



Microplastic removal via physical and chemical methods

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Received 11/7/2022, Accepted, 19/10/2022, Published 15/1/2023

A growing number of synthetic plastics derived from fossil fuels are produced, and improper plastic waste management has caused a lot of pollution problems. There are many microplastics in the environment, and they disintegrate slowly in soil and water. The properties of microplastics include long residence times, high stability, high fragmentation potential, and the ability to adsorb other contaminants. Invertebrates and planktonic organisms are easily able to accumulate microplastics in aquatic species. Therefore, microplastics (MPs) must be removed from the water and other media. This paper aims to review the occurrence, raw polymers and additives, and remediation methods for removing microplastics from the environment. Several methods are available for removing contaminants, including sorption, filtration, and chemical treatments. Various removal methods are discussed along with their methods, efficiency, and advantages.

Keywords: Microplastics; Physical; Chemical.

1. INTRODUCTION

Globally, plastic production increased dramatically from 1950 to 2015 to improve human quality of life [1]. As a result, plastic pollution has increased worldwide, posing a threat to environmental health [1]–[3], Approximately 0.59×10^9 particles of microplastic are released by sewage treatment plants each year into aquatic ecosystems [1]. The term

microplastic refers to plastics smaller than 5 mm in size, formed when many plastic-based products are exfoliated and degraded into ecosystems [4]. Marine sediments [5], urban and rural areas [6], freshwaters [7], and seawaters [8] have all been reported to contain microplastics. According to most studies, microplastics accumulate in aquatic environments, increasing the exposure of living organisms to microplastics and the degradation products created by them [9], [10]. Generally, microplastics (MPs) can be divided into primary microplastics, which are raw materials used in the manufacturing of household and personal care products, whereas secondary microplastics come from discarded materials or remnants of production, which are materials that develop through physical, chemical, and biological degradation in the environment [1], [11]. Microplastics are a concern for environmental scientists due to their long-term durability and their ability to easily travel between different habitats (Fig. 1) [1].

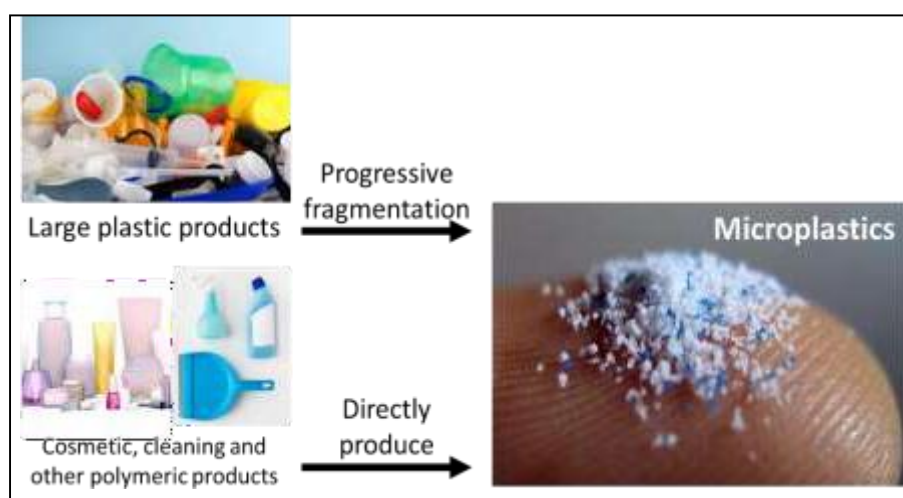


Figure 1 Common Microplastics sources (modified from [1]).

The most common raw polymers are polyethylene terephthalate (PET), polyurethane (PU), polystyrene (PS), polyvinyl chloride (PVC), polypropylene (PP), polyesters, polyethylene (PE), and polyamides (PA, nylon). Microplastics are ubiquitous because of poor plastic waste management [12,13]. Chronic exposure to microplastics is found to be toxic, but there is no evidence that they cause acute fatality [12], [13]. Chemical structure, additives used during polymerization, and how they are linked during polymerization control toxicity of microplastics [13], [14]. Microplastics, such as polystyrene, are capable of crossing into the bloodstream and disrupting the reproductive process of marine filter feeders [1], [8]. We discussed microplastic removal during this review. The sources of microplastic additives are discussed along with their occurrence, followed by a review of removal methods. Physical methods of removal include sorption and filtration, as well as chemical processes based on chemical phenomena. The potential toxicity of microplastics arises from unreacted monomers, oligomers, and chemical additives leaked from the plastic in the long run [1]. Types of microplastic toxicity are depicted in table 1.

Table 1 Kinds of microplastic toxicity.

