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Comparative Study of metal–doped carbon dots and Chitosan for effective dye removal from water

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Abstract

This study presents a comparative analysis of metal–doped carbon dots (CDs) synthesized from banana (CDs–B) and mandarin peels (CDs–M), alongside their chitosan–based composites, for the catalytic reduction of organic dyes Remazol Red (RR) and Methyl Orange (MO) in aqueous solutions. The CDs were produced via a hydrothermal method from fruit waste that naturally contains metal ions like K, Ca, Mg, Fe, and Zn, which enhance catalytic performance by improving electron transfer and structural stability. Characterization was performed using X–ray diffraction (XRD), UV–Vis spectroscopy, fluorescence emission, and Fourier–transform infrared (FTIR) spectroscopy. Kinetic studies, based on pseudo–first–order models, revealed that CDs–B outperformed CDs–M, exhibiting higher rate constants for RR (0.19954 min^{-1} vs. 0.13429 min^{-1}) and MO (0.14934 min^{-1} vs. 0.04201 min^{-1}). The incorporation of chitosan significantly enhanced the catalytic efficiency, with the chitosan/CDs–B composite achieving the highest rate constant of 0.34376 min^{-1} for RR reduction and showing the fastest reduction times (3 minutes for RR and 15 minutes for MO). These results suggest that banana peel–derived CDs, particularly when combined with chitosan, form a promising and sustainable nanocatalyst system for efficient dye remediation and wastewater treatment.

Key Words:

Carbon dots, Chitosan, Dye degradation, Water purification, green synthesis

1.Introduction:

The discharge of synthetic dyes into aquatic environments represents a serious ecological threat due to their non–biodegradable nature, high toxicity, and persistence. These dyes are widely employed in industries such as textiles, paper, and

pharmaceuticals. Their complex aromatic structures hinder natural degradation, contributing to long-term water pollution(Yang et al., 2025). Once present in water bodies, they obstruct light penetration, lower dissolved oxygen levels, and pose carcinogenic and mutagenic risks to humans(Crini & Badot, 2008).

Conventional dye treatment methods, including coagulation, filtration, and advanced oxidation, often face limitations such as high costs, low efficiency, and the potential generation of secondary pollutants(Vakili et al., 2014). These limitations have prompted researchers to investigate more sustainable alternatives, particularly those involving nanomaterials and biopolymers. Carbon dots (CDs), a novel class of carbon-based nanomaterials, have attracted attention for their potential in environmental remediation. Typically smaller than 10 nm, CDs exhibit tunable optical properties, high biocompatibility, and rich surface functionalities (John et al., 2024).Originally discovered as byproducts of carbon nanotube synthesis, CDs can now be purposefully synthesized from a range of carbon-rich sources, including organic waste, using hydrothermal treatment, microwave-assisted reactions, pyrolysis, and electrochemical exfoliation(Arul & Sethuraman, 2019; George et al., 2024; Pang et al., 2024).

Several studies validate the catalytic potential of CDs in pollutant degradation. For example, (Yang et al., 2025)synthesized metal-free CDs from tartaric acid and L-tryptophan with an average particle size of approximately 6 nm, achieving 70.26% removal of Cr(VI) at 150 μg/mL under visible light. Similarly, (Zourou et al., 2024)developed a magnetic CD-based hybrid that effectively adsorbed organic dyes, demonstrating the broad applicability of CDs in water purification.

The catalytic performance of CDs can be enhanced through metal doping (e.g., Ag, Cu) or heteroatom incorporation (e.g., N, O), which improves both electron mobility and dye-binding affinity(Dhanush & Sethuraman, 2020; John et al., 2024).

Chitosan, a naturally derived polysaccharide from chitin, has also shown strong potential in wastewater treatment. Its molecular structure contains abundant amino and hydroxyl groups that enable interactions with pollutants through electrostatic attraction, hydrogen bonding, and chelation(Crini & Badot, 2008). Previous studies have reported removal rates of up to 93% for iron and 92% for lead from Nile River water samples(Benha University, 2020). When modified with iron oxide nanoparticles, chitosan composites achieved over 95% removal efficiency for Pb(II) and Cd(II)(El-Nemr et al., 2021).

