaptation to various applications. This field is full of opportunities to explore and discover, indicating that there is still a fascinating journey ahead towards new frontiers in this area.

8. Conclusions

In conclusion, this review has undertaken a comprehensive exploration of diverse immobilization techniques applied to the $g-C_3N_4$ photocatalyst including encapsulation, electrospinning, casting, coating, layer-by-layer assembly, and integration into solid supports by incorporation into ink or innovative 3D structures. Additionally, a brief explanation of the synthesis procedures and fundamentals of each technique was provided. Through a critical analysis of their advantages and disadvantages, valuable insights into their efficacy are achieved that highlight specific areas for future prospects to overcome potential limitations and enhance the performance.

An essential observation is that these immobilization techniques have proven highly effective, enabling the facile recovery and reuse of $g-C_3N_4$. This promising outcome suggests a potential pathway for advancing continuous processes in wastewater treatment and warrants exploration at larger scales for practical applications such as energy production or H_2 generation. Concrete examples illustrating the success of these techniques open novel opportunities, contributing significantly to the sustainable and efficient development of applications centered around $g-C_3N_4$. Aligned with these advancements, this review not only enriches the understanding of immobilization techniques but also makes a meaningful contribution to the evolving landscape of scientific inquiry and technological innovation.

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