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Microplastics in Aquatic Environments: Recent Advances in Separation Techniques

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Abstract

Separation and removal of microplastic pollution from aquatic environments as a global environmental issue is classified as one of the major concerns in both water and wastewater treatment plants. Microplastics as polymeric particles less than 5 mm in at least one dimension are found with different shapes, chemical compositions, and sizes in soil, water, and sediments. Conventional treatment methods for organic separation have shown high removal efficiency for microplastics, while the separation of small microplastic particles, mainly less than 100 µm, in wastewater treatment plants is particularly challenging. This review aims to review the principle and application of different physical and chemical methods for the separation and removal of microplastic particles from aquatic environments, especially in water treatments process, with emphasis on some alternative and emerging separation methods. Advantages and disadvantages of conventional separation techniques such as clarification, sedimentation, floatation, activated sludge, sieving, filtration, and density separation are discussed. The advanced separation methods can be integrated with conventional techniques or utilize as a separate step for separating small microplastic particles. These advanced microplastic separation methods include membrane bioreactor, magnetic separation, micromachines, and degradation-based methods such as electrocatalysis, photocatalysis, biodegradation, and thermal degradation.

Keywords

microplastic separation, magnetic separation, membrane bioreactor, micromachines, wastewater treatment

1 Introduction to microplastic occurrence and hazards

Plastic-based materials are widely used in today's life and cause a growing threat due to releasing various forms of plastic waste such as nano-, micro-, and macro-plastics are releasing into the environment [1]. During the last decade, microplastic particles (MPs) have entered directly into marine and freshwater environments, affecting habitats and animals negatively. Firstly, in the early 1970s, microplastics were reported in North America as spherules in plankton tows in coastal waters of New England [2]. Subsequently, microplastics are penetrating oceans and water bodies, including rivers and lakes progressively. Accordance to the National Oceanic and Atmospheric Administration (NOAA), microplastic particles are defined as plastic particles smaller than 5 mm in length. Microplastics can be categorized into two major classifications as primary and secondary microplastics, depending on their source [3]. Primary microplastics consist of industrial products such as cosmetics as well as different kinds of textiles [4–6]. Secondary microplastics form by the fragmenting larger plastic items, caused by weathering (e.g., ultraviolet light) and during consumption or fabrication [6–8]. Annually more than 348 million tons of plastic waste releases into aquatic environments. Fragmented polymeric particles less than 5 mm has potential toxic risks in the ecosystem and human health [9]. Fragmentation of polymeric waste decreases the size of plastic particles to micro- and nano-scale, which may be due to the effect of tides and waves [10].

Recent research revealed that more than 100 billion microplastics can be released by a single wastewater treatment plant (WWTP) yearly; hence WWTPs are substantial contributors to the issue of microplastic pollution of surface waters [11]. Additionally, microplastic particles in the effluent of the wastewater treatment plant penetrate the

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water bodies and pile up in the environment eventually, taking into account WWTPs may remove some of microplastics in light of used treatment units [12, 13].

Typically, microplastics refer to plastic particles with dimensions ranging from 100 nm to 5 mm. This range includes sub-micron plastic particles (100 nm–1 µm), small microplastics (1–100 µm), and large microplastics (100 µm–5 mm). Plastic particles smaller than 100 nm are classified as nanoplastics [14–17]. However, a threshold limit of 1000 nm is used in some studies related to environmental nanotechnology [17, 18]. As discussed in a comprehensive review by Yin et al. [19] on the toxicity of microplastics and nanoplastics, depending on the target organs, microplastics and nanoplastics show different toxicity. Microplastics with small sizes are more toxic than large ones because of the higher bioavailability and retention time in the body. Generally, nanoplastics with higher surface area seem to be more toxic than microplastics. Micro- and nano-plastic particles can accumulate in various tissues [20]. Depending on the organ type, the accumulation of plastic particles with nano- and micron-size are different [21]. Plastic particles with different components show different toxicity which arises from differences in their physicochemical properties [19].

The abundance of some polymer types as a percentage in wastewater treatment identified by Raman Spectrometer is shown in Fig. 1 [22]. Polyethylene and polystyrene as hydrophobic polymers with densities like water are some of the most abundant microplastics in drinking and freshwater systems [23, 24]. Microplastics are present in aquatic environments, sediments, and water treatment plant effluents [25].

