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Carbon Nanodots Modulate Ciprofloxacin Adsorption onto Polylactic Acid Microplastics in Aquatic Environments: Behavior and Mechanistic Insights

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Abstract:

To investigate the influence of carbon dots (CDs) on the adsorption behavior of ciprofloxacin (CIP) by microplastics, willow leaves were used as a carbon source to synthesize CDs via a hydrothermal method, which were subsequently employed to modify polylactic acid (CDs-PLA). The materials were characterized by transmission electron microscopy (TEM), scanning electron microscopy (SEM), and Fourier-transform infrared spectroscopy (FTIR). Batch adsorption experiments were conducted to evaluate the adsorption performance and underlying mechanisms of both CDs and CDs-PLA toward CIP. The adsorption process was analyzed using the pseudo-second-order kinetic model, as well as the Langmuir and Freundlich isotherm models. The results demonstrated that the synthesized CDs exhibited a uniform spherical morphology with an average particle size of 5.95 ± 0.07 nm, an ordered surface lattice, and abundant oxygen-containing functional groups including C=O, -OH, -COOH, and C-O-C. The adsorption of CIP onto both

PLA and CDs-PLA followed the pseudo-second-order kinetic model and was well described by the Langmuir and Freundlich isotherm models, displaying both monolayer and multilayer adsorption characteristics, with chemisorption as the predominant mechanism.

Keywords: carbon dots; hydrothermal method; polylactic acid; ciprofloxacin; adsorption

1 Introduction

Carbon dots (CDs) are an emerging class of carbon nanomaterials with particle sizes typically below 10 nm(Chen *et al.*, 2019; Zheng, Tao *et al.*, 2023). They are characterized by low cost, low toxicity, high aqueous solubility, photoluminescence, and excellent biocompatibility (Chen *et al.*, 2022). Their surfaces can be readily functionalized via surface modification, enabling their wide application in medical diagnostics (Hasan *et al.*, 2020), photocatalysis (Yu *et al.*, 2024), drug detection (Rajendran *et al.*, 2020), fluorescent sensing (Hu *et al.*, 2021), and bioimaging (Bartkowski *et al.*, 2024). The surfaces of CDs are rich in hydrophilic groups, such as carboxyl, hydroxyl, and amino groups, which facilitate further modification (Sun *et al.*, 2019; Miao *et al.*, 2020) and provide considerable potential for polymer reinforcement. Biomass-derived CDs, which are prepared from biomass carbon sources, have attracted significant attention because of their low cost, abundant availability, and environmentally benign nature (Han *et al.*, 2020; Zhou *et al.*, 2020).

Microplastics (MPs) are plastic particles with a size of 5 mm or smaller. Owing to their low cost, ease of manufacture, durability, and broad applicability, plastics are widely used in industry, agriculture, and daily life (Deepak *et al.*, 2021). Because of their small size, diverse sources, and limited natural degradability, MPs have become widely distributed and accumulated in

environments such as oceans, lakes, rivers, farmland, and the atmosphere (Thompson *et al.*, 2004). Through bioaccumulation along the food chain, microplastics can induce various toxicological effects in organisms (Baumann *et al.*, 2016). Global microplastic pollution, therefore, represents an urgent environmental issue. Biodegradable polylactic acid (PLA) is increasingly replacing conventional plastics in many fields because of its biodegradability (Narancic *et al.*, 2017). However, PLA has been detected in various aquatic environments (Li *et al.*, 2024). Studies have shown that biodegradable plastics can exhibit greater adsorption capacities for organic pollutants than conventional plastics, thereby posing greater potential risks to organisms (Mejías *et al.*, 2023). Therefore, elucidating the adsorption mechanisms governing the interactions between PLA and organic pollutants is important for mitigating their combined pollution in the environment.

Ciprofloxacin (CIP) is a widely used second-generation fluoroquinolone antibiotic (Al-Buirahi *et al.*, 2022). Following administration, CIP is not completely absorbed and can be released into aquatic environments through excretion and other pathways, thereby contributing to water contamination (Shah *et al.*, 2022). Once released, it can accumulate in the natural environment and enter the human body through the food chain, leading to various adverse health effects (AI-Musawi *et al.*, 2021). Currently, methods such as adsorption, photocatalytic degradation, and biodegradation can be used to reduce CIP concentrations in the environment (Qi *et al.*, 2022). Among these methods, adsorption has received considerable attention because of its operational simplicity, low energy consumption, high adsorption capacity, and environmental compatibility.

