

C-doped g-C₃N₄@Polyaniline (PANI) Composites as High-Performance Electrode for Supercapacitor Applications

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ABSTRACT

Graphitic carbon nitride (g-C₃N₄) is a promising supercapacitor electrode material owing to its layered structure and nitrogen-rich framework, but its limited conductivity and moderate specific capacitance restrict practical applications. This study reports C-doped g-C₃N₄@Polyaniline (PANI) composites for supercapacitor electrodes. Undoped g-C₃N₄ was synthesized via thermal polymerization of melamine at 550°C, while C-doped g-C₃N₄@PANI composites with PANI loadings (3.2, 6.3, and 9.1 wt%) were prepared by ultrasonic-assisted wet impregnation. X-ray diffraction confirmed the preserved g-C₃N₄ structure with peak shifts and broadening, indicating successful carbon doping and reduced crystallite size. Fourier Transform Infrared spectroscopy verified the chemical integrity of both components and revealed strong hydrogen bonding between g-C₃N₄ and PANI. Field Emission Scanning Electron Microscopy showed morphological evolution from flake-like sheets to PANI-coated structures. Electrochemical measurements showed the 3.2 wt% PANI composite achieving the highest specific capacitance of 45 F/g (CV) and 42 F/g (GCD), compared to 25 F/g (CV) and 23 F/g (GCD) for undoped g-C₃N₄, with reduced charge transfer resistance and improved ion diffusion. The synergistic combination of electric double-layer capacitance from g-C₃N₄ and pseudocapacitance from PANI positions these composites as promising electrode materials for next-generation supercapacitors.

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INTRODUCTION

The escalating global energy crisis and the growing environmental concerns associated with the depletion of fossil fuels have necessitated the urgent development of sustainable, efficient, and environmentally friendly energy storage technologies (Gomari et al., 2024; Mohammed et al., 2024; Zakariyya et al., 2024; Umaru et al., 2023). Among the various electrochemical energy storage systems, supercapacitors have garnered significant attention due to their exceptional advantages, including high power density, rapid charge-discharge capabilities, excellent cycling stability, and environmental benignity (Kumar et al., 2023). These devices bridge the gap between conventional capacitors and batteries, offering intermediate energy density while maintaining superior power delivery. However, the relatively low energy density of supercapacitors compared to batteries remains a critical challenge limiting their widespread commercialization, necessitating the development of advanced electrode materials with enhanced electrochemical performance (Du et al., 2023).

Graphitic carbon nitride (g-C₃N₄) has emerged as a promising electrode material for supercapacitors due to its unique two-dimensional layered structure, nitrogen-rich composition, excellent chemical stability, and low-cost synthesis (Dankawu et al., 2023). The tri-s-triazine units in g-C₃N₄ provide abundant nitrogen-containing functional groups that

contribute to pseudocapacitive charge storage through reversible redox reactions, while its layered morphology facilitates ion intercalation and electric double-layer formation (Shuaibu et al., 2024). Despite these attractive properties, pristine g-C₃N₄ suffers from several inherent limitations, including poor electrical conductivity, limited surface area, and moderate specific capacitance, which significantly constrain its electrochemical performance and practical viability. Various strategies have been employed to overcome these limitations, including elemental doping, morphological modification, and the construction of composite materials with conductive polymers or carbonaceous materials (Aswathi & Sreeja, 2025).

Elemental doping has proven to be an effective approach for modulating the electronic structure and enhancing the electrical conductivity of g-C₃N₄. Carbon doping, in particular, introduces additional active sites and modifies the band structure by substituting nitrogen atoms in the g-C₃N₄ framework, thereby improving charge carrier mobility and electrochemical activity (Praveen et al., 2025). Simultaneously, conducting polymers such as polyaniline (PANI) have attracted considerable interest as supercapacitor electrode materials due to their high pseudocapacitive behavior, excellent electrical conductivity, facile synthesis, and environmental stability. PANI stores charge through reversible Faradaic redox transitions between its

leucoemeraldine, emeraldine, and pernigraniline states, involving the exchange of protons and electrons. However, PANI alone suffers from poor cycling stability due to volumetric changes during charge-discharge cycles, which can be mitigated by incorporating it into composite structures with robust supporting materials (Tchouank Tekou Carol et al., 2020).

The combination of g-C₃N₄ and PANI into composite materials offers a synergistic approach to developing high-performance supercapacitor electrodes. The layered g-C₃N₄ provides a stable structural scaffold and contributes electric double-layer capacitance, while PANI enhances pseudocapacitive charge storage and improves overall conductivity. The intimate interfacial contact between the two components facilitates efficient charge transfer, reduces internal resistance, and enhances the utilization of active sites. Furthermore, the incorporation of carbon doping into the g-C₃N₄ structure further enhances the electrical properties, creating a ternary synergistic system with superior electrochemical performance.

