

*Review Article***Microplastics Removal Strategies in Aquatic Environments****Edhi Widiyanto^{1,2}, Titik Istirokhatun^{1,2*}, Pertiwi Andarani^{1,2}, Heru Susanto^{2,3}, Ria Desiriani¹, Pamella Apriliana⁴**¹ Department of Environmental Engineering, Faculty of Engineering, Universitas Diponegoro, Semarang, 50275, Indonesia² Membrane Research Center (MeR-C), Integrated Laboratory for Research and Services, Diponegoro University, Semarang, 50275, Indonesia³ Department of Chemical Engineering, Faculty of Engineering, Universitas Diponegoro, Semarang, 50275, Indonesia⁴ Department of Chemical Science and Engineering, Graduate School of Engineering, Kobe University, Kobe, 657-8501, Japan*Corresponding Author, email: titik.istirokhatun@live.undip.ac.id**Abstract**

Microplastics (MPs) have been globally detected in aquatic environments. The abundance of MPs contributed to the negative effects on aquatic ecosystems. Thus, it's critical to create effective solutions for removing MPs from water. In this review, we compared several methods, including physical, physicochemical, and biological approaches, towards membrane filtration. The physical filtration technology is the simplest way in comparison with other methods. However, the removal ability of physical filtration against smaller MPs than 20 μm becomes a crucial concern. Then, the other option is an adsorption method. Although the adsorption option is an inexpensive method, the undesirable aspect during adsorbent usage may not be environmentally friendly in aquatic systems. The similar problem is also demonstrated by chemical approaches in terms of coagulation and electrocoagulation treatment. Consequently, the biological methods were found to be less toxic to the environment. Even though it provides safe conditions to the environment, the biological approach needs a long time to degrade MPs. To overcome their disadvantages, the membrane technology offers efficient removal of MPs and no addition of chemical usage. However, the main point to pay attention to is that each technology has benefits and drawbacks. Therefore, the application of multiple technologies for MPs removal is considered.

Keywords: Microplastics, removal, filtration, aquatics, environment**1. Introduction**

In the early decades of the 20th century, plastic was discovered in different industries due to its low weight, excellent strength, long-term stability, and low price (Mendonça et al., 2023). Therefore, this is unavoidable in almost every aspect of daily life. Plastics began to be manufactured commercially in the 1950s, then plastic production increased significantly from 335 metric tons of global plastic production in 2016 and is expected to reach more than 12,000 million metric tons in 2050 (Ferreira et al., 2023). The plastic production, garment washing, and textile industries are the primary contributors to donating plastic rubbish. Moreover, the exposure of plastic garbage every day under sunlight and weathering will break down plastic material into tiny bits, these are called microplastics (MPs). The size of MPs is mainly from 100 up to 500 μm . Commonly, the release of MPs was manufactured from different polymer-based products such as polyethylene (PE), polystyrene (PS), and polyethylene terephthalate (PET) (Zhao et al., 2024). For example, the overabundant consumption of masks during the COVID pandemic resulted in

the generation of significant amounts of microplastic (MPs) that flows into rivers accidentally or intentionally (Fadare and Okoffo, 2020). The presence of MPs of 58-193 items/m³ in March to 71-1265 items/m³ in October from Antua River, Portugal (Rodrigues et al., 2018). Alongside, the MPs was also coming from tire milling waste with a total amount of 0.81 kg/person per year worldwide (Harahap et al., 2024; Mahesh et al., 2023).

Due to the fact that the MPs is not perfectly dissolved in aquatic environments, the presence of plastic garbage has contributed to the negative effects to aquatic ecosystems. As illustrated in Figure 1, MPs can enter the food chain of aquatic organisms such as animals or plants, and bioaccumulate in their tissues. Then, the effect of MPs when accidentally consumed by humans as the top of the main food chain, resulting in implications for inflammation and cell function, even worse will cause tumor and cancer. Thus, the abundance of MPs not only has crucial risk to body water contaminant, but also to survival living things i.e plant, animal, and human (Al Mamun et al., 2023; Cverenkárová et al., 2021; Dissanayake et al., 2022). Hence, it is critical to create efficient and environmentally friendly ways for MPs removal.

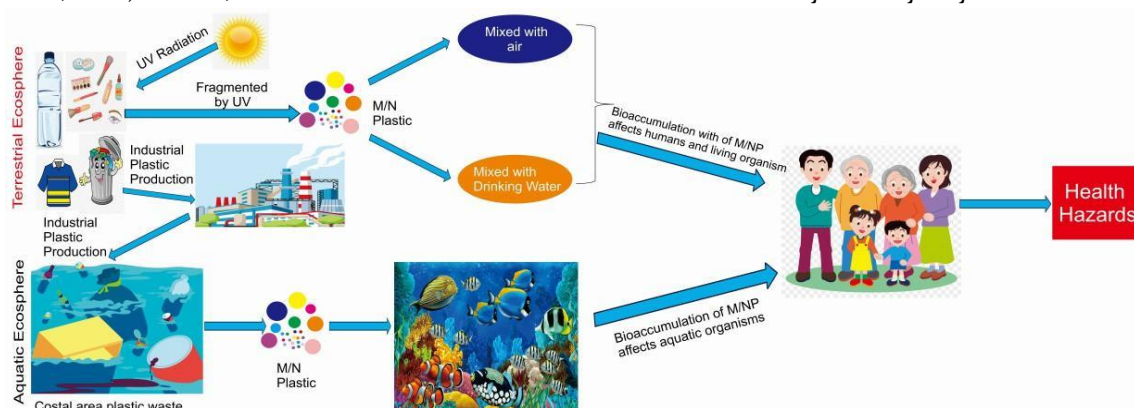


Figure 1. The pathways of microplastics enter the food chain processes.

Recently, various techniques with promising findings are broadly offered for efficient MPs removal, which can be divided into biological (microorganism aggregation and microbial degradation) (Amjad et al., 2023), physicochemical (adsorption technology, coagulation-flocculation-sedimentation, electrocoagulation) (Perren et al., 2018), physical (filtering, disk filter, and nylon filter) (Simon et al., 2019), and membrane filtration (Patterson, 2021; Poerio et al., 2019; Takeuchi et al., 2023). Even though each of these methods has its own applications, membrane technology has numerous benefits among other separation techniques, including low energy usage, flexibility, and environmental friendliness. Thus, this review explains recent numerous methods considered for MPs removal, especially membrane filtration, given observations regarding the strengths and limitations of various technologies.