To investigate the effect of carbon dots (CDs) on the adsorption of ciprofloxacin (CIP) onto polylactic acid (PLA), this study employed willow leaf-derived CDs to modify PLA and

comparatively examined the adsorption behavior and mechanisms of CIP before and after CD modification. Changes in the physicochemical properties of PLA induced by CD modification were also characterized. This work provides theoretical and practical insights into the prevention, control, and remediation of environmental microplastic pollution.

2 Materials and methods

2.1 Main materials

To investigate the effect of carbon dots (CDs) on the adsorption of CIP onto PLA, this study used willow-derived CDs to modify PLA and compared the adsorption behavior and mechanisms of CIP before and after CD modification. It also examined changes in the physicochemical properties of PLA before and after modification. This research provides theoretical and practical insights into the prevention, control, and remediation of environmental microplastic pollution.

2.2 Laboratory equipment

The main instruments used in this study included a freeze dryer (FD-1C-50, Shanghai Bilang Instrument Manufacturing Co., Ltd., China), a Fourier transform infrared (FTIR) spectrometer (Nicolet iS5, Thermo Fisher Scientific, USA), a scanning electron microscope (ZEISS Sigma 300, Carl Zeiss AG, Germany), a transmission electron microscope (JEM-2100F, JEOL Ltd., Japan), a UV–Vis spectrophotometer (T9, Beijing Purkinje General Instrument Co., Ltd., China), and an electronic balance (FA2004N, Shanghai Jinghai Instrument Co., Ltd., China).

2.3 Material preparation

2.3.1 Preparation of CDs

Willow leaves were washed, dried, ground into powder, and sieved through a 200-mesh sieve. Then, 4 g of willow leaf powder was weighed and dispersed in 50 mL of deionized water.

After thorough mixing, the suspension was transferred to a 100 mL PTFE-lined autoclave and heated at 180 °C for 10 h. After the reaction mixture had cooled to room temperature, it was first filtered and then purified through a 0.22 µm microporous membrane to remove impurities, and the dark-brown filtrate was collected. The resulting liquid product was transferred to a dialysis bag and dialyzed for 2-3 days until the solution in the beaker became nearly colorless. The dialyzed solution was freeze-dried for subsequent use. The preparation procedure for willow leaf-derived CDs is shown in **Fig. Error! Reference source not found.**

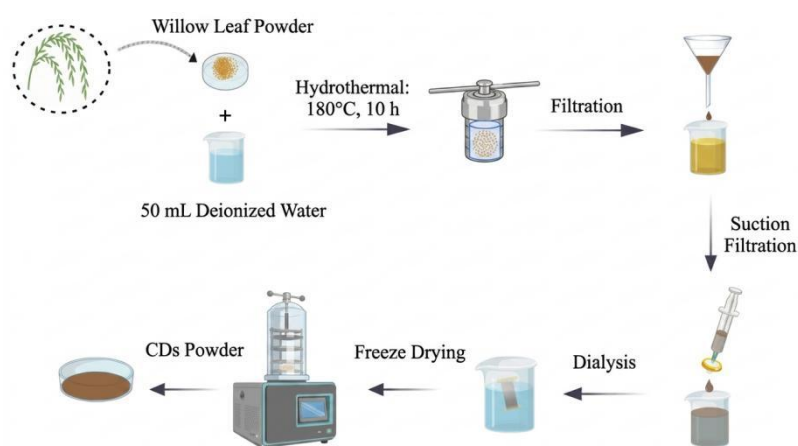


Fig.1 Flowchart for the preparation of willow leaf-derived carbon quantum dots.

2.3.2 Preparation of CD-Modified PLA

PLA particles passing through a 100-mesh sieve were immersed in a 0.5 mg mL⁻¹ CD ethanol solution for 17 h. The samples were then transferred to a Petri dish and dried at 40 °C for 5 h, yielding willow leaf-derived CD-modified polylactic acid (CDs-PLA).