In this study, we report the synthesis and comprehensive characterization of C-doped g-C₃N₄@PANI composites with varying PANI loadings (3.2, 6.3, and 9.1 wt%) for high-performance supercapacitor electrodes. The undoped g-C₃N₄ was synthesized via thermal polymerization of melamine at 550 °C, while the composites were prepared using ultrasonic-assisted wet impregnation to ensure uniform dispersion and intimate interfacial contact. The structural properties were investigated using X-ray diffraction, chemical composition and functional groups were analyzed through Fourier Transform Infrared spectroscopy, and surface morphology was examined using Field Emission Scanning Electron Microscopy. The electrochemical performance was systematically evaluated using cyclic voltammetry, galvanostatic charge-discharge, and electrochemical impedance spectroscopy in a three-electrode configuration. The objectives of this research are to elucidate the synergistic effects between C-doped g-C₃N₄ and PANI, optimize the PANI loading for maximum electrochemical performance, establish structure-property-performance relationships, and demonstrate the potential of these composites as advanced electrode materials for next-generation supercapacitor applications. The findings of this study contribute to the development of sustainable, high-performance energy storage materials and provide valuable insights into the design and optimization of g-C₃N₄-based composite electrodes for practical supercapacitor devices.

MATERIALS AND METHODS

Chemicals and Reagents

Analytic grade high purity (98-99%) chemicals and reagents were used without further purification for the synthesis. Aniline monomer, Ammonium per sulphate (APS), Absolute Ethanol, Melamine (C₃H₆N₆), and Citric acid were purchased from LOBA Chemie while Hydrochloric Acid was purchased from MOLYCHEM.

Synthesis Method

Synthesis of Undoped g-C₃N₄

The un-doped g-C₃N₄ was synthesized using thermal polymerization method. Typically, 45 g of melamine was weighed and added into a beaker containing 200 ml of distilled water. The mixture was then stirred using magnetic stirrer for about three hours to obtain a thick white solution and then dried in an oven at 80 °C for 12 hrs until the water fully evaporates. The dried sample was then grinded using mortar and pestle and then annealed using a furnace for 4 hrs at 550 °C.

Synthesis of C-doped g-C₃N₄

C-doped g-C₃N₄ was synthesized using thermal polymerization method by weighing 70g melamine 5%wt of citric acid (as carbon source) into a beaker containing 200ml of deionized water (Figure 1 (a)). The mixture was stirred using a magnetic stirrer for about 3 hrs until a homogeneous thick white solution is obtained. Afterward, it was dried in an oven at 80 °C for 12 hrs, and then grounded using mortar and pestle to obtain the precursor material which was then heated in a muffle furnace at 550 °C for 4 hrs. Dark yellow C-doped g-C₃N₄ was obtained as the final product.

Synthesis of PANI

The synthesis of PANI was carried out using oxidative polymerization. Typically, 50ml of aniline monomer was added in 300ml of hydrochloric acid in an ice bath and stirred for 20 mins, this is termed as solution A. 60g of ammonium per sulphate (APS) was measured and dissolved in 200ml of distilled water in a beaker and then stirred until a clear solution was obtained this is termed as solution B. Solution B was then gradually poured into solution A, the mixture was then stirred for 3 hrs for complete polymerization. The mixture was then filtered and washed several times with water, and dried at room temperature. The final product was then grinded using mortar and pestle to obtain the PANI nanoparticles.

Synthesis of the C-doped g-C₃N₄@PANI Composites

The C-doped g-C₃N₄@PANI composites were synthesized via ultrasonic assisted wet impregnation method. PANI nanoparticles were then loaded onto the C-doped g-C₃N₄ and four samples were made by measuring 5 g of C-doped g-C₃N₄ in each case at 0% wt, 3.2% wt, 6.3% wt and 9.1% wt of PANI. In the 3.2% wt sample, 0.165 g of PANI was added to 5 g of C-doped g-C₃N₄ and mixed in 50ml water in a beaker, and sonicated for 30 minutes to obtain uniform dispersion. The sample was then stirred until the water completely evaporates. The sample was dried in an oven at 60 °C for 12 hrs to obtain the C-doped g-C₃N₄@3.2wt%PANI composites. The C-doped g-C₃N₄@6.3wt%PANI composites and C-doped g-C₃N₄@9.1wt%PANI composites were synthesized by adding 0.336 g and 0.500 g of PANI respectively (Table 1).

Table 1: Electrode parameters from GCD curve

Sample	Weight of C-doped g-C ₃ N ₄ (g)	Weight of PANI (g)	PANI wt%
Undoped g-C ₃ N ₄	0	0	0
C-doped g-C ₃ N ₄ @3.2 wt% PANI	5	0.165	3.2
C-doped g-C ₃ N ₄ @6.3 wt% PANI	5	0.336	6.3
C-doped g-C ₃ N ₄ @9.1 wt% PANI	5	0.500	9.1

Characterization Technique

The analysis of purity, crystallinity and structure of the synthesized materials was carried out using X-ray Diffraction (ThermoFisher Scientific ARL'XTRA X-ray) with Cu-K α radiation and a wavelength (λ) of 1.5418Å in the 2θ range of 10°-80°C. Chemical composition spectroscopy, as well as functional group identification of the prepared samples, were carried out using FTIR spectroscopy (on Agilent Technologies FTS-3000). Surface morphological characteristics of the prepared samples were observed through FE-SEM (with FEI Quanta FEG 200 HR-SEM). Electrochemical measurements, including cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS), were conducted on a Metrohm Multi-Channel Autolab (Model: AUT.MAC.204) electrochemical workstation. The experiments were executed in a standard three-electrode cell assembly utilizing the synthesized

material as the working electrode, a reference electrode, and a platinum wire counter electrode, with data collection managed via Metrohm NOVA software.