Beyond its adsorption capacity, chitosan can function as a catalytic support when combined with metal nanoparticles like Cu, Fe, or Ag. These hybrid materials facilitate electron transfer from $NaBH_4$ to dye molecules, improving reaction kinetics(Elhady et al., 2024). Additionally, membranes containing optimized chitosan concentrations (e.g., 1.5%) have demonstrated favorable porosity and ion removal performance (Damascus University, 2024).

Chitosan-metal nanocomposites have been successfully employed in degrading synthetic dyes such as methyl orange, methylene blue, and 4-nitrophenol. For instance, copper-modified chitosan achieved over 90% reduction of methyl orange in under 20 minutes(Reviews, 2023), while iron oxide-embedded chitosan beads enabled complete degradation of 4-nitrophenol at ambient temperature. Furthermore, activated carbon–

chitosan composites outperformed activated carbon alone in removing methylene blue.

Both CD-based and chitosan-metal systems follow a catalytic reduction mechanism, where $NaBH_4$ acts as the electron donor facilitating dye degradation.

To evaluate the structural and optical properties of the synthesized CDs and their efficiency in dye reduction, analytical techniques such as X-ray diffraction (XRD), UV–Vis spectroscopy, Fourier-transform infrared spectroscopy (FTIR), and fluorescence spectroscopy will be utilized.

Although both CDs and chitosan-based materials are effective in wastewater treatment, their modes of action differ. CDs function primarily as redox catalysts facilitating rapid electron transfer, whereas chitosan operates mainly through adsorption, with catalytic properties emerging upon modification. Experimental data suggest that CDs can achieve over 90% dye reduction within 10–30 minutes, particularly under visible light in the presence of $NaBH_4$. Chitosan-metal composites, though slightly slower, often reach 80–90% efficiency within 20–60 minutes depending on the metal loading and dye type(Elhady et al., 2024; Reviews, 2023).

In this study, metal-doped carbon dots (CDs) were synthesized from banana peels (CDs-B) and mandarin peels (CDs-M) via a hydrothermal method. The resulting CDs were thoroughly characterized using X-ray diffraction (XRD), UV–Visible spectroscopy, fluorescence spectroscopy, and Fourier-transform infrared (FTIR) spectroscopy to determine their structural and optical properties. The primary objective of the work is to evaluate and compare the catalytic efficiency of these biomass-derived CDs, both individually and in combination with chitosan, for the reduction of organic dyes, specifically Methyl Orange (MO) and Remazol Red (RR), in aqueous environments. This approach aims to develop an environmentally friendly and effective nanocatalyst system for wastewater treatment applications..

2.Methods of Research and the tools used.

2.1 Materials:

Banana and mandarin peels were utilized as carbon sources for the synthesis of carbon dots, while shrimp shells served as the primary source of chitin for chitosan production. The study also employed organic dyes including methyl orange (MO), and Remazol Red (RB-133), and sodium borohydride ($NaBH_4$) as the reducing agent. Additional materials included sodium hydroxide (NaOH), glacial acetic acid, and hydrochloric acid (HCl).

