AN INSIGHT INTO THE ANTIBACTERIAL ACTIVITY OF COPPER DOPED GRAPHITIC CARBON NITRIDE COMPOSITE

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Abstract. The synthesis and doping of graphitic carbon nitride (g-C_3N_4) with copper (ii) chloride dihydride ($CuCl_2.2H_2O$) to the corresponding copper doped graphitic carbon nitride (Cu-g-C_3N_4) composite and also the application of the as-synthesized composite as an antibacterial agent is described. The graphitic carbon nitride (g-C_3N_4) was synthesized from melamine via condensation and then converted to copper doped graphitic carbon nitride (Cu-g-C_3N_4) by treatment with different percentage (5%, 10% and 20%) of the metal precursor to obtained Cu-g-C_3N_4. The resultant composite was characterized using XRD, FTIR, and FESEM. The antibacterial assay of the composite was assessed against the standard laboratory strains of Staphylococcus aureus (gram +ve), Salmonella typhi (gram –ve), Escherichia Coli (gram –ve) and Streptococcus pyogenes (gram +ve) using well diffusion method. The as-synthesized composite shows a good antibacterial activity against the test organisms (gram +ve and gram –ve) with zone of inhibition ranging between 2-21 mm. The results were found to be appreciable when compared with that of the standard antibiotic (Ampicillin). These indicate that the as-synthesized composites have good potentials for developing new antibiotics which will help in reducing healthcare costs and mortality rate due to resistance of bacteria to already manufactured drugs.
Keywords: *metal doped, composite, antibiotics, bacteria, resistance, healthcare costs*

Introduction

Nature has given bacteria great ability to transmit from one host to another and also resist against the drugs that are used for its inhibition (Muhammad et al., 2020; Halilu et al., 2016). During the past decades, various forms of antibiotic drugs have been produced by pharmaceutical industries to address this issue but still there is rapid increased in drug resistance by the microorganisms (Sreenivasa et al., 2012). Antibiotic resistance has become a major issue in today's generation which has led to increase in health care cost and mortality rate. Therefore, there is urgent need for the development of new drugs to address these challenges. In response to these challenges, graphitic carbon nitride (g-C_3N_4) has been extensively studied in many fields like photo-catalysis and photo-electrochemical water splitting (Shao et al., 2010), CO_2 reduction (Yang et al., 2013), and photocatalytic degradation of pollutants (Wang et al., 2015). Later on, it was widely studied as a versatile support for noble metals to accomplish photocatalytic hydroxylation of benzene, oxidation, and hydrogenation reactions, Suzuki and Sonogashira couplings, and Knoevenagel reaction because of its interesting electronic and optical properties (Verma et al., 2016). Several functional groups such as amines, polyethyleneimine (PEI), hydrazine, boronic acid, and phenyl groups have also been tethered with g-C_3N_4 for CO_2 capture, CO_2 reduction, water splitting to H_2 or even to enhance it photoluminescence and sensing properties (Li et al., 2016; Zhang et al.,

2013). In recent years, graphitic carbon nitride (g-C_3N_4) has also attracted great interest in the field of biomedicine due to its unique elemental composition and photoelectric features. Due to its carbon and nitrogen content, it possesses an outstanding biocompatibility, which makes it beneficial and suitable in the field of biomedicine. The fluorescent characteristics of g-C_3N_4 can be applied to biological imaging. Furthermore, its appropriate energy level (2.7 eV) can promote the deposition of electrons, making g-C_3N_4 useful for antibacterial materials and photodynamic therapy (PDT) (Liu et al., 2022). Recently, graphitic carbon nitride has been found to have great antibacterial activity against pathogenic bacteria (Klebsiella pneumonia and Escherichia coli) in wastewater (Rattan Paul and Nehra, 2021). Also nitrogen-plasma-treated g-C_3N_4 nanosheets were reported to have lipid heads, thus, resulting in an excellent antibacterial activity (Cui et al., 2019). More researchers have already documented the efficacy of g-C_3N_4 in eradicating Escherichia coli and Staphylococcus aureus under visible conditions (Thurston et al., 2017). In addition to these, the NH_2 functional group engineering of g-C_3N_4 to the corresponding quaternary ammonium hydroxide (g-C_3N_4-OH) as solid-base and its application as an antibacterial agent has been reported (Muhammad and Usman, 2023). But there is no report on the use of metal doped graphitic carbon nitride as an antibacterial agent.

