Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2023.29398

288 (2023) 51–59
March

Efficient removal of polyethylene and polyvinyl chloride microplastics from water using a modified coagulation process supported by the addition of a surfactant

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Received 29 September 2022; Accepted 12 February 2023

ABSTRACT

In water and wastewater, microplastics (MPs) are regarded as one of the emerging contaminants. Understanding how microplastics are removed in the current wastewater and water treatment system is important. The purpose of this study was to investigate how the coagulation process affected the removal of two different types of microplastics from tap water, including polyethylene (PE) and polyvinyl chloride (PVC) and their mixture. Regarding the effectiveness of microplastic removal, the effects of different types of coagulants, such as $AlCl_3·6H_2O$ and $FeCl_3·6H_2O$, doses of coagulants, solution pH, microplastic concentration, and water characteristics, as well as addition of surfactant sodium dodecylbenzenesulfonate (SDBS), were thoroughly examined. The neutral pH value of tap water and the coagulant dose of 0.05 g/L resulted in the highest removal of MPs. When tap water was compared with ultrapure water, tap water with humic acid, and tap water with NaCl, the best coagulation efficiency was observed in tap water. The effectiveness of PE and PVC removal in both Al salt and Fe salt coagulation was greatly improved by the addition of SDBS. The elimination of PE and PVC was more than 90% successful for both tested coagulants at a dose of 0.025 g/L. Optimal parameters were also used to remove the analyzed materials in the mixture of PE and PVC. The efficiency of microplastic removal with the use of Al and Fe coagulants and SDBS was obtained at the level of 95.92% and 98.9%, respectively.

Keywords: Microplastics; Polyethylene; Polyvinyl chloride; Coagulation; Detergent; Particle properties

1. Introduction

Recent years have brought increasing concern about the aquatic environmental risks posed by microplastics (MPs, plastic debris with dimensions of 1 to 5,000 μm) [1–7]. The most abundant MPs detected in the environment include polyethylene (PE), polypropylene (PP), polystyrene (PS), polyvinyl chloride (PVC), and polyethylene terephthalate (PET), of various colors and shapes [8,9]. Due to their origin, microplastics are divided into two groups, primary and secondary. Primary microplastics are plastics produced in microscopic sizes. They are used as a raw material in the plastics industry, in the production of medicines and articles for hygiene and personal care. These particles are a serious problem because they are not biodegradable and because of their small size, they are not removed in traditional wastewater treatment, thus ending up in natural water reservoirs. Secondary microplastics are the result of the breakdown of large plastic particles by physical, chemical,

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Presented at the 15th Scientific Conference on Micropollutants in the Human Environment, 14–16 September 2022, Częstochowa, Poland

and biological factors, which means that any single piece of plastic can be a potential source [10,11].

Water and wastewater treatment plants are recognized as important sources of microplastics [12]. The presence of microplastics in treated water and treated wastewater raises concerns about the sufficient effectiveness of the technologies used, and thus about the quality of the treated water or wastewater. Although the efficiency of microplastics removal in a wastewater treatment plant is over 90%, the amount of microplastic particles emitted to the aquatic environment in treated wastewater reaches over 10^9 particles, which corresponds to 200 PET bottles. This is confirmed by the fact that microplastics are present and move through the entire wastewater treatment system before being discharged into receivers [13–15]. Due to their small size and diverse chemical nature, microplastics can easily move through wastewater and water treatment processes, affecting technology at different levels and challenging operational and process stability. High concentrations of microplastics can affect the efficiency of water and wastewater treatment processes. In addition, microplastic particles moving through successive processes in a wastewater treatment plant face various influences such as mixing or pumping, which can break down the microplastic into smaller pieces, increasing the number of toxic nanoparticles released into the water. So far, research on the fragmentation of microplastics has focused only on the natural environment. However, there is no research focusing on the fate and transformation of microplastics in wastewater treatment plants or in water treatment technology. The mechanism and degree of influence of unit processes on the degradation of microplastic particles are unknown. Currently, research is conducted all over the world on mainly methods of quantifying microplastics in environmental samples. The problem of accurately estimating the amount of microplastics results from the need to use advanced equipment. The result of inaccurate analysis methods is the underestimation of the amount of microplastics present in wastewater and water. In addition, it was confirmed that, as a result of physical, chemical, and biological interactions during water and wastewater treatment, microplastic particles are further fragmented into, for example, nanoplastic. The statement that microplastics are removed during wastewater treatment may therefore be false due to conversion to nanoplastic and smaller particles [9,16].

Today, microplastics has become a global and complex problem. Due to chemical stability, plastic microparticles are stable in an aqueous environment and remain there for thousands of years [17,18]. The systematic influx of microplastics into natural water reservoirs is a threat to plants, animals, and people. This phenomenon causes irreversible and dangerous changes in the aquatic environment. The micrometric elements of plastic find their way into the food chains of aquatic organisms. Microplastics have a high specific surface area because of their small particle size. The microplastics present in water reservoirs absorb and transport many toxic substances, such as polycyclic aromatic hydrocarbons, pesticides, detergents, and heavy metals. Furthermore, plastic becomes toxic through additives [19]. An efficient and low-cost method to remove microplastics from water and wastewater is needed. In recent years, many technologies have been developed to remove MPs in water environments, which can be roughly divided into physical (filtration, adsorption), chemical (coagulation, photocatalysis, oxidation treatment), and biological technology (microbial degradation, bioreactors, other biological treatments) [20]. For MPs, filtering technology is a quick removal method. However, between medium filtration and membrane filtration, the removal efficiency differs. Additionally, small MPs or NPs are difficult to remove with quick filtration technology, which has good efficiency for removing large MPs. Although MPs can be effectively removed in a wide range of sizes by membrane filtration methods including ultrafiltration, microfiltration, and MBR, regular membrane fouling and membrane replacement would come at a relatively high cost [21]. Furthermore, when microplastics are successfully removed using the membrane approach, the deposition of microplastics can speed up the contamination of the membrane, which can speed up the contamination of other organic materials in the membrane. To prevent excessive organic matter and microplastic membrane contamination when using a membrane, a pretreatment procedure must be installed [22,23]. Adsorption is a straightforward technique to remove MPs, especially those less than 10 μm in size. However, in aquatic environments, additive sorbents can cause secondary contamination. The potential toxicity, repeatability, and degradation of the adsorbents should all be carefully taken into account when using them. Magnetic separation has recently been created to remove MPs. Large-surfaced magnetic nanoparticles were utilized as adsorbents throughout the removal process to combine with the MPs, and the magnetized MPs could then be swiftly and readily removed from water using magnetic force. However, it is typically necessary to add many magnetic adsorbents to ensure that their amount exceeds the number of MPs in the water. Consequently, the issue is how to completely remove the added magnetic components after the treatment process [24,25]. Advanced oxidation processes and biodegradation are based on the degradation and mineralization of MPs, a technique that is still in its infancy. In contrast, oxidation treatment is quick but requires the inclusion of chemical catalysts. Because of their high removal efficiency, NPs and small-size MPs are particularly well suited for chemical oxidation treatments such as photocatalysis. However, leftover oxidation intermediates and chemical catalysts provide a possible environmental danger. However, for the treatment of MPs, the biodegradation method requires a lengthy period of time. However, it does have the benefit of no chemical addition, though. Bioreactors offer an alternative engineering approach, but the effectiveness of degradation varied depending on the type of polymer [26–31]. In general, research on microplastics removal technology is still in its early stages and has a number of limitations, including the kind and size of target MPs, the quantification method of MPs, etc. Currently, it is believed that combining several methods will increase the effectiveness of eradicating MPs.