Preparation of the Electrode

Nickel foam (substrate) was purified via sonication process in diluted HCl, and then washed with water as well as acetone. The electrode ink was prepared by blending C-doped g-C₃N₄/PANI + Carbon Black + PVDF binder in the ratio of 8:1:1 followed by the addition of 10ml of NMP to the mixed composite. The uniform slurry was coated onto a nickel form substrate and then dried at 60° for 3hrs in a vacuum oven. The active mass utilized for the calculations was obtained by calculating the difference between the mass of the substrate before the depositing the electrode ink and the mass of the substrate after depositing the electrode ink.

RESULTS AND DISCUSSION

XRD Analysis

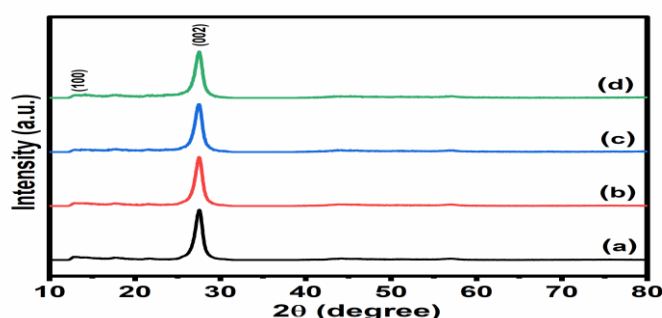


Figure 1: XRD pattern of (a) undoped g-C₃N₄, (b) C-doped g-C₃N₄@3.2wt%PANI, (c) C-doped g-C₃N₄@6.3wt%PANI, and (d) C-doped g-C₃N₄@9.1wt%PANI composites

The structural properties of undoped g-C₃N₄ and C-doped g-C₃N₄@PANI composites with varying PANI loadings (3.2, 6.3, and 9.1 wt%) were investigated using X-ray diffraction. Figure 1 (a), (b), (c) and (d) presents the XRD pattern of undoped g-C₃N₄, C-doped g-C₃N₄@3.2wt%PANI, C-doped g-C₃N₄@6.3wt%PANI, and C-doped g-C₃N₄@9.1wt%PANI composites respectively. The undoped g-C₃N₄ sample displayed two characteristic diffraction peaks at approximately 27.4° (002) and 13.1° (100), corresponding to the interlayer stacking of conjugated aromatic systems and the in-plane repeating tri-s-triazine units, respectively. The (002) peak yielded an interlayer spacing of approximately 0.326 nm calculated from Bragg's law, while the broad nature of these peaks confirmed the turbostratic structure with small crystallite dimensions typical of thermally polymerized g-C₃N₄. The absence of impurity peaks verified the successful synthesis of pure phase graphitic carbon nitride (Chetoui et al., 2025; Kundu et al., 2026).

For the C-doped g-C₃N₄@PANI composites, the characteristic g-C₃N₄ peaks were preserved in all samples, indicating that the fundamental crystalline framework remained intact despite PANI incorporation. However, systematic structural modifications were observed with increasing PANI content. The (002) peak exhibited slight shifts in 2θ position compared to undoped g-C₃N₄, which can be attributed to carbon doping-induced changes in interlayer spacing and lattice strain from PANI deposition on the surface. Progressive peak broadening was observed as PANI loading increased from 3.2 to 9.1 wt%, suggesting reduced crystallite sizes calculated from the Debye-Scherrer equation. This reduction results from restricted g-C₃N₄ crystal growth

during synthesis, disruption of ordered layer stacking by amorphous PANI, and introduction of structural defects through ultrasonic-assisted wet impregnation. The decreased crystallite size is electrochemically beneficial as it provides shorter ion diffusion pathways, greater active site exposure, and enhanced surface area for charge storage (Muthuganesh et al., 2025; Lü et al., 2025).

Analysis of interlayer spacing revealed systematic decreases in d-spacing with increasing PANI content, indicating slight compression of the interlayer distances upon carbon doping and PANI incorporation. This compression suggests stronger interlayer interactions and potential partial intercalation of PANI between g-C₃N₄ layers, creating intimate composite structures that facilitate charge transfer and ion transport. Notably, no distinct PANI diffraction peaks were observed in any composite sample, consistent with the amorphous nature of polyaniline synthesized via oxidative polymerization, the relatively low loading percentages below XRD detection limits, and the overlap of PANI's broad amorphous scattering (20-25° 2θ) with the g-C₃N₄ (002) peak. The broadening and slight asymmetry of the (002) peak at 9.1 wt% PANI may indicate underlying amorphous PANI scattering contribution (Tchouank Tekou Carol et al., 2020; Yin et al., 2020).

The observed structural modifications have significant implications for supercapacitor performance. Increased disorder and reduced crystallinity enhance pseudocapacitive behavior by providing more defects and active sites for Faradaic redox reactions essential for high specific capacitance. Reduced crystallite size facilitates faster ion diffusion and charge transfer kinetics, improving rate capability at high charge-discharge rates. Modified interlayer

spacing may optimize electrolyte ion intercalation/deintercalation, potentially enhancing charge storage capacity beyond individual components. The intimate interface between crystalline g-C₃N₄ and amorphous PANI creates heterojunction structures promoting efficient charge

transfer, reduced charge transfer resistance, and synergistic electrochemical activity. Additionally, carbon doping contributes to improved electrical conductivity, crucial for reducing internal resistance and achieving high power density.

