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Effect of embedding a sieving phase into the current plastic recycling process to capture microplastics

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ABSTRACT

This study proposes a systematic change to the current plastic recycling process by introducing a sieving stage in between the shredding and washing units to capture the microplastics being unintentionally generated and released. The benefit of adding the sieving stage to minimise microplastics release to wash water was highlighted by comparing the findings with the case where microplastics are released to wash water and a conventional coagulation process is used to remove microplastics from water. Two coagulants, aluminium sulphate ($Al_2(SO_4)_3.18H_2O$) and aluminium chloride ($AlCl_3.6H_2O$), were used to remove polyethylene terephthalate (PET) and polycarbonate (PC) from water. The size of the microplastic particles played a significant role on the removal efficiency. The maximum removal efficiency of PET by $AlCl_3.6H_2O$ was 99.2 % for the particles in 1.18–5 mm range, whereas the average removal efficiency over the whole tested size range of 0.15–5.00 mm was 76.1 % for the same plastic-coagulant combination. By contrast, the addition of a 5 mm sieve between the shredding and the washing units was found to capture 96–97 % of the microplastics generated. The findings of this innovative experiment demonstrate the beneficial impact that this strategy has on capturing microplastics prior to entering water matrix.

1. Introduction

The emerging environmental concerns that are associated with microplastic pollution have been driving research and innovation within the current literature. Microplastics are known to act as vectors for a wide range of pollutants [1,2] and can have detrimental toxicity effects on both fauna and flora [3]. The significant abundance of microplastics in the environment [4–6] and the limited available remediation procedures that completely collect the pollutant once in the environment are some of the greatest issues associated with the contaminant [7]. This prompts the need to identify potential sources of microplastic pollution and develop effective preventative measures that reduces the risk of environmental emissions of microplastics.

Recent research has discovered that the processes involved with the recycling of plastic waste may also be linked to the unintentional generation and environmental emission of microplastics [8–11]. When plastic waste enters a recycling facility, it is reduced in size through the process of shredding, which due to its mechanical nature, can generate large quantities of unwanted microplastics [12]. After the shredding process, the plastic waste material is then washed to remove any contaminates such as food waste or adhesives [13]. It is in this process where microplastics may unintentionally be left in the wastewater and pollute the environment [9,11].

Many plastic recycling facilities have onsite wastewater treatment plants targeted specifically for odour reduction and water recycling [14,15]. However, since microplastics were not a contaminant of concern at plastic recycling facilities until now, the wastewater treatment options may not have been designed to capture the pollutant. If environmental regulations are not as strict in the location of the recycling facility, there is also the potential that the wash water is directly emitted to the environment without any treatment [8].

The use of coagulation for microplastic removal has been well researched as it is a cost effective and simple wastewater treatment process [16–19]. Some studies have highlighted that coagulation can achieve microplastic removal efficiencies >90 % [20–24]. However, coagulation has not always been shown to be so effective. For example,

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the removal efficiency of polyethylene (PE) by the coagulant ferric chloride was only 29.7 % [25]. The type of plastic that is being processed by a plastic recycling facility will therefore determine if coagulation would be an effective method for removing microplastics from the wash water. There are still significant gaps in the current literature as most studies have focused primarily on the effectiveness of coagulation on the removal of PE and polystyrene (PS).

The purpose of this study is to investigate two approaches for capturing microplastics that are being generated during the plastic recycling process. The first method is to examine the effectiveness of coagulation on the removal of microplastics from the wash water. The focus will be on using two common coagulants (aluminium sulphate ($Al_2(SO_4)_3.18H_2O$) and aluminium chloride ($AlCl_3.6H_2O$)) to remove polyethylene terephthalate (PET) and polycarbonate (PC) from water. The second method is to investigate a systematic change to the current plastic recycling system, whereby an additional process is added between the shredding phase and the washing phase. The systematic change was developed based on the idea of preventative measures being more effective than remedial measures. The preventative measure is to capture the microplastics by dry sieving the shredded plastics prior to the material being washed. As far as the authors are aware, this is the first experimental paper to investigate a systematic change in the current plastic recycling process to capture microplastics being unintentionally generated.

2. Materials and methods

2.1. Removal of microplastics through pre-wash sieving

The effect of sieving plastic waste prior to the wash phase in a plastic recycling facility was simulated within a laboratory setting. Fig. 1 depicts two scenarios. Scenario 1 is the current process for washing plastic waste at a recycling facility [13]. Scenario 2 is the proposed systematic process change that aims to capture microplastics before entering the wash water.

2.1.1. Materials and sample preparation

Virgin polyethylene terephthalate (PET) bottles and polycarbonate (PC) sheets were commercially purchased and reduced in size using a GP20 hybrid plastic shredder (3devo, The Netherlands). Using a 5 mm sieve (microplastics size limit), a portion of the dry shredded plastics was subjected to a single pass through a sieve using the ASTM D1921-01 (ASTM, 2017) standard. This involved the mechanical shaking of the sieve and shredded plastic material for 10 min. The material collected on the 5 mm sieve was used for the analysis of Scenario 2 (Fig. 1).