The most efficient approach at the moment to remove microplastics is thought to be coagulation-based. Coagulation is frequently employed as a pretreatment method to remove large, heavy, or light MPs in conjunction with sedimentation and air flotation. Numerous studies have examined the effectiveness of removing microplastics from these procedures when using various coagulants, such as Fe and Al salts [21–33]. The coagulation of PE microplastics in conjunction with ultrafiltration was studied by Ma et al. [21]. The removal effectiveness was shown to increase with decreasing PE particle size. $AlCl_3·6H_2O$ produced a better PE removal outcome than $FeCl_3·6H_2O$. Using $FeCl_3$ and PAC (poly aluminium chloride) coagulation, Zhou et al. [32] investigated the effectiveness and method of removing polyethylene and polystyrene microplastics. This study proved that PAC was more effective in removing PE and PS than $FeCl_3$. For PE, the removal efficiency increased with decreasing particle size, whereas for PS, the removal efficiency decreased with decreasing particle size. Monira et al. [33] investigated the effect of coagulation process for the removal of PE and PP from synthetic stormwater. The results indicated that standalone alum and PAM (polyacrylamide) were ineffective in removing MPs, but a combination of both coagulants increased the removal efficiency. The maximum efficiency was up to 96% using 150 mg/L of alum and 15 mg/L of PAM coagulant at pH = 7. The presented articles concern research on the removal of one type of polymer. It is challenging to create ideal conditions for such a large collection of micropollutants, since there are so many different varieties of microplastics in water and wastewater, all with distinct materials, shapes and sizes, and diverse qualities. No research has attempted to eliminate a variety of mixtures of microplastics. Furthermore, more research is required to comprehend the removal of MPs taking into account their shape, size, and surface.

Microplastics in freshwater ecosystems need increased attention because freshwater was once thought to be a significant source of microplastics in the oceans. People's lives are directly impacted by freshwater, because most freshwater rivers supply people with their daily tap water. However, tap water has also been shown to contain microplastics. Coagulation is a crucial process in water treatment plants, which is related to the amount of microplastics entering people's lives. Water treatment facilities are the link between freshwater and tap water. According to reports, coagulation has a specific impact on the elimination of microplastics. However, there are different forms of microplastics in distinct bodies of water, and various coagulants have different outcomes. Furthermore, the precise mechanism is unclear [34,35]. Therefore, it is vital to investigate its coagulation efficiency and process in order to remove microplastics more efficiently. Due to the prevalence of microplastics in the aquatic environment and their detrimental effects on both the environment and living things, studies have been done to determine whether the coagulation process is effective in removing microplastics from tap water. The objective was to examine the effectiveness of removing PE and PVC microplastics using salt coagulation that is based on Al and Fe. The uniqueness of the research was the addition of an anionic surfactant, sodium dodecyl benzenesulfonate, to improve the coagulation process. The purpose of adding surfactant to water and wastewater during coagulation up to this point has been to ensure that the microplastics are evenly distributed throughout the water. Finally, because most of the research on MPs has concentrated on a single form of microplastic, this study examines the removal of a mixture of two significant plastic elements of MPs that are important from an environmental perspective.

2. Materials and methods

2.1. Reagents and materials

Commercially available PE (ultra-high molecular weight) and PVC (high molecular weight) microplastics were purchased from Sigma-Aldrich (USA). Information on the microplastics tested is presented in Table 1.

$AlCl_3·6H_2O$ and $FeCl_3·6H_2O$ as coagulants, sodium dodecylbenzenesulfonate anionic surfactant (SDBS), hydrochloric acid (HCl), sodium hydroxide (NaOH), humic acid (HA), sodium chloride (NaCl) and ethanol (C_2H_5OH) were obtained from Sigma-Aldrich (USA). All reagents used in this study were analytical grade. The water used in the study was tap water (Rzeszow), ultrapure water extracted by Milli-Q pure water mechanism, tap water with humic acid (25 mg/L), and tap water with NaCl (1 g/L), The selected parameters of tap water (June 2022) are presented in Table 2. The glass microfiber filters were purchased from Whatman (GF/A, 47 mm diameter, 1.6 μm pore size).

2.2. Coagulation experiments

Stuart flocculator with six rotators (SW6) was used for coagulation experiments. Each volume of water sample was 500 mL. The pH of the solution was maintained at 3, 5, 7, and 9 by adding HCl or NaOH solution. The doses of PE and PVC in the experiments were 0.05, 0.1, 0.15, and 0.2 g/L. Weighed samples of microplastics were added to tap water and the solutions were mixed. The dose of Al and Fe-based coagulant used in this study was 0, 0.025, 0.05, 0.1 and 0.2 g/L. The coagulants were dosed in the form of 0.1 g/mL aqueous solutions. Modification of the classical coagulation process consisted of adding 20 mg/L of SDBS surfactant to

Table 1
Characteristics of microplastics

Microplastic	Size (μm)	Density in 25°C (g/mL)	CAS number
Polyethylene	15–97[a]	0.94	9002-88-4
Polyvinyl chloride	50–162[a]	1.4	9002-86-2

[a]Own LDIR analysis.