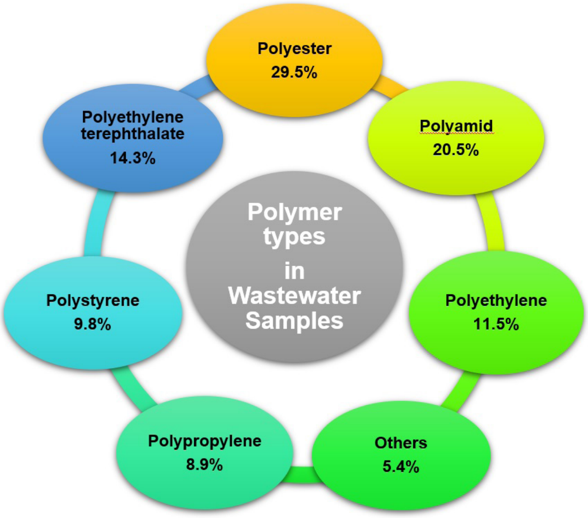

Fig. 1 Different types of polymers in wastewater samples detected by Raman Spectrometer [22].

Adsorption of hazardous substances such as metals or organic compounds on the surface of microplastic increases the chemical toxicity of hazardous. Long-term weathering of microplastics in aquatic systems provide sorption sites for metals or organic compounds [9, 26, 27]. Aquatic ecosystem is widely affected by plastic wastes as one of the most emerging contaminants with small size, low-density, and bioavailability to organisms. The hydrophobic nature and high surface area of microplastics facilitate the adsorption of organic pollutants and metals in aquatic systems [28]. Fossi et al. [29] studied the detection of microplastics as plastic debris on large filter feeders such as baleen whales and sharks. The results revealed that the concentration of Phthalate and organochlorines could be considered as a tracer for microplastic. Besseling et al. [30] reported microplastics of various types and sized in the baleen whale. FTIR analysis revealed the existence of various polymers such as polyethylene, polypropylene, polyvinylchloride, polyethylene terephthalate, and nylon of various shapes such as sheets and fragments with sizes larger than 1 mm. Gonçalves et al. [31] assessed the ingestion and excretion of microplastics by exposing the Mediterranean mussel to polystyrene microplastics of 2 and 10 mm µm. The histopathological results revealed the potential ability of the Mediterranean mussel to digest and exert microplastics. Several studies are conducted to detect microplastic particles in food such as honey, sugar, beer, table salt, and drinking water [1]. Oral digestion of microplastics in food has topological harmful and small microplastic particles less than 1.5 µm may penetrate organs; therefore, it is essential to develop high precise methods for detecting small microplastic particles in food [32].

Additionally, microplastics due to their physicochemical properties can interact with metallic, inorganic, and organic matters and pollutants, and nutrients and create a suitable condition for microorganisms' attachment and colonization [33–35]. The attached microorganisms via their surrounded extracellular biopolymers or so-called biofilms have been studied extensively [36–39]. In addition to some effects of biofilm formation such as protecting the microplastics from abiotic and environmental stress and destruction, growth and enrichment of pathogenic bacteria and fungi and possible genetic materials' exchange between bacteria are the main concerns [40–44]. Microplastics can act as vectors for transferring the microorganisms and contaminating the environments [45, 46]. It is worth noting that many researches are focused on another aspect of the interaction between microplastics and microorganisms. Due to the potential of some bacteria and fungi in the enzymatic digestion of plastics, biological degradation of plastic materials attracts much attention in the last decades [47–50]. Since the focus of the review is microplastics' separation techniques, we refer the readers to good reviews published in recent years for more information on microplastics and microorganisms interactions [44, 51, 52].

This review provides a critical discussion on various techniques for microplastic particles separation from aquatic environments. In addition to the separation methods conventionally utilized in the wastewater treatment process, more recent advanced separation techniques such as membrane bioreactors, magnetic-based separation, micromachines, and degradation-based separation are presented and reviewed. The challenges and limitations of conventional techniques as well as the advantages of advanced techniques to separate small micron-size plastic particles from water have also been presented and discussed.

2 Conventional methods of microplastic separation

Studies related to the occurrence and removal of microplastics have attracted the attention of researchers, mainly about the removal of microplastics by applying different treatment techniques. The potential hazards of microplastics in our everyday life and the development of efficient methods for the characterization and quantification of polymeric particles of micron size in aquatic environments and sediments have been investigated in many researches [53].

Among various treatment methods in water treatment plants [54, 55], clarification, sedimentation, density separation, coagulation and, or flocculation, activated sludge, sieving and filtration are considered conventional treatment processes in water treatment plants, and several studies have been focused on the removal efficiency of these treatment process for microplastic separation [56]. The basis of various physical, chemical, and biological methods conventionally applied in water treatment plants has been reviewed by Tirkey and Upadhyay and Zhang et al. [57, 58].

The sedimentation process is limited for the separation of low-density particles. Unlike the cake filtration method, dept filtration is a suitable technique for microplastic separation from large volume and dilute aquatic samples. In contrast, dept filtration method suffers from a large pressure drop in this process [53].

