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Supporting Information

Synthesis of chitosan-based flocculant by DBD modification and its flocculation performance in wastewater treatment

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The schematic of the experimental setup is shown in Figure S1. The setup consisted of a plasma power supply (CTP-2000K, Nanjing Suman Electronics Co., Ltd., China), an electrical detection system, and a dielectric barrier reactor. The electrical detection system was composed of an oscilloscope (MSO54B, Tektronix, USA), a high-voltage probe (P51000A, Tektronix, USA), and a current probe (A622, Tektronix, USA).

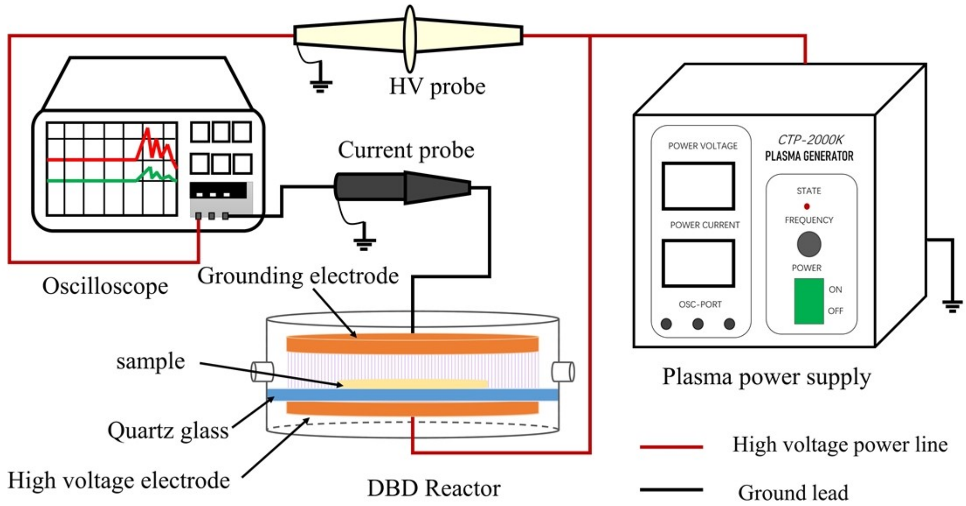

Figure S1. Schematic of the experimental setup

Table S1. Specific parameters of the DBD plasma reactor

Serial Number	Title	Description
1	dielectric material	quartz glass
2	diameter of dielectric material	100 mm
3	thickness of dielectric material	2 mm
4	discharge electrode	stainless-steel plate
5	diameter of electrode	80 mm
6	thickness of electrode	5 mm
7	discharge distance	5 mm