Table 2
Characteristics of tap water [36]

Parameters	Average value
Color, mg/L	<5
Turbidity, NTU	<0.20
pH	7.70
Conductivity, μS/cm	647
NO_3^-, mg/L	6.3
NO_2^-, mg/L	<0.05
Cl^-, mg/L	38
SO_4^{2-}, mg/L	36
Total organic carbon, mg/L	1.50
Total hardness, mg·$CaCO_3$/L	260

the selected solutions. The mixing speed was maintained at 300 rpm for 1 min, then 50 rpm for 15 min, with a subsequent sedimentation of 45 min. After sedimentation, the supernatant was collected to calculate the removal efficiency of microplastics.

2.3. Measurement of microplastics

All supernatants collected from each experiment were filtered through pre-weighed Whatman filters, and the remaining flocs on the surface of the MPs were eliminated using an HCl solution. The thermal research chamber was used to dry the MP-containing filters for 18 h at 60°C. After the MPs filters had reached room temperature, they were weighted. Every experiment was run a minimum of three times. Calculating the mass of each filter membrane before and after filtering allowed us to determine the mass of the remaining microplastic in the dried sample (which had not been removed). Dried microplastic samples were suspended in ethanol and deposited on infrared reflective glass slides (7.5 cm × 2.5 cm, MirrIR, Kevley Technologies, USA). The glass slides were analyzed in transflection by automated LDIR Imaging (with Quantum Cascade Laser) (Agilent 8700). The automated particle analysis protocol with Agilent Clarity software was used. The sensitivity was set to the maximum and the spectral resolution to 8 cm^{-1}. The following parameters of the microplastics were determined: diameter range and mean diameter, area, perimeter, circularity, and solidity.

2.4. Quality assurance and quality control

Only glassware and metal items were used to avoid plastic contamination. All glassware was rinsed with Mili-Q water and 30% ethanol and heated at 280°C for 5 h to remove impurities. All filters have been cleaned with ultrapure water prior to use. All experiments are carried out on a clean operating platform. The weight loss of the filter itself was also checked during heating at 60°C for 18 h (weight loss below 0.5% was observed).

3. Results and discussion

3.1. Variable study on microplastics removal

Previous studies have shown that Al-based coagulants are better than Fe coagulants in microplastics removal [21,37]. Therefore, $AlCl_3{\cdot}6H_2O$ was selected to assess the effect of the coagulant dose on the effectiveness of PE and PVC removal. The removal efficiencies of PE and PVC under different dosages of coagulant are shown in Fig. 1a. Coagulant doses of 0, 0.05, 0.1 and 0.2 g/L were used, which corresponds to the amount of Al, respectively, 0, 0.21, 0.42 and 0.84 mM/L. In the absence of coagulant, the removal efficiency of PE was only 8.44%, while that of PVC was about 65%, which is related to different density. PVC sinks easier as a result of the higher density. The trend is similar to a previous study [32]. Zhou et al. [38] studied the susceptibility of polystyrene and polyethylene microplastics to coagulation. Under the same coagulants, the removal efficiency of PS was much higher than that of PE, largely due to the higher density of PS. Although PS has a higher density than water, which settles easily, density is not the only element that influences the vertical distribution. According to certain research, microplastics that are denser than fresh water can still be found on the surface of the water. Meanwhile, sediments included microplastics whose density was lower than that of freshwater [39,40]. Although the density of PS microplastics with small diameters is higher than that of water, the effect of the water surface tension is likely to prevent settlement. Large PS microplastics are more likely to overcome surface tension and settle more readily, which may explain why their removal effectiveness is higher.

As seen in Fig. 1a, the best removal efficiency for both PE and PVC was reported for the dose of 0.05 g/L. The removal efficiencies of PE and PVC were 29.62% and 89.24%, respectively. Both lower and higher doses of coagulant resulted in less removal of the microplastics tested. This may be explained by the phenomenon that microflocs kept the state of single particles at a low concentration, but when the coagulant dose was too high, the flocs tend to loosen and break

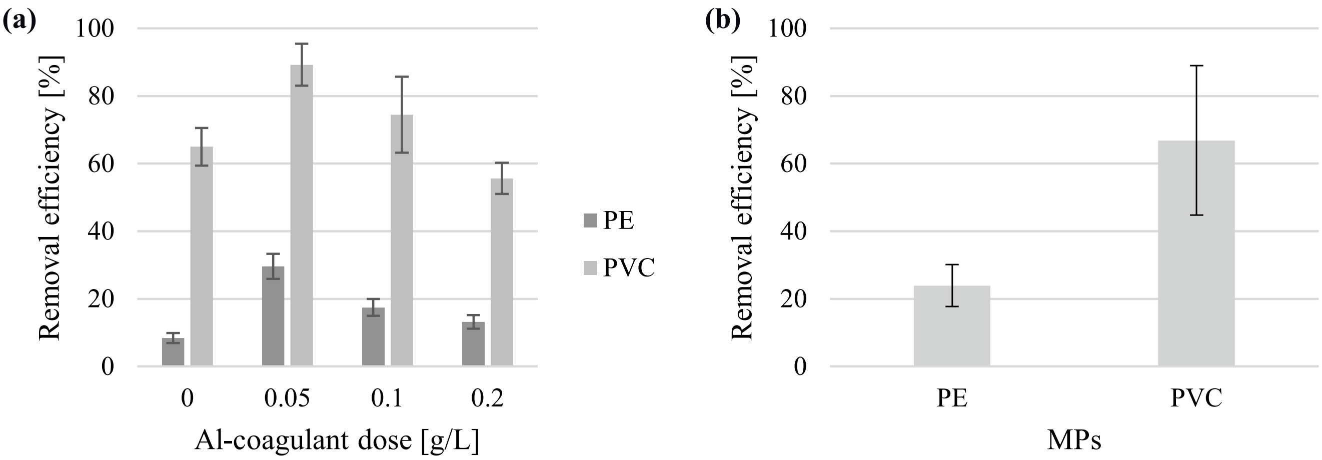

Fig. 1. Efficiency of removal of polyethylene and polyvinyl chloride microplastics in tap water under various dosages of Al- and Fe-coagulant. (a) Al-coagulant process ($[PE]_0 = [PVC]_0 = 0.1$ g/L, pH = 7) and (b) Fe-coagulant process ($[PE]_0 = [PVC]_0 = 0.1$ g/L, pH = 7, $[Fe\text{-coagulant}]_0 = 0.05$ g/L).

