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A Minireview on the Use of $g\text{-}C_3N_4$–Chitosan Biocomposite for Potential Applications

C. R Santiago-Ramírez[1]*, Pooja R Nair[1], C. A Vela-Monroy[1], C. G Aba-Guevara[2], N. A Ramos-Delgado[2] and M. A Gracia-Pinilla[1]*

[1]Facultad de Ciencias Físico-Matemáticas, Universidad Autónoma de Nuevo León, San Nicolas de los Garza, Mexico, [2]Centro de Investigación e Innovación Tecnológica, Instituto Tecnológico de Nuevo León, Apodaca, Mexico

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***Correspondence:**
C. R Santiago-Ramírez
clau.santiagor@gmail.com
M. A Gracia-Pinilla
miguel.graciapl@uanl.edu.mx

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The novel biocomposite based on graphitic carbon nitride ($g\text{-}C_3N_4$ (CN)) and Chitosan (CS) has been deeply studied and summarized in key points concerning various applications. The CN material is composed of the earth-abundant nature of C, N, and H and possesses excellent properties due to its two-dimensional structure, good chemical stability, and a narrow bandgap that allows its use in many applications. There is a lot of information on the role of CN as a potential photocatalyst, but not in association with other composites. In contrast, this minireview summarizes its applications not only in the field of photocatalysis but also in all fields reported on the biocomposite of CN with CS. The incorporation of chitosan helps to overcome the existing limitations of CN, like low-surface area, low light absorption, fast recombination of charges, and hydrophobic character. To introduce, CS is an attractive biomaterial, which is a low-cost alternative for the preparation of films and catalysts due to its unique characteristics such as biodegradability, antimicrobial activity, and film-forming properties that increase the popularity of CN. In this current minireview, a comprehensive study was conducted on the properties, synthesis, and applications along with the advancements of CN incorporated with CS. Finally, we hope to stimulate researchers to study the biocomposite of CN and CS to find new portals and ways to develop effective materials.

Keywords: g-C3N4, chitosan, biocomposites, photocatalysis, biological, photo-electrochemical

1 INTRODUCTION

A fundamental understanding of the properties of materials provides a general approach to determining their applications in different technological fields. A brief history of the CN material can be traced back to 1834 when Berzelius and Liebig developed a form of polymeric carbon nitride-like melon (LIEBIG, 1834). Later in 2009, Wang and co-workers reported for the first-time hydrogen generation using CN in the presence of visible light (Wang et al., 2009). From that moment onwards, CN is at the apex because of its properties such as having a layered structure with abundant hydrogen and covalent bonds, a narrow bandgap of 2.7 eV, where the highest occupied molecular orbital (HOMO) is at −1.3 V, and the lowest unoccupied molecular orbital (LUMO) is at +1.4 (vs. NHE, pH = 7) (Huang et al., 2020). The CN is one kind of robust material that has attracted attention due to the following reasons: The synthesis process is simple and employs low-cost raw materials, outstanding physicochemical stability, and a befitting electronic valence band structure (Li Y. P. et al., 2021; Fei et al., 2021). However, this composite also exhibits certain demerits in application like aggregation, the fast recombination rate of charge carriers, the hydrophobic character on the surface, low value of the surface area, and high costs of removal and reuse (Karimi-Nazarabad and Goharshadi, 2017; Li Z. et al., 2018; Song et al., 2021), thus limiting its applications. To address the above hindrance, various strategies have been developed that include doping with different elements (Wang et al., 2018; Huang et al., 2020; Tang et al., 2020; Aboubakr et al., 2021), construction of heterojunctions (Li et al., 2017, 2020b; Zhang et al., 2019; Cao et al., 2020; Bai et al., 2022), co-catalyst loading (Ye et al., 2017), and development of biocomposites of CN with different compositions of biomaterials (Faraji et al., 2018; Abdel-Moniem et al., 2021; Sher et al., 2021c), among which some are micro-fibrillated carboxymethyl cellulose (Gholami et al., 2020) and chitosan (Zhao et al., 2018). Conforming to CS being a biodegradable polymer with excellent water permeability, it has plenty of strengths like earth abundance, non-toxicity, low cost, and the ability to form a film (Jayash et al., 2021; Kibungu et al., 2021). As of now, CS, the most abundant linear polysaccharide in nature, has been studied to improve the properties of CN (Zhao et al., 2016; Ye et al., 2017; Duan et al., 2018; Li H. et al., 2018, Li et al., 2021 Q.-H.; Fan et al., 2021; Liu E. et al., 2021; Liu et al., 2021 X.-P.). The CS possesses certain unique characteristics such as biodegradability, high carbon content, low cost, rich yield, high concentrations of amino groups, excellent mechanical properties, and antibacterial properties (Crini and Badot, 2008; Liu E. et al., 2021; Liu et al., 2021 X.-P.; Hu et al., 2021; Jayash et al., 2021; Zou et al., 2022a). Therefore, researchers have been studying the addition of CS to CN to obtain improved properties and the usage in various applications such as degradation of organic pesticides (Vigneshwaran et al., 2019), heavy metals (Li Q.-H. et al., 2021), electrochemical determination of mercury (Amiri et al., 2016), electrochemical sensors (Amiri et al., 2016), CO_2 reduction (Hu et al., 2021), etc. There are a lot of reviews on the emerging material CN, but most of them do not fully emphasize its progress with CS. Our primary focus in this article is on summarizing the application studies of CN with CS.

