

Article

Unraveling the Effect of Synthesis Temperature and Metal Doping on the Structural, Optical, and Photocatalytic Properties of g-C₃N₄ for Enhanced *E. coli* Photodisinfection and Self-Cleaning Surface Applications

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Abstract

The development of efficient photocatalytic materials for waterborne pathogen inactivation and self-cleaning surfaces in biomedical applications remains a critical challenge due to the rising prevalence of antimicrobial-resistant bacteria. This study systematically investigates the structural, optical, and photocatalytic disinfection properties of graphitic carbon nitride (g-C₃N₄) synthesized at variable temperatures (450–600 °C) and doped with transition metals (Mn, Co, Cu). Through FTIR and UV/Vis spectroscopy, we demonstrate that synthesis temperatures between 450 and 550 °C yield a well-ordered polymeric network with enhanced π -conjugation and charge separation, while 600 °C induces structural degradation. Metal doping with Mn and Co significantly enhances photocatalytic disinfection, achieving complete *E. coli* inactivation (6-log reduction) within 6 h via optimized reactive oxygen species (ROS) generation. The best material (g-C₃N₄ synthesized at 500 °C and doped with Mn) was integrated into sodium alginate hydrogel surfaces, demonstrating reusable self-cleaning functionality with sustained bactericidal activity (5.9-log CFU/mL reduction after five cycles). This work provides a roadmap for tailoring metal-doped g-C₃N₄ composites for practical antimicrobial applications, emphasizing the interplay between synthesis parameters, ROS dynamics, and real-world performance.

Keywords: photocatalytic disinfection; graphitic carbon nitride; synthesis temperature optimization; transition metal doping; reactive oxygen species (ROS); bacterial inactivation; hydrogel-based photocatalytic coatings



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1. Introduction

Waterborne pathogenic bacteria represent a major public health concern due to their ability to spread antimicrobial resistance and persist in aquatic environments. Many of these microorganisms can form biofilms on surfaces, making their eradication particularly challenging [1]. These issues have spurred growing interest in advanced disinfection strategies, particularly those based on visible-light-activated photocatalytic materials [2].

In this sense, heterogeneous photocatalysis, a subset of advanced oxidation processes (AOPs), uses photocatalytic materials to generate reactive oxygen species (ROS) like hydroxyl radicals ($\text{HO}\bullet$), superoxide anions ($\text{O}_2\bullet^-$), and singlet oxygen ($^1\text{O}_2$), which degrade organic pollutants and inactivate microorganisms [3,4]. While TiO_2 is the most studied photocatalytic material, its limited visible-light absorption ($\lambda < 387 \text{ nm}$) and rapid charge recombination ($\sim 100 \text{ ps}$) limit its application [5]. Thus, the development of new semiconductors is required.

On the other hand, graphitic carbon nitride ($\text{g-C}_3\text{N}_4$), a metal-free semiconductor, offers advantages such as visible-light responsiveness (bandgap $\sim 2.8 \text{ eV}$, which correlates with a wavelength of 442 nm), thermal stability, and low toxicity [6,7]. However, its efficiency is constrained by its low surface area and sluggish charge transfer [6]. Recent advances highlight that optimizing synthesis parameters—particularly the polycondensation temperature of urea precursors—can tailor its structural and electronic properties [8].

Although several studies have examined the effect of synthesis temperature on the structural and optical properties of carbon nitride-based materials, most reports focus on narrow temperature intervals or on materials tailored for general photocatalytic applications, such as dye degradation or hydrogen evolution [8]. However, to date, there is no comprehensive study that systematically explores a wide synthesis temperature range and its impact on the photocatalytic disinfection performance of $\text{g-C}_3\text{N}_4$, particularly regarding the generation pathways of reactive oxygen species (ROS) and their correlation with bacterial inactivation efficiency. Furthermore, few studies translate powdered catalysts into functional self-cleaning surfaces, limiting real-world applicability.

In addition, critical gaps persist in understanding how synthesis temperature synergizes with metal doping to govern ROS dynamics and bactericidal efficacy. In fact, while transition metal doping (e.g., Mn, Co, Cu) is known to alter the electronic structure of $\text{g-C}_3\text{N}_4$ by introducing mid-gap states, prior works have predominantly studied doping effects independently of synthesis temperature [9]. For instance, Fe_3O_4 incorporation has been reported to enhance $\text{O}_2\bullet^-$ and $\text{HO}\bullet$ [10]. Yet, the interplay between optimized synthesis temperatures and metal doping, which are critical for tailoring ROS selectivity and bactericidal performance, has not been rigorously addressed.

Taking *E. coli* as an indicator of bacterial inactivation, this study bridges the above-mentioned gaps by systematically investigating how synthesis temperature ($450\text{--}600^\circ\text{C}$) governs the structural, optical, and electrochemical properties of $\text{g-C}_3\text{N}_4$, as well as how subsequent transition metal doping (Mn, Co, Cu) modulates ROS generation and disinfection kinetics. By integrating temperature-dependent structural analysis with metal doping strategies, we identify synergistic conditions for maximizing photocatalytic disinfection efficiency and translate these insights into functional self-cleaning surfaces.

2. Results and Discussion

2.1. Characterization of $\text{g-C}_3\text{N}_4$ Synthesized at Different Temperatures

2.1.1. Spectroscopic Characterization of Bare $\text{g-C}_3\text{N}_4$

The FTIR spectra shown in Figure 1 illustrate the structural evolution of $\text{g-C}_3\text{N}_4$ synthesized at different temperatures, with the spectrum of urea included as a reference. The initial spectrum of urea exhibits several sharp and intense bands characteristic of its molecular structure, particularly vibrations related to N–H stretching and C=O functional groups. Upon thermal treatment, these distinctive urea signals are modified, confirming the decomposition of the precursor and the subsequent formation of polymeric structures consistent with graphitic carbon nitride [11].

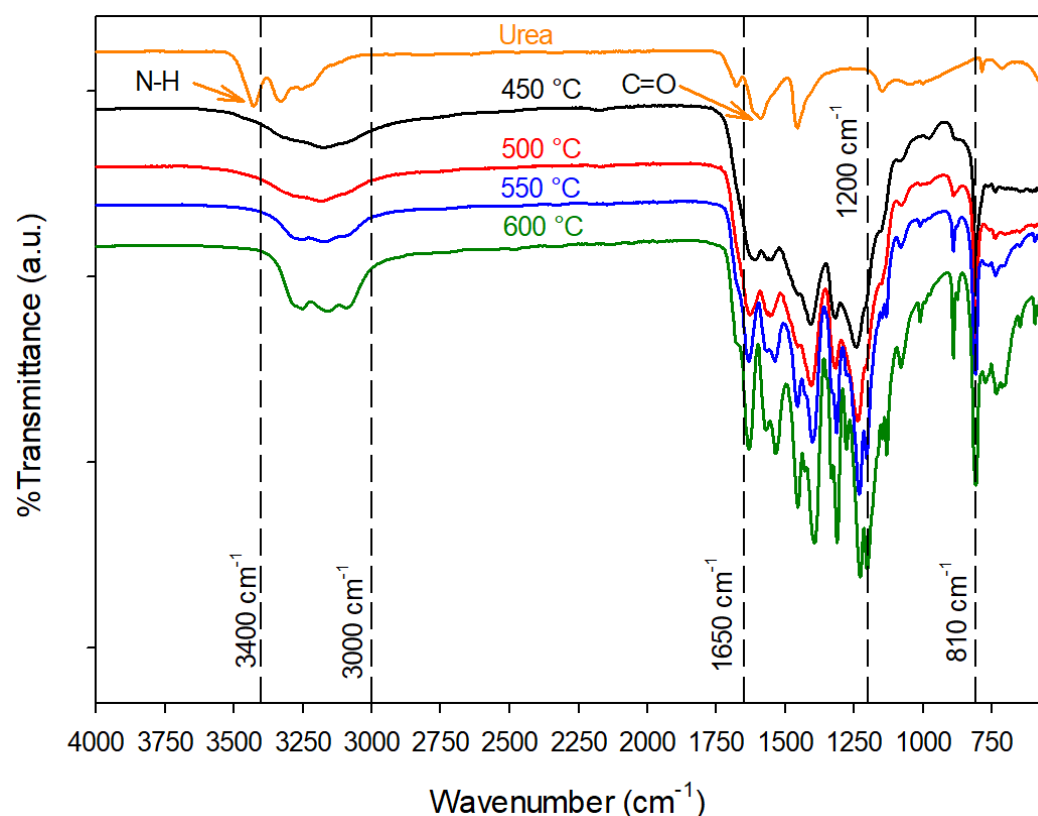


Figure 1. FTIR spectra for g-C₃N₄ synthesized at different temperatures.

In the spectra of the thermally treated samples, a prominent and sharp absorption band appears around 810 cm^{−1} across all cases, corresponding to the out-of-plane bending vibrations of the s-triazine rings. This feature is a well-known fingerprint of the g-C₃N₄ structure and indicates the successful formation of the heptazine-based polymeric network. Additionally, in the range of 1200–1650 cm^{−1}, multiple overlapping bands are observed, which are associated with the stretching vibrations of C–N and C=N bonds in the heterocyclic framework. These bands are clearly defined in samples synthesized between 450 and 600 °C, demonstrating the development and stability of the g-C₃N₄ structure within this temperature range [11].

