Filtration methods for microplastic removal in wastewater streams – A review

Usman Salahuddin[1], Jiyu Sun[1] , Chunxiang Zhu[1,2], Pu-Xian Gao[1, 2, *]
[1] *Institute of Materials Science, University of Connecticut, Storrs, CT 06269-3136, USA*
[2]*Department of Materials Science and Engineering, University of Connecticut, Storrs, CT 06269-3136, USA*
** Corresponding author e-mail address: puxian.gao@uconn.edu*

ABSTRACT

Microplastics are commonly recognized as environmental and biotic contaminants. The prevalent presence of microplastics in aquatic settings raises concerns about plastic pollution. Therefore, it is critical to develop methods that can eliminate these microplastics with low cost and high effectiveness. This review concisely provides an overview of various methods and technologies for removing microplastics from wastewater and marine environments. Dynamic membranes and membrane bioreactors are effective in removing microplastics from wastewater. Chemical methods such as coagulation and sedimentation, electrocoagulation, and sol-gel reactions can also be used for microplastic removal. Biological methods such as the use of microorganisms and fungi are also effective for microplastic degradation. Advanced filtration technologies like a combination of membrane bioreactor and activated sludge method show high microplastic removal efficiency.

Keywords: Microplastics, filtration, quantification, water treatment

INTRODUCTION

Microplastics are plastics with a size less than 5 mm, usually formed by degradation and exfoliation of large quantities of plastics released into the ecosystem [1]. A plethora of reports suggest accumulation of these microplastics in the marine environment and their harmful effects on the aquatic life [2] [3]. Microplastics in the ocean can be attributed from different resources, as shown in Figure 1a. In 2018, 35% of microplastics in the ocean come from synthetic textiles; 28% comes from car tires, and 24% is from city dust. It is estimated that microplastics account for 94 percent of the Great Pacific Garbage Patch in the ocean [4]. Besides the challenge of large quantity of microplastics accumulation. Microplastics broad size distribution and hard for accurate quantification make the microplastics study even more complicated. As indicated in Figure 1b, microplastics size distribution can range from 10 µm to 32000 µm. The particle size distribution is highly dependent on the quantification method and locations as comparing data from Cózar et al. (2014) versus Enders et al. (2015). What is more, waste plastics vary significantly in chemical composition, physical form, size, texture, and shape. These characteristics can even evolve while in use and after discard. Hence, the removal of these microplastics is a critical yet challenging topic that needs immediate attention of the research community. Currently, many novel technologies and approaches have been applied to achieve efficient microplastics removal from fresh and saltwater environment. This document entails the methods and technologies most widely reported in the literature for microplastics' removal from wastewater or marine environments.

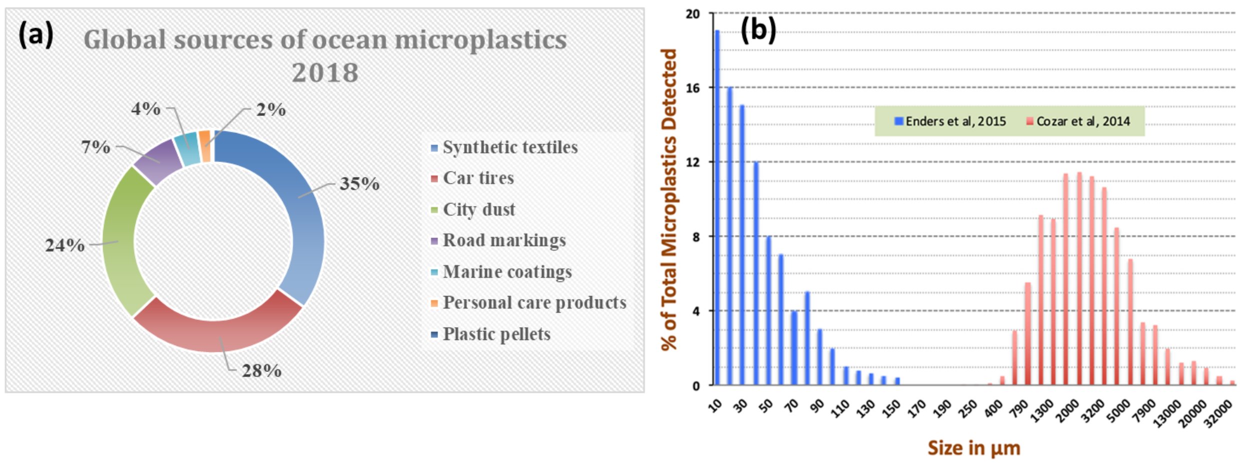

Figure 1. Global sources of ocean microplastics in 2018 and (b) Microplastic size distributions depend on sampling location (geographically and vertically in the water column) and analytical methods applied [4].

