

Synergistic Enhancement of Coagulation and Settling in Congo Red Synthetic Wastewater Treatment Using Magnetic Coagulation: Combination of Polyaluminium Chloride, Pectin, and Iron Oxide Nanoparticles

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Abstract

Conventional polyaluminum chloride (PAC) coagulation is widely used for treating dye wastewater. However, its performance is often limited by relatively slow settling and moderate removal efficiency. This study proposes a magnetic coagulation system by integrating pectin as a natural coagulant aid with iron oxide nanoparticles (IONPs) Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$ under an external magnetic field to enhance Congo red (CR) removal. The effects of pectin dosage, IONPs dosage, and magnetic field intensity on coagulation performance were investigated, while the coagulation mechanism was evaluated through kinetic analysis, control experiments, FTIR spectroscopy, SEM, and microscopic floc characterization. The optimum pectin dosage was 2 mg/L, increasing CR removal from 57.3% to 68.4% through polymer bridging. Subsequent addition of 20 mg/L IONPs further enhanced removal efficiencies to 83.2% and 81.3% for Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$, respectively, while substantially reducing the settling time required to reach equilibrium to approximately 20 minutes. An external magnetic field of 170 mT yielded the best coagulation performance, while stronger fields showed only a slight decline rather than a further gain. Kinetic analysis indicated that the pseudo-second-order (PSO) model best described the coagulation process, with increasing rate constants indicating accelerated floc formation in the presence of IONPs. FTIR, SEM, and microscopic observations further confirmed the synergistic roles of PAC-induced charge neutralization, pectin-mediated polymer bridging, and magnetically enhanced floc aggregation, resulting in denser and more compact sludge structures. These findings demonstrate that integrating natural polymer coagulant aids with magnetic coagulation offers a promising strategy for improving both the efficiency and kinetics of dye wastewater treatment.

Keywords: coagulant-aid, Congo red, magnetic coagulation, natural coagulant, pectin

Introduction

Water pollution remains a global issue with significant environmental and health consequences. Among the major contributors is Congo red (CR), a diazo carcinogenic synthetic dye widely used in the textile industry. Globally, dye production reaches up to 108 tonnes annually, with azo dyes (including CR) accounting for approximately 60–70% of the total dye production. The widespread use of azo dyes consequently generates large volumes of wastewater containing these dyes, contributing to environmental pollution (Harja et al., 2022). Due to its complex aromatic structure, CR exhibits resistance to biodegradation, leading to its persistence in wastewater. The presence of CR alters the color of aquatic environments, produces unpleasant odors, adversely affects aquatic organisms, and poses potential risks to human health (Setyaningtyas et al., 2019).

Among many water treatment methods, coagulation–flocculation is a conventional, cost-effective, and widely applied technique (Harja et al., 2022; Zainudin et al., 2023). However, the use of inorganic coagulants based on Fe and Al, which are commonly utilized, may introduce residual metal ions into treated water, alter its pH, and generate large volumes of toxic sludge. To address this problem, natural coagulant aids have been explored as sustainable alternatives to reduce the dosage of inorganic coagulants required in wastewater treatment (Alazaiza et al., 2022; Bhandari & Ranade, 2014; Koul et al., 2022).

Natural coagulants are typically high-molecular-weight polymers with several advantages, including effectiveness over a wide pH range, absence of residual metal ions, renewability, low toxicity, and biodegradability (Alazaiza et al., 2022; Koul et al., 2022). Pectin has gained attention as a promising natural coagulant (Cam Trang et al., 2025; Kristianto, Jennifer, et al., 2020; Naidu et al., 2018). Commercial pectin is obtained almost entirely from fruit processing residues, chiefly citrus peel and apple pomace, which arise in large volumes as by-products and remain substantially underused. Fruit processing alone can generate peel

residues equivalent to roughly 25–35 wt% of the total fruit mass (Ciriminna et al., 2015; Kristianto, Jennifer, et al., 2020; Sagar et al., 2018). At present, pectin is applied mainly in the food industry as a gelling and stabilizing agent, in biodegradable packaging, and in the pharmaceutical field for drug delivery systems and wound healing (Yi et al., 2024). In addition to these established applications, pectin has also been investigated for wastewater treatment, particularly as a natural coagulant, which is the interest of the present work.

Pectin is a negatively charged heteropolysaccharide built around a homogalacturonan backbone of α -D-(1,4)-linked galacturonic acid units, with rhamnogalacturonan regions carrying neutral side chains such as rhamnose, arabinose, galactose, and xylose (Cam Trang et al., 2025; Kristianto, Jennifer, et al., 2020). A fraction of the galacturonic acid units carries methyl ester groups, and that fraction defines the degree of methyl esterification (DM). Pectin with a DM below 50% is classified as low-methoxy (LMP). It is preferred for coagulation because the larger proportion of free carboxyl groups can interact with the multivalent cations and hydrolyzed metal species supplied by inorganic coagulants such as polyaluminum chloride (PAC), bridging adjacent chains and promoting network formation within the floc (Kristianto, Jennifer, et al., 2020; Yapo & Gnakri, 2014).

The use of pectin as a natural coagulant aid remains relatively underexplored. Haryanto et al. demonstrated a synergistic interaction between aluminum sulfate at 30 mg/L and pectin at 15 mg/L in treating CR synthetic wastewater at 50 mg/L, achieving a removal efficiency of 97.1% (Haryanto et al., 2024). Similarly, Buenaño Bautista et al. reported that the combination of 60 mg/L aluminum sulfate and 0.06 mg/L pectin effectively treated natural water with an initial turbidity of 5.32 NTU, resulting in 87.4% removal (Buenaño Bautista et al., 2019). Cam Trang et al. also achieved 85% removal of hydrazine sulfate and hexamethylenetetramine

97 solution with an initial turbidity of 28.1 NTU using 150 mg/L aluminum sulfate and 50 mg/L
98 pectin (Cam Trang et al., 2025).

99 Conventional coagulation–flocculation, however, has a key limitation, namely the long
100 settling time required, as the process primarily relies on gravitational forces for floc separation
101 (Lv et al., 2018). One approach to overcome this limitation is the application of magnetic
102 coagulation using IONPs, which are among the most used nanomaterials due to their unique
103 characteristics, including a high surface area-to-volume ratio, cost-effectiveness, low toxicity,
104 and high biocompatibility (Xu et al., 2012). Another advantage of magnetic coagulation with
105 IONP incorporation is its ability to enhance adsorption and enable faster floc separation under
106 an external magnetic field (Hu et al., 2024; M. Zhang et al., 2017). Among IONPs, there are
107 three distinct crystalline phases, namely magnetite (Fe_3O_4), maghemite ($\gamma\text{-Fe}_2\text{O}_3$), and
108 hematite ($\alpha\text{-Fe}_2\text{O}_3$) (Mbuyazi & Ajibade, 2024). Fe_3O_4 is commonly selected as a magnetic
109 coagulation material due to its superparamagnetic behavior at the nanoscale, which facilitates
110 manipulation under an external magnetic field while maintaining stability against aggregation
111 (Arrisujaya et al., 2025; Seabra et al., 2017).

112 Several studies have explored the incorporation of Fe_3O_4 to optimize coagulation
113 performance. Inorganic coagulants such as FeCl_3 and PAC have been combined with Fe_3O_4 to
114 enhance coagulation efficiency. FeCl_3 at a dosage of 20 mL/L was reported to achieve 95%
115 removal of 50 mg/L CR synthetic wastewater within 5 minutes of settling time with the
116 assistance of 150 mg/L Fe_3O_4 (Sugih et al., 2025). Likewise, PAC at a dosage of 7.5 mg/L
117 combined with 37.5 mg/L Fe_3O_4 achieved 81.7% removal of 50 mg/L CR synthetic wastewater
118 within 20 minutes of settling time (Suhardi et al., 2024). Similarly, humic acid and kaolin
119 synthetic wastewater at 0.8 mmol/L were treated with 15 mg/L PAC and 30 mg/L Fe_3O_4 ,

achieving 93.8% removal within 5 minutes of settling time under an applied magnetic field of 0.5 T (Lv et al., 2018).

On the other hand, natural coagulants such as *Leucaena leucocephala* and *Moringa oleifera* extracts have also been studied for their synergistic coagulation performance with Fe_3O_4 . *Leucaena leucocephala* at a concentration of 20 mL/L, combined with 3 mg Fe_3O_4 /mL extract, was reported to treat synthetic wastewater containing 50 mg/L CR, achieving 90% removal within 30 minutes of settling time (Kristianto, Tanuarto, et al., 2020). Conversely, a 20 mL saline extract of *Moringa oleifera* combined with 20 mg Fe_3O_4 at 600 mg/L achieved 70% removal of 50 mg/L tartrazine within 10 minutes of settling time (Mateus et al., 2018).

None of these studies, however, has combined an inorganic coagulant, a natural coagulant aid, and magnetic seeding within a single system, and $\alpha\text{-Fe}_2\text{O}_3$ remains untested as a seeding material for this purpose. Therefore, this study investigates the application of pectin as a coagulant aid in combination with Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$, alongside PAC, for the treatment of synthetic CR wastewater via magnetic coagulation. The effects of coagulant aid dosage, IONPs dosage and type, and external magnetic field strength were systematically evaluated. In addition to removal efficiency, settling kinetics were analyzed using appropriate kinetic models to elucidate the coagulation mechanism and assess the influence of each variable.