A challenge encountered when trying to compare results from microplastic surveys is the lack of comparable protocols, for the identification of microplastic in the samples. In sampling protocols microplastics are usually, classify based on source, type, shape, color, and degradation stage. Identification is primarily made by visual identification, often with the aid of stereomicroscope. Researchers for sampling and analyzing plastics from natural particles use different protocols. There are some protocols for analysis of microplastic in aquatic samples based on visual analyzing and FTIR spectroscopy. The most common analysis methods are FTIR and Raman spectroscopies [59, 60].

Here we provide a brief explanation of the most common techniques utilized in water and wastewater treatment plants to remove small polymeric particles of micron size.

Primary clarification aims to provide solid settling before the biological treatment. Primary clarifiers are also supported by surface skimmers to skim floating solids off the surface before the secondary treatment. Michielssen et al. [61] observed that 84–88% of microparticles with sizes ranging from 100–1000 μm, were eliminated through primary screening and primary clarification; Conley et al. [62] reported the loading of microplastics and their removal efficiency in three wastewater treatment plants with various treatment operations and service arrangements in USA for one year. The major wastewater treatment plant was using a primary clarification and demonstrated the highest microplastic removal efficiency of about 97.6 % that clarifies the impact of primary clarifiers on microplastic removal performance. The size fractions included microplastic particles larger than 418 μm, between 178–418 μm, and between 60–178 μm [62].

The sedimentation technique, which is based on gravitational settling, can remove suspended contaminants such as microplastic particles from aquatic systems. This method is used not only in primary treatment but also in secondary treatment. The removal efficiency of microplastics by sedimentation is affected by two crucial factors, including density and shape [63, 64]. This process can be used before other treatment techniques [63, 64] with removal efficiencies of 57%–64% in wastewater of South Korea [65], which microbeads and fragments were reported as the major kinds of microplastics in all wastewater samples and 91.7% [66, 67]. The major drawback of the sedimentation technique is the essence of utilizing some other appropriate techniques in the following to complete removal.

Flotation is based on four steps. The steps include bubble generation in the wastewater, contact between the gas bubbles and suspended particles/oil droplets, attachment of the particle/oil droplets to the bubble surface, and finally rising the air-solid mixture for skimming off the floating materials [68]. There are several types of floatation, depending on the bubble generation method, such as dissolved air floatation (DAF), Induced air floatation (IAF), Froth floatation, electrolytic floatation, vacuum floatation [69]. Flotation is one of the most widely used methods for separating low-density plastic particles from soil or sediment in dense liquids [70]. Dissolved air floatation allows to remove of low-density particles and algae effectively; however, this method is expensive to operate and maintain compared with the sedimentation process [71]. Coppock et al. [15] proposed a portable density floatation to separate microplastics with particle sizes ranging from 100 µm-10 mm from sediments with an average efficiency of 95.8%.

Conventional activated sludge process (CASP) is a common wastewater treatment process, relying on biodegradation using activated sludge. Microplastic particles could attach to the suspended matter and separate in the subsequent settling step [67, 72]. Magni et al. [73] conducted a grid chamber and conventional activated sludge process at a municipal WWT system in Italy for microplastic separation with a 64% removal rate. In this study, the size classes included 1–5 mm, 0.5–1 mm, 0.1–0.5 mm, and 0.01–0.1mm. The main drawbacks of CASP are producing an excessive sludge, extensive retention times, extensive sedimentation surface, and massive cost of energy and dumping. However, this process is flexible, appropriate for wide-scale treatments [63, 74–76]. The retention time and nutrient extent in wastewater are considered as the most important affecting factors on the efficiency of the activated sludge method for microplastic removal [12, 77].

To investigate the impacts of microplastics as emerging pollutants, it is required to collect different types of microplastics from aquatic environments for identification through sampling and extraction techniques.

Filters with different pore structure, pore size, and materials are used for extracting the microplastics from aquatic samples. Metal-based filters such as stainless steel and polymer-based filters such as polycarbonate, nitrocellulose, and nylon are utilized for the separation of microplastics from retained particles [78, 79]. Some filter materials have curvy and deep pore structures such as stainless steel and nylon filters. Some others exhibited narrow and straight circular pores such as polycarbonate filters. The particles employed in the mesh filtration technique were in the range of 50-1000 µm [80]. After sampling, the retained microplastics on the filter are analyzed quantitatively and qualitatively. The analysis of the abundance and size distribution of retained microplastics are termed quantitative analysis. The qualitative analysis includes evaluating the color, shape, and composition of the retained microplastics [81]. In some studies, manta trawls and neuston nets are utilized as a sampling system from large volume aquatic environments such as oceans and water column [82]. It may be possible secondary contamination of water by filter fibers in filtration method. Therefore, it may be checked that the secondary contamination is reasonable in comparison with the removal of the primary microplastics.

