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Article

Rhodamine B Dye–Functionalized Hydrophobic Carbon Quantum Dots with Dual Emission for White-Light Organic Optoelectronic Devices

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Abstract

Hydrophobic carbon quantum dots (hbCQDs) with tunable photoluminescence were synthesized via a solvothermal approach and further hybridized with Rhodamine B (RhB) to extend emission into the visible range. The hbCQDs exhibit quasi-spherical morphology with an average particle size of 8 nm and predominantly disordered graphitic structure, as confirmed by TEM and XRD analyses. FTIR and XPS characterizations reveal surface functional groups including C–N, C=O/C–O, and S–H, which govern the photoluminescence properties. Pure hbCQDs display blue emission at 453 nm under excitation, with a quantum yield (QY) of 6.2%. Incorporation of RhB leads to dual-emission behavior: the surface-state emission remains in the blue region, while molecular-state emission from RhB appears in the orange-red region. The 0.2 mL RhB–CQD composite exhibits optimal properties, including a QY of 13% and a production yield of 82%, emitting white light under 365 nm UV excitation. Increasing RhB loading to 0.4 mL results in a shift of emission peaks and a reduced QY (<9%), with weaker orange fluorescence. These findings demonstrate that controlled RhB hybridization effectively tunes the emission spectrum of hbCQDs, offering a simple and reproducible strategy to achieve dual-color and white-light emission. The optimized hbCQDs/RhB composites hold significant potential for applications in hydrophobic media-compatible organic optoelectronics, light-emitting devices, and bioimaging.

Keywords: hydrophobic carbon quantum dots; rhodamine B composites; photoluminescence; dual-emission; white-light emission; surface functionalization; quantum yield; solvothermal synthesis; organic optoelectronic materials

1. Introduction

Carbon quantum dots (CQDs) have emerged as an important class of zero-dimensional carbon-based nanomaterials owing to their tunable photoluminescence, excellent chemical stability, and low toxicity [1–3]. Their distinctive optical characteristics, including size-dependent emission and surface-state-mediated fluorescence, have enabled a wide range of applications in bioimaging, chemical sensing, photocatalysis, and optoelectronic devices [4] [5] [6] [7]. However, most reported CQDs are hydrophilic in nature [8] [9] [10], which limits their compatibility with nonpolar environments and restricts their integration into hydrophobic matrices or organic optoelectronic systems. In addition, extending the emission of CQDs toward the low-energy visible and red spectral regions remains a significant challenge. Typically, pristine CQDs exhibit dominant blue emission arising from surface defect states [11] [12], while red or white-light emission is relatively rare and usually requires complex surface passivation [13] [14] [15] or hybridization strategies [16] [17] [18].

To overcome these limitations, hybrid nanostructures that combine CQDs with luminescent organic dyes have recently attracted considerable attention. In such systems, CQDs can function as broadband surface-state emitters or energy donors, whereas organic dyes contribute strong and well-

defined molecular fluorescence in the visible region. Among the available dyes, Rhodamine B (RhB) has emerged as a promising candidate due to its high molar absorption coefficient, excellent photostability, and intense emission in the orange-red region of the spectrum [19] [20] [21].

The spectral overlap between the emission of CQDs and the absorption band of RhB facilitates efficient interfacial photophysical processes such as Förster resonance energy transfer (FRET) or photoinduced charge transfer, which can broaden the overall emission bandwidth and improve light-harvesting efficiency [22] [23] [24]. Through these interactions, the excited-state energy generated within CQDs can be partially transferred to RhB molecules, thereby modifying exciton recombination pathways and extending the emission toward longer wavelengths. As a result, CQD-RhB hybrid systems can exhibit dual-emission behavior and broadened photoluminescence spectra, which are highly desirable for achieving tunable visible-light emission and potentially near white-light generation under ultraviolet excitation [11] [12].

Although organic dyes such as RhB provide strong visible-light emission [25] [26], the direct incorporation of dye molecules into CQD systems to form stable and reproducible composites without fluorescence quenching remains insufficiently explored. In particular, systematic investigations on how the concentration of RhB influences the photophysical properties, quantum yield, and emission tunability of hydrophobic CQDs are still limited.

In this work, these challenges are addressed by synthesizing hydrophobic carbon quantum dots (hbCQDs) and fabricating hbCQDs/RhB hybrid composites with controlled dye loading. The study aims to elucidate the structural and optical interactions between CQDs and RhB, evaluate the contribution of molecular-state emission to surface-state fluorescence, and identify optimized compositions that enhance quantum yield and broaden visible-light emission. These findings provide valuable insights for designing CQD-based hybrid luminescent materials and advance their potential applications in white-light organic optoelectronic devices and hydrophobic media-compatible photonic systems.

