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COMPARATIVE ANALYSIS OF METAL-DOPED GRAPHITIC CARBON NITRIDE SYNTHESIS

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ABSTRACT

An improved photocatalyst for visible light dye degradation, iron-enriched graphitic carbon nitride (Fe-g-C3N4) was synthesized and studied in this work. We hoped to overcome the limits of traditional g-C3N4 by improving its photocatalytic activities by adding iron to its structure. Scanning electron microscopy (SEM), Fourier-transform infrared spectroscopy (FTIR), and BET were used to characterize the Fe-g-C3N4, which was produced utilizing a generic and easily scalable technique. These studies proved that the g-C3N4 matrix was successfully doped with iron, leading to notable enhancements in optical and structural properties. Degradation of popular organic dyes under visible light irradiation was used to evaluate the photocatalytic performance of Fe-g-C3N4, which showed significantly greater activity compared to undoped g-C3N4. Doping with iron reduces the band gap, boosts absorption of visible light, and facilitates efficient charge separation and transfer; all of which contribute to the improved performance. This research highlights the remarkable capabilities of Fe-g-C3N4 as a photocatalyst for environmental applications. It suggests a possible solution for sustainable wastewater treatment and environmental remediation by efficiently degrading harmful dyes.

Keywords: Visible Light, Photocatalytic Activity, Metal, Charge Separation.

1.1 INTRODUCTION

Because of its exceptional photocatalytic capabilities and novel electrical structure, graphitic carbon nitride (g-C_3N_4) has become an extremely attractive material. Materials science research has focused on g-C_3N_4 frameworks because metal doping greatly improves these characteristics. Modified electrical, optical, and catalytic properties are achieved through metal-doped g-CሰN_4 synthesis, which entails adding different metal ions to the g-CሰN_4 matrix. This study intends to compare and contrast several metal-doped g-CሰN_4 synthesis processes, as well as the kinds of metal dopants used and how they affected the material's characteristics.

Thermal polymerization, sol-gel procedures, hydrothermal/solvothermal synthesis, chemical vapor deposition, and thermal polymerization are some of the synthesis methods used to accomplish metal doping in g-C_tN_4. In regard to the distribution of dopants, structural integrity, and scalability, each approach has its own set of benefits and drawbacks. The most popular approach, thermal polymerization, often uses metal salts or metal-organic frameworks as dopant sources and direct heating of precursors like dicyandiamide, melamine, or urea. Despite its rarity, chemical vapor deposition produces very uniform dopant dispersion and permits fine control over the doping process. Nanoparticles with improved catalytic activity are frequently produced via hydrothermal and solvothermal techniques, which employ highpressure and high-temperature environments to enable the incorporation of metal ions into the g-C_3N_4 lattice. You can regulate the porosity and dopant inclusion of the material very well using the sol-gel technique, which involves turning a colloidal solution into a solid gel.

The characteristics of the final g-C_3N_4 are greatly affected by the metal dopant that is used. The capacity of transition metals like Fe, Co, Ni, and Cu to create new electronic states inside the bandgap of g-C_3N_4 increases its visible light absorption and catalytic efficiency, making them frequently utilized. Despite their high price tag, noble metals such as platinum, gold, and silver have unmatched stability and catalytic performance. Researchers are interested in rare earth metals like Ce and La because of their unusual electrical structures and the possibility that they could form defect sites that serve as active catalytic centers.

By comparing different synthesis methods, this study not only shows how different g-C_3N_4 characteristics may be tailored to different applications, but it also emphasizes how important metal doping is. For better photocatalysis, energy conversion, and environmental remediation catalyst designs, scientists need to know how synthesis processes, metal dopants, and material properties relate to one another.

1.2 REVIEW OF LITERATURE

Palani, Geetha et al., (2022). Population and industrial expansion have emerged as key issues in the modern world's energy and environmental crises. Inadequate access to clean water is primarily caused by extremely toxic contaminants in water, including heavy metal ions, dyes, antibiotics, phenols, and pesticides. Here, the nonmetallic polymeric substance graphite carbon nitride (GCN or g-C3N4) has found widespread use as a photocatalyst that responds to visible light for a range of ecological purposes. This review revolves around the latest advancements in the design and photocatalytic uses of nanomaterials based on metal-doped GCN for CO2 photoreduction, water splitting to produce hydrogen, disinfection of bacteria, and degradation of organic pollutants. This paper also covers how GCN-based materials can be optimized for use in metal deposition, ion doping, dye sensitization, and environmental applications.