2. Methodology Analysis

A complete literature search was undertaken in Web Science databases, Google Scholar, ScienceDirect, and Scopus using the keywords "microplastics" AND "removal" AND "membrane" AND "microfiltration" AND "ultrafiltration" AND "nanofiltration". Furthermore, the following searches were performed: "microplastic" AND "removal" AND "physical" AND "physicochemical" AND "biological" AND "treatment". The studies were selected for their relevance to the subjects covered in this review. They were thoroughly analyzed and discussed in the following parts. Fig. 2 was created by analysing articles with appropriate keywords. The years 2021 and 2022 had the most articles discovered reaching over 3400 – 5400 publications.

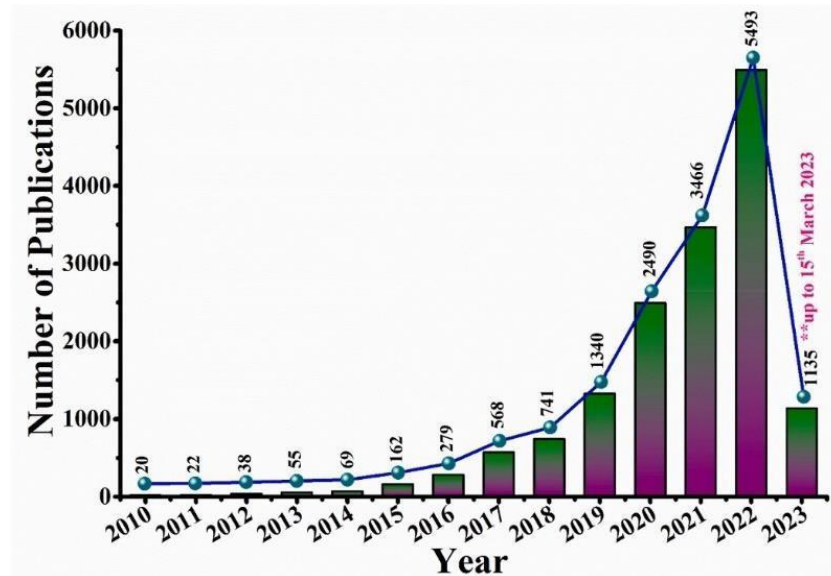


Figure 2. Number of publications on removal of MPs by varied technology on March 10, 2023.

3. MPs Measurement by Quantitative and Qualitative Methods

Currently, the general assessment of MPs is divided into quantitative and qualitative, as listed in Table 1. The visual observation mainly relies on human senses. The limitation of visual observation techniques using a microscope is that the size of the microplastic is smaller than the microscope's ability to detect it. So a weighting method is needed if the size of the parliamentarians is too small. Although the weighting methods demonstrate low energy cost, the inaccurate and complex preprocessing becomes a major challenge.

The instrument of chromatographic techniques was provided by chromatography-mass spectrometry (GC-MS) and liquid chromatography (LC) tools. Due to the chromatographic only analyze mass spectrum, the information of size and quality is not provided by GC-MS and LC. Meanwhile, the information of size and quality was given by a combining scanning electron microscopy (SEM) with energy dispersive X-ray spectrophotometry (EDX), surface-enhanced Raman spectroscopy (SERS), dynamic light scattering (DLS), and Fourier transform infrared spectroscopy (FTIR) measurements. Therefore, it is important to investigate the presence and concentration of MPs by existing instruments.

Table 1. Comparison of qualitative and quantitative M/NP analysis methods

Method	Qualitative/Quantitative	M/NP detection range	Properties
Visual Observation	Quantitative (number, size)	no size limit	Less cost and quality, prone to human error, need microscope analysis
Weigh in	Quantitative (mass)	no size limit	Less cost, require for other analysis (filtration)
Turbidity	Quantitative (concentration)	Not suitable for small-density M/NP	Simply to use, rapid, wide range measurement, vulnerable to external particle interference, unsuitable for calculating MPs density.
GC-MS	Qualitative	no size limit	High accuracy, unable provide size and number of MPs

Method	Qualitative/Quantitative	M/NP detection range	Properties
LC	Qualitative	no size limit	High accuracy, unable provide size and number of MPs
FTIR	Qualitative	$\geq 20 \mu\text{m}$	Not suitable for nano MPs, expensive, requires qualified analysis
Raman	Qualitative	$\geq 1 \text{ mikron}$	Fast and non destructive method, sensitive to non polar functional groups, susceptible to microorganism contaminants and organic or inorganic substances
SERS	Qualitative	$\geq 50 \text{ nm}$	High sensitivity
SEM-EDX	Quantitative (number, size)	no size limit	Provide high-resolution images of MPs, composition morphology and surface elements of MPs can be obtained, high cost, requires complicated pre-processing steps
DLS	Qualitative (size)	1 nm - 10 mikron	Fast and accurate, suitable for the determination of molecular weight and molecular size, sensitive to changes in temperature and viscosity

Source: (Liu et al., 2022)

4. MPs Removal through Physical, Chemical, and Biological Approaches

4.1. Physical Methods for MPs removal

4.1.1. Filtering

As presented in Table 2, the comparison of filtering technology for MPs removal in water. The filtering technology has been widely known as cheap and easy to use for MPs removal technology in water. Its mechanism is mainly dependent on trapping MPs through a multiple filter size. However, the limitless basic filtering process is less selectivity (Ahmed et al., 2024). Researchers recommend that filtration is an effective removal rate when the particle size is larger than $20 \mu\text{m}$ (Liu et al., 2024). Thus, key factors such as size and shape of MPs and filter characteristics consisting of pore size, thickness, and mesh size significantly affect the filtering process (Farooq et al., 2023).