easily [32,41]. Fig. 1b shows the results for the coagulation process using $FeCl_3{\cdot}6H_2O$ at a dose of 0.05 g/L (0.18 mM/L of Fe). For both PE and PVC microplastics, a lower average removal efficiency was observed for the Fe-coagulant. According to reports, aluminum coagulation is more effective and research on iron coagulation is not as well covered as that on aluminum coagulation. The different nature of the sludge could be the cause. The fact that microplastic particles are removed more effectively from aluminum sludge may be due to its higher water content compared to ferrous sludge [42]. Other experiments [43,44] showed that the concentration of coagulants based on Al and Fe should always be less than 20 mg/L (0.74 mM of Al, 0.36 mM of Fe) for drinking water treatment.

PE and PVC particles were transferred before and after the coagulation process to Kevley slides and analyzed using LDIR. The following parameters of the molecules were determined: diameter range, mean diameter, area, perimeter, circularity, and solidity. The results obtained are presented in Table 3. The aspect ratio of a geometric shape is the ratio of its sizes in different dimensions. Circularity is defined as the degree to which the particle is similar to a circle, taking into account the smoothness of the perimeter. A circularity value of 1.0 indicates a perfect circle. As the value approaches 0.0, it indicates an increasingly elongated shape. In turn, solidity is the ratio of the actual surface area of a particle to the surface area constituted by a thread stretched around the particle. By analyzing the data contained in Table 3, it is possible to observe a change in all the parameters tested. The detection of larger particles indicates the joining of smaller and larger particles and the formation of flocs during coagulation. On the other hand, the detection of particles smaller than the original ones confirms that the particles are subjected to different forces during the coagulation process and fragmented into smaller ones. The increase in the area and perimeter of the particles is related to the increase in the mean diameter. The mean circularity of the unremoved particles was higher in each coagulation process for PE and in Al-coagulation for PVC than in the circularity of the particles before coagulation. On the other hand, similarities were observed in the case of solidity. In the case of PE, this parameter increased, while in the case of PVC, this parameter did not change in the case of Al-based coagulation, whereas in the case of Fe-based coagulation, it decreased slightly.

The pH of the water plays a significant role in the coagulation performance for the removal of MPs. The pH value has an effect on the hydrolysis of the coagulant and the coagulation efficiency [45]. Fig. 2 shows the removal efficiencies of PE and PVC microplastics at different initial pH. The experiments were carried out at pH 3, 5, 7 and 9 to investigate the efficiency of MP removal. The results showed that the removal efficiency of PE and PVC was the highest under neutral conditions, both in the Al- and Fe-based coagulant. The lowest PE removal efficiency was observed for acidic conditions (pH = 3) for both coagulants used, and at pH = 5 and pH = 9 for the Fe coagulant. The lowest PVC removal efficiency was observed at pH = 3 and pH = 9 (Fe coagulant). Similar conclusions have been obtained in other studies [29,30]. For example, the study by Shen et al. [46] on the effect of electrocoagulation on the removal of PE, PMMA (poly(methyl methacrylate)) and PP showed that the final removal rate of each microplastic at pH 3 and pH 10 was lower than that of pH 5 and pH 7.2. Under neutral conditions, all microplastics are negatively charged, which is more conductive to combine with positively charged flocs to remove microplastics from water [46]. Due to the fact that the pH value of municipal wastewater is usually 6–9.2, the applicability of the coagulation process meant that it can be used effectively in almost all wastewater containing microplastics without adding many more chemicals to adjust the pH value [47]. On the other hand, in the studies of Monira et al. [33], it was shown that the removal efficiency of LDPE (low-density polyethylene), HDPE (high-density polyethylene) and PP was higher under acidic conditions (pH 3 and pH 5), while alum and PAM were used for coagulation.

The influence of the initial concentration of microplastics on the effectiveness of the coagulation process was analyzed. (Fig. 3). A coagulation process was carried out using Al salts in solutions containing PE and PVC in amounts of 0.05, 0.1, 0.15 and 0.2 g/L. The effectiveness of PE and PVC removal depended on the initial concentration

Table 3
Characteristics of microplastics before and after the coagulation process ($[PE]_0$ = $[PVC]_0$ = 0.1 g/L, $[\text{Al-coagulant}]_0$ = $[\text{Fe-coagulant}]_0$ = 0.05 g/L, pH = 7)

Particle parameters						
Coagulation process	Diameter range (μm)	Mean diameter (μm)	Mean area ($μm^2$)	Mean perimeter (μm)	Mean circularity	Mean solidity
Polyethylene						
Before	15–97	39.84	1,507	173.27	0.61	0.881
$AlCl_3{\cdot}6H_2O$	15–146	67.87	4,392	249.63	0.75	0.93
$FeCl_3{\cdot}6H_2O$	16–124	55.23	2,805	214.86	0.69	0.92
Polyvinyl chloride						
Before	50–162	98.18	8,146	368.09	0.72	0.96
$AlCl_3{\cdot}6H_2O$	27–197	134.58	15,276	484.79	0.76	0.96
$FeCl_3{\cdot}6H_2O$	19–277	143.79	17,970	571.84	0.68	0.93