Inspired by the previous literature reports and our continue interest in the development of new antibiotics prompted us on the synthesis and doping of graphitic carbon nitride (g-C_3N_4) using copper to the corresponding copper doped graphitic carbon nitride (Cu-g-C_3N_4) and also on the application of the as-synthesized composite as an antibacterial agent. The graphitic carbon nitride (g-C_3N_4) was initially synthesized from melamine via condensation and then converted to copper doped graphitic carbon nitride (Cu-g-C_3N_4) by treatment with different percentage of copper (ii) chloride dihydride as a metal precursors ($CuCl_2.2H_2O$) to obtain Cu-g-C_3N_4 respectively as represented in (*Figure 1*).

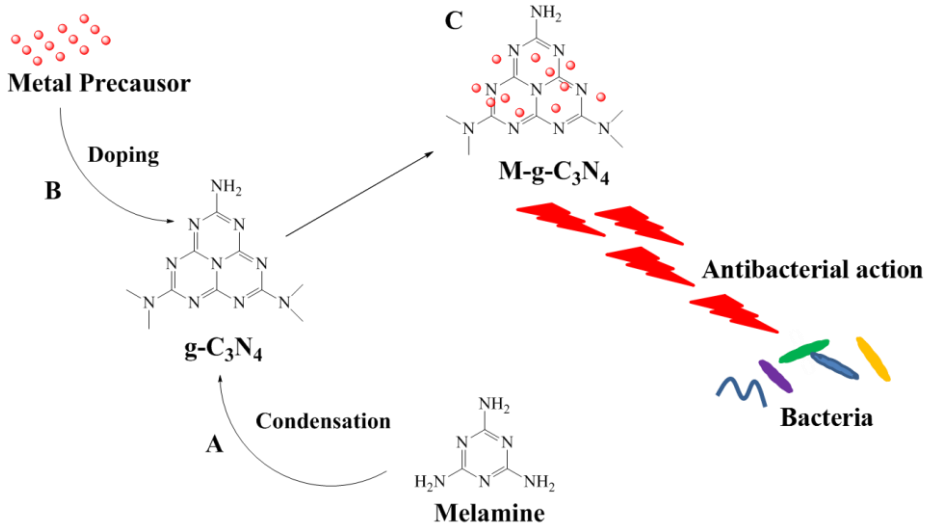

Figure 1. *Synthesis of copper doped graphitic carbon nitride (Cu-g-C3N4) composite and its antibacterial action.*

Materials and Methods

Melamine, copper (ii) chloride dihydride and all solvents were obtained from LobaChemie and used as received. Mular Hilton nutrient agar and broth were used for antibacterial analysis. Powder X-ray diffraction (XRD) was carried out using a Bruker diffractometer (D8 Advance, Davinci) with CuKα rays (λ=1.5418Å). The FTIR measurements was carried out on Bruker α-Eco-ATR IR spectrometer using ZnSe crystal in the wavenumber ranging from 400-4000 cm^{-1}. The morphology was observed with field emission scanning electron microscope (FESEM, JEOL JEM- 2100Plus). The test organisms were obtained from the Department of Microbiology, Faculty of Science, Sokoto State Univesity Sokoto, Nigeria. The microorganisms were standard laboratory strains of Staphylococcus aureus (gram +ve), Escherichia coli (gram –ve), Streptococcus pyogens (gram +ve) and Salmonella typhi (gram –ve).

Synthesis of g-C_3N_4

To synthesize graphitic carbon nitride (g-C_3N_4), 20 g of melamine was taken in a crucible and covered completely to avoid escape of fine particles. The crucible was then be transferred into a maffle furnace and heated at a temperature of 550 ^oC for 3 hours at a heating rate of 3 degree per minute. After cooling to normal room temperature, the resultant pale yellow material was obtain and grounded very well into fine powder using mortar and pestle, weighed and transferred into container which was stored in a desiccator for further treatment.