Based on the source of plastics, the waste plastics can be characterized into primary and secondary microplastics. Primary microplastics are plastics directly released into the environment in their original form. They can be a voluntary addition to products such as scrubbing agents in toiletries and cosmetics or they can come from the abrasion of large plastic objects during manufacturing, use or maintenance, such as the erosion of tires when driving or of the abrasion of synthetic textiles during washing. Secondary microplastics are microplastics originating from the degradation of larger plastic items into smaller plastic fragments once exposed to marine environment. This happens through photodegradation and other weathering processes of mismanaged waste such as discarded plastic bags or from unintentional losses such as fishing nets [5].

Microplastics can be toxic to marine life and the ecosystem. The potential toxicity of microplastics can be from unreacted monomers, oligomers and chemical additives that tend to leak out from the plastic in the long run or physical effect when absorbed and transferred to living bodies. The toxicity depends also physically on the size and shape of microplastics. Biota and humans are affected by toxic effects of microplastics via mechanisms including sorption and aggregation in different organs, ingestion, and exertion of physical damages. Although a lot of research have been conducted in terms of the microplastics toxicity, effort is still needed to clarify how microplastics induce tissue changes and pathological disorders [6].

As mentioned, accumulation of waste microplastics in the water system has been growing and resulting in significant environmental issues. The use of plastics will only increase in the foreseeable future. Thus, it is vital to find an effective solution to solve the microplastic pollution

and eventually solve the problem from the source. Recently, many efforts have been made and much progress has been achieved for waste microplastics removal in the water system, thus, in-time summary and understanding of the technologies development is necessary to guide future study.
Here, in this short review, we summarize the recent methods and technologies that are used for efficient microplastics removal from waste and marine water systems.

FILTRATION METHODS

Microplastics can be separated from wastewater using dynamic membranes. As shown in Figure 2, Li et al. have reported the effects of variables like particle concentration and influent flux on the removal efficiency of membranes formed on a diatomite platform with a 90 µm supporting mesh [7]. The membrane reduced the turbidity from 195 NTU to less than 1 NTU in just under 20 minutes [8]. Higher influxes of influents and higher concentrations of microplastics facilitate the formation of dynamic membranes.