2 DISCUSSION

2.1 Synthesis Methods

It is well known that CN can be synthesized using precursors rich in nitrogen like melamine, thiourea, and cyanamide. A large number of articles on the synthesis of CN can be found, for example, the traditional method of direct polycondensation (Wang et al., 2009; Li Z. et al., 2018, 2020a), the hydrothermal process (Zheng et al., 2015; Ong et al., 2016), the laser exfoliation method (Dong et al., 2014), the photodeposition method (Kumar et al., 2018), and pyrolysis (Mo et al., 2015; Song et al., 2022). This section focuses on the up-to-date synthesis strategies and intends to provide an approach to improve the properties of CN with CS. CS contains functional groups, such as -OH, $-NH_2$, and $-NHCOCH_3$, allowing their functionalization with CN through noncovalent and electrostatic interactions, hydrogen bonds, and Waals Forces (Li H. et al., 2018). The coupling between CN and CS depends on its synthesis method. In most two-step synthesis methods, CS acts as supporting material and does not involve in tailoring the bandgap energy. However, the presence of CS improves the properties of CN, as observed in **Table 1**. **Table 1** shows the summarized synthesis methods and the properties observed. For example, CN and CS form aggregates easily, affecting the adsorption properties and making it difficult to recycle the material. Furthermore, CS suffers chemical damage in an acidic pH. Qing-Hao et al. synthesized nanofibers of CS/CN/TiO_2 by a two-step method of electrospinning (Li Q.-H. et al., 2021). This method demonstrates that it is possible to create materials with a high specific area, and also proves to be a promising and easy method for its separation from water. Besides, the addition of CN/TiO_2 to CS improves the chemical damage that is caused by acidic solutions. In addition, the CN ultrathin layer sheet has a smooth surface, while the biocomposite CN–CS presents a rough one, causing an increase in the specific surface area (Liu X.-P. et al., 2021). Different interesting synthesis methods for the composite have been reported so far. Recently, Vigneshrawan et al. reported the synthesis of the CS/CN, where acetic acid and glutaraldehyde were mixed by the Sol-Gel method (Vigneshwaran et al., 2019). The CS/CN combination was successful and has been confirmed with the presence of stretching and bending vibrations at 3,400 cm^{-1} and 1,079 cm^{-1} in FTIR spectra. Another report included the synthesis of CN–CS beads by cross-linking to form a network structure (Zhao et al., 2018), wherein the CS molecules interact with each other to attain the structure. This synthesis method is seen to provide an excellent renewable material. An interesting, easy, and one-step method is doping the CN semiconductor with carbon, using CS as the carbon source. The methodology shows that is possible to develop self-assembly aggregates to prepare C–CN since CS contains a high quantity of carbon (Li H. et al., 2018; Liu et al., 2019, Liu et al., 2021 E.). The substitution of an N atom by a C atom provokes delocalized big π bonds, forming a high electrical conductivity, an extended visible light absorption, and an increase in the mobility of photoinduced electron–hole pairs. However, this kind of material does not form a biocomposite of CN–CS. Therefore, the C–CN semiconductors are not discussed in the minireview, but they can be found in the table included because it is an interesting idea to use CS in self-assembly synthesis.

