Treating Waste with Waste - Study on Visible-Light Photocatalytic Performance of Modified Banana Peel Biochar Loaded with TiO_2 and Its Reactor Integration Design

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Abstract

This study investigates the development of a low-cost composite material and a continuous-flow device for the removal of methylene blue from dye-contaminated wastewater. Industrial dye pollution poses significant environmental challenges, and agricultural waste materials provide a potential sustainable solution. In this work, banana peel powder was modified using different concentrations of citric acid to enhance its adsorption capacity, and titanium dioxide (TiO_2) was subsequently loaded onto the optimally modified substrate to achieve synergistic adsorption-photocatalytic degradation under UV photocatalysis.

Materials were prepared via esterification, impregnation, and ultrasonic dispersion. Characterization using SEM, FTIR, and BET confirmed increased porosity, successful carboxyl functionalization, and stable Ti–O–C bonding. Adsorption experiments showed that banana peel modified with 1.5 mol/L citric acid achieved 97.74% removal efficiency. The optimal composite (CA:TiO_2 mass ratio of 5:1.5) exhibited near-complete removal (99.56%) under UV light.

For practical application, an Archimedean screw-based continuous-flow reactor was designed and tested. The horizontal prototype integrated UV irradiation and used cotton to retain the composite material and prolong dye contact. Long-term evaluation over 40 hours demonstrated up to 46.68% removal, confirming continuous operation feasibility. Performance differences between material and device tests are attributed to variations in material-to-solution ratios and UV intensity, indicating clear optimization pathways.

In conclusion, this work successfully demonstrates a "waste-to-resource" strategy, proving that a citric acid-modified banana peel-TiO_2 composite offers strong synergistic effects for dye removal. The integrated spiral reactor provides a viable and scalable framework for continuous wastewater treatment, with significant potential for improvement through enhanced reactor design to fully exploit the material's capabilities.

1. Introduction

Pollution always exists manufacturing. One of the most serious pollution is waterbody pollution. According to statistics from the United Nations, only about 27% of the industrial wastewater is properly treated. One major portion of the polluted water is dye contaminated. Therefore, this paper aims to bring out an effective yet economical way to deal with dye-contaminated wastewater. Fortunately, titanium(IV) dioxide (TiO2) is oftentimes considered an effective tool. With the photocatalytic property, there will be electrons in the conduction band and holes in the valence band as TiO2 is exposed under UV light. In the water, the holes promotes the ionization of water to form hydroxyl radicals. These radicals can react with dyes like methylene blue (C16H18N3SCl) and results in the drgradation of the dyes. This process can be demonstrated by the following sets of chemical equations:

$$TiO_2 \xrightarrow{UV} TiO_2\ (e^-{}_{CB} + h^+{}_{VB})$$

$$TiO_2\ (h^+{}_{VB}) + H_2O \rightarrow TiO_2 + H^+ + OH^-$$

$$TiO_2\ (h^+{}_{VB}) + OH^- \rightarrow TiO_2 + OH\cdot$$

$$C_{16}H_{18}N_3SCl + 102\ OH\cdot \rightarrow 16CO_2 + 3NO_3{}^- + SO_4{}^{2-} + Cl^- + 57H_2O + 6H^+$$

This mechanism can help degrade and remove organic dye pollutants in water. However, it is of relative low efficiency when TiO_2 itself is used to do the degredation, so a carrier is probably needed to exemplify the effect of TiO_2. [1] [2] Through broad research, dehydrated banana peels stand out, not only because of the extreme low costs and pollution, but because of its capability to be modefied to form more pores through esterification as well. [3]

2. Methodology

2.1. Materials

Citric acid (CA), sodium hydroxide (NaOH), and methylene blue (MB) were of analytical grade and purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Glassware, including beakers, funnels, and glass rods, was supplied by Tianjin Glass Instrument Factory. Titanium dioxide (TiO_2, anatase phase, purity ≥ 99.8%) was purchased from Aladdin Industrial Corporation (Shanghai, China).

2.2. Banana Peel Modification

Fresh banana peels were cut into small pieces with areas controlled in the range of 1–4 cm². The pieces were then washed three times with ethanol and three times with ultrapure water to remove surface impurities. The cleaned banana peels were dried in an oven at 180 °C for 24 h, pulverized, and sieved through a 100-mesh sieve to obtain raw banana peel powder (denoted as Raw).

The raw banana peel powder was separately immersed in citric acid solutions with concentrations of 0.5 mol/L, 1.0 mol/L, and 1.5 mol/L and heated in a water bath at 60 °C for 2 h. After modification, the samples were filtered, rinsed three times with ultrapure water, dried at 100 °C, and ground into powder. The resulting citric acid-modified banana peel powders with different concentrations were designated as **CA, CA0.5, CA1.0, and CA1.5,** respectively. The experimental procedure is shown in Fig. 1.

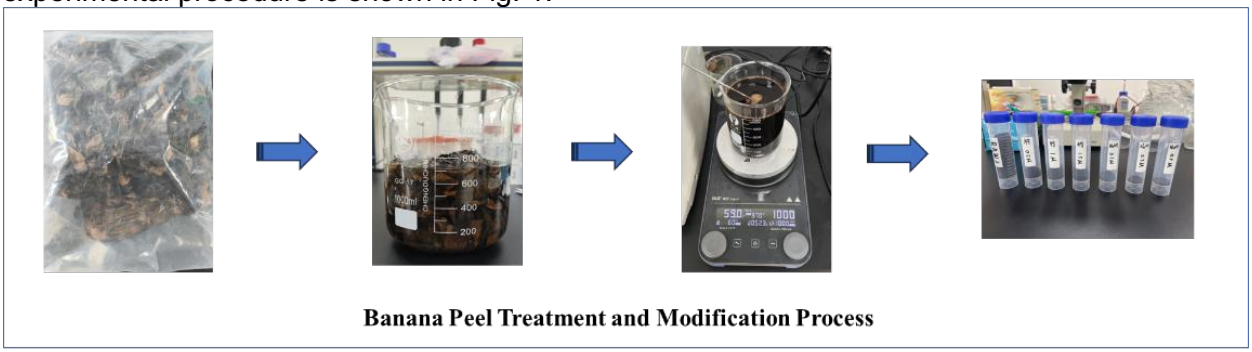

Figure 1 Schematic illustration of the banana peel modification procedure

2.3 Methylene Blue Adsorption Performance Test

The experimental procedure is illustrated in Fig. 2. A methylene blue solution with a concentration of **20 mg/L** was prepared as the target pollutant. Accurately weighed **0.5 g portions o**f the citric acid-modified banana peel powders (prepared using 0.5 mol/L, 1.0 mol/L, and 1.5 mol/L citric acid, respectively) were placed in beakers. Subsequently, **20 mL** of the