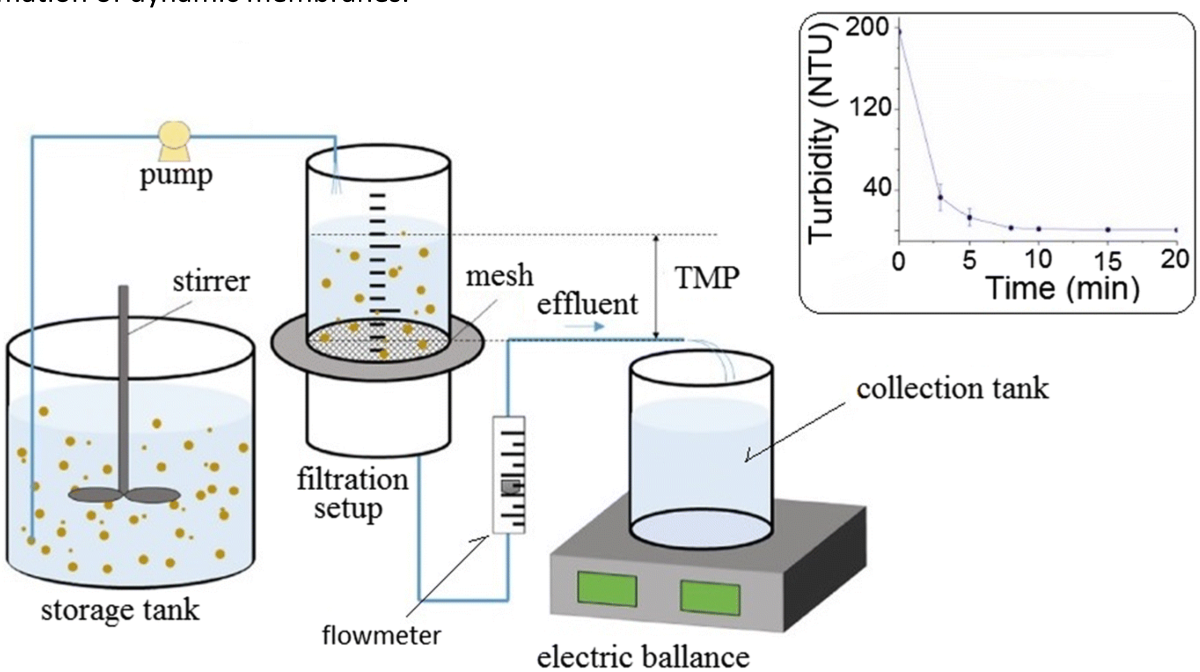

Figure 2. Dynamic membrane experimental setup and graph showing the decrease in turbidity with time when microplastics are removed. TMP transmembrane pressure, NTU nephelometric turbidity unit [6].

Membrane bioreactors is another technique that offers higher capacities relative to dynamic membranes [9]. This technique has the advantage of handling a large variety of wastewater and high strength contaminants. Talvitie et al. compared the removal efficiencies of various technologies such as, disk filters, rapid and filters, dissolved air floating and membrane bioreactors [10]. They showed that membrane bioreactor outperformed all other technologies by eliminating 99.9% of microplastics from the influent stream. The study also showed that membrane bioreactors can remove microplastic particles as small as 20 µm. The shape of the particles also did not seem to affect the efficacy of this method, as FTIR analysis on the effluent stream showed extremely low concentrations of microplastics. This fact hints towards good sorption capabilities of membrane reactors towards microplastics possessing various chemical structures.

CHEMICAL METHODS

Since the size range for microplastics is very small ranging from a couple of microns up to several hundred microns, their separation is a big challenge. Many commercial scale wastewater treatment plants using technologies like coagulation and sedimentation to circumvent this problem [11] [12]. Large contaminant particles that are formed after coagulation and sedimentation processes are easier to separate. These processes usually involve Al or Fe based salts that binds tiny microplastic particles using a ligand exchange mechanism [13]. A recent study investigated the coagulation of polyvinyl chloride particles < 50 µm, using ferric and aluminum sulphate [14]. The use of ferric and aluminum sulphate as coagulants resulted in the removal of about 80% of MPs. Specifically, polyvinyl chloride (PVC) MPs with a size less than 50 µm were successfully coagulated by ferric and aluminium sulphate, and MPs with a size of at least 15 µm were eliminated under optimized coagulation conditions (ferric sulphate at 20 mg L^{-1} and pH 7 or aluminum sulphate at 40 mg L^{-1} and pH 7).

Electrocoagulation is another technique used for microplastic removal that shows higher efficiency, incurs lower cost, allows sludge minimization, etc. [15]. Zeng et al reported the findings that aluminum anode was better than iron anode in the removal of microplastics, and the removal rate of was above 80% in all experiments [16]. As shown in Figure 1, the removal rate of four microplastics by electrocoagulation can reach more than 82 % in the range of pH 3–10, and the best removal rate was 93.2 % for PE, 91.7 % for PMMA, 98.2 % for CA and 98.4 % for PP at pH 7.2.