2.2 Applications

2.2.1 Environmental Pollution

In recent decades, environmental pollution has become a public health problem of particular concern since it affects living beings dangerously. Recently, a highly cited report was published in which Qing Haoli et al. studied the removal of Cr (IV) by CS/CN/TiO_2 nanofibers through adsorption and photocatalytic processes (Li Q.-H. et al., 2021). The adsorption and photocatalytic results were improved by the synergetic effect between CS and CN-TiO_2. For adsorption of Cr (IV), CS/CN/TiO_2 attained the highest adsorption capacity by the formation of chelates in contrast with CN/TiO_2, where the adsorption ability was negligible. For the photocatalysis process, the biocomposite of CS/CN/TiO_2 obtained a lower PL intensity and a 50% higher value of removal than the pure CN. This is following the lower

TABLE 1 | Synthesis, properties, and applications of biocomposites of CN and CS.

Biocomposite	**Synthesis**	**Type of binding**	**Properties**	**Application**
CS/CN composite (Vigneshwaran et al., 2019).	Direct polycondensation followed by the Sol-Gel method.	-OH and CH_2OH groups forms the binding between CN an CS.	• Bandgap of 2.7 eV. • Stable a low value of pH. • Unstable a high value of pH.	Removal of insecticides (chlorpyrifos).
CN/CS NCF (Thiyagarajan et al., 2020).	Calcination followed by the simple solution cast method.	Bond formation between $–NH_2$ and C-O and heptazine units.	• Bandgap of 2.65 eV. • The entrapment of CN into the CS matrix.	Photodegradation of methyl orange (MO).
CN–CS beads (Zhao et al., 2018).	Thermal polycondensation followed by the blend crosslinking method.	Bonding between $–NH_2$ and C-O and heptazine units.	• The CN is embedded in the CS matrix. • A low photoluminescence density.	Photodegradation of methylene blue (MB).
CN/SnS2/CS (Duan et al., 2018).	First, an electrode of CN/SnS2 is prepared and then the addition of CS.	Not reported.	• Excellent sensitivity, stability, repeatability, and dependability, as the proposed photoelectrochemical immunosensor.	Photoelectrochemical sensor for prostate-specific antigen.
Ag/CN/CS (An et al., 2017).	Direct heating followed by the phase chemical reduction method.	Not reported.	• Better separation efficiency of charges. • CS acts like as material support.	Photocatalytic reduction of 4-nitrophenol (4-NP).
Ag_2O/CN in Hydrogel of PVA/CS (Fan et al., 2021).	Condensation-polymerization. Then PVA and CS are mixed.	Bonding of $–NH_2$ and –OH with the semiconductors.	• Light weight • Self-floating • Good dispersion of the active material thought pores.	Photocatalytic inactivation of *Microcystis aeruginosa*.
Membrane of PVDF/CN/CS (Hassanzadeh et al., 2021).	Condensation-polymerization followed by the casting method.	Not reported.	• Addition of chitosan to the membrane increases hydrophilicity properties. • Biocomposite improves the mechanical properties of the membrane.	Remotion of Direct Blue 14.
Nanofibers of CS/CN/TiO_2 (Li et al., 2021a).	Electrospinning	Formation of heterojunction of TiO_2 and CN.	• Bandgap of 2.5. • Removal efficiency of 65% of removal of Cr (VI)	Remotion of Cr(VI) via adsorption/photocatalysis
C self-doped CN from CS (Li et al., 2018a).	Supramolecular self-assembly method	The substitution from N atoms to C atoms.	• Improved charge separation • H_2 evolution rate of 22 μmol h^{-2}	Photocatalytic H_2 production
C self-doped CN from CS (Liu et al., 2021a).	One-step copolymerization	Substitution from N atoms to C atoms.	• Specific area of 41.8 m^2g^{-1} • Pore widths in the interval from 2 to 15 nm. • Average pore diameter of 9.52 nm • visible-light absorption region to approximately 700 nm • Bandgap of 2.26 eV • The H_2-evolved rate of 61.2 $mmolh^{-1}$	Photocatalytic H_2 production
Ultrathin CN Nanosheets Grafted with Rare-Earth Up-Conversion Nanoparticles and CS (Zhao et al., 2016).	"green" liquid exfoliation route; thermal evaporation method	Chenical coupling between $–NH_2$ (or –NH) and –COOH.	• high optical transmittance (93.8% at550 nm) • excellent mechanical properties.	As a luminescent nano-paper with anti-counterfeiting effect for important documents
Film of CS with CN nanosheets (Liu and Wang, 2021).	Sonication-assisted exfoliation	Electrostatic interaction between the carboxyl and amine groups.	• The chitosan adjusted the distance among CN nanosheets. • An efficient and rapid fluorescence quenching.	A sensor for monitoring copper ions