The region between 3000 and 3400 cm^{−1} displays a broad absorption band in all samples, which can be attributed to the N–H stretching vibrations of the terminal amine groups and O–H stretching from adsorbed water or hydroxyl functionalities. As the synthesis temperature increases, the intensity of this broad band gradually increases, indicating progressive condensation of surface amine groups and a more hydrophilic surface [11]. This trend reflects the thermal evolution of the synthesized g-C₃N₄, which is consistent with the material undergoing further deamination and densification at higher temperatures.

A shoulder around 1170 cm^{−1} becomes more apparent in the spectra of samples treated at intermediate temperatures, particularly at 500 and 550 °C. This feature is associated with the presence of C–OH groups located at the edges of the g-C₃N₄ sheets [11]. Its relative prominence suggests that moderate temperatures favor the retention of certain edge functionalities that may be beneficial for further surface modifications or interactions.

The UV/Vis spectra presented in Figure 2 correspond to g-C₃N₄ samples synthesized at different temperatures, ranging from 450 °C to 600 °C. All spectra exhibit two main absorption features, characteristic of polymeric carbon nitride. The first is an intense absorption band below 260 nm, which is attributed to $\pi \rightarrow \pi^*$ electronic transitions within the aromatic s-triazine units. This band is prominent in all samples and confirms the

presence of the conjugated aromatic system typical of g-C₃N₄ [12]. The second feature is a broader absorption band extending from approximately 270 to 400 nm, which corresponds to $n \rightarrow \pi^*$ transitions involving lone pairs on nitrogen atoms in C–N bonds. This band reflects the presence of nonbonding electronic states and is sensitive to the local electronic environment, particularly to changes in structure and defects.

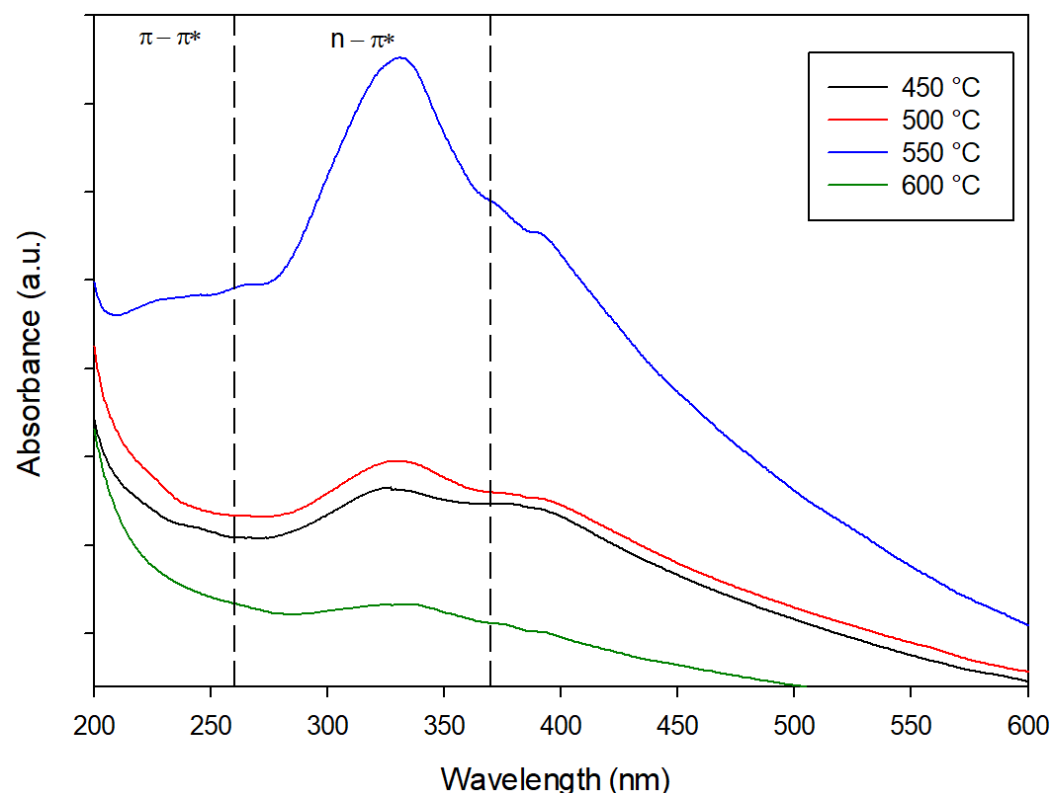


Figure 2. UV/Vis spectra for g-C₃N₄ synthesized at different temperatures.

The Tauc plots corresponding to g-C₃N₄ samples synthesized at different temperatures (450–600 °C) are shown in Figure 3. For the sample synthesized at 450 °C, the extrapolated bandgap is approximately 2.75 eV. This value is characteristic of a partially polymerized g-C₃N₄ structure, suggesting that the condensation process is still incomplete, and the π -conjugated system is not yet fully developed. The corresponding UV/Vis spectrum supports this interpretation, showing moderate absorbance in the UV region and limited extension into the visible range. Table 1 summarizes the experimental bandgaps and the correlated maximum wavelengths associated with the electron migration from the valence band to the conduction band. Moreover, it shows the electric resistance for the diffusion of the electrons through the semiconductor domains and the flat potential band of the conduction band measured by impedance electrochemical spectroscopy and the corresponding flat potential of the valence band.

At 500 °C, the bandgap decreases to 2.66 eV, indicating a more advanced degree of condensation and a better-organized structure. This correlates with the increase in the $n \rightarrow \pi^*$ transition band observed in the UV/Vis spectrum, pointing to an improved electronic configuration.

The sample obtained at 550 °C presents a higher bandgap compared to the preceding materials, 2.89 eV, and the most intense optical absorption among all samples. Although the bandgap increases slightly, this is likely due to a reduction in defect states and trap levels, which narrows the density of electronic states near the conduction and valence bands [14].

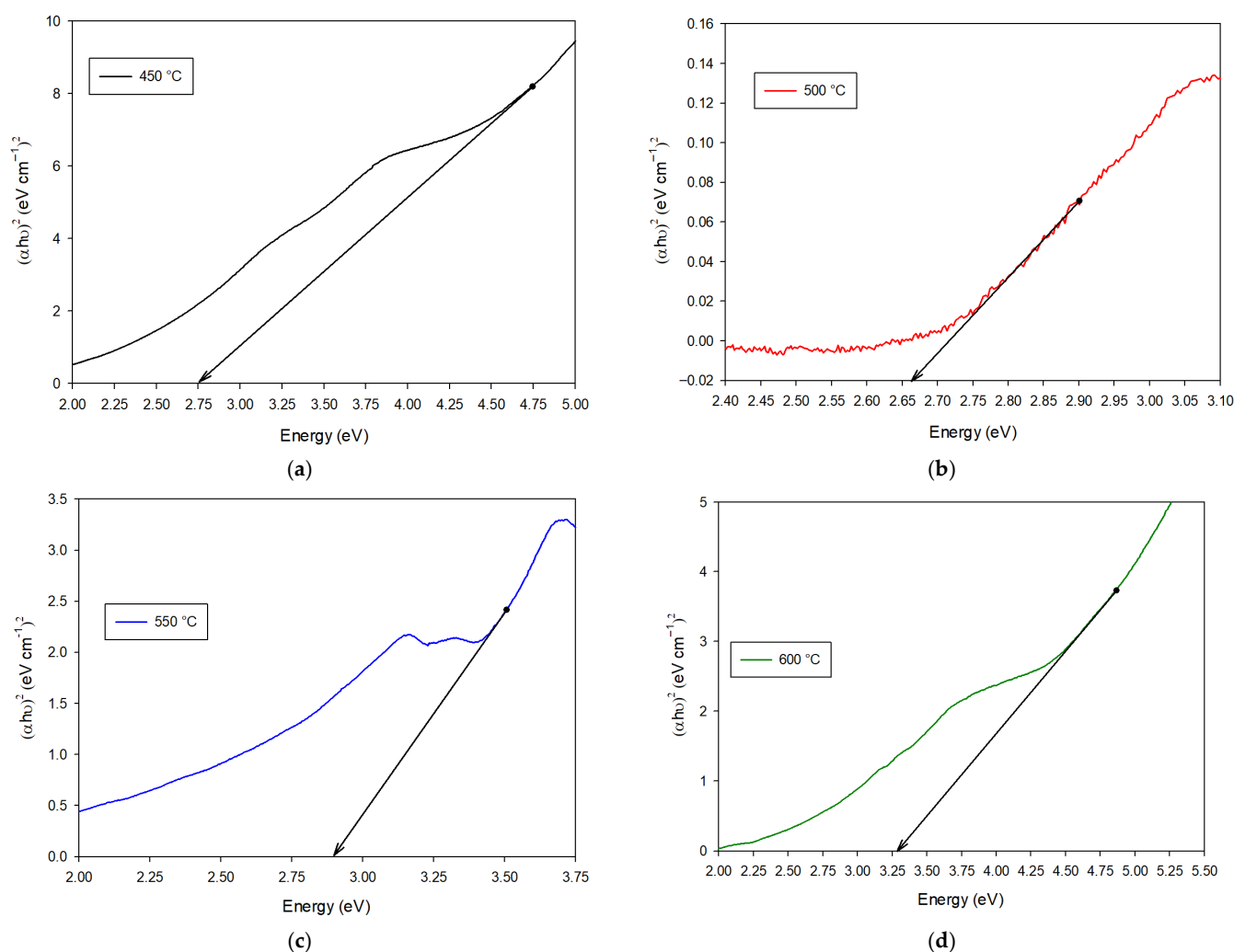


Figure 3. Tauc plots and bandgap energies of g-C₃N₄ synthesized at (a) 450 °C, (b) 500 °C, (c) 550 °C and (d) 600 °C.