2. Experimental Section

2.1. Materials

Melamine (MA) ($\text{C}_3\text{N}_3(\text{NH}_2)_3$), dithiosalicylic acid ($\text{C}_{14}\text{H}_{10}\text{O}_4\text{S}_2$), and Rhodamine B (RhB) ($\text{C}_{28}\text{H}_{31}\text{ClN}_2\text{O}_3$) were procured from Shanghai Adamas Reagent Co., Ltd. Acetic acid (CH_3COOH) was supplied by Guangdong Guanghua Sci-Tech Co., Ltd.. All reagents were of analytical grade and used as received without further purification. Deionized water was produced using a Milli-Q water purification system (Millipore) and was employed consistently throughout all experimental procedures.

2.2. Synthesis of Hydrophobic Powder CQDs

The hydrophobic carbon quantum dots (hbCQDs) were synthesized via a solvothermal route [27] [28] following the procedure schematically illustrated in Figure 1. Initially, melamine (MA, 0.00079 mol) and thiosalicylic acid (0.0064 mol) were dissolved in 20 mL of acetic acid under ultrasonic irradiation until a homogeneous and transparent solution was obtained, ensuring complete precursor dispersion and molecular-level mixing. Subsequently, the resulting solution was transferred into a 50 mL Teflon-lined autoclave reactor and heated in an oven at 180 °C for 10 h to promote carbonization and nucleation of CQDs under autogenous pressure. After completion of the reaction and natural cooling to room temperature, the as-prepared CQD solution was rapidly injected into 1 L of boiling deionized water (100 °C). This step induced flocculation of the hbCQDs, facilitating the removal of unreacted precursors and residual solvent through phase separation. Aggregated CQDs were thereby formed and isolated. The precipitated CQDs were collected by vacuum filtration and thoroughly washed. Finally, the purified CQD powder was dried in an oven at 70 °C until constant weight was achieved, confirming the effectiveness and reproducibility of the water-washing purification protocol.



Figure 1. Schematic diagram for the synthesis of carbon quantum dots by Solvothermal method.

2.3. Synthesis of Powder CQDs/Rhodamine B Hybrid Composite

To broaden the emission profile of the CQDs toward the low-energy (red) region—an area of considerable interest due to its limited availability and typically low production yield—a CQDs/RhB hybrid system was developed. The hybrid materials were synthesized following the same solvothermal protocol used for the preparation of hydrophobic CQDs, with the incorporation of Rhodamine B (RhB) as a luminescent modifier. Specifically, controlled volumes of RhB (0.2 and 0.4 mL) were introduced into the precursor solution prior to the solvothermal treatment. Two hybrid samples were thus fabricated with different RhB loadings to systematically investigate the influence of dye concentration on the optical properties and emission tunability of the resulting CQDs/RhB nanocomposites.

3. Instrumentation

X-ray diffraction (XRD) analysis was conducted using a Rigaku Ultima IV diffractometer. High-resolution transmission electron microscopy (HR-TEM) characterization was performed on a JEOL ARM-200F operating at 200 kV and equipped with dual Cs aberration correctors. Micrographs were captured using an Orius camera, and subsequent image analysis and data processing were carried out with Gatan software. Fourier-transform infrared (FTIR) spectra were acquired using a PerkinElmer FTIR spectrometer. Samples were mounted on a thin-film holder with the film surface oriented toward the incident IR beam, and spectra were recorded across the 400–4000 cm^{-1} wavenumber range. X-ray photoelectron spectroscopy (XPS) measurements were performed using an XPS system, and spectral deconvolution and analysis were completed with CasaXPS. For XPS characterization, CQD dispersions were drop-cast onto 10 mm × 10 mm indium tin oxide (ITO) substrates and oven-dried prior to measurement. Optical absorption spectra were obtained using a Jasco V-770 UV–Vis–NIR spectrophotometer. Photoluminescence (PL) measurements were conducted with a Jasco fluorescence spectrometer. The quantum yield (QY) of the CQD samples was determined using a 100 mm integrating sphere coupled to a Jasco ILF-835 system.

4. Results and Discussions

4.1. Structural Characterization

4.1.1. X-Ray Diffraction Analysis (XRD)

The XRD patterns of pure hbCQDs and hbCQDs/RhB composites are presented in Figure 2. The diffraction profile of pristine hbCQDs exhibits a broad peak centered at approximately 22.65°, corresponding to an interlayer spacing (d-spacing) of 0.39 nm, which is indexed to the (002) plane of

graphitic carbon. This broadened feature indicates a low degree of crystallinity and the presence of turbostratic or amorphous carbon domains. Additionally, a weak diffraction peak is observed around 41° , with a calculated interlayer spacing of 0.21 nm, attributed to the (100) in-plane lattice reflection. These structural features collectively confirm the formation of carbon quantum dots with predominantly disordered graphitic structures. For the hbCQDs/Rhodamine B composites, the characteristic (002) and (100) diffraction peaks are retained at low RhB loading, indicating that the fundamental carbon framework remains largely unaffected. However, at higher RhB content, the (100) reflection becomes indistinguishable, suggesting increased structural disorder or partial surface coverage by RhB molecules, which may disrupt the in-plane graphitic ordering of the CQDs.

