Mediated Growth of Carbon Nitride Films via Spray-Coated Seeding Layers for Photoelectrochemical Applications

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Polymeric carbon nitride (CN) has emerged as a promising semiconductor in photoelectrochemical devices, thanks to its unique electronic and catalytic properties, low price, stability in various chemical environments, and benign nature. Decent progress in the deposition and growth of CN layers on substrates has been achieved using several deposition and growth methods; however, the properties of the layer, including the quality of its contact with the substrate and its structural properties, are still largely dependent on the surface properties of said substrate. Here, a new approach is introduced in which a spray-coated seeding layer composed of CN monomers directs the growth of a homogenous, thick CN layer on a substrate by calcination at high temperature in the presence of melamine vapor. Uniform CN layers with strong adhesion to the substrate are obtained. The influence of the seeding layer and the vapor composition on the photoelectrochemical, optical, and structural properties is studied in detail. The best-performing electrode, based on urea as the seeding layer, demonstrates good activity as a photoanode in photoelectrochemical cells, reaching up to 300 µA cm⁻² in the presence of a hole scavenger.

1. Introduction

Polymeric carbon nitride materials (CNs) have emerged as low-cost, chemically stable photo- and electro-catalysts for various reactions, such as water splitting, CO₂ reduction, and other organic transformation reactions.[1–10] In the last few years, efforts to integrate carbon nitride films into photoelectronic and photoelectrochemical devices have increased, motivated by the unique photophysical properties of carbon nitride (energy band positions and tunable bandgap) and its stability under harsh chemical environments.[11,12] However, the deposition/growth of a CN layer in intimate contact with the substrate remains an ongoing challenge.[13–15] To this end, several gas-phase-based methods to grow CN layers on a substrate have been developed: chemical vapor deposition (CVD),[16,17] thermal vapor condensation (TVC),[18–21] microwave-assisted condensation,[22] alongside others. However, in all scenarios, one of the main bottlenecks for the growth of a homogenous and firmly attached layer is the poor interaction of CN with the surface of the substrate. This is especially the case for methods involving the adsorption of a gas-phase reactant (e.g., CVD) since solid–gas interactions between the substrate and the CN monomer are required for the reaction to occur.[23] Therefore, the CN coatings obtained so far have mostly consisted of thin, poorly attached layers.

One approach to directing the growth of materials by a gas-phase reaction is to use a seeding layer to facilitate the nucleation process.[24–27] Furthermore, a thicker layer can be prepared by this approach, owing to the large roughness induced by the seeding layer compared to a flat substrate. For a controlled gas-phase growth of a CN layer on a substrate, a homogenous seeding layer of controlled thickness consisting of CN monomers should be deposited before the thermal condensation. Spray coating is a facile, repeatable, and large-scale method to coat a substrate with a wide diversity of molecules and materials.[28,29] In this technique, a liquid is atomized into a mist of even micrometer-sized droplets using ultrasonic vibrations in the nozzle; it can then be easily and controllably deposited as a layer on a variety of (possibly heated) substrates.

Herein we report the use of spray coating to deposit a seeding layer of urea and other CN monomers, for the direct growth of CN films on substrates by calcination in the presence of melamine vapor. The seeding layer provides nucleation sites promoting the growth of the CN film in the form of a homogenous layer in intimate contact with the substrate, thus improving the photoelectronic properties of the electrode. The morphology, structural properties, and optical properties of the obtained CN films were investigated. The photoelectrochemical performance was measured for a model water-splitting photovoltaiccell (PEC) setup; specifically, hydrogen evolution reaction in the presence of a hole scavenger was measured, which accords with the United Nation’s sustainable development goals as a source of affordable and clean energy.[30–34]

2. Results and Discussion

2.1. Preparation and Characterization of Urea Films

The synthetic route is illustrated in Figure 1a. The first step is the deposition of a CN-precursor seeding layer of controlled
thickness. Here, an aqueous urea solution (375 g L\(^{-1}\)) was sprayed on fluorine-doped tin oxide (FTO) coated glass using an ultrasonic spray coater, forming a uniform layer of urea on the FTO. The spraying process includes a variable number of repetitive spray cycles; several films were prepared and labeled as \(U_x\), where \(x\) represents the number of spray cycles: 5, 10, 50, or 100. The X-ray diffraction (XRD) patterns of \(U_{10}\) and urea powder exhibit similar features (Figure 1b); the additional diffraction signals for \(U_{10}\) are attributed to the FTO substrate. The Fourier-transform infrared (FTIR) spectra of \(U_{10}\) and urea powder show similar N–H vibrations in the range of 3230–3410 cm\(^{-1}\), as well as a C=O stretching band at \(\approx 1660\) cm\(^{-1}\), which further confirms the successful deposition of urea on the FTO (Figure S1, Supporting Information). Top-view and cross-sectional scanning electron microscopy (SEM) images disclose the morphology of the obtained \(U_x\) films: as the number of spray cycles increases, the layer becomes thicker and rougher (Figure 1c; and Figure S2, Supporting Information).