Sakuna, Pichnaree et al., (2022). The photocatalytic conversion of acetic acid to carbon dioxide under UV-visible irradiation has demonstrated remarkable potential in metal-doped graphitic carbon nitride (MCN) materials, which outperform virgin carbon nitride (g-C3N4, CN). We looked at the surface and bulk structures of metal-doped CN samples and how metal dopants affected their physicochemical properties as well as their catalytic activity in the photooxidation of acetic acid. Although the light absorption range was extended by Fe doping, the crystalline phase, shape, and specific surface area of the CN materials were not substantially changed by the addition of metals. Nevertheless, the improvement in photocatalytic efficiency could not be adequately explained by the expansion of visible light absorption due to Fe doping. The materials were examined utilizing two supplementary methods, electron spin resonance spectroscopy (ESR) and reversed double-beam photoacoustic spectroscopy (RDB-PAS), to gain a better grasp of this matter. This study's findings point to a relationship between the electron trap density (between 2.95 and 3.00 eV), the threshold number of free unpaired electrons in CN-based materials, and the rate of CO2 evolution via photocatalytic oxidation of acetic acid.

Syrgiannis, Zois & Christoforidis, Konstantinos. (2021). A very young subfield of materials research is known as two-dimensional materials (2D). Here, graphene's star structure paves the way for the development, study, and application of a wide variety of two-dimensional materials based on carbon and non-carbon elements. In the past ten years, the production of graphitic carbon nitride has been facilitated by the demand for graphene doped with heteroatoms. It has a high nitrogen to carbon ratio and a mix of graphitic and polymeric structures; it is two-dimensional. These substances are heterocycles containing carbon and nitrogen atoms coupled by sp^3-bonded nitrogen atoms or -NH-groups to heptazine or triazine rings. Papers have been published detailing several synthetic procedures that rely on poly-condensation reactions. Analysis of composition, X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), vibrational spectroscopy (Infra-Red, Raman spectroscopy), solid-state nuclear magnetic resonance (NMR), optical spectroscopies, EPR, and various microscopies (AFM, SEM, TEM) have all been employed to characterize the synthesized materials. Photocatalysis, energy storage, catalysts, catalyst support materials, and materials for biological applications (phototherapy, antibacterial) all rely on the characteristics of carbon nitrides, which are best achieved by synthetic techniques.

Zhao, yin et al., (2021). The breakdown of antibiotic pollutants under visible light using semiconductor photocatalysts has recently gained a lot of attention. In this study, a new photocatalyst for degrading tetracycline was effectively built using the 60 °C oil bath approach. The catalyst is cadmium doped graphite phase carbon nitride. In addition, researchers looked at how starting TC concentrations, catalyst dosage, pH, and other parameters affected the reaction. This study provides a solid foundation for the synthesis of cost-effective, high-efficiency photocatalysts.

Bai, Yuhan et al., (2021). Doping carbon nitride materials with metals is a typical way to control their molecular structure. Electrocatalysis, organic synthesis, biosensors, nanozymes, and photocatalysis are just a few of the exciting new areas that have recently explored the fascinating potential of metal-doped carbon nitride (M-CN) materials. From a purely theoretical standpoint, M-CN represents a crucial subset of carbon nitride materials; it paves the way for the synthesis of single-atom catalysts and gives these materials some amazing new abilities. Extensive experimental and theoretical evidence has shown that many M-CN materials contain M-Nx structures as active sites. The most recent developments in M-CN preparation, characterization, and its many uses are outlined in this review. The paper concludes with a discussion of potential future directions for the production and use of M-CN materials.

Zhang, Hongping et al., (2018). This research investigated the possibility of using metal-doped g-C3N4 as an extremely sensitive molecular sensor for NO2 detection through the use of density function theory (DFT) estimates. An assortment of g-C3N4 sheets doped with different metals were taken into consideration. Strong chemical linkages were observed to allow molecules of CO, CO2, NH3, N2, and NO2 to adsorb on metal-doped g-C3N4. Gas molecules that were adsorbed onto metal-doped g-C3N4 created charge transfer complexes, in which the metal-doped g-C3N4 transferred charges to the gas molecules. Based on the adsorption energy, density of states, and isosurface of electron density difference, pure and doped g-C3N4 sheets showed promise as gas molecule capturers. It was discovered that the NO2 molecule clearly affected the various metal-doped g-C3N4 sheets.