Toxicity Type	Proposed effects	Ref
Structure-based toxicity	Ability to migrate from food packaging materials.	[15]
	Potentially absorbed residuals (i.e., Polystyrene bisphenol resins) by living tissues.	[16]
	Chemical additives are used during polymer manufacturing (i.e., phthalates from baby bottles) that enhance anomalous embryonic development.	[17]
	Chemicals released from plastics, like benzene, toluene, ethylbenzene, styrene, etc., may also cause chronic health effects.	[18]
Physicochemical toxicity	The large surface area/volume ratio of microplastics causes them to cause damage, that effect aquatic animals and then carries them to other habitats.	[19]
	Significant liver and brain tissue changes exposed to lowdensity polyethylene glycol microplastics containing phenanthrene.	[20]
	An adverse effect of microplastics on algae photosynthesis	[21]
	Ingested microplastics can also be toxic and absorbent depending on the shape and texture.	[22]
Microorganism toxicity	Pathogenic bacteria on some polyethylene, polypropylene, etc. may cause health impacts due to the micro-bacterial assemblages found in microplastics.	[23]
	Low concentrations of airborne microplastics in the air can cause cardiovascular diseases, respiratory diseases, and interstitial lung diseases.	[24]

2. MATERIALS, POLYMERS, AND ADDITIVES ASSOCIATED WITH MICROPLASTICS

2.1. Chemical additives for microplastics

Polyethylene, polypropylene, and polystyrene are the most common polymer components of primary microplastics, depending on the type of products being manufactured; while polyester, acrylic, and polyamide are the most common polymer components of secondary microplastics, forming fibers in the environment [25]. Polyethylene terephthalate and polypropylene were major types of microplastic, e.g., in Wuhan's inland freshwaters. 1650.0 ± 639.1 and 8925 ± 1591 numbers/m³ were the major types here. The

strongest type of microplastic has also been found to be low-density polyethylene [18]. Chemical additives such as bisphenol A, polybrominated diphenyl ethers, and phthalates are commonly found in raw plastics to enhance plasticity [26]. In addition to causing endocrine disruption, these additives may also be toxic. Such plasticizers are present in wide ranges in the plastic debris of remote and urban beaches: Bisphenol A is found in up to 35 ng/g of plastic debris on remote beaches, polybrominated diphenyl ether reaches up to 9900 ng/g on urban beaches, and phthalates are found in up to 3940 ng/g of plastic debris on urban beaches [6]. Most microplastic polymers have been detected with these plastic additives [25]. Additionally, researchers reported that silicone and polycarbonate microplastics could leach bisphenol A and nonylphenol [27]. There has also been a report of such chemicals accumulating in the human body through biological processes [28]. Microplastic exposure via food is one of the most alarming routes for humans [29], where the adverse effects of the chemical additives and mechanism of entry into the body are still under investigation. Thus, finding strategies for reducing the presence of microplastics in the environment must be a key objective. There have been reports on identifying the sources and occurrence of microplastics, their fate, methods for detection, and their environmental effects; however, to date, very few research and review papers have discussed how microplastics can be removed from a contaminated environment.

2.2. Sources and incidences of microplastics

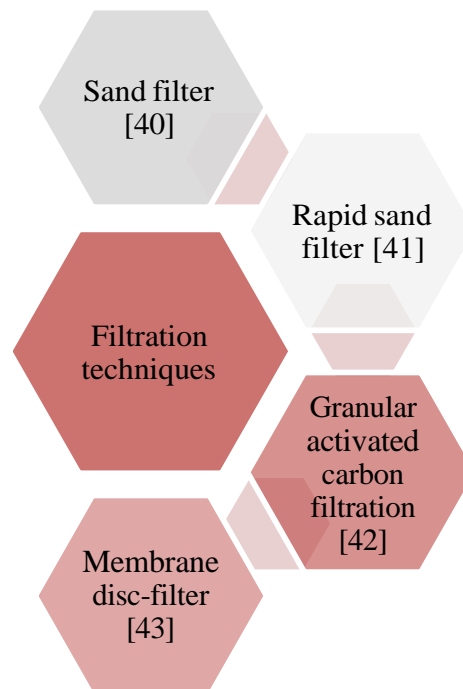
The transport phenomena involved in transporting microplastics such as wind and ocean currents contribute to their widespread presence in coastal regions and aquatic ecosystems worldwide [30]. Plastic pellets or powders used for air blasting are among the primary sources of polymers in household sewage discharge, including polymers from cosmetics and cleaning products [25]. In a secondary source of microplastics, the progressive fragmentation of larger plastic items in the environment (e.g., via mechanical degradation and UV exposure), contributes to the entry of substantial amounts into the environment through mechanical degradation and UV exposure [31]. By increasing plastic debris availability for ingestion by a wide range of organisms, we highlight the possibility of environmental hazards increasing [32]. It is also common for wastewater treatment facilities to release microplastics [33], [34]. Microplastics often bypass wastewater treatment and enter and accumulate in aquatic environments, even when larger plastic particles are effectively removed during wastewater treatment [35]. The treatment of wastewater frequently bypasses the removal of microplastic particles, which accumulate in aquatic environments despite the convenience of being removed from larger plastic particles [35]. There are many water treatment plants located near oceans and seawater, causing microplastics to be released into the environment. According to data from mainland China, out of 3340 wastewater plants, almost 1873 (58%) have treatment capacities of $78 \times 10^6 \text{ m}^3/\text{day}$ and are located along coasts where effluent can be discharged directly or indirectly into aquatic ecosystems [36]. To address this problem, many researchers are researching how microplastics are eliminated from water treatment plants by investigating their fate, occurrence, detection, and removal of these particles [37], [38].