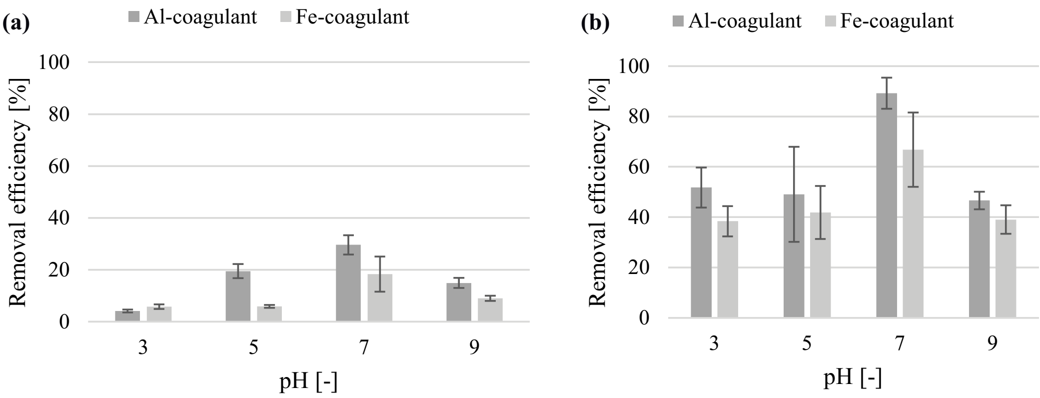

Fig. 2. Efficiency of removal of (a) polyethylene and (b) polyvinyl chloride microplastics in tap water at various pH value ($[PE]_0 = [PVC]_0 = 0.1$ g/L, $[\text{Al-coagulant}]_0 = [\text{Fe-coagulant}]_0 = 0.05$ g/L).

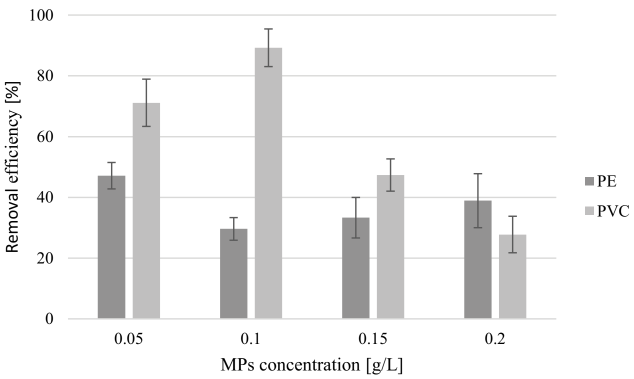

Fig. 3. Efficiency of removal of polyethylene and polyvinyl chloride microplastics in tap water at various concentrations of microplastics ($[\text{Al-coagulant}]_0 = 0.05$ g/L, pH = 7).

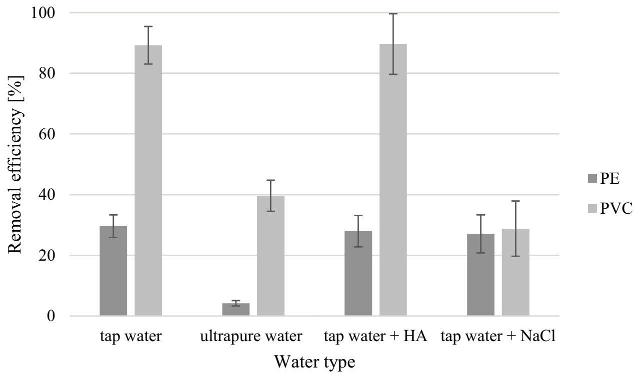

Fig. 4. Efficiency of removal of polyethylene and polyvinyl chloride microplastics in different types of water ($[PE]_0 = [PVC]_0 = 0.1$ g/L, $[\text{Al-coagulant}]_0 = 0.05$ g/L, pH = 7).

of microplastics. Microplastic PE removal was best at a concentration of 0.05 g/L, while the best PVC removal efficiency was reported at a concentration of 0.1 g/L.

The possibility of using the tested coagulation to remove microplastics from ultrapure water, tap water with HA, and tap water with NaCl salt was also checked. The results obtained were compared with the results for coagulation in tap water (Fig. 4). The PE microparticles were removed best from tap water (29.62%), then from tap water + NaCl (27.93%), tap water + HA (27.03%) and the worst from ultrapure water (4.17%). The additives and constituents of tap water used promoted coagulation and thus removal of PE. For example, microplastic removal efficiencies are higher in the present of CO_3^{2-}, due to the fact that the presence of these ions makes the solution alkaline and promotes the hydrolysis of the coagulants [32]. In turn, the efficiency of the removal of PVC was the highest in tap water + HA (89.66%) and in tap water (89.24%). The lowest effectiveness was observed in tap water with NaCl addition (28.79%). Unlike in the studies by Zhou et al. [32], the presence of a large amount of Cl^- ions had a significant impact on the PVC removal efficiency.

3.2. Effect of SDBS addition on PE and PVC removal efficiency

The effect of SDBS addition (20 mg/L) on the efficiency of PE and PVC removal is shown in Fig. 5. The addition of surfactant can also simulate the average concentration of surfactant in wastewater [30]. In the absence of coagulant, the removal efficiency of PE was 39.41%, while that of PVC was approximately 92.77%. Sodium dodecylbenzene sulfonate is a typical example of anionic surfactants with properties of detergency, moistening, foaming, emulsification, and dispersity. SDBS is commonly used as a detergent, emulsifier, and antistatic agent [48]. The effect of the presence of surfactant was analyzed for doses of $AlCl_3 \cdot 6H_2O$ and $FeCl_3 \cdot 6H_2O$ at a concentration of 0.025, 0.05 and 0.1 g/L. The addition of SDBS significantly increased the efficacy of PE and PVC removal in Al salt and Fe salt coagulation. For the coagulant dose of 0.025 g/L, for both tested coagulants, the effectiveness of PE and PVC removal was greater than 90%. In a study by Shen et al. [46], a SDBS was used only to ensure that the PE microplastics are completely dispersed in water. Microplastics are hydrophobic organic micropollutants, while the addition of surfactants changes their physical and chemical properties. Coexisting surfactants can be adsorbed into MPs and change their hydrodynamic attributes. The addition of anionic surfactants does not hinder the coagulation removal of MPs. However, nonionic surfactants can lead to a surfactant stealth effect that may lead to an apparent decrease in coagulation removal efficiency and possibly increase their discharge quantity up to tens of times in the