Dai, Hongzhe et al., (2013). The first step in creating graphitic carbon nitride (g-C3N4) sub-microspheres was a simple microwave synthesis involving a polymerization reaction of cyanuric chloride (C3N3Cl3) and sodium azide (NaN3) in acetonitrile (ACN) as the solvent. The results indicate that g-C3N4 cannot be dissolved in most common solvents, with the exception of DMSO. It has excellent chemical and thermal stability (below 650 °C), a particle size ranging from 0.076-0.137 µm, a surface area of 89.1 m2/g, and a band gap of 2.41 eV. In comparison to the conventional solvothermal method, the microwave method of preparing g-C3N4 yields a material with superior thermal stability, smaller particle radius, bigger surface area, narrower band gap, and stronger emission intensity. Lastly, we also suggest a way to influence the behavior of C3N4 sub-microspheres using microwaves.

1.3 MATERIAL AND METHODS

EXPERIMENTAL

In this experiment, melamine was utilized for g-C3N4 synthesis, FeSO4, CoSO4, and NiSO4(H2O) were bought from SDFCL in Mumbai, India, for the manufacture of corresponding metal doped g-C3N4, and Congo red (CR) (C32H22N6O6S2Na) was supplied by SRL in Taloja, India. We did not purify any of the precursors before using them because they were all of laboratory quality. Every process involved the use of distilled water. We used a set quantity of melamine as a precursor and heated the photocatalysts to 600 °C for 6 hours using the thermal condensation method. Metal doping causes modifications and changes in formation, as shown in Figure 1.1.

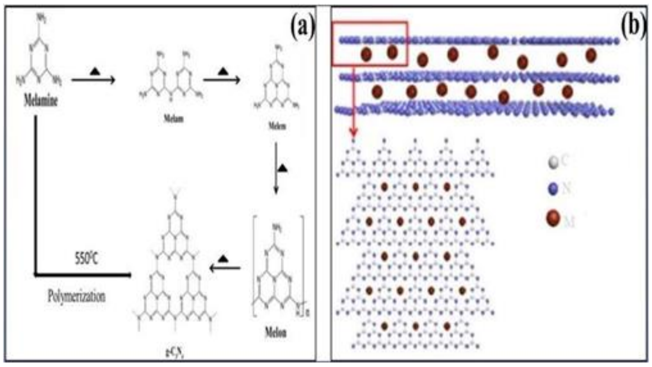


Fig.-1.1: Formation of (a) Undoped g-C3N4 and (b) Change in Structure after Metal Introduction

A solution of nitric acid (0.1mol/L) was used to rinse the produced yellow powder. A metal complex was added to the melamine in the same way, with 5% weight of melamine. A slurry was formed by adding cc of water to guarantee adequate mixing. In order to evaluate the photocatalytic activity of g-C3N4, a common hazardous pollutant, a congo red (CR) dye solution was utilized in a photoreactor that used visible light.

A photocatalyst is introduced to the solution before the photocatalytic reaction starts in order to achieve the adsorption-desorption equilibrium. Centrifuge the mixture at a steady speed for 5 minutes after the reaction is finished to separate the photocatalyst particles. A UV-Vis spectrophotometer set at 497 nm was used to measure the CR concentration.

Characterizations

Rigaku, Lead X-ray diffraction (XRD) spectra were utilized to investigate the catalyst's structure. The JCM-6000 Plus Bench Top Sem Neo scope, manufactured by JEOL Asia PTE Ltd., was used to obtain the SEM pictures. The Nicolet iS5 from THERMO Electron Scientific Instruments, LLC (model no.) was used to plot the FTIR spectra. The adsorption-desorption isotherms data was collected using the BELLSORP MAX II & BELCAT-II instruments by Microtrac BEL Corp, and the presence of metal doping in the catalyst was determined using EDX testing. We used Quanta 450 and EDAX for the EDX. Using a UV-visible spectrophotometer's absorbance as a function of wavelength, DRS was carried out to locate and ascertain the band gap.