FTIR Analysis

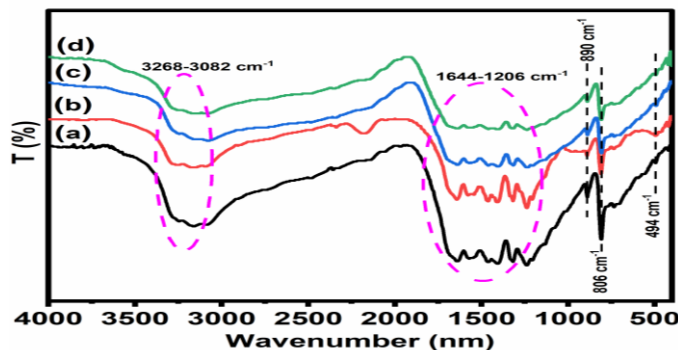


Figure 2: FTIR spectra of (a) undoped g-C₃N₄, (b) C-doped g-C₃N₄@3.2wt%PANI, (c) C-doped g-C₃N₄@6.3wt%PANI, and (d) C-doped g-C₃N₄@9.1wt%PANI composites

The FTIR spectra of undoped g-C₃N₄ and C-doped g-C₃N₄@PANI composites with varying PANI loadings (3.2, 6.3, and 9.1 wt%) provides essential information about chemical composition, functional groups, and molecular interactions between composite components. Figure 2 (a), (b), (c) and (d) presents the FTIR spectra of undoped g-C₃N₄, C-doped g-C₃N₄@3.2wt%PANI, C-doped g-C₃N₄@6.3wt%PANI, and C-doped g-C₃N₄@9.1wt%PANI composites respectively. For undoped g-C₃N₄ shown in Figure 2 (a), the spectrum exhibits characteristic absorption bands confirming successful graphitic carbon nitride formation. A broad band in the 3000-3500 cm⁻¹ region corresponds to N-H stretching vibrations and O-H groups from adsorbed water or surface hydroxyls. Sharp intense peaks in the 1200-1650 cm⁻¹ region represent C-N and C=N stretching vibrations of the heterocyclic rings forming the tri-s-triazine framework, with specific peaks at approximately 1630 cm⁻¹ (C=N stretching), 1400 cm⁻¹ and 1320 cm⁻¹ (C-N stretching of tertiary amines and bridging C-NH-C units). The intense peak at approximately 810 cm⁻¹ represents the out-of-plane bending vibrations and characteristic breathing mode of the tri-s-triazine ring system, serving as a fingerprint for the g-C₃N₄ structure (Ranganathan & Selwynraj Arunodayaraj, 2024; Bai et al., 2025).

For the composite spectra in Figure 2 (b-d), characteristic absorption bands of both g-C₃N₄ and PANI are observed, confirming successful composite formation. The g-C₃N₄ peaks, including the 810 cm⁻¹ breathing mode and C-N/C=N stretching vibrations, are preserved, indicating that the basic chemical structure remains intact after PANI incorporation. However, these g-C₃N₄ peak intensities show progressive reduction with increasing PANI content from 3.2 to 9.1 wt%, attributed to the dilution effect from increasing PANI concentration and surface shielding of g-C₃N₄ by PANI coating (Jing et al., 2019). The composites reveal characteristic PANI absorption bands confirming successful incorporation. C=C stretching vibrations of quinoid and benzenoid rings appear at approximately 1580 cm⁻¹ and 1480 cm⁻¹ respectively, confirming the maintained conjugated polymer structure of PANI. The presence of both ring vibrations is important as their ratio determines the doping state and conductivity of PANI. Bands at 1200-1300 cm⁻¹ correspond to C-N stretching of secondary amines, while

1100-1150 cm⁻¹ peaks are attributed to C-H stretching in aromatic rings. The intensity of PANI characteristic bands increases systematically with PANI loading, demonstrating progressive surface coverage. Distinct C-H out-of-plane bending vibrations in the 700-900 cm⁻¹ region confirm successful polymerization of aniline into PANI structure (Wang et al., 2024).

Significant peak shifts in the composite spectra indicate strong chemical interactions and hydrogen bonding between the components. The N-H stretching region (3000-3500 cm⁻¹) shows broadening and shifts upon PANI incorporation, suggesting hydrogen bond formation between amine groups of g-C₃N₄ and imine/amine groups of PANI. Shifts in C-N and C=N stretching vibrations confirm electronic interactions where electron-rich nitrogen sites in g-C₃N₄ interact with the conjugated PANI backbone, potentially enhancing its conductivity. The broadening of absorption bands, particularly in the 1200-1650 cm⁻¹ region, indicates increased structural disorder and more amorphous composite structure, consistent with XRD findings and beneficial for electrochemical applications by providing accessible active sites (Yin et al., 2020). The FTIR analysis also provides evidence for successful carbon doping through systematic shifts in C-N and C=N stretching frequencies, indicating modifications in the electronic environment of nitrogen atoms in the g-C₃N₄ framework. The absence of carbonyl (C=O) or carboxylic acid (COOH) bands confirms high purity and complete removal of precursors, with no detectable impurities or unreacted starting materials.