2.4. Adsorption experiments

2.4.1 Kinetic experiments

PLA and CDs-PLA samples (0.0500 g each) were separately placed in 40 mL amber EPA vials. A CIP working solution with an initial concentration of 5 mg/L was added to completely fill each vial, after which the vials were sealed with PTFE-lined caps and placed in a rotary incubator

at 15 r/min. Samples were collected at 0, 1, 2, 4, 6, 9, 24, 48, 72, 96, and 120 h. After filtration, the CIP concentration in the filtrate was measured at 278 nm. Each experimental treatment was conducted in duplicate, and a blank control was included.

2.4.2 Adsorption isotherm experiment

PLA and CDs–PLA samples (0.0500 g each) were separately placed in 40 mL amber EPA vials. CIP working solutions with different initial concentrations (0.2, 0.4, 1.0, 2.5, and 6.0 mg L⁻¹) were added to the calibration mark. The vials were placed in a rotary shaker at 15 r min⁻¹ and shaken for 120 h. After filtration, the CIP concentration was determined at 278 nm using a UV spectrophotometer. Each experimental group was performed in duplicate, and a blank control was included.

2.5 Data analysis and adsorption models

2.5.1 Adsorption kinetic model

The pseudo-second-order kinetic model was used to fit the CIP adsorption process onto pristine PLA and CD-modified PLA, as expressed in **Eq.1**:

$$t/q_t = 1/kq_e^2 + t/q_e \quad (\text{Eq.1})$$

where q_e and q_t denote the equilibrium adsorption capacity and the adsorption capacity at time t (mg kg⁻¹), respectively, and k is the pseudo-second-order adsorption rate constant.

2.5.2 Adsorption isotherm models

The adsorption experimental data were fitted using the Langmuir and the Freundlich isotherm models, as shown in **Eq.2** and **Eq.3**, respectively:

$$q = \frac{q_m \cdot K_L \cdot C_w}{1 + K_L \cdot C_w} \quad (\text{Eq.2})$$

$$q = K_F \cdot C_w^n \quad (\text{Eq.3})$$

where q denotes the equilibrium adsorption capacity (mg kg^{-1}); C_w is the equilibrium aqueous concentration of CIP (mg L^{-1}); K_L is the Langmuir adsorption constant ($\text{mL } \mu\text{g}^{-1}$); K_F is the Freundlich adsorption constant ($\text{mg}^{1-n} \text{L}^n \text{kg}^{-1}$); n is a constant describing the adsorption intensity; and q_m is the theoretical maximum monolayer adsorption capacity ($\mu\text{g g}^{-1}$).

2.6 Data processing and visualization

The experimental data were processed using Microsoft Office 2021, and the graphs were generated using Origin 2021.

3 Results and discussion

3.1 Characterization of CDs and CDs-PLA

3.1.1 Characterization of CDs

The morphology of the prepared CDs was examined by transmission electron microscopy (TEM), as shown in **Fig.** Error! Reference source not found.. As observed in **Fig.** Error! Reference source not found., the prepared CDs exhibited a nearly spherical morphology and were well dispersed. The lattice fringe spacing of the CDs was 0.212 nm, which corresponds to the (100) plane of graphene (Parvathy *et al.*, 2022). Particle-size distribution analysis showed that the CDs had an average diameter of 5.95 ± 0.07 nm.

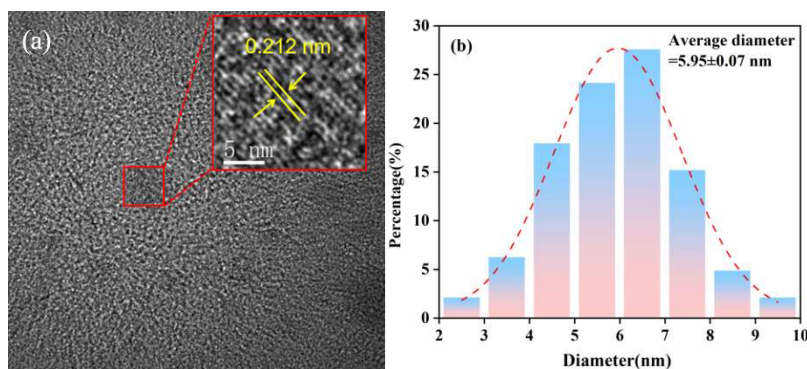


Fig.2 Morphological characterization of CDs. (a) TEM micrograph; (b) particle-size distribution.