2.2 Carbon quantum dot synthesis:

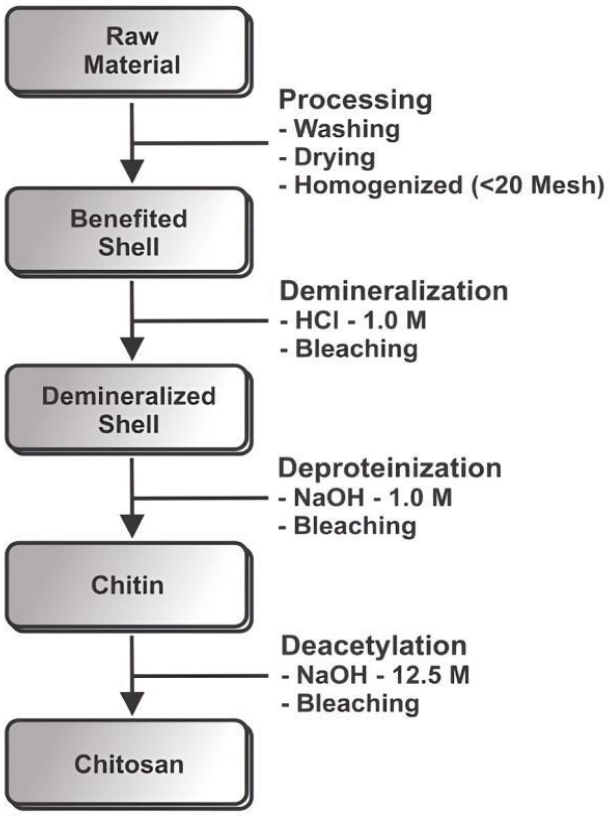
Carbon quantum dots (CDs) were synthesized via the hydrothermal method using banana and mandarin peels as carbon sources. following established protocols (Akhter & Aslam, 2025; Atchudan et al., 2021; Kasinathan et al., 2022; Sharma et al., 2025).As illustrated in Scheme **1**, the peels were washed, dried, and ground into powder, then mixed with deionized water and heated at 100 °C. The solution was transferred to a Teflon-lined autoclave and treated at 200 °C for 18 hours. Upon cooling to room temperature, a brownish solution was obtained, which was filtered and centrifuged to remove insoluble impurities and larger particles. The clear supernatant was further purified using a 1000 Da microporous membrane to eliminate larger aggregates. The obtained solution with intense yellow color was stored in a refrigerator. For subsequent characterization and application, the

solution was dried at 60 °C, yielding powdered carbon dots.

Scheme 1. A Schematic representation of the Synthesis of carbon dots from the banana peels by hydrothermal method.

2.3 Extraction of chitosan

As illustrated in Scheme **2**, chitosan was extracted from shrimp shells through a sequential process involving demineralization, deproteinization, and deacetylation. Initially, the cleaned and sun-dried shells were treated with 1.0 M hydrochloric acid (HCl) to remove minerals, followed by 0.5 N sodium hydroxide to eliminate proteins. The obtained chitin was thoroughly washed, dried, and converted into chitosan via alkaline reflux using concentrated NaOH. The final product was filtered, neutralized, and dried for further use. (Abdou et al., 2008; Liu et al., 2012; Sagheer et al., 2009).



Scheme 2. A schematic illustration showing the steps of chitosan extraction from shrimp shells.

2.4 Procedure for Catalytic Reduction of Dyes.

The catalytic efficiency of the synthesized carbon dots (CDs) and chitosan was evaluated by monitoring the reduction of organic dyes, specifically Methyl Orange (MO) and Remazol Red RB-133 (RR), in the presence of sodium borohydride ($NaBH_4$) as a reducing agent. In a typical procedure, 3 mL of a 5×10^{-5} M dye solution was mixed with 100 μL of CDs solution, followed by the rapid addition of 0.5 mL of freshly prepared 1% $NaBH_4$. The reduction process was monitored by recording UV–Visible absorption spectra at regular time intervals(Vinoth Kumar et al., 2022). A significant decrease in absorbance and

color intensity indicated successful dye degradation. Control experiments performed without the catalyst validated the role of CDs and chitosan in enhancing the reduction process. In the reduction experiments, UV–vis spectra at 520, and 460 for RR, and MO, respectively, were taken at different time intervals to track the evolution of dyes.

3. Results and discussion

3.1. Characterization of CDs

The structural and optical properties of the synthesized carbon dots (CDs) were analyzed using X-ray diffraction (XRD), Fourier-transform infrared (FTIR) spectroscopy, UV–Visible spectroscopy, and fluorescence spectroscopy.