2.2. Preparation and Characterization of CN Films

The calcination of the \(U_x\) films in the presence of melamine powder (0.5 g at the bottom of a closed, but not sealed, reaction glass tube at 550 °C under \(N_2\) atmosphere for 4 h) led to the formation of CN electrodes (labeled as CN–\(U_{10}\)). During the thermal treatment, the melamine sublimes, and the vapor is deposited on the urea-coated FTO substrate, where the urea coating serves as a seeding layer for the growth of the CN. Motivated by the high photocurrent density of CN–\(U_{10}\), we prepared another electrode by spraying 10 cycles of urea solution as a seeding layer, followed by calcination in the presence of 1.0 g of melamine (labeled as CN–\(U_{10}\)) instead of the 0.5 g that was used in all other cases.

To evaluate its importance in the formation of the final CN film, we performed a similar calcination process in the absence of the seeding layer (namely, the CN film was created by deposition of melamine vapor on bare FTO, labeled as CN–\(M_{0.5}\)). Without the seeding layer, the process resulted in incomplete coverage of the substrate by the CN film. Indeed, top-view and cross-sectional SEM images show that CN–\(M_{0.5}\) consists of a patchy, uneven distribution of CN structures that only partially covers the FTO surface (Figure 2a,b). In contrast, while CN–\(U_{10}\) has a similar morphology to that of CN–\(M_{0.5}\), it provides better coverage of the substrate (Figure S3a,b, Supporting Information). This difference proves the critical role of the seeding layer in the formation of a uniform CN film on the FTO. We also performed the same calcination process in the absence of melamine powder in the reaction tube; most of the urea film sublimed during calcination, and no substantial CN film was formed on the FTO. Therefore, we conclude that both the seeding layer and the melamine vapor are essential for the growth of a CN layer that fully covers the FTO.

The top-view and cross-sectional SEM images of CN–\(U_{10}\), CN–\(U_{10}\), and CN–\(U_{10}\), all show uniform coating of the FTO. However, their cross-sectional SEM images reveal differences: CN–\(U_{10}\) is very compact and has the fullest contact to the FTO and a uniform layer thickness, whereas the contact of CN–\(U_{10}\) and CN–\(U_{10}\) with the FTO is less homogenous. Besides, CN–\(U_{10}\) shows...
significant variations in its thickness, which is likely due to an excess of melamine powder during calcination.

The XRD patterns of the obtained CN electrodes (Figure 3a) exhibit two typical CN signals corresponding to an in-plane (100) diffraction at 12.8° and an interplanar stacking (002) diffraction at 27.6°. The FTIR spectra of the CN electrodes (Figure S4, Supporting Information) show signals in the 1100–1700 cm$^{-1}$ range that correspond to C$\equiv$N heterocycles stretching vibrations.

Figure 2. a) Top-view (inset: digital photo) and b) cross-sectional SEM images of CN–M$_{0.5}$. c) Top-view (inset: digital photo) and d) cross-sectional SEM images of CN–U$_{10}$M$_{0.5}$.

Figure 3. a) XRD patterns of the CN electrodes; the diffraction signals at 26.4°, 33.6°, 37.7°, and 51.5° are from the FTO substrate, and (bottom, black) XRD pattern of a bare FTO substrate. b) Absorbance spectra of the CN electrodes. c) Tauc plot analysis for the determination of the optical bandgaps of the CN electrodes. d) PL spectra of the CN electrodes; $\lambda_{ex} = 390$ nm.
modes. The peaks at \( \approx 800 \text{ cm}^{-1} \) are attributed to the breathing vibration of the heptazine units in CN films, whereas the broad bands at \( \approx 3000–3600 \text{ cm}^{-1} \) match the stretching vibration of N–H in the amino groups resulting from incomplete polymerization. The X-ray photoelectron spectroscopy (XPS) spectra of CN–U\(_{10}\)M\(_{0.5}\) (Figure S5, Supporting Information) confirm the successful growth of CN films. The C1s spectrum presents three peaks located at 284.9, 288.0, and 289.4 eV, which correspond to C–C from the carbonaceous environment, N–C=N coordination, and O–C=O, respectively, with the latter possibly resulting from partial oxidation of the CN film.[37] The N1s spectrum reveals three peaks located at 398.4, 399.5, and 400.8 eV, attributed to C–N–C, N–(C)\(_3\), and N–H, respectively. Hence, the XRD, FTIR, and XPS results confirm the formation of tri-s-triazine-based CN.