2.1.2. Simulation of the washing process

To determine the effectiveness of sieving shredded plastics prior to the washing stage on the reduction of microplastics remaining in the wash water, a lab scaled simulation of the washing process was performed. The methodology used to simulate the wash process was adapted from previous literature [14]. 10.00 g (M_1) of dried shredded plastic was accurately weighed (minimum range of 1.0×10^{-4} g) and was placed into a conical flask filled with 250 mL of tap water. The mixture of shredded plastics and water was mixed using a multi-flask rotary shaker. The rotation speed was set to 200 RPM for 10 min. After mixing is completed, the contents of the conical flask are emptied over a 5 mm sieve, representing the mesh screen paddles (Fig. 1) used in the plastic recycling industry to remove the washed plastic flakes from the wash bay. The conical flask is rinsed thoroughly and emptied onto the sieve to ensure all material is collected. The water and contents passing through the sieve are collected. The material collected on the sieve is then oven dried, accurately weighed and recorded (M_2). Results were reported as grams of microplastics remaining in the wash water per kg of washed plastics. All experiments were repeated in triplicates (n = 3) for repeatability.

2.2. Lab-scale coagulation treatment of microplastics in water

2.2.1. Materials and sample preparation

Virgin polyethylene terephthalate (PET) bottles and polycarbonate

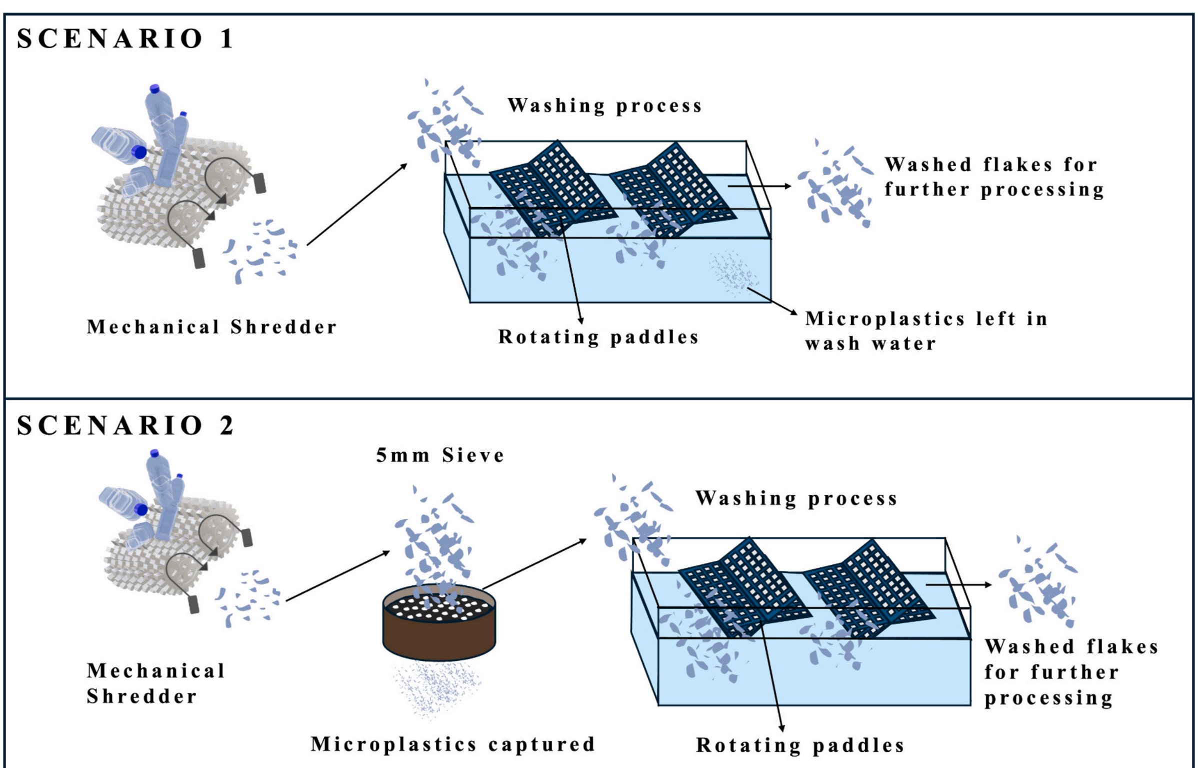

Fig. 1. Experimental schematic to determine the influence of dry sieving on the reduction of microplastic contamination in the wash water.