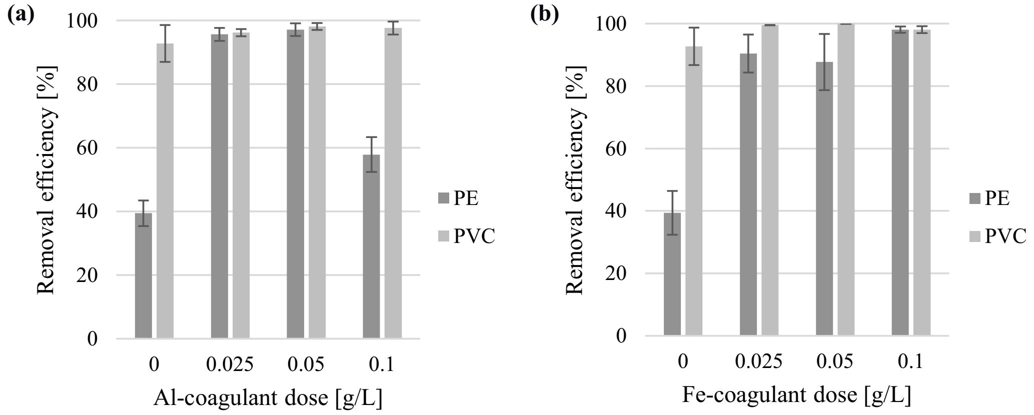

Fig. 5. Impact of the addition of SDBS (20 mg/L) on the removal efficiency of polyethylene and polyvinyl chloride microplastics in tap water ($[PE]_0$ = $[PVC]_0$ = 0.1 g/L, $[\text{Al-coagulant}]_0$ = $[\text{Fe-coagulant}]_0$ = 0.05 g/L, pH = 7).

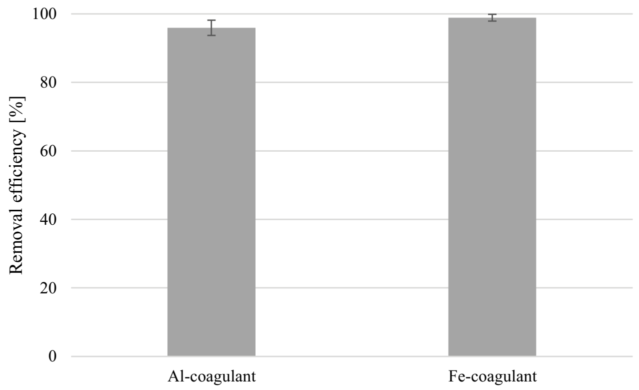

Fig. 6. Efficiency of removal of mixed polyethylene and polyvinyl chloride microplastics in tap water ($[MPs]_0$ = 0.1 g/L (0.05 g/L of polyethylene + 0.05 g/L of polyvinyl chloride), $[\text{coagulant}]_0$ = 0.05 g/L, pH = 7).

effluent. As non-ionic surfactants are frequently used in our lives, eg foods and personal care products, extra attention should be paid to non-ionic surfactants when we try to eliminate MPs from wastewater by coagulation and other water treatment processes [49].

The studies also attempted to use assisted coagulation in tap water that contains a mixture of PE and PVC in equal amounts by weight. The efficiency of microplastic removal in coagulation with the use of Al- and Fe-coagulants was obtained at a level of 95.92% and 98.9%, respectively (Fig. 6). This is a very important aspect, as a result of the presence of a mixture of microplastics in water and sewage, with no single materials. Therefore, studies on the removal of pollutants should not concern individual substances, but the whole group of them. Often the method developed and its parameters are effective in removing only this one substance. For industrial-scale applications in water and wastewater treatment technology, universal technology capable of removing a wide variety of pollutants is required.

After the coagulation process, the unremoved PE and PVC particles were analyzed to determine which particles coagulated less. For this, the dried microplastics were transferred to Kevley slides and analyzed using LDIR.

Table 4
Percentages of individual microplastics

Microplastic	Percentages		
	Before coagulation	After Al-coagulation	After Fe-coagulation
Polyethylene	86.1%	90.3%	79.6%
Polyvinyl chloride	13.9%	9.7%	20.4%

The percentages of individual microplastics before and after the coagulation processes are presented in Table 4. The use of $AlCl_3 \cdot 6H_2O$ coagulant was observed to increase the percentage of PE from 86.1% to 90.3%, while in coagulation with $FeCl_3 \cdot 6H_2O$ it decreased to 79.6%. In the case of PVC, a reverse trend was observed. This means that PE is more prone to be removed by Fe coagulant, while PVC microplastic is more prone to Al-salt coagulation.

4. Conclusions

Most studies on microplastics have focused on their sources, distributions, detection techniques, and ecotoxicological impacts, particularly in the ocean. Microplastics have become a major problem across the globe. Understanding the microscopic plastic removal features within the current drinking water treatment methods is crucial given their gradual discovery in surface water. Therefore, in this study, the removal behavior of PE and PVC was investigated during traditional coagulation and coagulation supported by SDBS, with the main conclusions as follows. This study has shown that single microplastics (PE and PVC) and their combinations may be effectively removed from tap water using the coagulation procedure. In this study, different concentrations of $AlCl_3 \cdot 6H_2O$ and $FeCl_3 \cdot 6H_2O$ at various pH levels of the solution. The use of the Al salt coagulant is more effective than that of the Fe salt coagulant in removing PE and PVC microparticles. The best removal efficiency for both PE and PVC was reported for the dose of 0.05 g/L. Finding the ideal dose of the coagulant is essential because microplastic efficacy is not improved by using doses that are either too low or too high. The removal efficiency of PE and PVC was the highest in water at pH 7. The best coagulation efficiency was observed in tap water compared to ultrapure water, tap water with humic acid, and tap water with NaCl. The effectiveness of coagulation was affected by the initial concentration of microplastics in the water. Low and high concentrations have a negative impact on the effectiveness of microplastic removal from tap water. PE and PVC concentrations of 0.05 and 0.1 g/L, respectively, showed the best performance. The results indicate that the coagulation process is effective up to a certain degree for removing PE and PVC microplastics, but the addition of surfactant increased the removal efficiency even to 100%. It is feasible to lower the dose of coagulant while maintaining high effectiveness of PE and PVC removal by adding SDBS (20 mg/L). SDBS-assisted coagulation is effective in removing a mixture of PE and PVC using both $AlCl_3{\cdot}6H_2O$ and $FeCl_3{\cdot}6H_2O$.