Table 1. Summary of the characterization of the materials synthesized. E_{BG}: bandgap energy calculated from Tauc plots. λ_{Exc,max}: maximum excitation wavelength obtained from Tauc plot [13]. Electric resistance and E_{CB} (conduction band) and E_{VB} (valence band) potential were obtained from the electrochemical characterization.

Material	E _{BG} (eV)	λ _{Exc,max} (nm)	Electric Resistance (kΩ)	E _{CB} (V) vs. NHE	E _{VB} (V) vs. NHE
450 °C	2.75	451	1.9	−0.471	2.279
500 °C	2.66	466	1.1	−0.377	2.283
550 °C	2.89	429	2.3	−0.305	2.585
600 °C	3.30	375	1.1	−0.355	2.945

For the 600 °C sample, the bandgap increases to 3.30 eV, yet the UV/Vis spectrum reveals a significant drop in absorbance intensity, implying the beginning of structural degradation. The loss of chromophoric density and extended conjugation could adversely affect the optical performance of the material.

2.1.2. Electrochemical Characterization of Bare g-C₃N₄

The electrochemical impedance spectroscopy (EIS) results presented in Figure 4a show the Nyquist plots for g-C₃N₄ materials synthesized at different temperatures (450–600 °C),

while Figure 4b depicts the equivalent electrical circuit model used to fit the experimental data. The Nyquist plots represent the imaginary impedance component ($-Z''$) as a function of the real component (Z') and provide valuable insight into the charge transport, interface properties, and resistive/capacitive behavior of the materials.

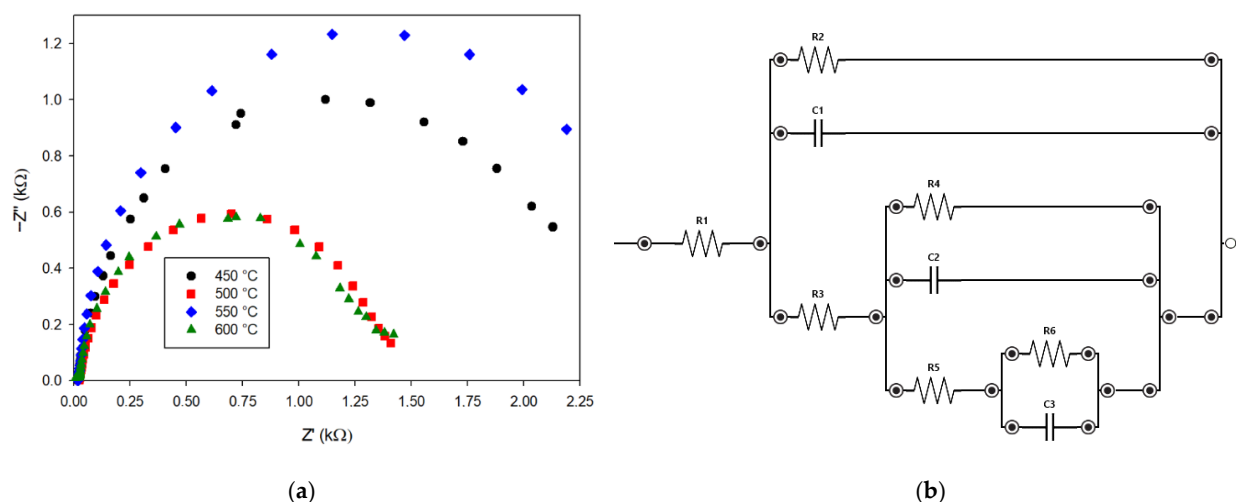


Figure 4. (a) Nyquist plot obtained at -0.3 V vs. Ag/AgCl electrode of all bare $g\text{-C}_3\text{N}_4$ materials. (b) Equivalent electric circuit fitted for the Nyquist plots.

All spectra exhibit semicircular shapes, characteristic of mixed charge-transfer and diffusion-controlled processes in porous or semiconducting materials. The diameter of the semicircle is typically associated with the charge-transfer resistance, while deviations from a perfect semicircle suggest complex interfacial phenomena, such as distributed capacitance or multiple relaxation processes [15].

Among the samples, the one synthesized at 550 °C (blue diamonds) shows the largest semicircle diameter, corresponding to the highest charge-transfer resistance. This behavior is consistent with the highly ordered structure and lower defect density observed for this sample in the UV/Vis analysis in Figures 1 and 2.

In contrast, the samples synthesized at 500 °C (red squares) and 600 °C (green triangles) show smaller semicircles, indicating lower charge-transfer resistance and potentially enhanced ionic or electronic conductivity [15]. The 450 °C sample (black circles) also exhibits relatively high resistance, limiting effective charge transport.

The Mott–Schottky (M–S) plots shown in Figure 5 correspond to $g\text{-C}_3\text{N}_4$ samples synthesized at different temperatures (450–600 °C) and were obtained with EIS. These plots display the inverse squared capacitance ($1/C^2$) as a function of the applied potential versus Ag/AgCl, and they allow the estimation of the flat-band potential of the conduction band (E_{CB}), as well as insights into the semiconductor type and charge-carrier behavior of the materials.

All curves exhibit a positive slope, characteristic of n-type semiconductors, which confirms that the majority carriers in the $g\text{-C}_3\text{N}_4$ samples are electrons. The flat-band potential (Table 1) can be extracted from the x-intercept of the linear region of the plots. As seen, it varies significantly with the synthesis temperature.

These values indicate a clear trend: the flat-band potential shifts toward more positive values as the synthesis temperature increases up to 550 °C, suggesting a progressive improvement in electronic structure and band alignment. The E_{CB} can be seen in the fifth column of Table 1. Beyond 550 °C, the flat-band potential shifts back toward more negative values, as seen for the 600 °C sample.

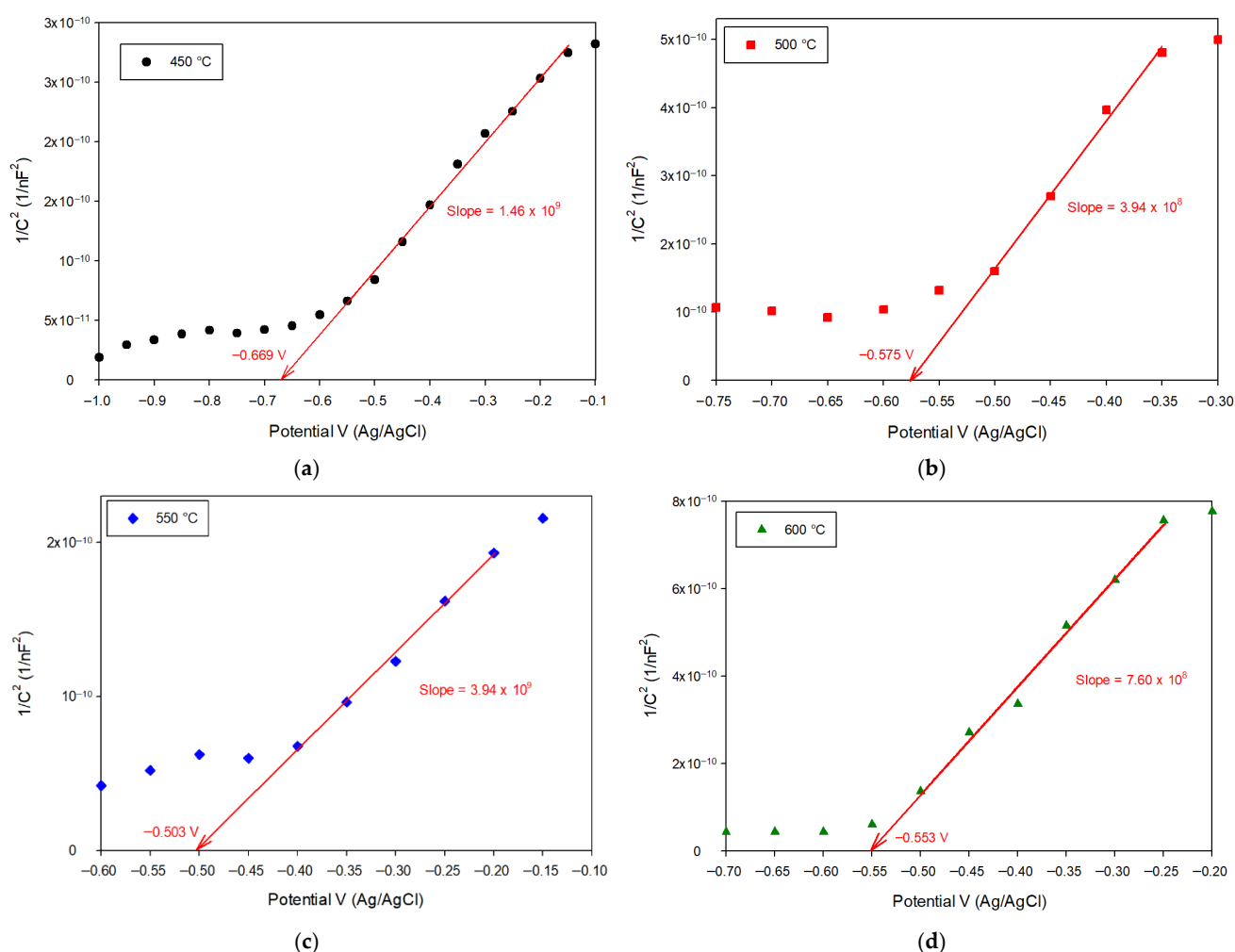


Figure 5. Mott–Schottky plots representing the Ag/AgCl of the $g-C_3N_4$ synthesized at (a) 450 °C, (b) 500 °C, (c) 550 °C and (d) 600 °C.

