

Possibilities of removing microplastics from the aquatic environment using membrane processes

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ABSTRACT

According to the literature, a lot of microplastics and nanoplastics enter the aquatic environment each year. Various physical, chemical and biological treatment processes can be used to the removal of microplastics, among them membrane processes can play an important role. Pressure-driven membrane techniques, that is, micro- ultra-, nanofiltration and reverse osmosis, can be used in the context of micro- and nanoplastic removal as a third step in integrated wastewater treatment systems. The most effective solution in this regard are membrane bioreactors (MBRs), which combine the process of biological wastewater treatment with membrane separation. MBRs can increase the removal rate of microplastics and nanoplastics from primary wastewater to 99.9%, especially in different sizes and shapes, which is significantly more compared to other advanced treatment methods. Microplastics are being detected in drinking water, increasing concerns about the effectiveness of water treatment plants. The ultrafiltration process combined with coagulation/ flocculation can be one of the main technologies for removing not only organic contaminants but also microplastics in current water treatment plants. Significant progress has been made in the removal of microplastics and nanoplastics using membrane processes, but further progress is needed to minimize fouling, extension MBR efficiency, and scale-up issues in implementing membrane processes into industrial practice.

Keywords: Micro- and nanoplastics removal; Water and wastewater treatment; Membrane processes; Membrane bioreactors

1. Introduction

Plastics production has been growing worldwide on a large scale since 1950. In 2014 it was 299 million tons, in 2016 335 million tons, in 2017 348 million tons, in 2018 359 million tons, and in 2019 367 million tons [1,2]. Due to the COVID-19 pandemic, global plastics production in 2020 recorded a 0.3% decrease compared to 2019, but at the same time, other plastic wastes appeared in large quantities, such as single-use masks, disposable gloves, and other protective equipment [3]. Most plastics are produced in Asia (about 50%), followed by Europe (>18%), North America (about 18%), the Middle East, Africa (7.7%), South America (4%) and the Commonwealth of Independent States (2.6%). Plastic production in Europe reached a record high of more than 62 million tons of manufactured goods in 2018. The largest demand for such plastics is in the packaging market, as well as in building materials and automotive. This significant growth and spread of plastic production around the world is generating a huge amount of plastic waste, which penetrate the surface and underground water [4]. Microplastics (MPs) (size range of <5 mm) are therefore an urgent issue to solution due to their negative impacts on the environment and human health [5,6]. Plastics enter the environment with municipal and industrial wastewater, surface runoff and by breaking down larger plastics into

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smaller fragments [7,8]. MPs can be found in practically all water sources. In natural waters, it is mainly found: polypropylene (PP), polyethylene (PE), polystyrene (PS), polyvinyl-chloride (PVC), polycarbonate (PC), polyamides (PA), polyester (PES), polyurethane (PU), cellulose acetate (CA) and polyethylene terephthalate (PET) [9].

Potential MPs toxicity to humans from unreacted monomers and chemical additives includes respiratory irritation, obesity, cardiovascular disease, asthma and cancer [10,11]. When the concentration of MPs and plastic additives reaches the limit values, they can be penetrated into the body through various routes [12]. This is because MPs can become heavily contaminated with organic compounds due to their large specific surface area and intensive ability to bind hydrophobic organics on the surface [13–15]. Type of plastics and such properties as ionic strength and pH can impact the MPs adsorption efficiency [16].

The removal of MPs and nanoplastics (NPs) from the water and wastewater is a big challenge for scientists and water and wastewater technology specialists. To date, few publications have appeared on the use of membrane processes to remove MPs from the aquatic environment. This article demonstrates the applicability of membrane processes in the removal of MPs. Membrane technology makes it possible to manipulate membrane properties by selecting pore size and distribution, as well as the type of membrane-forming material, which will determine the membrane's mechanical and thermal resistance. We think that membrane technology will be useful not only in the removal of MPs, but also in their reuse.

2. Microplastics source and occurrence

Recently, MPs have been found in lakes and rivers, and thanks to river and wind transport, plastic trash is reaching the coasts and oceans, and can even be found in the Arctic Sea [17–19]. Now, wastewater treatment plants (WWTPs) are considered one of the main responsible for plastic pollution in the environment, as 98% of MPs are retained in them, with the remaining 2% being particles smaller than 20 μ m [7,20–23].

Firstly, fibres lost from textiles during washing [24] and plastic beads used for exfoliation or purification in cosmetics and personal care products enter WWTPs through domestic discharge systems. The use of detergent appeared to affect the total mass of fibres released to the environment, yet the detergent type or overdosing of detergent did not significantly influence MPs release. Despite different release quantities, the overall microplastic fibre length profile remains similar regardless of wash condition or fabric structure, with the vast majority of fibres ranging between 100 and 800 μ m in length irrespective of programme selected on the washing machine [24]. This indicates that the fibre staple length and/or debris encapsulated inside the fabric from the yarn spinning could be directly responsible for releasing stray fibres.