The UV–vis absorbance spectra of all CN electrodes exhibit similar features except that of CN–M\(_{0.5}\), which displays the highest transmittance, owing to its uneven appearance of CN–M\(_{0.5}\) (Figure 3b). A Tauc plot analysis (Figure 3c) reveals an optical bandgap in the 2.60–2.64 eV range for the CN–U\(_{10}\)M\(_{y}\) films, as expected for CN materials.[38] The CN–U\(_{10}\)M\(_{0.5}\) film has the narrowest estimated bandgap (2.60 eV). It also exhibits the lowest photoluminescence (PL) intensity (Figure 3d), suggesting nonradiative recombination paths of photogenerated electron–hole pairs caused by surface states.

### 2.3. Adhesion Test

To evaluate the mechanical stability and adhesion of the CN films to the FTO, we covered CN–U\(_{10}\)M\(_{0.5}\), CN–M\(_{0.5}\), and CN–U\(_{10}\)M\(_1\) with scotch tape and placed a heavy object (\( \approx 1.5 \text{ kg} \)) on top of each sample for two hours (Figure S6, Supporting Information). Removal of the scotch tape revealed that CN–U\(_{10}\)M\(_{0.5}\) largely remained uniform, whereas CN–M\(_{0.5}\) was affected to a greater extent, clearly illustrating the benefit of using a seeding layer. CN–U\(_{10}\)M\(_1\) peeled off partially, probably because of the thickness differences in different areas of the electrode, as its cross-sectional SEM image shows (Figure S3h, Supporting Information).

### 2.4. Versatility of the Method

The deposition of a urea seeding layer by spray coating followed by the growth of a CN film from melamine vapor during calcination was also achieved on a glass slide and carbon cloth (Figure S7a–c, Supporting Information). A similar procedure was successfully carried out on a 9 x 1.2 cm\(^2\) FTO slide to show the applicability of the method to larger surfaces (Figure S7d, Supporting Information). Furthermore, for the sake of example, we successfully used thiourea and alanine instead of urea as the seeding layer (Figure S7e,f, respectively, Supporting Information) and substituted thiourea for melamine as the precursor that undergoes sublimation in the test tube (Figure S7g, Supporting Information). These proof-of-principle results indicate that the CN electrodes can be prepared from different monomers and that the seeding layer promotes the growth of a CN film regardless of the type and size of the substrate. However, further optimization is needed for each system.

### 2.5. PEC Performance of the CN Electrodes

The activity of the CN electrodes as water-splitting photoanodes was tested in a standard three-electrode system. Chronoamperometric measurements of the CN electrodes were performed at 1.23 V versus reversible hydrogen electrode (RHE) in 0.1 m KOH under 1-sun front- and back-side illumination (Figure 4a; and Figure S8, Supporting Information). The highest photocurrent density (\( \approx 110 \mu A \text{ cm}^{-2} \)) was obtained using a CN–U\(_{10}\)M\(_{0.5}\) photoelectrode under back-side illumination. We ascribe this to the electron diffusion length being the limiting factor in charge separation and transport.[39] Additional chronoamperometry measurements were conducted in the presence of triethanolamine (TEOA) as a hole scavenger (10% v/v TEOA in 0.1 m KOH) to test the efficiency of electron transport, assuming most photogenerated holes are consumed by TEOA (Figure 4b; and Figure S9, Supporting Information). Under these conditions, both photocurrent density values and stability were improved in all the CN electrodes, thanks to the improved hole extraction, which hinders the recombination of photogenerated electron–hole pairs and the self-oxidation of the CN layer. Linear sweep voltammetry (LSV) curves in the dark and under 1-sun back-side illumination (Figure 4c) disclose a low onset potential of 0.17 V versus RHE for CN–U\(_{10}\)M\(_{0.5}\). All the CN electrodes exhibited a linear increase in photocurrent density with increased potential—a typical PEC behavior—and a sharp increase in current at \( \approx 1.7 \text{ V} \) versus RHE attributed to electrocatalytic water oxidation.