Table 2. Types of filtration methods for MPs removal

Filtration method/device	Pore size	MPs Removal Rate (%)	Reference
Elution divace	1 mm	50.2	(Zhu, 2015)
Disc filter	$10 \mu\text{m}$, $20 \mu\text{m}$ (disc filter)	40 – 98.5	(Talvitie et al., 2017)
Disc filter	$18 \mu\text{m}$	87	(Simon et al., 2019)

Filtration method/device	Pore size	MPs Removal Rate (%)	Reference
Nylon filter (NY)	8 μm	± 92	(Cai et al., 2020)
Filter container	100 μm , 50 μm , 10 μm	79	(Funck et al., 2021)

4.2. Physicochemical Methods for MPs Removal

4.2.1. Adsorption Technology

The exploration of adsorption according to the adsorption mechanism between MPs and adsorbents. The adsorption technique is remarkably successful at removing MPs from aquatic environments due to being eco-friendly and easily accessible in the ecosystem. During the adsorption process, the effects of electrostatic interactions, hydrogen bond interactions, and π - π interactions, as demonstrated in Figure 3. Many researchers introduced granular or pulverous adsorbent for removal of MPs (Aguiar et al., 2022; Rout et al., 2022; Verma et al., 2024). The granular adsorbents are typically larger than pulverous adsorbents, thus they have a relatively large surface area and are commonly used in packed bed columns for large-scale industrial applications. The most widely known granular adsorbents are activated carbons and zeolite granular. A previous study observed the modified cationic surfactant with zeolites for MPs removal. The result showed that polyethylene (PE) and polyamide (PA) as MPs had a significant removal (>96%) in comparison with basic rapid sand filter (63%) (Shen et al., 2021). The modified materials in adsorption technology presented a wide scope of potential for removal of MPs in water or wastewater treatment processes. The granular activated carbon as adsorbent demonstrated MPs removal up to 95.5% (Amirah Mohd Napi et al., 2023).

Meanwhile, the pulverous adsorbents are composed of finely powdered materials and mainly used for applications which need rapid adsorption kinetics such as in catalysts. The silica gel powder and activated alumina has been found as part of pulverous adsorbents. The significant removal of MPs was observed by synthesized sponge material with graphene and biochar compounds [85]. The sponge material rich of silica was synthesized by chitin and graphene oxide (ChGO) combined with OeC_3N_4 demonstrated high removal efficiency of 81,2% (Sun et al., 2020). Not only effective to remove MPs from water, the ChGO sponge was friendly and non-toxic for the environment. The other comparison method of MPs removal by adsorption method has been listed in Table 3.

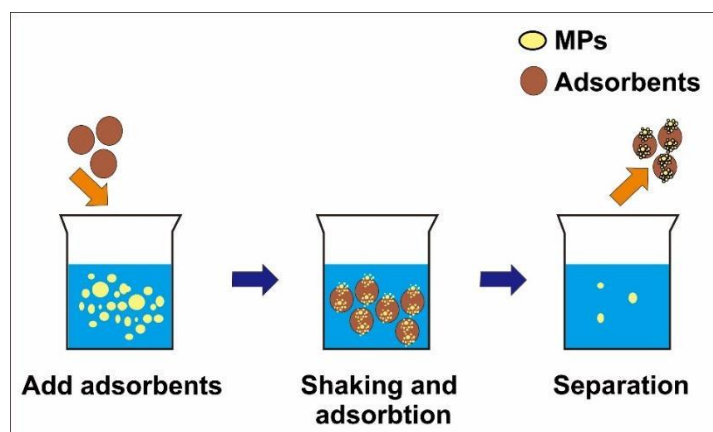


Figure 3. Schematic of MPs removal by adsorption process

Table 3. Comparison study of MPs removal by adsorption method

Adsorbents	M/NP	Related parameter	Removal rate (%)	Adsorption capacity (mg/g)	References
Fe ₃ O ₄ particles (magnetic seeds)	PVC (2.06 µm), PMMA (5.98 µm)	Time : 0-10 min, pH : 1-11	95	-	(Rhein et al., 2019)
Hydrophobic Fe nanoparticles	MPs (<8 mm)	Adsorbent : 2 mg, Type of matrices : artificial seawater, fresh water, sediment	92-93 (artificial seawater), 84 (freshwater), 78 (sediment)	-	(Grbic et al., 2019)
Three-dimension reduced graphene oxide	PS (5 µm)	Adsorbent : 1.5 mg, MPs : 0.1-0.8 g/L, pH : 2-10, Time : 2 h	89	617.2	(Yuan et al., 2020)
Biochar, Fe ₃ O ₄ -biochar	PS (0.02, 0.2, 2 µm)	MPs : 4 mg/L, Adsorbent : 10 cm in column, Flow rate : 0.73 mL/min	83.5-92.5	-	(Tong MeiPing et al., 2020)
Chitin and graphene oxide sponge	PS, PS-NH ₂ , PS-COOH (1 µm)	MPs : 1 mg/mL, Adsorbent : 1 x 1 x 1 cm, pH : 4, 6, 8, 10, Temperatur : 25, 35, 45°C	72.4-89.8	5.8-8.4	(Sun et al., 2020)
Geothite, magnetite, kaolinite, montmorillonite	PS (50 nm)	Adsorbent : 20 mg, MPs : 0.10 mg/L, Time : 12 h, pH : 2-11, Ions : Na ⁺ , Ca ²⁺ , Fe ³⁺	40-80 (kaolinite and montmorillonite no effect)	-	(Yangyang Zhang et al., 2020)

Adsorbents	M/NP	Related parameter	Removal rate (%)	Adsorption capacity (mg/g)	References
Magnetic polyoxometalate (POM) supported ionic liquid phase	PS (1, 10 µm)	MPs : 1 g/L, 5 mL, Adsorbent : 50 mg, Time : 24 h	100	-	(Diagbaya et al., 2020)
Chitin based sponges (Ch, ChCN, ChGO, ChGO-CL, ChGO-CT)	PS, PS-NH ₂ , PS-COOH (1 µm)	MPs : 1 mg/mL, Adsorbent : 1 x 1 x 1 cm, pH : 4, 6, 8, 10, Temperatur : 25, 35, 45°C, Time : 48 h	63,3-92,1	4.87-12.9	(Siipola et al., 2020)
Magnetic carbon nanotubes	PE, PET, PA (48 µm)	MPs : 5 g/L, Adsorbent : 2-7 g/L, Times : 5 h	-	1650 (PE), 1400 (PET), 1100 (PA)	(C. Sun et al., 2021)
Magnetic material (Fe-modified fly ash)	PS (80 nm)	MPs : 1-30 mg/L, 10 mL, Adsorbent : 20 mg, pH : 3-10, Time : 0.5-30 h, Temperatur : 25, 35, 45°C, kons : Ca ⁺ , Mg ²⁺ , Na ⁺ , K ⁺ , SO ₄ ⁺ , Cl ⁻ , NO ₃ ⁻ , PO ₄ ³⁻	-	82.8-89.9	(Tang et al., 2021)

4.2.2. Coagulation Technology

Coagulation plays an essential purpose in removing MPs pollutants (Lee and Jung, 2022). In concept, the MPs removal method is divided into three steps including coagulant rapid mixing (M), flocculation (F), and sedimentation (S), as illustrated in Fig. 4. First, the MPs were dispersed by coagulant via a mixing process. Then, the coagulant rapidly chains the MPs targeted. Finally, the MPs become aggregated in the form of sediment.