The XRD patterns of CDs-B and CDs-M, shown in Fig.1, display broad diffraction peaks centered near 22° (2θ), which correspond to the (002) planes of graphite-like carbon. The broadness and lack of sharp crystalline peaks confirm the amorphous or non-crystalline nature of the CDs, which is characteristic of disordered carbon-based nanomaterials(Chen et al., 2025; Kasirajan et al., 2024a; Meng et al., 2025a)

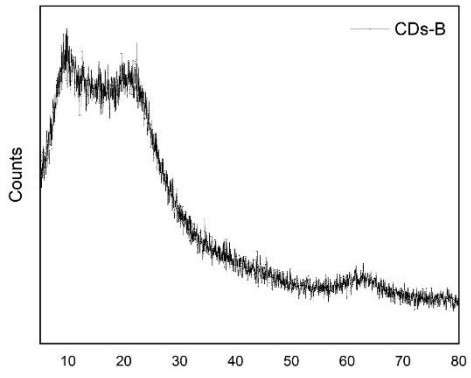


Fig 1. XRD pattern of the synthesized CDs from banana peels.

The functional groups and surface chemical structure of carbon quantum dots (CDs) synthesized from banana and mandarin peels were investigated using Fourier-transform infrared (FTIR) spectroscopy, as depicted in Figure 2.Both samples exhibit a broad absorption band around 3400 cm^{-1}, which corresponds to the O–H stretching vibration, indicating the presence of hydroxyl groups. However, this band is more intense in CDs-M, suggesting a greater number of –OH groups compared to CDs-B. In CDs-M, a strong peak appears near 1720 cm^{-1}, which is associated with C=O stretching, confirming the presence of carboxylic acid groups. On the other hand, CDs-B displays weaker signals in the 1600–1700 cm^{-1} range, which may reflect a smaller amount of carbonyl or carboxyl groups. A noticeable absorption band around 1400 cm^{-1}, linked to C–H bending, likely arises from aliphatic $–CH_2$ or $–CH_3$ groups and is clearer in CDs-M. Additionally, both types of dots show peaks between 1000 and 1250 cm^{-1}, corresponding to C–O stretching, typically related to ether or ester functionalities. These peaks are again more pronounced in CDs-M, suggesting a higher degree of surface oxidation or functionalization. (Atchudan et al., 2021; Kasirajan et al., 2024a; Meng et al., 2025a)

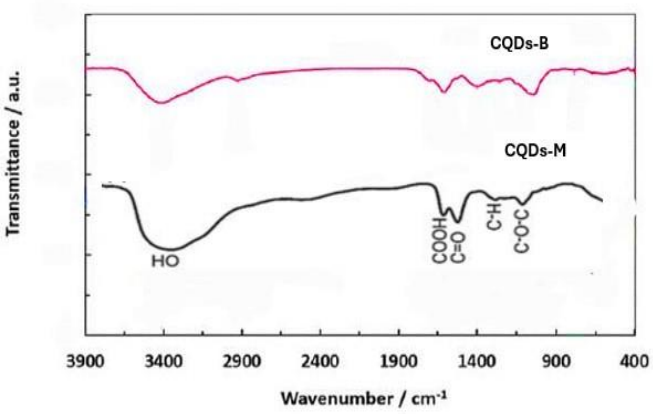


Fig.2 FT-IR Spectrum of CDs samples.

The UV–vis spectrum presented in Fig.3a exhibits an absorption peak near 272 nm, which corresponds to the π–π* electronic transition of the aromatic C=C bond. Additionally, a broad absorption band centered around 323 nm is attributed to the n–π* electronic transition of the C=O bond. The inset highlights that CDs have a yellowish appearance under daylight but emit bright blue fluorescence when exposed to UV light at 365 nm (Meng et al., 2025).To explore the optical characteristics of carbon dots (CDs), UV–Vis spectroscopy was employed, with the results illustrated in Fig.3b. A distinct absorption peak at 240 nm was identified, which corresponds to π→π* transitions within the C–C bonds of sp2 carbon domains. Meanwhile, a second prominent band appeared at 334 nm, associated with n→π* transitions arising from C–O bonds and also from C–N/C–N bonds. These findings provide deeper insights into the electronic transitions that influence CDs behavior (Kasirajan et al., 2024b).