The FTIR results have significant implications for electrochemical performance. The tri-s-triazine units in g-C₃N₄ provide nitrogen-rich active sites for pseudocapacitive charge storage through reversible redox reactions, while PANI contributes additional pseudocapacitance through redox transitions between leucoemeraldine, emeraldine, and pernigraniline states involving reversible proton and electron exchange. The intimate hydrogen bonding and electronic interactions between components facilitate efficient charge transfer at the interface, reducing charge transfer resistance and improving overall electrochemical performance. The preservation of both structures ensures that beneficial properties of each component are maintained while synergistic effects enhance overall activity.

Morphology and Elemental Analysis

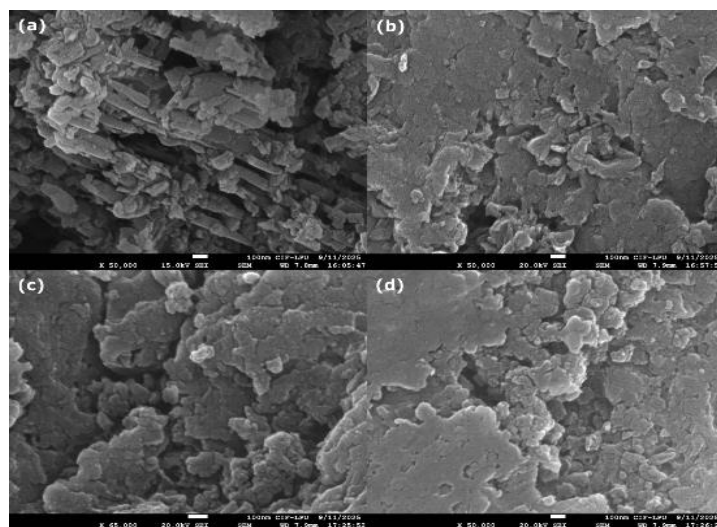


Figure 3: FESEM micrographs of (a) undoped g-C₃N₄, (b) C-doped g-C₃N₄@3.2wt%PANI, (c) C-doped g-C₃N₄@6.3wt%PANI, and (d) C-doped g-C₃N₄@9.1wt%PANI composites

The surface morphology of undoped g-C₃N₄ and C-doped g-C₃N₄@PANI composites with varying PANI loadings (3.2, 6.3, and 9.1 wt%) was examined using FESEM. The analysis provides critical visual evidence of morphological evolution upon PANI incorporation and its implications for electrochemical performance. Figure 3 (a), (b), (c) and (d) presents the FESEM micrographs of undoped g-C₃N₄, C-doped g-C₃N₄@3.2wt%PANI, C-doped g-C₃N₄@6.3wt%PANI, and C-doped g-C₃N₄@9.1wt%PANI composites respectively. The undoped g-C₃N₄ shown in Figure 3 (a) exhibits irregular, flake-like sheet structures with stacked layered morphology characteristic of graphitic carbon nitride synthesized via thermal polymerization (Jiang et al., 2025). The material displays a rough, porous surface with visible agglomerates, consistent with the turbostratic structure identified in XRD analysis. The sheet-like morphology with folded edges and thin nanosheets provides high surface area and abundant active sites for charge storage, while the porous structure facilitates electrolyte penetration and ion diffusion, contributing to enhanced electrochemical accessibility. Upon PANI incorporation, significant morphological changes are observed in the composite micrographs (Figure 3 (b-d)). At 3.2 wt% PANI loading (Figure 3 (b)), the composite exhibits flake-like g-C₃N₄ sheets coated with a thin, uniform PANI layer resulting in slightly smoother surfaces. The sheet structure remains discernible but with reduced edge sharpness, indicating partial surface coverage while preserving the overall g-C₃N₄ morphology (Saminathan et al., 2025; Zghab et al., 2025). The uniform coating demonstrates good dispersion and intimate contact achieved through ultrasonic-assisted wet impregnation.

At 6.3 wt% PANI loading (Figure 3 (c)), the morphology shows more pronounced surface coverage with a thicker, continuous PANI coating. The g-C₃N₄ sheets appear more rounded and embedded within the PANI matrix, which fills voids and interstitial spaces between sheets. This results in a more compact, densely packed structure with reduced porosity and increasingly smooth, uniform surfaces. This morphology creates a continuous conductive network through the PANI phase, facilitating efficient charge transport while

maintaining access to active sites on both components. At 9.1 wt% PANI loading (Figure 3 (d)), the highest PANI content sample exhibits the most significant morphological changes, with g-C₃N₄ sheets almost completely obscured by a thick PANI layer. The composite appears as agglomerated particles with predominantly smooth, polymer-like surface texture. The PANI forms a dense, continuous matrix with embedded g-C₃N₄ sheets, though some PANI aggregation is observed at this high loading due to excessive polymer content. While this ensures excellent electrical connectivity, the thick PANI layer may partially block access to g-C₃N₄ active sites, suggesting the optimal PANI loading for maximizing performance may lie below 9.1 wt% (Fan et al., 2015; Prabakaran et al., 2024). The progressive morphological evolution from exposed g-C₃N₄ sheets at low PANI content to PANI-coated and PANI-encapsulated structures at high content confirms successful composite formation with tunable surface characteristics. The transition demonstrates intimate component interaction consistent with FTIR findings and allows systematic optimization of electrochemical properties by balancing the contributions of both components. The morphological features have direct implications for electrochemical performance. The sheet-like g-C₃N₄ morphology provides a high surface area substrate, while porous structures facilitate electrolyte infiltration. Uniform PANI coating at moderate loadings creates continuous conductive networks enhancing charge transfer and reducing resistance, while maintaining access to active sites. Intimate interfacial contact promotes synergistic effects where pseudocapacitive properties of both materials are optimally utilized.