As shown in **Fig.** Error! Reference source not found., the FTIR spectrum of the CDs showed a

broad absorption band centered at 3381 cm^{-1} , which was assigned to O-H stretching vibrations, possibly overlapping with N-H stretching vibrations (Asadi *et al.*, 2025). The weak band at 2936 cm^{-1} was attributed to aliphatic C-H stretching vibrations (Pansari *et al.*, 2025). The intense band at 1614 cm^{-1} was mainly assigned to the asymmetric stretching vibration of carboxylate (-COO-) groups, with a possible contribution from C=C stretching within the carbon core (Wang *et al.*, 2024), whereas the band at 1410 cm^{-1} corresponded to the symmetric stretching vibration of -COO- groups. In addition, the band at 1120 cm^{-1} was ascribed to C-O and C-O-C stretching vibrations (Tang *et al.*, 2023), whereas the low-wavenumber band at 612 cm^{-1} could be associated with skeletal vibrations or bending modes related to surface functional groups (Zhong *et al.*, 2025). These results indicate that the prepared CDs were rich in surface oxygen-containing functional groups.

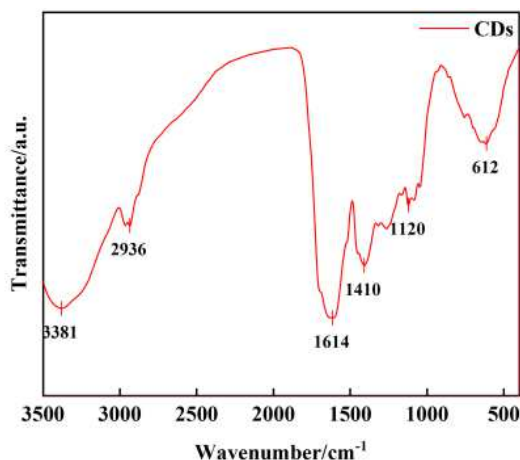


Fig.3 FTIR spectrum of CDs.

3.1.2 Characterization of CD-Modified PLA

(1) SEM analysis

Fig. Error! Reference source not found. shows SEM images of PLA and CDs-PLA. As shown in Figs. Error! Reference source not found. a and Error! Reference source not found. b, the PLA

surface was composed of fibrous structures with abundant pores and a relatively rough morphology. In contrast, **Figs.**Error! Reference source not found.**c** and Error! Reference source not found.**d** show that the granular CDs were well dispersed on the PLA surface. A large number of CDs adhered to the PLA surface and filled the surface voids, resulting in a smoother and more uniform surface morphology.

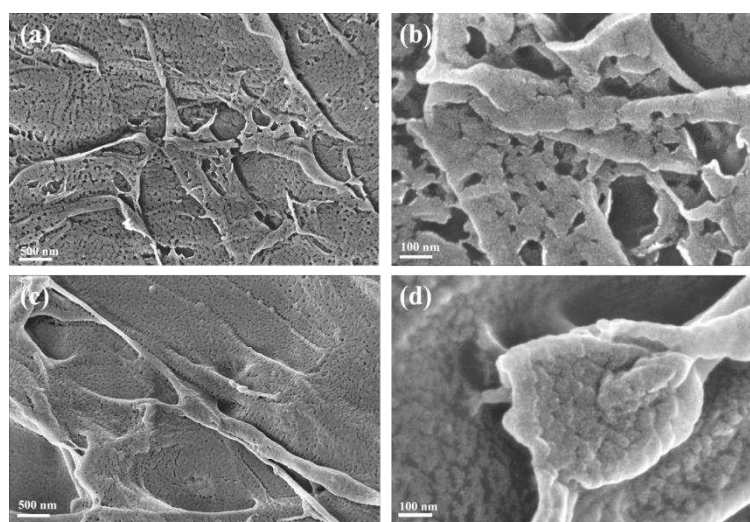


Fig.4 SEM images of PLA and CDs-PLA.(a,b) PLA; (c,d) CDs-PLA.