Secondly, industrial plastics used in surface blasting, moulding and many other processes are discharged into municipal wastewater collection systems before entering WWTPs [25–27].

The third factor responsible for MPs contamination in WWTPs is the wet sedimentation process. Fine plastic debris found in the atmosphere or in concrete and highway structures that results from the breakdown or abrasion of other plastics, such as packaging, textiles and tyres, can enter wastewater through stormwater runoff [26-29]. Car tyres release wear particles through mechanical abrasion. Wear and tear from tyres significantly contributes to the flow of (micro-)plastics into the environment. The estimated per capita emission ranges from 0.23 to 4.7 kg/y, with a global average of 0.81 kg/y. The emissions from car tyres (100%) are substantially higher than those of other sources of microplastics, for example, airplane tyres (2%), artificial turf (12%–50%), brake wear (8%) and road markings (5%). Emissions and pathways depend on local factors like road type or sewage systems. The relative contribution of tyre wear and tear to the total global amount of plastics ending up in our environment is estimated to be 5%-10%.

Finally, WWTPs can receive MPs from landfill leachate, where due to harsh environmental conditions, landfilled plastic waste is fragmented into MPs, which are then transferred with leachate discharge to enter WWTPs [30]. He et al. [31] investigated twelve leachate samples from four active and two closed municipal solid waste landfills. MPs were found in all the landfill leachate samples. In total, seventeen different types of plastics were identified in the leachate samples with calculated concentration ranging from 0.42 to 24.58 items/L. Polyethylene and polypropylene were the predominant polymer types. 99.36% of MPs were derived from the fragmentation of plastic waste buried in landfills. The size of 77.48% of the microplastics was between 100 and 1,000 µm. The study shows that the generation, accumulation and release of MPs in landfills is a long-term process.

Along with wastewater, MPs can enter the environment via sewage sludge. Sewage sludge can contain from 20 to more than 180 particles of MPs per gram of dried sludge, depending on sludge management and testing methods [32,33]. Due to their relatively high phosphorus and nitrogen content, in many countries sludge is applied to agricultural land or used in landscaping [34]. According to Horton et al. [35], the amount of MPs in terrestrial environments can be 4 to 23 times higher than in the oceans. In addition, airborne MPs that have been emitted by the plastics industry and vehicles also enter WWTPs [29] WWTPs are therefore considered the main recipients of terrestrial MPs before they enter natural aquatic systems [36,37]. It has been proven that untreated MPs are commonly discharged from WWTPs, enter water bodies, and eventually accumulate in the environment [20,38-40].

Surface water, (i.e., rivers, lakes and reservoirs), and groundwater are the main sources for drinking water production, however, freshwater sources are limited. Therefore, seawater is also sometimes used for this purpose, however, desalination of seawater involves high costs and high energy consumption [41]. Natural waters are often polluted by agriculture and industry, and wastewater from industrial animal husbandry and that is why MPs have been detected in natural surface waters [42]. The average abundance of MPs in these waters ranges from several to millions of tons [43]. Such large differences are primarily due to location, natural conditions, human activity, etc.

3. Classification of microplastics

Considering the size of the particles, they are divided into macroplastics (>25 mm), mesoplastics (5-15 mm), microplastics (<5 mm), and nanoplastics (<100 nm) [36]. MPs have diameters less than 5 mm, which makes them resistant to (bio)degradation [44]. According to IUPAC (International Union for Pure and Applied Chemistry) as MPs are considered particles of dimensions 0.1-100 µm [45]. However, in recent years it has been accepted to define MPs as plastic fragments whose longest dimension is less than 5 mm. This is also the definition used by the European Commission, the EPA (U.S. Environmental Protection Agency) and Asia-Pacific countries [23]. On the other hand, NPs are defined as particles (nanospheres, nanofibers, nanotubes and nanofilms) with dimensions from 1 to 100 nm [32,46,47]. It is important insofar as particles below this diameter, unlike MPs, may be capable of damaging the cell membrane [48].

Taking into account the origin, a distinction is made between also primary and secondary MPs and NPs [49]. Primary MPs are small pieces of specially manufactured plastics, such as hand and face wash, shower gels, toothpaste, peelings, eye shadows, deodorants, blush powders, makeup foundation, mascara, shaving cream, baby products, bubble bath lotions, hair colouring, nail polish, insect repellents and sunscreens, as well as plastic pellets and vectors for drugs [2,32]. Table 1 presents examples of primary MPs fabricated in Europe in 2017 [50].

Table 1 shows that in Europe, the most significant products wrapping primary MPs are personal care products, and the main polymer produced as MPs is polyurethane. In 2017, primary MPs containing polyurethane accounted for nearly 50% of total MPs production in the cosmetics and personal care market, as well as detergents.

