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Review

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# Microplastic removal by coagulation: a review of optimizing the reaction conditions and mechanisms

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# Abstract

Coagulation is a widely employed technique for removing suspended particles from water and wastewater, and recently, it has gotten attention as a popular method for the removal of microplastics (MPs). Studies on coagulation-based removal of MPs are still in their infancy, and few findings are available about this treatment approach, its mechanism, and removal efficiency. Given these gaps, this study was designed to comprehensively investigate recent advances in the removal of MPs via coagulation. The influence of various experimental factors such as coagulant type, dose of the coagulant, pH of the solution, and shape of the MPs are critically reviewed. The study findings showed that optimizing environmental conditions during the coagulation process is crucial for improving the removal of MPs and reducing energy costs. The study findings showed that the coagulation efficiency of MPs depends on optimal reaction conditions, which may vary depending on the type and



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concentration of MPs and the characteristics of the water or wastewater being treated. Optimizing these reaction conditions is, therefore, critical to achieving maximum removal efficiency. More extensive research is required to reveal the mechanisms of coagulation in controlling floc density and removing pollutants from effluent. Consequently, the current review aims to highlight the gaps and challenges associated with coagulation techniques for the removal of MPs during wastewater treatment. Current advancements in the synthesis and chemical modification of bio-based coagulants and their coagulation performance for the removal of MPs could constitute a paradigm shift in ecosystem protection and sustainability. The use of eco-friendly coagulants and combining coagulation with other techniques are suggested to increase the efficacy and viability of this method. This review will provide significant insights for field researchers, guiding their future investigations and contributing to the advancement of knowledge.

Keywords: Adsorption, charge neutralization, coagulation, microplastics, sweep flocculation

#### INTRODUCTION

Plastic production has gradually increased and reached nearly 400 million tons in 2020 alone<sup>[1]</sup>. Compared to the early 1950s, the current production of plastic has seen a considerable upsurge. Plastic products are widely used by people daily, ranging from single-use items such as packaging and bottles to enduring goods such as electronic devices, furniture, and automobiles<sup>[2-4]</sup>. In addition, the COVID-19 pandemic has exerted significant pressure on the preexisting plastic waste management infrastructure due to overconsumption, extensive production, and inadequate disposal practices pertaining to personal protective equipment (PPE)<sup>[5,6]</sup>. People's preference for using plastic items in their daily lives can be linked to its physiochemical characteristics, which include low cost, durability, reliability, transparency, lightweight, and ease of manufacture and availability. Examples include personal care products, toys, cars, clothing, construction materials, and packaging [Figure 1A and B]<sup>[6,7]</sup>. Following their use, these plastic products weather and degrade due to various physical and mechanical forces, culminating in microplastics (MPs)<sup>[8,9]</sup>. These "tiny" plastic particles first came to the attention of scientists in the 1970s after being identified as a large component of ocean floor debris, and they received considerably greater attention after Thompson et al. coined the term "microplastics"<sup>[10]</sup>. MPs are plastic particles ranging from 100 nm to 5 mm, while particles smaller than 100 nm are termed nanoplastics (NPs)<sup>[11,12]</sup>. The presence of MPs in aquatic and terrestrial ecosystems constitutes a substantial danger to these ecosystems' health and long-term sustainability. Because of the large surface area, persistence, and mobility of MPs in aquatic environments, this hazard extends to human food security and public health<sup>[8,13]</sup>. Thus, developing technologies and strategies for preventing and managing MPs at the upstream and downstream processes are crucial.

Currently, there are several methods available for removing MPs, such as membrane filtration, biodegradation<sup>[14]</sup>, adsorption<sup>[9,15]</sup>, photocatalytic degradation<sup>[16]</sup>, coagulation<sup>[17]</sup>, and electrocoagulation<sup>[18,19]</sup>. These technologies are installed in combination or individually for the efficient and effective removal of MPs from aquatic environments. However, the efficiency of each technique varies according to the MPs' dimensions, form, and chemical composition. Furthermore, some of these technologies are either less efficient or consume more energy. As a result, environmental practitioners are now attempting to develop more effective and efficient MP removal technologies.

Coagulation is a promising technique employed in wastewater treatment plants (WWTPs) to remove suspended particles from the effluent. Recently, it has attracted a lot of scientific interest as a viable method for removing MPs from aquatic environments due to its simplicity, reduced operational costs, low carbon footprint, and eco-friendly features<sup>[13,15,17,20]</sup>. Furthermore, the coagulation technique is more viable for large-scale wastewater treatment applications with less time required for purification as compared to other



Figure 1. (A) MP generation sources and fate in the environment; (B) most commonly generated MPs and their sources. MPs: Microplastics.

conventional methods. Adding synthetic or natural polymers to water during flocculation helps settle unstable particles of MPs by fusing with a flocculant to form bigger microflocs<sup>[7]</sup>. Various inorganic flocculants can remove MP particles by compressing the electric double layer (EDL). However, the low removal efficiency of MPs was reported with common coagulants due to variations in size, density, and shape, as well as inadequate coagulant hydrolysis<sup>[15,21]</sup>. According to Ma *et al.* (2019), the removal efficiencies of FeCl<sub>3</sub>·6H<sub>2</sub>O and AlCl<sub>3</sub>·6H<sub>2</sub>O coagulants were below 12.7% and 36.9%, respectively, for polyethylene (PE) particles with a size range of 0.5-5 mm<sup>[20]</sup>. Recent research by Zhang *et al.* found that Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> had a removal efficiency of less than 2% for MPs (PE and PS) (180 nm - 125  $\mu$ m)<sup>[22]</sup>. The floc adsorption and sedimentation capacities, floc volume, and floc density, as well as the removal of MPs via coagulation, can all be enhanced by the use of coagulant aids<sup>[20]</sup>. The removal efficiency of MPs (PE) (0.5 mm) was increased from 25% to 61% when cationic polyacrylamide (PAM) was added to AlCl<sub>3</sub>·6H<sub>2</sub>O<sup>[20]</sup>. As a result, various experimental conditions must be used to improve the removal efficiency of MPs during coagulation processes.

Coagulation by charge neutralization involves the addition of highly charged cations that neutralize the surface charge on colloidal particles, leading to their aggregation and removal from the water. In this process, the positively charged cations such as Al<sup>3+</sup> or Fe<sup>3+</sup> are attracted to the negatively charged colloid particles and adsorb onto their surface, which neutralizes their surface charge and eliminates repulsive forces between them. As a result, strong van der Waals forces are generated, which cause the aggregation of particles into larger flocs. These flocs have the ability to capture and remove colloidal particles. However, hydrolysis may occur, leading to the formation of insoluble hydroxides, depending on the concentration of the coagulant and the pH of the solution. These hydroxides can contribute to the development of even bigger aggregates, which can then be removed through sweep coagulation<sup>[23]</sup>. To fathom the coagulation mechanisms, it is necessary to investigate the basic principles that govern the interaction between MPs and coagulants or coagulant aids. Recent studies found that the main factor for the flocculation of aluminum

oxide particles and surface-functionalized 2-acrylamido-2-methylpropionic acid polymer nanoparticles was electrostatic interaction<sup>[13,24]</sup>. In addition, Fourier transform infrared spectroscopy (FTIR) research showed that MPs' surfaces had formed new Al-O and Fe-O bonds, which contributed to their enhanced settling<sup>[15]</sup>. It is necessary to conduct additional research on the chemical interactions that take place between MPs and flocs, as well as research into the prevailing factors (Brownian motion or buoyancy force) that determine the settling of MPs. Despite the enormous studies conducted on coagulation technology, there are few literature reviews that comprehensively summarize removal mechanisms along with the gaps and progress on the coagulation technology for eliminating MPs from aqueous solution<sup>[25,26]</sup>. Considering this, the current review has been designed with the aim of (i) assessing the effectiveness of the coagulation technique for MP removal from water and wastewater; (ii) investigating the effect of experimental factors such as coagulant type, characteristics of MPs, and operating conditions on the removal efficiency of MPs via coagulation; (iii) conducting a systematic analysis of the mechanism by which removal of MPs is accomplished by coagulation; (iv) exploring the potential of bio-based and eco-friendly coagulants for the removal of MPs; and (v) highlighting the opportunities and challenges of coagulation for efficient MP removal. The findings of the current study can be used to increase the removal efficiency of MPs from water via coagulation in WWTPs by providing insight into the mechanism by which particle size and water chemical conditions affect the removal of nanosized plastic particles.

# **METHODOLOGY**

The literature review was conducted using the Scopus database, focusing on studies published between 2008 and 2023 [Figure 2]. Some of the significant concepts and definitions that emerged beyond the target time frame were also included<sup>[27]</sup>. The primary search term used was "microplastics", which was combined with other keywords such as "removal", "coagulation, "flocculation", "wastewater treatment", and "management". The search in the database continued until no new relevant studies were found, reaching a point of saturation. Additionally, relevant information was gathered from reputable organizations such as the World Health Organization (WHO) and the National Oceanic and Atmospheric Administration (NOAA). The keyword search covered the period from 2009 to 2023.

Figure 2 shows the trend of MP research over the period from 2009 to 2023, where a significant increase has been observed in the MP publications. Notably, there has been a growing trend in research on microplastics in the past five years, particularly regarding the development of effective approaches for their detection and removal. However, Figure 2 shows that limited research has been conducted regarding MP removal through coagulation and flocculation, which seeks more attention from the research community.

# MP REMOVAL PERFORMANCE THROUGH COAGULATION

The coagulation process is used to remove pollutants and MPs from wastewater. Chemical coagulants such as metallic salt of ferric and aluminum are applied to neutralize the surface charge present on MPs, forming flocs that can then be removed via skimming or settling. Talvitie *et al.* initially found that the sedimentation process of a WWTP effectively removes MPs from influent<sup>[28]</sup>. Perren *et al.* applied electrocoagulation technique initially to remove MPs, thus paving the way for additional research into MP removal using coagulation flocculation<sup>[18]</sup>. The study carried out by Xue *et al.* mentioned that adding 30 mg/L of aluminum sulfate  $[Al_2(SO_4)_3]$  resulted in the removal of 75.6% and 85.2% of 6 µm polystyrene (PS) microspheres from Ria Grande and Lake Erie water<sup>[29]</sup>. According to Zhou *et al.*, the removal efficacy of aluminum-based coagulants was higher than that of iron-based coagulants<sup>[15]</sup>. The removal of polyethylene (PE) by poly aluminum chloride (PAC) and FeCl<sub>3</sub> at a dose of 90 mg/L was reported to be 29.7% and 17%, respectively. It has been determined that PAC has a higher effect on neutralizing charges than FeCl<sub>3</sub>. The removal efficiency of



Figure 2. Annual publication trend for MPs research from 2009 to 2023. The data were collected from the Scopus database between 2009 and 2023. MPs: Microplastics.