(2) FTIR

The FTIR spectra of PLA and CDs-PLA are shown in **Fig.**Error! Reference source not found.. As shown in **Fig.**Error! Reference source not found., the main characteristic peaks of PLA and CDs-PLA appeared at nearly the same positions, and their overall peak profiles showed only minor changes. Previous studies have shown that carbon-doped PLA composites exhibit FTIR spectra similar to those of neat PLA, with no formation of new functional groups but more distinct characteristic peaks ([Vorkapić et al., 2025](#)). The strong absorption peak of PLA at 1750 cm^{-1} was attributed to C=O stretching vibrations, whereas the characteristic peaks at 1181 and 1080 cm^{-1} were assigned to C-O-C stretching and asymmetric stretching vibrations, respectively.

Compared with PLA, CDs-PLA showed no obvious shifts in the main characteristic peaks,

including those of C=O and C-O-C, indicating that the introduction of CDs did not alter the fundamental chemical structure of PLA. Previous research has also shown that carbon incorporation does not significantly change the basic chemical structure of PLA (Pathek *et al.*, 2024).

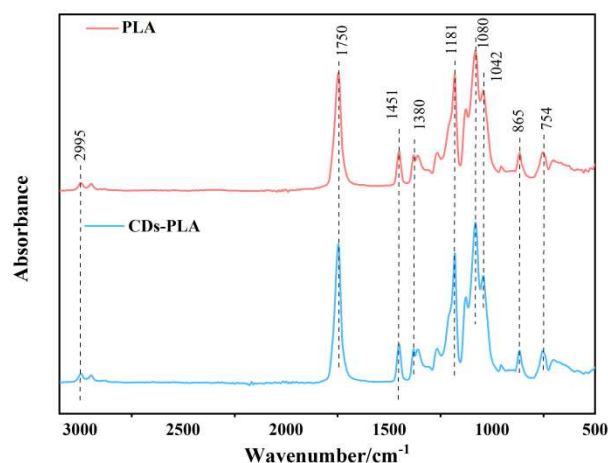


Fig.5 FTIR spectrum of PLA and CDs-PLA.

3.2 The effect of CDs on the CIP of PLA adsorption

3.2.1 Adsorption kinetics

Fig.Error! Reference source not found. shows the pseudo-second-order kinetic fit for the adsorption of CIP by PLA and CDs-PLA. As shown in **Fig.**Error! Reference source not found. and Error! Unknown switch argument., the pseudo-second-order kinetic model provides a good fit for the adsorption of CIP by PLA and CDs-PLA, with R^2 values of 0.999 and 0.998, respectively. Furthermore, the theoretical values ($q_{e,cal}$) and experimental values ($q_{e,exp}$) obtained from the pseudo-second-order kinetic model are very close. Therefore, the adsorption of CIP by PLA before and after CDs modification conforms to the pseudo-second-order kinetic model. The pseudo-second-order adsorption kinetics model assumes that the rate of the adsorption process is determined by the square of the number of unoccupied adsorption sites on the adsorbent surface. The adsorption rates of PLA and CDs-PLA are positively correlated with the square of the number

of unoccupied adsorption sites on the surface (Mezenner *et al.*, 2008). Therefore, the adsorption of CIP onto PLA before and after the CDs modification is a chemical adsorption process.

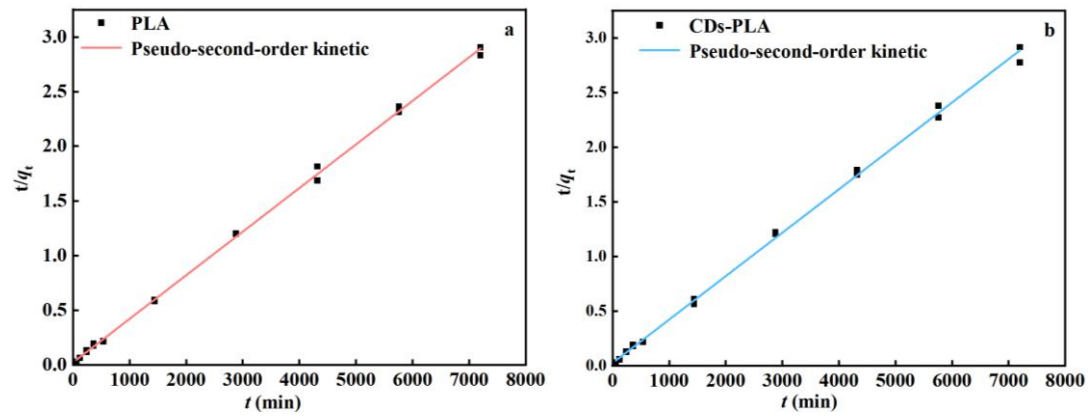


Fig.6 Quasi-second-order kinetics of CIP adsorption by different PLA materials (a) PLA; (b) CDs-PLA.