1.4 EXPERIMENTAL RESULTS

FTIR Analysis

The FTIR spectra of the nanocomposites g-C3N4/Fe, g-C3N4/Co, and g-C3N4/Ni, shown in Figure-1.2, confirm that these elements are present in the powdered form, mostly as oxides. A notable bandwidth in the 3100-3400 cm-1 region, caused by the presence of moisture, is one of the unique features shown by the spectra. The stretching vibrations of primary (-NH2) and secondary (-NH-) amines, which come from residual amino groups in the heterocyclic CN structure's peripheries, are responsible for this. In addition, a peak at 1250 cm-1 shows that gC3N4 contains -CN- in amide. This peak disappears after a certain treatment, which is probably because a metal atom has been substituted for a nitrogen atom. It is worth mentioning that there are multiple absorption peaks between 1200 and 1640 cm-1, which can be associated with the stretching vibrations of C-N bonds and C-N bonds in the CN aromatic row.

In addition, absorption peaks at 885 cm-1 and 800 cm-1 confirm the presence of unique out-of-plane vibrations linked to the triazine/s-triazine aromatic repeating units.

It is worth mentioning that the wavenumber-wise peak locations in the spectra of g-C3N4/Fe and g-C3N4/Co are same. Having said that, their transmittance values are different. This indicates that, when contrasted with g-C3N4 doped with Co, g-C3N4 doped with iron has better transmission characteristics, showing more light absorption and higher transmittance.

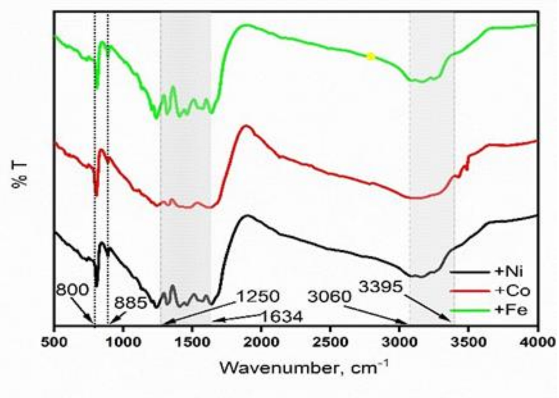
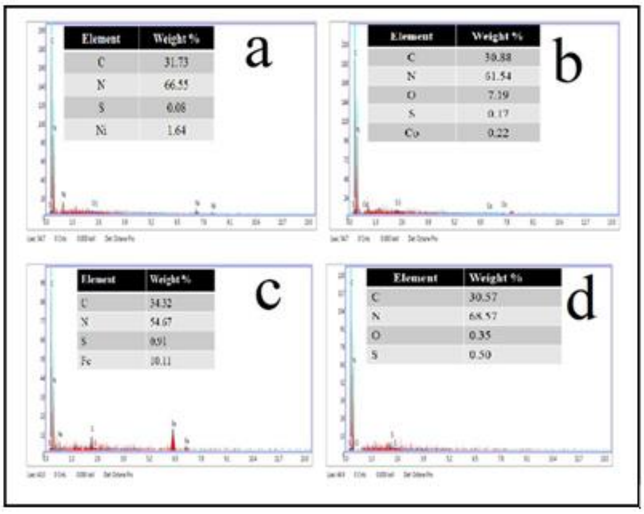


Fig.-1.2: FTIR Spectrum of g-C3N4 Doped with Fe, g-C3N4 Doped with Co, and g-C3N4 Doped with Ni EDX

Metal doping caused changes to the elemental composition of g-C3N4, as seen by the EDX study. For each metal, the percentage increase in weight (% wt) was as follows: Ni 1.64%, Co 0.22%, and Fe 10.1%, which is significantly greater. The substitution of iron atoms for nitrogen atoms yields the highest yield with the same amount of precursor, as seen by the significant increase in % wt for Fe.

The fact that Fe underwent a far more pronounced transformation than Ni and Co led to this conclusion.



Element	Weight %
C	31.73
N	66.55
S	0.08
Ni	1.64

Element	Weight %
C	30.88
N	61.54
O	7.19
S	0.17
Co	0.22

Element	Weight %
C	34.32
N	54.67
S	0.91
Fe	10.11

Element	Weight %
C	30.57
N	68.57
O	0.35
S	0.50

Fig.-1.3: EDX Spectrum of Graphitic Carbon Nitride Doped with (a) Ni (b) Co (c) Fe and (d) Undoped UV – DRS Spectra

The visible light absorbance of g-C3N4/Fe is greater than that of pure g-C3N4 and the highest of the three metals we tested, indicating that it is an active photocatalyst in this region. On the other hand, the photocatalysts exhibit a greater absorption of ultraviolet light (UV) at wavelengths below 380 nm, and their upward trend after 720 nm indicates that they are active in the infrared region of the solar spectrum