bandgap obtained in the biocomposite (2.50 eV) compared to CN-TiO_2 (2.60 eV). On the other hand, Hassanzadeh et al. have prepared and characterized a composite of PVDF/CN/CS for the direct removal of blue 14 dye (Hassanzadeh et al., 2021). The increment in the percentage of addition of CS to the composite increased its hydrophilic nature, increasing the permeability, rejection of ions, high antifouling property, and removal of the dye. The main reason for this phenomenon is the enhancement of electrostatic force, which causes the adsorption of sodium ions and the repulsion of chloride ions on the surface of the membrane. For the degradation of insecticides like the chlorpyrifos (O, O-diethyl-3,5,6- trichloro-2-pyridyl phosphorothioate (CPFS)), CS/CN shows a maximum removal efficiency of 94% at a pH of 3–5 employing an indirect route where the reaction of $^{-}O_2 + H_2O_2 \cdot OH$ generates ·OH radicals, as CN does not oxidize directly (Vigneshwaran et al., 2019). The highest efficiency was obtained by the sample containing the pesticide in an aqueous solution containing organophosphate anions. The anionic ligand can donate a pair of electrons (Lewis's base), strongly adsorbing the cationic CS/CN biocomposite. Up to this point, the mechanism for the above examples is the following; first, CS attracts the molecules of interest to the surface by electrostatic forces, and this step is called "*in situ* adsorption". The next step depends on the molecule and the pH of the solution. Taking into consideration the band structure, the exposure of the biocomposite under light produces photo-generated electrons from Valence Band (VB) to form holes. Then, the photo-generated electrons jump to the Conduction Band (CB) for interaction with the molecules. For the molecule of Cr(IV) in an acidic pH, the photo-generated electrons formed on the surface and protons reduce to Cr(III). At a similar pH (~ 3–5 pH), the degradation of CPFS was achieved. The authors have described that CN cannot directly form •OH radicals, and hence, •OH radicals are produced in the route $^{-}O_2 + H_2O_2$ •OH, concluding that the main active species are holes and •OH radicals.