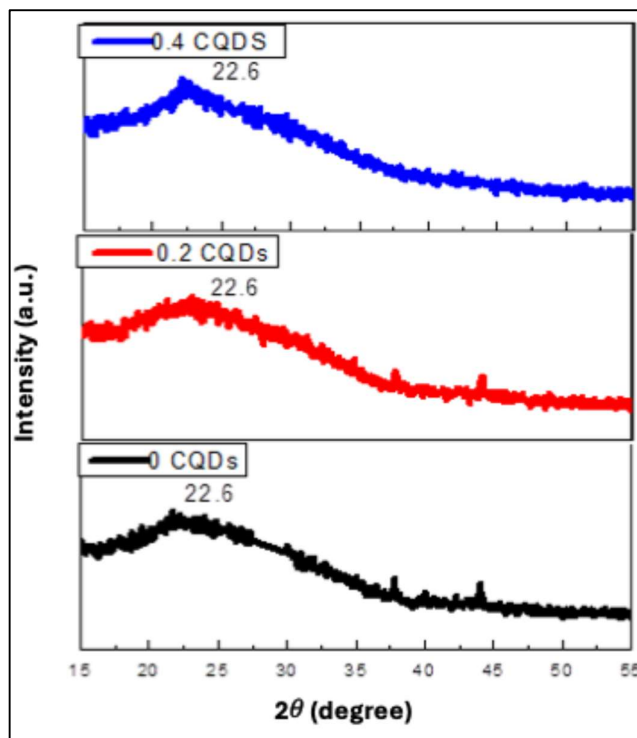


Figure 2. X-ray diffraction (XRD) patterns of hydrophobic carbon quantum dots (hbCQDs) and hbCQDs/Rhodamine B composite solid powders prepared with varying RhB loadings.

4.1.2. Transmission Electron Microscopy Analysis (TEM)

Transmission electron microscopy (TEM) was employed to examine the morphology, dispersion behavior, and particle size distribution of the carbon quantum dots. The low-magnification TEM image (scale bar: 2 μm) in Figure 3(a) illustrates the aggregation behavior of hbCQDs in aqueous media. The particles exhibit a pronounced tendency to form irregular, randomly shaped clusters, reflecting their hydrophobic surface characteristics and limited colloidal stability in water. High-magnification TEM images (Figure 3(b)) reveal that the hbCQDs are quasi-spherical and partially crystalline in nature. Lattice fringes observed in certain regions indicate localized graphitic ordering within the carbon core. Statistical analysis of particle diameters, derived from multiple TEM micrographs, shows a size distribution ranging from 2 to 20 nm, with an average particle size of approximately 8 nm, as summarized in the histogram presented in Figure 3(c).

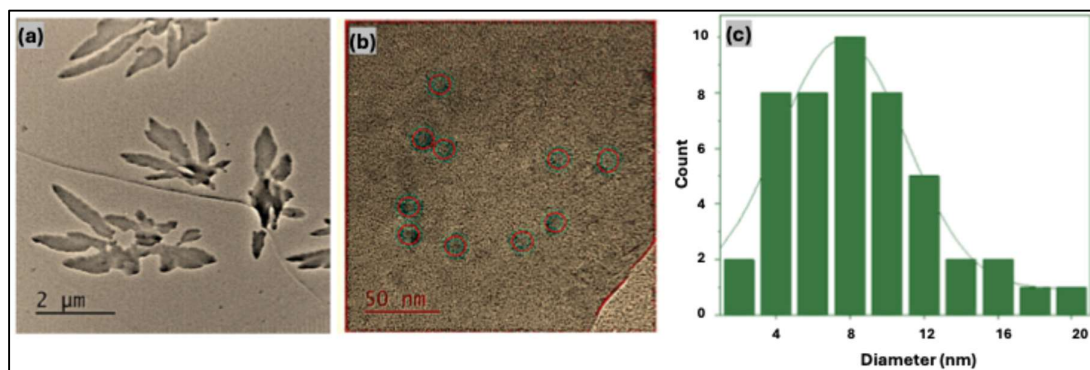


Figure 3. TEM images of hbCQDs: (a) low-magnification image (scale bar: 2 μm) showing particle aggregation behavior; (b) high-magnification image (scale bar: 50 nm) illustrating quasi-spherical morphology and structural features; and (c) particle size distribution histogram obtained from TEM analysis using ImageJ software.