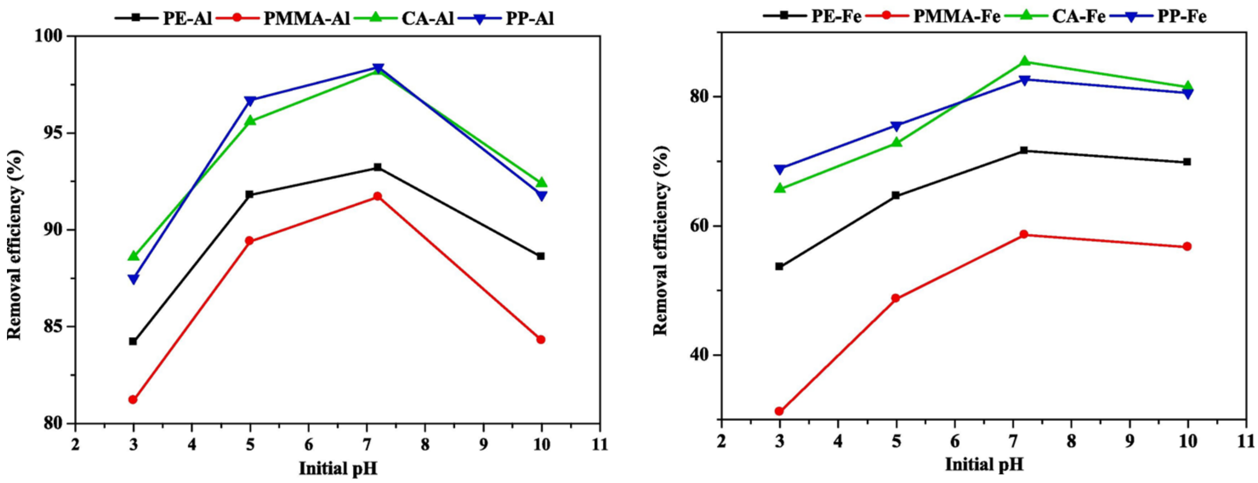

Figure 3. Effect of initial pH on removal of microplastics during electrocoagulation [16]*. (Copyright © 2021 Elsevier B.V.)*

Herbort et al. demonstrated an agglomeration method based on alkoxy-silyl bond formation via a sol-gel reaction [17]. The sol-gel formed in this method resembles a hybrid organic-inorganic silica gel that can be used in a wide variety of applications [18]. Photocatalytic degradation is a well-established method for the destruction of polymeric chains [19]. Li et al have suggested a aqueous phase photochemical method for the degradation of microplastics, in which the degradation reaction is promoted by hydroxyl radicals [20].

Moreover, heterogenous photocatalytic degradation of low-density polyethylene over rod like ZnO nanoparticles in aquatic media has been recently investigated by Tofa et al. [21]. Results showed that the nanorod surface area greatly affected the photocatalytic performance of the reaction. Another study showed the efficacy of titania based nanodevices for microplastic photocatalytic treatment [22]. A similar study shows the ability of protein based TiO_2 photocatalyst to degrade polyethylene microplastics in solid and aqueous media. The results showed a mass loss of 1.1 % and 6.4 % respectively for solid and aqueous phases.

BIOLOGICAL METHODS

Microorganisms including eukaryotes, bacteria and archaea have excellent potential in degradation of microplastics in coastal sediments and marine environments. Dawson et al. studied polyethylene fragmentation and size alteration by Antartic Krill, a planktonic crustacean [23]. The stock suspension contained beads with a mean diameter of 31.5 µm. When particles were isolated from krill, their mean size was 78% smaller than the original beads, with some fragments reduced by 94% of their original diameter, resulting in an average size of 7.1 µm. Fecal material also contained smaller particles with a mean size of 6.0 µm. Moreover, the size distribution of particles within the krill and excreted particles were significantly different from the beads in the exposure stock. The smaller size of plastic particles found in krill and their fecal pellets suggests that the Antarctic krill are physically breaking down the beads after ingestion. Cocca et al. reported a four-month long study on the removal of high-density polyethylene microplastics in seawater using two types of indigenous marine communities, namely, Agios consortium and Souda consortium [24]. The study showed that microplastics acted as a rich carbon source for the microorganisms. Paco et al. demonstrated the use of fungus Zalerion maritimum for the biodegradation of polyethylene in a batch reactor [25]. The results showed that the fungus used microplastics as a nutrient source. Sangale et al. discovered the elite polythene deteriorating fungi (isolated from the rhizosphere soil of Avicennia marina) [26]. The degradation potential of polythene by fungi was evaluated at different pH levels (3.5, 7, and 9.5). The study was carried out by subjecting the polythene samples to continuous shaking at ambient temperature for 60 days and monitoring changes in weight and tensile strength. The BAYF5 isolate (pH 7) exhibited the highest reduction in weight (58.51 ± 8.14), while PNPF15 (pH 3.5) showed the greatest reduction in tensile strength (94.44 ± 2.40). The results of degradation of the polythene strips by the fungal isolates were confirmed by the formation of the cracks/holes/scions and were visualized in Scanning electron microscopic photographs as shown in Figure 4 B.