In this context, Zhao et al. have developed a regenerable photocatalyst based on CS–CN beads for methylene blue (MB) photodegradation (Zhao et al., 2018). The removal efficiency of TOC by CN photocatalysts reached a value of 40%. Another study reported the photodegradation of methyl orange (MO) using CN/CS NCF nanocomposites (Thiyagarajan et al., 2020), where the degradation of dyes up to 95% under visible light irradiation was shown by reusability studies. The highest TOC removal observed was 86%. The mechanism for photodegradation of MB and MO is similar. The first step is when the photoexcitation of CN occurs owing to visible light. The photo-generated electrons from the VB migrate to the CB, thus producing holes. These electrons can convert O_2 to $O_2^{-\bullet}$. At the same time, the holes created and the species of $O_2^{-\bullet}$ oxidate the dyes to produce -OH, CO_2, and H_2O. Considering the bandgap of 2.65 eV of the biocomposite (**Table 1**), the mechanism does not describe how the addition of CS improves photoactivity. CS is a superior material in terms of surface electron mobility (Senthil Kumar et al., 2015), and hence, the photo-generated electrons generated migrate from CN to CS, preventing the recombination of electron–hole pairs. Yet another example is the photocatalytic reduction of 4-nitrophenol (4-NP) to 4-aminophenol (4-AP), using sodium borohydride ($NaBH_4$) as the reducing agent and a visible-light-driven catalyst of Ag/CN/CS (An et al., 2017). The addition of Ag/CN/CS to the solution caused it to turn out colorless within 25 min, and the peak at 400 nm almost disappeared, after the appearance of a new peak at 300 nm (maximum absorbance of 4-AP). The authors assigned the catalytic property of Ag/CN/CS for the reduction of 4-NP to the transfer of electrons and hydrogen atoms from BH_4^- to organic compounds.

2.2.2 Biological

These biocomposites have been successfully tested in the elimination of gram-positive and gram-negative bacteria (Zhao et al., 2021; Ni et al., 2022). For example, Zou and co-authors synthesized membranes of CN and CS (CTS/CN/HPAN) by membrane ultrafiltration, and the antimicrobial activity of CTS/CN/HPAN membranes was studied using *Escherichia coli* as a model. The microbial suspensions were spread on sterilized CTS/CN/HPAN membranes, and the results were analyzed (Zou et al., 2022a). The antibacterial activity was determined by the content of colony-forming units and was compared with a control group and with CN/HPAN membranes. Using the CN/HPAN membranes (membranes without CS), it was possible to obtain a bacterial reduction of 80.4% after incubation for 24 h, while with the CTS/CN/HPAN membranes, a reduction of 95.6% was achieved. With these results, they confirmed that the use of CS did not reduce the effect but rather increased the antibacterial activity due to the presence of the nanosheets of CN and CS (Zou et al., 2022a). These results were shown to be better than those obtained by Reddy et al. who synthesized V-doped CN nano-architectures by direct calcination of urea and ammonium metavanadate. They tested its antibacterial effectiveness utilizing the percentage of antibacterial activity in suspension under visible light, using *Escherichia coli* as a model organism. As a result, they obtained ~90% inhibition with the V-doped CN while with the undoped CN only ~70% inhibition was obtained (Reddy et al., 2021).

Another example of CN biocomposites was the work of Ni and his co-authors. They manufactured biocomposite films of CS, CN, and curcumin (CS-HCNS-Cur) to test their antimicrobial activity. The CS-HCNS-Cur biocomposites were prepared by the solution casting method, and their antimicrobial activity was tested by inhibition zones and antibacterial efficiency using *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) as model organisms (Ni et al., 2022). As a result of their research, they were able to record an inhibition diameter of 8 and 8.5 mm for *E. coli* and *S. aureus*, respectively. While in the antibacterial activity, they had an antibacterial efficiency of ±85% in both bacteria, *E. coli,* and *S. aureus* (Ni et al., 2022). Presenting the greater antibacterial activity against *S. aureus* than against *E. coli*, this may be due to the structure of each of the bacteria because *E. coli* is a gram-negative bacterium and *S. aureus* is gram-positive (Parvathy et al., 2009).

Even though there is very little information on CN biocomposites with antimicrobial activity, the studies that have been carried out is shown to be competitive with composites and naked CN (Du et al., 2020; Qamar et al., 2020; Sun et al., 2020; Chen et al., 2021; Rao et al., 2021; Reddy et al., 2021; Xiong et al., 2021). It has been studied that the process by which they eliminate microorganisms is during irradiation, producing holes and electrons in the VB and CB of the photocatalyst. These charge carriers generate superoxides that primarily attack the membranes of bacterial cells, damaging these membranes and inactivating genetic material by disrupting phosphate and hydrogen bonds (Khan et al., 2018; Chen et al., 2019). The ·OH radicals and reactive H_2O_2 species are also produced, which participate in the photocatalytic inactivation of bacterial cells, mainly attacking the membranes of bacterial cells (Sher et al., 2021a; Sher et al., 2021b; Rao et al., 2021).