In addition to the shift in E_{CB} , the slope of the linear region also provides qualitative information about the carrier density. A smaller slope implies a higher charge-carrier density [15], and in this regard, the sample at 500 °C again appears optimal, with a moderately sloped linear region with respect to the other materials, indicating a balance between sufficient charge-carrier concentration and minimized recombination losses.

In summary, the combined structural, optical, electronic, and electrochemical analyses show that the properties of $g-C_3N_4$ are significantly influenced by the synthesis temperature.

Figure 6 presents the redox potentials (referenced to the Normal Hydrogen Electrode, NHE, at pH 7.0) of selected reactions associated with the generation of reactive oxygen species (ROS), superimposed on the estimated conduction and valence band edges of the synthesized $g-C_3N_4$ materials. The band positions were inferred from the Tauc and Mott–Schottky analyses discussed previously.

The conduction band edges for all samples lie at more negative potentials than the $O_2/O_2\bullet^-$ redox couple (-0.305 V vs. NHE), indicating that photoexcited electrons in each material can thermodynamically reduce molecular oxygen to generate superoxide radicals ($O_2\bullet^-$). However, differences are observed in the positions of the valence bands. Only the materials synthesized above 500 °C have a valence band edge more positive than the redox potential required for the oxidation of water to hydroxyl radicals ($HO\bullet$), which takes place at 2.310 V vs. NHE. The valence band positions of the other samples fall below this

threshold, meaning they cannot oxidize water to $\text{HO}\bullet$ via direct hole-driven oxidation under standard conditions.

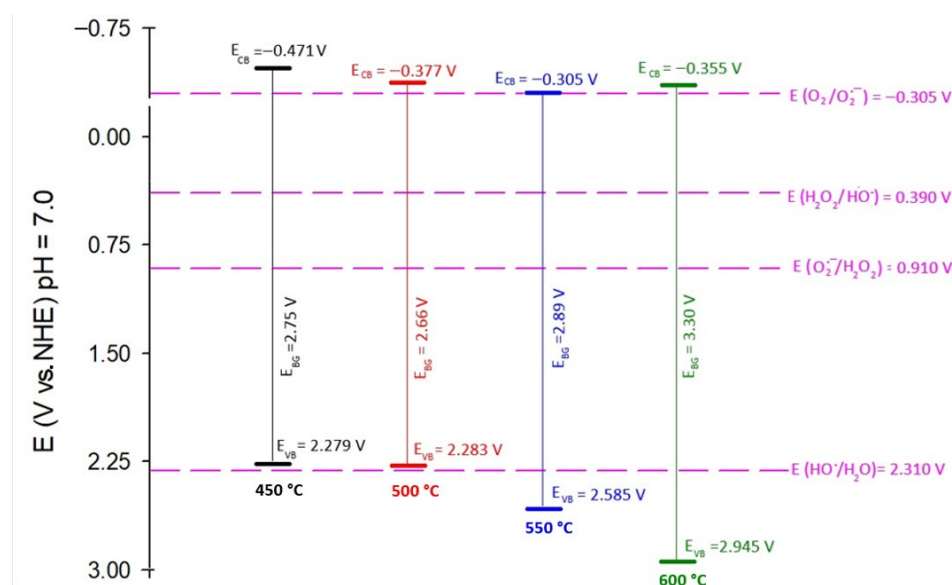


Figure 6. Redox potentials (E vs. NHE at pH 7.0) for selected reactions are graphically represented with the conduction and valence bands for all bare $\text{g-C}_3\text{N}_4$ materials.

As documented in the literature [16], the photocatalytic generation of reactive oxygen species (ROS) by $\text{g-C}_3\text{N}_4$ can also include the production of singlet oxygen ($^1\text{O}_2$). Mechanistic studies propose a sequential redox pathway involving (i) the reduction of molecular oxygen (O_2) by photogenerated electrons (e^-) to form superoxide anion radicals ($\text{O}_2\bullet^-$), followed by (ii) the oxidation of $\text{O}_2\bullet^-$ by valence band holes (h^+) to yield $^1\text{O}_2$ (Equations (1) and (2)):



However, the thermodynamic feasibility of this two-step mechanism remains ambiguous due to the absence of experimentally validated standard reduction potentials $E(\text{O}_2\bullet^- / ^1\text{O}_2)$. While the $E(\text{O}_2 / \text{O}_2\bullet^-)$ is well-established (-0.305 V vs. NHE at pH = 7.0), the potential required for $\text{O}_2\bullet^-$ oxidation to $^1\text{O}_2$ has not been systematically characterized.

Alternatively, $^1\text{O}_2$ generation may proceed via energy transfer from photoexcited $\text{g-C}_3\text{N}_4$ to ground-state triplet oxygen ($^3\text{O}_2$). This pathway involves intersystem crossing (ISC) to populate triplet-excited states of $\text{g-C}_3\text{N}_4$, which transfer energy to $^3\text{O}_2$ and subsequently promote the singlet-state ($^1\Delta_g$) formation.

The reactive oxidative species (ROS) predominant in a photocatalytic process depends on the relative contributions of these competing mechanisms. Further investigation should be carried out to identify the ROS formed under work conditions.

Based on these results, the samples synthesized at 450, 500, and 550 °C are considered promising, as they maintain structural integrity and present favorable optical and electronic properties. Meanwhile, the sample treated at 600 °C is classified as less promising due to signs of thermal degradation that negatively affect its physicochemical performance. Further photocatalytic testing will be necessary to confirm the practical implications of these findings.

2.2. Photocatalytic Evaluation of Bare $g\text{-C}_3\text{N}_4$ Synthesized at Different Temperatures

The disinfection profiles presented in Figure 7 show the photocatalytic inactivation of *E. coli* over time using $g\text{-C}_3\text{N}_4$ materials synthesized at different temperatures (450–600 °C). The results reveal a strong dependence of the antibacterial activity on the synthesis temperature, consistent with the structural, optical, electronic, and electrochemical characterizations previously discussed.

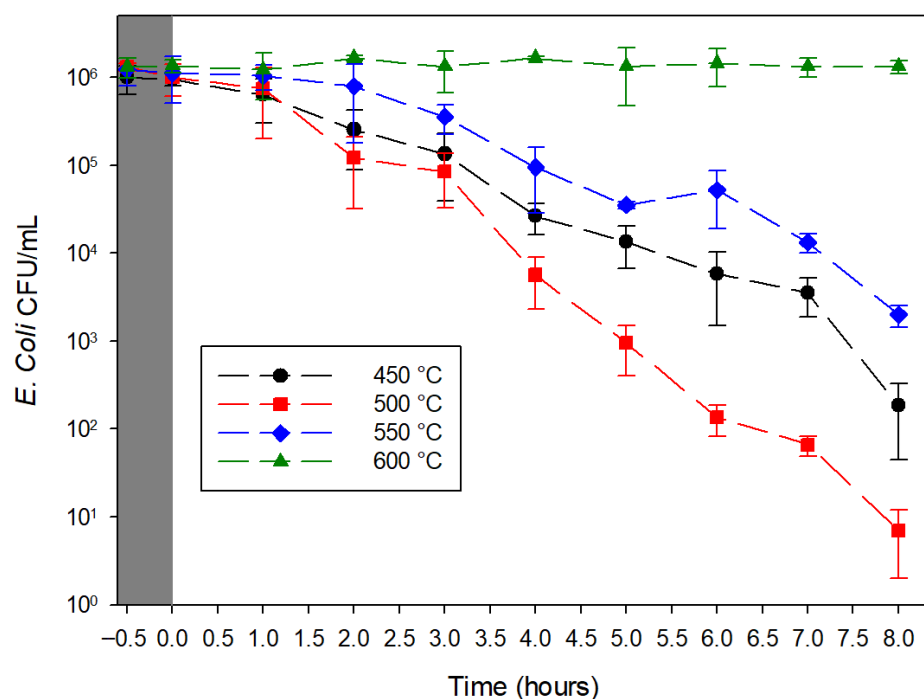


Figure 7. Kinetic profiles of *E. coli* disinfection promoted by $g\text{-C}_3\text{N}_4$ obtained at different temperatures under irradiation of indoor light. Conditions: 10 mL and 5 mg of the corresponding material. The error bars represent the standard deviation from duplicate treatments ($n = 2$). The dark region denotes the time in which the LEDs were turned off.