Secondary MPs are formed during the decomposition of larger waste plastics through physical, biological and chemical processes, both at sea and on land [32]. Polymer degradation can be divided into biodegradation, photodegradation, thermo-oxidative degradation and hydrolysis [51]. The process of maximally reducing the molecular weight of a polymer is called plastic degradation, which results in pieces of plastic becoming brittle [23]. UV solar radiation is the most efficient degradation to plastic waste especially exposed to air [51].

It is important to mention that even though there are many degradation processes that can occur in the water and coast environment, however, common plastics used in everyday applications do not biodegrade at a fast enough velocity that could be beneficial. As a general rule, plastics commonly used in everyday use do not biodegrade fast enough, because microorganisms capable of metabolizing polymers are rarely found in the aquatic environment. However, there are biodegradable biopolymers, such as natural chitin and chitosan, and a few synthetic polymers, such as aliphatic polyesters [23].

4. Removal of microplastics from the aquatic environment

Most water bodies have been contaminated with MPs, but it is difficult to remove them from the aquatic environment. MPs have been detected in lakes, rivers, oceans, and municipal wastewater, so their removal from the aquatic environment is a new and urgent challenge, given their negative effects on humans and aquatic flora and fauna [52-56]. None of the current water and wastewater treatment technologies are designed to remove plastic particles because they have been developed to remove and degrade dissolved and suspended pollutants, and solid waste [57,58]. To control the level of MPs in water, it is necessary to reduce their transport chain. In general, two systematic approaches should be considered in the fight against MPs contamination of water. The first is to stop or reduce the introduction of MPs into natural waters, while the second is to remove MPs from water. Generally, water and wastewater treatment technologies for MPs removal are commonly based on physical, chemical and biological treatments (Fig. 1) [36].



Fig. 1. Basic classification of MPs removal methods, own compilation based on [36].

Table 1

Primary microplastics fabricated in Europe in 2017, amount of their production and type of polymers, compiled on the basis of Scudo et al. [50]

Product	Production (T/y)	Type of polymers
Personal care products	714	PU, PE, CA, polylactic acid, Nylon 11
Detergents	142	PU, PES, PA, acrylic polymers, poly(methyl methacrylate), PET, PE
Paints and coatings	220	Acrylic polymers, fibers of PA, polyacrylonitrile

Based on the available literature, physical methods are studied more than chemical and biological ones. Physical methods are mainly filtration, adsorption, sedimentation, flotation and others. Most of them have been tested in the laboratory and on a pilot scale, and some have even been implemented on a full scale. Among various physical methods with high MPs removal efficiency include: carbon based adsorbents like biochar, magnetic carbon nanotubes, magnetic polyoxometalate-based ionic liquid phase adsorbents, disc filters, rapid sand filter and dissolved air flotation, magnetic separation processes, and others [36,59,60]. Studies have shown that filtration methods have higher MPs removal efficiency than others. The order of MPs removal by physical methods can be ranked as follows: filtration > flotation > adsorption > membrane techniques > magnetic and density separation [61].

Chemical methods are also used in the treatment of water and wastewater containing MPs, either alone, or in combination with, others to enhance the effectiveness of physical processes (e.g., sedimentation, membrane processes). Several methods, like ozonation, advanced oxidation processes, coagulation and electrocoagulation, Fenton processes and photocatalysis are most commonly used for plastic removal/degradation [62]. The efficiency of MPs removal by chemical methods can be ranked as follows: photo-Fenton process > electrocoagulation > ozonation > electro-Fenton process > coagulation > modified Fenton process [62]. Unfortunately, no single removal method is capable of removing MPs from wastewater when used alone. Therefore, hybrid methods should be used, combining chemical methods with other physical or biological methods. Often chemical methods, such as coagulation and electrocoagulation, produce by-products as well as secondary sludge that require further treatment.

Physical and chemical treatment methods can be applied to remove a wide range of MPs from water, with their average removal efficiencies summarised in Table 2. The wide range of MPs removal rates is due to the different process conditions and the different sizes of particles removed.

Biological methods use organisms to degrade and remove MPs present in the environment. A number of organisms have been studied for their ability to degrade MPs in water and wastewater. The greatest ability to degrade MPs is demonstrated primarily by microorganisms [81]. Biological methods of removing MPs have been used primarily to treat wastewater, both municipal and industrial. Wastewater treatment can be divided into three main groups: pretreatment, biological (second-stage) treatment, and third stage treatment, also known as final treatment, applied in the case of their reuse [22] (Fig. 2). At this last stage of purification, membranes have great potential [1,23].