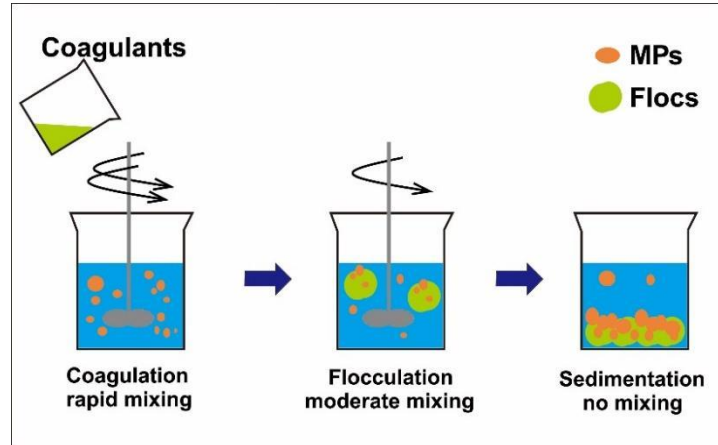


Figure 4. The schematic process of coagulant method for MPs removal

During the coagulation process, coagulants destabilize and aggregate suspended MPs, then the large flocs are formed due to interaction between coagulants and MPs. The use of coagulant to remove MPs from water has been widely used. Iron and aluminum-based salts are generally employed as coagulants. For instance, the coagulant of $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ was used for remove polyethylene(PE) (Ma et al., 2019a) (see Table 4 for details). During the filtration process, the addition of polyacrylamide (PAM) helps to improve the removal efficiency from 12.65% and 36.89% for without PAM and 61.19% for with PAM, respectively (Skaf et al., 2020). Furthermore, the comparison between coagulant FeCl_3 and polyaluminum chloride (PAC) was also employed for polysulfone (PS) removal. The result demonstrated that the removal rate of FeCl_3 was slightly higher than PAC, where 99.4% for FeCl_3 and 98.2% for PAC, respectively (Chen et al., 2020).

Table 4. The relevant study of MPs removal through coagulant method

Tyoes of coagulant/flocculant	MPs	Coagulant method	Removal rate (%)	References
Fe-based coagulant ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$)	PE (<5 mm)	M: 300 rpm, 1 min ; F: 100rpm, 14 min ; S: 30 min	85.21-90.91 (with anionic PAM)	(Ma et al., 2019a)
Al-based coagulant ($\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$), fe-based coagulant ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$)	PE (<5 mm)	M: 300 rpm, 1 min ; F: 100rpm, 14 min ; S: 30 min	12.65 (Fe), 36.89 (Al), 61.19 (Al with anionic PAM)	(Ma et al., 2019b)
FeCl_3 , polyaluminum chloride (PAC)	PS (1 ; 6.3 μm)	M: 400 rpm, 1 min; F: 40 rpm, 20 min; S: 30 min	99.4 (FeCl_3), 98.2 (PAC)	(Rajala et al., 2020)
Ca/Al dual flocculant	PS (100 nm)	M: 120 rpm, 2 min ; F: 80 rpm, 20 min ; S: 6 h	>80	(Chen et al., 2020)

Types of coagulant/flocculant	MPs	Coagulant method	Removal rate (%)	References
$\text{Al}_2(\text{SO}_4)_3$ coagulant	PP, PS (180 nm-125 μm)	M: 200 rpm, 1 min ; F: (i) 70 rpm, 2 min (ii) 39 rpm, 5 min (iii) 5 rpm, 5 min ; S: 15 min	≤ 1.8 ; ≤ 13.6 (with PolyDADMAC)	(Yongli Zhang et al., 2020)
Alum [$\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$]	PE (5-15 μm), rayon (8.7-20.6 μm)	M: 100 rpm, 1 min ; F: 30 rpm, 30 min ; S: 30 min	86-99	(Skaf et al., 2020)
Al_3 coagulant (AlCl_3)	PET, weathered PET (500 \pm 2.5 nm)	M: 200 rpm, 1.5 min ; F: 40 rpm, 10 min ; S: 30 min	100 (PET) ; 92 (weathered PET)	(Lu et al., 2021)
$\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$	PS (53-500 μm), PE (500-1000 μm)	M: 300 rpm, 1 min ; F: 50 rpm, 15 min ; S: 30 min	83.3 ± 3.9 (PS) ; 59.4 ± 5.2 (PE)	(Arvaniti et al., 2021)
$\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$	PE in wastewater	M: 350 rpm, 1 min ; F: 100 rpm, 15 min ; S: 30 min	96.10 ($\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$) ; 70.56 ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$)	(Esfandiari and Mowla, 2021)

*M: mixing, F: flocculation, S: sedimentation

4.2.3. Electrocoagulation

The principle of electrocoagulation technology is an electrolytic cell, where the submerged anodes and cathodes are connected by direct current (DC) power source (Kim and Park, 2021). In the electrocoagulation process, there are three stages of the ion. First, the cations are separated from the anode by the electric field to obtain "microcoagulants"; second, microcoagulants combine and crash with MPs, thus MPs can remove from water (Mateo et al., 2024). In the current year, the utilization of electrocoagulation has been used to remove MPs contaminants from water or wastewater. Al, Fe, Cu are the most commonly used as electrodes (Shen et al., 2022). In principle, Al and Fe electrodes served as anode, while Cu electrodes acted as the cathode, as shown in Fig. 5.