The sieving method of water samples is also used to separate microplastics plentifully, resulting in sorting particles into different size ranges depending on the choice of sieve mesh size categories [83, 84]. The sieve physically traps the microplastic particles, enabling water to get lost from the sample [85]. Olivatto et al. [86] studied separate microplastics found in samples of the Guanabara Bay in Brazil via the sieving and manual sorting. Microplastic particles less than 5 mm were isolated in the laboratory by wet sieving using two meshes including 355 µm in the bottom and 4.75 mm in the top. The most common sieving system for the separation of microplastics from water and sediment samples is multi-step sieving, which is using a series of sieves with different mesh size [87]. A cost-effective separation and quantifying method with less environmental footprint was presented by Gimiliani et al. [88] comprising sieving of 2.0, 1.0, 0.5, and 0.25 mm mesh sizes, sediment collection, drying, and stereomicroscopic evaluation of the samples maintained on each sieve [88].

Density separation of microplastics is based on their different densities and is usually conducted by adding brine solutions to allow separating lower density particles from denser matrices after settlement [6, 15, 89, 90]. Konechnaya et al. [91] reported that $ZnCl_2$-based density separation is an appropriate method for separating polymeric particles from a sandy sample for isolating the particles with sizes including 1–5 mm, 400–1000 µm, 200–400 µm, and 100–200 µm. Applying a centrifugation step after density separation in saline solutions can enhance the plastic-sediment separation ability and improve the extraction capacity of microplastic fibers and granules from sediments [90, 92].

3 Recent progress in conventional separation methods

Removal of small microplastic particles less than 100 µm is challenging since particles larger than 100 µm can be sufficiently separated in today's water treatment plants [56, 93]. Wang et al. [56] studied the presence of various microplastics of 1–100 µm in size such as polyethylene terephthalate, polyethylene, polypropylene, polyacrylamide with fiber, sphere, or fragment shapes in the effluent of different treatment processes of an advanced drinking water treatment plant (ADWTP) . Xia et al. [94] utilized the Fluorescence imaging method to evaluate the effect of tween 20 surfactants in ppm level on the coagulation of polystyrene microplastics of 1 µm. A flexible and hydrophilic layer formed on the microplastic particles by surfactant molecules hinders the deposition of bentonite. It inhibits agglomeration resulting in a decrement in the removal efficiency with increasing surfactant concentration, as shown in Fig. 2 [94]. In contrast, anionic surfactants such as sodium dodecyl sulfate will not hinder the coagulation of microplastic particles since negative charges induced by surfactant adsorption are neutralized in the presence of Al^{3+} ions resulting in the precipitation of microplastic particles. As shown in Fig. 2, the coagulation removal efficiency was not affected by increasing sodium dodecyl sulfate surfactant [94]. The coagulation integrated with sedimentation is an appropriate choice for contaminant removal [95]. Pivokonský et al. [96] reported microplastic removal of 88% using a multi-step process such as coagulation-flocculation with sedimentation. Coagulation-flocculation with sedimentation was quite effective for the elimination of microplastics, and additional MP removal was obtained by filtration and granular activated carbon processes. Ma et al. [97] examined microplastic removal in coagulation/sedimentation and ultrafiltration in controlled tests using Al- and Fe-based salts, observing a removal efficiency lower than 40%.

Filtration system integrated with various separation techniques such as clarification, floatation, or reverse osmosis has been investigated in some studies [70, 98, 99].

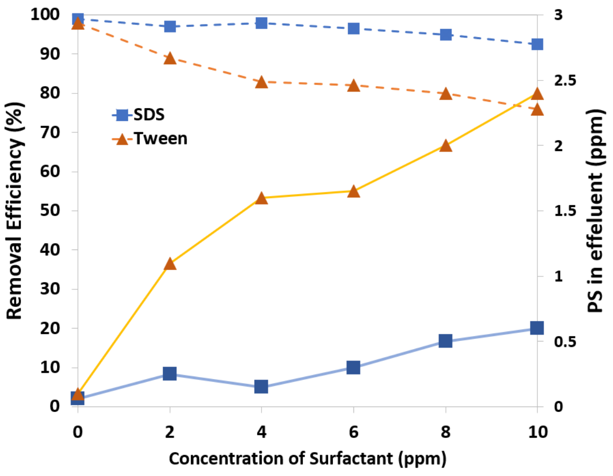

Fig. 2 The effect of surfactants on (left) the removal efficiency of polystyrene microplastics and (right) the residue concentration of microplastic particles in the effluent [94].