Electrochemical Analysis

The electrochemical performance of undoped g-C₃N₄ and C-doped g-C₃N₄@PANI composites with varying PANI loadings (3.2, 6.3, and 9.1 wt%) was evaluated using Cyclic Voltammetry (CV), Galvanostatic Charge-Discharge (GCD), and Electrochemical Impedance Spectroscopy (EIS) in a three-electrode configuration, providing comprehensive insights into charge storage mechanisms, specific capacitance, and ion transport kinetics.

Cyclic Voltammetry Analysis

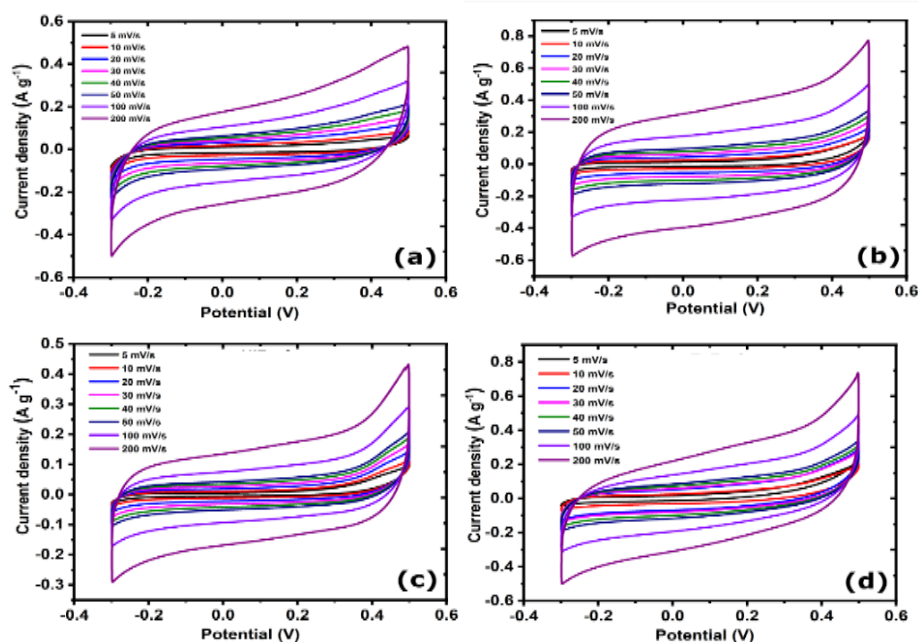


Figure 4: CV plot of (a) undoped g-C₃N₄, (b) C-doped g-C₃N₄@3.2wt%PANI, (c) C-doped g-C₃N₄@6.3wt%PANI, and (d) C-doped g-C₃N₄@9.1wt%PANI composites

Table 2: Electrode Parameters from CV Curve

Sample	Approx. CV Area (A·V/g)	Scan Rate (V/s)	ΔV (V)	C _s (F/g)
Undoped g-C ₃ N ₄	0.10	0.005	0.8	25
C-doped g-C ₃ N ₄ @3.2 wt% PANI	0.18	0.005	0.8	45
C-doped g-C ₃ N ₄ @6.3 wt% PANI	0.09	0.005	0.8	22
C-doped g-C ₃ N ₄ @9.1 wt% PANI	0.17	0.005	0.8	42

Figure 4 (a), (b), (c) and (d) presents the CV plot of undoped g-C₃N₄, C-doped g-C₃N₄@3.2wt%PANI, C-doped g-C₃N₄@6.3wt%PANI, and C-doped g-C₃N₄@9.1wt%PANI composites respectively. The CV plots in Figure 4 (a-d) reveal distinct electrochemical behavior across the samples. The capacitance is calculated from the integrated area under the curve using the relation

$$C = \frac{1}{2v\Delta V \cdot m} \int IdV \quad (1)$$

where v is the scan rate, ΔV is the potential window, m is the mass loading, I is the current, V is the potential and $\int IdV$ is the integrated area. Undoped g-C₃N₄ (Figure 4 (a)) displays a quasi-rectangular curve with broad redox peaks, indicating a combination of electric double-layer capacitance from its layered structure and pseudocapacitance from nitrogen-containing functional groups, though the small integrated area suggests moderate specific capacitance due to limited active sites and modest conductivity (Zhang et al., 2021). Upon PANI incorporation, the composite electrodes in Figure 4 (b-d) exhibit significantly enhanced CV responses with increased current densities and larger integrated areas. The maintained quasi-rectangular shapes with well-defined redox

peaks (0.2-0.6 V) confirm the combination of EDLC and pseudocapacitive mechanisms, with the redox peaks corresponding to PANI's Faradaic transitions between leucoemeraldine, emeraldine, and pernigraniline states. Current density increases progressively with PANI loading from 3.2 to 9.1 wt%, attributed to more redox-active sites and improved conductivity from the PANI network. The sustained quasi-rectangular shape even at 9.1 wt% indicates excellent rate capability and reversibility (Thosare et al., 2025; Okafor et al., 2025). The specific capacitance values obtained from CV curves are summarized in Table 2. The undoped g-C₃N₄ electrode exhibited a specific capacitance of 25 F/g. Upon PANI incorporation, the capacitance increased to 45 F/g at 3.2 wt% PANI, attributed to the synergistic effect between carbon-doped g-C₃N₄ and the pseudocapacitive PANI. However, at 6.3 wt% PANI, the capacitance dropped to 22 F/g, likely due to PANI agglomeration blocking active sites. At 9.1 wt% PANI, the capacitance recovered to 42 F/g, suggesting the formation of a percolated conductive network. Overall, 3.2 wt% PANI yielded the optimal electrochemical performance (Aswathi & Sreeja, 2025).