The material synthesized at 500 °C displays the fastest and most efficient bacterial inactivation, reducing the viable cell concentration by over 5 orders of magnitude within 8 h. This rapid disinfection correlates with the well-defined polymeric structure and properties of this sample, related to the strong light absorption and appropriate bandgap (UV/Vis and Tauc analyses), favorable flat-band potential (Mott–Schottky analysis), and relatively low charge-transfer resistance (EIS). These features likely contribute to more efficient generation and separation of photogenerated charge carriers, enhancing the production of reactive oxygen species (ROS), such as $\text{O}_2^{\bullet-}$ or $^1\text{O}_2$, which play a dominant role in microbial inactivation.

The 550 °C sample also shows photocatalytic disinfecting properties, though to a lower extent than those observed for the prepared samples at 450 and 500 °C. Despite its excellent optical absorption, its higher charge-transfer resistance (with EIS) may limit the efficiency of ROS generation and delay the inactivation kinetics. Meanwhile, the 450 °C sample shows intermediate activity, consistent with its capacity to generate $\text{O}_2^{\bullet-}$ (Figure 6) or $^1\text{O}_2$. However, its relatively lower light absorption, as shown in Figure 2, and incomplete polymerization may result in a decrease in photocatalytic activity.

In stark contrast, the sample synthesized at 600 °C exhibits negligible disinfection activity over the 8 h period. This might be due to the relatively high E_{BG} needed to activate the catalysts. In fact, wavelengths lower than 375 nm are required for this sample, while, as shown in Figure S1, the LEDs used have an emission spectrum of $\lambda \geq 425$ nm.

Considering all the physicochemical, optical, electronic, and photocatalytic results, the sample synthesized at 500 °C demonstrates the most balanced and effective performance. Therefore, this catalyst was selected to evaluate the influence of metal doping on photocatalytic activity.

2.3. Morphological, Crystallographic, and Spectroscopic Characterization of the Metal-Doped 500 °C g-C₃N₄ Material

Co-, Mn-, and Cu-doped g-C₃N₄ synthesized at 500 °C was obtained. Figure 8 shows some selected SEM micrographs of the undoped and metal-containing samples. The undoped material exhibits the typical morphology reported for g-C₃N₄ materials corresponding to a sheet structure with a low thickness and a relatively low porosity [17]. Upon doping the organic semiconductor with the metal domains, modifications in the morphology are observed. For instance, for 500-Co-25% and 500-Cu-25%, solid particles with a relatively low diameter appear. However, there is not a noticeable difference regarding the porosity with respect to the undoped material. Interestingly, for the sample 500-Mn-25%, a marked difference in the porosity of the sample is appreciated with the development of meso- and microporosity.

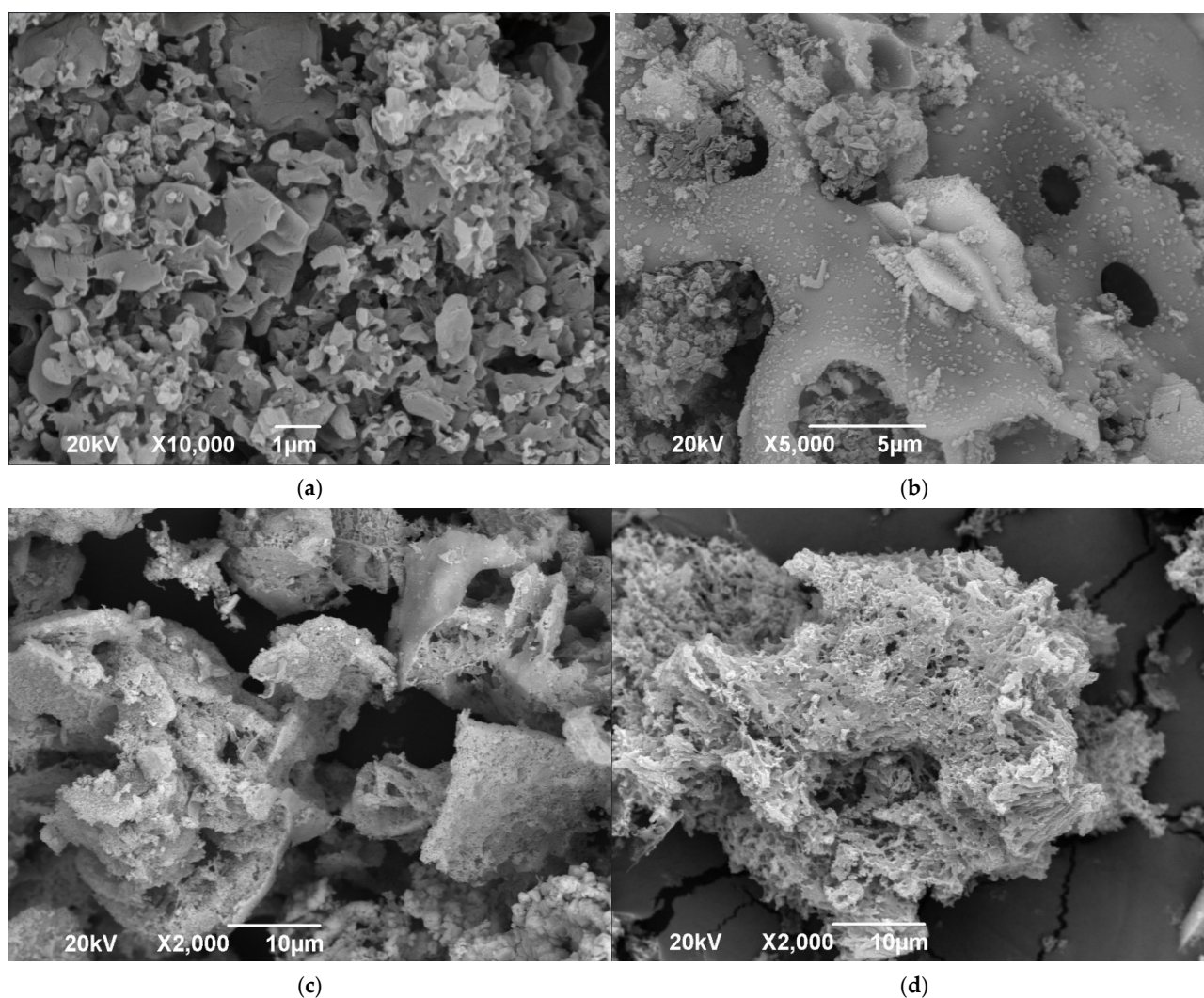


Figure 8. Selected SEM micrographs of the (a) undoped 500 °C, (b) 500-Co-25%, (c) 500-Cu-25%, and (d) 500-Mn-25% samples.

Energy-dispersive spectrometer (EDS) analysis evidenced the presence of C and N atoms for all samples. The undoped material showed a C/N ratio of 1.475, which

differs from the theoretical 0.75 value. This difference might be due to the corresponding processing for the obtention of the SEM micrographs. Nevertheless, the doped materials showed differences in this ratio with values of 1.445, 1.998, and 1.746 for 500-Mn-25%, 500-Cu-25%, and 500-Co-25%, respectively. These values seem to suggest that although Mn doping modifies the morphological properties of the semiconductor, it does not seem to substantially modify the semiconductor structure, while Co and Cu doping results in the loss of N fractions in the organic moieties. Regarding the metal-containing samples, values of 9.10, 46.51, and 18.96% were elucidated for the metals in the same order as previously mentioned.

To elucidate the crystalline nature of the samples, XRD analysis was performed, and the corresponding diffractograms are shown in Figure 9. The undoped material sample exhibits the typical diffractogram for pristine $g\text{-C}_3\text{N}_4$ with diffraction peaks at 12.9° and 27.6° , which correspond to the (1 0 0) and (0 0 2) planes of the in-plane tri-s-triazine repeated units due to $\pi\text{-}\pi$ stacking [18]. Interestingly, the introduction of metals to the systems modifies the diffractograms. The peak corresponding to the plane (0 0 2) moves to higher values of 2θ for the samples 500-Co-25% and 500-Cu-25%, while under the experimental conditions, 500-Mn-25% does not seem to modify this value. These results suggest that Co and Cu atoms are being introduced into the organic structure of $g\text{-C}_3\text{N}_4$, while Mn does not seem to be introduced into the semiconductor structure. This analysis agrees with the invariant C/N ratio only for the 500-Mn-25% sample elucidated through EDS.