Kim and An [100] developed a vacuum-based method for separating microplastic LDPE films, for including two differently sized cylindrical sieves to accumulate film samples. Wang et al. [99] studied the occurrence of phthalate esters and microplastics at the effluent of four wastewater treatment plants, receiving water bodies in winter and spring. The main techniques were clarification, filtration, and reverse osmosis with removal rates of 42.7%–69.2%, 25.3%–59.3%, and 22.6%–51.0%, respectively. The total removal rates of phthalate esters and microplastics in the four RWTPs were 47.7%–81.6% and 63.5%–95.4%, respectively. The results revealed that the surrounding environment considerably affected the amount of phthalate esters and microplastics in surface waters.

Classification of microplastics before analysis seems to be required and useful since sedimentation velocity depends on particle density and size. Polymeric particles have various densities; some polymers are denser than water, and some others have densities close to or less than water. For small-size microplastics with a low sedimentation velocity of 1 cm/h, a filtration system should be designed based on the particle size rather than particle density [101]. Bannick et al. [101] developed a filtration system for analyzing microplastic samples in water using a thermal extraction-desorption gas chromatography-mass spectrometry (TED-GC-MS). The filtration system was validated for artificial water samples and was applied in the effluent of a WWTP in Berlin. Artificial water samples composed of spherical polyethylene and polystyrene particles with various sizes ranging from 22–150 µm and 298 µm, respectively. The filtration system included several stainless-steel filters to classify microplastics into different size classes of 1000–500 µm, 500-100 µm, 100-50 µm, and 50–10 µm. The results revealed that the sampling and separation method depends on the size class of microplastic in water. This filtration method showed a recovery percent of 80–110, depending on the type and size of particles. For analyzing quantic samples using TED-GC-MS, the larger particles required a larger volume of water required to be sampled. For particles smaller than 50 µm, significantly smaller than 10 µm, it is required to apply pressure filtering due to the small effect of density in sedimentation of small polyethylene and polystyrene particles [101].

Anna Markiewicz et al. [102] assessed the performance of a pilot plant for the removal of non-particulate organic pollutants from urban runoff in Sweden. The separation system included a sand-column as pre-filter, which is in series with a granulated activated carbon, Sphagnum peat, or Pinus sylvestris bark column. All filters exhibited an effective removal of total suspended particles larger than 1.2 µm.

Automatic recognition of different microplastics, including polyethylene, polypropylene, and polystyrene with a size of 100 µm, is reported by Zhu et al. (2020) using a near-infrared hyperspectral imaging (HSI) technique. Gold-coated polycarbonate and glass microfiber filters showed a suitable performance for the identification of microplastics using the HSI technique [103].

4 Advanced methods of microplastic separation

Increasing demand for removing parts of microplastics that pass through conventional water and wastewater treatment plants is comprehensible. As reported in the literature [104–106], several advanced methods and technologies for removing micropollutants have been evaluated on a large scale in several countries such as Germany, Sweden, and Switzerland. Here we represent and discuss some of the advanced methods, including magnetic-based techniques such as magnetic seed filtration and magnetic micro-submarines [107], photocatalytic micro-motors [108], membrane bioreactors coupled with activated carbon filters, rapid sand filtration, or CAS [109] and degradation-based techniques such as electrocatalysis [110], photocatalysis [111, 112], biodegradation [113], and thermal degradation [114, 115].

4.1 Membrane bioreactors

Different technologies have been studied to remove microplastics from municipal and industrial wastewaters in real or pilot scales. Membrane bioreactor (MBR) is a established process for removing microplastics from wastewaters in real WWTPs or pilot scales [67, 98, 109]. The removal efficiency of the MBR process in several studies conducted in the Netherlands, China, the United States, the United Kingdom, and Finland are in the range of 64.4 to 99.9% [12, 13, 67, 109, 116, 117]. Membrane bioreactor is a growing technology in conventional water and wastewater treatment plants for replacing the conventional activated sludge technology in some countries such as Sweden. Membrane bioreactor is a combination of biological activated sludge process and membrane separation, which results in significant advantages over conventional activated sludge process for removing micropollutants in both municipal and industrial wastewater treatment plants [106]. Baresel et al. [106] evaluated a membrane bioreactor coupled with granulated active carbon-based biofilter for the removal of various kinds of micropollutants, including microplastics and organic compounds from real wastewater of Stockholm's main WWTP Henriksda with a hydraulic retention time of 10 hours. An ultrafiltration system was applied after the biological reactor. The effluent of the membrane bioreactor, with qualities of lower than 0.2 mg TP/L and 6 mg TN/L, was pumped to a granulated active carbon-based biofilter with a total area of 0.3 m^2. A screening technique using a 20 µm filters was applied to separate the microplastics from water samples. A stereo microscope with 50 times magnification was applied for counting and dividing microplastics into three groups of fragments, flakes, and fibers. Baresel et al. [106] found 100 percent removal efficiency for microplastics in the MBR effluent. Rapid sand filtration is a tertiary treatment in WWTPs, and its removal efficiency is compared with other technologies such as ozonation, membrane disc filter, and membrane bioreactor in some studies [65, 109]. Bayo et al. [109] found 14 polymer types in wastewater samples using membrane bioreactor and rapid sand filtration technologies and polyethylene, including low-density polyethylene (LDPE) and high-density polyethylene (HDPE) with 75.76%, was the most common type in the samples. Different forms of microplastics including fibers, films, fragments, and beads with size ranging from 210 µm to 6.3 mm were isolated in this study. About 58.90% of microplastics had sizes smaller than 1 mm. Membrane bioreactor showed a removal percentage of about 79%, which was more than that of rapid sand filtration, i.e., 75.5%. Among various types of polymers, LDPE, nylon, and polyvinyl were remained in RSF effluent and melamine in MBR effluent [109].