Materials and Methods

Materials

All chemicals used in this study were of analytical grade and employed without further purification. Polyaluminum chloride (PAC) served as the primary inorganic coagulant and commercial low-methoxy pectin (LMP) as the natural coagulant aid; both were obtained from a local supplier and used as received. The degree of methyl esterification of the pectin was not determined in this work, and the material is therefore treated simply as a commercial low-

methoxy grade. Iron oxide nanoparticles (IONPs) consisting of Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$ with an average particle size of 50 nm (Sigma-Aldrich) were used as magnetic seeding materials to enable magnetic coagulation. Synthetic wastewater was prepared using Congo red (CR) dye (Sigma-Aldrich). Distilled water was used for all solution preparations, and the pH was adjusted as required using sodium hydroxide (NaOH) and hydrochloric acid (HCl) solutions (BrataChem).

Jar Test Experiment

A stock solution of CR was prepared by dissolving 5 g of CR in 1,000 mL of distilled water. Working solutions with the desired initial concentration of 50 mg/L were prepared by diluting the stock solution, which was then adjusted to its optimal coagulation pH of 3 using 0.5 M NaOH or HCl, and measured with a calibrated digital pH meter (± 0.01 accuracy). A PAC stock solution was prepared by dissolving 100 mg of PAC in 100 mL of distilled water, then diluting to the desired initial concentration. Pectin solution was prepared separately by dissolving the required amount of pectin powder in 10 mL of deionized water. IONPs were weighed according to the required dosage and dispersed in the PAC working solution by ultrasonication immediately before each run, so that PAC and IONPs were dosed together as a single suspension. A constant PAC dosage of 5 mg/L was used throughout the experiment at a constant CR initial concentration of 50 mg/L and pH of 3, followed by variation of the pectin dosage (0–3 mg/L) at a constant 5 mg/L of PAC, the IONPs dosage was varied (0–60 mg/L), and lastly, the external magnetic field was varied. The coagulation pH of 3 was chosen based on previous research (Sudirgo et al., 2023). The variations in this study are presented in **Table 1**.

A coagulation study was conducted using a standard jar test apparatus at room temperature. The PAC–IONPs suspension was added to the synthetic wastewater working

solution and mixed rapidly at 200 rpm for 1 min, followed by the addition of the pectin solution and a further 2 min of rapid mixing at the same speed, and then slow mixing at 60 rpm for 30 min. In the runs without IONPs, the PAC working solution alone was dosed at the same point in the sequence. After coagulation–flocculation, the mixture was left to settle over a permanent magnet placed beneath the beaker. The magnetic flux density quoted throughout this work was measured at the outer base of the beaker, that is, at the point closest to the settling flocs, using a gaussmeter (Tunkia, TD8620). Samples were collected every 5 minutes for 60 minutes and measured using a UV-Vis spectrophotometer (Thermo Scientific Genesys 150) at CR's maximum wavelength (570 nm).

Table 1. Variation of pectin, IONPs, and external magnetic field in this study

Parameter	pH	PAC (mg/L)	Pectin (mg/L)	IONPs (mg/L)	External Magnetic Field (mT)
Pectin dosage	3	5	0-3	0	170
IONPs dosage	3	5	Best pectin dosage	0-60	170
External magnetic field	3	5	Best pectin dosage	Best IONPs dosage	0-250

The removal efficiency (%removal) of CR from synthetic wastewater was calculated using Eq. (1), where C_0 and C (mg/L) refer to the initial and final concentration of CR. Sludge volume was measured using an Imhoff cone with a settling time of 60 min and calculated using Eq. (2). The formed flocs were visually observed under a light microscope with an external camera. All experiments were conducted in duplicate, and all the data in this study are presented as mean value $\pm$ standard deviation, where applicable. Statistical analysis was performed using two-way analysis of variance (ANOVA) with least significant difference (LSD) post hoc test in SPSS ver. 20.0.

Control experiments were conducted under the same jar test conditions to determine the contribution of each component. In the runs performed without PAC, the IONPs were dispersed in an equal volume of deionized water rather than the PAC working solution. They

were dosed at the same point in the mixing sequence, ensuring identical dosing order and mixing regime across all runs. These controls were used to determine whether the observed performance arises from the combination of PAC, pectin, and IONPs rather than from any single component acting alone.

$$\% \text{removal} = \frac{C_0 - C}{C_0} \times 100 \quad (1)$$

$$\text{Sludge volume} = \frac{\text{Measured sludge volume (mL)}}{\text{Synthetic wastewater volume (L)}} \quad (2)$$

Settling Kinetic Model

Coagulation proceeds through charge neutralization and interparticle bridging, and the capture of dye molecules by growing flocs follows the same functional form as a sorption process. On that basis, and following earlier coagulation studies (Beltrán-Heredia et al., 2009; Suhardi et al., 2024), pseudo-first-order and pseudo-second-order expressions are adopted here as empirical rate models for the removal-versus-time data. Because q is obtained by rescaling the measured removal by a constant factor, the fitted parameters describe how fast and how far removal proceeds. The removal capacity (q_e and q_t) was calculated using Eq. (3) where q (mg dye/mg PAC) refers to removal capacity, C_0 (mg/L) to the initial CR concentration, C_e (mg/L) to the concentration at equilibrium, C_t (mg/L) to the concentration at time t , V (L) to the sample volume, and m (mg) to the mass of PAC dosed. Normalization is performed to the PAC dose alone, since PAC is held constant across all runs while the IONPs are treated as a seeding additive rather than as part of the coagulant dose (Suhardi et al., 2024).

The kinetic models for the removal of CR were approached using adsorption models, i.e., the pseudo-first-order (PFO) and PSO, as shown in Eqs. (4) and (5), respectively. For these models, q_t (mg dye/mg PAC) refers to removal capacity in time t , q_e (mg dye/mg PAC) refers to removal capacity at equilibrium, t (min) represents the time when the sample was taken, k_1 (min^{-1}) is the rate constant of PFO model, and k_2 (mg PAC/mg dye·min) is the rate constant of

the PSO model. The experimental data were fitted to the models using nonlinear regression, and goodness-of-fit was evaluated using the adjusted R^2 and the reduced chi-square (χ^2).

$$q = \frac{(C_0 - C_t)}{m} \times V \quad (3)$$

$$q_t = q_e(1 - e^{-k_1 t}) \quad (4)$$

$$q_t = \frac{k_2 q_e^2 t}{1 + k_2 q_e t} \quad (5)$$

Sludge Characterization

Physical characterization of the sludge produced was performed using Fourier Transform Infrared Spectroscopy (FTIR, Bruker, Alpha II), Scanning Electron Microscopy (FESEM, SU3500, Hitachi), and light microscopy (Olympus, CX22RFS1). For FTIR and SEM analyses, sludge samples were oven-dried at 50 °C until completely dry, then scraped off and analyzed. For microscopic evaluation, sludge was extracted using a pipette after a 60-minute sedimentation period. Observations were conducted using a light microscope equipped with an OptiLab digital camera and OptiLab Viewer 4 software, capturing images at 4×, 10×, and 40× magnification. To quantitatively assess floc morphological complexity, these images were processed using the FracLac plugin (version 2.5) in ImageJ (version 1.54g) to determine fractal dimensions.

Results and Discussion

Effect of Pectin Dosage in Coagulation

The effect of pectin addition on CR removal efficiency and sludge volume at different concentrations is presented in **Fig. 1A**. The removal of CR showed a positive trend as pectin dosage increased. This result suggests that pectin and PAC have a synergistic effect. The PAC forms small, loose flocs via the charge neutralization mechanism (Z. Zhang et al., 2018). Then, pectin acts as a coagulant aid through a polymer bridging mechanism, further supporting floc formation by charge neutralization, thereby improving coagulation performance (Haryanto et

al., 2024). However, adding pectin above the optimal level decreased removal, as seen at 2.5 and 3 mg/L. This occurs because an overdose of pectin increases repulsive forces between the coagulant and the CR, dispersing flocs and hindering settling (Sanghi et al., 2007). It is concluded that, at a base PAC dosage of 5 mg/L, varying the pectin concentration from 1 to 3 mg/L yielded the highest removal efficiency at 2 mg/L. Adding 2 mg/L pectin raised removal from 57.3% to 68.4%; above that dosage, removal fell back rather than leveling off, reaching values close to the pectin-free baseline. Sludge volume followed the same trend as removal, reaching 19.5 mL/L at 2 mg/L pectin. Sludge volume is reported throughout this work as an indicator of floc structure rather than treatment quality; a rise in volume alongside an increase in removal indicates that more material has been swept out of solution into an open, bridged floc network. This pectin dosage was used in the subsequent experiments.

Effect of IONPs Dosage in Coagulation

The effect of IONPs addition on CR removal efficiency and sludge volume at different concentrations is presented in **Fig. 1B**. The results indicate that IONPs addition exerted minimal influence on removal performance at concentrations up to 10 mg/L. However, a substantial increase in removal efficiency was observed at 20 mg/L, reaching 83.24% and 81.27% for Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$, respectively, representing the highest performance among the tested dosages. Beyond this concentration, a slight decline in removal efficiency was observed within the 30–60 mg/L range. A similar, but more pronounced, trend was evident in sludge volume, which increased with IONPs dosage up to 20 mg/L before progressively decreasing at higher concentrations. These results indicate that 20 mg/L is the optimum IONPs dosage, providing the most favorable coagulation performance. Two-way ANOVA indicated that IONPs type (Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$) did not significantly affect CR removal ($p > 0.05$), whereas IONPs dosage significantly influenced the removal efficiency ($p < 0.05$). Post hoc LSD

analysis showed that the highest removal efficiency was achieved at 20 mg/L, which differed significantly from the other dosage levels.