3. REMOVAL OF MICROPLASTICS USING PHYSICAL METHODS

This study reviewed various physical techniques that are efficiently applied for the removal of microplastics (MPs) from treatment water.

3.1. Advanced filtration technology for microplastic removal

Several filtration techniques are utilized for MPs removing classified under physical methods as shown in Scheme. 1.



Scheme 1 Filtration techniques for microplastic removal.

Recently, Lares et al. (2018) applied a combination of membrane bioreactors/conventional activated sludge to investigate the performance of a municipal wastewater treatment plant. They sampled every 2 weeks for 3 months [44]. Finland's Mikkeli city center is home to a municipal water treatment plant that was used to collect samples of wastewater. As long as the conventional activated sludge system contains an aeration tank, where the wastewater is mixed with air to activate micro-organisms, and a sedimentation tank, where the treated wastewater is separated from the sludge, for subsequent biological treatment and secondary purification, the efficiency should also be improved [44]. It is also possible to use a conventional activated sludge system is also expected to be improved by the addition of aeration tanks for mixing water with air and sedimentation tanks for separating the sludge from the treated wastewater for biological degradation and secondary purification [45] [46]. Membrane bioreactors were significantly better at removing microplastics (99.4%) than conventional activated sludge treatment systems (98.3%). It was estimated that in the water effluent of the former system the microplastic concentration was 0.4 ± 0.1 MP/L, which was lower than that found in the water effluent of the latter system (1.0 ± 0.4 MP/L). Moreover, the study authors pointed out that using slightly different processing steps and wastewater samples in their study may have contributed to the narrow range of final microplastic concentrations [35], [47].

By observing the dimensional changes, abundance, shape, and color occurring during the removal steps, researchers in Changzhou, China, evaluated microplastic removal efficiency at their wastewater treatment plants [48]. Almost all plants using a combination of floating and sedimentation tanks, as well as filtration processes eliminated over 90% of microplastics from the influents. The final removal of microplastics reached 97.15 percent. Depending on the volume of processing daily, the type of raw water, and

the type of treatment process, the removal efficiency may vary considerably. This was my previous report, which reported less abundance of large microplastics in the effluents [49]. Furthermore, these microplastics were mainly composed of fiber rayon and polyethylene terephthalate, as evidenced by the high removal rates [49]. A study published by Yang et al. 2019 presented the results of a study by researchers in Beijing, China in which microplastics were removed from municipal sewage treatment plants [50]. Anoxic, aerobic, and anaerobic A₂O treatments were used during the initial treatment process of the influents. This included an aerated grit chamber, primary and secondary sedimentation tanks, and an aerated grit chamber. Denitrification, ultrafiltration, ozonation, and ultrasound are techniques used to remove microplastics from wastewater and complete the treatment process [50].

Polyethylene terephthalate and polyester rank first in abundance in the effluent, with 42.26 and 19.1%, respectively, according to FTIR analysis. Microplastics were removed from influents with an efficiency of 58.84% following the primary treatment using aerated grit, and thus 71.67% following the advanced treatment procedures. It was comparable to the efficacy of dissolved air flotation and sand filters despite the current sewage treatment plant's 90.166 percent removal efficiency being significantly lower than the 99.9% average of membrane bioreactors. [50], [51]. The current treatment systems are not effective enough to remove microplastics from sewage treatment plants. Although these processes do not eliminate all microplastics from wastewater, they do eliminate a good percentage of them.

3.2. Membrane-based technology for removal

During the study of Li et al. (2018), dynamic membranes were used to effectively remove microplastics from synthetic wastewaters, Fig. 2 shows the decrease in turbidity (39) when microplastics are removed from synthetic wastewaters. The study applied dynamic membranes for the effective removal of microplastics. Influence of flux and particle concentration during filtration of synthetic wastewater on the removal efficiency of dynamic membranes formed on a diatomite platform with 90 µm of supporting mesh.