4.1.3. Fourier-Transform Infrared Analysis (FTIR)

Fourier-transform infrared (FTIR) spectroscopy was conducted to identify the surface functional groups of the hydrophobic CQDs. The FTIR spectra of pristine hbCQDs and hbCQDs/RhB composites are presented in Figure 4. The spectrum of pure hbCQDs exhibits relatively weak absorption features, indicating a surface with limited oxygen-containing functionalities. The broad region between 4000 and 2000 cm^{-1} shows minimal prominent peaks, confirming that the structure is predominantly carbonaceous with low surface polarity. This observation is consistent with the hydrophobic nature of the synthesized CQDs [29]. Upon incorporation of a low amount (0.2 mL) of Rhodamine B (RhB), new low-intensity functional group signals emerge. A noticeable O–H stretching vibration appears around 2852 cm^{-1} , indicating the introduction of hydrophilic groups associated with RhB molecules. With further increase of RhB content to 0.4 mL, the O–H stretching band becomes significantly more intense, suggesting enhanced surface functionalization. Consequently, the surface character transitions from hydrophobic to hydrophilic, enabling improved water dispersibility of the composite [30] [31]. All samples display characteristic absorption bands corresponding to C≡N stretching (2034 cm^{-1}), S–H stretching (2650 cm^{-1}), and amide carbonyl (C=O) stretching at 1682 cm^{-1} . Additional peaks are observed at 1469 cm^{-1} (C=C), 1415 cm^{-1} (C–N), 1264 cm^{-1} (aromatic C–NH), 1155 cm^{-1} (C–O), and 690 cm^{-1} (C–S). A weak band near 491 cm^{-1} is attributed to S–S vibrations, likely arising from aggregated RhB molecular structures. Moreover, the asymmetric stretching mode of –N=C=S at 2150 cm^{-1} confirms the presence of isothiocyanate groups derived from RhB [32]. For the composite with higher RhB loading, the spectrum clearly exhibits the characteristic absorption features of RhB in the 1600–1000 cm^{-1} region, reflecting the increased contribution of dye-related functional groups to the overall chemical structure.

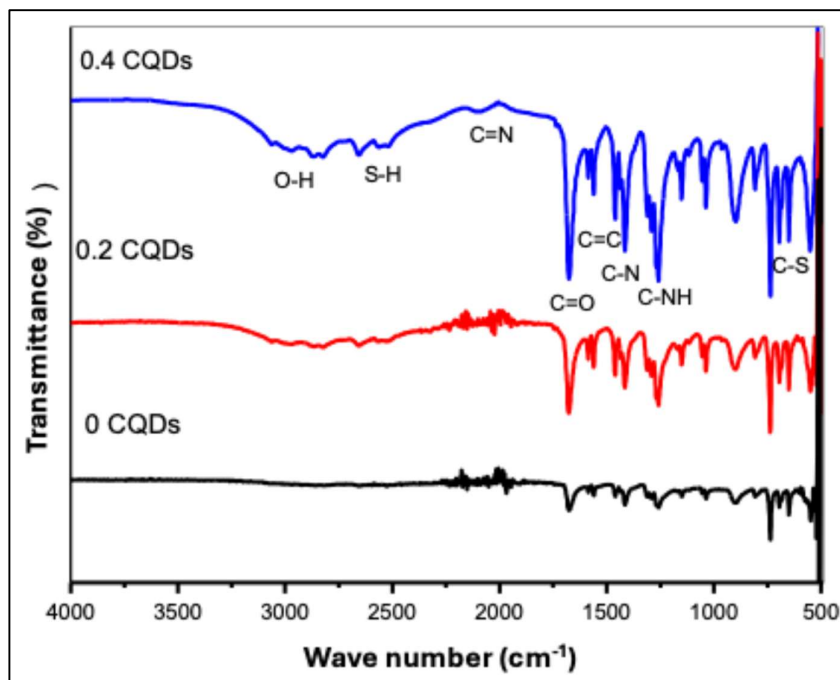


Figure 4. FTIR spectra of hbCQDs and hbCQDs/Rhodamine B composites: pure hbCQDs (black), 0.2 mL RhB-hbCQDs (red), and 0.4 mL RhB-hbCQDs (blue), highlighting the evolution of surface functional groups with increasing RhB content.

4.1.4. X-ray Photoelectron Spectroscopy Analysis (XPS)

X-ray photoelectron spectroscopy (XPS) was employed to investigate the elemental composition and chemical bonding of the hbCQDs, as illustrated in Figure 5. The survey spectrum reveals four prominent peaks at binding energies of 531 eV [33], 400 eV [33], 285 eV, and 164 eV [35], corresponding to the O 1s, N 1s, C 1s, and S 2p core levels, respectively. These results confirm the presence of oxygen, nitrogen, carbon, and sulfur in the hbCQDs and provide insight into the surface chemical environment of the synthesized nanomaterials.