FeCl<sub>3</sub>, PAC, and polyamine in removing MPs smaller than 10 µm in actual municipal wastewater was studied by Rajala et al.<sup>[17]</sup>. The size of the MPs affected their removal, with 1 and 6.3 µm MPs showing removal rates higher than 95% and 76%, respectively. This suggests that the effectiveness of the treatment method may depend on the size of the MPs present in the wastewater. Wang et al. investigated the impact of several MP removal treatment procedures<sup>[30]</sup>. The results show that the coagulation and sedimentation approach brought the MP concentration down from  $6,614 \pm 1,133$  to  $3,472 \pm 502$  MPs/L. The overall removal efficiency for all MPs ranged from 40.5% to 54.5%, while the total removal efficiency of water plants ranged from 82.1% to 88.6%. Pivokonský et al. conducted a study at the drinking water treatment plant (DWTP) Plzeň in the Czech Republic to evaluate the removal of MPs in the water treatment process<sup>[31]</sup>. Their findings showed that coagulation and sedimentation were the most effective methods, removing 62% of MPs. Meanwhile, only 20% and 6% of MPs are removed by the employed technique of deep filtration and granular activated carbon (GAC), respectively. Cherniak et al. conducted a study that showed the effectiveness of coagulation, flocculation, and sedimentation methods in removing 71% of full-sized MPs from raw water compared to untreated water<sup>[32]</sup>. However, less than 1% of the MPs were removed due to filtration. Dalmau-Soler et al. found that 78% of MPs could be removed during the first stage of water treatment, where the employed treatment technique is reported as coagulation, sedimentation, and sand filtration<sup>[33]</sup>. This is the highest recorded efficiency for this type of water treatment method. However, it is important to note that the sand filtration step also contributes to the removal of MPs, so the actual effectiveness of the coagulation-sedimentation process alone may be lower. Hidayaturrahman and Lee studied three WWTPs to evaluate the removal of MPs at various stages of treatment<sup>[34]</sup>. Compared to the primary and secondary treatments (56.8%-64.4%), the removal efficiency of MP coagulation in tertiary treatment increased from 47.1% to 81.6%. Additionally, Ruan et al. reported that 78.2% of MPs were removed through coagulation sedimentation, whereas sedimentation treatment alone only removed 41.7% of MPs<sup>[35]</sup>.

A study by Wang *et al.* investigated the efficiency of coagulation for removing MPs from wastewater<sup>[36]</sup>. In the study, the removal efficiency of MPs was found to be 96.1% when ferric chloride (FeCl<sub>3</sub>) was added at an optimal dose of 50 mg/L. A study on advanced WWTPs used dissolved air flotation (DAF) cells with aluminum- and iron-based coagulants to remove PE from wastewater. The results indicated that the Al-based coagulant reduced PE by up to 96.10% more than the Fe-based coagulant, which reduced PE by up

to 70.56%<sup>[37]</sup>. During water treatment, flocculants have been widely used to improve coagulation<sup>[38,39]</sup>. Overall, these studies demonstrate the efficiency of coagulation for MP removal from water bodies. The selection of the appropriate coagulant and the optimal dosage may differ depending on the properties of the aqueous medium and the characteristics of the MPs being treated.

Research results indicate that the use of flocculant aids can improve the removal efficiency of MPs. Studies suggested that MP removal efficiency was improved with the addition of coagulant aids. The removal of MPs (PE smaller than 0.5 mm) by an Al-based coagulant (135 mg/L), including anionic and cationic PAM at pH 7, was investigated by Ma *et al.*<sup>[40]</sup>. The removal efficiency with the addition of 15 mg/L anionic PAM increased from 27% to 64%, as shown by the results. 45.4% of the PE was removed when cationic PAM was added at 15 mg/L, and 61.4% was removed when anionic PAM was added at the same dose. The results showed that anionic PAM was more effective than cationic PAM in removing PE. Shahi et al. found that the treatment of polyethylene MPs (d < 0.5 mm) with 20 mg/L alum and 500 mg/L PC sand resulted in a removal efficiency of 92.7%<sup>[41]</sup>. This was significantly higher than the removal efficiency achieved with sole alum (65.9%). Furthermore, ultrafiltration contributes to MP removal by assisting coagulation. After coagulation removal, it was observed that the remaining polyethylene MPs were unable to pass through a membrane with an average pore diameter of 30 nm, likely due to their significantly larger particle size<sup>[40]</sup>. Nonetheless, membrane fouling was observed as a result of the flocs that were rejected, which quickly formed a soft, cake-like layer on the membrane surface. Furthermore, FeCl<sub>3</sub>·6H<sub>2</sub>O dosage positively influenced the level of membrane fouling. Zhang et al. used a combination of PAM and PAC to remove polyethylene terephthalate (PET), and also determined that a 91.45% removal efficiency was achieved with a high PAM dosage<sup>[42]</sup>. This study suggests that combining these two chemicals can efficiently remove PET from water.

Separating solids from liquids or sorting particles based on their density or size are both possible with the help of a hydrocyclone, a device that works on the principle of centrifugation<sup>[43]</sup>. If the apparatus design parameters are changed, hydrocyclones can be used in other unit processes. In water resource recovery facilities (WRRFs), hydrocyclones can be used for various purposes, such as grit separation, sludge thickening, digester cleaning, and electromagnetic particle separation<sup>[44]</sup>. Since the operation of a hydrocyclone primarily depends on apparatus design criteria and operational parameters, specifically feed flow, the hydrocyclone is thought to have low energy demands. Energy consumption is limited to moving water through the hydrocyclone, as it is a single-bodied vessel with no moving parts<sup>[45]</sup>. Hydrocyclones also have a significant role in the removal of MPs. However, according to Kikuchi et al., earlier attempts primarily concentrated on influential MPs (239 m-4 mm)<sup>[46]</sup>. Hydrocyclones with a 10-mm diameter were used by Liu *et al.* to remove polyamide and low-density polyethylene particles<sup>[47]</sup>. In order to filter out MPs of a smaller size, mini-hydrocyclones with diameters of less than 15 mm have recently been proposed<sup>[48]</sup>. The effectiveness of a mini-hydrocyclone in removing PMMA (polymethyl methacrylate) particles with a diameter of 10 m was studied by He et al.<sup>[48]</sup>. The sedimentation process was initially used in WWTP for the removal of MPs; later, the electrocoagulation method was used by some researchers. Effective removal of MPs was observed using coagulation, flocculation, and sedimentation techniques. The removal efficiency of MPs in the tertiary treatment process is higher than in the primary and secondary treatment processes. Aluminum-based coagulants have a higher coagulation efficiency than iron-based coagulants. Additionally, it was discovered that the coagulant aids increased the effectiveness of the removal of MPs. For instance, adding PC sand to alum improved the removal efficiency of MPs. The size of the MPs also has an impact on MP removal efficiency as shown in Table 1.

# Table 1. MP removal efficiency of various coagulants

Type of media	MPs polymer type	MPs size	Coagulant	Dosage	pH range	Removal efficiency	Ref.
Simulated water	PE	≤270 µm	$Mg(OH)_2$ and $Fe_3O_4$	Mg(OH) <sub>2</sub> (50-250 mg/L), Fe <sub>3</sub> O <sub>4</sub> (40-200 mg/L)	7	98%	[49]
Wastewater	Polystyrene	1 and 6.3 μm	Ferric chloride, polyaluminum chloride and polyamine	0.017 and 1.4 mmol/L	6.5	99.4%	[17]
Ultrapure water + humic	PE	< 500 µm	Ferric chloride, polyacrylamide	0.25 and 46.4 mg/L	7.3	98.20%	[20]
acids + kaolin				0.2 mmol/L	7	13.27% ± 2.19% to 89.32%	
Ultrapure water + humic acids kaolin	PE	< 500 µm	Aluminum chloride, ferric chloride, polyacrylamide	15 mM	6-8	61%	[40]
Deionized water	PP	0.25 mm	Polyaluminium chloride	PACI concentration of 200 ppm, PAM concentration of 21 ppm	5-9	$18.00\% \pm 1.43\%$ and $19.69\%$	[50]
Deionized water	Polystyrene	50-1,000 nm	Aluminum chlorohydrate and polyacrylamide	400 PAC and 20 mg/L PAM	6-9	98.50%	[13]
Drinking water	Polyethylene	0.1 mm and 15 μm to 0.9 and 1.3 mm	Aluminum sulfate	0 and 43.2 mg/L	4-7	-	[51]
Deionized water and river water	Polystyrene	10-90 μm	Aluminum chloride hexahydrate	0 to 10 mg/L	4.3, 6.0, and 8.5	33%-95.3%	[52]
Low-density polystyrene	PS	100 µm	Aluminium sulphate	3.4 mg Al/L	5	$98.9 \pm 0.94\%$	[53]
Synthetic water	PE	10-100 μm	Alum coagulant and alum combined with cationic PC sand	30	7 ± 0.5	70.70%	[41]
Synthetic stormwater	LDPE, HDPE, PP	-	Alum and PAM	50-250 and 5-25	3-5	92%, 84% and 96%,	[54]
Water	PET	100-400 μm	PAC, PAM, sodium alginate and activated silicic acid	PAC 200 mg/L, PAM 100 mg/L	3-9	91.45%	[42]
Water	PET	100-400 μm	Polyaluminum chloride, and activated silicic acid	PAC 200 mg/L, ASA 100 mg/L	3-9	73.35%	[42]
Wastewater	PE	≤270 μm	Magnetic magnesium hydroxide and PAM	Mg(OH) <sub>2</sub> 200 mg/L, PAM 4 mg/L	5-9	87.1%-92%	[55]
Deionized water	PE, PS	< 500 µm	PAC	30 to 180	7	29.70%-77.83%	[15]
Ultrapure water	PET/weathered PET	500 ± 2.5 nm	FeCl <sub>3</sub> AlCl <sub>3</sub>	0.01-0.20 mmol Al/L	6-8	92%-100%	[56]
Distilled water	Polystyrene beads	0.5 μm (45-53 μm)	Plant-derived tannic acid	-	6-8	> 97% for PS	[57]
Drinking water	Polyethylene and PS microspheres, and PEST fibers	15-140 μm	Alum and aluminum chlorohydrate	(0.45-3.64 mg Al/L) and PAM (0.05-0.30 mg/L)	7	97% and 99% were measured for PEST and PE, respectively	[21]
Wastewater	PE	300-355 μm	Iron electrodes	-	7.5	90%-99.24%	[18]
Real surface water, deionized water	PS-COOH	50 nm	PAC, $\operatorname{FeCl}_3$ and $\operatorname{AlCl}_3$	PAC (2.5-40 mg/L), AICl <sub>3</sub> (2.5-40 mg/L), FeCl <sub>3</sub> (2.5-40 mg/L)	3-10	96.60%	[58]
Lake water	Amidine PS	110 ± 25 nm	PAC	10 mg/L	3.0 ± 0.2 to 11.0 ± 0.2	-	[59]