The mechanisms of electroagulation divided into two distinct operates. First, Al^{3+} effeiciently binds to water molecules, leading the formation of $\text{Al}(\text{H}_2\text{O})_6^{3+}$ ions. Then, the presence of H^+ ions in $\text{Al}(\text{H}_2\text{O})_6^{3+}$ was released by hydrolysis procces, resulting in the formation mononuclear hydroxides such as $\text{Al}(\text{H}_2\text{O})_3\text{OH}^{2+}$, $\text{Al}(\text{H}_2\text{O})_4\text{OH}^{2+}$, and $\text{Al}(\text{H}_2\text{O})_5\text{OH}^{2+}$. Secondly, Al^{3+} reacts with OH^- to form $\text{Al}(\text{OH})^{2+}$ and $\text{Al}(\text{OH})^3$. The unsaturated hydroxyl groups in hydroxyl Al ions facilitate for polymerization with supplementary Al^{3+} ions, resulting in hydroxyl bridges. Consequently, the hydroxyl binds the polymer network of MPs (Liowska-Bizukojć and Olejnik, 2020). The procedure of removing MPs by electrocoagulation technique as described in Fig. 6.

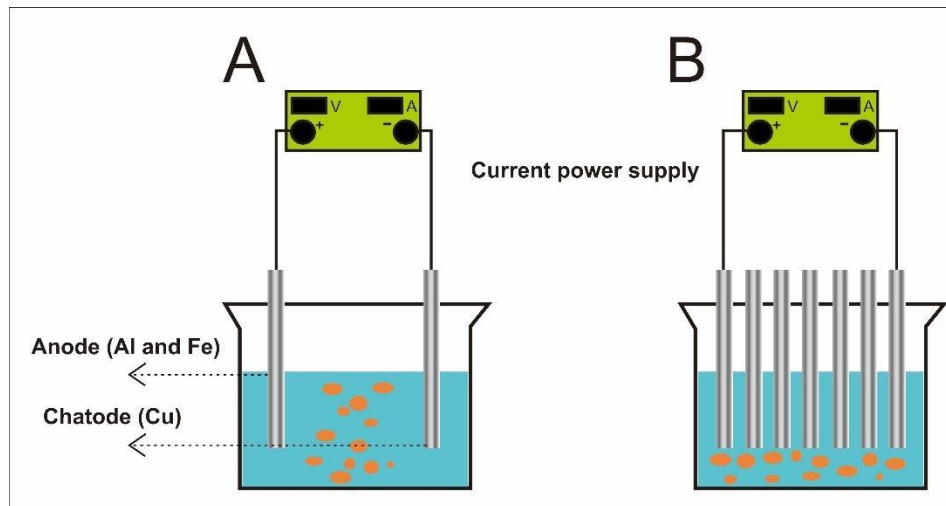


Figure 5. Electrocoagulation method for MPs removal (A) Monopolar electrode and (B) Bipolar electrode

The electrocoagulation has been used for several MPs removal, as presented in Table 5. For example, a previous study demonstrated that the four types of electrodes (Al-Fe, Al-Al, Fe-Fe, and Fe-Al) were used as anode and cathode for removal of MPs of polyamide. The results showed that the highest removal of MPs was obtained by Fe-Al electrodes, followed by Fe-Fe, Al-Fe, and Al-Al, respectively. The promising outcomes of Fe-Al electrodes is due to the fact that Fe and Al has generated each other in electrolytic cell (Hu et al., 2023).

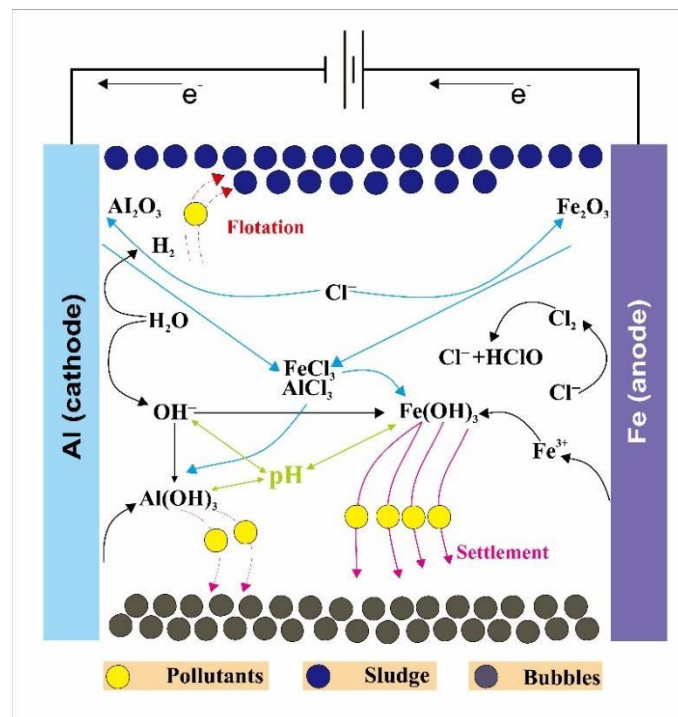


Figure 6. The mechanism of removing pollutants by electrocoagulation technique.

Table 5. Comparison study of electrocoagulation methods for MPs removal

Electrodes	MPs types	Removal rate (%)	References
Seven aluminum electrodes (90 mm x 30 mm x 1 mm)	Microbeads (300-355 µm)	99.24	(Perren et al., 2018)
Activated carbon (AC) electrode (75 µm)	PS, aged PS (40 nm)	0.707 plastic/g AC (PS) ; 0.322 g plastic/g AC (aged PS)	(Akarsu et al., 2022)
Aluminum plates (30 cm x 2.54 cm x 0.25)	Polyester (25-65 µm)	99 ; 96.5 (real wastewater)	(Elkhatib et al., 2021)
Seven aluminum electrodes (100 mm x 50 mm x 2 mm)	Microbeads (38-45 µm)	90	(Senathirajah et al., 2023)
Al-Fe plates, Fe-Al plates (90 mm x 60 mm x 1 mm)	PE	100	(Liu et al., 2023)
Al-Fe plates, Fe-Fe plates, Fe-Al plates (90 mm x 60 mm x 1 mm)	MPs in laundry wastewater (15.804 MPs/L)	98	(Akarsu et al., 2022)
Anode: Al plate or Fe plate ; Cathode: Cu plate (4 cm x 6 cm x 0.1 cm)	PE (6.3 µm 286.7 µm), polymethylmethacrylate (PMMA) (6.3 µm 286.7 µm), cellulose acetate (CA) (1-2 mm), PP (1-2 mm)	93.2 (PE) ; 91.7 (PMMA) ; 98.2 (CA) ; 98.4 (PP)	(Krishnan et al., 2023)