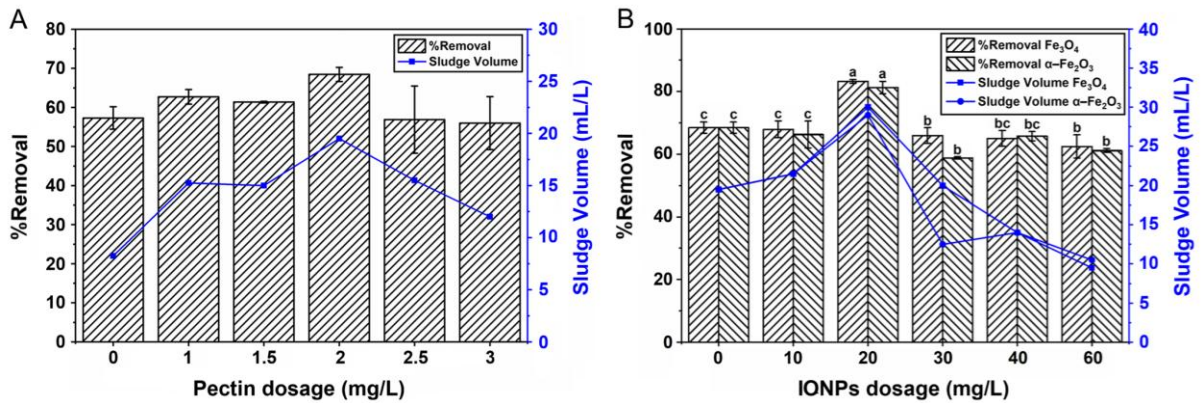


Fig. 1 Effect of (A) pectin dosage and (B) IONPs dosage on CR %removal and sludge volume

The observed enhancement in coagulation performance at moderate IONPs dosages can be attributed to magnetic nanoparticles serving as nucleation sites that facilitate floc growth and aggregation. The presence of IONPs promotes particle collisions and accelerates floc formation, while their magnetic properties enable stronger interactions under an external magnetic field, enhancing floc settling and separation efficiency (Lv et al., 2018; Suhardi et al., 2024). Consequently, larger, denser flocs form, resulting in improved contaminant removal and increased sludge accumulation at the optimum dosage. In contrast, the decrease in performance observed at IONP dosages above 20 mg/L is likely attributed to nanoparticle overdosing. Excessive concentrations of IONPs can disrupt the coagulation process by increasing particle crowding within the suspension, interfering with effective interparticle bridging and floc stabilization. Furthermore, the elevated abundance of nanoparticles intensifies Brownian motion and particle–particle interactions, potentially destabilizing developing flocs and increasing the likelihood of floc breakage (Hu et al., 2024).

The kinetics of CR removal as a function of settling time and kinetic model fitting are presented in **Fig. 2**. The results shown in **Fig. 2A** and **Fig. 2B** demonstrate that coagulation performance increased rapidly during the initial 5 min of settling, followed by a gradual approach toward equilibrium, which was reached within approximately 20 min (inset figure). This represents a significant reduction in the settling time required to achieve equilibrium compared with the system without IONPs addition. The accelerated removal behavior indicates that IONPs incorporation not only influences coagulation efficiency but also substantially enhances the rate at which flocs are separated from the suspension.

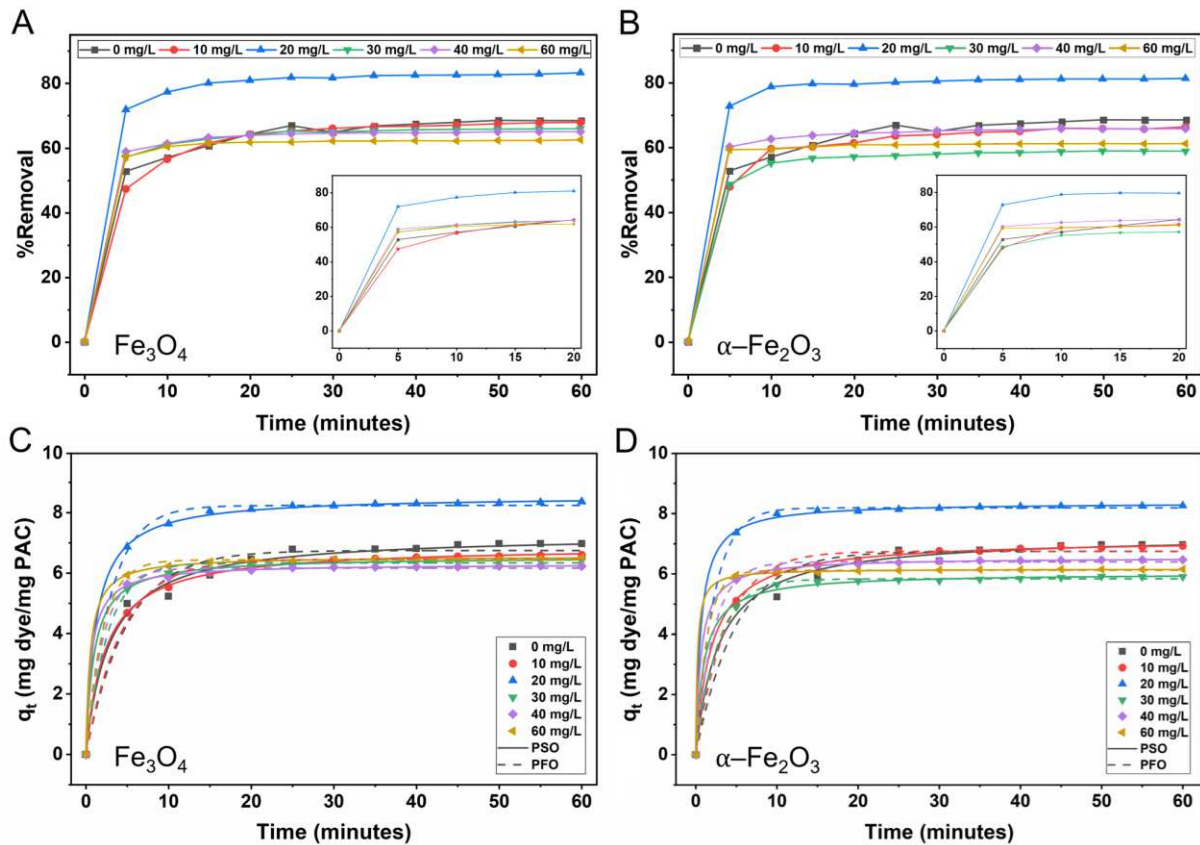


Fig. 2 CR removal performance: Time-dependent removal of various (A) Fe_3O_4 and (B) $\alpha\text{-Fe}_2\text{O}_3$ dosages, comparison of kinetic model fitting for (C) Fe_3O_4 and (D) $\alpha\text{-Fe}_2\text{O}_3$

This trend is further supported by the kinetic parameters obtained from model fitting shown in **Fig. 2C** and **2D**, and as summarized in **Table 2**. A comparison between PFO and

PSO kinetic models based on their respective Reduced Chi-Square (χ^2) values confirms the superior suitability of the PSO model for describing the coagulation process. A corresponding increase in the PSO k_2 parameter accompanied an increase in the IONPs dosage. Although higher IONPs dosages did not necessarily result in higher q_e , the larger k_2 values indicate that equilibrium was achieved more rapidly. This behavior suggests that the presence of additional IONPs facilitates faster floc formation and settling under the applied magnetic field, thereby reducing the time required for the system to reach equilibrium. While q_e may decrease slightly at excessive IONP dosages, the k_2 increases due to enhanced magnetic responsiveness. Similar observations have been reported in previous studies, in which larger k_2 values were associated with shorter times to attain maximum adsorption or removal capacity (Huang et al., 2021; Suhardi et al., 2024). Based on these results, an IONPs dosage of 20 mg/L was selected for subsequent variations.

One observation deserves explicit comment. Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$ belong to different magnetic classes and therefore respond very differently to an applied field, yet the two-way ANOVA found no significant effect of IONP type on removal ($p > 0.05$). Had the enhancement been governed mainly by magnetic force, a substantial gap between the two oxides would be expected. Its absence indicates that most of the benefit at 20 mg/L comes from particles acting as ballast and nucleation sites, raising floc density and collision frequency, while the applied field exerts a smaller effect, showing up chiefly in the settling rate rather than in the equilibrium removal. The kinetic parameters support this reading, since k_2 increases with IONP dosage, even when q_e does not.