Conversion of g-C_3N_4 to Cu-g-C_3N_4

To a mixture of methanol (25.0 mL) and graphitic carbon nitride (1.0 g) in a round bottom flask, 0.1 g of $CuCl_2.2H_2O$ was added and covered with foil paper supported by a masking tape to avoid vaporization of the solvent. The content in the flask was then be stirred for 24 hours at 25 ^oC and then centrifuge at 3000 rotation per minute (rpm) for 5 minutes, then wash thoroughly with ethanol to remove excess chloride ions to obtained the copper doped graphitic carbon nitride composite (Cu-g-C_3N_4). The composite was dry in an air circulating oven for 24 hours at 80 ^oC, grounded to fine powder and then stored for further use. In addition to these different concentrations of metals loading was used to obtain 5%, 10% and 20% concentration of each metal on the graphitic carbon nitride. The resultant composite material was characterized using FTIR, XRD, and FESEM and also subjected to antibacterial activity test.

Antibacterial tests

The antibacterial test was conducted using the well diffusion method described by Finch et al. (2010). The Nutrient Agar plates was prepared according to manufacturer's instruction and was allowed to solidify for 15 minutes at room temperature and incubated without inoculum for 24 hours at 37^oC to ensure the sterility of the medium. The dilution ratio for Gram-positive bacteria and Gram-negative bacteria was 1:1000 and 1:5000, respectively, using normal saline water. The Nutrient Agar plates were flooded with 1 mL of the inoculum and the excess was removed using Pasteur pipette. Four wells (cups) of about 4 mm in diameter was cut on each Nutrient Agar plate using a sterile cork borer and the agar plugs was removed using sterile ampoule file. The composite solution (0.5 mL) was placed in each of the wells and allowed to settle for

two hours at room temperature before incubation for 24 hours at 37^{o}C. The inhibition zone was observed and then recorded in millimetre using a transparent ruler. Standard antibiotic (Ampicillin) was used as reference.

Minimum Inhibitory Concentration (MIC)

This was carried out as described by Usman and Osuji (2007). Minimum Inhibitory Concentration (MIC) was defined as the lowest concentration where no visible turbidity would be observed in the test tubes. The MIC was determined for the micro-organisms that showed reasonable sensitivity to the test composites. In this test, the micro-organisms were prepared using the broth dilution technique. The stock composite concentration of 10 mg/mL, 15 mg/mL and 20 mg/mL were made by dissolving 0.1g, 0.15g and 0.2g, respectively, of the composite in 10 ml of sterile distilled water and the working concentrations prepared by two-fold serial dilution technique that ranged from 0.098 mg/mL to 50 mg/mL using nutrient broth and later inoculated with 0.2 mL suspension of the test organisms. After 24 hours incubation at 37°C, the tubes were observed for turbidity. The lowest concentration where no turbidity is observed was taking and noted as the Minimum Inhibitory Concentration (MIC).

Minimum Bactericidal Concentration (MBC)

The minimal bactericidal concentration were determine from broth dilution test resulting from the MIC tubes as described previously by inoculating the content of each test tube on a nutrient agar plate. The plates were then incubated at 37°C for 24 hours. The lowest concentration of the composite that showed no growth was noted and recorded as the minimum bactericidal concentration (Usman and Osuji, 2007).

Results and Discussion

Materials characterization

The crystallinity of pure g-C_3N_4 and Cu-g-C_3N_4 were determined using powder X-ray diffraction (*Figure 2*). The pure g-C_3N_4 shows a peak of high intensity at 27.4º that correspond to (002) plane, which is due to the interlayer-stacking of graphite (Sanad et al., 2023; Pan et al., 2022; Wu et al., 2018; Elavarasan et al., 2016; Han et al., 2014; Chu et al., 2012); whereas other peak that clearly appears at 13.1º with very low intensity corresponds to (100) plane and is assigned to the motif structural packing of in-plane tris-s-triazine. On conversion to Cu-g-C_3N_4 as shown in *Figure 2*, the intensity of (100) and (002) peak planes in the Cu doped g-C_3N_4 sample indicated an improved crystallinity relative to undoped g-C_3N_4. No powder XRD peaks corresponding to Cu metal was detected as expected due to the low Cu loading. Nevertheless, a slight shifting of (002) peak to a lower 2 theater angle was observed for Cu doped g-C_3N_4 sample, which suggested some modification of the graphitic stacking of g-C_3N_4 as a consequence of Cu doping resulting in an increased interlayer distance (Zhang et al. 2018). However, varying the Cu dopant does not result in any significant differences in the diffraction patterns. The X-ray diffraction results indeed confirm the high stability of g-C_3N_4 toward metal loading. The FTIR spectra of pure and Cu doped composite are shown in *Figure 3*. The broad peak absorption observed at 2900-3400 cm^{-1} is due to the presence of N-H stretching vibration (Liu et al., 2014). The peaks observed between 1100-1700 cm^{-1} are characteristics peaks of tris-s-triazine assigned to C=N and C-N