4.2 Magnetic based separation

Microplastic recovery and extraction of small size lower than 150 µm are challenging. Magnetic separation and extraction of microplastics such as polyethylene, polyethylene terephthalate, polystyrene, polyurethane, polyvinyl chloride, and polypropylene from various environmental matrices such as seawater, freshwater, and sediment can be considered as a post density separation step or a standalone process to produce a drinking water [107]. Magnetic seed filtration (MSF) technique includes two general steps: hetero-agglomeration of microplastic particles with magnetic nanoseeds, and separation of magnetized agglomerates using magnetic force [53, 107]. Compared with classic filtration methods, MSF has a lower pressure drop. There is no limit on the minimum size of microplastics for separation using the MSF method since the size dependency in this method can be tuned by varying the size and type of magnetic nanoparticles. Magnetic removal of organic dyes and heavy metals from wastewater using magnetic nanoparticles have been reported in the literature [118]. Moreover, the magnetic seed filtration technique showed promising results in large-scale water treatment. Hydrophobic and electrostatic interactions are introduced as main driving forces in the magnetic separation of microplastics from aquatic environments so far [53, 107]. Grbic et al. [107] developed a magnetic method based on using hydrophobic coated iron nanoparticle to recover microplastics even small-sized microparticles from seawater. This method was based on the magnetization of microplastic surfaces using hexadecyl trimethoxy silane functionalized-iron nanoparticles, microplastic with a higher surface to volume ratio, i.e., smaller microplastics, can be extracted more efficiently, see Fig. 3(A) [107]. The results revealed a separation recovery of 92% for microplastic particles smaller than 20 μm, including polyethylene and polystyrene beads, see Fig. 3(B), (C) [107]. Also, a recovery of 93% obtained for microplastic particles larger than 1 mm, including polyethylene, polyethylene terephthalate, polystyrene, polyurethane, polyvinyl chloride, and polypropylene. This method also applied for the separation of microplastics of 200 μm to 1 mm from freshwater and sediments with recovery percent of 84% and 78%, respectively.

4.3 Micromachines

One of the most promising technologies for environmental remediation and removal of contaminants such as oil, organic compounds, heavy metals, and microplastics from aquatic systems are self-propelled micro/nano-scale devices such as magnetic micro-submarines [119] and photocatalytic micro-motors [108]. Photocatalytic micromotors provide fascinating features, including an on/off switch, using water as green fuel and light as a renewable energy source [119]. Sun et al. [119] fabricated hollow