Table 2. Kinetic parameters of IONPs dosage variations

IONPs	Model	Parameter	IONPs dosage (mg/L)					
			0	10	20	30	40	60
Fe ₃ O ₄	Pseudo-First-Order	q_e model (mg dye/mg PAC)	6.7474	6.4556	8.1357	6.3384	6.1655	6.4383
		k_1 (min ⁻¹)	0.2033	0.2305	0.4960	0.3801	0.4714	0.4983
		Standard Error q_e	0.1222	0.0548	0.0360	0.0313	0.0306	0.0201
		Standard Error k_1	0.0253	0.0146	0.0359	0.0216	0.0354	0.0255
		Reduced Chi-Sqr (χ^2)	0.1306	0.0275	0.0138	0.0101	0.0099	0.0043
		Adj. R-Square (R^2)	0.9640	0.9916	0.9973	0.9967	0.9966	0.9987
	Pseudo-Second-Order	q_e model (mg dye/mg PAC)	7.2904	6.8986	8.3028	6.5450	6.3070	6.5571
		k_2 (mg PAC/mg dye·min)	0.0484	0.0620	0.2142	0.1583	0.2497	0.2946
		Standard Error q_e	0.1167	0.0202	0.0144	0.0125	0.0127	0.0066
		Standard Error k_2	0.0069	0.0018	0.0108	0.0056	0.0131	0.0092
		Reduced Chi-Sqr (χ^2)	0.0442	0.0015	0.0011	0.0007	0.0008	0.0002
		Adj. R-Square (R^2)	0.9878	0.9996	0.9998	0.9998	0.9997	0.9999
α -Fe ₂ O ₃	Pseudo-First-Order	q_e model (mg dye/mg PAC)	6.7474	6.7542	7.9092	5.8374	6.4026	6.1190
		k_1 (min ⁻¹)	0.2033	0.2627	0.4688	0.3637	0.4620	0.7241
		Standard Error q_e	0.1222	0.0589	0.0195	0.0181	0.0263	0.0146
		Standard Error k_1	0.0253	0.0189	0.0173	0.0124	0.0278	0.0616
		Reduced Chi-Sqr (χ^2)	0.1306	0.0331	0.0040	0.0034	0.0073	0.0023
		Adj. R-Square (R^2)	0.9640	0.9907	0.9992	0.9987	0.9977	0.9992
	Pseudo-Second-Order	q_e model (mg dye/mg PAC)	7.2904	7.1600	8.0669	6.0283	6.5485	6.1682
		k_2 (mg PAC/mg dye·min)	0.0484	0.0718	0.2152	0.1656	0.2379	0.8022
		Standard Error q_e	0.1167	0.0350	0.0192	0.0325	0.0043	0.0095
		Standard Error k_2	0.0069	0.0040	0.0145	0.0161	0.0040	0.0907
		Reduced Chi-Sqr (χ^2)	0.0442	0.0048	0.0019	0.0050	0.0001	0.0005
		Adj. R-Square (R^2)	0.9878	0.9987	0.9996	0.9981	1.0000	0.9998

327

328 Effect of External Magnetic Field Strength in Coagulation

329 The influence of external magnetic field strength on coagulation performance was
 330 evaluated over the range 0–250 mT, with results presented in **Fig. 3**. Removal efficiency
 331 remained relatively unchanged at lower magnetic field strengths, with no improvement
 332 observed up to 150 mT. Subsequently, an increase in removal efficiency was observed at 170
 333 mT, yielding the highest removal efficiency and indicating this magnetic field strength as the
 334 optimum for the coagulation process. Beyond this point, removal efficiency fell slightly at 200
 335 and 250 mT. Magnetic saturation accounts for the absence of any further gain, since the
 336 magnetization of Fe₃O₄ approaches its plateau in this range, but it does not by itself account

for a decline. A more plausible contribution is that a strong field pulls the IONPs together before they have been fully incorporated into flocs, producing dense particle chains that settle separately from the dye-bearing flocs and therefore carry less dye out of suspension. Two-way ANOVA demonstrated that magnetic field intensity significantly influenced CR removal efficiency ($p < 0.05$), whereas the IONPs type did not ($p > 0.05$). Post hoc LSD analysis further showed that the removal efficiency at 170 mT was significantly higher than those at the other magnetic field intensities, regardless of the IONPs used.

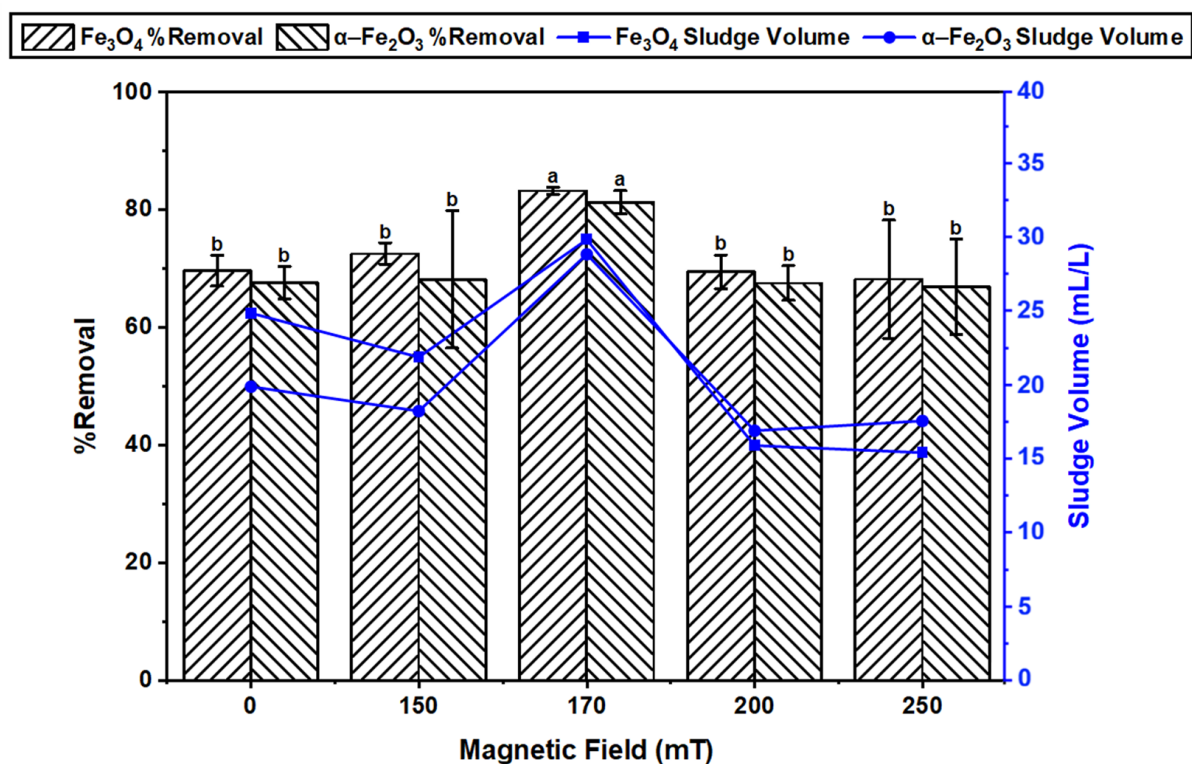


Fig. 3 The effect of magnetic field strength on CR removal and sludge volume

Sludge volume behaved differently under the applied field. Following the same reading used above, a fall in volume at constant or higher removal indicates that the same amount of material has been packed into a denser floc rather than that less material was captured. Volume decreased across most field strengths, except at the optimum condition, where it rose with removal. Increasing field intensity therefore promotes closer packing of the magnetic

aggregates and yields denser, more compact sludge (Yu et al., 2020). From an operational standpoint, this is the preferred outcome, as it reduces the volume of sludge requiring handling. Under the strongest field applied, α -Fe₂O₃ produced a somewhat more voluminous sludge than Fe₃O₄.

The two oxides belong to different magnetic classes; Fe₃O₄ is ferrimagnetic and, at the nanoscale, superparamagnetic, whereas α -Fe₂O₃ is antiferromagnetic and shows only weak canted ferromagnetism (Seabra et al., 2017). Under the same applied field, the magnetization induced in α -Fe₂O₃ is therefore far smaller; the magnetic force acting on the α -Fe₂O₃ flocs is correspondingly weaker, and less field-driven compaction is expected. Studies on this variable remain scarce, but the two-sided behavior observed here matches that described by Gu et al., who attributed poor performance at low field intensity to insufficient coagulant–pollutant interaction and poor performance at high field intensity to disruption of the coagulant system itself (Gu et al., 2026). Field strength therefore behaves as an operating parameter with a genuine optimum rather than one to be maximised.

Aside from removal efficiency and sludge volume production, the kinetic parameters presented in **Table 3** also provide important insights into the effect of magnetic field intensity. For both IONPs, a similar trend was observed, where the k_2 and q_e generally increased with increasing magnetic field strength. However, the q_e values initially increased up to 170 mT, but then declined at higher intensities (200 and 250 mT), indicating that q_e no longer increased proportionally with k_2 beyond this point. Although the removal efficiencies obtained at 200 and 250 mT remained comparable to those observed at lower magnetic field intensities (0 and 150 mT), the corresponding kinetic behavior differed substantially. At lower magnetic field (up to 170 mT), increases in k_2 were accompanied by relatively consistent increases in q_e , suggesting that the optimum condition had not yet been reached, since both the removal rate and the removal capacity continued to improve simultaneously.