When artificial wastewater was filtered with a diatomite platform with a 90 µm mesh of supporting mesh, the effect of influent flux and particle concentration was determined. Microplastics were filtered to near-zero turbidity in 20 minutes by reducing the influent turbidity from 195 NTU to less than 1 for the effluent [49], [52]. Input fluxes and microplastic concentrations are both factors that facilitate membrane formation. Based on elongated polymer coatings and mesh screens, researchers have developed an efficient microplastic removal tool. According to him, the tool has good durability, can be easily fabricated from common materials, and is durable. Additionally, there are no mechanical or electrical devices with these tools [1], [53].

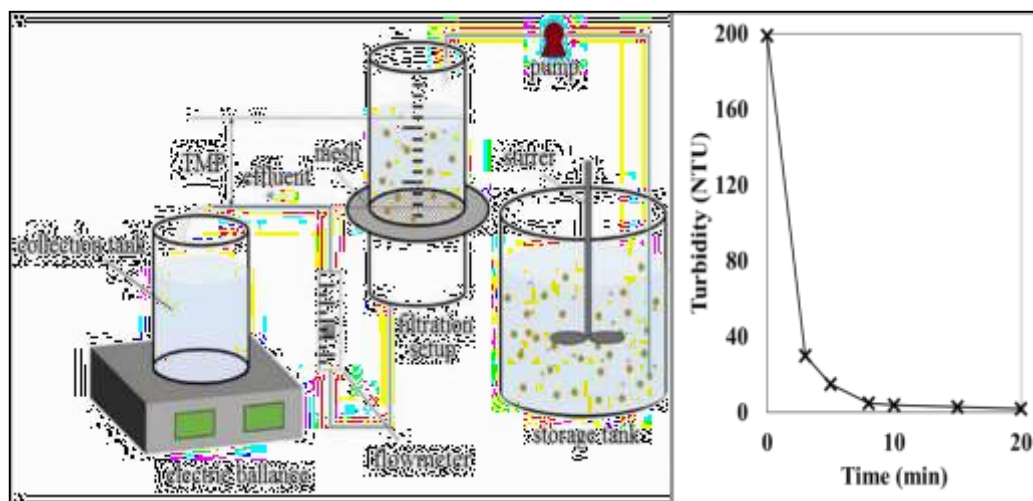


Figure 2 Graph and set up for the dynamic membrane experiment (modified from [54]).

There is, however, a greater capacity for the removal of micro-sized plastics within membrane bioreactors than in simple dynamic membranes [55]. Kno- block et al. (1994) explored the possibility of purifying a combined system by taking advantage of porous membranes along with biological processes [56]. The successful use of membrane bioreactors to remove high-level contaminants such as polymeric debris and microplastics confirms the suitability of this technology to handle complex industrial wastewater [46]. Talvitie et al. [46] conducted a study to examine how microplastics were removed from wastewater treatment plant effluents by using advanced endstage technologies, including membrane bioreactors, disk filters, rapid sand filters, and dissolved air flotation as shown in (Fig. 3).

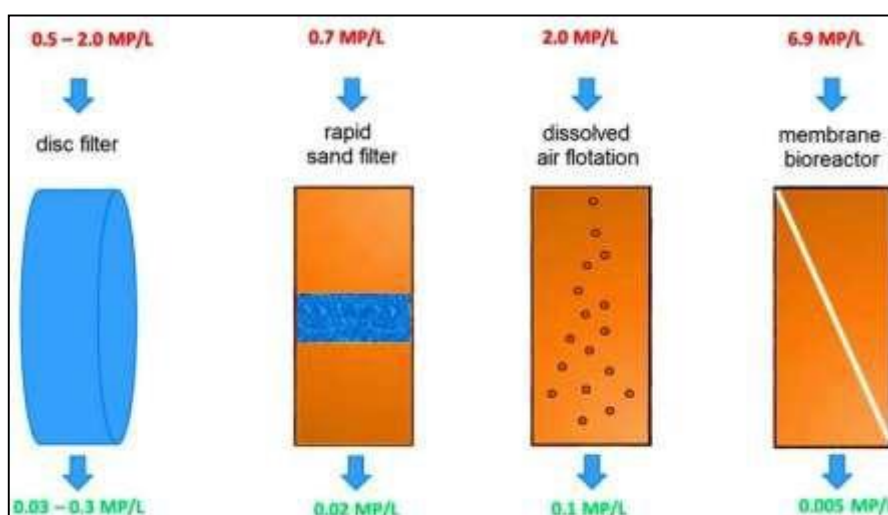


Figure 3 Amount of microplastics removed with final-stage technologies, measured in microplastics per liter, MP/L (modified from [51]).