Lake water, bottled mineral water	PS	90 nm	PAC and $\operatorname{FeCl}_3$	PAC (0-4 mg/L), FeCl <sub>3</sub> (0-10 mg/L)	3.0 ± 0.2 to 11.0 ± 0.2	-	[59]
Lake water	PS	100 nm	$AICI_3$ and $CaCI_2$	10 mg/L	3-9	-	[7]
Lake water, WWTP effluent	Carboxylated PS	500 nm	Lysozyme amyloid fibrils	10 g/L	5-8	-	[60]
Tap water	PS and PE	PS 53-500 μm	FeSO <sub>4</sub>	FeSO <sub>4</sub> (496-993 mg/L)	6-8	PS-92.4	[61]
Park pond water	PS	PE 500-1,000 μm	FeCl <sub>3</sub>	FeCl <sub>3</sub> (483-964 mg/L)	-	PE-72.1	[53]
		100 µm	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	10 mg/L		$98.9 \pm 0.94$	

ASA: Activated silicic acid; HDPE: high-density polyethylene; LDPE: low-density polyethylene; MPs: microplastics; PAC: poly aluminum chloride; PAM: polyacrylamide; PACI: polyaluminum chloride; PC: polyaminecoated; PE: polyethylene; PEST: polyester; PET: polyethylene terephthalate; PP: polypropylene; PS: polystyrene; PS-COOH: carboxyl-modified polystyrene; WWTP: wastewater treatment plant.

The density of the MPs smaller than 5 mm may be lower than the water in which they are suspended, making the MP removal through coagulation challenging. MPs can be removed from water using coagulation. The efficiency of the removal of MPs in water treatment facilities can be enhanced by coagulation in combination with other treatment processes. Numerous factors can affect the removal of MPs through coagulation. The efficiency of coagulation in MP removal is size-dependent. Coagulation removes larger MPs better because they form flocs and settle out of the water. The coagulant and its dosage must be carefully chosen. Some coagulants effectively neutralize the charges on MPs and encourage their agglomeration into flocs. Adjusting coagulant dosage and pH may improve performance. Larger and denser flocs are more likely to form with proper mixing and flocculation. It is vital to mix gently to prevent the breaking up of delicate flocs containing MPs. The MPs in flocs may settle after coagulation and flocculation. The larger and denser flocs can be formed by promoting mixing and flocculation. MPs in flocs may settle. Depending on the size and density of the MPs, sand or membrane filtration may be required to capture smaller suspended particles. Some water treatment facilities may use advanced treatment technologies to improve MP removal, such as granular activated carbon (GAC) adsorption or advanced oxidation processes (AOPs), due to the challenges associated with MP removal.

# COMMONLY USED COAGULANTS

Table 2 demonstrates the use of various coagulants for the removal of MPs in water and wastewater treatment. Several inorganic coagulants such as aluminum and iron-based coagulants, MgCl<sub>2</sub>, magnetic coagulants, as well as organic polymer coagulants and bio-flocculants have all been used for MP removal from water and effluent<sup>[62]</sup>. It has been found in previous studies that aluminum-based coagulants perform better than iron-based coagulants in MP removal<sup>[15,37,40,52]</sup>. Zhou *et al.* found that at a concentration of 90 mg/L, polyaluminum chloride (PAC) and ferric chloride (FeCl<sub>3</sub>) were found to remove approximately 29.7% and 17% of polyethylene, respectively<sup>[15]</sup>. Similar results were also obtained by Ma *et al.*<sup>[40]</sup>. In their study, they assessed the removal efficiency of FeCl<sub>3</sub>·6H<sub>2</sub>O and AlCl<sub>3</sub>·6H<sub>2</sub>O for the removal of PE and found that AlCl<sub>3</sub> was more effective for the removal of PE during coagulation. Wang *et al.* reported a contrary finding, demonstrating that FeCl<sub>3</sub>·6H<sub>2</sub>O was more effective than PAC in removing MPs<sup>[36]</sup>. In all circumstances where the dosage was between 5-35 mg/L, FeCl<sub>3</sub>.6H<sub>2</sub>O eliminated between 5.9% and 73.1% of MPs, whereas PAC removed only 3.5% to 53.6% of MPs. Tang *et al.* found that the PS

Coagulants	Chemical formula	Merits	Demerits
Aluminum sulfate	Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ·18H <sub>2</sub> O	Simple to use and apply; creates less sludge than lime; most efficient between pH $6.5$ and $7.5$	Water is treated in the form of dissolved solids (salts), which makes it effective throughout a narrow pH range
Sodium aluminate	Na <sub>2</sub> Al <sub>2</sub> O <sub>4</sub>	Efficient in hard water; usually, only a small dose is needed	Frequently used with alum; expensive; unreliable in soft waters
PAC	AI <sub>13</sub> (OH) <sub>20</sub> (SO) <sub>4</sub> CI <sub>15</sub>	In certain applications, the floc yielded is denser and settles faster than alum	Rarely used; little full-scale data, unlike other aluminum derivatives
Ferric sulfate	Fe <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	Efficiently work between pH 4-6 and 8.8-9.2	Add dissolved solids (salts) to water; alkalinity is usually required
Ferric chloride	FeCl <sub>3</sub> ·6H <sub>2</sub> O	Effective within a pH range of 4 to 11	Adds dissolved solids (salts) to water, and it requires twice as much alkalinity as alum
Ferrous sulfate	FeSO <sub>4</sub> ·7H <sub>2</sub> O	Less sensitive to pH than lime	Adds dissolved solids (salts) to water; usually requires alkalinity
Lime	Ca(OH) <sub>2</sub>	Commonly used; very effective; no salts added to effluent	Dependent on pH, can generate significant amounts of sludge, and an overdose can lead to poor-quality effluent
Aluminum chloride hexahydrate	AICI <sub>3</sub> ·6H <sub>2</sub> O	Aluminum ion has been used as a highly effective flocculant in sewage treatment Low cost, great efficiency, and low toxin levels	pH-dependent
Anhydrous calcium chloride	CaCl <sub>2</sub>	Under high pH conditions, calcium ion also demonstrates outstanding sedimentation performance for impurities Low cost, high efficiency, and low toxicity	pH-dependent
Magnetic magnesium hydroxide	Mg(OH) <sub>2</sub>	Highest removal efficiency for PE	Need to be tested for other types of MPs
Chitosan	$C_{18}H_{35}N_3O_{13}$	Good removal efficiency Additionally, it is an eco-friendly coagulant without harmful impacts	The economic viability of designing large-scale treatment facilities still needs to be determined. Need to understand the flocculation mechanism
Chitin Graphene oxide sponges	-	High elasticity enables them to retain their high porosity and achieve a stable, high absorbent capacity during recycling	Water insolubility of chitosan

#### Table 2. Common coagulant for the removal of MPs

MPs: Microplastics; PAC: polyaluminum chloride; PE: polyethylene.

microspheres used in the experiments lacked a charge, so wrap sweeping was more effective than charge neutralization in removing MPs<sup>[62]</sup>. Different mechanisms of action are expected to affect the removal efficiency of MPs due to factors affecting the coagulation process. Due to changes in the mechanism of action when the same coagulant is used under different conditions, aluminum-based coagulants cannot be assumed to be more effective than iron-based coagulants.

Lee and Jung found that as salinity increased from 3% to 15%, the MP removal efficiency of aluminum trichloride, aluminum sulfate, and aluminum chloride decreased from 71.6% to 64.3%<sup>[63]</sup>. On the other hand, the removal efficiency of MPs increased from 63.1% to 79.1% through aluminum sulfate. Water with a salinity of 15% is thought to inhibit the hydrolysis of Al ions in aluminum trichloride due to the abundance of Cl ions in the water. The number of charged

MPs, the rate of MP degradation, and the types of water ions vary between water bodies due to the complexity of actual water. As a result, the ability of an inorganic salt coagulant to remove MPs may vary. Zhang *et al.* employed a combination of magnetic Fe<sub>3</sub>O<sub>4</sub> particles and the formation of magnesium hydroxide for PE removal<sup>[55]</sup>. The researchers changed the Mg<sup>2+</sup> to OH ratio during the formation process to form three magnetic magnesium hydroxides (MMHCs). There was a significant increase in MP removal efficiency (87.1%) when MMHCs were prepared with a 1:1 ratio of Mg<sup>2+</sup> to OH. As a potential method for MP removal from water bodies, layered double hydroxides (LDH) formed by divalent and trivalent metal cations have been investigated in other studies. According to Huang *et al.*, LDHs may be particularly effective at removing negatively charged MPs due to their ability to adsorb such particles via the positive charge generated on their surface during coagulation<sup>[64]</sup>. The salinity of water efficiently reduced the removal efficiency of MPs.