Galvanostatic Charge-Discharge Analysis

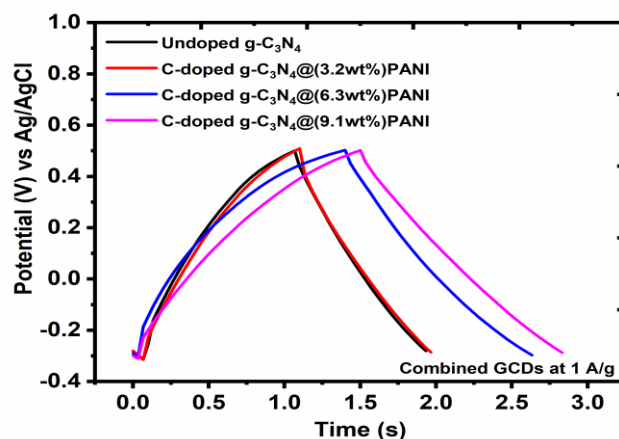


Figure 5: GCD plot of (a) undoped g-C₃N₄, (b) C-doped g-C₃N₄@3.2wt%PANI, (c) C-doped g-C₃N₄@6.3wt%PANI, and (d) C-doped g-C₃N₄@9.1wt%PANI composites

Table 3: Electrode Parameters from GCD Curve

Sample	Peak Potential (V)	End Potential (V)	C _s (F/g)
Undoped g-C ₃ N ₄	0.5	0.3	23
C-doped g-C ₃ N ₄ @3.2 wt% PANI	0.5	0.3	42
C-doped g-C ₃ N ₄ @6.3 wt% PANI	0.5	0.3	20
C-doped g-C ₃ N ₄ @9.1 wt% PANI	0.5	0.3	39

Figure 5 (a), (b), (c) and (d) presents the GCD plot of undoped g-C₃N₄, C-doped g-C₃N₄@3.2wt%PANI, C-doped g-C₃N₄@6.3wt%PANI, and C-doped g-C₃N₄@9.1wt%PANI composites respectively. The GCD plots in Figure 5 (a-d) provide quantitative capacitance values. Undoped g-C₃N₄ (Figure 5 (a)) shows non-linear curves with short discharge times, indicating moderate specific capacitance. The capacitance from the GCD curve is calculated using equation (2)

$$C = \frac{I \Delta t}{\Delta V} \quad (2)$$

where I is the discharge current, Δt is the discharge time, and ΔV is the potential window. The composite electrodes exhibit significantly prolonged discharge times with increasing PANI content. The GCD curves maintain non-linear features with distinct plateau regions confirming pseudocapacitive contributions from PANI. Specific capacitance increases systematically with PANI loading, with the 9.1 wt% sample exhibiting the highest value. The specific capacitance values obtained from GCD curves are

summarized in Table 3. The undoped g-C₃N₄ electrode delivered a specific capacitance of 23 F/g. Upon PANI incorporation, the capacitance increased to 42 F/g at 3.2 wt% PANI, consistent with the CV results and confirming the synergistic effect between carbon-doped g-C₃N₄ and the pseudocapacitive PANI. However, at 6.3 wt% PANI, the capacitance dropped to 20 F/g, likely due to PANI agglomeration blocking active sites. At 9.1 wt% PANI, the capacitance recovered to 39 F/g, suggesting the formation of a percolated conductive network. Overall, 3.2 wt% PANI yielded the optimal electrochemical performance, and the GCD values are in good agreement with those obtained from CV. This enhancement results from the synergistic combination of g-C₃N₄'s EDLC, PANI's pseudocapacitance, improved conductivity from carbon doping, and the conductive PANI network facilitating charge transport. Good capacitance retention at higher current densities indicates favorable rate capability (Yan et al., 2026; Mahmood et al., 2025).

Electrochemical Impedance Spectroscopy Analysis

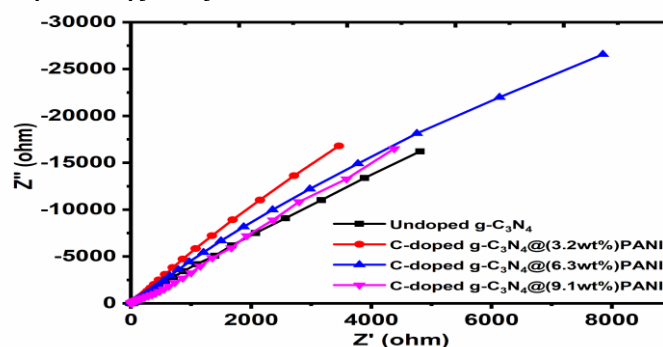


Figure 6: EIS plot of (a) undoped g-C₃N₄, (b) C-doped g-C₃N₄@3.2wt%PANI, (c) C-doped g-C₃N₄@6.3wt%PANI, and (d) C-doped g-C₃N₄@9.1wt%PANI composites.