During pretreatment, large-diameter suspended solids are removed, but effluent still contains a significant concentration of suspended solids, and the removal rate of MPs is about 25%. During biological treatment (second-stage), despite the higher efficiency, the concentration of MPs can be reduced by 75% [9]. In second-stage (biological) treatment, aerobic or anaerobic methods are used to remove biodegradable organics. In addition, it uses an alternating system anaerobic, anoxic & oxic (A²O) for biological nutrient removal. Activated sludge (AS) and biological beds (BF) (effluent filters/biofilters), membrane bioreactors (MBR), and hydrotreatment plants (constructed wetlands) are most commonly and widely used technologies for secondary treatment of wastewater and most effective methods for MPs removal [9]. During tertiary treatment, the efficiency of MPs removal is of the order of 98%, which makes it possible to obtain water of a quality similar to drinking water [11–14]. The fact of infrequent use of tertiary treatment in wastewater treatment plants is the source of large amounts of MPs in treated wastewater and sludge. The use of advanced wastewater treatment technologies in this stage is therefore a necessity. As 3-stage treatment processes, membrane techniques can effectively solve the contamination of MPs and NPs in the environment. A comparative overview of the biological methods, their advantages and drawbacks are summarised in Table 3. The removal of MPs via biological methods decreased in the order: MBR > CWs > activated sludge > microbe processes. The MBR process and CWs have potential in leading biological methods of MPs removal.

5. Membrane technology

Membrane technology can be used to remove MPs from the aquatic environment, as membrane techniques are energy efficient and most have been implemented on a technical scale. Membranes, moreover, are characterized by simplicity of operation, chemical and thermal stability, and, as in seawater and brackish water desalination, enable treatment of large volumes of water [23].

Pressure-driven membrane processes, that is, microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), are the most widely used in water and wastewater technology [41,59]. Table 4 presents the fundamental properties of membrane techniques. Other membrane techniques, as forward osmosis (FO), membrane distillation (MD) and processes used ion exchange membranes (electrodialysis, electro-deionization) are also considered [59]. Membrane properties, that is, its capacity and separation properties, depend on the material from which the membrane is made, as well as on the porosity (pore size and proportion of pores in the membrane volume) and surface roughness of the membrane.

According to Enfrin et al. [60] MPs interact with the membrane surface due to hydrophobicity, charge and surface roughness. Membranes are highly effective in removing MPs < 100 µm in diameter and NPs. Various researchers investigated the MPs removal using the wastewater treatment system as a third stage of purification. The membrane then provides a physical barrier to MPs and NPs, because the diameter of MPs is large or similar size as that of membrane pores, which enable completely removal MPs from waters [60,92]. Membranes can remove MPs from the water with higher efficiency and stable permeate quality. Comparing the treatment efficiency of membrane techniques as a third stage of wastewater treatment with rapid sand filter, disk filter and dissolved air flotation, the MPs removal efficiency for MBR is 99.9% after the first stage of treatment, while the classical processes achieve 97%, 98.5% and 95%, respectively [9,33].

Table 2

Advantages,	limitations	and effectivenes	s of physical a	nd chemical	treatment	technologies	in MPs removal,	own c	compilation	based
on [7,9,21,63-	-80]									

Process type	Efficiency	Advantage	Disadvantage
Adsorption (activated and biochar carbon)	100%	Sufficient surface area and suitable porosity effectively retained large size MPs	10 μm spherical MPs did not absorb as efficiently
Rapid sand filtration	97.2%	Low operational and maintenance cost	Fouling take place; backwash is needed. MPs are broken into smaller particles
Disc filters	98.5%	Sludge cake formation Float MPs are especially removed	Backwash needed due to membrane fouling
Ultrafiltration	41.7%	PE particles can be completely bound by the UF membrane	Fouling
Dynamic membranes	>90%	Low energy consumption and trans-membrane pressure, low filtration resistance, low cost	Membrane fouling Not effective for large scale water treatment
GAC filtration	99.9%	Remove small size MPs with biological activity	Clogging is the main problem
Sedimentation	78.34%	Low-cost process Effective for large MPs	Need to secondary and tertiary treatment to remove small MPs
Flotation	95%	-	Removes contaminants by trapping low-den- sity MPs PE, PP), and the medium-density plastics (PS, and PA)
Magnetic separation	78%–93%	Efficient for smaller MPs Better for drinking water treatment	MPs recovery from sediment is lower
Ozonisation	89.9%	Efficient tertiary treatment	Difficulties in ozone production High operational cost
Modified Fenton	25.49%	Reagents availability	Lower efficacy
process		Cost-effective process	Applied for specific type MPs Optimal and cost-efficient reagents
Electro-Fenton pro- cess	75%	Eco-friendly process Highly efficient method	Excessive cost requirements Required modifications
		Lower reagent costs	Required investigation for application on different MPs
Photo-Fenton process	>99%	Highly efficient method No requirements of excessive catalysts or reagents	pH should be maintained in an optimum level More investigation is required for practical uses
Photocatalytic	Possible complete	No additional chemicals	Lower efficacy
degradation	mineralization	Eco-friendly process Efficient mineralization of particles with the help of solar energy	Generation by-oduts High energy-consuming process Require photo-reactor and difficult to recover
Coagulation	61%	Controllable operational conditions Adequate for small MPs	Inappropriate for large-sized MPs Uses of additional chemicals
Electrocoagulation	>90%	Minimal sludge Adequate for smallest MPs Cost-effective No secondary contaminants	Sacrificial anodes are required to be replaced repeatedly Cathode passivation is observed Electricity is required