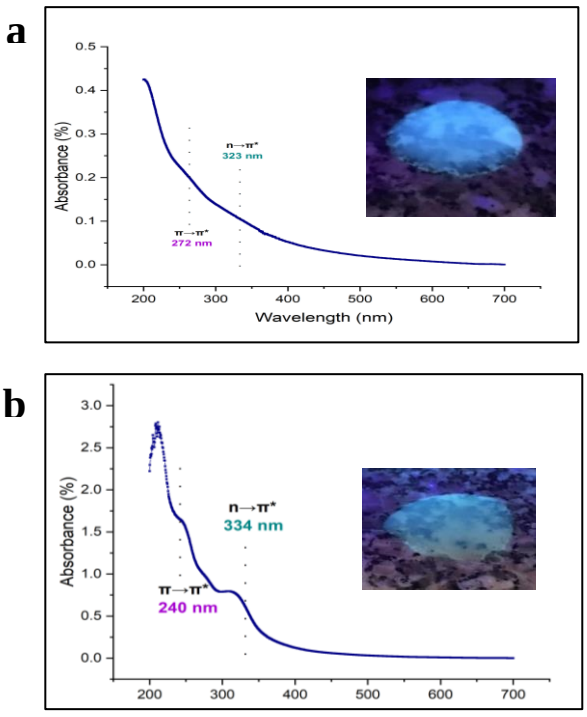


Fig.3 Absorption spectra of (a) CDs-B and (b) CDs-M

The fluorescence emission spectra (Fig. 4) exhibited an intense peak at around 500 nm for banana and mandarin peels upon the excitation of 379nm for banana peels, the excitation of 450 nm for mandarin peel, CDs commonly exhibit excitation dependent emission characteristics (Reddy et al., 2024)

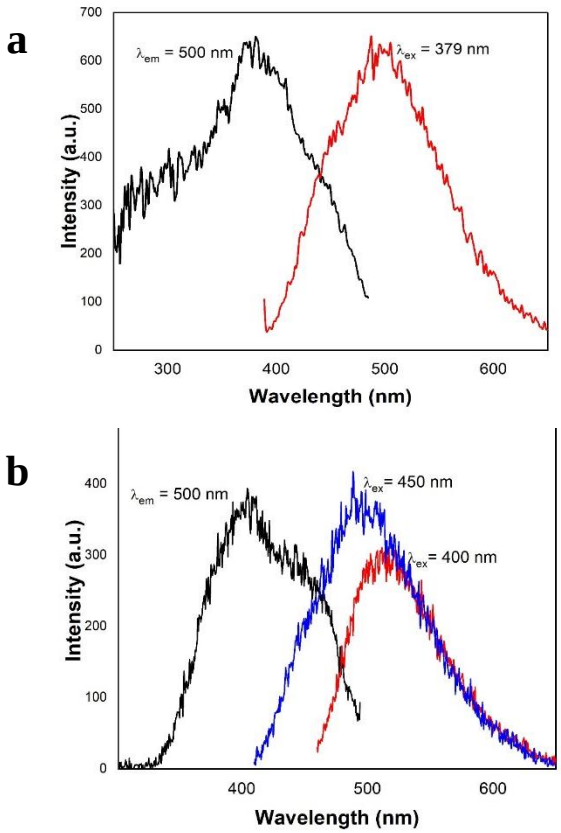


Fig.4 FL excitation and emission spectra of (a) CDs-B and (b) CDs-M

3.2. **Characterization of Chitosan**.

The XRD pattern of pure chitosan, illustrated in Figure **5a**, displays prominent peaks at 2θ angles of 10°, 20°, 23°, 25°, and 29°, which highlight its crystalline structure. The peak observed at 10° corresponds to the hydrated crystalline form, indicating the inclusion of water molecules within the crystal lattice—an aspect that influences both the solubility and flexibility of the material. In