The composite metal calcium-aluminum (Ca/Al) ions were used as a coagulant to remove PS NPs (100 nm) from water in a study by Chen *et al.*<sup>[7]</sup>. Using this coagulant allowed the researchers to remove the NPs with an efficiency of up to 80%, and they discovered that it worked best at pH values under 5. At this pH, the NPs were unstable and aggregating because the negatively charged surface layer, or double electric layer, of the NPs was disrupted by the positively charged calcium, aluminum, and hydrogen ions in the coagulant. As the pH rose, calcium and aluminum ions precipitated and became trapped inside the NPs, forming separate or collective crystals that helped remove them. Similarly, Mg/Al LDH was employed. Similarly, Chen *et al.* removed PS NPs using Mg/Al LDH as a coagulant<sup>[65]</sup>. Removing PS NPs from water was more effective with a Mg/Al double flocculation system than with a single Mg or Al flocculation system. This indicates the possibility of using multiple coagulants to increase removal efficiency. Previous research demonstrates promising methods for removing NPs from water and highlights the importance of pH for coagulant performance.

Coagulants made of organic polymers have a high molecular weight and consist of strings of similar units (monomers) grouped together through covalent bonds. Based on their ionic composition, these coagulants are anionic, cationic, or nonionic<sup>[66]</sup>. These organic coagulants can cause particle aggregation by charge neutralization or adsorption bridging as a result of hydrolysis because of their high molecular size surface charge properties. Organic polymer coagulants are used to remove MPs, and some of these include PAM, polyamines, n-butyl trichlorosilane, and diallyl dimethylammonium chloride<sup>[67]</sup>. For the elimination of MPs, these organic polymer coagulants can be employed by adding them directly to water bodies. Lee and Jung extracted MPs from simulated marine water using silane-based coagulants<sup>[63]</sup>. They found that these coagulants maintained their removal efficiency even at high salinities, reaching > 93%, and performed better than iron- and aluminum-based coagulants in seawater. A cationic polymeric polyamine substance named C-577 was used in a study by Rajala *et al.* to remove MPs from samples of WWTP effluent<sup>[17]</sup>. The removal of MPs from the C-577 system reached a maximum of 65%, and the researchers found a connection between this removal rate and the system's absolute zeta potential value. This shows that organic polymeric coagulants may cause the removal of MP via charge neutralization to become unstable<sup>[62]</sup>. PAM has been found to significantly enhance the removal of MPs by aluminum and iron-based coagulants, with anionic PAM being more effective than cationic PAM<sup>[20,40]</sup>. According to Jarvis *et al.*, the particle size and density of flocs are increased by PAM, resulting in improved settling performance of the flocs<sup>[68]</sup>. Additionally, Ma et al. noted that aluminum-based and iron-based flocs carry a positive charge at pH 7.0, allowing them to bond with anionic PAM, resulting in the formation of larger flocs that can capture a higher number of MPs for settling<sup>[40]</sup>. MPs can also be effectively removed by using nonionic PAM. Zhang et al. found that a PAC concentration of 400 mg/L combined with a nonionic PAM concentration of 20 mg/L resulted in a 98.5% removal rate for 500 nm PS NPs<sup>[13]</sup>. The PAM hydrolyzing well in water contains a -CH<sub>2</sub>-CH-C(O)-NH<sub>2</sub> group, which helps in the adsorption of negatively charged particles of MPs (PS) and flocs through the bridging mechanism, thus accounting for the high removal rate. Furthermore, the PS NPs can be attached to the PAC/PAM flocs via the newly formed Al-O bond, making PS NP removal easier. For the purpose of removing PET MPs, the removal efficiency of PAC in combination with PAM and activated silicic acid (ASA) was investigated by Zhang et al.<sup>[42]</sup>. The use of PAC alone resulted in a removal rate of only 35.50%, while the addition of any of the three coagulants significantly enhanced the removal efficiency. The conventional dosage of ASA resulted in a significant increase in coagulation effect, leading to a 19% increase in removal efficiency. On the other hand, PAM showed the most significant improvement in the coagulation effect at the high dosage, resulting in a 56% increase in removal efficiency. Shahi et al. found that the use of alum alone resulted in the removal of 65.9% of polyethylene (PE)<sup>[41]</sup>. However, when alum was combined with cationic polyamine-coated sand (PC-sand) at a concentration of 500 mg/L, the removal rate significantly increased to 92.7%. This was observed when 20 mg/L of alum was combined with PC-sand. Due to their useful properties, hydrophobic organic coagulants derived from inorganic coagulants have become increasingly popular in wastewater treatment. Natural and synthetic organic polymers can be distinguished, with natural polymers being non-toxic, abundant, and eco-friendly, whereas synthetic polymers have the opposite characteristics. They are frequently combined with inorganic coagulants to increase their effectiveness because they cannot always be adjusted to meet specific requirements. Synthetic polymers can be optimized during production for broader utilization, but some may have toxic properties. Organic polymers come in various forms, including linear, branched, and cross-linked structures<sup>[67]</sup>.

Natural biological coagulants are gaining popularity as a green alternative to inorganic coagulants. Polysaccharide materials such as starch, chitosan, and cellulose are the main sources of these coagulants. Natural coagulants have functional groups that neutralize negatively charged MPs as a result of their macromolecular structure. Despite being less researched compared to their inorganic counterparts, limited studies have demonstrated the efficient ability of natural coagulants to remove MPs. The efficiency of a new natural biological coagulant in the removal of PS MPs was investigated by Peydayesh et al.[60]. The coagulant, lysozyme amyloid fibrils, were formed by hydrolyzing and reassembling the lysozyme monomer at elevated temperatures. Under the same conditions, the PS MPs were more efficiently removed by the lysozyme amyloid fibrils than by either natural lysozyme monomers or conventional metal coagulants. With a removal efficiency of 98.2%, lysozyme amyloid fibrils remove PS MPs via charge neutralization. This study highlights the potential of natural biological coagulants as effective alternatives for the removal of MPs. Park et al. modified the surface of PS microspheres with chitosan and tannic acid to create surface phenolic MPs<sup>[57]</sup>. The flocculation of MPs and flocs was then facilitated by the addition of a metal coagulant using coordinate bonds between metal and phenol. The results demonstrated that the combination of chitosan and tannic acid improved removal efficiency by 49% compared to using a metal coagulant. Natural polymer chitosan is safe for various applications because it is non-toxic. In water purification process, it has been utilized to treat waste from food processing, remove metal ions, and condition sludge. Additionally, it has been utilized to remove the color from the effluents of dye houses. Investigating environmentally friendly coagulants that are effective at removing MPs without negatively impacting the environment through sludge production is essential because excessive use of inorganic coagulants could lead to the generation of toxic sludge.

Water source, MP characteristics, and treatment goals determine the coagulant. Alum is widely used as a coagulant in water treatment and is frequently used in MP removal. When added to water, it forms flocs of aluminum hydroxide. MPs can be trapped and removed with the help of these flocs. Additionally, ferric chloride, a common coagulant, can effectively remove MPs. MPs and other suspended particles can be removed because they form dense flocs. Due to its higher charge density and better performance in some

water conditions, poly aluminum chloride (PAC) is sometimes preferred over traditional aluminum sulfate or ferric chloride as a coagulant. The effectiveness of primary coagulants can be improved by using polyacrylamides (PAM) as coagulant aids. They can enhance the formation and sedimentation of flocs, which can aid in removing MPs. The primary purpose of calcium hydroxide (lime) in water treatment is to adjust the pH, but it can also act as a coagulant and aid in MP removal under certain conditions. Coagulant aids are used to remove MPs, and some organic polymers, such as cationic polymers, are used for this purpose. They can enhance flocculation by filling in the spaces between the particles. Other specialized coagulants or coagulant blends may be used to achieve the best results, depending on the particular water quality and the characteristics of the MPs. Removing all MPs by coagulation alone may not be possible, especially for the smaller particles. Coagulation may need to be combined with additional treatment steps like adsorption and advanced oxidation to remove MPs from water sources effectively.

# FACTORS AFFECTING COAGULATION

The coagulation of MPs from wastewater is a complex process influenced by multiple factors such as coagulant choice, coagulant dose, pH of the water, mixing and flocculation, MP characteristics, interfering substances such as organic matter, and contact time [Figure 3].

# Solution pH

As shown in Table 1, the solution pH has a vital influence on the removal of MPs through coagulation. According to Sillanpää et al., pH controls the hydrolysis of coagulants and the characteristics of flocs, which is a key factor in coagulation<sup>[69]</sup>. Shen et al. found that coagulation efficiency is affected when pH is changed from 6 to 9, which applies to the majority of wastewater scenarios<sup>[70]</sup>. Removal efficiencies of PSNPs were found to be significantly higher at pH 8.0 (90.7%  $\pm$  0.7%) and pH 9.0 (93.8%  $\pm$  0.3%) in 30 min compared to pH 6.0 and 7.0<sup>[13]</sup>. The removal efficiency of MPs at pH of 7.3 and 6.5 was raised when the coagulant dosage was increased<sup>[17]</sup>. However, pH had a minor effect on removal performance, and there was a slight decrease in removal efficiency in the PAC system when the pH was low (pH = 3), as reported by Zhang et al.<sup>[42]</sup>. The efficiency of aluminum sulfate nanoparticles was not affected when the pH of the solution ranged from 3 to  $9^{[71]}$ . Similarly, when pH changed from 6 to 10, it was found that there was not too much effect on color removal using ferric chloride APAM<sup>[72]</sup>. The removal of hydrophobic MPs ranged from 54% to 91% at acidic (pH 1-5) conditions. The removal of MPs was only 70% after adjusting the pH to > 6.8 and using  $Al_2(SO_4)_3$ as flocculant. Aluminum sulfate has a high surface potential when the pH is adjusted to 5, which favors the removal of hydrophobic MPs<sup>[73]</sup>. Comparing the removal efficiencies at pH 6, 7, and 8, the maximum removal efficiency was observed at pH  $6^{[20]}$ . In addition, the effect of pH on PE and PS elimination by PAC was studied. PS and PE removal efficiency were relatively constant between pH 5 and 8, but significantly increased at pH 9. With increased floc size, MPs sweep, and sedimentation, alkaline conditions accelerate PAC hydrolysis<sup>[42]</sup>. Ma et al. observed a clear trend in PE removal using AlCl<sub>3</sub>·6H<sub>2</sub>O<sup>[40]</sup>. As the solution pH increased from 6 to 8, the removal efficiency of PE, regardless of size, decreased at a high dosage of AlCl<sub>2</sub>·6H<sub>2</sub>O (5 mmol/L). The surface charge of MPs is affected by pH. The negative charge on the surface of the particles increases as pH increases<sup>[74]</sup>. Initially, the zeta potential of the solution of MPs was -47.9 mV, but it decreased to -54.7 mV as the pH increased<sup>[42]</sup>. As a result, as the pH rises, the coagulant's ability to neutralize the charge on MPs decreases. If sweeping flocculation is unsuccessful, the zeta potential of the MPs in the system may shift from zero potential, which was achieved after coagulation<sup>[37]</sup>. pH also affects the particle size of flocs, with larger flocs in alkaline conditions better suited for netting and sweeping than those in acidic conditions<sup>[15]</sup>. Smaller, more dispersed flocs form at pH 6 and 7, while larger, denser flocs form at pH 8 and 9.