MPs removal depends on their shape, size, mass and polymer type. The separation mechanisms of membrane processes, which describe the removal of contaminants from water, are mainly sieve mechanism, hydrophobic and electrostatic interactions. The removal of MPs from water and wastewater can therefore be influenced by both the characteristics of MPs (size, shape and polymer type) and membranes (structure, pore size and membrane material) [93]. An important parameter is the shape of MPs, which determines the effectiveness of their removal by membrane methods and interactions with other contaminants, including microorganisms [94]. There are the following shapes of

108

Preliminary and primary treatment									
 Sedime degasif Chlorir 	 Sedimentation, screening, aeration, filtration (above 1µm), flotation and skimming, degasification. Chlorination, ozonation, neutralization, coagulation adsorption, ion exchange. 								
	Secondary treatment								
 Aerobic processes: activated sludge process, trickling filtration, oxidation ponds, lagoons, aerobic digestion. Anaerobic processes: anaerobic digestion, septic tanks, lagoons. 									
		Tertiary trea	atment						
Disinfect dosing.	tion, oxidation, softenin Microfiltration - MF Ultrafiltration - UF Nanofiltration - NF Reverse osmosis- RO	g, membrane Dynamic membranes (DM)	Forward osmosis (FO)	n exchange, chemical Membrane bioreactor (MBR)					

Fig. 2. Stages of wastewater treatment, own compilation based on [9,20,21].

Table 3

Efficiency and types of MPs removed in biological wastewater treatment process, own compilation based on [7,9,15,32,33,49,64, 67,68,82–90]

Treatment process	Efficiency (%)	Type of microplastic
		removed
Submerged MBR (KUBOTA)	100.0	_
Submerged MBR	100.0	_
MBR	99.9	20–100 μm MPs
MBR	99.4%	PES, PE, PA and PP
MBR	99	PVC fragments, fibres
BAF biological aerated filter	99	PE 100–300 μm
AS	98.3%	Various types
MBR	97.6	PES fibres and PE fragments
OD oxidation ditch	97	Fragments, fibres
AS USA	95.9	SAL
ASP	93.8	Microbeads
A ² O	93.7	PE, PP, PE and acrylic fibres
AS and clarification	92	Fragments, fibres
TF and AS	89.8	Microbeads
MBR, AS, and settling tank	83.1–91.9	Fragments
MBR	79.01	Fibres, PP, PS
AS South Korea	75–91.9	Primary and secondary MPs
A/A/O	71.67 ± 11.58	Not mentioned
AS	66.7	Polystyrene
AS, sedimentation	64	Fibres
A ² O	54.4	_
AS Slovenia	52	PE < 100 μm
A ² O	28.1	PET, PE, PES, PAN, PAA
Anoxic tank, aeration basin, clarifier	2.4	Smaller microplastics

MPs: fibers, pellets, fragments, films and foams. The most abundant form of MPs in wastewater is fiber, which comes from domestic and industrial washing of fabrics containing synthetic polymers [95]. Cai et al. [96] showed that membranes made of different materials, and with different structures and pore sizes, also differed in MPs removal efficiency. They showed that fibers of 3568 μm were retained membranes with a pore size of 1,000 μm , while MPs

Membrane techniques	Cut off, kDa	Transmembrane pressure (bar)	Membrane structure	Average permeability (L/m²·bar)	Compounds retained
MF	100–500	1–3	Porous symmetric	500	Bacteria, colloids, organics with cut-off >100 kDa
UF	20–150	2–5	Porous asymmetric	150	Bacteria, viruses, organics cut-off 50–100 kDa, MPs
NF	2–20	5–15	Thin film composite	10–20	divalent cations and anions, micropollutants
RO	0.2–2	15–75	Thin film composite	5–10	All compounds including monovalent ions

Table 4	4												
Main 1	prop	perties	of	pressure-	driven	membrane	processes,	own	com	pilation	based	on	[44,91]

fragments of 37.2 μ m were found on membranes with pores of 50 μ m. MPs in the form of fibers can pass through membranes more easily than other shapes of MPs, even when the fiber dimensions are smaller than the membrane pore size. In a study by Michielssen et al. [77], fibers accounted for ~80% of MPs in wastewater from a three-stage treatment consisting of sand filtration and MBR, while in wastewater from an activated sludge process the MPs content was only <44%.