contrast, the 20° peak is linked to the anhydrous crystalline form, reflecting the well-organized structure of chitosan chains without water content. This form enhances the material's rigidity and mechanical strength. Additional peaks at 23°, 25°, and 29° provide further evidence of chitosan's crystalline nature, pointing to varying degrees of crystallinity and diverse crystal orientations. The overall crystalline organization plays a significant role in shaping the physical characteristics of chitosan, including its mechanical durability, thermal resistance, and adsorption capability. (Elhady et al., 2024) Characterization of the prepared Chitosan. The FT–IR spectra of Chitosan was illustrated in Fig.**5b**. The spectra of Chitosan Showed the Characteristic absorption. Peak of OH and NH_2 at 3,361 and 3,291 cm^{-1}. The peaks at 2,921 and 2,877 cm^{-1} are attributed to C–H Stretching vibration. The characteristic absorption Peaks of NH, $–CH_3$ and the second hydroxyl absorption peak (OH) happen at 1,645 – 1,423 – 1,325 respectively. This suggests the formation of inter- and intra-molecular hydrogen bonds in the presence of free amino groups Chitosan. (Abdou et al., 2008; Liu et al., 2012; Sagheer et al., 2009).

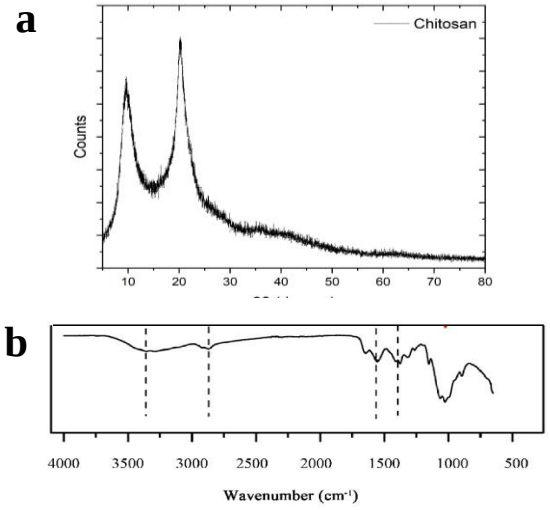
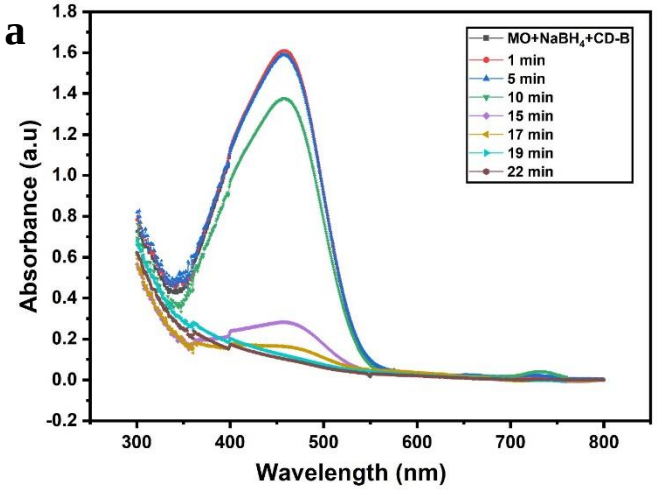


Fig.5 (a) XRD pattern and (b) FT-IR Spectrum of chitosan.

3.3. Catalytic efficiency of the CDs in the reduction of RR and MO dyes

The catalytic activity of metal-doped carbon dots (CDs) synthesized from banana peels (CDs-B) and mandarin peels (CDs-M)—both rich in natural metal ions such as K, Ca, Mg, Fe, and Zn—was tested for the reduction of organic dyes Remazol Red (RR) and Methyl Orange (MO) in the presence of sodium borohydride ($NaBH_4$). These inherent metal dopants, originating from the fruit biomass during hydrothermal method, enhance the electron transfer capabilities and structural stability of the CDs, contributing to their catalytic efficiency (Atchudan et al., 2021).Based on fig. 6 and 7 kinetic studies, assuming pseudo-first-order behavior with respect to dye concentration in systems containing only CDs revealed that CDs-B significantly outperformed CDs-M. Specifically, CDs-B exhibited rate constants of 0.19954 min^{-1} for RR and 0.14934 min^{-1} for MO, compared to 0.13429 min^{-1} and 0.04201 min^{-1} for CDs-M, respectively. This superior activity of CDs-B is attributed to higher density of surface functional groups and more efficient electron transfer mechanisms.