Figure 3. Effect of various factors on the removal of MP efficiency (A) MP size; (B) pH; (C) typical ions; (D) dissolved organic matter<sup>[42]</sup>. MP: Microplastic.

#### Inorganic ions

The performance of coagulation may be impacted by the presence of several types of inorganic ions in natural water. This is because coagulants' stability and ability to remove contaminants might be affected by interactions between the ions and coagulants. Therefore, the efficiency of coagulation can be influenced by the concentration and composition of ions in the water<sup>[26]</sup>. Comparing the removal efficiency of MPs when using PAC and FeCl<sub>3</sub>, researchers found that  $SO_4^{2^\circ}$  and  $CO_3^{2^\circ}$  have a greater impact on the removal of MPs than Cl<sup>-</sup> ions<sup>[15,40]</sup>. Lee and Jung<sup>[63]</sup> found that a high Cl<sup>-</sup> concentration (up to 15% salinity) reduced the MP removal efficiency of AlCl<sub>3</sub> coagulant. Additionally, Al<sup>3+</sup> hydrolysis in AlCl<sub>3</sub> is inhibited by the high Cl<sup>-</sup> concentration. Furthermore,  $CO_3^{2^\circ}$  and  $HCO_3^-$  can increase coagulant hydrolysis, floc volume, and specific surface area, which can increase the removal efficiency of MPs<sup>[15,55]</sup>. Typical ions like Cl<sup>-</sup>,  $SO_4^{2^\circ}$ , and  $HCO_3^-$  were studied for their effects on coagulation. In the PAC system,  $SO_4^{2^\circ}$  serves as a bridge<sup>[75]</sup>. The HCO<sub>3</sub><sup>-</sup> in the PAC-PAM system hydrolyzed PAC and PAM, forming flocs with a larger volume and surface area that settled better<sup>[55]</sup>. SO<sub>4</sub><sup>2-</sup> reduced the positive charges of the hydrolysate and made the metal coagulants unstable, reducing the efficiency of PAC coagulation to remove PS and PE MPs<sup>[76]</sup>. SO<sub>4</sub><sup>2-</sup> has also been reported to inhibit MP removal by Zhou *et al.*<sup>[15]</sup>.

#### MP size and type

Recent research suggests that the removal effect varies with the MP type, which was confirmed by the researchers<sup>[15,21,61]</sup>. Different MPs have different densities, which might lead to an effective coagulation process in terms of settling efficiency<sup>[21,61]</sup>. The impact of MP particle size on collision efficiency and settling behavior affects the performance of coagulation removal<sup>[13]</sup>. Several studies have found that larger MPs particles are more efficiently removed<sup>[13,17,41]</sup>, while some experts feel that smaller MPs can be eliminated more easily than larger MPs<sup>[20,21,26]</sup>. The existing analysis regarding the effect of MP size on their removal efficiency needs to be more consistent. Different coagulation mechanisms may be the primary driving force

behind MPs. Zhang et al. found that flocs adsorb enveloped particles were essential to remove large-size MPs<sup>[13]</sup>. Charge neutralization and adsorption effect of coagulants help in the removal of small-sized MPs. The removal efficiency of MPs with larger particle sizes increases when charge neutralization becomes the dominant mechanism during the coagulation process. Compressive double-layer effects make large MPs more unstable and prone to aggregation than small particles<sup>[42]</sup>. When sweep flocculation dominates coagulation, MP size decreases, which also reduces the removal efficiency. Ma et al. conducted a study in which they discovered that smaller PE particles facilitate the capture and removal of particles by flocs<sup>[20]</sup>. A floc of a given size likely captures smaller particles more frequently<sup>[21]</sup>. The particles that can be wrapped are larger if the floc is larger<sup>[77]</sup>. Larger MPs can only be effectively removed by flocs that are of a similar size and bulk, whereas flocs of varying particle sizes can remove smaller MPs. According to Xue et al., MPs with 3 µm size were easier to remove than 45 and 90 µm MPs<sup>[29]</sup>. Most of the studies found that MP particle size increased the removal efficiency of MPs<sup>[30,78,79]</sup>. The main reasons are: (i) Hydraulic mixing may fragment large MPs during coagulation. Therefore, the removal efficiency of large-size MPs must be increased to increase the abundance of small-size MPs<sup>[78]</sup>; (ii) The ability of larger MPs to attach to flocs increases their possibility of settling in water<sup>[30]</sup>. Due to their higher flocculation rate and significant Brownian motion during flocculation, smaller MPs are easier to remove<sup>[80]</sup>. During coagulation, the MP shape is more likely to be adsorbed on the floc surface, so fibers are removed with the highest efficiency  $(50.7\%-60.6\%)^{[30]}$ . The reason for the lower removal rates of polypropylene (PP) and PE, which have a density of less than 1 g/cm<sup>3</sup>, is their tendency to float on the surface of water. In contrast, polyvinyl chloride (PVC) and VINYON, which have a density greater than 1 g/cm<sup>3</sup>, have removal rates of 82.66% and 75.01%, respectively<sup>[79]</sup>. PET accounts for 55.4% to 63.1% of MPs in raw water, is fibrous, and has high removal rates<sup>[30,81]</sup>. Thus, laboratory research is needed to determine the removal efficiency of various particle sizes of MPs via coagulation and sedimentation.

#### **MP** shape

The shape of MPs can also influence their removal efficiency through coagulation. Shahi *et al.* found that coagulation was the best method for removing fibrous MPs in laboratory analyses<sup>[41]</sup>, consistent with water treatment plant findings<sup>[30]</sup>. Fibrous MPs are more likely to combine with flocculants despite having the same volume due to their higher specific surface area. Compared to smooth MPs, the removal efficiency of MPs with a rough surface is high<sup>[41]</sup>. This is because adsorption bridging is more prominent on rough surface MPs than smooth ones, as the rough MP has a higher adsorption capacity<sup>[82]</sup>. Rough MPs have more asymmetric surfaces compared to smooth MPs, which results in the application of different forces on their surfaces. The alignment of the Feret's diameter of asymmetric MPs along the flow direction can lead to increased aggregation of asymmetric particles under certain flow conditions<sup>[62,83]</sup>.

#### **Coagulant dose**

The coagulant dose determines MP removal and disinfection by-product components distribution. Sibiya *et al.* compared three coagulants, alum (A), eggshells (E), and ferromagnetite (F), and their dosages (FA, FE, and FEA) to find a cost-effective coagulant for industrial wastewater treatment<sup>[84]</sup>. The 10-20 mg/L dosage reduced colloidal agglomeration and destabilization. Other studies have also shown that co-agglomeration saturation decreases or stabilizes performance after increasing coagulant dosage<sup>[74,85]</sup>. Yang *et al.* found that charge neutralization dominates coagulation and increases removal efficiency with coagulant dosage because the coagulant's positive charges gradually decrease MPs' zeta potential<sup>[86]</sup>. When MPs' particle zeta potential is zero or nearly zero, coagulant dosage is optimal, and removal efficiency is the highest. According to Zhang *et al.*, continuous injection of coagulant can result in the re-stabilization of MPs particles, leading to a reduction in removal efficiency<sup>[55]</sup>. The coagulation system includes various mechanisms such as charge patching, adsorption bridging, and sweeping flocculation, and the use of high coagulant dosages and multiple coagulation mechanisms can inhibit MPs re-stabilization. The absolute

value of zeta potential showed a weak correlation with MP removal, and FeCl<sub>3</sub> was found to be effective in removing MPs when sweeping flocculation and charge neutralization were involved, as reported by Rajala *et al.*<sup>[17]</sup>. Thus, MPs' zeta potential in water is significantly greater than zero when removal efficiency is highest. Higher coagulant doses produce denser flocs with stronger adsorption and sweeping effects. Zhang *et al.* indicated a greater removal efficiency because a higher floc density was produced at 200 mg/L PAC than 20 mg/L when PAM dosage was constant<sup>[42]</sup>. Thus, MP removal efficiency increases with coagulant or coagulant aid.

#### Organic matter and surfactants

During the coagulation process, most coagulants have a stronger affinity for the hydrophobic fraction of organic matter compared to the hydrophilic fraction. As a result, hydrophobic organic matter can be effectively eliminated<sup>[87]</sup>. The presence of humic substances on MPs can facilitate more efficient removal during coagulation by serving as a bridge to the coagulant. This is particularly effective when the hydrophobic fraction of organic matter adheres well to the MPs<sup>[21]</sup>. During coagulation, the MP removal efficiency increases when flocs capture organic matter, which increases floc mass and makes sediment removal easier<sup>[42]</sup>. According to a study by Monira *et al.*, the presence of organic matter can reduce the adsorption capacity and removal efficiency of MPs by modifying their functional groups and hydrophobic properties<sup>[54]</sup>. Another study conducted by Zhang et al. found that the absolute electric potential of polystyrene (PS) increases with the addition of humic acid, resulting in a decline in the zeta potential from -49.4 to -61.0 mV as the humic acid concentration increased from 5 to 20 mg/L<sup>[13]</sup>. This indicates that PS binding to flocs is facilitated by increased electrostatic repulsion on particle surfaces. Surfactants can adsorb on MPs' surfaces, affecting their physicochemical properties and coagulant binding capacity<sup>[88]</sup>. Skaf *et al.* found that surfactants significantly changed MP surface zeta potential, increasing MP dispersion in solution<sup>[51]</sup>. Surfactants had little effect on MP coagulation because this experiment used sweeping flocculation to remove MPs. Xia et al. found that nonionic surfactants inhibited the coagulation of MPs but not ionic ones<sup>[49]</sup>.