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heterocyclic aromatic rings units as observed in the case of XRD (Yang et al., 2013). The intensity of these peaks decreases with increasing metal content (i.e Cu), suggesting that g-C_3N_4 contains numerous triazine rings that comprise Sp^2 C-N bond such as N=C-N and C–N=C. However, the graphitic structure of g-C_3N_4 was partly damage by metal doping. In particular, some triazine units were broken by deamination upon metal precurssor addition. All these results imply that the g-C_3N_4 frame work partly changed upon metal doping. Some triazine rings were broken and Sp^2 C-N bonds were transformed into C≡N triple bonds (Thomas et al., 2008; Montigaud et al., 2000).

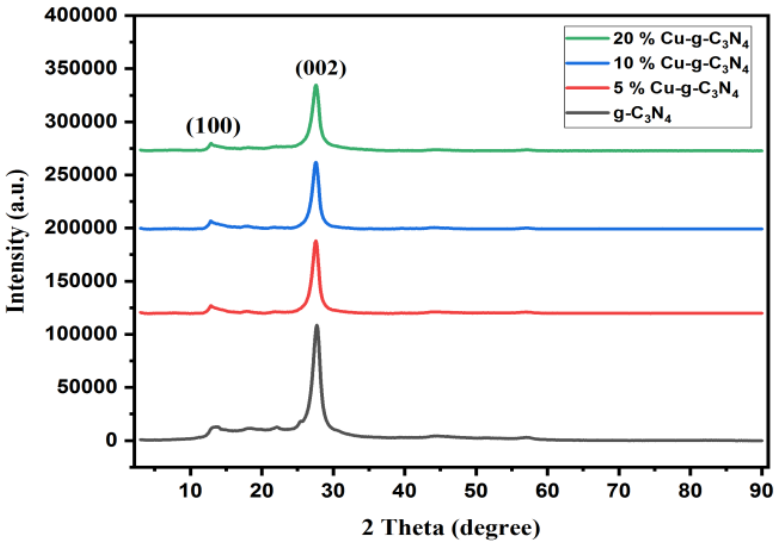


Figure 2. *XRD patterns of g-C_3N_4 doped with various amount of Copper.*

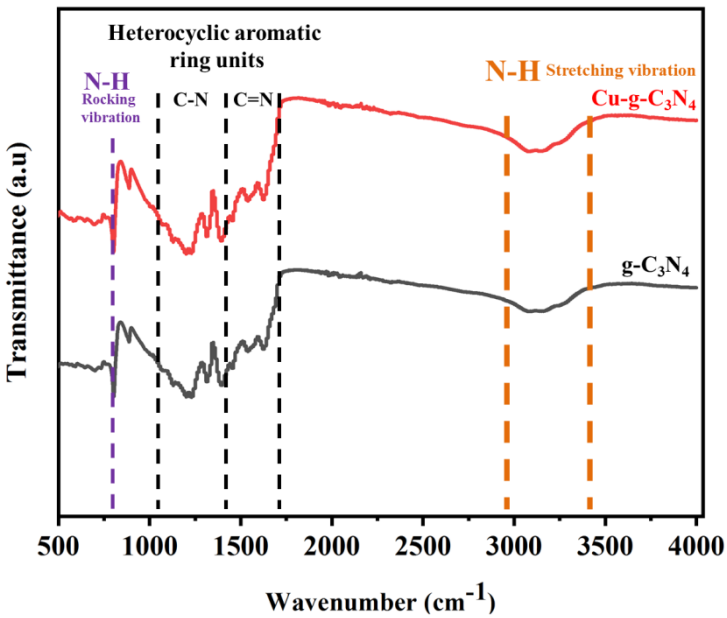
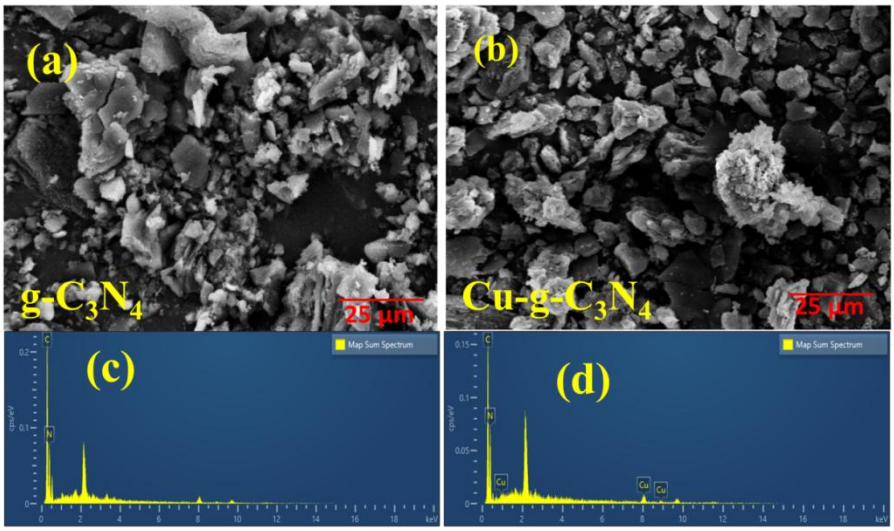


Figure 3. *FTIR spectra of g-C_3N_4 and Cu-g-C_3N_4.*

The author further investigated the morphological features with scanning electron microscope (SEM) and EDX. Scanning electron microscope of pristine graphitic carbon nitride (g-C_3N_4) and copper-doped graphitic carbon nitride (Cu-g-C_3N_4) composite showed rod-like morphology (*Figure 4(a) and Figure 4(b*); the particle size of pristine graphitic carbon nitride (g-C_3N_4) was found to be between 1-8 µm in length with about 150-135 nm thickness: whereas the post-synthetically modified Cu-g-C_3N_4 composites were to be in the range of 1-6µm in length. The morphological features were further investigated using EDX, the presence of C, N and Cu in the pure g-C_3N_4 and modified g-C_3N_4 samples can be confirmed using energy dispersive spectroscopy (EDX). The results of EDX analysis (*Figure 4(c) and Figure 4(d*) unambiguously indicated the presence of copper together with C and N in the as-synthesized composite.