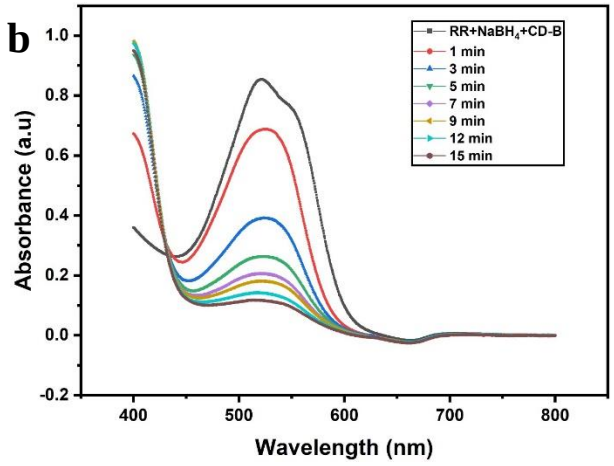


Fig. 6 UV-vis absorbance spectra of (a) MO and (b) RR aqueous solutions in the presence of the CDs-B and $NaBH_4$ at different degradation times

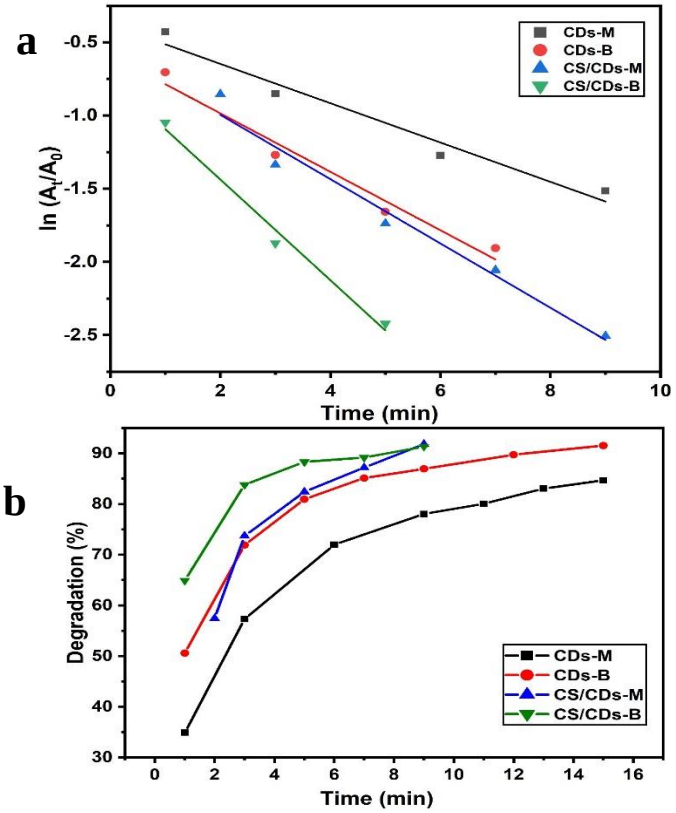


Fig. 7 (a) pseudo-first order kinetic plots, and (b) degradation efficiencies (%) of RR dye solutions in presence of CDs samples and chitosan.

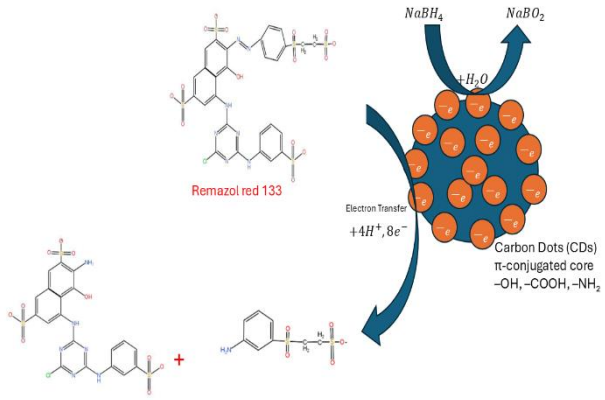
Incorporation of chitosan further improved catalytic activity. The chitosan/CDs-B composite yielded the highest rate constant of 0.34376 min^{-1} for RR reduction, outperforming both individual components. This synergistic enhancement is attributed to chitosan's functional groups (–OH and $–NH_2$), which promote dye adsorption and facilitate electron transfer. Similarly, the chitosan/CDs-M composite also showed improved kinetics, underscoring the catalytic advantage of the hybrid structure. Interestingly, systems containing chitosan deviated from simple pseudo-first-order kinetics, likely due to more complex surface interactions, enhanced adsorption mechanisms, and potential dual-site catalytic behavior—trends supported by prior studies on polymer-functionalized nanocatalysts (Barbosa et al., 2016; Deng et al., 2021).Overall, the results (summarized in **table 1**) indicate that banana peel-derived CDs exhibit greater catalytic potential than those from mandarin peels and, when coupled with chitosan, form a highly effective, sustainable nanocatalyst system suitable for wastewater treatment and dye remediation applications.