methylene blue solution was added to each beaker, and the mixtures were stirred at room temperature for 1.5 h at 800 rpm. After adsorption, the suspensions were filtered, and the absorbance of the filtrate was measured using a UV-Vis spectrophotometer at 665 nm. The removal efficiency was calculated according to the following equation to determine the optimal citric acid concentration for modification:

$$Removal\ efficiency\ (\%) = [(C_0 - C_e)/C_0] \times 100\% \quad (1)$$

where C_0(mg/L) is the initial concentration of methylene blue, and C_e(mg/L) is the equilibrium concentration of methylene blue after adsorption.

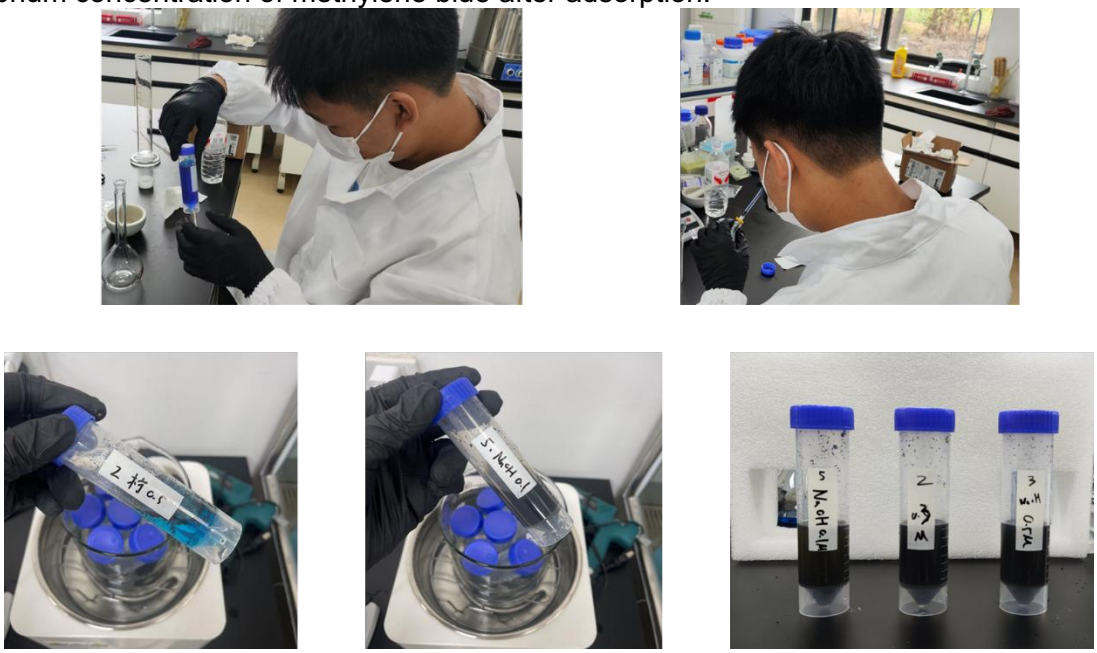

Figure 2 Methylene blue adsorption experiment

2.4 Preparation of Titanium Dioxide-Banana Peel (CA-TiO_2) Composites

The CA powder exhibiting the highest adsorption performance in section 2.3 was selected as the substrate, and TiO_2 was loaded onto it via the impregnation method. Exactly 5 g of the CA powder was weighed, and 0.5 g, 1.0 g, or 1.5 g of TiO_2 powder together with 50 mL of ethanol were added. The mixtures were ultrasonically dispersed for 1 h to ensure uniform mixing, then allowed to stand at 60 °C for 6 h. Subsequently, the samples were dried in an oven at 60 °C for 12 h and ground into fine powder. The resulting CA- TiO_2 composites with different TiO_2 loadings were designated as **CA-TiO_2-0.5, CA- TiO_2-1.0, and CA- TiO_2-1.5**, respectively.

2.5 Photocatalytic Adsorption-Degradation Performance Test for Methylene Blue

A methylene blue solution with a concentration of **80 mg/L** was prepared to evaluate the synergistic adsorption-photocatalytic degradation performance of pure CA powder and the CA-TiO_2 composites with varying TiO_2 loadings. Pure CA powder served as the control group, while the experimental groups consisted of CA- TiO_2 composites with CA-to- TiO_2 mass ratios of 5:0.5, 5:1, and 5:1.5. For each group, **0.5 g** of the material was added to **20 mL** of the simulated wastewater, and the mixture was reacted under UV irradiation for 1 h. The total removal efficiency of methylene blue (adsorption + photocatalytic degradation) was calculated by measuring the absorbance of the filtrate at 665 nm, using the method described in Equation (1). The experimental procedure is illustrated in Fig. 3.

Figure 3 Photocatalytic adsorption-degradation experiment for methylene blue

2.6 Material Characterization

Scanning electron microscopy (SEM) was used to examine the microscopic surface morphology of the unmodified banana peel, citric acid-modified CA powder, and CA-TiO_2 composites, allowing comparison of the effects of citric acid modification on banana peel surface morphology and evaluation of the TiO_2 particle loading.