4.3. Biological Method

Biodegradation is a cost-effective and environmentally friendly technique of removing MPs. Although most microplastics are biodegradable, some microorganisms and insects have been discovered to destroy traditional plastic. Meanwhile, several biological approaches have been represented to effectively remove MPs from the aquatic. In aquatic environment, the formation of biofilms was provided by microbiotas. The hydrophobic surfaces of biofilm can attract MPs. Commonly, the type of hydrophobic surfaces was natural or artificial membranes. A previous research demonstrated that the varied of biofilm such as fungus, bacteria, and enzyme may cause damage of MPs by modifying MPs and eventually decomposing MPs, as demonstrated in Table 6-8. The construction of biofilm have been used in MPs removal treatment. For example, the *Phanerochaete chrysosporium*, *Bacillus subtilis*, and *Aspergillus tubingensis* were significantly remove MPs of PP, HDPE, and LDPE, respectively. However, the limitless biological methods required a long time of degraded MPs and less degradation rate in comparison with basic filter, coagulation, and electrocuagulation method.

Table 6. The comparison study of MPs removal through microbial degradation

Microorganisms	MPs	Time (days)	Temperature (°C)	Degradation rate (%)	References
<i>Phanerochaete chrysosporium</i>	PP film (50 µm)	360 days	30	18.8	(Jeyakumar et al., 2013)
<i>Engyonitium album</i>	PP film (50 µm)	360 days	30	9.42	(Jeyakumar et al., 2013)
<i>Aspergillus spp</i>	HDPE (40 µm)	30	30	6.02-851	(Devi et al., 2015)
<i>Pseudomonas aeruginosa E7</i>	Low molecular weight PE	80	37	40.8	(Jeon and Kim, 2015)
<i>Bacillus subtilis</i>	LDPE film (18 µm), HDPE film (41 µm)	30	32	9.26	(Vimala and Mathew, 2016)
<i>Bacillus spp.</i>	High Impact PS film	30	30	23	(Mohan et al., 2016)
<i>Pseudomonas spp</i>	High Impact PS film	30	30	< 10	(Mohan et al., 2016)
<i>Stenotrophomonas pavani</i>	LDPE film (21 µm)	56	30	< 25	(Mehmood et al., 2016)
<i>Bacillus ceureus</i>	PE, PET, PS (75 µm)	40	room temperature	1.6 (PE), 6.6 (PET), 7.4 (PS)	(H. S. Auta et al., 2017a)
<i>Bacillus gottheilii</i>	PE, PET, PS, PP (75 µm)	40	room temperature	6.2 (PE), 3.0 (PET), 5.8 (PS), 3.6 (PP)	(H. S. Auta et al., 2017a)
<i>Bacillus ceureus</i>	PP MPs	40	room temperature	12	(S. H. Auta et al., 2017)
<i>Sporosarcina globispora</i>	PP MPs	40	room temperature	11	(S. H. Auta et al., 2017)
<i>Comamonas sp., Delftia sp., Stenotrophomonas sp.</i>	PE film (100 µm)	90	No data	46.7 (viscous area)	(Peixoto et al., 2017)
<i>Zalerion maritimum</i>	PE (250-1000 µm)	28	25	43	(Paço et al., 2017)
<i>Bacillus sp.</i>	PP MPs	40	room temperature	4	(H. S. Auta et al., 2017b)
<i>Rhodococcus sp.</i>	PP MPs	40	room temperature	6.4	(H. S. Auta et al., 2017b)
<i>Brevibacillus sps. & Aneurinibacillus sp.</i>	LDPE, HDPE, PP (2.5 mm)	140	50	58 (LDPE), 47 (HDPE), 56 (PP)	(Skariyachan et al., 2018)
<i>Pseudomonas sp.</i>	PP (250 µm-4 mm)	40	10	17.3	(Habib et al., 2020)
<i>Rhodococcus sp.</i>	PP (250 µm-4 mm)	40	10	7.3	(Habib et al., 2020)

Table 7. The relevant study of MPs degradation with fungus strains

Microorganisms	Strains	MPs	Efficiency of degradation	References
Fungi	<i>Malbranchea cinnamomea</i>	Dipropyl phthalate, Dibutyl phthalate, Dihexyl phthalate	> 90%	(Duan et al., 2019)
Fungi	<i>Aspergillus glaucus</i>	Polyethylene	23.11% weight Loss	(Sangale et al., 2019)
Fungi	<i>Humicola insolens</i> <i>Cutinace (Hic)</i>	Low crystallinity polyethylene terephthalate (PET) films	97% weight loss	(Srikanth et al., 2022)
Fungi	<i>Rhizopus Delemer</i>	Polyester type polyurethane	53% of degradation	(Srikanth et al., 2022)
Fungi	<i>Aspergillus fumigatus</i>	Polyhydroxy butyrate (PHB), Poly (butyrate succinate (PBS), Polyethylene Succinate (PES)	95% weight loss	(Kaushal et al., 2021)
Fungi	<i>Phanerocheate chrysosporium</i>	Polyvinyl chloride (PVC) films	31% weight loss	(Temporiti et al., 2022)

Table 8. The comparison study of enzyme utilization for MPs degradation

Enzymes	Organisms	Types of Plastic	Reaction parameters	MPs degradation rate (%)	References
Laccases	<i>Rhodococcus ruber</i> C208	LDPE film	Incubation for 30 min at 37°C and pH 7.0 along with the addition of copper	20	(Santo et al., 2013)
	<i>Pleurotus ostreatus</i>	LDPE	Incubation for 30 Days	27	(Gómez-Méndez et al., 2018)
	<i>Aspergillus flavus</i> PEDX ₃	Polyethylene	Incubation for 28 days	3.9	(J. Zhang et al., 2020)
	<i>Trichoderma harzianum</i>	PE films	Incubation for 15 min at 30°C	40	(Temporiti et al., 2022)