The charge transfer efficiency of CN–U\(_{10}\)M\(_{0.5}\) reached up to 53% at 1.6 V versus RHE (Figure S10, Supporting Information). Incident photon-to-current conversion efficiency (IPCE) measurements of CN–U\(_{10}\)M\(_{0.5}\) were performed at 1.23 V versus RHE in both 0.1 m KOH and 10% v/v TEOA in 0.1 m KOH (Figure 4d). The highest efficiency was achieved at 380 nm in both cases; the presence of a hole scavenger improved the IPCE value from 1.7% to 3.9%. In addition, CN–U\(_{10}\)M\(_{0.5}\) showed a good stability in the presence of TEOA, with a photocurrent value of \( \approx 60 \mu A \text{ cm}^{-2} \) even after 16 h of continuous illumination (Figure S11, Supporting Information). Additional chronoamperometric measurements of CN–U\(_{10}\)M\(_{0.5}\) were performed at 1.23 V versus RHE in a neutral aqueous solution (0.1 m phosphate buffer, pH 7) under 1-sun front- and back-side illumination (Figure S12, Supporting Information). Under these conditions, a lower photocurrent density (\( \approx 65 \mu A \text{ cm}^{-2} \)) was obtained compared to the photoanode’s performance in 0.1 m KOH; no apparent change in stability was observed. Using Mott–Schottky analysis of the electrodes (Figure S13, Supporting Information) and the optical bandgaps (Figure 3c), we calculated the energy diagram presented in Figure S14 (Supporting Information), which shows the band positions of all the CN electrodes. Moreover, despite morphological changes of the CN film following PEC measurements in alkaline conditions, its contact with the substrate remained quite homogenous (Figure S15, Supporting Information). The XRD patterns of CN–U\(_{10}\)M\(_{0.5}\) before and after PEC measurements do not exhibit any significant difference.
(Figure S16, Supporting Information), while an energy dispersive X-ray spectroscopy (EDS) analysis of the photoanode post-PEC measurements reveals a minor increase in oxygen quantity, suggesting oxidation of CN film during the measurements (Figure S17 and Table S1, Supporting Information).

The measured PEC performances of the various CN photoelectrodes, together with the morphology analysis of the urea films before calcination and the corresponding CN photoelectrodes, show that a seeding layer (∼12 µm) is of great benefit for the formation of a homogenous CN film firmly attached to the substrate.

3. Conclusions

To conclude, we reported a facile and scalable method to grow a homogenous CN layer on a substrate, with good adhesion, by providing nucleation sites for the growth of CN from the gas phase. To do so, we coated the substrate with CN precursors, using a spray coating technique before the thermal condensation. Our results indicate that the seeding layer promotes the growth of a uniform CN film regardless of the type and size of the substrate. The influence of the spray coating parameters (composition, thickness, and solvent) and the vapor content were studied in detail and correlated with the structural, photophysical, and photoelectrochemical properties of the CN layer. The new electrodes exhibit good performance as photoanodes in photoelectrochemical cells, i.e., low onset potential, high photocurrent density, and moderate photostability. This approach facilitates the growth of CN on various substrates with controlled properties; its extension to other CN and metal-free materials on various substrates with controlled properties should accelerate their integration into photoelectrochemical devices.

4. Experimental Section

Materials: All reagents and solvents were used as received from their respective commercial vendors, without further purification: urea (98%) from Glentham; thiourea (99%) from Acros Organics; L-alanine (>95%) from Fisher Bioreagents; melamine (99%) from Sigma-Aldrich. KOH (85%) and Na2SO4 (99%) from Loba-Chemie; Na2HPO4 (98%) and NaH2PO4 (96%) from Alfa Aesar; acetone (99.8%) and ethanol (absolute, 99.9%) from Bio-Lab Chemicals. Deionized water (18.2 MΩ cm resistivity at 25 °C, purified using a Millipore Direct-Q3 system) was used for all aqueous solutions. FTO-coated glass (12–14 Ω sq−1) was purchased from Xop Glass company, Spain. Before use, the FTO was cut into rectangular pieces (1.3 × 2.5 cm2) and sonicated with an aqueous detergent solution (Alconox, 1% m/v), acetone, and ethanol successively, for 15 min each, then dried in an air oven at 60 °C.