Removing suspended particles and impurities from water and wastewater requires complex coagulation. Numerous factors can affect the efficiency of coagulation. Coagulation can be significantly impacted by the type, size, and charge of the MPs in the water. One of the important factors includes the selection of an appropriate coagulant and its dosage. Different coagulants (e.g., aluminum sulfate, ferric chloride, and poly aluminum chloride) have different chemical properties and may perform better under particular water conditions. Effective coagulation requires optimal dosage without overdosing, which can lead to residual coagulants in treated water. Coagulation also depends on pH. Adjusting the pH to the appropriate range can enhance coagulation efficiency. Alum is best in slightly acidic to neutral conditions, while ferric chloride is used in a wider pH range. Temperature has an impact on the rate of chemical reactions, including coagulation. The coagulation process, and consequently the formation and subsequent settling of floc, can be sped up using slightly warmer water. The presence of natural organic matter (NOM), such as humic and fulvic acids, can interfere with coagulation by competing with particles for coagulant ions. The concentration of ions in the water can influence coagulation. Higher ionic strength can affect the aggregation of particles and may require adjustments in coagulant dosage. Higher turbidity and smaller particle sizes may require more coagulants and longer contact times for effective coagulation. Coagulant aids, such as polymers or organic chemicals, can be added to improve the coagulation process. These aids can help bridge particles together, enhance floc formation, and improve settling. Successful coagulation requires careful consideration and control of these factors to achieve the desired water quality and removal of impurities.

# COAGULATION MECHANISM FOR MP REMOVAL

Coagulation is a process that destabilizes MPs particles in water to promote their removal by sedimentation. The coagulant's hydrolysate plays a key role in this process, mainly through charge neutralization, adsorption, and sweep flocculation<sup>[89]</sup>. Figure 4A and B explains MP removal mechanism through coagulation. The introduction of charged coagulant particles causes the destabilization of MPs particles during coagulation. Electrostatic forces, caused by the attraction between the negative charges of MPs particles and the positive charges of coagulant particles, cause MPs to aggregate and create flocs, which eventually settle<sup>[15]</sup>. Charge neutralization reduces the electrostatic repulsion of MPs by neutralizing their surface charge. According to Xu *et al.*<sup>[26]</sup> and Wu *et al.*<sup>[90]</sup>, adsorption refers to the coagulant hydrolysate adsorbing oppositely charged particles, such as MPs, which are otherwise removed by precipitates. When there is no coagulant or the concentration is very low, charge neutralization dominates. Adsorption and widespread flocculation increase significantly as the coagulant dosage increases. Several studies have investigated the coagulant mechanism. Adsorption and sweep flocculation become more important with increasing coagulant dosages, but adsorption is preferred at higher doses due to its physical and chemical effects<sup>[42,89]</sup>.

#### Adsorption

The adsorption mechanism is based on electrostatic interaction between the particles having opposite charges. The flocs expand as more MPs are adsorbed, finally settling at the bottom of the water column or being eliminated through filtering. Furthermore, the coagulant might destabilize the MPs and induce them to agglomerate, which increases their removal via adsorption. Coagulant particles can also adsorb onto the surface of MPs, causing them to grow and settle out of the water. In the study conducted by Zhou et al., Al(OH)<sub>3</sub>(am) is produced during PAC hydrolysis, and positively charged monomers adsorbed the surrounding particles before being carried away by amorphous precipitation<sup>[15]</sup>. Under the influence of electrostatic gravitational forces, Van der Waals forces, and chemical bonds, polymers combine with MPs that have not yet fully destabilized via active sites, creating an adsorption bridging effect<sup>[13]</sup>. According to Zhou et al., very little Fe and Al remained in the water after coagulation, indicating that most coagulants formed flocs and precipitated<sup>[15]</sup>. Lapointe *et al.* conducted a study and found that electrostatic adsorption and hydrogen bonding were responsible for the removal of MPs by aluminum-based coagulants<sup>[21]</sup>. These mechanisms were found to interact with MPs via the positively charged cations Al<sup>3+</sup> and amorphous Al(OH)<sub>3</sub> produced by the hydrolysis of these coagulants. When a particle of MP is adsorbed by one end of a polymer chain and another particle of MPs is adsorbed by the other end, a structure known as "MPspolymer-MPs" flocculent is formed<sup>[62]</sup>.

The adsorption mechanism is further explained by the FTIR analysis carried out for MPs and flocs formed during coagulation. For instance, the chemical bonds of MPs and flocs are shown in Figure 5. According to relevant literature on FTIR spectra, the broad peak observed in the 3,000-3,500 cm<sup>-1</sup> range in the PAC spectrum is attributed to the stretching vibration of the hydroxyl group, which may result from PAC hydrolysis. In the case of variable angle vibration of absorbed water, crystal water, and coordinated water, the bending vibration of hydroxyl groups can be linked to the adsorption peak detected at 1,620 and 1,650 cm<sup>-1</sup>. Hydrogen bonds between MPs, PAC hydrolysate, and coagulant aid hydrolysate are confirmed by the presence of O-H. Al-O bond bending and stretching vibrations may also be associated with the observed peak at 535.7 cm<sup>-1</sup>. Coagulant aids can promote PAC hydrolysate adhesion to MPs' particles, as indicated by the large peaks created by the stretching vibration of the hydroxyl group in Figure 5B. Similarly, in Figure 5C, the spectra of the precipitate show broad peaks formed by the stretching vibration of the hydroxyl group. This observation has confirmed that the addition of coagulant aids has an effect on the attachment of the PAC hydrolysate to the MP particles [Figure 5D]. The PET + PAC + PAM system exhibited a new peak at 1,661.96 cm<sup>-1</sup> compared to the PET and PAC + PAM spectra. This peak may be



**Figure 4.** (A) Mechanisms of MP removal by coagulation<sup>[62]</sup>; (B) MP/NP elimination mechanisms with and without coagulation<sup>[59]</sup>. MP: Microplastic; NP: nanoplastic.

attributed to amide, indicating that the MPs' surface has been effectively adsorbed with the hydrolyzed forms of PAC and PAM. Figure 5D shows that the  $SO_4^{22}$  and Si-O-Si lattice vibrations are attributed to the 1,113.59 and 437.86 cm<sup>-1</sup> peaks, respectively. After coagulation, these peaks were observed in the sediment, indicating the efficient adsorption of the coagulant<sup>[42]</sup>. According to Zhou et al., the H-OH vibration of water molecules, C-H stretching, and bending vibration all contributed to a prominent peak in the FTIR spectrum of PS around 3,430 and 2,960 cm<sup>-1[15]</sup>. The presence of the C=O stretching band was indicated by the peak at 1,726 cm<sup>-1</sup>, while the benzene ring substitution was indicated by the peaks at about 870 and 730 cm<sup>-1[91]</sup>. Similar peaks at 3,431, 2,964, 3,430, and 2,960 cm<sup>-1</sup> were exhibited in the FTIR spectrum of flocs, indicating H-OH and C-H vibrations similar to those observed in PS. The substituted group of the benzene ring, which is characteristic of PS, was represented by weak absorption peaks at 871, 729, 869, and 728 cm<sup>-1</sup>, which did not significantly change after coagulation<sup>[15]</sup>. According to Lu et al.<sup>[56]</sup> and Liu et al.<sup>[92]</sup>, the stretching vibration of the hydroxyl groups in the Al-based hydrolysis products is responsible for the strong and broad peak in the PAC spectrum observed around 3,400-3,500 cm<sup>-1</sup>. The hydroxyl group bending vibration, which may be the variable angle vibration of absorbed water and coordinated water, was attributed to the 1,620-1,650 cm<sup>-1</sup> peak<sup>[42,93]</sup>. The existence of a weak peak at 608.53 cm<sup>-1</sup> can be attributed to Al-O bond bending and stretching vibrations. The stretching vibration of the NH group in APAM can explain the large peak found in the 3,400-3,500 cm<sup>-1</sup> range. The stretching vibration of C=O was represented by the peak at 1,621.80 cm<sup>-1</sup> and the bending vibration of NH was represented by the weak peak at 1,660.65 cm<sup>-1</sup>. Characteristic peaks of CH<sub>2</sub> were attributed to PE and remained relatively unchanged after



**Figure 5.** FTIR spectra of MPs and flocs before and after PAC coagulation. Spectra of the precipitate (A) before and (B)-(D) after PAC coagulation supported by coagulant aid<sup>(42)</sup>. ASA: Activated silicic acid; FTIR: fourier transform infrared spectroscopy; MPs: microplastics; PAC: poly aluminum chloride; PAM: polyacrylamide; PET: polyethylene terephthalate; SA: silicic acid.

coagulation in various systems. These peaks were observed at 2,918.77, 2,850.19, 1,473.01, and 718.47 cm<sup>-1[94,95]</sup>.

The adsorption mechanism is further explained by the SEM analysis. Figure 6 shows the shape of flocs generated during various coagulation systems. Figure 6A reveals that the particles appear dispersed and separate, indicating a lack of coagulation. This observation is further supported by Figure 6B, where the presence of distinct individual particles is still evident. Additionally, in Figure 6C, the lack of particle aggregation or clustering is apparent, underscoring the ineffective clumping of the MPs and coagulant particles. The morphology of formed flocs in the composite pollutants system is shown in Figure 6D. It was observed that the flocs formed solely by PAC coagulation [Figure 6E] had a smoother surface compared to those formed by the combination of PAC and coagulant. This observation was further supported by Figure 6F. The addition of coagulant caused the surface of the flocs to become rough, with the appearance of small particles on the surface shown in Figure 6G. The formation of enlarged flocs after the addition of coagulants indicates that adsorption is occurring. In some cases, cracks have been observed on the MPs surface, which might be due to the plastic crushing, as can be seen in Figure 6H. These cracks can increase the specific surface area of MPs, enhance their adsorption capacity, and become carriers of other particles and contaminants [Figure 6]. When a higher concentration of the coagulant (5 mg/L) was introduced, long-chain polymer agglomerates were observed on the surface of the flocs, providing additional evidence for the occurrence of adsorption during the coagulation process<sup>[95]</sup>. Uniform and dense flocs have been found to exhibit higher removal efficiency due to increased collision between the coagulant and particles present in water. However, increasing the concentration of OH- can yield different results, such as the formation of sparse structures with holes on their surface instead of particles. In such cases, the surface of the flocs may appear relatively flat, which could lead to lower removal efficiency. The removal of the pellet and sparse hole structures, which can play a significant role in promoting the coagulation process by providing sites for particle attachment and surface area for chemical reactions, may be responsible for chemical reactions<sup>[55]</sup>.