Fouling is a major problem in membrane filtration, due to the deposition of particles on the surface and in the membrane pores. During wastewater treatment, colloids and macromolecular compounds, including MPs, interact with the membrane and deposit on its surface or pores. As a result, the membrane pores narrow, causing fouling [32]. Fouling decreases membrane filtration performance which resulted in higher energy cost, operation time and maintenance [23]. Shear stress during membrane filtration causes fragmentation of MPs into NPs, which are easily deposited on the membrane surface. On the other hand microorganisms are settled on the surface of MPs through biofilm formation, causing biofouling of the membrane [60,97].

Comparison of literature data on MPs and NPs removal efficiency is difficult due to the different composition of wastewater in terms of particulate size and the lack of uniformity in removal studies plastics [47].

5.1. Ultrafiltration and microfiltration

Low-pressure membrane processes (i.e., MF and UF) are used in the treatment of drinking water, thanks to the low energy consumption and transmembrane pressure 1-10 bar, high efficiency and compactness of the equipment [41,59,98]. Porous MF/UF membranes (symmetrical or asymmetrical), with a pore size of 0.05-10 µm for MF and 1-100 nm for UF, can remove suspended solids and macromolecules (e.g., proteins), and bacteria, viruses and protozoa from wastewater or natural waters. Thus, many drinking water treatment plants using MF/UF in the purification process protect treated water from contamination with Cryptosporidium, Giardia and other microorganisms [41,61]. That is why, MF/UF can replace classical processes, that is, coagulation and sedimentation, classical filtration and chlorination used in plant of water and wastewater treatment. In particular, MF/UF can enable the reuse

of water from industrial plants that consume and discharge large amounts of water (chemical, steel, plastics, paper, pharmaceutical, food, etc. industries) [98]. Water and wastewater treatment plants use membranes made of polymers and ceramics. The latter are characterized by greater chemical and thermal stability and can be operated much longer than membranes made of polymers [99].

As already mentioned, fouling is one of the biggest problems in the operation of MF and UF membranes, due to their hydrophobicity. Controlling fouling should ensure long-term stable operation of MF and UF membranes [100]. Fouling involves surface fouling of the membrane due to concentration polarization [78] and deposition of contaminants, including MPs, which leads to the formation of a cake layer [92]. Methods to reduce fouling usually include methods to improve the hydrophilicity of membranes by modifying hydrophobic polymers before membrane fabrication, and grafting or coating the membrane surface with hydrophilic polymers. In addition, membranes are also subjected to hydraulic and chemical cleaning after various period of operation, depending on membrane and raw stream properties [101]. Nevertheless, after a certain period of time, the membranes must be replaced, due to a reduction in pore size or even blockage [99].

Another effective method of reducing fouling is pretreatment of raw water or wastewater, and thus the effect of MPs on membranes by using for example coagulationflocculation-sedimentation (CFS). Higher concentrations and smaller dimensions of MPs lead to greater fouling of UF/ MF membranes, thus pretreatment is necessary [79,102,103]. Li et al [82] investigating the effect of PE particle size on membrane fouling found that 1-µm MPs caused the greatest fouling than 0.1, 1, 10 and 18 µm MPs, suggesting a critical PE particle diameter of 1 µm. MPs increase also membrane biofouling due to production of extracellular polymeric substances (EPS), which underwent MPs-stimulated accumulation, and the resulting biofilm on the membrane surface became more hydrophobic [104]. EPS stimulated by the presence of MPs is the main factor causing membrane biofouling.

It should be noted, however, that MF/UF membranes do not completely remove MPs that remain in treated water or wastewater [9,41,58]. Nevertheless, UF membranes with a nominal size of 0.2 μ m in MBR are believed to provide 100% MPs removal [62]. A diagram of microplastic removal by membrane filtration is shown in Fig. 3. During MF/UF of water/wastewater containing MPs, there is a significant decrease in the final membrane/permeate flux capacity [96]. This indicates a significant interaction between MPs and membrane surface and pore interior, which involves adsorption characterized by a high rate. As the operating time increases and MPs particles accumulate, more and more of them permeate through the membrane into the permeate. Often MF/UF membranes are made of polysulfone and, like MPs, are negatively charged and hydrophobic. Thus, the forces of polar attraction are balanced by repulsive electrostatic forces caused by the surface charge of the membrane and MPs [78]. Intermolecular repulsion of MPs and the membrane surface is the main mechanisms in MPs removal by UF [78].

Yahyanezhad et al. [105] used a microfiltration (MF) membrane with a pore size of 0.1 μ m to eliminate MPs from biologically treated wastewater. A reduction in MPs concentration of 10⁶ MPs/L to only 2 MPs/L was achieved. Thus, the MF membrane can remove up to 98% of MPs.The number of MPs was also found to affect the efficiency of MF, as with more MPs, the efficiency of MF removal decreased. In addition, the particles detected after using the MF system were smaller than 0.1 μ m, meaning that the particles were within the nanoscale.