Map Sum Spectrum	
Element	Atomic %
C	44.92
N	55.08
Total	100.00

Map Sum Spectrum	
Element	Atomic %
C	42.28
N	57.59
Cu	0.13
Total	100.00

Figure 4. *SEM Images of (a) g-C_3N_4 (b) Cu-g-C_3N and EDX images of (c) g-C_3N_4 (d) Cu-g-C_3N_4.*

Antibacterial activity of g-C_3N_4 and Cu-g-C_3N_4

After characterizing the g-C_3N_4 and Cu-g-C_3N_4, the efficacy of the composite was investigated against standard laboratory strains of Staphylococcus aureus (gram +ve), Escherichia coli (gram –ve), Streptococcus pyogens (gram +ve) and Salmonella typhi (gram –ve). *Table 1*, *Table 2* and *Table 3* show the details of the results obtained from the antibacterial activity test of pure g-C_3N_4 and Cu-g-C_3N_4 composite and the standard antibiotic (Ampicillin). *Table 1* shows the detailed of the antibacterial activity of copper doped graphitic carbon nitride (Cu-g-C_3N_4) composite. The pure g-C_3N_4 shows no activity on all the test organisms while Cu-g-C_3N_4 has activity on all the bacterial strains with zone of inhibition ranging from 2.00-21.00 mm. *Table 2* show the detailed of the antibacterial activity of standard antibiotic (Ampicillin) which has activity on all the bacterial strains with zone of inhibition ranging from 15.00-22.00 mm. The result indicate that pure g-C_3N_4 has no antibacterial activity on all test organisms. The results deduced that Cu-g-C_3N_4 has activity on all the bacterial strains with zone of inhibition