**Figure 6.** SEM images of flocs obtained by composite systems. (A)-(D) Changes in floc appearance for the same solutions treated with increasing alum dose<sup>[51]</sup>; (E)-(G) The morphology of the inner structure of different flocs in composite pollutants system<sup>[15]</sup>; (H) and (I) Agglomeration adsorption in both PAC-PS system and FeCl<sub>3</sub>-PS system<sup>[48]</sup>. PAC: Poly aluminum chloride; PS: polystyrene; SEM: scanning electron microscopy.

#### Charge neutralization

Solution pH has a significant role in charge neutralization and has an effect on the MP removal efficiency [Figure 7A]. According to Tang et al., charge neutralization is a process for destabilizing colloidal particles that is based on the diffused double-layer model and the DLVO theory<sup>[62]</sup>. Most colloidal particles in water have negative charges, which provide relative dynamic stability to the system through electrostatic repulsion. The addition of a metal salt coagulant raises both the medium's ionic strength and the concentration of counter ions in the diffuse double layer. This is due to the rapid hydrolysis of the metal salt into several cations. Colloidal particle charge and zeta potential are both decreased as a result of this process, resulting in an increase in the frequency and efficiency of collisions between the particles. This exacerbates the system's instability, as Zhang et al.<sup>[42]</sup> and Zhou et al.<sup>[15]</sup> observed. Since MPs have a negative charge on their surfaces, charge neutralization has been regarded as an important factor for their removal efficiency. The application of a coagulant neutralizes the surface charge of MPs, minimizing electrostatic repulsion and facilitating the formation of large flocs. Al<sup>3+</sup> neutralized the surface potential of PET, leading to its removal via charge neutralization, as Lu et al. observed<sup>[56]</sup>. However, Yang et al. pointed out that when coagulant concentrations are too high, MP particles can absorb too much positive charge and re-stabilize<sup>[86]</sup>. Using cationic polymer polyamines resulted in a re-stabilization of MPs, as Rajala et al. demonstrated<sup>[17]</sup>. According to Zhang et al., at low dosages of coagulant, charge neutralization may contribute to the overall removal mechanism for MP destabilizing; however, for complete removal, other mechanisms are also involved<sup>[42]</sup>.

Figure 7 shows that the zeta potential was strongly negative prior to coagulation, indicating the existence of charged particles. The addition of coagulant resulted in a significant reduction in the zeta potential values of the particles, indicating a decrease in their overall charge. This reduction in charge can be attributed to the occurrence of charge neutralization, which took place in both single and composite systems<sup>[15]</sup>. Charge neutralization is an essential step in the coagulation process because it increases MP removal efficiency from water. The optimal dosage of coagulant applied at the appropriate pH can neutralize the negative



Figure 7. Effect of pH on MP removal efficiency and Zeta potential. (A) removal efficiency; (B) Zeta potential<sup>[55]</sup>. MP: Microplastic.

charge on the surface of MPs, bringing the system's zeta potential close to zero. This method results in higher charge neutralization, which increases the likelihood of successful MP removal from water after the addition of coagulants. These results suggest that charge neutralization was more effective in the composite system than in the single systems of PE and NOR<sup>[95]</sup>. The removal efficiency of MPs from water is greatly improved by charge neutralization, making it an essential step in coagulation. The zeta potential of the system can be reduced to zero by using the optimal amount of coagulant at an appropriate pH to neutralize the negative charge on the surface of MPs.

Depending on the characteristics of the MPs and the adsorbent material, different mechanisms can be used to adsorb MPs. Activated carbon, zeolites, clays, and modified polymers are commonly used as adsorbents because they effectively remove MPs from water. The specific properties of the MPs and the treatment objectives determine the choice of adsorbent to use. The surface chemistry of the adsorbent material and the MPs play a significant role in adsorption. MPs are typically hydrophobic, meaning they have low polarity and a strong affinity for nonpolar surfaces. The hydrophobic properties of absorbent materials used for the removal of MPs are frequently considered. The Van der Waals force is primarily responsible for the adsorption of MPs. Strong Van der Waals forces held MPs on the surface of the adsorbent material. Various water and wastewater treatment processes to remove MPs require charge neutralization. The presence of functional groups or ions on the surfaces of MPs causes them to exhibit surface charges. The charges on MPs can cause electrostatic repulsion, preventing them from aggregating or adhering to solid surfaces, making removal more challenging. These surface charges are neutralized to facilitate the aggregation and removal of MPs. The efficiency of charge neutralization and the removal of MPs depends on the type of coagulant or flocculant used, the MPs' size, surface charge density, water quality parameters, and other impurities or substances in the water.

#### ECONOMIC AND ENVIRONMENTAL IMPACT ANALYSIS OF COAGULATION TECHNOLOGY

Economic and environmental impact analysis is a common decision-making tool used to determine the practical feasibility of any existing or novel technologies. The cost of treatment to remove MPs from produced industrial water should be much lower for the effective commercialization of coagulation technology. Coagulant costs to remove MPs from generated water are factored into the overall cost of the coagulation-flocculation process. Identifying the optimal operating conditions of the coagulation process, such as coagulant dose, water volume, and solution pH, is crucial in calculating overall cost. In terms of cost and feasibility, the utilization of natural polymer-based coagulants appears to be a more viable option compared to synthetic chemicals<sup>[96]</sup>. It is also important to conduct the cost analysis of coagulant recovery

after the process to determine the feasibility of the coagulants in large-scale applications<sup>[97]</sup>. However, most of the published studies on MP removal through chemical coagulation lack the cost analysis of the treatment. In contrast, comprehensive investigations have been conducted to analyze the costs associated with electrocoagulation research for MP removal. In a study conducted by Perren *et al.*, the authors examined the comparative operating costs of employing electrocoagulation for the treatment of polyethylene and other contaminants present in wastewater<sup>[18]</sup>. The findings revealed that the operational cost associated with treating polyethylene was significantly lower than that of other contaminants, amounting to  $0.05 \notin$  per m<sup>3</sup>. In addition to conducting a cost analysis of the coagulation process, it is imperative to assess the environmental implications of the coagulants, produced sludge, and associated chemicals used during operation prior to implementing the process on a wide scale. The assessment of the life cycle of coagulants can be conducted for the intended objective.

The cost-effectiveness and sustainability of coagulation technology in water and wastewater treatment depend on economic and environmental impact analyses. The economic and environmental impacts of coagulation technology can be evaluated. An in-depth analysis of coagulation technology's economic and environmental impacts requires data on operational parameters, treatment effectiveness, chemical use, energy consumption, waste generation, and associated costs. Decision-making processes, water treatment technology selection, and efforts to reduce the environmental impact of water and wastewater treatment facilities can all benefit from such analyses.

# CHALLENGES AND PERSPECTIVE

Due to its comprehensive removal capabilities, coagulation can be a cost-effective and efficient approach for eliminating a wide spectrum of contaminants from water and wastewater if certain conditions are met<sup>[89]</sup>. In general, the coagulation/flocculation procedure could aid in the efficient removal of MPs from water and wastewater. According to the previous studies, this approach was highly dependent on pH value, size, shape, and types of MPs, and dosage and type of coagulant and flocculant aids<sup>[13,15,42,55,62]</sup>. Until now, there has been a scarcity of coagulation studies for MP removal, particularly in wastewater treatment systems. Future research must focus on determining the best coagulants/flocculant aids and ideal circumstances for MP removal and colloid elimination. Few studies have recently demonstrated that coagulation can successfully remove MPs from water and wastewater. Coagulation for MP removal, on the other hand, is still in its infancy, and more research is needed to optimize the process and understand the underlying principles. Future research should focus on developing new coagulant materials, optimizing the reaction conditions for different types of MPs, and investigating the potential impact of coagulant residues on the environment. In addition, the combination of coagulation with other treatment methods such as membrane filtration and advanced oxidation processes may further improve the removal efficiency of MPs and increase the economic viability of the treatment process<sup>[62]</sup>.

Enhanced coagulation [Figure 8] is a practical and cost-effective technique for removing colloidal particles, including MPs, from water. This process encompasses a range of disciplines, such as physics, chemistry, hydraulics, physical chemistry, colloid chemistry, and others, which necessitate collaboration between multidisciplinary experts and scholars to strengthen coagulation through theoretical research and practical exploration<sup>[74]</sup>. Coagulation is a commonly employed method for water treatment in water supply facilities, and as drinking water standards continue to improve, higher standards have been set for its effectiveness. This has resulted in the development of new coagulants and coagulation technologies, including the production of specialized flocculants with a variety of options to choose from. To achieve optimal results, different flocculants should be used for varying water temperatures, pH values, and types of suspended and dissolved matter. Different flocculant products have been created for tap water by developing and utilizing



Figure 8. Conditions required for enhanced coagulation.

aluminum salts. However, it is important to produce specialized flocculants that are tailored to specific needs to ensure proper production and sales, especially since water quality is typically similar in the same geographic region. The ideal flocculants should be easily accessible, have simpler production procedures, be user-friendly, and have lower costs with competitive market prices. Continuous research and development efforts are necessary to generate effective flocculants that meet various water treatment requirements. This requires continuous research and development efforts to produce effective flocculants that meet the needs of different water treatment scenarios.