Based on their analyses, the membrane bioreactor eliminated 99.9% of microplastic particles from 6.9 to 0.005 per L of water (Table 2). Additionally, microplastics of any size, even those of 20 to 100 microns, were removed by membrane bioreactors, rapid sand filtration, and dissolved air flotation [51].

Table 2 Average concentrations of microplastics before and after treatment with various technologies [51].

Treatment	Effluent type	Before (MP/L)	After (MP/L)	Removal (%)
Disk filter 10 ^a	Secondary	0.5	0.3	40.0
Disk filter 20 ^a	Secondary	2.0	0.03	98.5
Rapid sand filter	Secondary	0.7	0.02	97.1
Dissolved air flotation	Secondary	2.0	0.1	95.0

Each microplastic concentration is measured in microliters of effluent. The pore size is μm . Furthermore, A significant amount of microplastic was effectively removed from influents and effluents during treatment, irrespective of the shape of the microplastic. An analysis of the samples using Fourier transform infrared spectra (FTIR) revealed a marked decrease in polymers in the final effluent by the membrane bioreactor, highlighting the enzyme's ability to bind various chemical structures of microplastics [1], [9], [49], [51]. Technologies based on membranes have been effective in removing microplastics from polluted aquatic ecosystems. Microplastics are removed more efficiently over durable membranes, have a large influent flux, and are both large and concentrated. Biological processes combined with porous membranes could enhance removal efficiency by 99.9%.

Algae adsorption

Table 3 REACH Annex XIII provides a list of persistent contaminants in different media [61].

Compartment	Half-life (days)
Marine water	> 60
Fresh or estuarine water	> 40
Marine sediment	> 180
Fresh or estuarine sediment	> 120
Soil	> 120

Because of the potential for entanglement and bioaccumulation of microplastics in aquatic environments, they are more critical than other pollutants [57], [58]. Microplastics can cause multiple harmful effects as well as the death of aquatic organisms, e.g. reptiles, fishes, mammals, and birds. Since they are persistent and low-degradable, removal methods are required. Since they are persistent and poorly degradable, removal methods are necessary. Most microplastics are classified as persistent materials, but their nature and chemical structure determine how quickly they degrade. If the half-life times are lower than those determined by REACH criteria for consistency (Table 2), then these microplastics are degradable and are not hazardous to the environment [59]. It's well known that microplastics adsorb and carry a wide range of contaminants from water on their surfaces, carrying them into nearby habitats and desorbing them [60]. Because of their high surface area to volume ratio, other contaminants are likely to adhere to them. According to Sundbaek et al. (2018), fluorescent microplastic particles adhered well to the surface of seaweed, *Fucus vesiculosus*, an edible marine microalga. Microchannels within the plant cells of the sorbent limit the translocation of polystyrene microplastics into tissues due to the polystyrene microplastics' 20 nm diameter. The results showed very high sorption (94.5%) of microplastics to seaweeds around the cut surfaces, which is attributed to the release of alginate compounds from the cutting processes [62]. Alginate acts as a gelatinous substance that can be used to improve the adhesion of polystyrene to the surface of seaweed due to its anionic character [63]. Microplastics and microalgae's surface characteristics are influenced by surface charge in this paper and other studies about microalgae's ability to adsorb plastic particles [64], [65]. Researchers examined the

adsorption of polystyrene particles of 20 to 500 nm on bicellular algae, *Pseudokirchneriella subcapitata*, Nolte et al. (2017) considered positively charged rather than negatively charged microplastics for the most efficient adhesion [65]. Microplastics' ability to bind to algae surfaces is strongly influenced by their surface charge. An anionic polysaccharide in the chemical structure of algal cells explains why positively charged microplastics tend to be absorbed more efficiently [66].

4. MICROPLASTICS: CHEMICAL TREATMENTS

Flotation and agglomeration processes are commonly used in wastewater treatment plants to produce larger constituent particles that are easier to separate [67]. Through the use of Fe- and Al-based salts along with other coagulants, these processes bind tiny particles by inducing uptake-complexation mechanisms that are initiated by exchanges of ligands, thus forming strong bonds between waste particles [68]. Using iron and aluminum salt coagulants and ultrafiltration, Aza-Tarazona et al. (2019) determined the effects of anionic polyacrylamide (PAM), pH, and the formation of Al-based flocs on the removal efficiency of microplastics. The results are shown in Figure 4. The experiments were conducted with Al^{3+} and Fe^{3+} ions at different concentrations, and the results indicated that Al^{3+} performs better than Fe^{3+} . Furthermore, the removal efficiency of microplastics was not significantly affected by pH in the presence of low concentrations of Al coagulant source, 0.5 mM, although removal efficiency decreased when pH was raised with small microplastics of diameter less than 0.5 mm. A high Al dosage of 5 mM did not improve the removal efficiency of small microplastics as well as it did for large particles when using polyacrylamide (PAM), an enhancing coagulant. In the presence of cationic polyacrylamide, small microplastics grow at an accelerated rate. The removal efficiency of smaller microplastics ($d < 0.5\text{mm}$) was significantly enhanced when anionic polyacrylamide was used, from 25.83% without polyacrylamide to 61.19% with 15 mg/L polyacrylamides; however, the growth rate increased by just 4.27% to 18.34% for large microplastics (2–5mm diameter) [69] [70].