(PC) were shredded and separated into the four size ranges of 0.15–0.5 mm, 0.5–0.85 mm, 0.85–1.18 mm, 1.18–5 mm for further analysis. Analytical reagent grade aluminium sulphate ($Al_2(SO_4)_3.18H_2O$) and aluminium chloride hexahydrate ($AlCl_3.6H_2O$) and RCI premium grade 32 % hydrochloric acid (HCl) were purchased from ChemSupply, Australia.

2.2.2. Coagulation analysis

To simulate the removal of microplastics using coagulation, a JLT6 flocculation apparatus (VELP Scientifica, Italy) was employed. $Al_2(SO_4)_3.18H_2O$ and $AlCl_3.6H_2O$ were used in this study as aluminium based coagulants are widely used in the wastewater treatment industry [26]. The methodology used was adapted from previous literature [21,22,27]. Microplastics in the size range of 0.5–0.85 mm were accurately measured and recorded using a precision scale (minimum range 1.0×10^{-4} g) and then mixed with tap water in glass mixing beakers to make the desired microplastic concentration of 50 mg/L. Six predefined coagulant concentrations (0, 20, 40, 60, 80, 100 mg/L) were added to the beakers during a slow mixing stage (50 RPM for 30 s). Rapid mixing (200 RPM) was maintained for 60 s to ensure the coagulants dissolved. This was followed by a flocculation period (50 RPM for 10 min). A final settling period (0 RPM for 30 min) occurred prior to further analysis of removal efficiency. A pH and conductivity meter were used pre and post coagulation to determine the effect of the coagulants on the characteristics of water matrix. This process was repeated in triplicates ($n = 3$) to confirm reproducibility of data.

To elucidate the effect of microplastic size and concentration on the removal efficiency of coagulation, the methodology depicted in Fig. 2 was followed. To elucidate the effect of microplastic size and concentration on the removal efficiency of coagulation, the methodology depicted in Fig. 2 was followed. The coagulant concentration that yielded the highest microplastic removal efficiency was selected as the standard for subsequent experiments. The impact of particle size on the efficiency of the coagulation treatment was analysed across four predefined microplastic size ranges (0.15–0.5 mm, 0.5–0.85 mm, 0.85–1.18 mm, and 1.18–5 mm). In all experiments assessing the effect of microplastic size, the concentration was maintained at 50 mg/L. To evaluate the influence of microplastic concentration on coagulation removal efficiency, three predefined concentrations (50 mg/L, 100 mg/L, and 150 mg/L) were tested, with the particle size range held constant at 0.5–0.85 mm. The coagulation treatment protocol described above was applied uniformly across all experiments investigating the effects of microplastic size and concentration on removal efficiency.

To verify the testing procedure, laboratory blanks and spikes were utilised. To reduce the risk of unwanted microplastic contamination, glass beakers were used for the jar test procedure and glass conical flasks were used in the simulated washing procedure.

2.2.3. Removal efficiency calculation

To measure the microplastics removal efficiency by conventional coagulation a previously reported protocol was followed [27]. Microplastics were washed with 1 M HCl to remove residues and oven dried at 60 °C for 12 h. Precise weights of microplastics (measurable to 1.0×10^{-4} g) were added to jar tests (M_1). After the jar test had concluded, the accumulated microplastics at the bottom were carefully removed with a 50 mL syringe. 1 M HCl solution was used to remove residues off microplastic samples and were vacuum filtered using a 0.45 μm membrane. Microplastics were carefully scraped from the membrane and oven dried at 60 °C for 12 h. Microplastics were subsequently weighed (M_2), and a removal efficiency was determined.

The removal efficiency was then calculated using the following equation:

$$\textit{Removal efficiency}\ (\%) = \frac{M_1 - M_2}{M_1} \times 100\% \tag{1}$$

Where M_1 is known mass of microplastic placed into the jar test and M_2 is the mass of the microplastics collected.

2.2.4. Statistical analysis

To determine if microplastic concentration and size influenced the removal efficiency of microplastics using coagulation an ANOVA single factor analysis was performed using Microsoft Excel [28]. A significant variation between the means of the two populations was reported with a $p < 0.05$, indicating a rejection of the null hypothesis.