Fourier transform infrared spectroscopy (FTIR) was performed in the wavenumber range of 4000–400 cm^{-1} to characterize the functional groups and their changes in the above three materials, thereby verifying the regulatory effect of citric acid modification on functional groups and the interactions between TiO_2 and CA.

The specific surface area, pore volume, and average pore diameter of the modified CA powder and CA- TiO_2 composites were determined using a Brunauer-Emmett-Teller (BET) analyzer through nitrogen adsorption-desorption isotherms, to investigate the influence of TiO_2 loading on the pore structure of the modified banana peel.

2.7 Engineering structural establishment

The next step of this work is to fabricate a water purification device using CA-TiO_2 composite material. Since this material requires UV light for photocatalysis, the conventional filter packing method cannot provide sufficient light irradiation for the material. In addition, the material is in powder form, so using a traditional filter may easily cause the powder to enter the water and result in secondary pollution. Therefore, it is necessary to redesign a reactor to achieve water purification with the CA-TiO_2 composite.

When considering liquid delivery, the Archimedes screw pump was first considered. It has the potential to transport liquid from a lower level to a higher level, especially when the liquid cannot flow naturally. Moreover, the screw is in full contact with the liquid. For this reason, the helical structure is adopted as the main body of the reactor. It is envisioned that a UV lamp can be placed in the center of the screw, so that the lamp can fully irradiate and catalyze the material. The design sketch is shown below.

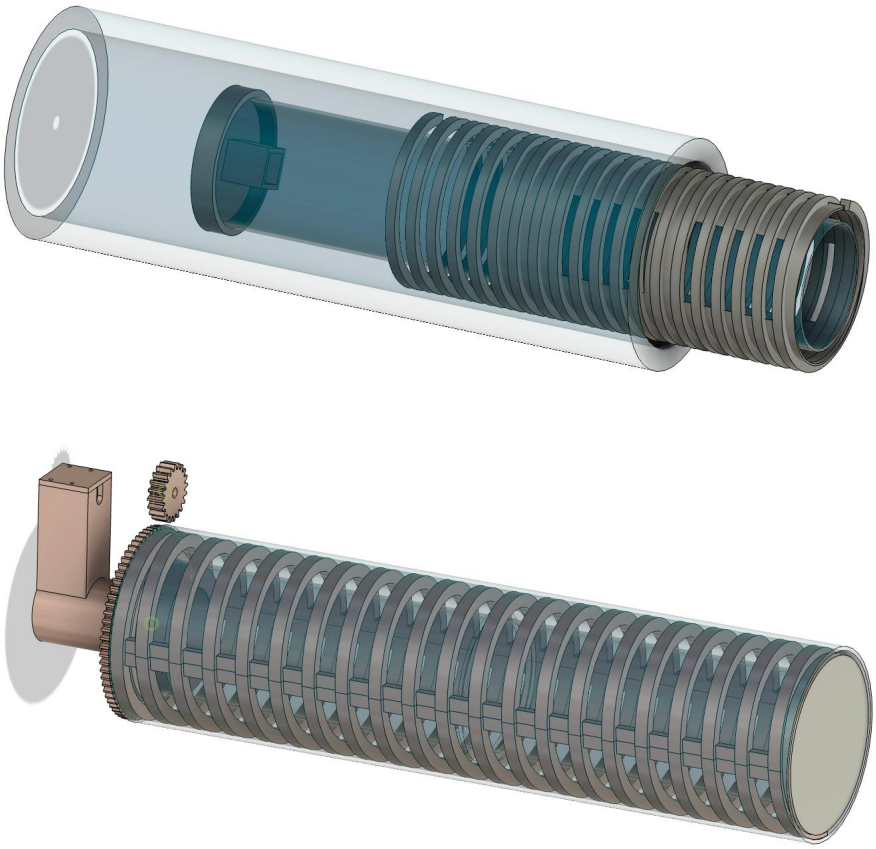

Figure 4 Archimedean screw reactor design blue-prints

During the design and fabrication of the new reactor, many problems were encountered. The most critical one was how to keep the UV lamp isolated from the liquid while preventing it from rotating with the screw. To solve this, a bearing structure was designed to support the lamp, ensuring that the UV lamp would not rotate with the entire device, which could otherwise cause wire twisting or even breakage, as shown on the left side of the figure below.

Another important issue was how to arrange the powder material inside the screw. Various fixing methods were proposed and tested, and the final selected scheme was to add additional clamping slots in the screw and fix the powder in the slots with filter cotton, as shown on the right side of the figure below. This design not only solves the problem of powder immobilization but also allows the filter cotton to further filter large-particle impurities in the water.

The screw was printed using a transparent material. When the device is operating, the entire screw acts as a light-emitting chamber, allowing UV light to fully contact the material and achieve efficient photocatalytic reaction.