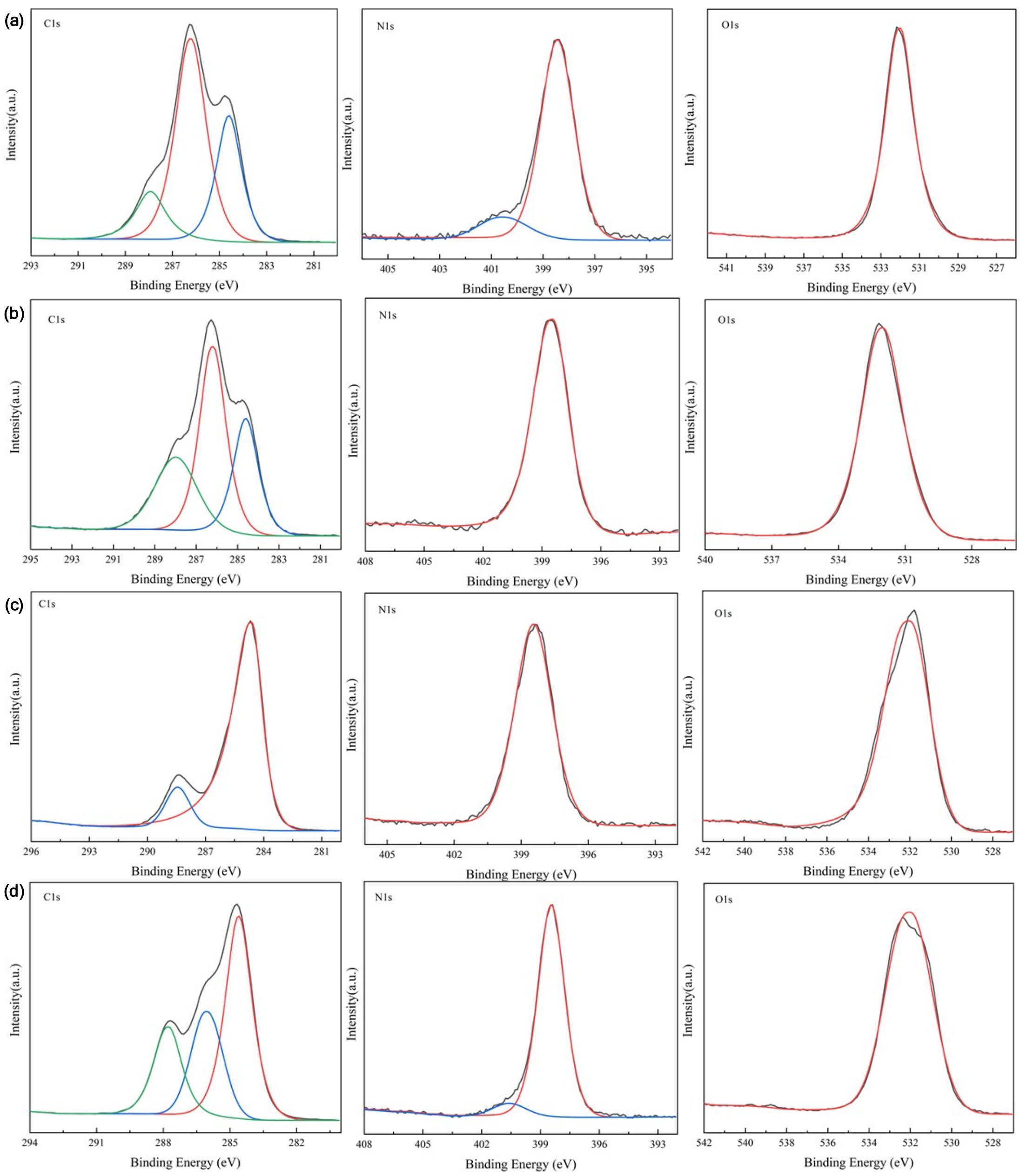

Figure S2. XPS peak fitting diagram of C、N、O: (a) CS; (b) Plasma modified CS; (c) CS-AM; (d) AM

Table S2. The ratio of different functional groups of four substances

	Sample	Binding energy（eV）	Functional group	Ratio（%）
C1s	CS	284.6	C-C; C-H	29.59
		286.2	C-O;	56.50
		287.9	C=O; O-C-O;	13.91
	Plasma modified CS	284.6	C-C; C-H	27.62
		286.21	C-O;	45.08
		287.9	C=O; O-C-O;	27.29
	CS-AM	284.6	C-C; C-H	49.67
		286.2	C-O;	28.10
		287.8	C=O; O-C-O;	22.22
	AM	284.6	C-C; C-H	88.18
		288.4	C=O;	11.82
N1s	CS	398.5	$-NH_2$	87.19
		400.6	$-CONH_2$	12.81
	Plasma modified CS	398.5	$-NH_2$	100.00
	CS-AM	398.5	$-NH_2$	93.44
		400.6	$-CONH_2$	6.56
	AM	398.45	$-NH_2$	100.00