A comparative study of MF and UF ceramic membranes was conducted to evaluate their ability to remove MPs and their effect on fouling [106], using synthetic wastewater (deionized water + MPs in the form of 80 μ m nylon fibers) as a raw wastewater and actual wastewater from an industrial laundry. In the case of MF, the critical flux value was 200 L/m²h, which indicates high fouling of the MF membrane. No critical flux was obtained for UF, which clearly indicates less fouling than in MF. In both cases, 100% fiber



Fig. 3. UF scheme in microplastic removal, own compilation based on [62,76,78].

removal was achieved. For real wastewater, the critical flux value for MF was 90 L/m²h after 20 min, and for UF was 50 L/m²h after 60 min. Over 4 d of filtration, the decrease in permeability of MF was ~95%, while that of UF was only ~37%, demonstrating the greater suitability of UF in laundry wastewater filtration applications.

Studies have also been conducted on the removal of MPs from industrial wastewater using membranes made of polyacrylonitrile with reduced graphene oxide (rGO/PAN) [107]. It was found that increasing the concentration of rGO from 0.11 to 0.83 wt.% in the PAN matrix results in membranes containing uniform diameters (~150 nm), which makes MPs removal easier. The rGO/PAN membranes tested also exhibited anti-fouling properties and ease of filter cake removal. Thus, it was shown that composite membranes containing GO nanoparticles may be more useful for removing MPs from wastewater than classic UF/MF membranes.

In many cases, MF/UF are integrated with classical technologies used in water and wastewater treatment, such as sedimentation, classical filtration, flotation, biological and advanced oxidation processes [41,78,98]. UF integrated with the coagulation is very frequently used in water treatment plants, thanks to high removal of organic matter. Fig. 4 presents schematic diagram of MPs removal during coagulation and UF processes [79,101].

High concentrations of MPs in natural waters necessitate an in-depth study of the behaviour of MPs during the hybrid coagulation–UF process, since this is the technology used to produce drinking water [41,79,102].

Ma et al. [79,102] studied coagulation-UF process in drinking water treatment using FeCl, 6H,O and AlCl, 6H,O coagulants and PE in raw water. The density of PE (0.92-0.97 g/cm³) is very close to that of water, making it difficult to remove by sedimentation or flotation. PE particles were removed in 100%, due to the small pore diameter of the UF membrane and low fouling at the dose of conventional coagulant, especially for large PE particles. For larger PE particles, a heterogeneous floc layer was formed for the Al coagulant, resulting in less membrane fouling. With an increase in the coagulant dose, membrane fouling gradually increased as a thicker cake layer was formed on the membrane surface. The intensity of fouling in the coagulation-UF process depends on parameters related to both the type of UF membrane and the properties of the MPs (polymer type, size and shape). Another parameter affecting MPs removal efficiency is the pH of the solution, with removal efficiency generally decreasing with increasing pH at low concentrations of coagulant and small MPs sizes (<0.5 mm) [77].



Fig. 4. Scheme of the coagulation–UF process for removal of MPs, own compilation based on [41,78,79,102].

The addition of anionic polyacrylamide (PAM) resulted in higher removal efficiency of smaller MPs, improved performance (Fig. 5) and reduced fouling due to the opposite charge of anionic polyacrylamide (PAM) as coagulant-based flocs. At a FeCl₃-6H₂O dose of 2 mmol/L, the rate of PE removal increased with increasing solution pH, while other environmental parameters (ionic strength, presence of organic matter and turbidity) had little effect on MPs PE removal efficiency [79]. For the Al coagulant, the removal rate was also increased when anionic flocculant (PAM) was applied for the removal of smaller MPs (d < 0.5 mm). Removal efficiency increased from 25.83% without PAM to 61.19% using PAM 15 mg/L, while for larger MPs of 2–5 mm, efficiency increased from 4.27% to 18.34% [102].

Systematically studies have shown, that the removal of MPs by coagulation–UF process have can be used for drinking water treatment.

5.2. Dynamic membranes

Dynamic membranes (DM) are formed on the support (membrane, mesh or filter fabric) during filtration of a solution containing membrane-forming components in suspension. These components, which normally cause fouling of the membrane, become trapped in the filtration layer,



Fig. 5. Removal efficiency of polyethylene (PE) using $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and anionic polyacrylamide (PAM), own compilation based on [79,102].

preventing clogging of the carrier material [108]. Fig. 6 shows the dynamic formation of a dynamic membrane layer, which has the relevant retention and transport properties. The deposited layer plays the role of a "secondary" membrane formed on the supporting material (membrane or mesh) [80,109]. Immersion of the porous supports in a suitable colloidal suspension of the membrane-forming material and drying is also used. Porous carbon electrode tubes, hard polyvinyl chloride, sintered metal powders and ceramic tubes are used as porous materials. Organic polyelectrolytes and hydrated metal oxides in colloidal form are most commonly used as film-forming components.