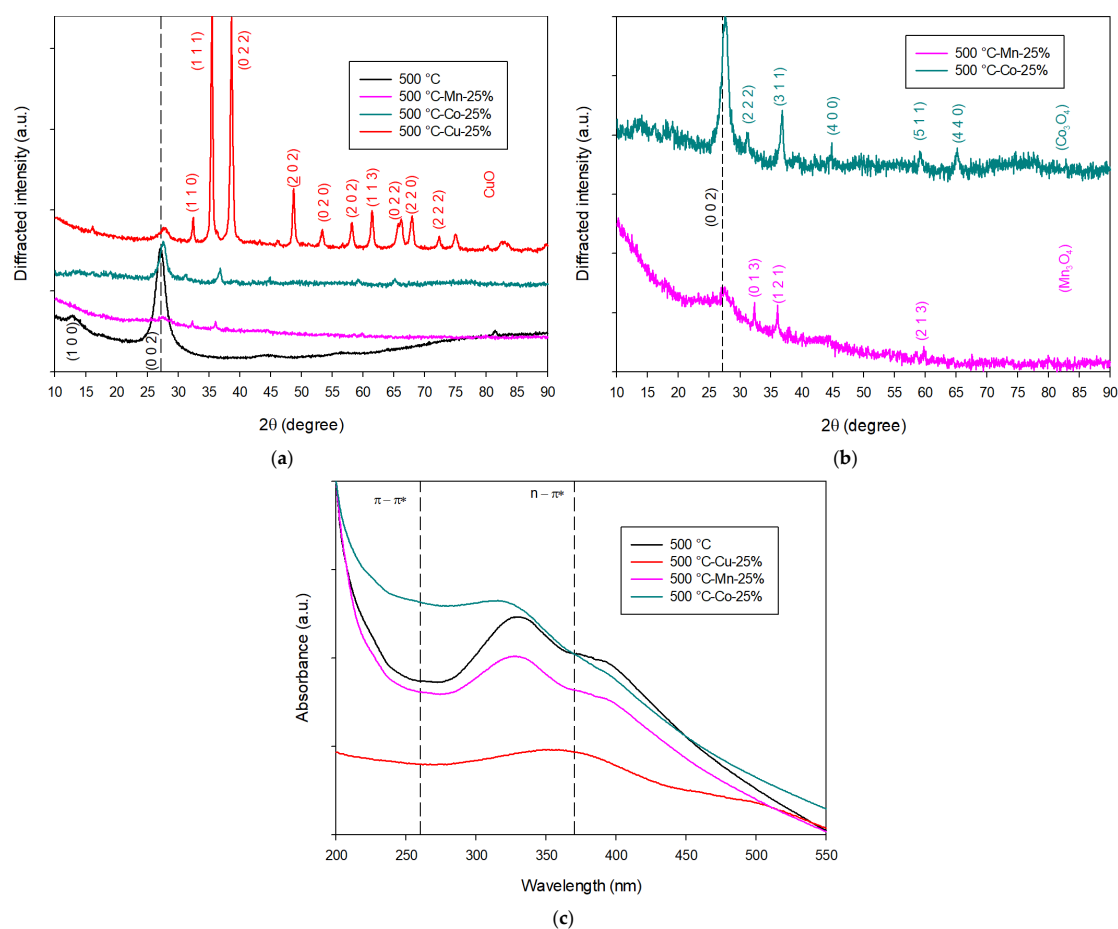


Figure 9. (a) X-ray diffractograms for the synthesized materials at 500 °C. (b) Magnification of 500-Mn-25% and 500-Co-25%. (c) UV/Vis spectra of the synthesized materials at 500 °C.

Nevertheless, in all cases, metal oxide crystalline structures were elucidated. The corresponding crystalline planes are shown directly in Figure 9, which corresponds to CuO

(JCPDS Card No. 48-1545), Co_3O_4 (JCPDS Card No. 42-15467), and Mn_3O_4 (JCPDS Card No. 01-089-4837) for 500-Cu-25%, 500-Co-25%, and 500-Mn-25%, respectively.

Figure 9c shows the UV/Vis spectra of the metal-doped materials. Interestingly, 500-Cu-25% exhibited marked differences when compared to the pristine 500 °C sample. In agreement with the previous characterization results, this might suggest that Cu doping might affect the organic domain, substantially modifying the optical properties of the composites. On the other hand, although 500-Co-25% retains the main spectral features of the pristine 500 °C sample, a blueshift of the $n \rightarrow \pi^*$ transition is seen. Instead, 500-Mn-25% shows an almost identical spectrum with lower absorption coefficients.

The E_{BG} from the Tauc plot suggests the modification from 2.66 eV for the pristine 500 °C sample to 2.51 and 2.45 eV for 500-Mn-25% and 500-Co-25%, respectively. Nevertheless, for the 500-Cu-25% sample, the bandgap corresponds to 1.55 eV, which agrees with the reported value for CuO particles.

2.4. Evaluation of the Metal Type to Enhance the Photocatalytic Disinfection Performance of the 500 °C $g\text{-C}_3\text{N}_4$ Material

The disinfection profile shown in Figure 10 demonstrates the influence of metal doping on the photocatalytic activity of $g\text{-C}_3\text{N}_4$ synthesized at 500 °C, using Mn, Co, and Cu at 25% molar loading. The light and dark controls for the undoped material (blue circles) show a negligible reduction in the *E. coli* concentration over the 8 h period. Moreover, Figure S2 shows the same behavior in dark conditions in the presence of the doped materials. These latter results suggest that neither visible light nor the catalyst alone are sufficient for bacterial inactivation. This confirms that the observed disinfection is due to the photoinduced activity of the materials and not directly related to the oligotoxic effect.

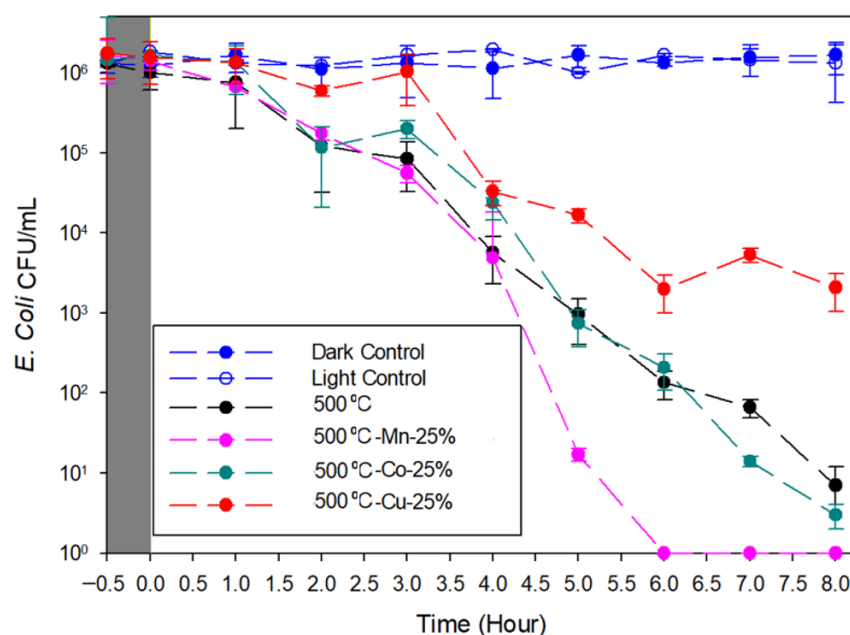


Figure 10. Kinetic profiles of *E. coli* disinfection promoted by $g\text{-C}_3\text{N}_4$ obtained at 500 °C doped with 25% of the metal under irradiation of indoor light. The dark profile shown corresponds to the absence of any material. Conditions: 10 mL and 5 mg of the corresponding material. The error bars represent the standard deviation from duplicate treatments ($n = 2$). The dark region denotes the time in which the LEDs were turned off.

The undoped 500 °C sample exhibits notable photocatalytic disinfection, achieving a reduction of over 5 log units in the *E. coli* concentration after 8 h. This performance agrees with its previously discussed favorable physicochemical properties.

Upon doping with Mn, a dramatic enhancement in photocatalytic performance was observed. The Mn-doped sample (500-Mn-25%) achieved total inactivation of *E. coli* within 6 h, outperforming all other materials. This improvement can be attributed to the role of Mn species in enhancing charge separation, reducing electron–hole recombination [19,20], and possibly participating in Fenton-like or redox cycling processes that amplify ROS generation [21]. In a previous paper, Wang et al., based on computational modeling, demonstrated the doping g-C₃N₄ with Mn affects the atomic arrangement and molecular orbital distribution of the g-C₃N₄ semiconductor, leading to an enhancement in photoinduced carrier separation [22].

The Co-doped sample (500-Co-25%) showed similar performance compared to the undoped catalyst, reaching nearly complete inactivation within 8 h. This result is consistent with the absorption spectra of both materials in the range of light emitted by the UV lamps used (Figure 9). This suggests that under experimental conditions, doping g-C₃N₄ with Co does not result in a kinetic improvement in the photodisinfection properties of the bare carbon nitride.

In contrast, the Cu-doped sample (500-Cu-25%) showed significantly lower photocatalytic activity. While a moderate reduction in bacterial concentration was observed in the first 3–4 h, the inactivation curve reached a plateau at 6 h, indicating limited further disinfection. This is consistent with the reduced light absorption of the material (Figure 9), which could be due to excessive modification of the pristine organic semiconductor structure induced by the introduction of Cu ions, as indicated by XRD and SEM-EDS analysis.

Overall, the data clearly demonstrate that metal doping has a strong influence on the photocatalytic disinfection performance of g-C₃N₄. These findings confirm the importance of dopant selection and its effect on charge dynamics. In this sense, several metal-doped g-C₃N₄ materials have been developed in the literature looking for improved *E. coli* disinfection (Table 2). Because of key differences such as the reactor type, volume of the reaction, matrix conditions, type of bacterial strain, catalyst charge, etc., a direct comparison among them is not an easy task. Considering the relatively low power used here, the reported efficiency of the developed materials in the current work is promising, which makes them of particular significance for incorporation into self-cleaning surfaces for indoor environments.