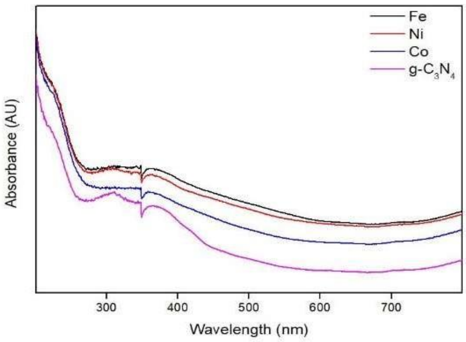


Fig.-1.4: UV-Vis Spectra of Prepared Photocatalysts

1.5 BET

The UV-Vis spectra results for g-C3N4 doped with iron were the best of the three, therefore researchers have been using it for things like BET characterisation, studying the effects of three parameters (catalyst dose, dye effects, and light intensity), and more. on Scales for adsorption and desorption are greater than those for pure g-C3N4. A mesoporous photocatalyst with an average diameter of around 35 nm and the ability to create a monolayer on its surface at p/p0 = 0.2 to 0.8 was demonstrated by both the particle size and the isotherm. Surface area ramped up to 36.372 m2/g from 18.467 m2/g, pore volume dramatically increased from 0.157 to 0.287 cm3/g, and pore diameter was lowered from 33.9 nm to 31.612 nm, according to adsorption-desorption tests. Gas adsorption reflects the impact of the surface area increase. Since nearly all adsorbed molecules desorbed, the desorption isotherm shows that there are plenty of sites available for photocatalytic reactions, and an increase in pore volume improves the number of sites available. Many particles in (c) and (d) have small pore sizes, as seen by the huge number of particles in the pore size distribution curve. On the other hand, when comparing g-C3N4/Fe to pure g-C3N4, gas adsorption rose from 100 cm3/g to 200 cm3/g, and cumulative pore volume quadrupled from 0.16 cm3/g to 0.30 cm3/g.

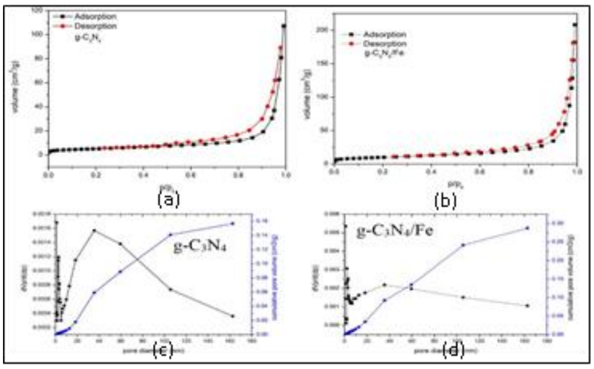


Fig.-1.5: BET Results: (a) Pure Graphitic Carbon Nitride and (b) Graphitic Carbon Nitride Doped with Fe; Pore Surface Area of (c) Pure Graphitic Carbon Nitride and (d) Graphitic Carbon Nitride Doped with Fe

While graphitic carbon nitride's activity was enhanced with the addition of metal, it demonstrates comparatively low activity in the VL area and higher activity in the UV. Fe had the most activity under VL conditions of the three metals.

Table 1.1: Percent Removal for Different Catalyst

Catalyst	Removalbyadsorption	Removalbyphotocatalysis
g-C3N4	0.20	0.21
g-C3N4/Ni	0.51	0.24
g-C3N4/Fe	0.15	0.60
g-C3N4/Co	0.25	0.35

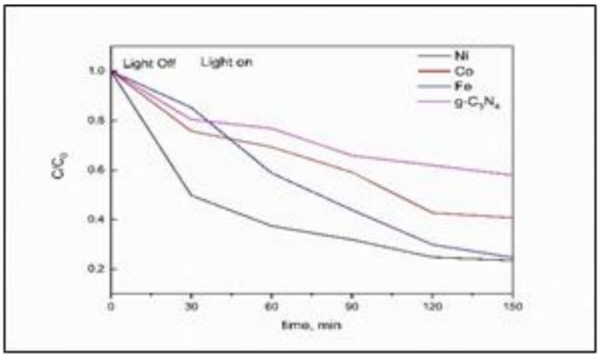


Fig.-1.6: Dye Degradation with Time

This figure compares pure g-C3N4 with g-C3N4 doped with various metals over time, showing the proportional changes in the concentration of a congo red dye solution (20 ppm). The catalyst was added to the dye solution and left alone for 30 minutes before the light was turned on. A decrease in concentration was seen prior to activating the light because of adsorption on the photocatalyst surfaces. An initial photocatalytic reaction was initiated upon illumination. The results demonstrated that with Ni, the change in concentration was most influenced by adsorption, but with g-C3N4 impregnated with Fe, the change in concentration was most influenced by photocatalysis. Both the light intensity and the amount of catalyst in the dye solution remain constant throughout the procedure.