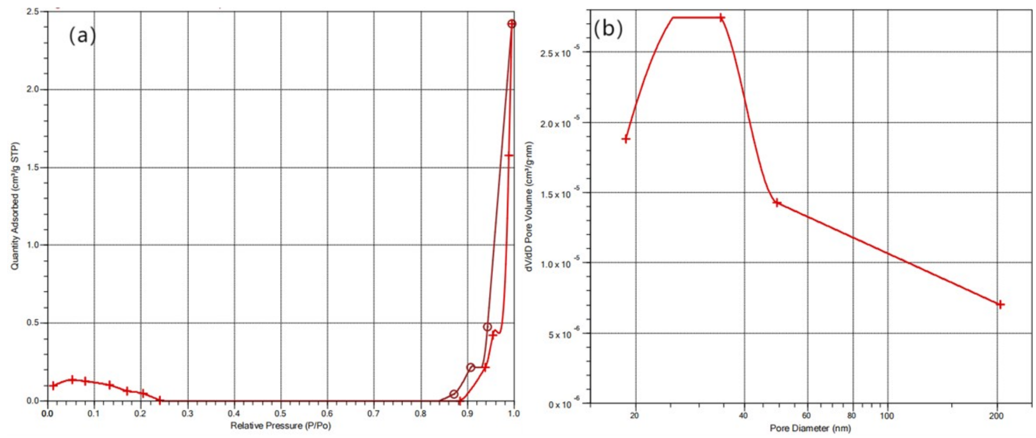

Figure S3. (a) Nitrogen adsorption and desorption isotherm and (b) pore size distribution of CS-AM (60 W, 5 min, m(CS):m(AM)=1:2, 3h, 70°C).

Table S3. BET and average pore size of CS-AM

	CS-AM
Single point surface area at P/Po = 0.052246386	0.5573 m^2/g
BET Surface Area	0.6142 m^2/g
Single point adsorption total pore volume of pores less than 361.3488 nm diameter at P/Po = 0.994681580	0.003746 cm^3/g
Adsorption average pore diameter (4V/A by BET)	24.40 nm
BJH Desorption average pore diameter (4V/A)	32.2720 nm

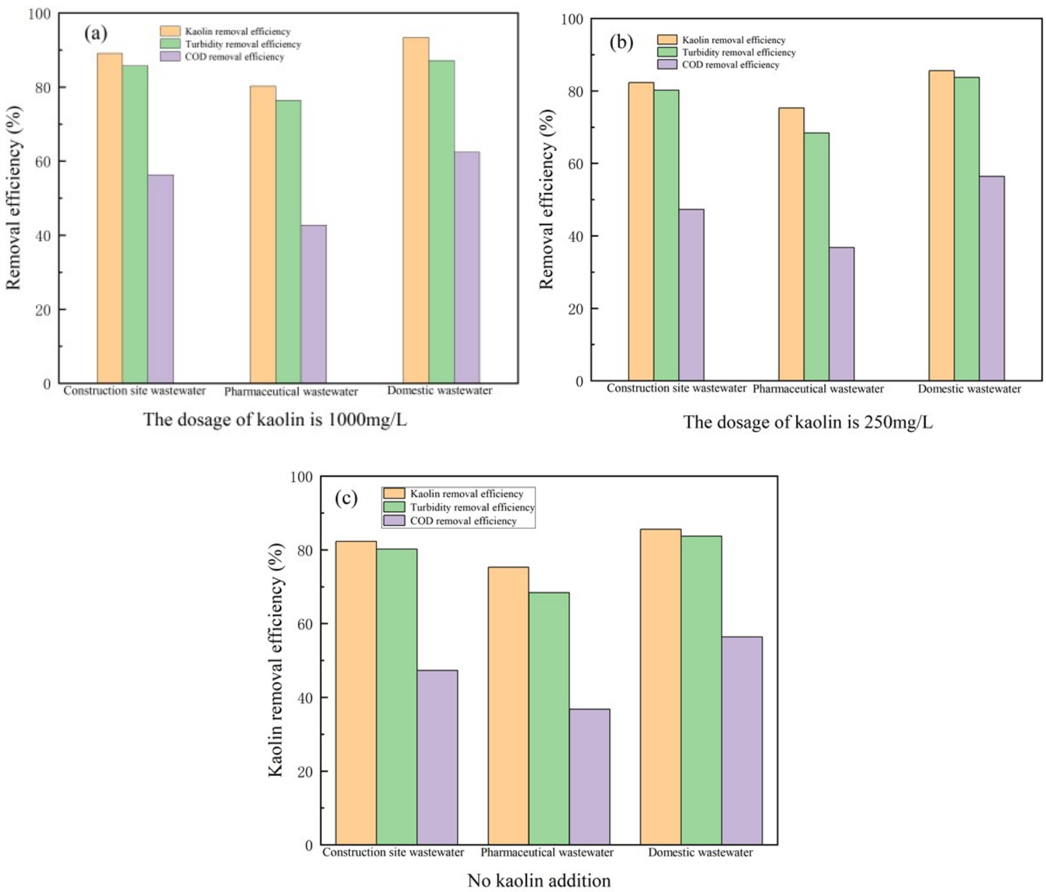

Figure S4. Actual wastewater treatment (a)the dosage of kaolin is 1000mg/L; (b) the dosage of kaolin is 250mg/L; (c) the dosage of kaolin is 0（dosage of flocculant is 5mg/L, stirring intensity is 250rad/min, flocculation time is 30 minutes）