More research on understanding the MPs removal mechanism needs to be investigated. Both laboratory and pilot-scale applications of the coagulation process for the elimination of MPs from tap water need to be investigated in future work. More research is needed to apply assisted coagulation to remove a wide range of microplastics and their mixtures present in the aquatic environment.

Funding

This publication reflects the implementation of Research Project No. 2021/05/X/ST8/00567 funded by the Poland National Science Center.

References

[1] B. Mrowiec, Microplastic-environmental and drinking water problem, Desal. Water Treat., 186 (2020) 65–71.
[2] H. Golwala, X. Zhang, S.M. Iskander, A.L. Smith, Solid waste: an overlooked source of microplastics to the environment, Sci. Total Environ., 769 (2021) 144581, doi: 10.1016/j.scitotenv.2020.144581.
[3] A.D. Vethaak, J. Legler, Microplastics and human health, Science, 371 (2021) 672–674.
[4] M. Kida, S. Ziembowicz, P. Koszelnik, CH_4 and CO_2 emissions from the decomposition of microplastics in the bottom sediment—preliminary studies, Environments, 9 (2022) 91, doi: 10.3390/environments9070091.
[5] M.N. Issac, B. Kandasubramanian, Effect of microplastics in water and aquatic systems, Environ. Sci. Pollut. Res., 28 (2021) 19544–19562.
[6] C. Wang, J. Zhao, B. Xing, Environmental source, fate, and toxicity of microplastics, J. Hazard. Mater., 407 (2021) 124357, doi: 10.1016/j.jhazmat.2020.124357.
[7] Y. Liu, W. Liu, X. Yang, J. Wang, H. Lin, Y. Yang, Microplastics are a hotspot for antibiotic resistance genes: progress and perspective, Sci. Total Environ., 773 (2021) 145643, doi: 10.1016/j.scitotenv.2021.145643.
[8] P. Krystynik, K. Strunakova, M. Syc, P. Kluson, Notes on common misconceptions in microplastics removal from water, Appl. Sci., 11 (2021) 5833, doi: 10.3390/app11135833.
[9] M. Enfrin, L.F. Dumée, J. Lee, Nano/microplastics in water and wastewater treatment processes–origin, impact and potential solutions, Water Res., 161 (2019) 621–638.
[10] M. Cole, P. Lindeque, C. Halsband, T.S. Galloway, Microplastics as contaminants in the marine environment: a review, Mar. Pollut. Bull., 62 (2011) 2588–2597.
[11] Y. Pico, A. Alfarhan, D. Barcelo, Nano-and microplastic analysis: focus on their occurrence in freshwater ecosystems and remediation technologies, TrAC, Trends Anal. Chem., 113 (2019) 409–425.
[12] S.M. Mintenig, I. Int-Veen, M.G. Löder, S. Primpke, G. Gerdts, Identification of microplastic in effluents of wastewater treatment plants using focal plane array-based micro-Fourier-transform infrared imaging, Water Res., 108 (2017) 365–372.
[13] S. Ziajahromi, P.A. Neale, F.D. Leusch, Wastewater treatment plant effluent as a source of microplastics: review of the fate, chemical interactions and potential risks to aquatic organisms, Water Sci. Technol., 74 (2016) 2253–2269.
[14] J. Talvitie, M. Heinonen, J.P. Pääkkönen, E. Vahtera, A. Mikola, O. Setälä, R. Vahala, Do wastewater treatment plants act as a potential point source of microplastics? preliminary study in the coastal Gulf of Finland, Baltic Sea, Water Sci. Technol., 72 (2015) 1495–1504.
[15] S.A. Carr, J. Liu, A.G Tesoro, Transport and fate of microplastic particles in wastewater treatment plants, Water Res., 91 (2016) 174–182.
[16] Y. Chae, Y.J. An, Effects of micro-and nanoplastics on aquatic ecosystems: current research trends and perspectives, Mar. Pollut. Bull., 124 (2017) 624–632.
[17] M. Shen, T. Hu, W. Huang, B. Song, M. Qin, H. Yi, Y. Zhang, Can incineration completely eliminate plastic wastes? an investigation of microplastics and heavy metals in the bottom ash and fly ash from an incineration plant, Sci. Total Environ., 779 (2021) 146528, doi: 10.1016/j.scitotenv.2021.146528.
[18] J. Li, H. Liu, J.P. Chen, Microplastics in freshwater systems: a review on occurrence, environmental effects, and methods for microplastics detection, Water Res., 137 (2018) 362–374.
[19] E. Wiśniowska, M. Włodarczyk-Makuła, Evaluation of the adsorption efficiency of carcinogenic PAHs on microplastic (polyester) fibers—preliminary results, Appl. Water Sci., 12 (2022) 1–7.
[20] W. Gao, Y. Zhang, A. Mo, J. Jiang, Y. Liang, X. Cao, D. He, Removal of microplastics in water: technology progress and green strategies, Green Anal. Chem., 3 (2022) 100042, doi: 10.1016/j.greeac.2022.100042.
[21] B. Ma, W. Xue, C. Hu, H. Liu, J. Qu, L. Li, Characteristics of microplastic removal via coagulation and ultrafiltration during drinking water treatment, Chem. Eng. J., 359 (2019) 159–167.
[22] H. Park, B. Park, Review of microplastic distribution, toxicity, analysis methods, and removal technologies, Water, 13 (2021) 2736, doi: 10.3390/w13192736.
[23] N. Yahyanezhad, M.J. Bardi, H. Aminirad, An evaluation of microplastics fate in the wastewater treatment plants: frequency and removal of microplastics by microfiltration membrane, Water Pract. Technol., 16 (2021) 782–792.
[24] X. Shi, X. Zhang, W. Gao, Y. Zhang, D. He, Removal of microplastics from water by magnetic nano-Fe_3O_4, Sci. Total Environ., 802 (2022) 149838, doi: 10.1016/j.scitotenv.2021.149838.
[25] Y. Tang, S. Zhang, Y. Su, D. Wu, Y. Zhao, B. Xie, Removal of microplastics from aqueous solutions by magnetic carbon nanotubes, J. Chem. Eng., 406 (2021) 126804, doi: 10.1016/j.cej.2020.126804.
[26] F. Miao, Y. Liu, M. Gao, X. Yu, P. Xiao, M. Wang, X. Wang, Degradation of polyvinyl chloride microplastics via an electro-Fenton-like system with a TiO_2/graphite cathode, J. Hazard. Mater., 399 (2020) 123023, doi: 10.1016/j.jhazmat.2020.123023.
[27] M. Kiendrebeogo, M.R. Karimi Estahbanati, A.K. Mostafazadeh, P. Drogui, R.D. Tyagi, Treatment of microplastics in water by anodic oxidation: a case study for polystyrene, Environ. Pollut., 269 (2021) 116168, doi: 10.1016/j.envpol.2020.116168.
[28] I. Nabi, K. Li, H. Cheng, T. Wang, Y. Liu, S. Ajmal, L. Zhang, Complete photocatalytic mineralization of microplastic on TiO_2 nanoparticle film, iScience, 23 (2020) 101326, doi: 10.1016/j.isci.2020.101326.
[29] C.N. Muhonja, H. Makonde, G. Magoma, M. Imbuga, Biodegradability of polyethylene by bacteria and fungi from Dandora dumpsite Nairobi-Kenya, PloS One, 13 (2018) e0198446, doi: 10.1371/journal.pone.0198446.