Enzymes	Organisms	Types of Plastic	Reaction parameters	MPs degradation rate (%)	References
Esterase	<i>Pseudomonas aeruginosa</i>	Polyester PUR film	Incubation for 12 days	2	(Shah et al., 2016)
Proteus	<i>Impranil DLN</i>	Incubation for 51 days at 37 °C	Incubation for 51 days at 37°C	33	(Venkatesh et al., 2021)
Alkane monooxygenase B1	<i>Pseudomonas aeruginosa E7</i>	Low molecular weight Polyethylene (LMWPE)	Incubation in LB broth for 50 days at 37°C in a shaker incubator at 120 rpm	14.4	(Jeon and Kim, 2016)
Hydroquinone peroxidase	<i>Azotobacter beijerinckii HM121</i>	PS Film	Incubation of medium containing hydrogen peroxide at 30°C for 10 min	77	(Ru et al., 2020)
Peroxidase	<i>Trichoderma harzianum</i>	PE film	Incubation at 30°C for 15 min	0.6	(Temporiti et al., 2022)
Cutinase HiC	<i>Humicola insolens</i>	PET	Incubation at 70°C for 96 h	97	(Taniguchi et al., 2019)
Cutinase FoCut5a	<i>Fusarium oxysporum</i>	PET	Incubation at 40°C, pH 6.0 for 10 min	6	(Temporiti et al., 2022)

4.4. Comparison of Physical, Physicochemical, and Biological Methods

The various kinds of technology used to remove MPs were further compared in Table 9. The basic of filtration technology has simple operation and high removal rate of large-size MPs. However, the weakness of filtration was obstacle to remove small MPs, thus advanced filtration technology was required to enhance their performance. Meanwhile, the approach of adsorption is a simple method for removing microplastics (MPs) smaller than 20 μm . Eventhough, the high removal rate of adsorption method, the potential toxicity of additive sorbent may cause secondary pollution in aquatic environments. There is a similar problem of chemical approaches in terms of coagulation and electrocoagulation treatment. Thus, the natural potential of the biological method has been another promising method for MPs removal. Although the biological process take long time to degrade MPs, biological method offer less toxicity to environment. After a comparison of varied methods for MPs removal, the membrane filtration provided efficient removal of MPs, both low and large-size MPs were retained by the membrane. Further, no addition of chemicals is used in membrane filtration. However, the possibility of a combination of multiple technologies is considered to enhance the efficiency of removing MPs in aquatic environments.

Table 9. Comparison of MPs removal methods among physical, physicochemical, and biological methods.

Category	Removal method	Advantage	Disadvantage	Application
Physical (Alrbaihat and Abu-Afifeh, 2023; Osman et al., 2023)	Filtering	Easy operation, no chemical treatment, and great efficiency	Efficiency is limited to MP >20 µm, requires regular cleaning and maintenance, and can damage the filter	Aquatic environments such as water treatment plants
	Density separation	Simple operation, no chemical treatment	Saline water types need to be adjusted, vulnerable to M/NP interference	Operation in static liquid environment M/NP
	Power spinning filtration	High removal efficiency, low energy consumption, high selectivity, mechanical strength, hydrophilicity	Complex synthesis process, possible fouling of filters, clogging of pores	Liquid environment, not yet widely used in practice
	Superhydrophobic materials	With high removal efficiency, organic solvents can be removed at the same time	Additional chemicals are required to achieve superhydrophobicity and separation and transfer of M/NP to the organic phase	Liquid environment, not yet widely used in practice
Physicochemical (Karapanagioti and Kalavrouziotis, 2019)	Coagulation, flocculation, sedimentation	Fast process, controlled operating conditions, simple mechanical equipment, suitable for removing small-sized M/NP, precipitated floc can be removed easily	Heterogeneous removal efficiencies, not suitable for large-sized M/NP; usage of too much coagulant may harm the environmental matrix	Water Environment

Category	Removal method	Advantage	Disadvantage	Application
	Electrocoagulation	Precipitated flocs may be easily removed, possess minimal conductivity requirements, no risk of secondary contamination, appropriate for removing tiny size M/NP, energy efficiency, and low cost	Continuous anode replacement and cathode passivation need sufficient current density to avoid high energy consumption and are not accessible in non-electrical settings	Conductive water environment
	Adsorption and magnetization	Simple and fast process, high removal efficiency, adsorbent can be modified and recycled	The results rely on the materials employed, desorption possibilities, the required adsorbent synthesis, and magnetized materials must be superparamagnetic	Water environment
	Aggregation of microorganisms	Easy to remove, M/NP can be released during recovery	Highly dependent on the microorganisms used, and has low efficiency	Water environment
	Microbial degradation	Simple and safe operation, low cost, widely applicable, safe by-products	Difficult to control environmental conditions, less producibility, suitable microbial groups required, very slow process, low filtration efficiency after tens of days of degradation	Aquatic environment and soil environment

5. MPs Removal through Membrane Technology

5.1. Conventional Membrane Technology

Membrane filtration is a widely used as wastewater treatment process due to its accessibility, high rejection capacity, and low cost retrofit ability. The membrane technology demonstrated > 90% of MPs removal, as listed in Table 10. The main factor during MPs removal with membrane are adsorption and sieving. The sieving mechanisms was applied by hydraulic membrane pressure (microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO)). In principle, the each membrane separation process has varied separation mechanisms in MPs removal, thus the following sections explain each of them in detail. Meanwhile, the illustration of MPs filtration using membrane was shown in Fig. 7. The absence of MPs in permeate solution was demonstrated after MPs in feed water was filtered by membrane.

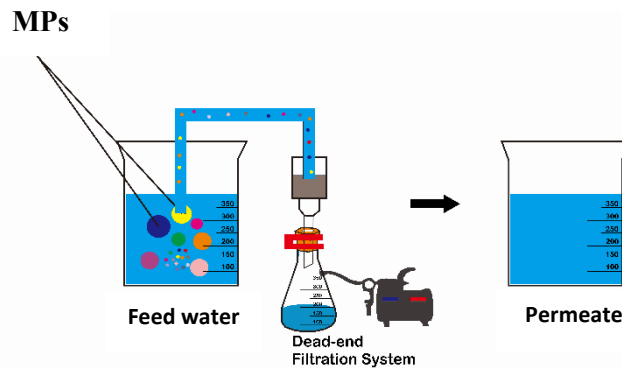


Figure 7. The sequence uses membranes in a lab scale.