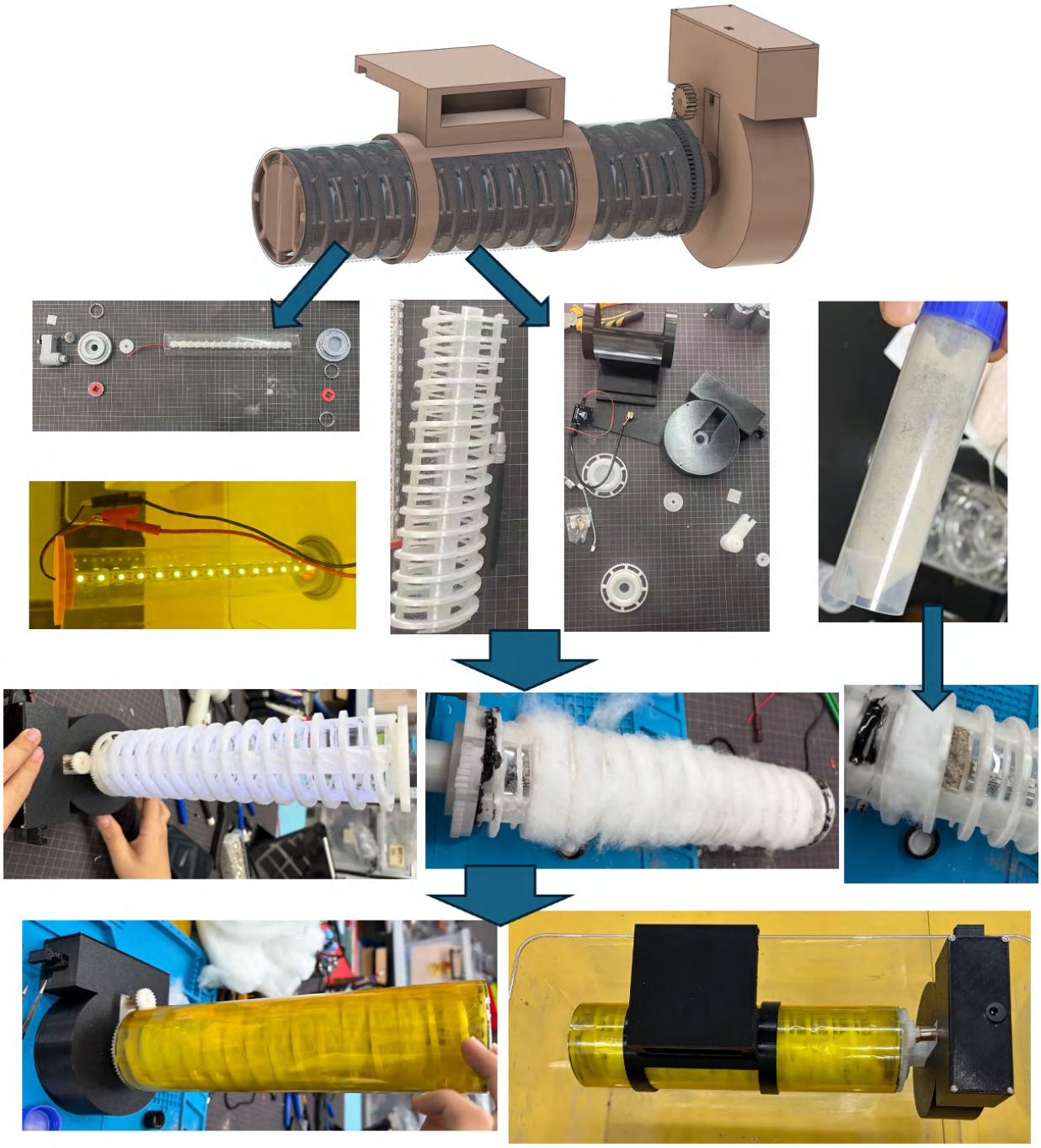

Figure 5 Archimedean screw reactor prototype building process

As shown in the final image above, this is the reactor prototype. In addition to the UV lamp chamber and the Archimedean screw structure mentioned earlier, the latest prototype is also equipped with a transmission mechanism and a suspension device, where all power supply components and the motor are installed. Extra waterproof treatment has been applied to protect these key components.

3. Results and Discussion

3.1 Methylene Blue Adsorption Efficiency

With controlled adsorption time, temperature, material dosage, solution volume, and concentration, the effect of citric acid modification concentration on the adsorption removal efficiency of methylene blue by the modified CA was investigated. The results (Fig. 4) revealed that as the citric acid concentration increased from 0.5 mol/L to 1.5 mol/L, the removal efficiency of methylene blue by CA gradually increased. The sample modified with 1.5 mol/L citric acid exhibited the highest removal efficiency of **97.74%**, which represented increases of 67.82, 45.42, and 25.78 percentage points compared to the unmodified sample (29.92%), the 0.5 mol/L modified sample (52.32%), and the 1.0 mol/L modified sample (71.96%), respectively. This enhancement can be attributed to the sufficient reaction between high-concentration (1.5 mol/L) citric acid and banana peel cellulose. On the one hand, esterification introduces a large number of carboxyl groups, whose electronegativity generates strong electrostatic attraction with cationic methylene blue, thereby strengthening adsorption. On the other hand, deep etching of the material surface widens existing pores and creates new hierarchical pore structures, providing ample diffusion channels and adsorption sites for methylene blue molecules.

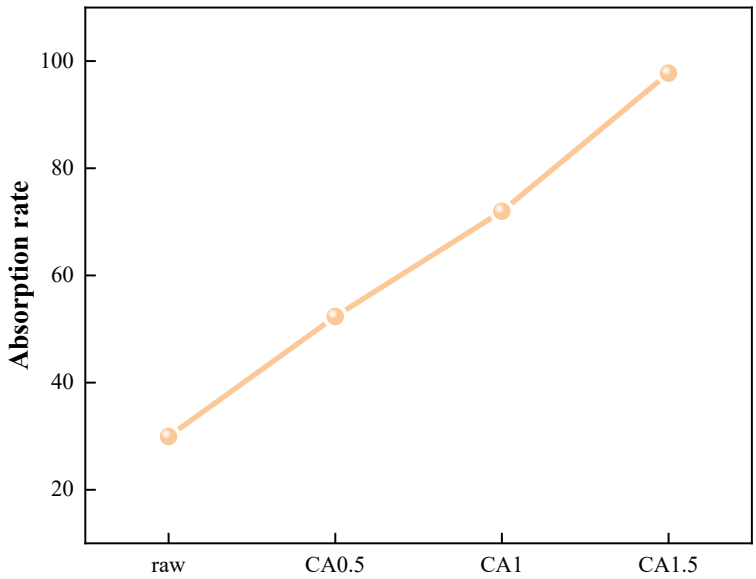

Figure 6 Effect of citric acid concentration on methylene blue adsorption performance