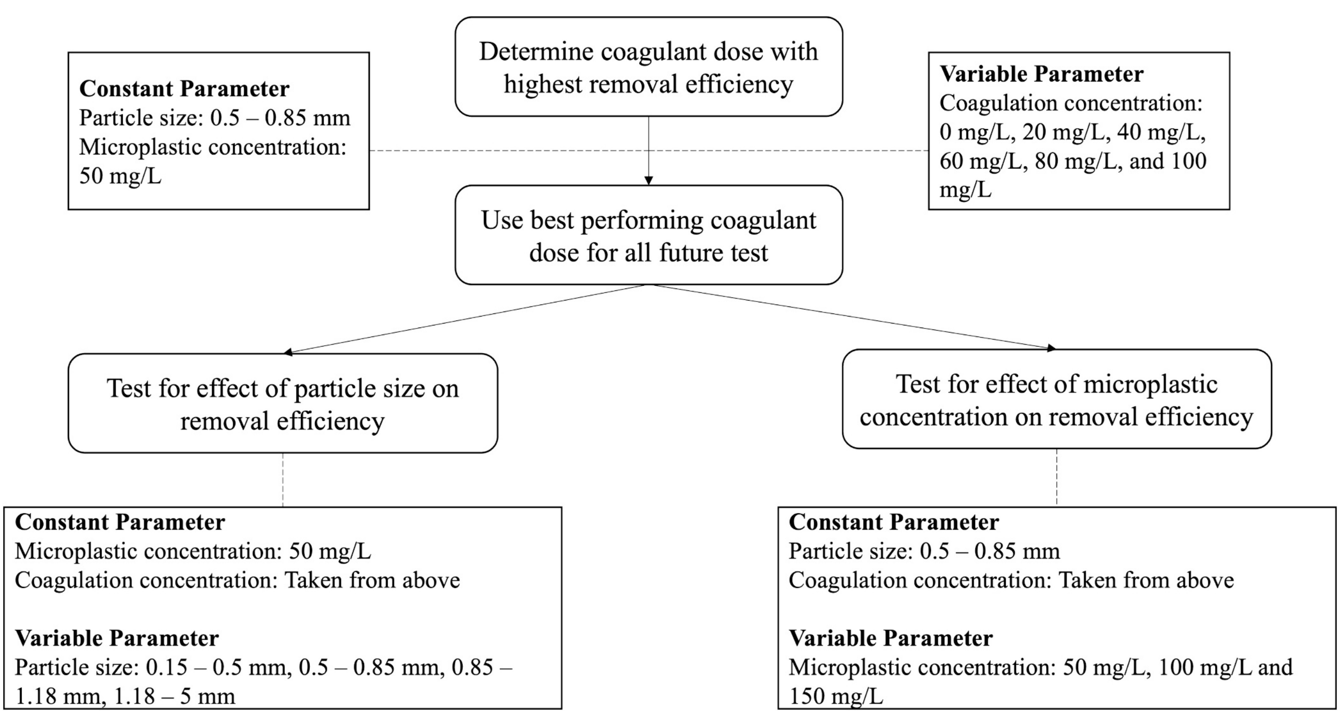

Fig. 2. Methodology used to determine the effect of microplastic size and concentration on the removal efficiency of microplastics by coagulation (based on [20,25]).

3. Results and discussion

3.1. Removal of microplastics using sieves

Fig. 3 depicts a significant reduction in microplastics remaining in the wash water when a dry sieve was implemented immediately after the shredding process. A consistent result was obtained regardless of the plastic type with PET and PC achieving a 96.8 ± 0.8 % and 96.3 ± 0.9 % reduction, respectively. Within this experimental design, the shredded plastic was subjected to only a single pass through the 5 mm sieve under the ASTM D1921-01 [43] standard. It is suspected that 100 % reduction was not achieved through a single pass as microplastics may be attached to the cracks and folds found in larger plastic that are produced as a consequence of being mechanically shredded [10]. Once the shredded plastics enter the washing phase, the contact with water allows for the microplastics to detach from the larger plastic flakes and remain in the water matrix. This process of adsorption and desorption of microplastics from larger plastic flakes is the likely cause for the microplastics remaining in the wash water as seen in Fig. 3. This is the first study investigating a new approach to capture microplastics generated from the plastic recycling process. The design and protocol for the sieving process are still in its infancy, however the results derived from this novel experiment show the significant effect this approach can have on capturing microplastics that are unintentionally being generated.

3.2. Removal of microplastics via coagulation

Within this section, the efficiency of the coagulation process for removing microplastics was assessed. The effect of the type of the coagulant ($Al_2(SO_4)_3.18H_2O$ and $AlCl_3.6H_2O$), the concentration of coagulant (0–100 mg/L), the size of microplastics (0.15–5.00 mm), and the microplastic concentration (50–150 mg/L) on the microplastic removal efficiency by coagulation was investigated. Optimal coagulation doses that were derived in Section 3.2.1 were maintained for experiments conducted in Section 3.2.2.