[30] H.S. Auta, C.U. Emenike, S.H. Fauziah, Screening of *Bacillus* strains isolated from mangrove ecosystems in Peninsular Malaysia for microplastic degradation, Environ. Pollut., 231 (2017) 1552–1559.

[31] S. Dobretsov, R.M. Abed, M. Teplitski, Mini-review: inhibition of biofouling by marine microorganisms, Biofouling, 29 (2013) 423–441.

[32] G. Zhou, Q. Wang, J. Li, Q. Li, H. Xu, Q. Ye, J. Zhang, Removal of polystyrene and polyethylene microplastics using PAC and $FeCl_3$ coagulation: performance and mechanism, Sci. Total Environ., 752 (2021) 141837, doi: 10.1016/j.scitotenv.2020.141837.

[33] S. Monira, M.A. Bhuiyan, N. Haque, B.K. Pramanik, Assess the performance of chemical coagulation process for microplastics removal from stormwater, Process Saf. Environ. Prot., 155 (2021) 11–16.

[34] D.W. Skaf, V.L. Punzi, J.T. Rolle, K.A. Kleinberg, Removal of micron-sized microplastic particles from simulated drinking water via alum coagulation, Chem. Eng. J., 38 (2020) 123807, doi: 10.1016/j.cej.2019.123807.

[35] M. Shen, Y. Zhang, E. Almatrafi, T. Hu, C. Zhou, B. Song, G. Zeng, Efficient removal of microplastics from wastewater by an electrocoagulation process, Chem. Eng. J., 428 (2022) 131161, doi: 10.1016/j.cej.2021.131161.

[36] https://mpwik.rzeszow.pl, [Access: 26.08.2022].

[37] C. Akarsu, H. Kumbur, A.E. Kideys, Removal of microplastics from wastewater through electrocoagulation-electroflotation and membrane filtration processes, Water Sci. Technol., 84 (2021) 1648–1662.

[38] J. Wang, Z. Tan, J. Peng, Q. Qiu, M. Li, The behaviors of microplastics in the marine environment, Mar. Environ. Res., 113 (2016) 7–17.

[39] X. Wen, C. Du, P. Xu, G. Zeng, D. Huang, L. Yin, R. Deng, Microplastic pollution in surface sediments of urban water areas in Changsha, China: abundance, composition, surface textures, Mar. Pollut. Bull., 136 (2018) 414–423.

[40] Y. Zheng, J. Li, W. Cao, X. Liu, F. Jiang, J. Ding, C. Sun, Distribution characteristics of microplastics in the seawater and sediment: a case study in Jiaozhou Bay, China, Sci. Total Environ., 674 (2019) 27–35.

[41] K. Wu, H. Wang, R. Liu, X. Zhao, H. Liu, J. Qu, Arsenic removal from a high-arsenic wastewater using in situ formed Fe–Mn binary oxide combined with coagulation by poly-aluminum chloride, J. Hazard. Mater., 185 (2011) 990–995.

[42] P. Krystynik, K. Strunakova, M. Syc, P. Kluson, Notes on common misconceptions in microplastics removal from water, Appl. Sci., 11 (2021) 5833, doi: 10.3390/app11135833.

[43] M. Sillanpää, M.C. Ncibi, A. Matilainen, M. Vepsäläinen, Removal of natural organic matter in drinking water treatment by coagulation: a comprehensive review, Chemosphere, 190 (2018) 54–71.

[44] Q. Xiangli, Z. Zhenjia, W. Nongcun, V. Wee, M. Low, C.S. Loh, N.T. Hing, Coagulation pretreatment for a large-scale ultrafiltration process treating water from the Taihu River, Desalination, 230 (2008) 305–313.

[45] X. Han, H. Lu, Y. Gao, X. Chen, M. Yang, The role of in situ Fenton coagulation on the removal of benzoic acid, Chemosphere, 238 (2020) 124632, doi: 10.1016/j.chemosphere.2019.124632.

[46] M. Shen, B. Song, G. Zeng, Y. Zhang, F. Teng, C. Zhou, Surfactant changes lead adsorption behaviors and mechanisms on microplastics, Chem. Eng. J., 405 (2021) 126989, doi: 10.1016/j.cej.2020.126989.

[47] M. Ahsan, A. Riaz, M.J. Jaskani, M. Hameed, Physiological and anatomical response of fragrant *Rosa* species with treated and untreated wastewater, Int. J. Agric. Biol., 19 (2017) 17957, doi: 10.17957/IJAB/15.0160.

[48] IRO Coating Additive. Available at: https://www.irocoatingadditive.com, [Access: 26.08.2022].

[49] Y. Xia, X.M. Xiang, K.Y. Dong, Y.Y. Gong, Z.J. Li, Surfactant stealth effect of microplastics in traditional coagulation process observed via 3-D fluorescence imaging, Sci. Total Environ., 729 (2020) 138783, doi: 10.1016/j.scitotenv.2020.138783.