Using CA1.5 as the base material, the synergistic adsorption-photocatalytic removal of high-concentration (80 mg/L) methylene blue by CA- TiO_2 composites with CA-to- TiO_2 mass ratios of 5:0.5, 5:1, and 5:1.5 was investigated. The results (Fig. 5) showed that the removal efficiencies of all three composites were significantly higher than that of pure CA1.5 (70.69%) and increased progressively with TiO_2 loading. The composite with a CA-to- TiO_2 mass ratio of 5:1.5 exhibited the best performance, achieving a removal efficiency of 99.56%, corresponding to nearly complete degradation of methylene blue. The removal efficiencies for the 5:1 and 5:0.5 composites were 91.30% and 83.02%, respectively, both falling short of complete degradation. The lower efficiencies of the 5:0.5 and 5:1 composites can be attributed to insufficient TiO_2 loading, which limited the number of TiO_2 nanoparticles per unit mass of CA, resulting in fewer photocatalytic active sites. In contrast, the higher TiO_2 loading in the 5:1.5 composite, coupled

with effective ultrasonic dispersion that prevented nanoparticle agglomeration, generated dense and well-dispersed photocatalytic active sites, ultimately achieving approximately 100% removal. These findings confirm that a CA-to- TiO_2 mass ratio of 5:1.5 is the optimal loading ratio.

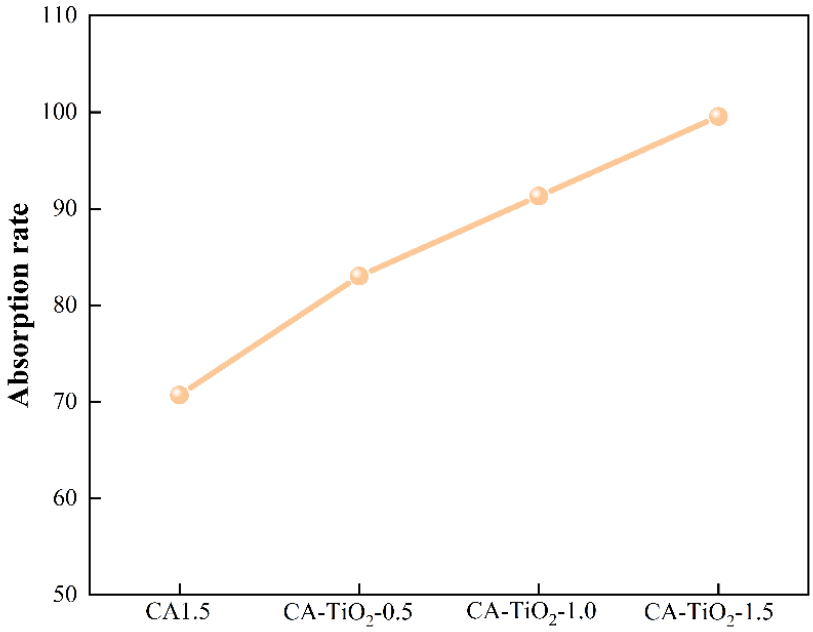

Figure 7 Photocatalytic adsorption-degradation performance of methylene blue

3.2 SEM Characterization Analysis

The surface of the unmodified banana peel exhibited a relatively smooth lamellar structure with a limited number of pores and uneven pore sizes, which are inherent morphological characteristics of natural biomass materials without etching treatment. This resulted in restricted adsorption sites for methylene blue. After modification with 1.5 mol/L citric acid, the surface roughness of the CA powder increased significantly, forming numerous uniformly distributed porous structures. The original lamellar structure was etched into a loose, network-like morphology. This can be attributed to the esterification reaction between citric acid molecules and banana peel cellulose, which etched the surface and enlarged the pores, providing ample space for methylene blue adsorption and subsequent TiO_2 loading.

The surface of the CA- TiO_2 composites showed uniformly attached nanoscale TiO_2 particles that were tightly bound to the inner walls and surfaces of the CA pores, with no obvious agglomeration observed. This indicates that ultrasonic dispersion combined with the impregnation method enabled stable loading of TiO_2 onto the CA surface. The TiO_2 loading did not completely block the pore structure of CA, preserving the loose morphology and ensuring structural support for the diffusion and adsorption of methylene blue molecules as well as the exposure of photocatalytic active sites. These observations are consistent with the enhanced removal efficiency of the composites observed in the subsequent performance tests.

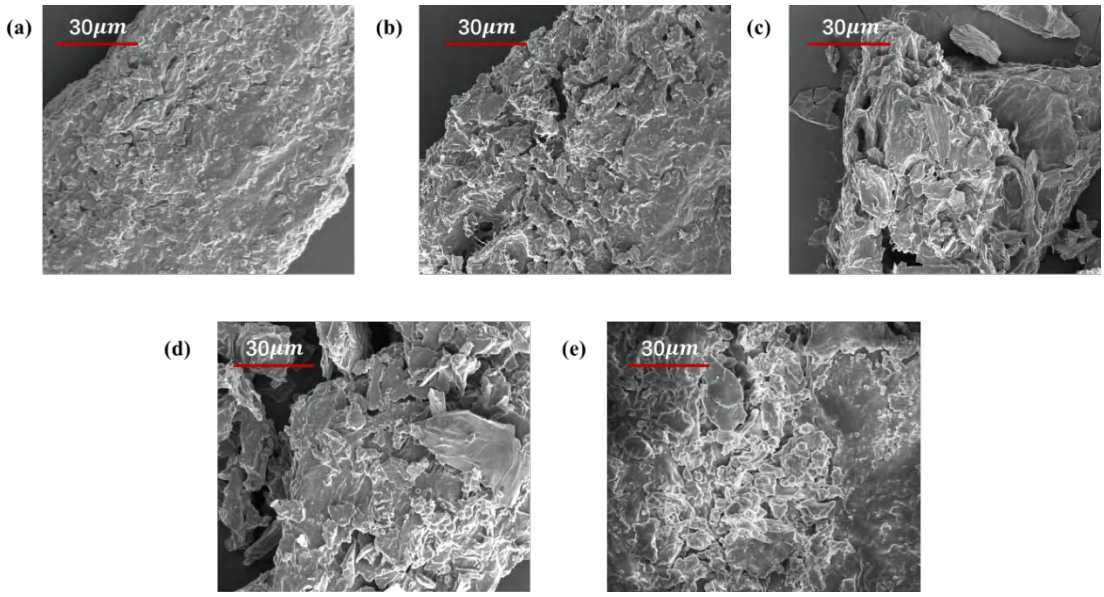

Figure 8 SEM images of (a) unmodified banana peel, (b) CA0.5, (c) CA1.0, (d) CA1.5, and (e) CA- TiO_2-1.5