3.2.1. Optimal coagulation concentration

The concentrations of $Al_2(SO_4)_3.18H_2O$ and $AlCl_3.6H_2O$ were increased from 0 to 100 mg/L to determine the optimal dose for microplastics removal (Fig. 4). For $Al_2(SO_4)_3.18H_2O$, the optimal dosage for the removal of PET and PC was found to be 40 mg/L. The optimal dosage for removing PET and PC using $AlCl_3.6H_2O$ was 20 mg/L. Investigation of the pH and conductivity at the end of coagulation process revealed that the maximum microplastic removal for both coagulants, regardless of the dosage, occurred at a pH of 5.9 and an electrical conductivity of 130 µs/cm (Fig. 4). Previous research has indicated the importance of pH and conductivity on the efficiency of coagulation due to its relationship to the microplastics zeta potential [29]. The conductivity and pH of the solution therefore plays a critical role in the effectiveness of coagulation, as the underlying mechanism is to manipulate the zeta potential of the microplastics with the aim to generate colloids that will settle and can be removed [17,30]. For both coagulants, after the optimal coagulant concentration is achieved the removal efficiency begins to decrease. This is likely due to the pH decreasing to a level whereby electrostatic interactions within the water matrix become unfavourable for the formation of colloids [22,31].

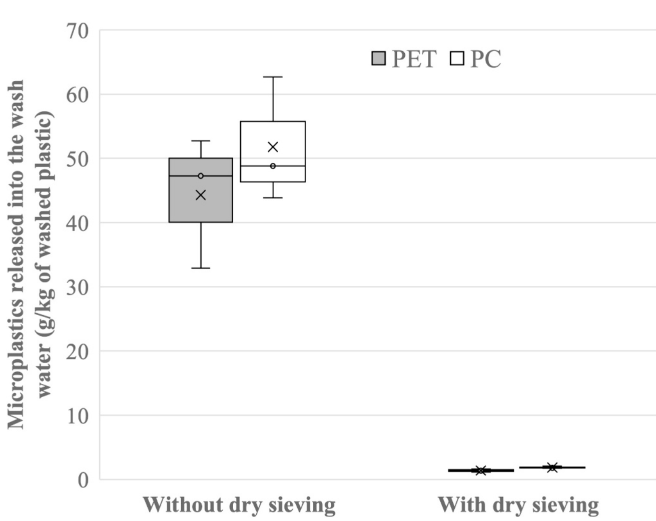

Fig. 3. Effect of dry sieving shredded plastic flakes on preventing microplastics from entering the wash water. Error bars are showing mean, median, and standard deviation for $n = 3$.

Maximum removal efficiency using $Al_2(SO_4)_3.18H_2O$ for PET and PC was found to be 66.1 ± 3.7 % and 32.9 ± 4.0 %, respectively. For $AlCl_3.6H_2O$, maximum removal efficiency for PET and PC was 66.3 ± 6.7 % and 43.9 ± 3.4 %, respectively. The differences in removal efficiencies between PET and PC is potentially linked to the variances in their densities (PET = 1.38 g/cm^3 and PC = 1.20 g/cm^3). As PET is denser, less flocs are required to form on the microplastic surface to bring it out of suspension. This finding is consistent with a previous study that investigated the effect of coagulation on the removal of PE and polyvinyl chloride (PVC) microplastics [32]. Maximum removal efficiency for PE was found to be 8.44 % whereas the removal of PVC microplastics by coagulation was up to 89.73 %. This was attributed predominately to the significant differences in density of the two polymers (PE = 0.88–0.96/cm^3 and PVC = 1.38 g/cm^3) allowing PVC to settle out of suspension at greater rates.

Compared to the maximum removal efficiency of 66.1 % of PET in this study, a previous study reported 100 % removal of PET using the same coagulant [21]. One plausible reason for the discrepancies in removal efficiencies may be due to the differences in surface morphology affecting coagulation performance [22]. Microplastics used within this study were generated using a mechanical shredder, which has been shown to create significant deformations on the surface of the particles [10]. The addition of cracks and holes on the surface may attract air bubbles during the coagulation process and therefore reduce the performance of the process. Within the previous study, PET microplastics were commercially purchased and as such may have exhibited a more uniform surface morphology [21]. Bubble entrapment has been seen to disrupted coagulant flocs, causing them to rise instead of settle [33]. Research into the effect of surface morphology on coagulation performance should be further investigated, with a specific focus on the impact of bubble entrapment. Comparable research on the effectiveness of using any coagulant for the removal of PC microplastics is not currently available.

3.2.2. Effect of microplastic size and concentration

The size and concentrations of microplastics in the wash water at a plastic recycling facility will vary significantly. For this reason, the effect that microplastic size and concentration has on the removal efficiency of coagulation was investigated (Fig. 5).