Figure 6 (a), (b), (c) and (d) presents the EIS plot of undoped g-C₃N₄, C-doped g-C₃N₄@3.2wt%PANI, C-doped g-C₃N₄@6.3wt%PANI, and C-doped g-C₃N₄@9.1wt%PANI composites respectively. The EIS Nyquist plots reveal charge transfer resistance and ion diffusion characteristics. Undoped g-C₃N₄ (Figure 6 (a)) exhibits a large high-frequency semicircle indicating significant charge transfer resistance (R_{ct}), followed by a low-frequency Warburg impedance line corresponding to ion diffusion. The composite electrodes show progressively smaller semicircle diameters with increasing PANI content, indicating reduced charge transfer resistance. This reduction is attributed to the conductive PANI network facilitating electron transport and carbon doping enhancing g-C₃N₄'s conductivity. The 9.1 wt% sample exhibits the smallest semicircle, suggesting the lowest R_{ct} and fastest Faradaic reactions. The low-frequency region shows more vertical lines for composites, indicating improved capacitive behavior and faster ion diffusion, with the 6.3 wt% and 9.1 wt% samples showing the most ideal capacitive behavior (Lan et al., 2026; Ghaharpour et al., 2026).

Performance Correlation and Optimization

The electrochemical results demonstrate a clear trend of enhanced performance with PANI incorporation. The 9.1 wt% sample shows the highest specific capacitance and most favorable charge transfer characteristics. This enhancement results from multiple synergistic effects: PANI provides additional pseudocapacitance through redox-active sites; carbon doping improves g-C₃N₄ conductivity; the conductive PANI network facilitates charge transport; and intimate interfacial contact promotes efficient charge transfer.

The electrochemical performance correlates well with structural and morphological characterization. The progressive capacitance increases with PANI loading aligns with FTIR-confirmed PANI content and FESEM-observed surface coverage. Reduced charge transfer resistance corresponds to the morphological transition from exposed g-C₃N₄ sheets to PANI-coated structures enhancing electrical connectivity. Improved ion diffusion kinetics relates to the porous structure and intimate component interaction. While the 9.1 wt% sample exhibits the highest capacitance, the 6.3 wt% loading may represent the optimal balance for practical applications, providing significant enhancement while maintaining structural integrity and active site accessibility, as excessive PANI at 9.1 wt% may lead to aggregation and potential cycling instability.

CONCLUSION

The comprehensive characterization of undoped g-C₃N₄ and C-doped g-C₃N₄@PANI composites with varying PANI loadings (3.2, 6.3, and 9.1 wt%) through XRD, FTIR, FESEM, and electrochemical analysis has successfully established the structural, chemical, morphological, and electrochemical properties of these materials, demonstrating their significant potential as high-performance supercapacitor electrodes. XRD analysis confirmed the successful synthesis of pure g-C₃N₄ with characteristic (002) and (100) peaks, while the preserved peaks with systematic shifts, broadening, and reduced intensity upon PANI incorporation confirmed carbon doping and composite formation. The progressive decrease in crystallite size and modified interlayer spacing with increasing PANI content indicated enhanced disorder beneficial for electrochemical applications by providing more active sites and shorter ion diffusion pathways. FTIR analysis verified the chemical integrity of both components, with

characteristic g-C₃N₄ bands (N-H at 3000-3500 cm⁻¹, C-N/C=N at 1200-1650 cm⁻¹, tri-s-triazine at 810 cm⁻¹) and PANI bands (quinoid/benzenoid at 1580/1480 cm⁻¹, C-N at 1200-1300 cm⁻¹) present in composites. Systematic intensity changes and peak shifts confirmed increasing PANI loading and strong hydrogen bonding interactions essential for efficient charge transfer.

FESEM morphological analysis revealed progressive evolution from exposed flake-like g-C₃N₄ sheets at low PANI content to uniform thin coating at 3.2 wt%, thicker continuous coating at 6.3 wt%, and complete encapsulation with some aggregation at 9.1 wt%, confirming successful composite formation with tunable surface characteristics. Electrochemical analysis through CV, GCD, and EIS demonstrated significantly enhanced performance with PANI incorporation, showing increased current densities, prolonged discharge times, systematically increasing specific capacitance with PANI loading, and progressively reduced charge transfer resistance with improved ion diffusion kinetics.

The synergistic combination of structural modifications, chemical interactions, morphological integration, and electrochemical enhancements establishes these composites as promising electrode materials. The C-doped g-C₃N₄@3.2 wt% PANI composite exhibited the highest specific capacitance of 45 F/g from CV and 42 F/g from GCD, substantially outperforming undoped g-C₃N₄ (25 F/g and 23 F/g, respectively), confirming the synergistic enhancement from PANI incorporation. The excellent electrochemical properties, including high specific capacitance, low charge transfer resistance, and good rate capability, position these composites as competitive candidates for next-generation energy storage devices. The successful demonstration of structure-property-performance relationships provides a solid foundation for further optimization and development of g-C₃N₄-based composite materials for sustainable energy applications.

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