3.3 FTIR Characterization Analysis

Fourier transform infrared (FTIR) spectroscopy is a core technique for characterizing functional group structures, chemical bond types, and interfacial interactions in materials. The spectra (Fig. 7) reveal that the unmodified banana peel displays a broad and intense absorption peak near 3400 cm^{-1}, corresponding to the stretching vibration of hydroxyl groups (-OH) in cellulose molecules. Peaks near 1630 cm^{-1} and 1050 cm^{-1} are attributed to the bending vibration of adsorbed water and the stretching vibration of C-O-C bonds, respectively, reflecting the primary functional group characteristics of natural banana peel.

For the citric acid-modified CA powder, significant peak fluctuations are observed in the 1500–2000 cm^{-1} range, indicating strong interactions between citric acid molecules and the banana peel. The carboxyl groups (-COOH) of citric acid undergo esterification with hydroxyl groups on the banana peel surface, while the carboxylate groups formed by carboxyl ionization establish hydrogen bonds or ionic bonds with amino and hydroxyl groups in the biomass molecules. This results in superposition and shifting of the original absorption peaks (e.g., C=O stretching and O-H bending vibrations) within this wavenumber range, leading to disordered peak profiles. These structural changes confirm the successful grafting of citric acid onto the banana peel matrix.

In the FTIR spectra of the CA- TiO_2 composites, the characteristic absorption peaks of CA are retained, but the C-O-C absorption peak at 1050 cm^{-1} shifts and increases in intensity, confirming the formation of Ti-O-C chemical bonds. This demonstrates that TiO_2 and CA are not merely physically mixed but form a stable composite through chemical bonding. Such chemical bonding facilitates the transfer of photogenerated electrons between CA and TiO_2, reduces the recombination rate of electron-hole pairs, and enhances photocatalytic degradation performance.

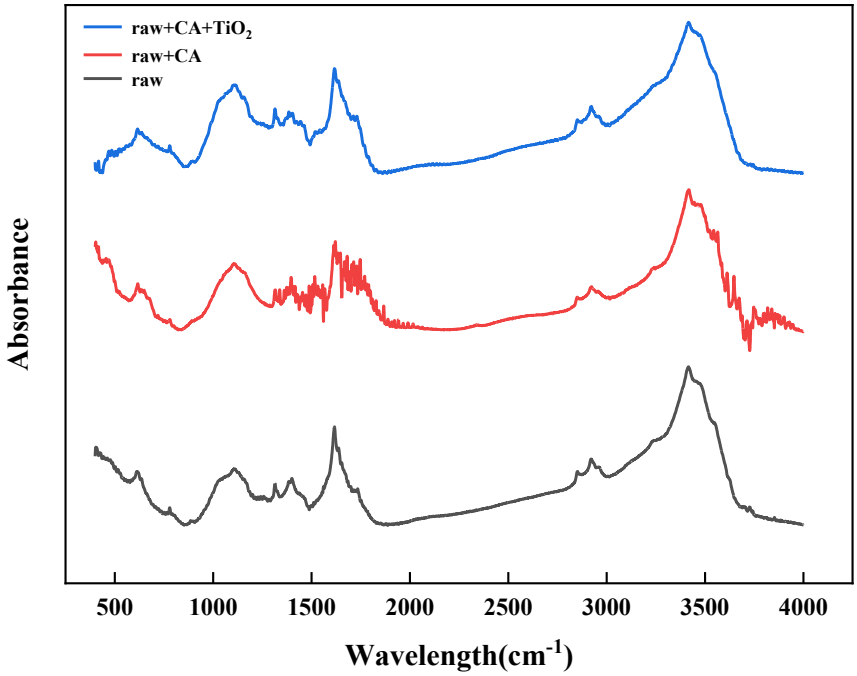

Figure 9 FTIR spectra

3.4 BET Characterization Analysis

BET analysis is a key technique for characterizing the surface properties and pore structure of porous materials. It enables calculation of specific surface area, pore volume, and average pore diameter from nitrogen adsorption-desorption isotherms, providing a quantitative basis for evaluating adsorption capacity and mass transfer efficiency. The pore structure parameters of the optimally modified CA powder and the CA- TiO_2-1.5 composite were determined using BET analysis. **According to IUPAC classification, the average pore diameters of both samples fall within the 2–50 nm range, exhibiting typical mesoporous characteristics**.

As shown in Table 1, the CA powder has a specific surface area of 244.4837 m^2/g, a pore volume of 0.309719 cm^3/g, and an average pore diameter of 5.4996 nm. After TiO_2 loading, the specific surface area of the composite decreased to 221.8197 m^2/g, the pore volume reduced to 0.287239 cm^3/g, and the average pore diameter contracted to 5.2154 nm. The reductions in specific surface area and pore size are primarily attributed to pore-filling and surface-coating effects: TiO_2 nanoparticles penetrate into the mesopores of CA, occupying part of the internal space and thereby decreasing the effective pore dimensions.

Table 1 Pore structure parameters of the CA powder and CA- TiO_2-1.5 composite.

	BET Surface Area (m^2/g)	Pore Volume (cm^3/g)	Average pore diameter (nm)
Raw+CA	244.4837	0.309719	5.4996
Raw+CA+TiO_2	221.8197	0.287239	5.2154

Furthermore, the nitrogen adsorption-desorption isotherm of the CA- TiO_2-1.5 composite exhibited an **H3-type hysteresis loop** (Fig. 8), indicative of slit-shaped pores formed by the

aggregation of TiO_2 particles. This pore structure increases the number of adsorption sites and prolongs the residence time of methylene blue molecules, thereby compensating for the slight decrease in specific surface area and ensuring effective synergistic adsorption-photocatalytic performance.