For both coagulants investigated, the microplastic size had a notable effect on the removal efficiencies. In all experiments, microplastic removal was the lowest for the smallest particles (0.15–0.5 mm) and increased drastically to 80–95 % for the largest particle size tested (1.18–5 mm). Significant variance was confirmed through the ANOVA analysis which resulted in $p < 0.05$ for all scenarios tested. The results found within this study challenge a previous study stating smaller particles are removed with higher efficiencies [20]. Lapointe et al. [20] investigated the variations in removal efficiencies of PE microplastic spheres and reported higher removal of 0.015 mm compared to 0.14 mm particles; however, the differences in the removal efficiencies were only 7 %. The study was limited by only testing two sizes which are comparatively closer to one another in the microplastic size range (0.001–5 mm). In the current study, the results from the ANOVA

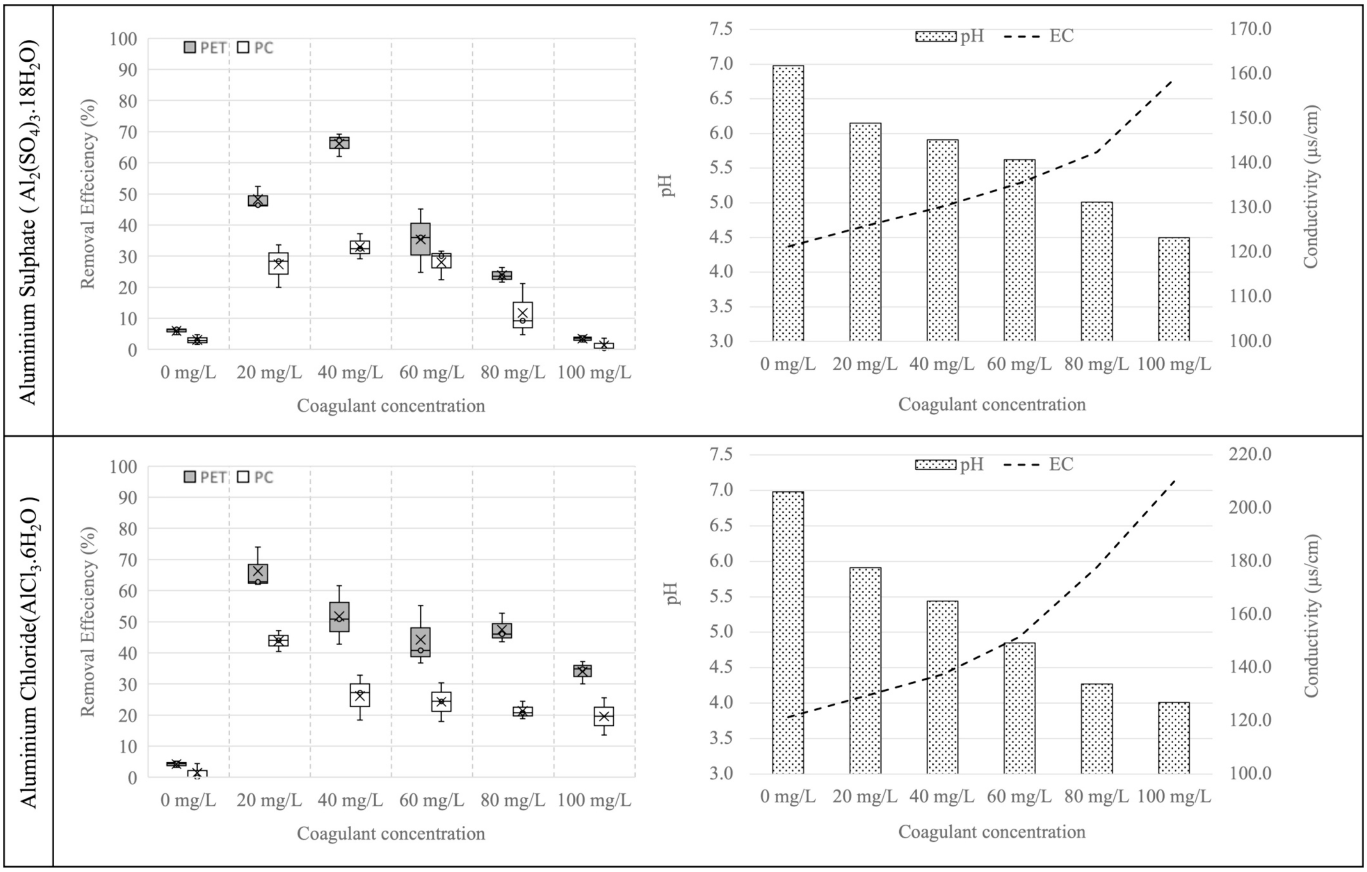

Fig. 4. Determination of optimal coagulation concentration for the removal of PET and PC from water (Box and whiskers). Error bars show mean, median, and standard deviation (n = 3). The bar and line graphs show the effect of coagulant concentration on the pH and conductivity of the water. Microplastic size and concentration was maintained at 0.5–0.85 mm and 50 mg/L, respectively.

analysis indicated that the microplastic concentrations that were investigated (50 mg/L – 150 mg/L) had no significant effect on the removal efficiencies ($p > 0.05$). Our results are consistent with a previous study that showed that variations in microplastic concentrations (100–1000 mg/L) had little effect on the removal efficiencies [25].