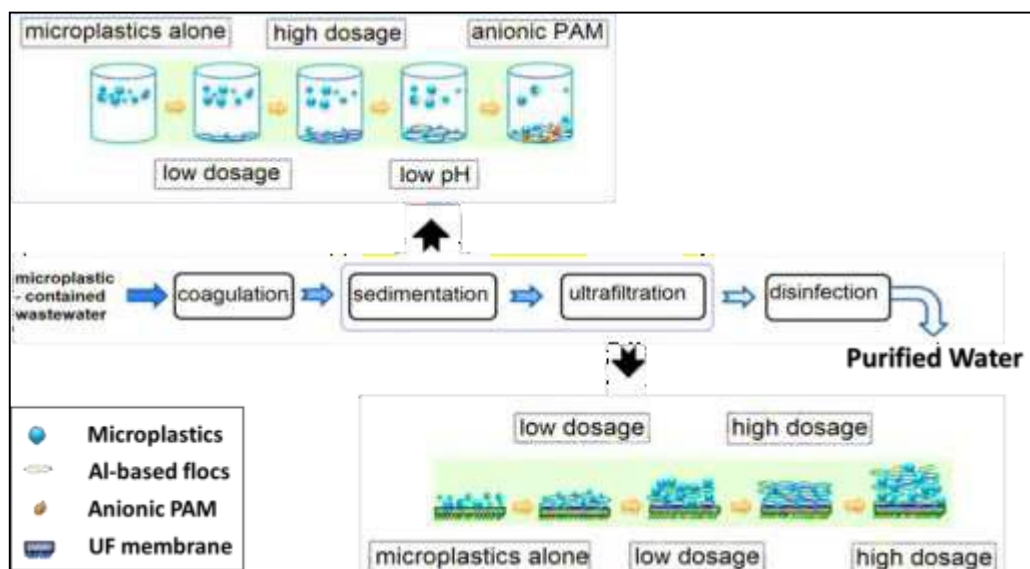


Figure 4 A coagulation, sedimentation, and ultrafiltration process for removing microplastics (modified from [48]).

In addition, Ma et al. (2019) used the same method to remove microplastics but applied a $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ coagulation agent instead (Fig. 5). Their experiments demonstrated that, at neutral pH, the removal of microplastics was enhanced with increasing concentrations of coagulants. This trend was especially clear for microplastics of less than 0.5 mm in diameter [48].

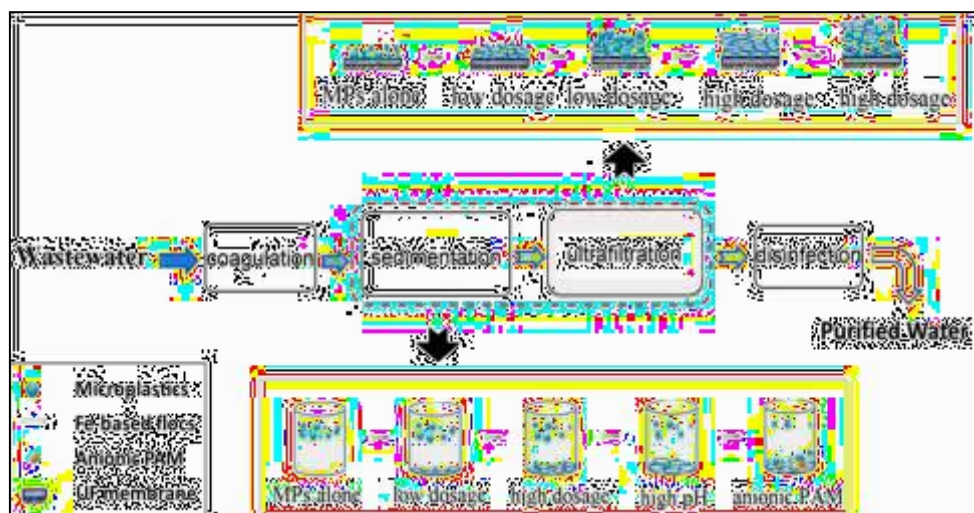


Figure 5 A coagulation, sedimentation, and ultrafiltration procedure for eliminating polyethylene microplastics from wastewater (modified from [48]).