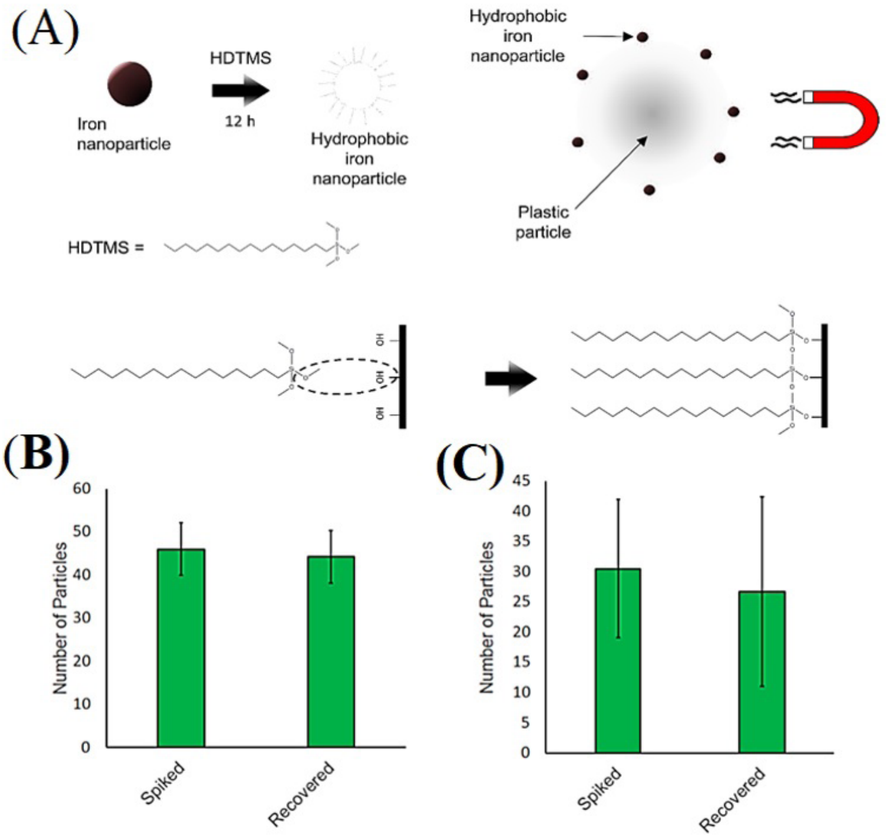

Fig. 3 (A) a schematic of synthesis procedure of hydrophobic iron nanoparticles using hexadecyltrimethoxysilane (HDTMS) functionalization and application for surface magnetization of microplastics for efficient extraction of small microplastic particles; (B) Number of small polyethylene spheres of less than 20 μm in 1μL of spiked sample counting using microscope compared with magnetic extraction recovery; (C) Number of small polystyrene spheres of 15 μm in 1μL of spiked sample counting using microscope compared with magnetic extraction recovery. Adapted with permission from [78]. Copyright (2019) American Chemical Society [107].

magnetic micro-submarines on a large-scale through sequential acidolysis and sputtering of natural sunflower pollen grains and applied for effective removal of oil and plastic particles from water. Hollow magnetic micro-submarines provided a recyclable, eco-friendly, and chemical-free method for microplastic removal through non-contact shoveling because of different fluid flow forces induced by the motion of micro-submarines in the aquatic environment. Wang et al. [108] developed photocatalytic micro-motors in the form of individual micro-motors and assembled a chain of catalytic particles based on Au@Ni@TiO_2 structures. The results proved the ability of light-driven micro-motors for catalytic elimination of microplastics from aquatic samples.

4.4 Degradation based separation

Efficient degradation of microplastics into small and valuable substances, known as chemical recovery methods, is one of the promising and under-developing approaches to decrease the serious environmental severe of realizing fine polymeric particles such as polyvinyl chloride in aquatic systems. The produced substances can be reused as fuel or chemical feedstock. Chlorine residue in oil products obtained from polyvinyl chloride waste limits the application of chemical recovery methods. Simultaneous dichlorination and degradation of the polymeric chain are required to develop a sustainable and rapid process for polyvinyl chloride waste chemical recovery [110]. Kang et al. [120] synthesized magnetic spring-like carbon nanotubes (Mn@NCNT) and evaluated polyethylene-based microplastics degradation performance of robust hybrid carbon based-catalysts via oxidation and hydrothermal hydrolysis with 50% removal efficiency. Toxicity analysis proved a green strategy since all organic intermediates were eco-friendly to the aquatic organisms. Highly stable catalytic performance of Mn@NCNT hybrid catalyst was attributed to the synergetic effects of robust structure, Mn encapsulation, and nitrogen doping, which reduce required activation energy. Miao et al. [110] applied a heterogeneous electro-Fenton like approach for degradation of polyvinyl chloride (PVC) in water using TiO_2/graphite cathode through simultaneous reductive dechlorination and radical oxidation of PVC with 56 wt% removal and dechlorination efficiency of 75% at -0.7 V, 100 °C for 6 h. During the electrocatalytic process, polyvinyl chloride microplastics obtained electrons from the cathode which resulted in the removal of chlorine followed by oxidation of polymeric chain and production of organic intermediates such as carbocyclic acids, alcohols, and esters, which finally converted to CO_2 and H_2O [110]. Ariza-Tarazona et al. [121] studied the visible light catalytic degradation of HDPE microplastics from water using protein-derived C,N-TiO_2 semiconductor catalyst. The best degradation performance was obtained at a low temperature of 0° C and a low pH value of 3 due to the combined effect of pH and temperature on releasing more H^+ ions to the aquatic system and polymer fragmentation. Nabi et al. [122] studied the photocatalytic degradation of polystyrene microspheres and polyethylene microplastic particles using TiO_2 nanoparticle films as a green and cost-effective removal method. Over 12 h illumination of UV light, over 98% degradation of 400 nm polystyrene microspheres was achieved, while faster photocatalytic degradation of polyethylene microplastic was reported over 36 h of UV illumination.