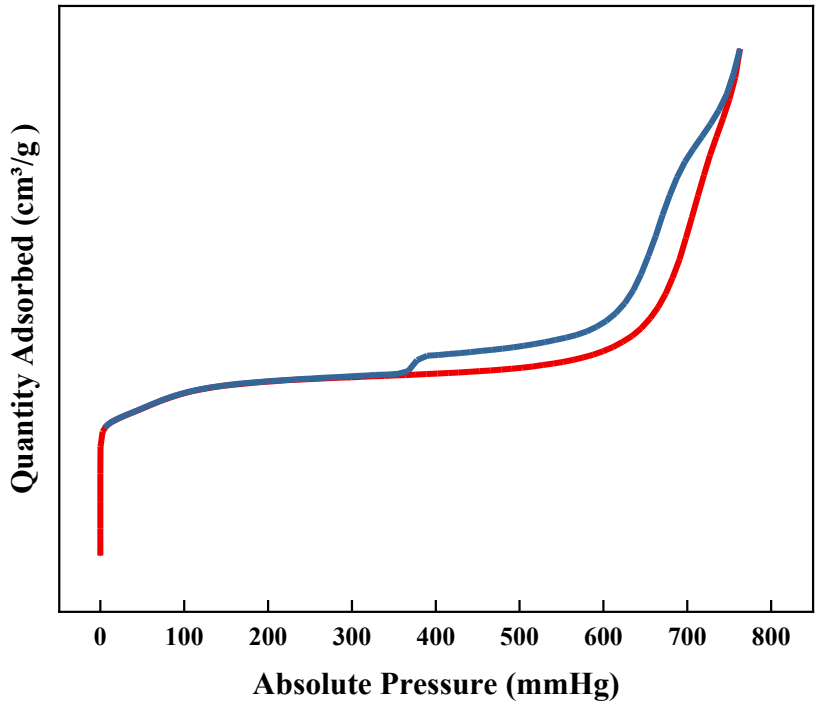

Figure 10 Nitrogen adsorption-desorption isotherms of CA-TiO_2-1.5

4. Application and further experiments on the material and model

The proposed working routine is proposed below:

1. Polluted water flows into the depollution spiral from the lefthand side.
2. The water is absorbed and trapped in the cotton and therefore is in contact with the material.
3. Under the photocatalysis of UV light, the methylene blue in the water which is kept in the cotton is almost degraded and depleted
4. the new polluted water will fill into the cotton again, pushing the purified product out into the tank from the right side.

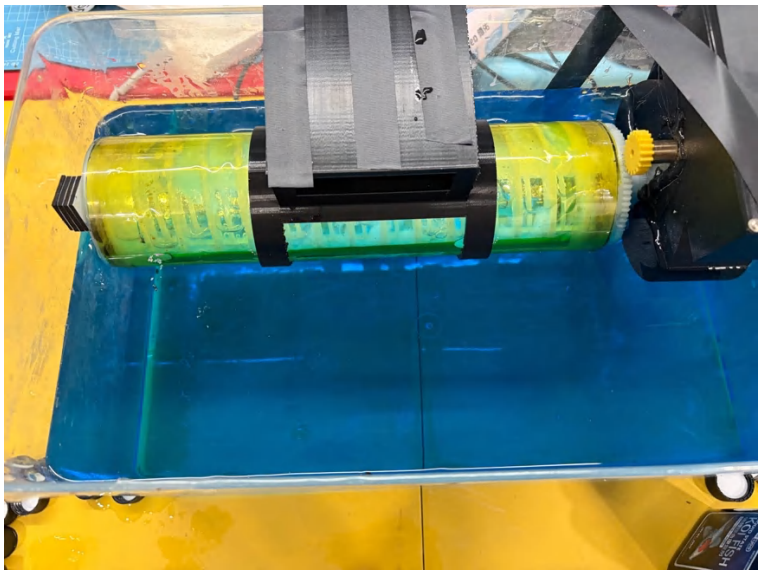

Figure 11 Picture of the application experiment

This process will last until the material reaches its absorption capacity. Therefore, this structure will theoretically lead to the continuous decrease in concentration of the methylene blue solution. Therefore, the absorbance of this solution is expected to decrease. To validate this, an experiment is done.

This experiment aims to uncover the effect of degradation of the device over time. The effect in the short run and in the long run are both considered. Samples are collected directly from the tank for specified time periods (0hr 0.5hr 1hr 1.5hr 18hr 40hr). The samples are shown below, corresponding to the duration of purification shown above from left to right.

Figure 12 Samples from the experiment

The samples are all tested by the spectrophotometer to measure the absorbance. The raw data is plotted in the table below.

Table 2 Raw Data for the Testification of Effectiveness of the Device

	Absorbance (unit = A)					
	T=0hr	T=0.5hr	T=1hr	T=1.5hr	T=18hr	T=40hr
1	0.1108	0.0937	0.0838	0.0887	0.075	0.0574
2	0.1097	0.093	0.0822	0.0867	0.0754	0.0568
3	0.1097	0.0926	0.0828	0.0877	0.0756	0.0569
4	/	/	/	0.0885	/	0.0566
5	/	/	/	0.0834	/	0.0607
6	/	/	/	0.0885	/	0.0609
7	/	/	/	/	/	0.0615

To clearly show the effect, the percentage of methylene blue absorbed respectively is calculated. The processed data table is shown below.

Table 3 Percentage of Absorption of Methylene Blue for Each Group

Groups	Percentage of Absorption (%)
T=0hr	0
T=0.5hr	0.154149
T=1hr	0.24651726
T=1.5hr	0.20729861
T=18hr	0.31556632
T=40hr	0.46681665

What is more, a graph of percentage of absorption against time is plotted.