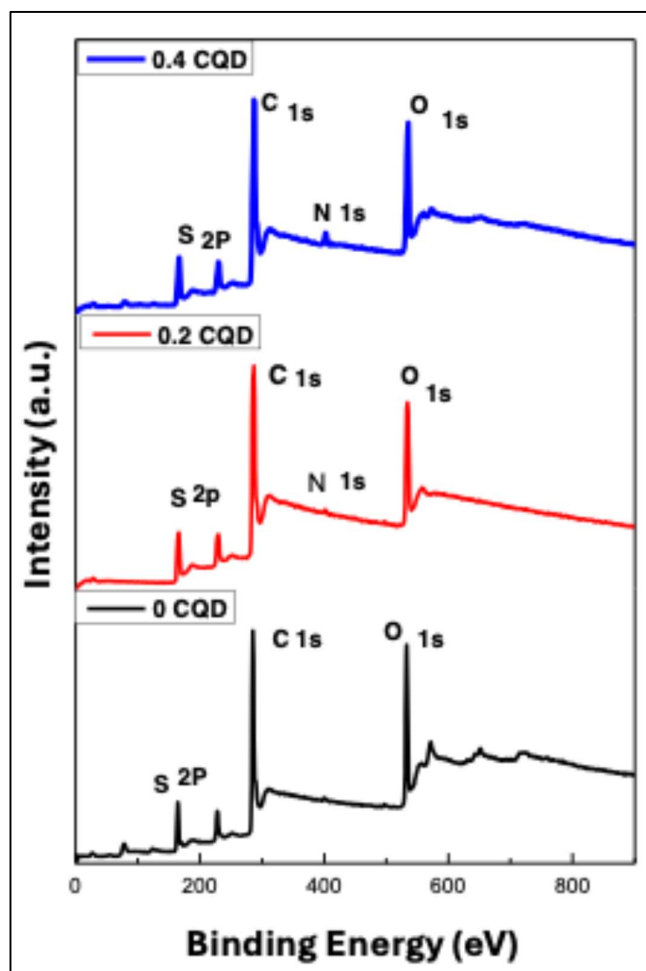


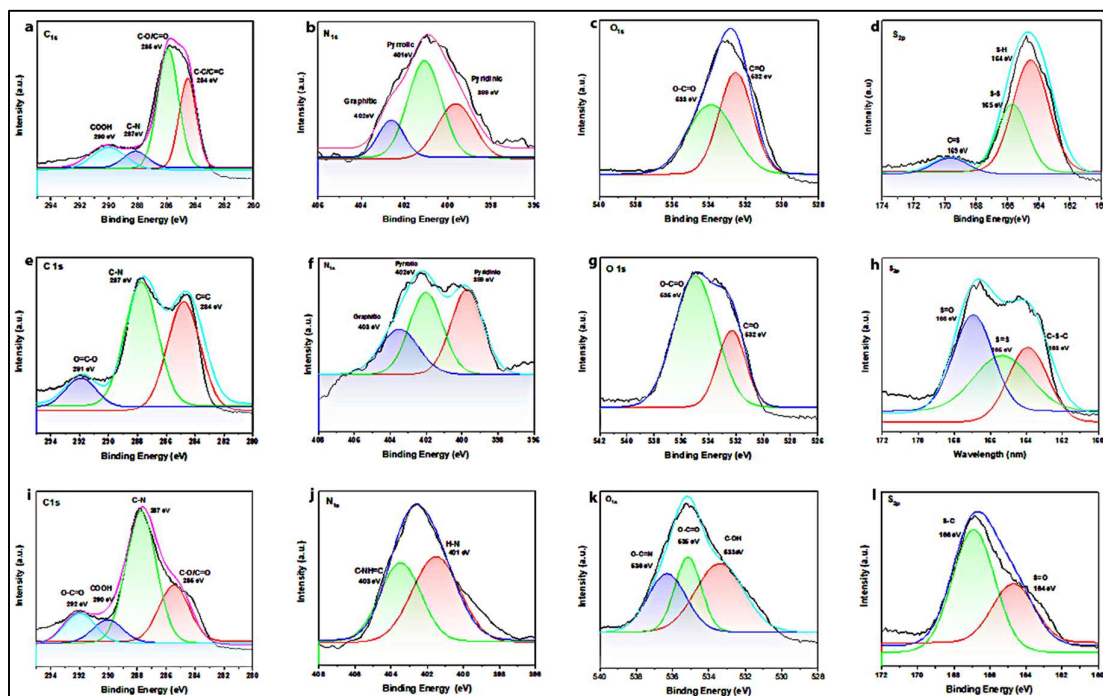
Figure 5. XPS survey spectra of hbcQDs and hbcQDs/Rhodamine B composites: pure hbcQDs (black), 0.2 mL RhB-hbcQDs (red), and 0.4 mL RhB-hbcQDs (blue), illustrating the elemental composition and chemical bonding variations with increasing RhB content.