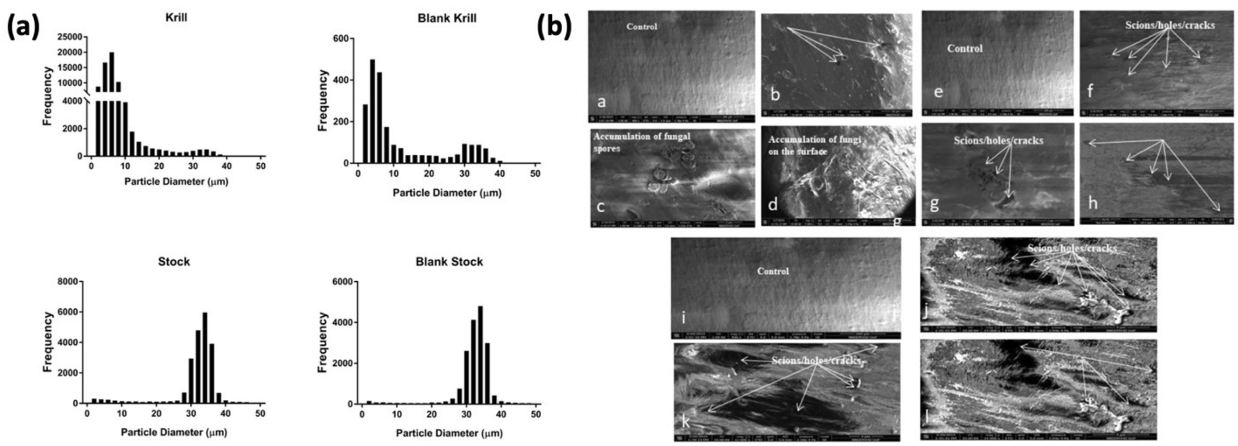

Figure 4. (a) Size distribution of particles isolated from: sample Antarctic krill (Euphausia superba), unhomogenized and enzyme digested Antarctic krill, stock suspension, and stock suspension enzyme digested. Particles between 25 and 50 µm indicate a whole bead; values below this range are fragments. [23] *(Copyright ©, Springer Nature 2018), (b) Scanning Electron Microscopic image of the polythene strips: (a–d) SEM of the PE with most %weight gain (24.4%) by JAMNF at pH9.5; (e–h) SEM of PE strip with maximum (94) % loss in TS by PNPF-15 at pH 3.5; (i–l) SEM of PE strips of maximum %WL (41%) by MANGF1* [27] *(Copyright © 2019, Springer Nature Singapore Pte Ltd. 2019)*

Figure 5. A, Size distribution of particles isolated from: sample Antarctic krill (Euphausia superba), unhomogenized and enzyme digested Antarctic krill, stock suspension, and stock suspension enzyme digested. Particles between 25 and 50 µm indicate a whole bead; values below this range are fragments. [23] *(Copyright © The Author(s) 2018); B, Scanning Electron Microscopic image of the polythene strips: (a–d) SEM of the PE with most %weight gain (24.4%) by JAMNF at pH9.5; (e–h) SEM of PE strip with maximum (94) % loss in TS by PNPF-15 at pH 3.5; (i–l) SEM of PE strips of maximum %WL (41%) by MANGF1* [27] *(Copyright © The Author(s) 2019)*