Table 3. Kinetic parameters of magnetic field variations

IONPs	Model	Parameter	Magnetic Field (mT)				
			0	150	170	200	250
Fe ₃ O ₄	Pseudo-First-Order	$q_{e \text{ model}}$ (mg dye/mg PAC)	6.5377	7.2949	8.1357	7.0487	7.4824
		k_1 (min ⁻¹)	0.2298	0.4091	0.4960	0.4932	0.5834
		Standard Error q_e	0.0675	0.0320	0.0360	0.0315	0.0180
		Standard Error k_1	0.0177	0.0225	0.0359	0.0356	0.0304
		Reduced Chi-Sqr (χ^2)	0.0417	0.0107	0.0138	0.0105	0.0035
		Adj. R-Square (R^2)	0.9876	0.9974	0.9973	0.9972	0.9992
	Pseudo-Second-Order	$q_{e \text{ model}}$ (mg dye/mg PAC)	6.9903	7.5015	8.3028	7.1950	7.5770
		k_2 (mg PAC/mg dye·min)	0.0607	0.1615	0.2142	0.2441	0.3839
		Standard Error q_e	0.0766	0.0128	0.0144	0.0120	0.0041
		Standard Error k_2	0.0066	0.0058	0.0108	0.0116	0.0092
		Reduced Chi-Sqr (χ^2)	0.0209	0.0008	0.0011	0.0007	0.0001
		Adj. R-Square (R^2)	0.9938	0.9998	0.9998	0.9998	1.0000
α -Fe ₂ O ₃	Pseudo-First-Order	$q_{e \text{ model}}$ (mg dye/mg PAC)	6.2840	5.9548	7.9092	7.0375	6.9011
		k_1 (min ⁻¹)	0.2379	0.3279	0.4688	0.6159	0.6380
		Standard Error q_e	0.0722	0.0226	0.0195	0.0269	0.0211
		Standard Error k_1	0.0209	0.0123	0.0173	0.0571	0.0512
		Reduced Chi-Sqr (χ^2)	0.0483	0.0051	0.0040	0.0078	0.0048
		Adj. R-Square (R^2)	0.9845	0.9981	0.9992	0.9979	0.9987
	Pseudo-Second-Order	$q_{e \text{ model}}$ (mg dye/mg PAC)	6.7272	6.1914	8.0669	7.1315	6.9789
		k_2 (mg PAC/mg dye·min)	0.0637	0.1301	0.2152	0.4092	0.4901
		Standard Error q_e	0.0407	0.0266	0.0192	0.0177	0.0140
		Standard Error k_2	0.0039	0.0086	0.0145	0.0455	0.0510
		Reduced Chi-Sqr (χ^2)	0.0059	0.0032	0.0019	0.0017	0.0011
		Adj. R-Square (R^2)	0.9981	0.9988	0.9996	0.9996	0.9997

378

379 In contrast, at higher field intensities, k_2 continued to rise, while q_e increased only
380 slightly before leveling off, indicating that the removal capacity had reached a plateau. Floc
381 formation and dye capture therefore proceeded faster without any corresponding gain in the
382 amount of dye that could ultimately be removed. As a result, the slight decline in removal
383 efficiency beyond the optimum condition further indicates that excessively high magnetic field
384 intensities may disrupt coagulant–pollutant interactions, thereby adversely affecting the overall
385 coagulation–flocculation performance.

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Control Experiments

To evaluate the contribution of each component and confirm the synergistic effect among PAC, pectin, and IONPs, control experiments using individual components and selected combinations were conducted. The results are presented in **Fig. 4**. In the absence of PAC, removal remained below 3% regardless of the presence of Pectin or IONPs. The addition of 2 mg/L pectin alone also did not significantly improve the removal efficiency, indicating that pectin and IONPs alone were insufficient to destabilize the colloidal particles. A similar trend was observed for sludge volume, where no measurable sludge was produced. The addition of 5 mg/L PAC together with 20 mg/L IONPs increased both removal and sludge volume, indicating that PAC played a major role in particle destabilization and floc formation. However, the removal efficiency was still lower than that observed in the presence of pectin.

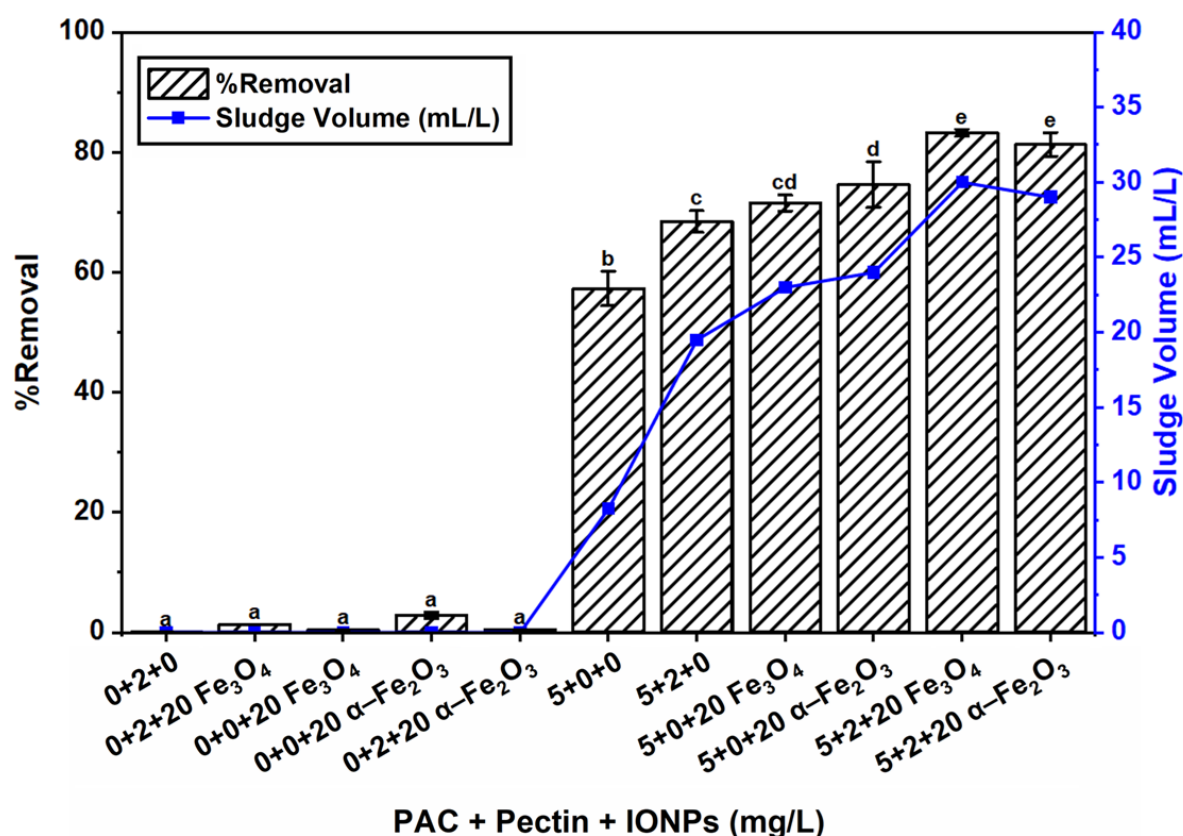


Fig. 4 Control experiments of individual components and their combinations on %removal and sludge volume

Sludge Characterization

Further evidence of these synergistic interactions was obtained by comparing the Fourier Transform Infrared (FTIR) spectra of the pristine materials with those of the sludge generated after the coagulation–flocculation process, which reveal the functional groups involved and the potential interactions among the constituent materials. **Fig. 5** presents the IR spectra of the pristine components, namely PAC, pectin, Fe_3O_4 , and $\alpha\text{-Fe}_2\text{O}_3$. Across the spectrum, the broad absorption band around 3400 cm^{-1} corresponds to O–H stretching vibrations of hydroxyl groups and adsorbed water (Pavia et al., 2009). Additionally, the band at 1640 cm^{-1} across all spectra can be attributed to the bending vibrations of adsorbed water (Gai et al., 2021). In the PAC spectrum, the region below 1200 cm^{-1} is associated with Al–O and Al–OH vibrations characteristic of hydrolyzed aluminum species at 622 cm^{-1} and 847 cm^{-1} respectively (Gai et al., 2021; Xiang et al., 2023).