In addition, the removal efficiency was further intensified with high pH and 2 molar mass (MM) coagulant concentrations, as well as for smaller microplastic particles. Under these conditions, anionic polyacrylamide performed far better than cationic polyacrylamide under a low dosage, 2 mM. As a result, the rates of removing microplastics from polyethylene were improved substantially. A mechanistic explanation can be put forward regarding the facile formation of Fe-based flocs during the coagulation process by using anionic polyacrylamides to make the products dense enough to be concentrated and trapped [48] [71]. Several techniques have been successfully used by researchers to remove polyethylene microplastics from a stirred-tank batch reactor (Fig. 6), including

electrocoagulation, charge neutralization, and floc formation then to clean water with sedimentation, which is an environmentally safe, energy-efficient, cost-effective, and highly automated method [72].

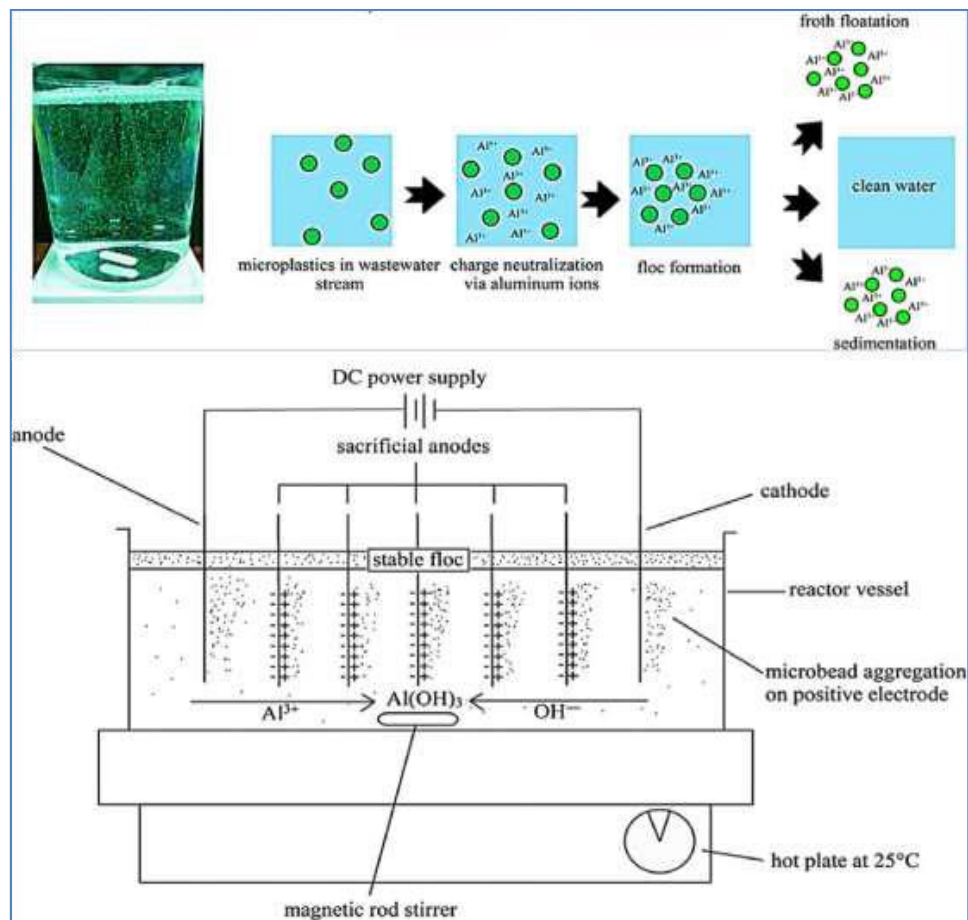


Figure 6 An electrocoagulation reactor setup, in which Al^{3+} acts as a coagulation agent, is used for microplastic removal (modified from [72]).

In coagulation, colloids are broken apart and the surface charges of microparticles are stabilized. Through van der Waals forces, the particles can interact sufficiently close to one another [73]. Concurrently, the microplastics in the wastewater sample are trapped by the coagulants, forming a sludge blanket. Based on the results of all experiments using electrocoagulation, the removal efficiency was higher than 90%. Using pH 7.5 and NaCl concentrations between 0 and 2 g/L, 99.24% of the contaminants were removed. A further study found that the case of the $11 A/m^2$ tested current density, the lowest tested current density in terms of energy use, resulted in the highest removal rate [73] [74]. Microplastics are not fully understood in terms of their degradation mechanisms. Brandon et al. (2016), studied the chemical changes in the structure of polypropylene, polyethylene, and other microplastics throughout 3 years of simulated realistic weather conditions [75]. According to FTIR analyses, some metabolites, such as carbonyl, hydroxyl, and carbon-oxygen bonds, exhibited slight nonlinear changes with time, indicating that microplastics take a long time to degrade [75]. The degradation of microplastics by elements [76], microorganisms [11], and catalysts [77] has been extensively studied, but the degradation of microplastics has been relatively neglected. Liu et al. (2019) investigated the long-term aging behaviors of polystyrene and polyethylene