ADVANCED FILTRATION TECHNOLOGIES

A combination of membrane bioreactor and activated sludge method was recently studied by Lares et al. at a pilot scale [28]. They collected samples every 2 weeks for 3 months. The pilot scale setup included an aeration tank for microorganism activation, a sedimentation tank for sludge separation and a secondary purifier. A microplastic removal efficiency of 99.4 % was reported. Similarly, a study in China showed a comparison of 11 different wastewater treatment plants for the efficiency to remove microplastics [29]. An efficiency of above 90 % was witnessed by all the plants that used multiple filtration steps.
Moreover, another Chinese study analyzed the removal methodology used by municipal sewage treatment plants for the removal of microplastics [30]. The study showed that the influents were treated initially using a series of processes, such as grit chamber, primary and secondary sedimentation tanks, anerobic, aerobic and anoxic treatments. Advanced treatment processes such as, denitrification, ultrafiltration, ozonation and ultraviolet treatment were later applied to remove the microplastics. The overall efficiency of plastic removal was reported to be 95.16 % which was still less than membrane bioreactors (99.9 %) The study also reported that sewage treatment plants release a large quantity of microplastics into the aquatic ecosystem as these plants are not primarily designed for microplastic removal.

Table 1. Comparison of available microplastic filtration methods [9] [10]

Technology	**Membrane Filters**	**Chemical/ Electrochemical Methods**	**Biological Methods**	**Other multistage filtration techniques**
Filtration size range (µm)	> 90	2000-5000	1-30	> 90
Efficiency (%)	90-99	60-65	< 15	97-99
Regeneration	Highly energy intensive	Not possible	Easy	Complicated
Scalability	Scalable but usually high cost	Scalable but of chemical and or electrical current constraints	Limited due to low plastic consumption rates of microorganisms,	Possible but costly due to multistage processes
Limitations	Expensive regeneration & costly membrane filters	Water quality affected by chemical treatment,	Slow kinetics of biological digestion	Multi step processes with high cost

	requiring post treatments	

CONCLUSIONS AND OUTLOOK

Microplastics are widely acknowledged as contaminants in both the environment and biota. Their omnipresence in aquatic environments has raised concerns about plastic pollution, highlighting the urgent need for effective and affordable removal methods. This paper presents an overview of the various techniques and technologies used to remove microplastics from marine environments and wastewater. Dynamic membranes and membrane bioreactors are effective in removing microplastics from wastewater, while chemical methods such as coagulation and sedimentation, electrocoagulation, and sol-gel reactions can also be used. Biological methods, such as the use of microorganisms and fungi, have also proven effective for microplastic degradation. Advanced filtration technologies, such as a combination of membrane bioreactor and activated sludge method, demonstrate high microplastic removal efficiency.

As the detrimental impacts of microplastics become more apparent, the demand for effective filtration methods continues to grow. In the future, it is likely that research will focus on developing more cost-effective and scalable technologies for removing microplastics from wastewater. Additionally, there may be a shift towards more sustainable and eco-friendly filtration methods, such as biological methods. Advances in nanotechnology may also lead to the development of more efficient and precise filtration methods. This review clearly shows the need for a robust, cheap, and efficient method for microplastic removal from waste streams. Conventional wastewater treatment plants are not typically designed to filter microplastics and hence any supplementary microplastic filtration devices that can be retrofitted to the existing wastewater treatment plants could really solve this problem. However, these retrofitting need to be modular and scalable in nature to accommodate the variable volumes of wastewater.

As of now, there are no regulations or laws restricting the water treatment plants for assessing and minimizing the microplastics concentrations in their effluent water. If regulated, these treatment plants will be forced to retrofit their treatment plants with microplastic capturing filters. In addition to the methods reported in this paper, there has been an increasing interest in using ceramic based filters for microplastics filtration. These ceramic filters have proven to be a low cost and robust alternative to all other filters reported in this review.

Another challenging problem in microplastic filtration is microplastic quantification in water-based samples. Since microplastic particles are neutral in charge, non-reactive and extremely resistant to any solvent or chemical, it is extremely difficult to quantify them. Available methods include microscopic counting and fluorescent tagging, but these methods only quantify in terms of number of particles per unit volume of water. What is needed is a scalable technique that can accurately provide a weight-based quantification of microplastics in water samples. The significance of microplastic quantification is two folds. First, weight-based quantification is crucial to assessing water quality. Second, it also helps in assessing the efficacy of a given microplastic filtration device.

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