Effect of Light Intensity

For photocatalysts to work better, the amount of light reaching them must be higher than their band gap energy. Figure 7 shows that iron doping causes a very noticeable change to the band gap of g-C3N4.

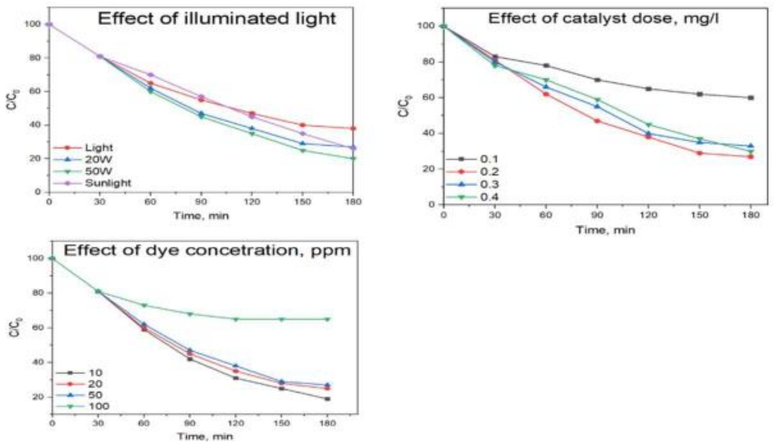


Fig.-1.7: Effect of Different Parameters on g-C3N4/Fe

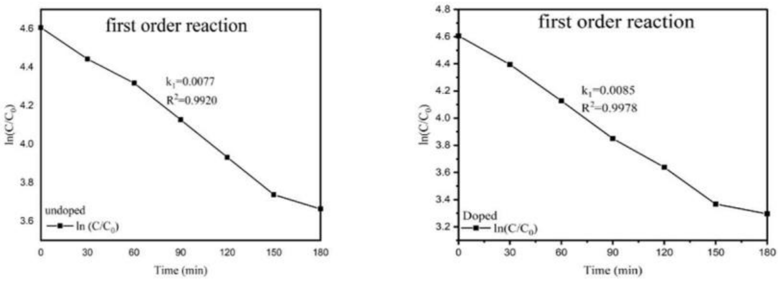


Fig.-8: Rate Constants of (a) g-C3N4 (b) g-C3N4/Fe

1.5 CONCLUSION

Ultimately, the study of metal-doped graphitic carbon nitride (g-C_3N_4) synthesis shows how various synthesis techniques and metal dopants significantly affect the material's characteristics and possible uses. The structural and electrical properties of the doped g-C_3N_4 are greatly affected by the synthesis approach chosen, which might range from thermal polymerization and chemical vapor deposition to hydrothermal/solvothermal techniques and sol-gel processes. Dopant dispersion, structural integrity, and scalability are three aspects that each approach improves upon, and these factors affect the material's performance as a whole.

The electrical, optical, and catalytic properties of g-C_3N_4 can be further tuned by adding different metal dopants, such as rare earth metals, transition metals, and noble metals. By creating new electronic states inside the bandgap, transition metals like Fe, Co, Ni, and Cu improve the absorption of visible light and the efficiency of catalysis. The increased expense is justified by the improved stability and catalytic performance offered by noble metals such as Pt, Au, and Ag. The particular benefits for certain applications are created by defect sites that operate as active catalytic centers, which rare earth metals like Ce and La generate.

This comprehensive study highlights the significance of comprehending the complex interplay between g-C_3N_4 synthesis techniques, metal dopants, and the consequent characteristics. Photocatalysis, energy conversion, and environmental remediation are just a few of the many uses for g-C_3N_4-based catalysts that may be created by optimizing these characteristics. Advancements in material science and solutions to global energy and sustainability concerns can be achieved through the ongoing investigation and improvement of metal-doped g-C_3N_4 synthesis.

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