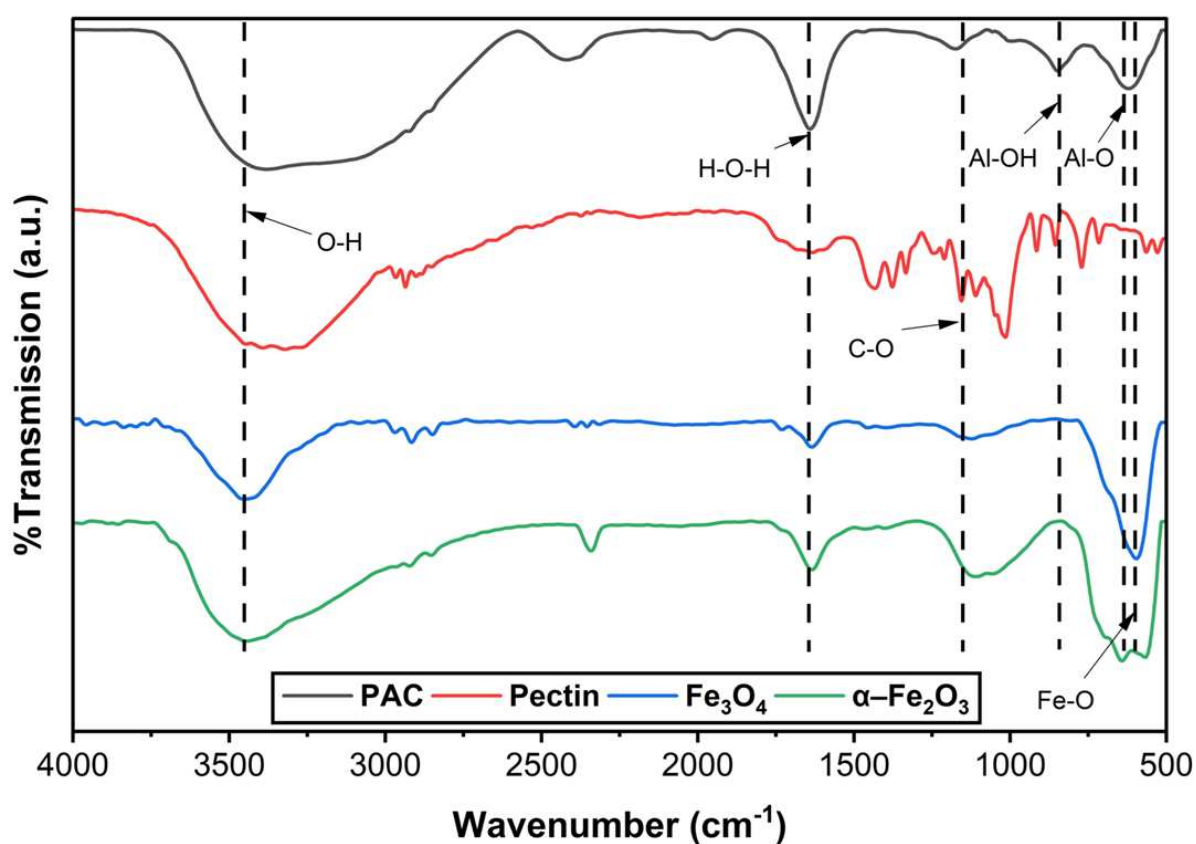


Fig. 5 FTIR analysis of pristine components

The pectin spectrum shows a band near 1740 cm^{-1} arising from the ester carbonyl of methyl-esterified galacturonic acid units and a stronger band in the $1640\text{--}1600\text{ cm}^{-1}$ region arising from asymmetric stretching of free carboxylate groups (COO^-); the relative intensity of the two reflects the degree of methyl esterification. Several peaks between 1246 and 1015 cm^{-1} correspond to C–O stretching of the polysaccharide backbone (Pavia et al., 2009). The Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$ spectra both show strong absorption below 700 cm^{-1} from Fe–O stretching. In Fe_3O_4 , the band near $570\text{--}590\text{ cm}^{-1}$ arises from the tetrahedral and octahedral Fe–O sites of the inverse spinel lattice, whereas $\alpha\text{-Fe}_2\text{O}_3$, which contains only Fe^{3+} , shows bands near 630 and 530 cm^{-1} associated with its corundum-type structure (Farahmandjou & Soflaee, 2015; Mohapatra et al., 2015). The $\alpha\text{-Fe}_2\text{O}_3$ spectrum shows a broader, more intense Fe–O region than that of Fe_3O_4 , reflecting differences in crystal structure and surface chemistry (Farahmandjou & Soflaee, 2015). The presence of abundant hydroxyl, carboxyl, and metal oxide surface groups suggests that PAC, pectin, and IONPs possess multiple reactive sites capable of participating in intermolecular interactions during sludge formation.

Compared with the pristine PAC and pectin spectra, the sludge spectra shown in **Fig. 6** exhibit a broader and slightly shifted O–H stretching region ($3400\text{--}3200\text{ cm}^{-1}$), indicating the formation of hydrogen bonding interactions between hydroxyl groups of pectin and hydroxylated aluminum species originating from PAC. Simultaneously, changes in the intensity and profile of absorption bands within the $1700\text{--}1000\text{ cm}^{-1}$ region were observed relative to the pristine materials, indicating the involvement of multiple functional groups in the coagulation–flocculation process. These spectral modifications suggest interactions among hydrolyzed aluminum species derived from PAC, the carboxyl functionalities of pectin, and characteristic functional groups associated with CR, particularly the sulfonate (S=O) stretching vibrations ($1375\text{--}1140\text{ cm}^{-1}$) and N–H bending vibrations ($1640\text{--}1550\text{ cm}^{-1}$) (Pavia et al., 2009). These interactions indicate that pectin is not merely physically blended with PAC but is

integrated into a hybrid polymer-metal network, promoting floc formation and stabilizing the resulting floc's structure.

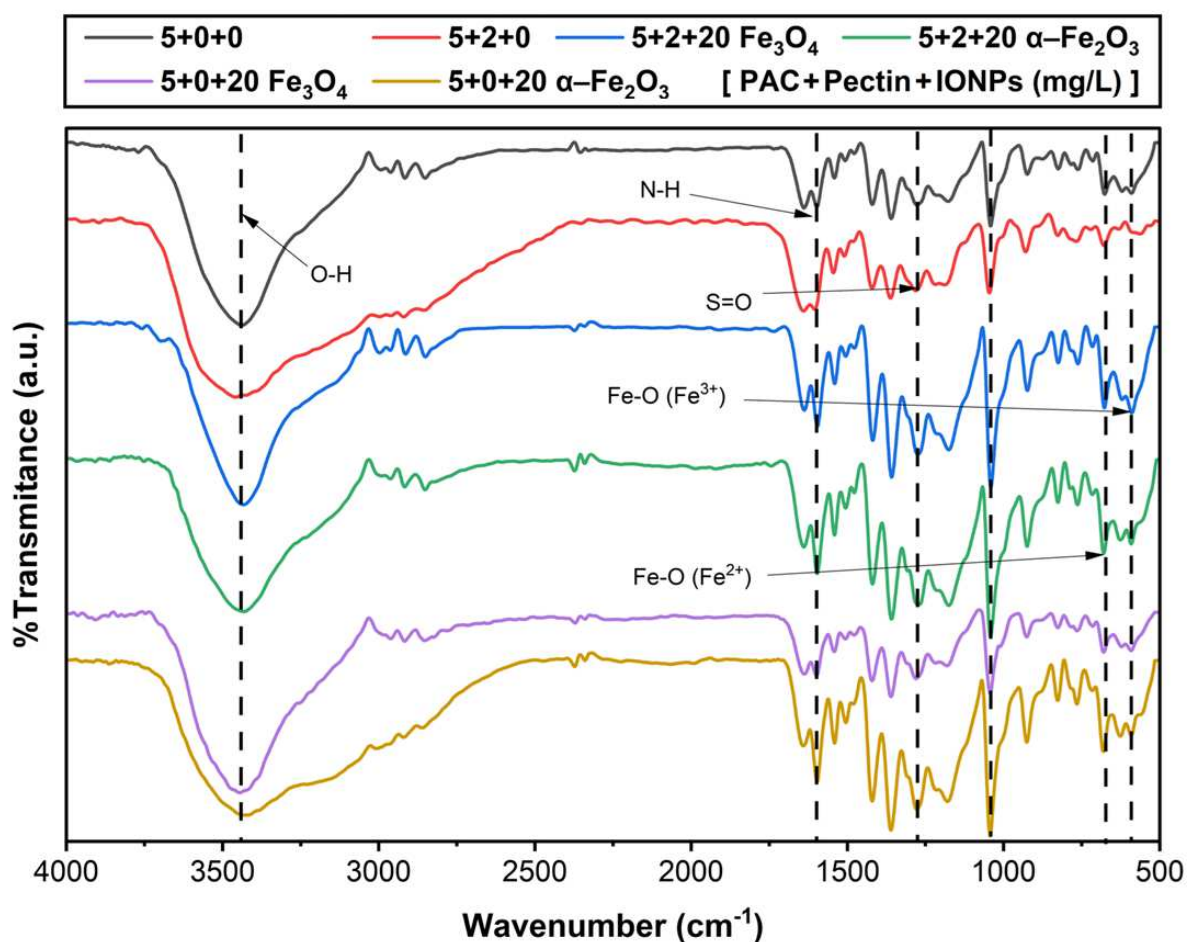


Fig. 6 FTIR analysis of PAC + Pectin + IONPs formulations

A comparison between the Fe_3O_4 -containing and $\alpha\text{-Fe}_2\text{O}_3$ -containing sludges revealed that the $\alpha\text{-Fe}_2\text{O}_3$ formulations exhibited more pronounced spectral features within the 3400–3200 cm^{-1} region, accompanied by a more pronounced broadening of the O–H stretching band. These observations suggest a higher abundance of accessible surface hydroxyl groups on $\alpha\text{-Fe}_2\text{O}_3$, which may enhance its interaction with the hydroxyl and carboxyl groups from pectin, as well as hydrolyzed aluminum derived from PAC. Consequently, $\alpha\text{-Fe}_2\text{O}_3$ appears to interact more strongly with PAC and pectin within the composite matrix, as reflected by the more pronounced spectral changes compared to the Fe_3O_4 -containing formulations. Despite the

stronger interactions indicated by FTIR analysis, the coagulation–flocculation performance of α -Fe₂O₃-containing sludge was comparatively lower, as shown in **Fig. 1B**. This behavior may be attributed to the excessive availability of surface hydroxyl groups at higher α -Fe₂O₃ loadings, leading to overdosage and subsequent colloidal restabilization. Under such conditions, the excess positively charged or hydroxylated surface sites may inhibit effective particle aggregation by re-establishing electrostatic repulsion between destabilized particles, thereby reducing floc growth and diminishing overall coagulation–flocculation efficiency (Kristianto et al., 2023; Sanghi et al., 2006). These findings suggest that although stronger chemical interactions can be achieved with α -Fe₂O₃, an optimum balance between interfacial association and colloidal destabilization is required to maximize treatment performance.