Table.1 Pseudo-second-order kinetic fitting parameters for CIP adsorption onto PLA and CDs-PLA.

Pseudo-second-order kinetic model			
MPs types	$q_e/(\text{mg/kg})$	$k_2/(\text{g} \cdot (\text{mg} \cdot \text{h}^{-1})^{-1})$	R^2
PLA	2488	5.11×10^{-6}	0.999
CDs-PLA	2492	4.22×10^{-6}	0.998

3.2.2 Adsorption isotherms

Fig.Error! Reference source not found. shows the adsorption isotherms of CIP onto PLA and CDs-PLA. As shown in the figure, the adsorption capacity of both PLA and CDs-PLA increased gradually with increasing initial CIP concentration, and CDs-PLA exhibited a markedly higher CIP adsorption capacity than PLA. The adsorption data for CIP onto both PLA and CDs-PLA were well fitted by the Langmuir and the Freundlich isotherm models (Error! Unknown switch argument. and **Fig.**Error! Reference source not found.), with R^2 values ranging from 0.983 to 0.997. These results suggest the coexistence of monolayer and multilayer adsorption, which is consistent with previous findings (Zheng, Wu *et al.*, 2023).

In the Langmuir isotherm model, both the maximum adsorption capacity (q_m) and the Langmuir constant (K_L) of CDs-PLA were higher than those of PLA. According to the Langmuir model, q_m represents the maximum adsorption capacity when the adsorbent surface reaches

monolayer saturation, whereas K_L is the adsorption equilibrium constant and is positively associated with adsorption affinity. These results indicate that CD modification significantly enhanced the CIP adsorption capacity of PLA, and that CDs-PLA exhibited a stronger affinity for CIP.

For the Freundlich isotherm model, the fitting parameter n for CIP adsorption onto PLA was less than 1, indicating nonlinear adsorption, whereas the n value for CIP adsorption onto CDs-PLA was close to 1, suggesting near-linear adsorption behavior. The Freundlich constant (K_F) of CDs-PLA was higher than that of PLA, further indicating the stronger adsorption affinity of CDs-PLA for CIP. Previous studies have shown that hydrophilic or oxygen-containing surface modifications can enhance the affinity of PLA-based materials toward hydrophilic contaminants such as CIP, thereby increasing their adsorption capacity.

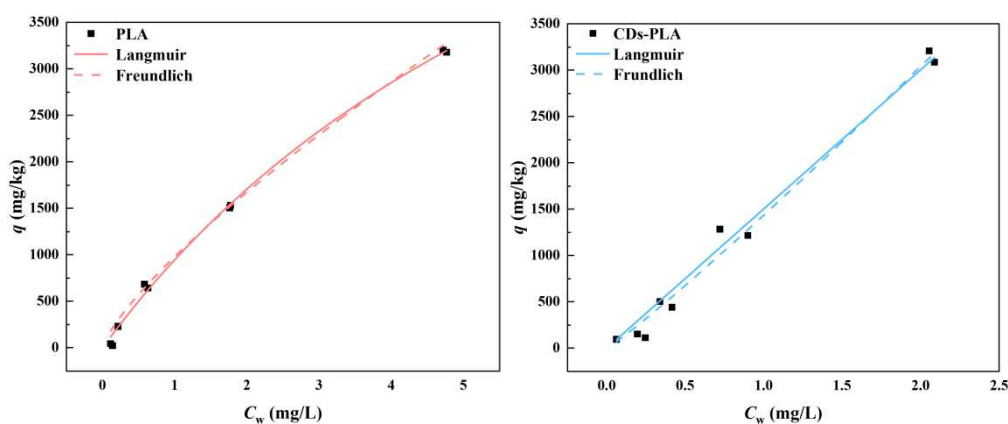


Fig.7 Langmuir and Freundlich isotherm fitting curves for CIP adsorption onto different PLA-based materials: (a)

PLA and (b) CDs–PLA.