Among the tested dopants, Mn provides the most substantial enhancement in antibacterial activity, followed by Co, while Cu appears to suppress the performance of the base material. Thus, 500-Mn-25% was selected as the most efficient material for *E. coli* disinfection. In the case of heterogenous photocatalysts involving metallic catalysts, the lixiviation of the metal and its consequent contribution to the disinfection process is of special interest. Indeed, manganese is toxic in excess, but it has also been recognized as a critical micronutrient which acts as a cofactor for superoxide dismutase and protects bacteria against oxidative agents [23]. To further investigate this, Mn²⁺ ions in solution at the end of the treatment were determined by atomic absorption spectroscopy, and a concentration of 12.8 mg L^{−1} was found. Then, a new set of experiments to test the participation of Mn²⁺ ions (MnSO₄) in *E. coli* disinfection was carried out, and the results are shown in Figure S3. As seen, no disinfection activity was observed even after 8 h of treatment. This result ruled out the participation of dissolved Mn²⁺ in the disinfection process.

Table 2. Performance of the different metal-doped g-C₃N₄ materials in *E. coli* disinfection.

Material	Power (W)	Initial CFU/mL	Final CFU/mL	Time (h)	References
Mn/g-C ₃ N ₄	45	10 ⁶	0	6.0	This work
Co/g-C ₃ N ₄	45	10 ⁶	10 ^{0.5}	8.0	This work
Cu/g-C ₃ N ₄	45	10 ⁶	10 ³	6.0	This work
MoS ₂ /g-C ₃ N ₄	100	10 ⁷	0	0.3	[24]
Ni/g-C ₃ N ₄	24	10 ⁸	0	2.2	[25]
AgS ₂ /g-C ₃ N ₄	300	10 ⁷	0	1.5	[26]
MgTiO ₃ /g-C ₃ N ₄	1000	10 ⁷	0	3.0	[27]
TiO ₂ /g-C ₃ N ₄	300	10 ³	0	0.5	[28]
AgBr/g-C ₃ N ₄	300	10 ^{6.5}	0	1.0	[29]

2.5. Evaluation of the Metal Type to Enhance the Photocatalytic Disinfection Performance of the 500 °C g-C₃N₄ Material in Self-Cleaning Surfaces

The disinfection kinetics depicted in Figure 11 illustrate the antibacterial efficacy of self-cleaning surfaces functionalized with sodium-alginate-hydrogel-embedded g-C₃N₄-based photocatalysts, including pristine and transition metal-doped variants (Mn, Co, Cu). To enable direct comparison with liquid-phase suspended photocatalytic systems, an equivalent initial bacterial concentration of 10⁶ CFU/mL was employed, though this microbial load presents a stringent challenge for solid-phase disinfection platforms due to the inherent physicochemical constraints of hydrogel matrices. The observed elongation of disinfection times in immobilized systems (Figure 11), relative to their suspended counterparts, arises from diffusional limitations imposed by the hydrogel's crosslinked polymeric network, which limits reactive oxygen species (ROS) mobility and bacterial cell accessibility. Spatial heterogeneity in photocatalyst distribution, coupled with biofilm-like aggregation of *E. coli* at high cell densities, further exacerbates shielding effects, protecting subsurface cells from ROS-mediated oxidative damage. Additionally, photon flux attenuation within the hydrogel matrix—attributable to light scattering and absorption by the alginate scaffold—reduces the effective irradiation of embedded g-C₃N₄ active sites, diminishing photocatalytic activation efficiency. These collective factors underscore the critical influence of material architecture on disinfection kinetics, highlighting the need to optimize hydrogel porosity, catalyst dispersion, and optical transparency for applications where microbial loads are typically orders of magnitude lower than the experimental benchmark employed here.

Under visible-light irradiation for 24 h, the surfaces containing Mn-doped, Co-doped, and bare g-C₃N₄ all achieved a 6-log reduction in *E. coli* concentration, indicating complete bacterial inactivation. The key difference among these three materials lies in their kinetic response. The Mn-doped surface (500-Mn25%) exhibited the fastest inactivation, reaching more than 4 logs of reduction within 12 h and full disinfection by 24 h. As indicated, this performance enhancement is attributed to the capacity of Mn to promote efficient charge separation and increased ROS generation, likely through redox cycling and band structure modulation.

The Co-doped surface (500-Co25%) also demonstrated excellent antibacterial performance, achieving complete disinfection within the 24 h period, though at a slower rate than the Mn-doped system. This agrees with the suspended disinfection results.

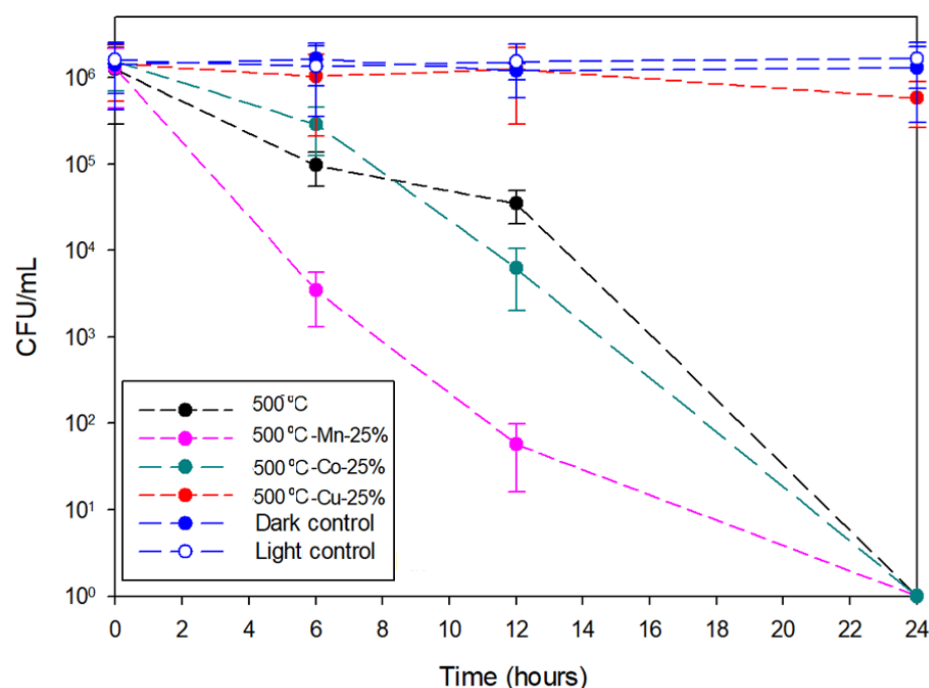


Figure 11. Kinetic profiles of *E. coli* disinfection promoted by g-C₃N₄ doped with 25% of the corresponding metal-based surfaces under irradiation of indoor light. Conditions: 10 mL of bacterial suspension and 10 cm diameter. The error bars represent the standard deviation from duplicate treatments (n = 2).

In contrast, the Cu-doped surface (500-Cu25%) showed negligible antibacterial activity, with *E. coli* concentrations remaining nearly unchanged. This result is consistent with previous suspension-phase tests, where Cu incorporation was found to hinder photocatalytic activity.

The dark and light controls exhibited no significant change in bacterial concentration, confirming that the observed disinfection was exclusively due to photocatalytic processes and not due to thermal effects, photolysis, or passive adsorption by the hydrogel matrix.

Overall, the results confirm that g-C₃N₄ materials synthesized at 500 °C can be successfully integrated into alginate-based hydrogels to form functional self-cleaning surfaces with excellent photocatalytic disinfection capabilities. Among the tested dopants, Mn provides the fastest antibacterial response, followed by Co, while Cu fails to improve or maintain photocatalytic activity in the immobilized system.

3. Materials and Methods

3.1. Reagents

Urea (97% w/w), copper (II) sulfate (CuSO₄), cobalt (II) sulfate hydrate (CoSO₄·H₂O), and manganese (II) sulfate hydrate (MnSO₄·H₂O), all with a purity of 97% w/w, and ammonium hydroxide (NH₄OH), 30% v/v, were provided by Sigma Aldrich, Louis, MO, USA. Type I water was obtained from a Synergy® water purification system (18 MΩ).

3.2. g-C₃N₄ Synthesis

In all cases, 10 g of urea was placed in porcelain crucibles covered with a lid and with aluminum foil. The system was heated from 25 °C using a heating ramp of 1.6 °C min^{−1} to the corresponding temperature and maintained for 3 h. The obtained powders were resuspended 3 times in 100 mL H₂O to remove any soluble impurity. The material was labeled with the temperature used in the synthesis. For instance, 500 is g-C₃N₄ pyrolyzed at 500 °C.

3.3. $g\text{-C}_3\text{N}_4$ -Metal Composite Synthesis

For the synthesis, 2 g of graphitic carbon nitride ($g\text{-C}_3\text{N}_4$) was suspended in 150 mL of water under constant mechanical stirring for 30 min. Subsequently, a specific mass of the selected metal chloride precursor (Mn, Co, or Cu) was added. The metal precursor quantities correspond to 0.573 mmol per gram of $g\text{-C}_3\text{N}_4$.

Following the precursor's addition, the system was kept under continuous magnetic stirring for two hours to ensure equilibrium in the adsorption–desorption process. Afterward, 10 mL of NH_4OH (25% v/v) was added to the mixture. The obtained material was centrifuged and washed three times with distilled water, then dried at 80 °C for 24 h.