The fate and toxicity of coagulants after treatment depend on various factors, including the type of coagulant, the treatment process, and the specific environmental conditions<sup>[98]</sup>. The effectiveness of the treatment process can influence the fate and toxicity of coagulants<sup>[99]</sup>. The treated water or wastewater may still contain some coagulant residuals after treatment. Dosage, treatment efficiency, and post-treatment process (such as filtration and disinfection) are all factors that affect the level of residual coagulants. Inorganic salts and other chemicals used in water treatment can be costly and harmful to the environment. They can produce toxic waste, be difficult to break down naturally, and cause contamination of water with metals, which can be a threat to human health. For example, the use of aluminum salts has been linked to Alzheimer's disease. Additionally, the dispersion of acrylamide oligomers, which can result from these chemicals, can be dangerous to human health as the acrylamide monomer is known to be carcinogenic and neurotoxic to humans, as stated in a study by Salehizadeh et al.[100]. Compliance with drinking water or effluent discharge standards requires monitoring of coagulant residuals. Coagulants themselves can have varying degrees of toxicity depending on their composition and concentration. However, the levels of coagulants used in water and wastewater treatment are typically below the threshold for acute toxicity to humans<sup>[66]</sup>. If the treated water is discharged into the environment, the fate of coagulant residuals will depend on the specific conditions of the receiving water body. Dilution, natural degradation processes, and interactions with sediments can affect the persistence and transport of coagulant residuals. Some coagulants, such as aluminum-based ones, may undergo hydrolysis and form insoluble precipitates in water, which can settle and become part of the sediment<sup>[101]</sup>.

Therefore, exploring less costly, more efficient, and more eco-friendly options for MP removal is important<sup>[102]</sup>.  $Mg(OH)_2$  is a water treatment agent that is considered environmentally friendly due to its many benefits, such as its ability to adsorb strongly, its stability, its ability to act as a stable buffer, its non-

corrosive nature, and its safety and lack of toxicity. Li *et al.*<sup>[103]</sup> and Zhang *et al.*<sup>[55]</sup> conducted a study using magnesium hydroxide Mg(OH)<sub>2</sub> or a magnetic coagulant made of Mg(OH)<sub>2</sub>, Fe<sub>3</sub>O<sub>4</sub>, and PAM to treat simulated natural water containing PE. Their findings indicated that the addition of magnesium ions alone did not result in enough floc formation to remove PE particles. However, the addition of PAM after the formation of magnesium hydroxide significantly improved the removal efficiency of PE particles, achieving 84.9%  $\pm$  3%. Furthermore, the addition of 5 mg/L of anionic PAM resulted in the production of larger and denser flocs with an average size of 57.19 µm. The formation process of flocculation was found to be dependent on the Mg(OH)<sub>2</sub> produced during the rapid mixing period, while the growth of flocculation was dependent on the bridging function of PAM.

One of the biggest challenges of the coagulation technique is the recovery of spent coagulant after MPs treatment, which is not comprehensively investigated in most of the published studies. Recovery of spent coagulants is crucial in mitigating the environmental toxicity associated with the coagulant while simultaneously enhancing the cost-effectiveness of the treatment process. Several recovery methods have been investigated, including alkalization, ion exchange, acid digestion, and membrane separation. The acid digestion method has been extensively employed due to its notable efficacy in recovering substances. Nevertheless, the coexisting pollutants that are soluble in acidic conditions, such as heavy metals and organic substances, are also recovered during acid digestion, which may result in secondary pollution. Therefore, ion exchange and membrane separation are currently popular methods for recovering spent coagulants with great efficiency. However, developing a multifunctional membrane with bio-based green materials over a conventional one, as well as improving the efficiency of ion exchange, are the greatest challenges and the subject of further exhaustive research.

Another pressing issue of coagulation technology is the management of recovered MPs. Open disposal of recovered MPs may create secondary pollution. The preparation of functional carbon materials via high-temperature carbonization using recovered MPs could be an effective approach for a sustainable solution. Several treatment methods such as anoxic pyrolysis carbonization, catalytic carbonization, pressure carbonization, flash joule heating, and microwave conversion have been employed for preparing functional carbon materials from plastic wastes.

Renewable materials derived from natural sources, such as cellulose and chitin, are being explored as potential solutions for mitigating MP pollution due to their biodegradable and biocompatible properties. Chitin, a polysaccharide found in the exoskeleton and internal structures of invertebrates, is an attractive option for MP removal due to its abundance, non-toxicity, and biodegradability. However, the practical use of chitin is limited by the molecular depolymerization that occurs during chitin modifications, resulting in reduced mechanical strength<sup>[104]</sup>. Chitin sponges have been reinforced with graphene oxide (GO) or oxygen-doped carbon nitride to increase their strength and enhance pollutant removal through hydrogen bonding and other interactions. These composite sponges have demonstrated high removal efficiency (71.6%-92.1%) for neutral and charged MPs, and are not significantly affected by seawater or dissolved organic matter<sup>[105]</sup>. Both GO and carbon nitride composites perform similarly, but carbon nitride is a more cost-effective option. These sponges can be reused multiple times without significant reduction in performance. In a previous study, chitin and GO sponges were synthesized using double cross-linking to remove MPs<sup>[104,106]</sup>.

Bio-based flocculants can efficiently generate flocs by reacting with MPs, which can then be easily removed by separation or filtration. Given their low-cost, environmentally friendly nature, and regeneration properties, lignin and cellulose are potential options for removing MPs<sup>[107,108]</sup>. It also helps in the precise

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monitoring of flocculation kinetics and aggregate size variations. Biopolymers are available in nature and are considered to be sustainable options for MP removal<sup>[102]</sup>.

The innovative electrocoagulation (EC) approach for water and wastewater treatment combines the advantages of coagulation, flotation, and electrochemistry<sup>[109]</sup>. The process of EC involves the generation of cations from metal electrodes under an electric field, which leads to the formation of "micro-coagulants". These micro-coagulants result in the destabilization of suspended particles, and subsequently, the interaction between coagulants and particles forms flocs<sup>[110]</sup>. EC produces coagulants directly in situ using metal electrodes instead of chemical coagulation, which requires the external addition of coagulants. As a result, the coagulation process is straightforward (easily automated), there is no chance of secondary pollution, and significantly less sludge is produced<sup>[109,111,112]</sup>. Drinking water, groundwater, and wastewater from refineries have all been treated with EC because it is a green technology that can remove even the most pervasive pollutants<sup>[109,110,113]</sup>. Perren *et al.* studied the efficacy of EC in removing polyethylene MPs from wastewater in a stirred-tank batch reactor<sup>[18]</sup>.

# CONCLUSION

MPs have become a major environmental concern due to their potential impact on ecosystems and human health. Coagulation has emerged as a promising method for removing MPs from wastewater and surface water due to its effectiveness and relatively low cost. However, the success of coagulation as MP removal method depends on several factors, including the pH of the solution, the type and concentration of the coagulant, temperature, stirring speed, settling time, and MP shape. The key mechanism responsible for the removal of MPs by coagulation is charge neutralization, adsorption, and sweep flocculation. Although initial results have indicated that inorganic coagulants are effective for MP removal, certain concerns regarding toxic sludge generation have been reported. This article looks at the benefits of employing ecologically friendly organic coagulants as well as the operational considerations involved in water and wastewater treatment. However, there is also a need to investigate the feasibility of commercializing and industrializing composite polymerization and the impregnation of organic polymers with inorganic coagulants as a promising study topic.

Changes in MP density can improve coagulation settling. The effectiveness of coagulation removal is influenced by MP particle size due to its effect on collision efficiency and settling behavior. Compared to larger MPs, smaller MPs are easier to remove. pH of the solution has a significant impact on the removal of MPs by coagulation. Hydrolysis of coagulants and floc characteristics are regulated by pH, which is an essential factor in coagulation. pH 6 and 7 form smaller, more dispersed flocs, while pH 8 and 9 form denser flocs. Several types of inorganic ions in natural water may impact coagulation performance. This is because interactions between ions and coagulants may affect their stability and capacity to remove contaminants. Comparing the removal efficiency of MPs,  $SO_4^{2^{-2}}$  and  $CO_3^{2^{-2}}$  have a more significant impact on the removal of MPs than Cl<sup>-</sup> ions. MP shape affects coagulation removal efficiency. The higher specific surface area of fibrous MPs makes them more likely to combine with flocculants despite having the same volume. Compared to smooth MPs, rough MPs particles are removed more efficiently. MP removal and disinfection by-product component distribution depend on the coagulant dose. The zeta potential of MPs decreases with a positive charge, so coagulant dosage increases coagulation efficiency. Humic substances on MPs can act as a bridge to the coagulant during coagulation, enabling more efficient removal. When flocs capture organic matter during coagulation, MP removal efficiency increases, increasing floc mass and sediment removal.

To optimize the efficiency of coagulation for MP removal, it is important to carefully consider the factors and to conduct further research to better understand the coagulation mechanisms. Developing more effective and efficient coagulation methods could have significant implications for the protection of aquatic ecosystems and public health and the sustainability of water resources. Furthermore, consideration of the potential environmental impacts of using coagulants, such as the generation of additional waste and the potential for unintended consequences on aquatic ecosystems, is essential. One of the most significant limitations of the coagulation process is the generation of a huge volume of sludge and recovered MPs, which can cause secondary contamination if not properly managed. Currently, a variety of technologies have been implemented for the purpose of sludge disposal, prioritizing both environmental safety and public health. However, finding a beneficial reuse approach has emerged as a critical aspect of sludge management. Likewise, the preparation of functional carbon materials via high-temperature carbonization could be an effective approach for the management of recovered MPs. It is therefore critical to continue to investigate novel and innovative techniques for removing MPs via coagulation, as well as to strive towards establishing more sustainable and environmentally friendly management for generated sludge and recovered MPs.

# DECLARATIONS

# Authors' contributions

Conceptualization, data curation, methodology, validation, writing - original draft: Khan MT Methodology, data curation, writing - original draft, software: Ahmad M Methodology, writing - review and editing: Hossain MF Writing - review and editing, validation, software: Nawab A Methodology, writing - original draft: Ahmad I Data curation, methodology, validation: Ahmad K Validation, writing - review and editing, supervision: Panyametheekul S

#### Availability of data and materials

The data for presenting the manuscript is provided in the text; further inquiries can be directed to the corresponding author.

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All authors declared that there are no conflicts of interest.

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