The high-resolution XPS spectra of pure hbcQDs were deconvoluted to analyze the chemical states of the constituent elements, as presented in Figure 6. Figure 6(a) shows the C 1s spectrum of pure hbcQDs, revealing characteristic peaks corresponding to C=C (284.6 eV), C=O/C-O (285.9 eV), and C-N (287.5 eV) bonds. The hbcQDs/RhB composites exhibit similar C 1s profiles; however, a slight positive shift in the C-N binding energy is observed with increasing RhB concentration, suggesting the formation of additional amide linkages within the hybrid structure (Figure 6(i)).

The O 1s spectra, depicted in Figure 6(b), indicate the presence of C=O groups at 532.2 eV in samples with lower RhB content. At higher RhB loading, a new peak emerges at 533.0 eV, corresponding to C-O/C-OH functionalities, reflecting enhanced surface oxidation and functionalization. Figure 6(c) presents the N 1s spectra, showing contributions from pyrrolic nitrogen (399.6 eV) and pyridinic nitrogen (398.17 eV). With increasing RhB content, the pyridinic nitrogen peak becomes more prominent, indicating the expansion of conjugated aromatic domains within the carbon framework (Figure 6(j)).

Finally, the S 2p spectra, shown in Figure 6(d), confirm the presence of multiple sulfur species, including thiol S-H (164.5 eV), thiophene C-S-C (163.3 eV), disulfide S=S (165.1 eV), and sulfoxide S=O (169.0 eV). With higher RhB concentrations, partial oxidation of S-OH groups occurs, leading to the formation of SO_x species, which reflects the chemical interaction between CQDs and the RhB molecules. This comprehensive XPS analysis demonstrates that RhB incorporation modifies the

surface chemistry of hbCQDs, promoting functional group formation and enhanced aromaticity within the composites.



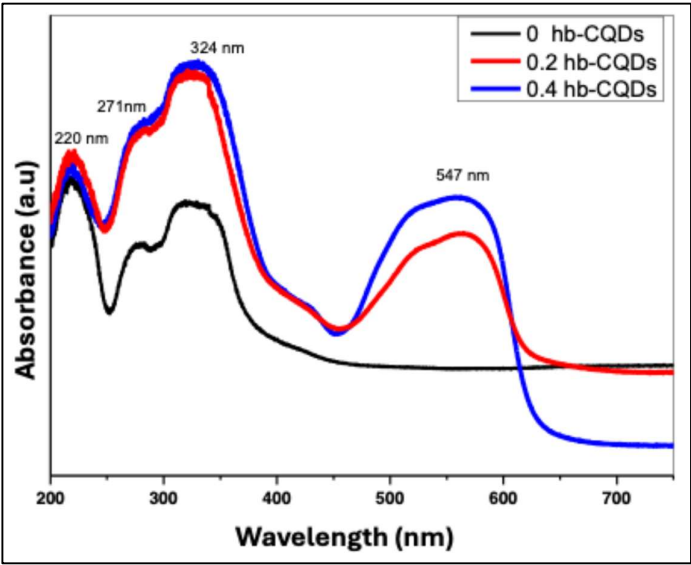


Figure 7. UV–Vis absorption spectra of hbCQDs and hbCQDs/RhB composites: pure hbCQDs (black), 0.2 mL RhB–hbCQDs (red), and 0.4 mL RhB–hbCQDs (blue), highlighting the influence of increasing RhB content on the optical absorption characteristics.

4.2.2. Photoluminescence (PL) Spectroscopy Analysis

4.2.1.1. Pure Hydrophobic CQDs Without RhB

Figure 8 illustrates the photoluminescence (PL) behavior of pure hbCQDs (0% RhB) under varying excitation wavelengths. Across this excitation range, the CQDs exhibit a characteristic blue emission, with the maximum emission peak centered at approximately 453 nm. The inset displays photographs of the powdered sample under daylight and upon 365 nm UV illumination, clearly demonstrating a vivid blue emission, which confirms the primary luminescent behavior of the CQDs. This emission originates predominantly from surface functional group states rather than the carbon core, with a measured QY of 6.2%.