Preparation of Urea/FTO Films: First, an aqueous urea solution (375 g L−1) was prepared by dissolving 15 g of urea in 40 mL of water and then shaking the mixture for 15 min. Next, the solution was sprayed on an FTO substrate heated to 45 °C using a Sono-Tek ExactaCoat ultrasonic spray coater, with a varying number of spray cycles (5, 10, 50, or 100) and the following constant spray parameters: shaping air pressure of 0.5 psi (∼3.5 kPa), dwell time of 10 s between spray cycles, and distance of 4.0 cm between the spray nozzle and the FTO. Finally, the urea/FTO films were dried in an air oven at 60 °C (these films are denoted as Ux, x being the number of spray cycles). The choice of solvent and CN precursor to be sprayed is determined mainly according to solubility—urea was chosen to be deposited as a seeding layer due to...
its high solubility in water, which enables the formation of uniform films with controlled thickness by the spray coater.

Preparation of CN Films: Each piece of urea/FTO was inserted into a glass test tube (16 mm diameter × 100 mm length) filled with the chosen amount of melamine powder (0.5 or 1.0 g) at its closed end. The tube was purged with N₂ for 1 min and closed with an Al foil. Lastly, the tube was submitted to a calcination process of 4 h at 550 °C with a heating ramp of 4.2 °C min⁻¹ under N₂ atmosphere to yield CN electrodes denoted as CN–U₉M₁₀, i.e., deposition of a seeding layer by 10 spraying cycles of urea, then calcination in the presence of 0.5 g of melamine. For the growth of a CN film on a 9 × 1.2 cm² FTO slide, the procedure for the preparation of CN–U₉M₁₀ was applied, only in a larger glass test tube (16 mm diameter × 150 mm length). The deposition of thiourea and L-alanine as seeding layers was achieved by spraying 10 cycles of aqueous thiourea (150 g L⁻¹) or aqueous L-alanine (125 g L⁻¹) solution, respectively.

Characterization: The morphology of the samples was characterized using a SEM (FEI VERIOS 460L) operated at 3.5 kV, 25 pA, and using an Everhart–Thornley secondary electron detector; the samples were sputtered with ~7 nm gold before analysis. EDS was performed using the aforementioned microscope, operated at 6 kV with a probe current of 13 nA. X-ray diffraction (XRD) patterns were measured using a PANalytical Empyrean diffractometer (equipped with an X'Celerator position-sensitive detector) with a scanning time of ~7 min for a 2θ range of 5°–60°, using Cu Kα radiation (λ = 1.54 Å)–operation parameters: 40 kV, 30 mA. FTIR spectra were collected on a Thermo Scientific Nicolet iS5 using a diamond #7 ATR. The chemical states of key elements were deduced from XPS measurements conducted on a Thermo Fisher Scientific ESCALAB 250 using monochromated Kr X-rays; all the binding energies in the XPS spectra were calibrated relative to the C 1s peak at 284.8 eV. UV–vis spectra were recorded on a Cary 100 spectrophotometer equipped with a diffuse reflectance accessory in both transmission (T) and reflectance (R) modes; the absorbance was calculated as: 100 − T − R (%). A Horiba Scientific FluoroMax 4 fluorimeter was used to collect PL spectra, with an excitation wavelength of λ_ex = 390 nm.

Photoelectrochemical Measurements: All photoelectrochemical measurements were performed using a three-electrode system with a Pt plate (1.0 cm²) and Ag/AgCl (saturated KCl) as the counter and reference electrodes, respectively. The electrolyte was either 0.1 m KOH or phosphate buffer (pH ≈ 13), 0.1 m KOH solution containing 10% v/v TEOA. It was assumed that the extraction rate of photogenerated holes in the system is 100% after the addition of the hole scavenger. Mott–Schottky measurements were carried out in 1.0 m Na₂SO₄ aqueous solution (pH = 6.3).

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
Research data are not shared.

Keywords
carbon nitride, photoelectrochemical cells, spray coating, water splitting

The charge transfer efficiency was calculated using:

\[ \eta(\%) = \frac{J_{\text{TEOA}} \times 100}{J_{\text{KOH}}} \]  \hspace{1cm} (3)

\[ J_{\text{KOH}} \] is the photocurrent density obtained in 0.1 m KOH aqueous solution, while \( J_{\text{TEOA}} \) is the photocurrent density obtained in 0.1 m KOH containing 10% v/v TEOA. It was assumed that the extraction rate of photogenerated holes in the system is 100% after the addition of the hole scavenger.

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