The resulting dried powder was subsequently calcined at 450 °C for 4 h. The calcined powders exhibited distinct coloration compared to the pristine graphitic carbon nitride precursor. The materials were purified by resuspension three times in water and ultrasonicated three times and then dried at 80 °C.

Figure 12 represents the synthesis procedure for obtaining the metal-doped composites.

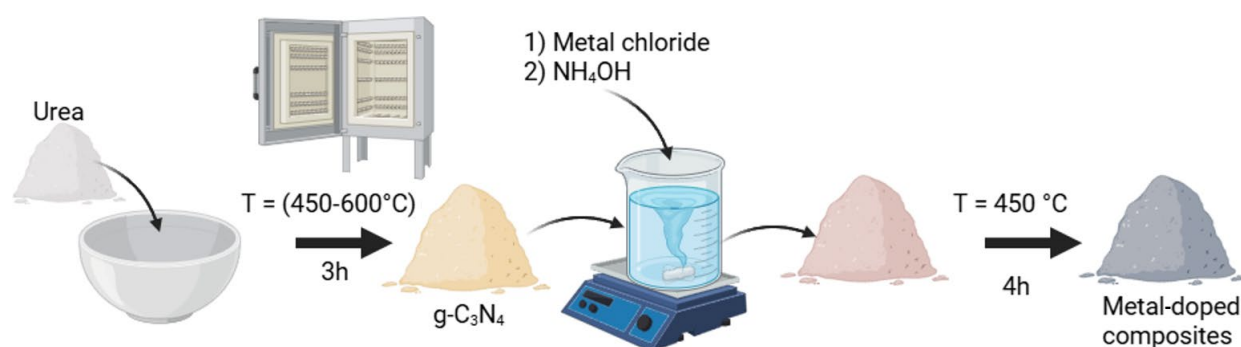


Figure 12. Schematic representation of the synthesis route of the metal-doped composites.

3.4. Preparation of $g\text{-C}_3\text{N}_4$ -Based Self-Cleaning Surfaces

A modified version of a previously reported protocol was employed [30]. Briefly, 1.2 g of sodium alginate was dissolved in preheated water at 90 °C under continuous stirring for 2 h. Simultaneously, 0.3 g of the selected $g\text{-C}_3\text{N}_4$ was dispersed in 20 mL of water and subjected to ultrasonication for 15 min before being incorporated into the alginate solution. The resulting mixture was stirred (300 rpm) at 25 °C for 24 h. Subsequently, 20 mL of the prepared suspension was transferred into a 10 cm diameter Petri dish and left undisturbed at 25 °C for 36 h.

3.5. *E. coli* Disinfection Tests

Disinfection experiments were conducted using an *Escherichia coli* strain (*E. coli* ATCC 8739) in a 0.8% saline solution. The irradiation source consisted of eight LED lamps with visible irradiation with a total power of 45 W. The corresponding irradiance spectrum is shown in Figure S1. *E. coli* was activated 24 h before disinfection, having undergone a cultivation process to ensure a bacterial population in the stationary phase. Following activation, the concentration was adjusted to 1.5×10^8 CFU/mL based on the 0.5 McFarland scale, equivalent to an absorbance range of 0.08 to 0.13 at 650 nm.

The experiments were performed in duplicate, utilizing 50 mL flasks. In each flask, 10 mL of saline solution (0.8% wt) containing 5 mg of the respective material was subjected to 30 min of ultrasound action (50 kHz and 50 W/L). Subsequently, 100 μL of the McFarland bacterial suspension was added, resulting in an initial concentration of 1.5×10^6 CFU/mL. The systems were placed on an orbital shaker operating at 150 rpm. Samples of 100 μL were extracted at 60 min intervals, except for the first 30 min.

For the final step, samples collected at the selected times were diluted to a 10^{-4} or 10^{-3} concentration and placed on agar plates for incubation at 37 °C during 24 h, prior to the counting of the growing colonies.

Another set of experiments was performed using self-cleaning surfaces (*vide infra*). For this purpose, the previous method was modified. In total, 10 mL of saline solution containing 1.5×10^6 CFU/mL was deposited onto a Petri dish containing the g-C₃N₄-based surface. The system was then introduced into an incubator at 37 °C for 45 min to promote solvent evaporation. Then, the Petri dish was irradiated, as previously discussed. The recovery of the *E. coli* from the surface was conducted with two steps of 10 mL of saline solution, which was further diluted and treated, as indicated previously.

3.6. Material Characterization

UV/Vis spectroscopy of the materials was conducted to identify characteristic absorption bands associated with metallic and organic moieties. Measurements were performed using a PerkinElmer Lambda 25 UV/Vis spectrophotometer manufactured by PerkinElmer (Hopkinton, MA, USA), equipped with a BaSO₄ integrating sphere, operating in the 200–800 nm wavelength range. For the analysis, 2 mL of a 100 ppm aqueous suspension of each material was placed in a quartz cell with an optical path length of 1 cm.

The bandgap energy of the materials was determined using a Tauc plot derived from the UV/Vis spectra, following Equation (3):

$$(\alpha h\nu)^n = A(h\nu - E_{BG}) \quad (3)$$

where α represents the absorption coefficient, h is Planck's constant, ν is the photon frequency, and n is the exponent defining the type of electronic transition. A is a proportionality constant, and E_{BG} corresponds to the bandgap energy. The E_{BG} value was estimated by extrapolating the linear region of the $(\alpha h\nu)^n$ vs. $h\nu$ plot.

The functional groups on the material surfaces were characterized using infrared (IR) spectroscopy with a Fourier transform infrared spectrophotometer (FTIR), model IRTracer-100 from Shimadzu (Beijing, China). The measurements were performed in the 4000–550 cm^{−1} range with a resolution of 4 cm^{−1}. The analysis was conducted using an attenuated total reflectance (ATR) crystal, allowing direct measurement of the samples without the need for additional preparation.

Electrochemical characterization of the materials was carried out following the protocol previously reported [18]. The materials were supported over Fluorine-doped Tin Oxide (FTO). The measurements were conducted using a PalmSens4 Electrochemical interface provided by PalmSens BV (Houten, The Netherlands) and the corresponding software PSTrace 5.9.415.

The surface structure was investigated through scanning electron microscopy (SEM) utilizing a JEOL JSM-6490LV model manufactured by JEOL (Akishima, Tokyo, Japan). Furthermore, an energy-dispersive spectrometer (EDS) was integrated with SEM to ascertain the elemental composition.

The confirmation of the crystalline phase of selected samples was conducted using the X-ray diffraction (XRD) technique in a PANalytical Empyrean X-ray 2012 powder diffractometer (manufactured by Malvern Panalytical (Almelo, The Netherlands)), operated in reflection transmission spinner geometry with Cu-K α radiation at 40 mA and 45 kV and in the reflection mode between 10 and 90° (2 θ angle).

To evidence Mn leaching, atomic absorption spectroscopy was used. The final bacterial suspension and particles were centrifuged and then acidified with 1 mL of HNO₃ [31].

4. Conclusions

In this study, g-C₃N₄ photocatalysts were successfully synthesized at different temperatures and systematically characterized to identify the optimal conditions for photocatalytic disinfection. Comprehensive structural, optical, electronic, and electrochemical analyses revealed that materials synthesized at 450–550 °C maintained favorable properties, with the 500 °C sample demonstrating the best balance between band structure, charge transport, and ROS generation capacity. Photocatalytic disinfection tests against *E. coli* confirmed that the 500 °C material exhibited the most rapid bacterial inactivation kinetics in aqueous suspension, leading to its selection for further modification via metal doping.

Doping the 500 °C g-C₃N₄ with transition metals (Mn, Co, Cu) revealed that Mn significantly enhanced the photocatalytic performance, while Cu doping hindered activity. Mn doping led to the most rapid inactivation kinetics.

The selected materials were then integrated into sodium alginate matrices to fabricate self-cleaning surfaces. These surfaces retained photocatalytic activity under visible light and achieved complete *E. coli* inactivation (6-log reduction) within 24 h. Among them, the Mn-doped g-C₃N₄ surface displayed the fastest response, achieving near-total disinfection within just 12 h, followed by the Co-doped and undoped surfaces. The Cu-doped material showed negligible activity in its immobilized form, which was consistent with its poor performance in suspension.

Overall, this work demonstrates the successful development of metal-doped g-C₃N₄-based self-cleaning surfaces with enhanced disinfection kinetics under visible light. The Mn-modified materials, especially in hydrogel form, offer a promising strategy for practical antimicrobial applications where rapid photocatalytic response is essential.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/inorganics13080262/s1>: Figure S1: Irradiance spectrum of the used LEDs. Figure S2. *E. coli* disinfection kinetic profiles promoted by g-C₃N₄ obtained at 500 °C doped with 25% of the corresponding metal under dark conditions. Conditions: 10 mL of solution and 5 mg of the corresponding material. The error bars represent the standard deviation from duplicate treatments (n = 2) and Figure S3. *E. coli* disinfection kinetic profiles in presence of soluble Mn²⁺ (12.8 mg/L) and under irradiation of indoor light. The error bars represent the standard deviation from duplicate treatments (n = 2).

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