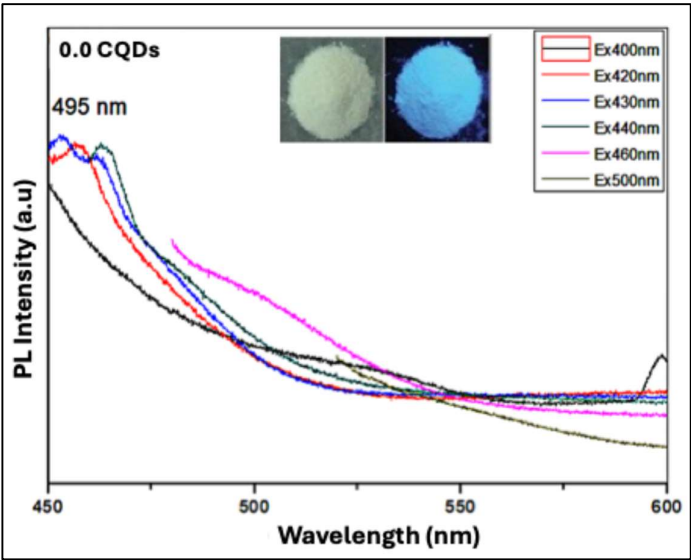


Figure 8. Photoluminescence (PL) spectra of pure hbCQDs (0% RhB). The inset shows comparative photographs of the sample under natural daylight and 365 nm UV illumination, highlighting the characteristic blue emission of the CQDs.

4.2.1.2. Hydrophobic CQDs Composite with RhB of 0.2 mL

Figure 9 presents the PL spectrum of the hydrophobic CQDs/RhB composite prepared with 0.2 mL RhB. The spectrum exhibits two distinct emission peaks: a higher-energy peak originating from carbon surface states, and a lower-energy peak attributed to molecular states derived from organic fluorophores used as precursors in CQD synthesis. These fluorophores may be covalently bonded to the carbon core or coupled to edge carbon atoms or functional groups, giving rise to $n-\pi^*$ and $\pi-\pi^*$ transitions. The surface-state emission shows excitation-dependent behavior, with maximum excitation/emission at 458 nm (blue), whereas the molecular-state emission is excitation-independent, with a peak at 605 nm (orange). The inset of Figure 9 shows a photograph of the 0.2 mL RhB–CQD composite powder. Under ambient light, the powder appears bright pink (left), while under 365 nm UV illumination, it emits a strong white light (right), resulting from the combination of the blue and orange emissions from the composite. This composite demonstrates optimal properties, with a high QY of 13% and a production yield of 82%, making it a promising candidate for future white-light organic optoelectronic applications.

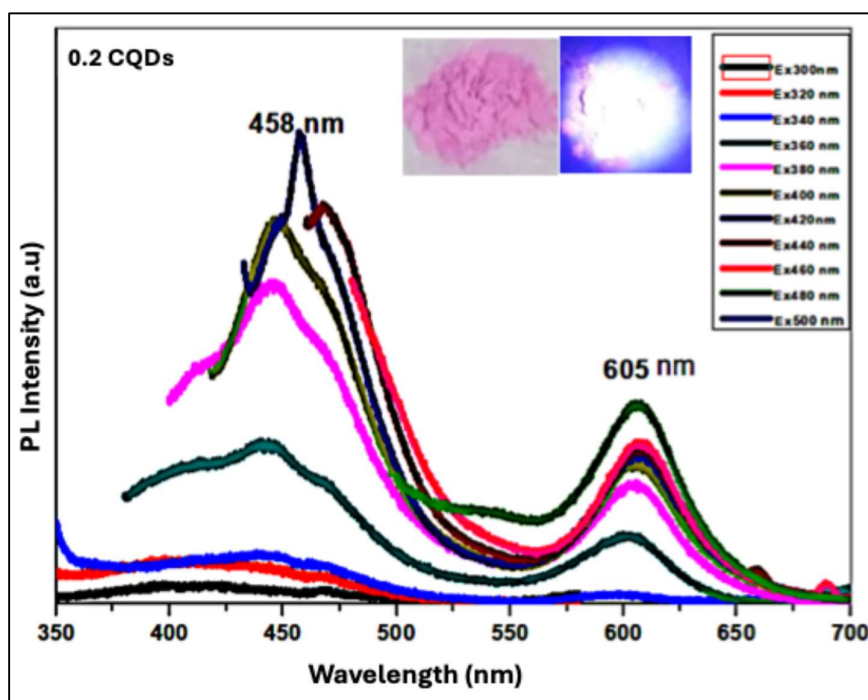


Figure 9. PL spectra of hbCQDs/RhB composite with 0.2 mL RhB. The inset shows comparative photographs of the powder under natural daylight and 365 nm UV illumination, highlighting the color change from bright pink to white emission.