ranging from 2.00-21.00 mm. The Cu-g-C_3N_4 composite exhibited excellent antibacterial activity on all the test organisms with zone of inhibition ranging between 2.00 mm to 21.00 mm (*Table 1*). The composite showed a high level of activity on both gram negative bacteria and gram positive bacteria. The results when compared with Ampicillin (Standard Antibiotic as in *Table 2*, the zone of inhibition produced by the antibiotic against the test organisms was found to be very appreciable in relation to those activities produced by the composite under study. Furthermore, according to Usman and Osuji (2007), diameter of zones of inhibition ≥10 mm is considered active. The as-synthesized composite (Cu-g-C_3N_4) was subjected to MIC and MBC determination (*Table 3*). The results of MIC and MBC in *Table 3* showed that Cu-g-C_3N_4 composite have an MIC of 100 mg/mL on S. pyogens, 55.00 mg/mL on S. aureus, 50.00 mg/mL on both E. coli and S. typhi and MBC obtained 100 mg/mL on S. pyogens, 50.00 mg/mL on both E. coli, S. typhi, S. aureus

Table 1. *Antibacterial activity of copper doped graphitic carbon nitride (Cu-g-C_3N_4).*

Metal loading	Concentration (mg/mL)	Zone of inhibition (mm)			
		E.coli	S. typhi	S. aureus	S. pyogens
5%	25.00	2.00	0.00	2.00	0.00
	50.00	4.00	0.00	6.00	0.00
	100.00	6.00	5.00	8.00	2.00
10%	25.00	4.00	6.00	8.00	4.00
	50.00	8.00	7.00	10.00	8.00
	100.00	14.00	10.00	12.00	12.00
20%	25.00	7.00	12.00	12.00	10.00
	50.00	10.00	16.00	18.00	12.00
	100.00	20.00	18.00	21.00	19.00
g-C_3N_4	25.00	0.00	0.00	0.00	0.00
	50.00	0.00	0.00	0.00	0.00
	100.00	0.00	0.00	0.00	0.00

Table 2. *Antibacterial activity of standard antibiotic (Ampicillin).*

Antibiotic concentration	Zone of Inhibition (mm)			
	E.coli	S. typhi	S. aureus	S. pyogens
Ampicillin (50 mg/mL)	15.00	15.00	21.00	22.00

Table 3. *Minimum Inhibitory Concentration (MIC) and Minimum Bactericidal Concentration (MBC) of copper doped graphitic carbon nitride (Cu-g-C_3N_4).*

Test organism	MIC (mg/mL)	MBC (mg/mL)
E.coli	50.00	50.00
S. typhi	50.00	50.00
S. aureus	55.00	50.00
S. pyogens	100.00	100.00

Conclusion

The author has described a simple and efficient method of doping graphitic carbon nitride g-C_3N_4 to corresponding (Cu-g-C_3N_4) by treating the graphitic carbon nitride g-C_3N_4 with metal precursors ($CuCl_2.2H_20$). The as-synthesized composite was characterized using XRD, FTIR, SEM and EDX and the results shows that the graphitic carbon nitride g-C_3N_4 retains its structural integrity even after doping with different

percentage of metal. The evaluation of antibacterial activity of the Cu-g-C_3N_4 composite using some selected bacterial strains (Staphylococcus aureus (gram +ve), Escherichia coli (gram –ve), Streptococcus pyogens (gram +ve) and Salmonella typhi (gram –ve) was found to be appreciable when compared with the standard antibiotic (Ampicillin). The results were found to be very useful in academics and can be extended to pharmaceutical applications as well. The findings of this research have significant importance for addressing the growing threat of antibiotic resistance.

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Conflict of interest

The authors confirm that there is no conflict of interest involve with any parties in this research study.

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