microplastics in the aquatic environment via a heat-activated persulfate-Fenton combined method [78]. They concluded that the O/C ratio and microplastic size were important factors determining microplastic adsorption capabilities and surface properties, which influence microplastic oxidation rates significantly [78]. Recently, studies have been published assessing the structural and morphological alterations of polyethylene microplastics under dark and UV light [11], [79]. According to FTIR analysis, artificial seawater was significantly more degradative than UV illumination compared to the initial materials and products (Fig. 7).

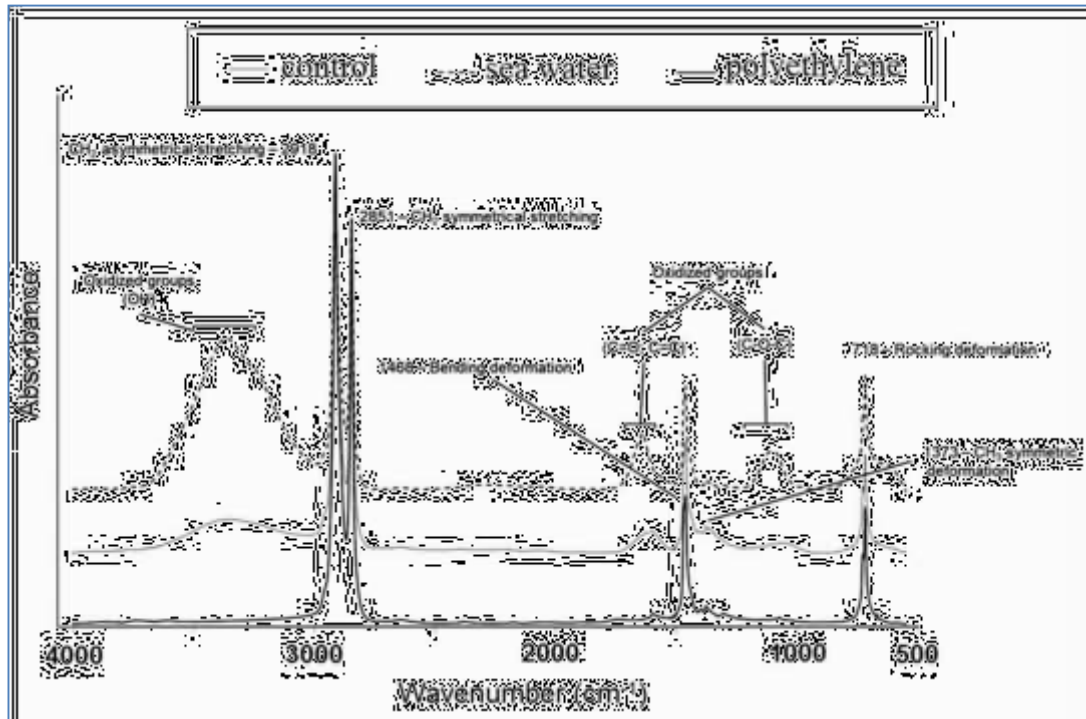


Figure 7 Polyethylene microplastics FTIR spectra before and after being treated with artificial seawater for 8 weeks (modified from [79]).

A greater organic content in the medium confirmed this. Moreover, when microplastics were exposed to UV light for the same period, no critical changes in their chemical structure were observed, showing that salt is necessary to form oxidized sites. Microplastic surface morphology was also affected by salt, revealing observable cracking lines in SEM images. Such findings confirm the important role of salinity in microplastic degradation [1], [53], [80], [82].

5. CONCLUSIONS

In this review, major physiochemical approaches to removing microplastics have been summarized. A variety of methods are available to remove them from an environment, including chemical and physical methods. Microplastics are greatly reduced in influent water entering the treatment plants by a combination of filtration and membrane bioreactors, but these systems act as sources of microplastics every day because effluents are directly released into aquatic environments. The conventional activated sludge treatment strategy is used in water treatment plants as well as membrane bioreactor technologies, but it shows less efficiency than the latter method, which results in it being a less popular treatment method. Microplastics can also be effectively separated by electrocoagulation and agglomeration, but these techniques must be combined with

significant additional filtration steps. To understand any structural alterations during degradation, FTIR and electron microscopy analysis are widely used. Based on the FTIR bands of the treated microplastics, it appears that the biological removal occurred via the oxidation of hydroperoxide and hydroxyl groups, carbonyl groups, and double bonds.

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