Further insight into the sludge characteristics was obtained through Scanning Electron Microscopy (SEM), as shown in **Fig. 7**, which reveals distinct morphological variations in sludge from different coagulation components. Sludge generated solely with PAC (**Fig. 7A**) exhibited relatively smaller, more dispersed aggregates and less interconnected floc structure. Upon the addition of pectin (**Fig. 7B**), the sludge morphology became increasingly interconnected, indicating the formation of a more extensive floc network through polymer-assisted bridging interactions. Similar behavior has been reported for natural polymer flocculants, in which bridging interactions facilitate the formation of larger flocs and enhance the structural connectivity among destabilized particles (Zheng et al., 2017).

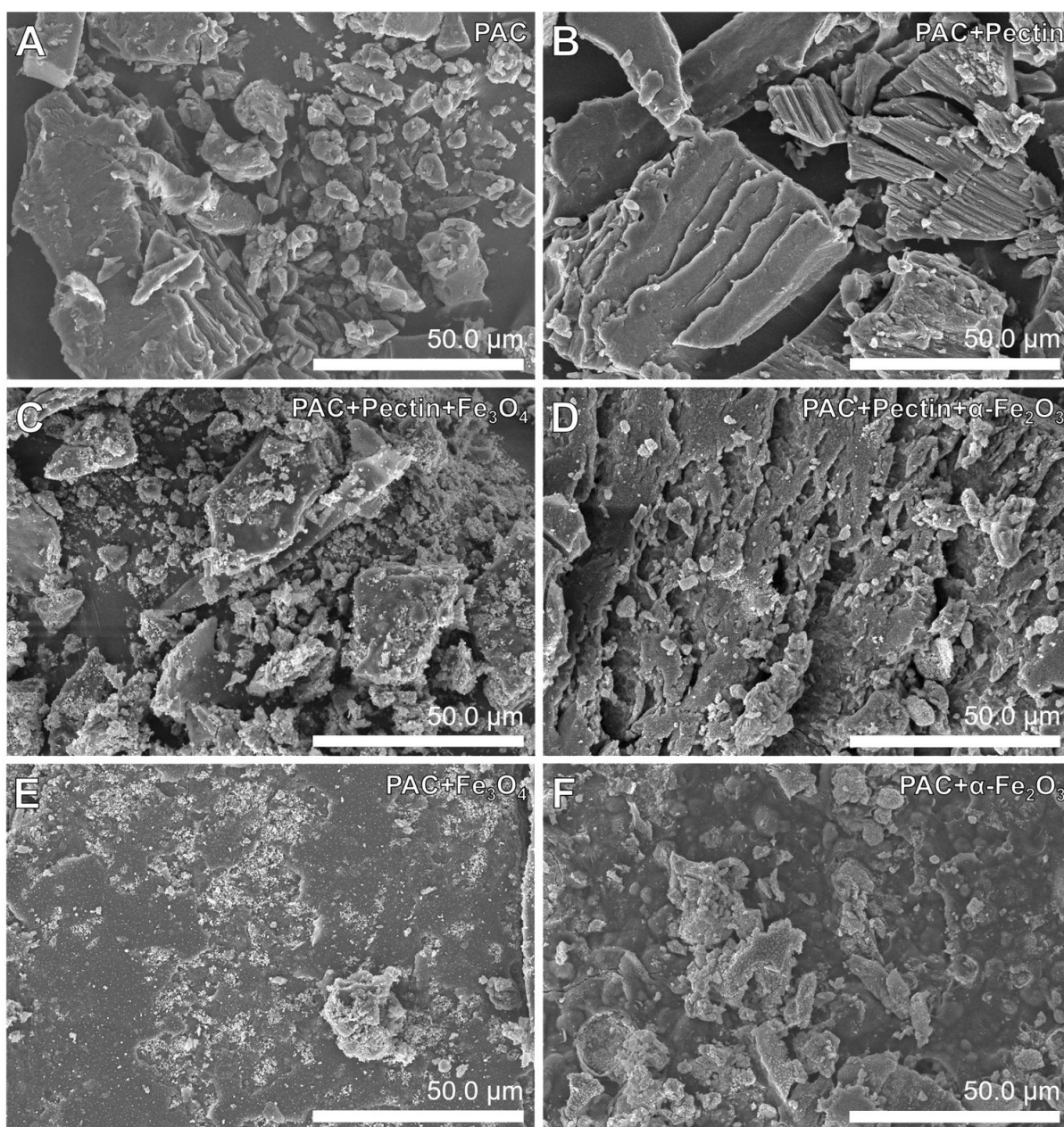


Fig. 7 SEM analysis of (A) PAC sludge, (B) PAC + Pectin sludge, (C) PAC + Pectin + Fe_3O_4 sludge, (D) PAC + Pectin + $\alpha\text{-Fe}_2\text{O}_3$ sludge, (E) PAC + Fe_3O_4 sludge, (F) PAC + $\alpha\text{-Fe}_2\text{O}_3$ sludge

Nevertheless, the presence of noticeable voids between aggregates suggests the formation of a relatively voluminous, porous sludge matrix. A more pronounced morphological transformation was observed following the incorporation of IONPs (**Fig. 7C** and **Fig. 7D**). Both Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$ promoted denser and more compact sludge structure, characterized

by the substantial reduction or disappearance of aggregate voids, which can be attributed not only to the ability of the nanoparticles to occupy spaces within the floc matrix and facilitate additional particle–particle interactions, but also to the influence of the external magnetic field applied during the settling stage. The magnetic responsiveness introduced by the embedded IONPs enables the flocs to experience an additional attractive force toward the magnetic field source, promoting closer packing of aggregates and accelerating floc consolidation. As the magnetically responsive sludge is drawn and concentrated under the applied field, the spaces between flocs are progressively reduced, resulting in a more densely packed structure.

Magnetic coagulation studies have similarly reported the formation of dense network aggregates and compact floc structures arising from magnetic interactions among Fe_3O_4 -containing particles, which enhance aggregate cohesion beyond that achievable through conventional coagulation alone (Liu et al., 2023). This phenomenon is reflected in the surface morphology, where a similar nanoparticle coverage pattern has been reported for Fe_3O_4 -coated adsorbents and magnetic flocculants, in which IONPs partially cover the host surface, generating rough, heterogeneous morphologies through nanoparticle attachment and agglomeration (Belibagli et al., 2024).

Furthermore, the sludge surfaces appeared to be extensively coated with IONPs, suggesting a strong affinity between the nanoparticles and the coagulated flocs. This observation is further supported by the SEM images in **Fig. 7E** and **7F**, which depict sludge produced in the presence of PAC and IONPs without pectin addition. The distribution of nanoparticulate deposits across the sludge surface demonstrates the successful attachment of IONPs to the floc matrix. Such behavior is consistent with the adsorption of nanoparticles onto coagulated particles, enabling their integration within the sludge structure and contributing to the enhanced compactness observed in the magnetic coagulation systems. The extensive surface coverage further indicates the effective participation of IONPs in the floc-formation

process, supporting their role in modifying sludge morphology and improving aggregate stability. In general, the sludge surface exhibits morphological characteristics comparable to those reported in the literature, including a rough and heterogeneous topography with abundant hills, protrusions, and cracks (Al-Musawi et al., 2021).

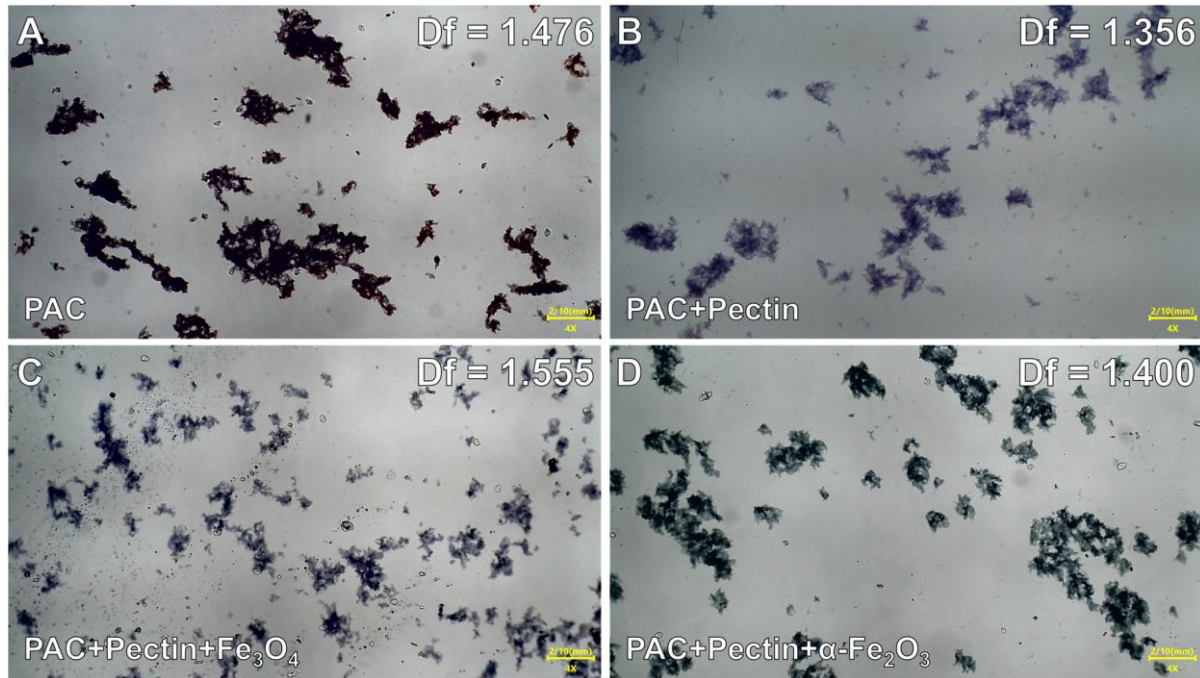


Fig. 8 Microscope observation of floc formation at 4× magnification for (A) PAC sludge, (B) PAC + Pectin sludge, (C) PAC + Pectin + Fe_3O_4 , (D) PAC + Pectin + $\alpha\text{-Fe}_2\text{O}_3$ in optimum conditions