To the best of the authors knowledge, this is the first study to investigate the efficacy of coagulation on removing PC microplastics from water. The findings show that when compared to PET, irrespective of the coagulant types or concentrations tested, the removal efficiency was consistently lower. Based on the tested parameters, coagulation was found only to be effective at removing larger PET and PC particles (0.85–5.00 mm). The information gained from this research can be used in conjunction with previous literature to determine optimum polymer-coagulant combinations to remove microplastics from water sources.

3.3. *Preventative measures vs remedial measures*

This research presented an approach to effectively capture and manage the microplastics that are being generated during the plastic recycling process. By capturing microplastics through the use of sieving technology prior to entering the wash water, there is a reduced risk of complications that are associated with the removal of microplastics from water matrices [34]. The effectiveness of the proposed approach was compared to the conventional wastewater treatment option of coagulation, with the experimental results summarised in Table 1. Through the preventative process of sieving, 96–97 % of microplastics generated are captured prior to entering the wash water. In comparison, the highest average removal efficiency for coagulation was 76.1 % when $AlCl_3.6H_2O$ was used to remove PET. Although coagulation was able to reach high removal efficiencies (99.2 % PET (1.18–5 mm) removed using $AlCl_3.6H_2O$), they were not consistent across the coagulation types, size ranges or plastic types tested (Fig. 5). Similar findings are expressed within the literature where it is apparent that there is no universal coagulation methodology that can effectively and consistently remove microplastics from water [29,35–37].

Alternative wastewater treatments options are available that can potentially achieve microplastics removal efficiencies >95 % e.g., membrane bioreactors (MBRs), sand filters and disc filters [38–40], nonetheless, associated issues with the technologies are still prevalent. An investigation into the efficiency of MBRs on removing microplastics found they are capable of achieving a removal rate of 99.69 % from the aqueous phase [39]. However, microplastics then accumulate in the MBR sludge. A common beneficial reuse practice for wastewater treatment sludge is to apply to land as a fertilizer, therefore potentially releasing the microplastics into the environment [41]. Filtration units (sand filters, disc filters etc.) also have the ability to remove microplastics from wastewater, however, they also can incur the issues of fouling and blockages [40] and how to safely re-capture microplastics out of the media.

To the best of the author's knowledge, this method for capturing microplastics generated during the mechanical recycling of plastics has not yet been investigated. The approach offers a preventative measure to capture microplastics before they enter the washing phase where issues with the recovery of the contaminant can occur. The proposed method offers a simple design that can easily be implemented to reduce the unintentional release of microplastics from the plastic recycling industry.