A large portion of released microplastics to aquatic systems are related to textile microfibers such as polyethylene terephthalate, and cellulose-based fibers entered to wastewater system from the effluent of cloths laundering [25]. The pure carbon structure of some extensively useful polymers, including polypropylene, polyethylene, and polyethylene terephthalate, restrict biodegradation using conventional techniques [123]. Compared with photocatalysis [111], electrocatalysis [110], and thermal degradation [114] methods, biodegradation of microplastics has particular strengths such as low operational cost, no need of chemicals, and being applicable for various polymeric particles [48, 124]. Periphytic biofilm was used by Shabbir et al. [48] for biodegradation of different microplastics, including polypropylene, polyethylene, and polyethylene terephthalate in the presence of glucose as an additional carbon source. The results revealed a weight loss ranging from 5.95–14.02% for PP, from 13.24 to 19.72% for PE and from 13.24–19.72% for PET biodegradation after 60 days. Li et al. [125] investigated the effect of prothioconazole as a broad-spectrum fungicide on the degradation of polyethylene and polybutyleneadipote-co-terphthalate (PBAT) microplastics. Biodegradable PBAT microplastics were degraded faster than polyethylene. Degradation of Polyglycerol maleate microbeads of 30 µm as a biodegradable microplastic was evaluated by Hsieh et al. [126] in different aquatic systems such as buffer solution, enzyme solution, deionized water, and seawater. Complete decomposition of microplastics was observed in alkaline solution for 45 min, attributed to surface erosion mechanism. Biodegradation of LDPE using Pseudomonas aeruginosa

ISJ14 via biofilm formation on the polymer surface by microorganisms was proved by Gupta and Devi [127].

5 Conclusion

The occurrence and impacts of plastic particles in water bodies progressively spread worldwide. As reported in the literature, million tons of plastic particles of micron- and nano-size are released into the aquatic environment annually. Studies on microplastic hazards and separation have been growing over the past decade. Many methods have been developed and evaluated on account of the current studies on microplastic particles, which will facilitate to fill the research gap in the future.

Some advanced separation techniques capable of removing small microplastic particles include membrane bioreactors, magnetic-based recoveries, electrocatalytic degradation, photocatalytic degradation, biological degradation, and thermal degradation techniques. Some studies reported almost the same separation efficiency using membrane bioreactor and rapid sand filtration compared with the conventional methods such as activated sludge and removing fiber-like microplastics seems to be challenging by these advanced methods. Magnetic-based adsorbents are introduced as a novel recyclable approach with high adsorption efficiency for microplastic separation with economic feasibility. In some studies, magnetic separation is introduced as an efficient and fast extraction method for clean samples, and it is recommended to utilize as a post-density or post-digestion step in water treatment plants. Applying a continuous collecting system such as rotary magnetic drums in magnetic separation is recommended as well. Recyclable and reusable microsubmarines are developed as a new and environmentally adaptive approach for removing microplastic particles with no need to adding other chemicals. Regarding UV-, or visible light photocatalytic degradation of microplastic particles using carbon nitrides, functionalized ZnO, and TiO_2, there is a gap in evaluating the operating parameters such as pH and temperature. It is also required more investigations to develop a new photocatalyst for complete degradation of microplastics in water. Fragmentation of microplastic particles through the photocatalytic process facilitates degradation through increasing the surface area and interaction between plastic particles and photocatalyst.

There are two general approaches to developing new separation methods in the literature. The first approach is sampling and identifying microplastics in water samples of freshwater bodies or the effluent of WWTPs based on sieving, filtration, and density separation methods. The other approach is removing microplastic particles of various types and sizes using conventional wastewater treatment processes or using integrating new techniques with conventional treatments.

6 Future remarks

There are few reports on the mathematical analysis and modeling of conventional and advanced separation techniques in wastewater treatment to have an effective plant operation. For a more accurate understanding of the environmental consequences of microplastic particles, future investigations should concentrate on the development of new modeling techniques to evaluate the transport route of microplastic particles in the soil, sediments, and water. It is also required to evaluate the impact of organism adsorption on the surface properties of microplastics and their fragmentation.

Despite conducting many attempts to develop approaches for separating and identifying microplastic particles, establishing practical and reliable standard protocols for quantifying microplastic particles with different shapes, sizes, and densities in water bodied and wastewater treatment plants is essential. It is demanding to standardize sieving, chemical digestion, density separation, and visual separation methods in the wastewater treatment plants. In conclusion, an appropriate remedy can be the identification and removal of microplastic resources and penetration routes to monitor inventories of materials or employ novel devices and methods.

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