A visual analysis using light microscopy was performed to characterize floc formation further, as shown in **Fig. 8**. The images were also analyzed using the box-counting method in the FracLac plugin in ImageJ to quantify the two-dimensional (2-D) fractal dimensions (Df) of each floc. The results show that PAC-alone-formed flocs (**Fig. 8A**) have an average Df of 1.476. Flocs formed with the addition of pectin (**Fig. 8B**), however, while increasing %removal, reduce the average Df to 1.356 relative to PAC alone. This reduction is attributed to the polymer-bridging nature of pectin, which creates branched structures within the flocs, consistent with previous findings that bridging flocculation produces flocs with the lowest Df

(Li et al., 2006). Conversely, the addition of IONPs reversed this effect, increasing collision frequency and promoting the formation of more compact, larger flocs, thereby increasing Df under a magnetic field (Wang et al., 2016). Flocs formed with the addition of Fe_3O_4 (**Fig. 8C**) have an average Df of 1.555, exceeding that of PAC alone. Flocs formed with the addition of $\alpha\text{-Fe}_2\text{O}_3$ (**Fig. 8D**) reached an intermediate Df, above that of the pectin-only system but below that of the Fe_3O_4 system. The ordering is consistent with the much weaker magnetization of $\alpha\text{-Fe}_2\text{O}_3$, which offers less field-driven compaction, although the difference is modest and rests on a limited number of images.

The integration of PAC, pectin, and IONPs enables each material to compensate for the limitations of the others, resulting in enhanced floc formation, improved settling characteristics, and superior overall coagulation performance compared with the individual components operating independently. Based on the analysis, the proposed interactions and their respective contributions to floc development are illustrated in **Fig. 9**. The proposed coagulation mechanism (**Fig. 9B**) involves the sequential interaction of PAC, CR dye, pectin, and IONPs. As shown in **Fig. 9A**, initially, PAC interacts with the negatively charged CR molecules, forming small, loose microflocs primarily via charge neutralization (Z. Zhang et al., 2018). Then, pectin functions as a coagulant aid by initiating a polymer bridging mechanism, connecting PAC-CR microflocs and facilitating their growth into larger flocs, thereby improving overall coagulation performance (Haryanto et al., 2024). IONPs attached to the polymer matrix then act as magnetic cores, promoting denser floc formation under an external magnetic field (Mohamed Noor et al., 2022).

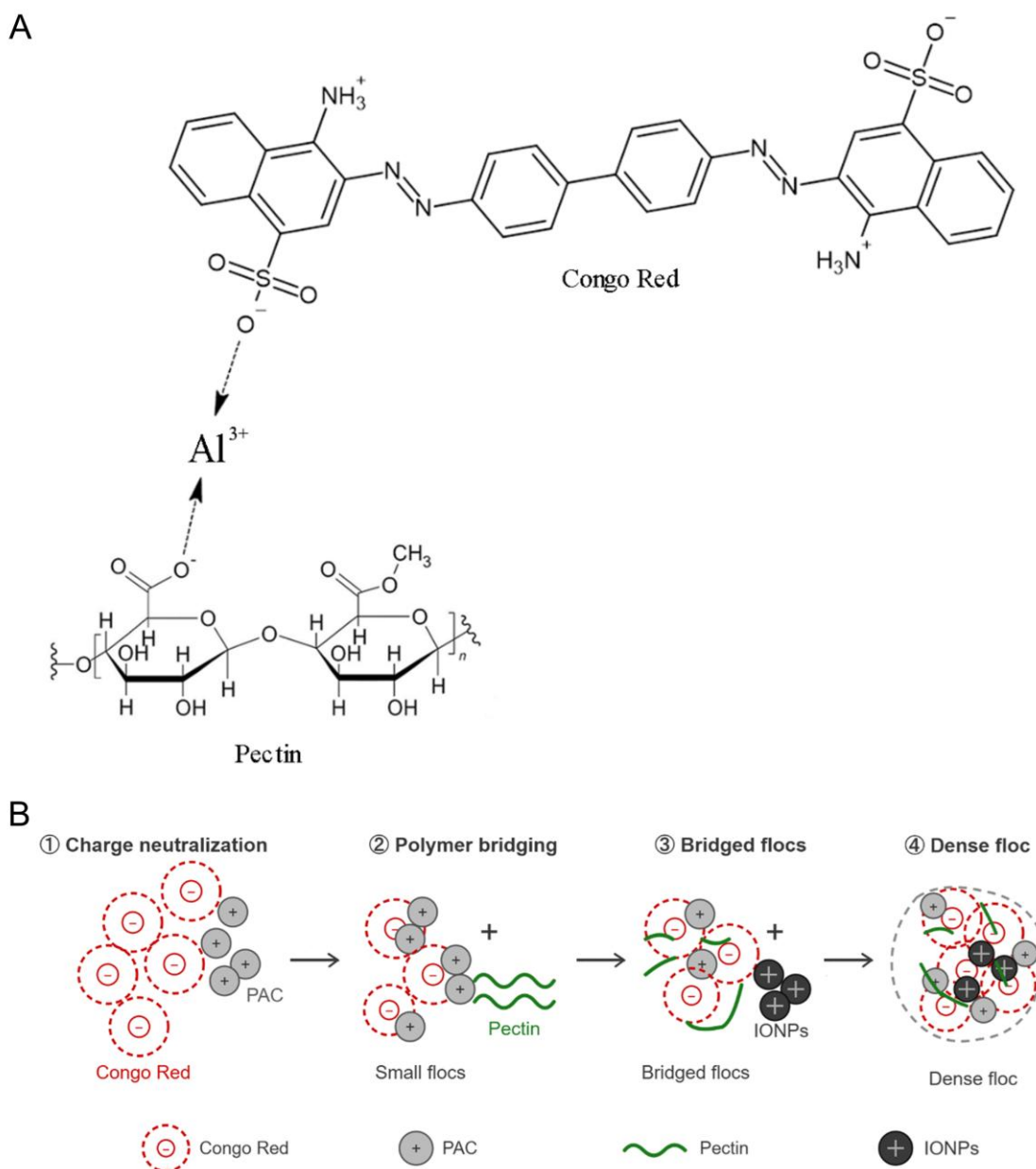


Fig. 9 (A) Schematic illustration of the intermolecular interactions between PAC, pectin, and CR, and (B) proposed synergistic coagulation–flocculation in the presence of IONPs

Conclusions

This study explores the benefits of combining pectin with IONPs (Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$) and PAC to treat synthetic CR wastewater. The optimal coagulation performance was achieved with 5 mg/L PAC and 2 mg/L pectin, resulting in synergistic charge neutralization and polymer bridging and achieving 68.4% removal. The addition of IONPs increased removal and

shortened the settling time required to reach equilibrium, with an optimum dosage of 20 mg/L for both IONP types, resulting in 83.2% and 81.3% removal for Fe_3O_4 and $\alpha\text{-Fe}_2\text{O}_3$, respectively, at a magnetic field intensity of 170 mT, with equilibrium attained within approximately 20 min. Furthermore, varying the magnetic field showed different effects on removal and settling kinetics, with 170 mT as the optimal magnetic field intensity. The kinetic analysis in this study shows that the PSO kinetic model best describes the coagulation of CR synthetic wastewater across all variations, as evidenced by lower Reduced Chi-Square values compared to the PFO kinetic model. The results are further validated by FTIR and SEM analysis, which confirm the synergistic interactions among PAC, pectin, and IONPs. Taken together, the results show that a low dose of PAC combined with pectin and IONPs under a moderate magnetic field removes CR more effectively and settles considerably faster than PAC alone. Each component contributes a distinct function, and the control runs confirm that none of them is effective in isolation. Further work is needed to test the same combination on real dye-bearing effluents and on other classes of wastewater.

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Author Contribution

PJL, TAS, HK: Investigation and formal analysis, data visualization, writing – original draft; AKS, SP, DT: Conceptualization, supervision, writing – review and editing, project

administration, funding acquisition; HK: Conceptualization, resources, validation, writing – review and editing.

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Data Availability

All data generated or analyzed during this study are presented in this manuscript and available from the corresponding author upon reasonable request.

Ethical Approval Declarations

This study did not involve human or animal subjects or the use of data that requires ethical review.

Conflict of Interests

The authors declare that they have no conflicts of interest to disclose.

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