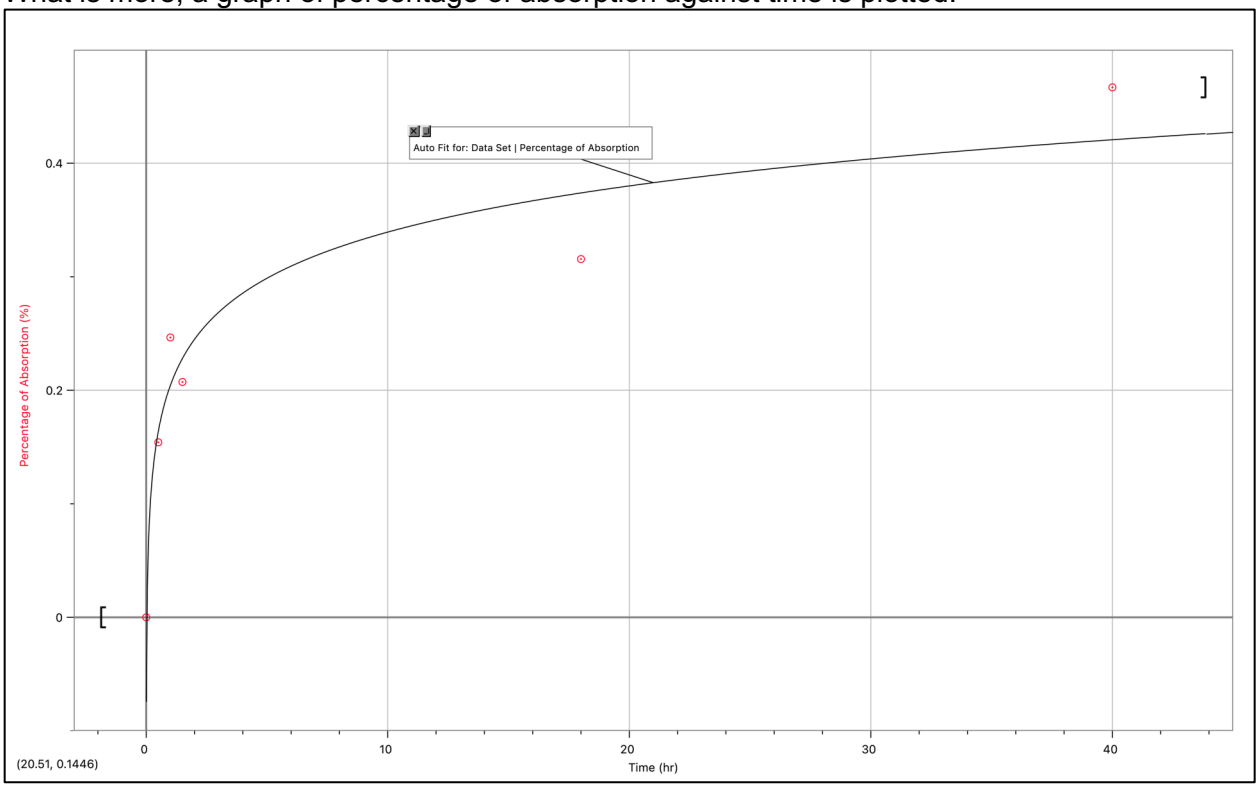

Figure 13 Percentage of Absorption of Methylene Blue in the solution against Time Graph

As the graph shows, the rate of absorption of methylene blue by this device is rather high in the first few hours. Though there is decrease in rate of absorption, it still has the potential to absorb about 60% percent of the methylene blue in a longer period of time.

This result is to some extent conflicting with the previous result of the experiment done on the very same material. However, this is attributed to the difference in environments as well as amounts. Firstly, the experiment done before is merely focusing on the functioning of the material itself. Therefore, the ratio of amount of the material to the amount of methylene blue is a lot higher than that in the latter experiment, which focuses on the effectiveness of the device. What is more, the previous experiment uses UV light with higher power, and the reaction occurs in a black box. That is, the UV light intensity on the material is a lot higher, making it reasonable that the effect of the degradation is a lot better. However, such level of UV light will cause possible harm to human body. Therefore, a weaker UV light is used. This can also be a possible cause of the discrepancy in the results of the two experiments.

Conclusions

In this study, citric acid modification of banana peel achieved simultaneous regulation of functional groups and morphological optimization, introducing carboxyl groups and constructing a porous structure that significantly enhanced adsorption performance. TiO_2 was stably integrated with CA through Ti-O-C bonds, enabling synergistic adsorption-photocatalysis effects: the mesoporous structure of CA rapidly adsorbed and enriched methylene blue, while TiO_2 generated reactive oxygen species under visible light to degrade the dye; additionally, the slit-shaped pores prolonged the residence time of dye molecules, further improving treatment efficiency.

The SEM, FTIR, and BET characterization results corroborated each other, clarifying the regulatory mechanisms of the modification and compositing processes on material morphology, functional groups, and pore structure, thereby providing structural and chemical evidence for the observed improvementsldg in removal efficiency. By optimizing the citric acid modification concentration and TiO_2 loading ratio, a highly efficient CA- TiO_2 composite was successfully prepared. This work also offers a feasible approach for the resource utilization of agricultural waste banana peels and the development of low-cost materials for dye wastewater treatment.

As for the reactor device specially designed to fit this material, it shows an overall excellent function, not only considering from the perspective of operation of the material (will not break over extended period of time), but also from the potential of implementing methylene blue dye degredation and absoprtion. Appearently, though, there’re still signs showing that this device has not yet being able to outshow the potential of the material to the greatest extent, it is firmly believed that by revising the design, more holes with higher volumes will appear on the spiral, more of the material will be able to be inserted and higher efficiency of degredation of organic dye will be possible. Thus, there still exists remarkable spaces for improvement. This devices will be continueously and dedicatedly revised and tested in the near future.

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