The flocculation effects of CS-AM on three different types of actual wastewater were also investigated. We added 250mg/L and 1000mg/L kaolin into construction site wastewater, pharmaceutical wastewater, and domestic wastewater, for flocculation treatment. As shown in Fig. S4 (a), it could be seen that when the dosage of kaolin was 1000mg/L, CS-AM had the best flocculation effect on domestic wastewater (initial pH is 6.21), with kaolin removal efficiency was 93.4%, turbidity removal efficiency was 87.2%, and COD removal efficiency was 62.38%. The removal efficiency of kaolin in construction site wastewater (initial pH was 6.91) reached 89.1%, turbidity removal efficiency reached 85.7%, and COD removal efficiency reached 56.3%. The removal efficiency of kaolin in pharmaceutical wastewater (initial pH was 6.01) reached 80.3%, turbidity removal efficiency reached 76.4%, and COD removal efficiency reached 42.7%.

From Fig. S4 (b), it could be seen that when the dosage of kaolin was 250mg/L, CS-AM still had the best flocculation effect on domestic wastewater. The removal efficiency of kaolin reached 85.6%, turbidity removal efficiency reached 83.7%, and COD removal efficiency reached 56.4%. The removal efficiency of kaolin for construction site wastewater reached 82.3%, turbidity removal efficiency reached

80.2%, and COD removal efficiency reached 47.3%. The removal efficiency of kaolin for pharmaceutical wastewater reached 75.3%, turbidity removal efficiency reached 68.4%, and COD removal efficiency reached 36.8%.

From Fig. S4 (c), it could be seen that CS-AM had a certain flocculation effect on these three types of raw water without adding kaolin. The removal efficiency of proteins, carbohydrates, etc. in domestic wastewater reached 74.1%, turbidity removal efficiency reached 68.3%, and COD removal efficiency reached 42.9%. The removal efficiency of construction waste in construction site wastewater reached 60.3%, turbidity removal efficiency reached 56.8%, and COD removal efficiency reached 34.6%. The removal efficiency of tetracycline and other pollutants in pharmaceutical wastewater reached 55.6%, turbidity removal efficiency reached 50.6%, and COD removal efficiency reached 14.3%. It can be seen that CS-AM also had good effects on actual wastewater.

Table S4. Comparison between CS-AM and other flocculants.

Flocculant	Synthetic process	Flocculation performance	Advantage/disadvantage	Reference
Chitosan-grafted Polyacrylamide	Thermal initiation	The removal efficiency of p-azo dye reached 86.4%	Large dosage and long flocculation time.	(Zheng et al. 2020)
Chitosan nanoparticles -g-acrylamide	Ultraviolet-assisted	Flocculation of both kaolin suspension and Cu^{2+} wastewater effectively.	Soluble, thermally stable, complex synthetic process.	(Ma et al. 2016)
Chitosan-Acrylamide-Lignin	Microwave assisted method	CAML terpolymer exhibited maximum percentage removal of 99.3% and 67.0% for reactive orange C-3R and methyl orange, respectively.	Environmental protection, high efficiency, good water solubility; fewer types of pollutants are treated.	(Lou et al. 2018)
Methacryloxyethyl trimethyl ammonium chloride	Ultraviolet initiation	The optimal chemical oxygen demand (COD) and the chromaticity removal rates in the dye wastewater were 79.9% and 83.9%	Soluble, high removal efficiencies, complex synthetic process, expensive.	(Sun et al. 2021)
Chitosan-graft-poly (N, N-Dimethylacrylamide)	Polymerization-grafting	The decolorization efficiency of dyes ranges from 83.17% to 99.4%	Good water solubility, large dosage, pH application range is small.	(Jin et al. 2023)
Chitosan-AM (CS-AM)	Dielectric barrier discharge plasma initiation	The removal efficiency of kaolin suspension and CNTs suspension can reach up to 95.9% and 90.2%	CS-AM has the advantages of wide pH application range, good water solubility and less flocculant dosage.	This research