5.1.1. MF and UF

MF and UF membranes are utilized at low-pressure range (0.1-10 bar). The largest pore size in the membrane process is MF, followed by UF. The membrane pore in MF and UF facilitate the circulation of different substances, the mechanism is known as size exclusion process (Gonzalez-Camejo et al., 2023). On the other hand, the adsorption process is also possible demonstrated during water purification and wastewater treatment by MF and UF membrane (Ma et al., 2019b). Thus, the MF/UF membranes provide a variety size for MPs removal.

Several organic polymers, including polyvinylidene fluoride (PVDF), cellulose acetate (CA), polysulfone (PSU), and polyethersulfone (PES), are frequently used for membrane fabrication. MF membranes effectively remove MPs, with rates ranging from over 90% to 100% (Ramos et al., 2023). Meanwhile, the UF membranes demonstrated higher MPs removal than MF membrane, with values over 95% (Takeuchi et al., 2023). Even so, the utilization of membrane potential is used as a secondary treatment. For example, a previous study observed the anionic polyacrylamide and iron coagulation as pretreatment in the UF filtration process. Thus, the result showed that UF membranes could remove 91% of polyethylene (PE) as the main commodity of plastic (Pramanik et al., 2021). High removal of 92% of MPs in wastewater was presented by UF membrane after pretreatment with alum coagulant (Zhang et al., 2023). The MPs removals in surface and groundwater using UF process within range from 72-86% (Yang et al., 2023).

Beside of the concentration feed and particle size, the main factor in membrane properties is very depend on the performance filtration of MPs removal. Nowadays, the researchers attention on MF or UF modification with metal organic frameworks (MOF) for MPs removal. MOF could modify functional groups in membrane structure to become more hydrophilic, resulting improved membrane performance in MPs removal. For instance, the modified PSF membrane using MIL-100(Fe) could retained 99% of MPs including of polyethylene (PE) and polyvinyl chloride (PVC) through electrostatic interaction and sieving mechanisms (Gnanasekaran et al., 2021). Similar mechanisms was also showed in modified membrane

using Ni-MOF/nyloNi-MOF/nylon effectively remove 99% MPs of polysulfone (PS) (Han et al., 2023). Thus, it is essential to study interaction during MPs removal mechanism to improve membrane performance.

5.1.2. NF

NF membrane process with pore size and separation capabilities between UF and RO. Until now, only limited research focused in MPs removal using NF membranes. The configuration NF after membrane bioreactor (MBR) has successfully removed MPs of 99.83% (J. Sun et al., 2021). The result is not surprisingly that NF demonstrated high removal against MPs, the majority size of MPs within range between 20-50 μm . Meanwhile, the NF membrane has size 0.1-10 nm (Van der Bruggen, 2009). Thus, the MPs were easily retained by NF membrane. This is also proven by a previous study. The result mentioned that no MPs observed in six varied of NF effluent samples. NF is considerably more effective than traditional water treatment technologies (Barbier et al., 2022). However, the depth investigation of how effective is utilization of NF membranes to separate (MPs) are still required.

5.1.3. RO

RO membranes require pressure above osmotic pressure (10-30 bar for freshwater, brackish water, and wastewater) to remove particles smaller than 0.001 μm , including salts, organic contaminants, ions, viruses, bacteria, and colloids (Acarer, 2023). RO membrane generates high-quality waste water for a variety of applications, including water recycling in industry and the separation of chemicals from waste byproducts (Dang et al., 2016). However, the utilization of RO consumes more energy and is susceptible to fouling. A study investigated the removal of MPs from wastewater through a comprehensive strategy consisting of screening (mesh size of 3 mm), sedimentation, biological treatment, flocculation, disinfection/de-chlorination, UF, RO, and decarbonation. The effluent contains 0.21 MP/L (42% PE, 36% PET, 15% PS, and 8% PP) in the size range of 100-190 μm . Even though the RO removed 90.45% of the MP, the permeate samples still remaining contained fibers and irregular forms (Ziajahromi et al., 2017). Eventually, the presence of MPs in permeate after treatment comes from the release of polymeric components in the membrane or the presence of MPs in the surrounding air. As a result, further confirmation is required.

6. Conclusion

In this review, we thoroughly presented, described, and examined numerous technologies, including filtering, coagulation, electrocoagulation, and biological methods in comparison with membrane filtration with their potential for MPs removal. Meanwhile, the ways to identify the absence of MPs in water was also discussed. Nowadays, MPs from plastic waste are expected to reach more than 12,000 million metric tons in 2050. The numerous amounts of MPs in water bodies could be assessed by a variety of methods, including gas chromatography-mass spectrometry (GC-MS), liquid chromatography (LC), scanning electron microscopy (SEM) combined with energy dispersive X-ray spectrophotometry (EDX), surface-enhanced Raman spectroscopy (SERS), dynamic light scattering (DLS), and Fourier transform infrared spectroscopy (FTIR).

This review study of recent technology for MPs removal. The physical filtration technology is the simplest way in comparison with other methods. However, the removal ability of physical filtration against smaller MPs than 20 μm becomes a crucial concern. Then, the other option is an adsorption method. Although the adsorption option is an inexpensive method, the undesirable aspect during adsorbent usage may not be environmentally friendly in aquatic systems. The similar problem is also demonstrated by chemical approaches in terms of coagulation and electrocoagulation treatment. Consequently, the biological methods were found to be less toxic to the environment. Even though it provides safe conditions to the environment, the biological approach needs a long time to degrade MPs. The removal by physical, physiochemical, and biological methods was generally 40-90%, 40-95%, and 1-60%, respectively. The highest of 90-99% MPs removal performance was demonstrated by conventional

membrane technology among all MPs removal methods. Thus, the membrane filtration is one of the promising technologies for MPs removal in aquatic environments providing efficient removal of MPs. Although, the significant possibility of combining several technologies is particularly considered to improve the efficiency of MP removal in aquatic environments.

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