4.2.1.3. Hydrophobic CQDs Composite with RhB of 0.4 mL

Figure 10 presents the PL spectra of the hydrophobic hbCQDs/RhB composite prepared with 0.4 mL RhB. The spectrum displays two emission peaks: a higher-energy peak arising from carbon surface states and a lower-energy peak associated with molecular states from the incorporated RhB. Compared to the 0.2 mL RhB–hbCQDs sample, the positions of both peaks show slight shifts, reflecting the higher RhB loading. The QY of this sample is reduced to below 9%, indicating decreased emission efficiency at higher dye content. The inset of Figure 10 shows a photograph of the composite powder. Under ambient light (left), the powder appears purple, while under 365 nm UV illumination (right), it emits a weak orange fluorescence. This observation demonstrates that increasing RhB content alters the emission profile and diminishes the overall photoluminescence intensity.

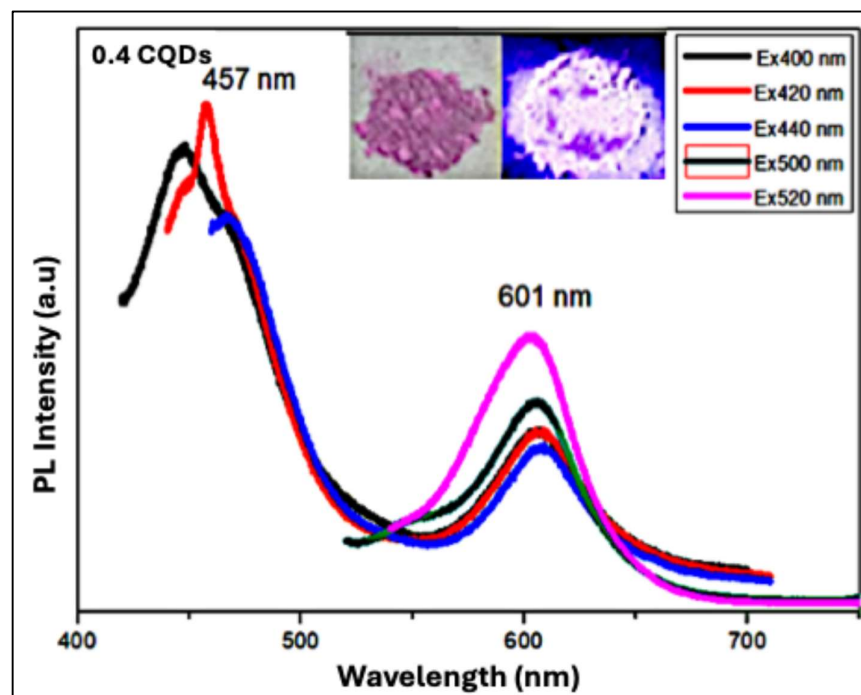


Figure 10. PL spectra of hbCQDs/RhB composite with 0.4 mL RhB. The inset shows comparative photographs of the powder under natural daylight and 365 nm UV illumination, highlighting the color change from purple under ambient light to weak orange emission under UV excitation.

5. Conclusion and Future Work

In this study, hydrophobic carbon quantum dots were successfully synthesized via a solvothermal method and subsequently hybridized with Rhodamine B (RhB) to achieve tunable photoluminescence. The pure hbCQDs exhibited blue emission at 453 nm with a quantum yield of 6.2%, while the incorporation of 0.2 mL RhB enabled dual-emission, combining surface-state blue emission and molecular-state orange emission, resulting in white-light emission with a QY of 13% and a production yield of 82%. Higher RhB loading (0.4 mL) caused slight peak shifts and reduced QY (<9%), demonstrating the critical influence of RhB concentration on optical performance. Structural and chemical analyses (TEM, XRD, FTIR, XPS) confirmed the quasi-spherical morphology, partially crystalline carbon core, and the presence of functional groups (C–N, C=O/C–O, S–H) that mediate photoluminescence. Overall, these results indicate that controlled RhB hybridization offers a simple and reproducible strategy to extend the emission of hbCQDs into the visible spectrum, making them suitable candidates for optoelectronic applications, including white-organic-light-emitting devices and hydrophobic media-compatible photonic systems.

Future investigations should focus on optimizing the emission balance between surface-state and molecular-state contributions to maximize white-light efficiency. The stability of hbCQDs/RhB composites under prolonged UV exposure, varying temperatures, and different solvent environments should be systematically evaluated. Furthermore, exploring other organic dyes or co-dopants could expand the emission range and enhance quantum yield. Integration of these composites into device architectures, such as LEDs or flexible optoelectronic systems, will be essential to translate the material's photophysical properties into practical applications. Additionally, understanding the precise mechanism of energy transfer between CQDs and organic fluorophores at the molecular level would provide insights for designing next-generation hybrid luminescent nanomaterials.

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Data Availability Statement: The authors confirm that the data supporting the findings of this study are available and can be accessed within the article.

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Conflicts of Interest: The authors declare no conflict of interest.

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