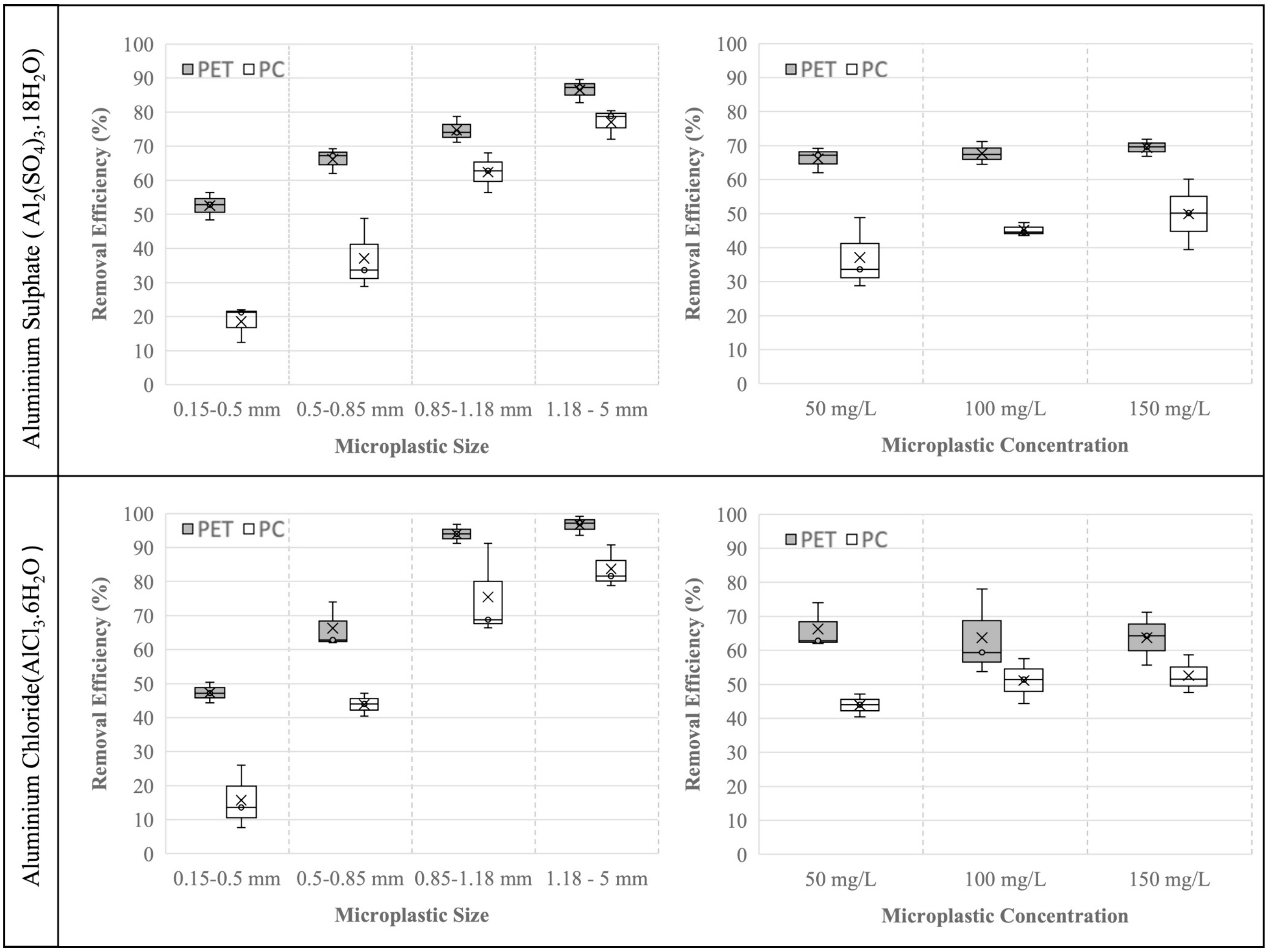

Fig. 5. Effect of microplastic size and concentration on the removal efficiency by coagulation. Coagulation dose was maintained at 40 mg/L for $Al_2(SO_4)_3.18H_2O$ and 20 mg/L for $AlCl_3.6H_2O$ as per optimised dose (Fig. 4). For the microplastic size experiment, microplastic concentration was 50 mg/L. For the microplastic concentration experiment, microplastic size was 0.5–0.85 mm. Error bars show mean, median, and standard deviation for n = 3.

Table 1
Comparison between the two approaches to capture microplastics generated during the plastic recycling process. For coagulation, microplastic removal efficiency is the average removal efficiency over the size ranges 0.15–5 mm (Fig. 5).

Configuration	Coagulant	Plastic Type	Microplastics Removed
Dry sieving[a]	–	PET	96.8 %
		PC	96.3 %
Coagulation	$Al_2(SO_4)_3.18H_2O$	PET	70.0 %
		PC	48.7 %
	$AlCl_3.6H_2O$	PET	76.1 %
		PC	54.7 %

[a] Note: For dry sieving removal % indicate retention before mixing with wash water, while for coagulation, removal % indicate the removal after all the generated microplastics were released into wash water.

4. Recommendations

Based on the findings of this research, the following recommendations have been made for the plastic recycling industry and for future research in this field:

1. Previous research has indicated plastic recycling to be a large generator of microplastic material [8–11]. Due to the simplicity of the technology, sieving shredded plastic waste should be considered and optimised to capture any microplastics that are being generated within the plastic recycling process.
2. This study was conducted on PC and PET. The effectiveness of the additional sieving stage on other widely recycled plastics e.g. high-density polyethylene and polypropylene needs to be investigated.
3. Optimisation of not only the sieving process but also the shredding process to reduce the amount of microplastics being generated needs to be conducted.
4. An investigation into the cost of incorporating a mechanical sieve into the plastic recycling process. Mechanical sieving technologies are used in other industries, e.g., mining [42], to remove impurities and can be used as a comparative for this analysis.
5. Investigation of the particle size distribution of the microplastics remaining in the wash water after dry sieving and determination of factors (shredder used, polymer type, water temperature, detergent used etc.) that influence the distribution.

5. Conclusions

This study proposes a new approach to the current plastic recycling practice that efficiently captures the microplastics that are unintentionally generated through the shredding process. The proposal will see the implementation of a sieving stage being employed between the shredding and washing process. Based on the PC and PET microplastics that were used within this study, sieving the shredded plastics captured

96–97 % of the generated microplastics. Furthermore, this study compared the findings of the proposed method to the conventional wastewater treatment option of coagulation to showcase the benefits of preventative measures compared to remedial measures. The maximum average removal efficiency (76.1 %) was found for PET over the size range of 0.15–5.00 mm using $AlCl_3.6H_2O$. Issues that coagulation currently faces are inconsistencies with removal efficiencies among the coagulants, size ranges and plastic types tested. The simplicity of the sieving process makes this modification appealing and easy to implement in the plastic recycling process.

CRediT authorship contribution statement

Michael J. Staplevan: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Ashley J. Ansari:** Writing – review & editing, Supervision. **Aziz Ahmed:** Writing – review & editing, Supervision. **Faisal I. Hai:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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