Table 1 : Comparative analysis of the catalytic reduction of methyl orange (MO) and Remazol Red (RR) using $NaBH_4$ in presence of CDs samples and Chitosan

Dyes	Catalyst	Reduction time (min)	Rate constant (k, min^{-1})
RR	CDs-B	5 min	0.19954
	CDs-M	11 min	0.13429
	Chitosan/CDs-B	3 min	0.34376
	Chitosan/CDs-M	5 min	0.21961
MO	CDs-B	15 min	0.14934
	CDs-M	30 min	0.04201

3.4. Proposed catalytic reduction mechanism for dye pollutants

In aqueous solutions, sodium borohydride ($NaBH_4$) undergoes hydrolysis, producing electrons that are crucial for initiating reduction reactions. These liberated electrons play a vital role in breaking down dye compounds (Vakili et al., 2014). Carbon dots (CDs) then act as effective mediators for electron transfer due to their conjugated carbon structure and the presence of surface functionalities such as hydroxyl, carboxyl, and amine groups. These structural features enhance redox behavior by facilitating efficient reception and transmission of electrons from $NaBH_4$. When further functionalized with electron-donating groups, the catalytic activity of CDs increases significantly (Panigrahi et al., 2023) Synthetic dyes such as methyl orange (MO) and Remazol Red (RR) become adsorbed onto the CD surfaces through multiple interactions, including electrostatic forces, hydrogen bonding, and π–π stacking, which help stabilize the dye molecules prior to reduction (Crini & Badot, 2008). Once adsorbed, the dye molecules receive electrons transferred from the CDs, leading to the cleavage of azo (–N=N–) linkages. This reduction process results in decolorization and the formation of less harmful aromatic amine derivatives (Dhanush & Sethuraman, 2020), Ultimately, the degradation yields colorless or low-toxicity aromatic compounds, significantly improving water quality and reducing toxicity (John et al., 2024)



Scheme 3: Mechanism of the catalytic reduction of Remazol Red (RR) and using CDs in the presence of sodium borohydride.

4. Conclusion

The present study highlights the superior catalytic performance of metal-doped carbon dots (CDs) synthesized from banana and mandarin peels, and their enhanced activity when combined with chitosan for the reduction of synthetic dyes Remazol Red (RR) and Methyl Orange (MO). Among the materials tested, CDs-B made from banana peels were more effective than CDs-M, with rates of 0.19954 min^{-1} for Remazol Red (RR) and 0.14934 min^{-1} for Methyl Orange (MO), indicating better surface reactivity and electron transfer efficiency.

Adding chitosan greatly increased the catalytic ability, especially in the chitosan/CDs-B composite, which achieved the highest observed rate constant of 0.34376 min^{-1} for RR and the shortest reduction time. This enhancement is attributed to the combined effects of the metal-rich surface of CDs and the adsorptive and functional properties of chitosan, enabling synergistic dye degradation.

Moreover, while CDs followed pseudo-first-order kinetics, the chitosan-based composites displayed

deviations suggestive of more complex adsorption and surface interaction mechanisms, aligning with prior studies on polymer-functionalized nanocatalysts. These findings highlight the promise of combining carbon materials made from biowaste with natural polymers to develop sustainable, efficient, and low-cost solutions for environmental remediation, particularly in wastewater treatment applications

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