2.2.3 Sensors

Liu and Wang (Liu and Wang (2021) reported the first study for the detection of Cu^{2+} using the biocomposite film of CN nanosheet/CS. The film presented an absorption band at 240 nm in the presence of Cu^{2+}, high dispersion, and high fluorescence intensity due to the interaction between Cu^{2+} and groups of COOH and OH. The important remark is that CS adjusts the distance among CN nanosheets, causing the nanosheets to get closer between each. This closed stacking provokes an effective electron transport.

PEC sensing can encourage an analytical approach, revealing a superior performance. In addition, CN is nontoxic, has good biocompatibility, is inexpensive, and presents a great capacity to be used in a PEC sensor application. Recently, an ultrasensitive immunosensor photoelectrode based on FTO/CN/SnS_2 was developed for prostate-specific antigen (PSA) (Liu E. et al., 2021). The FTO/CN/SnS_2 achieves a wide detection range from 10 fg ml^{-1} to 10 ng ml^{-1}. In this context, the CdS@Au-CN photoelectrode was used to detect the same molecule (PSA) (Cao et al., 2020), showing that the PEC intensity was enhanced by the LSPR effect. Both studies presented the reduction in the recombination rate of photo-generated electron–hole pairs, obtaining a larger photocurrent. On the other hand, Liu Xing-Pei et al. constructed a biosensor with CN/CdS nanocomposites for detecting the urokinase-type plasminogen activator (u-PA) (Liu X.-P. et al., 2021). Their results show favorable characteristics like a wide detection range from 0.1 pg ml^{-1} to 1 μg ml^{-1} for target u-PA detection. Similarly, the $LaFeO_3$@CN photoelectrode had been fabricated for a sensor of streptomycin (Xu et al., 2020). The sensor showed high sensitivity and selectivity, and a low detection limit of 0.0033 nM. All examples showed good characteristics for a PEC sensor, such as repeatability, specificity, and stability. Overall, CS was used as a binding agent in all studies presented.

3 CONCLUSION

In a nutshell, CS improves the characteristics of CN according to the different synthesis methods such as good chemical stability, increased specific surface area, improved hydrophilic character, formation of nanosheets, extended light-capturing ability, and generation of terminal adsorptive bonds. Unfortunately, there is little research performed on the CS–CN biocomposite, and no one focuses on theoretical studies of the biocomposite. Therefore, the theoretical–experimental studies of the characterization and reaction mechanisms are necessary to achieve a complete understanding. On the other hand, the CS–CN can be considered as a new kind of biocomposite because it can act generally in two ways. CS can attract molecules on the surface of the biocomposite and produce primarily "an *in-situ* adsorption" and then the "interaction and participation" between reactions, considering that CS has high electron mobility. In comparison with the other conventional materials, the advantages of the biocomposite of CN and CS include high values of surface area and roughness, presence of mesopores, interconnection of pore channels, antifouling properties, and exhibiting a synergetic effect. Furthermore, it is possible to successfully synthesize the biocomposite owing to the high quantity of functional groups on their surface that aid strong hydrogen bond formation between both biomaterials (Li Q.-H. et al., 2021), electrostatic forces, and coupling chemicals (**Table 1**). Finally, a wide range of applications is described in our minireview, hoping that this will encourage you to explore the immense potential of CS–CN.

AUTHOR CONTRIBUTIONS

MAG-P and NAR-D organized and contributed to conception and design of the minireview. CRS-R summarized the information and wrote the first draft of the manuscript. PR, CAV-M, CGA-G, NAR-D, and CRS-R wrote section of the manuscript. All authors contributed to manuscript revision, read, and approved the submitted version.

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