Table.2 Langmuir and Freundlich isotherm fitting parameters for CIP adsorption onto PLA and CDs–PLA.

MPs types	Langmuir			Freundlich		
	$q_m/(\mu\text{g}\cdot\text{g}^{-1})$	$K_L/(\text{mL}\cdot\mu\text{g}^{-1})$	R^2	$K_F/(\text{mg}^{1-n}\cdot\text{L}^n\cdot\text{kg}^{-1})$	n	R^2
PLA	8672 ± 889	0.122 ± 0.0183	0.996	979 ± 1.956	0.773 ± 0.001	0.997
CDs-PLA	3131736 ± 4.119	4.799 ± 0.063	0.983	1434.593 ± 71.854	1.083 ± 0.071	0.985

3.3 Analysis of adsorption mechanisms

Fig. Error! Unknown switch argument. shows the FTIR spectra of PLA and CDs-PLA before and after CIP adsorption. No substantial changes in the spectral profiles of PLA or CDs-PLA were observed after CIP adsorption, and no new characteristic peaks appeared. The main characteristic peaks of PLA and CDs-PLA were associated with C=O stretching at 1750 cm⁻¹ and C-O-C vibrations at 1080 and 1181 cm⁻¹ (Sun *et al.*, 2023; Yu *et al.*, 2025). After CIP adsorption, the peak intensities increased, following the order CDs--PLA + CIP > CDs-PLA > PLA + CIP > PLA. This result suggests that hydrogen-bonding-mediated chemisorption may play an important role in CIP adsorption onto PLA and CDs-PLA. The increase in characteristic peak intensity after CIP adsorption was more pronounced for CDs-PLA than for PLA, indicating stronger hydrogen-bonding interactions between CDs-PLA and CIP, as well as a higher adsorption capacity. Previous studies have also shown that CIP adsorption onto PLA is strongly influenced by hydrogen bonding (Liang *et al.*, 2023).

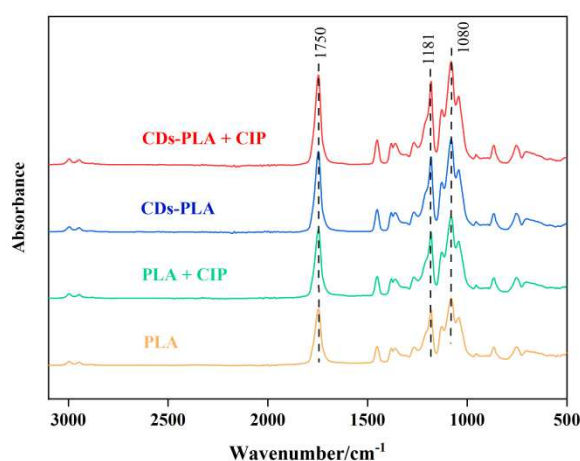


Fig.8 FTIR spectra of PLA and CDs–PLA before and after CIP adsorption.

4 Conclusions

In this study, carbon dots were synthesized using willow leaf powder as the precursor and were further used to prepare CD-modified polylactic acid (CDs-PLA). Adsorption experiments were conducted to evaluate the CIP adsorption performance of PLA and CDs-PLA, leading to the

following conclusions:

(1) Water-soluble carbon dots (CDs) were synthesized from willow leaf powder via a hydrothermal method. TEM and FTIR analyses showed that the CDs were spherical, with an average particle size of 5.95 ± 0.07 nm, and exhibited well-defined lattice fringes. The FTIR results further confirmed that the CDs contained abundant surface oxygen-containing functional groups.

(2) CDs were successfully introduced onto PLA, providing additional adsorption sites for CIP. Both PLA and CDs-PLA served as carriers for CIP, while CDs-PLA exhibited a higher CIP adsorption capacity than pristine PLA.

The adsorption of CIP onto PLA and CDs-PLA was well described by the pseudo-second-order kinetic model, as well as the Langmuir and the Freundlich isotherm models. These results indicate the coexistence of monolayer and multilayer adsorption, with the adsorption process primarily governed by chemisorption.

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Declarations

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277 Code availability: Not applicable.

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279 manuscript.

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