The filtration of a system containing DM differs from the filtration mechanism of MF/UF, as the filtration resistance is induced by the cake layer [110]. The formation of DM depends on the characteristics of the supports (type of material and its porosity), the characteristics of the membrane-forming material (particle size, concentration), and the operational parameters of DM formation (operating pressure, transverse flow velocity) [111].

The major advantages of DM membranes over MF/ UF include: (i) the possibility of using low-cost materials (mesh, nonwoven, woven materials), (ii) the filtration layer is formed by impurities contained in the feed, (iii) greater compactness of the installation, (iv) higher permeate flux, which reduces the number of membrane modules installed, (v) low energy consumption is lower because DM operation is by gravity, (vi) lower filtration resistance and easy cleaning [1,109]. Recently, DM is becoming an attractive technology for the treatment of municipal wastewater, surface water, oily water, industrial wastewater and sewage sludge [109].

DM is especially effective for the removal low density (slow settling) contaminants and undegradable MPs due to the rapidly forming secondary membrane (DM layer) with microparticles [109]. Studies were carried out for MPs removal from synthetic wastewater in gravity mode using a DM laboratory filtration kit (90 μ m support mesh), obtaining about 90% of MPs removal [109]. The turbidity of the permeate has been reduced to <1 NTU (Nephelometric Turbidity Unit), which confirmed the rapid formation of DM resulting in better MPs removal efficiency, and the



Fig. 6. Demonstration of the dynamic cake layer, own compilation based on [80,108].

transmembrane pressure (TMP) during the DM filtration was 16 times lower than for MF and UF processes. The formation of DMs was dependent on the concentration of particles in the influent, as evidenced by an increase in TMPs (at a constant rate) during DM filtration. Higher particle concentrations in the influent caused more MPs to be filtered through the booster mesh.

DMs have high permeate yields and high MPs elimination rates as well as lower membrane fouling. It has been suggested that the integration of DM with coagulation or activated sludge wastewater treatment can effectively increase the removal of micropollutants and MPs [109]. It should be noted, however, that the 90 μ m mesh generally used in DM will pass smaller MPs. Although DM systems may be more efficient than UF membranes in some performance parameters, they cannot replace them [23]. Further research is needed to unravel the mechanism of DM layer formation.

5.3. Reverse osmosis

The reverse osmosis (RO) process is based on the phenomenon of natural osmosis, in which there is a spontaneous flow of solvent from a solution of higher to lower concentration through a semi-permeable membrane [92,99]. If the pressure in the solution exceeds the osmotic pressure, there is transport of the solvent from the more concentrated solution to the dilute one, thus the opposite of natural osmosis (Fig. 7) [92,99]. The transmembrane pressures used in RO are high and are mostly 2–6 MPa [112].

RO is mainly used for desalination of brackish and seawater but also for removal of organic and inorganic (micro) contaminants [41]. In industry, RO is used in the food industry, power generation, biopharmaceutical production, water demineralization, and the recovery of water from industrial and municipal wastewater [113].

Wang et al. [114] studied the removal of phthalate esters (dimethyl phthalate, dibutyl phthalate, diisobutyl phthalate and di(2-ethylhexyl) phthalate) and MPs from wastewater simultaneously at four WWTPs and reservoirs. Clarification, filtration and RO were used, and the retention of phthalate esters and MPs in all wastewater treatment plants was 47.7%–81.6% and 63.5%–95.4%, respectively. MPs in the form of granules and fragments (<0.01 mm in size) were present in wastewater with a concentration of 276–1,030 MPs/L and in water receivers –103–4,458 MPs/L.

To a greater extent, RO is used to remove MPs as a third or even fourth stage of wastewater treatment. Ziajahromi



Fig. 7. The principle of natural osmosis and reverse osmosis (C – concentration of the solute), own compilation based on [59,94].

et al. [7] investigated the effectiveness of the MPs removal by RO in wastewater treatment plant in Sydney, Australia, as a fourth-stage treatment. Treatment includes screening and sedimentation, biological treatment, disinfection/ dechlorination processes, ultrafiltration and RO. MPs fibers were found to be present after the RO process, with only 90.45% removal efficiency for MPs of >25 μ m [7]. After first, second and third stage treatment and RO, the wastewater treatment plant continues to release ten million plastic wastes per day into the natural aquatic environment [7].

The paper [115] presents the results of MPs removal in an integrated membrane system and classical activated sludge treatment. The classical system included grids, sand filter, sedimentation tank, activated sludge and secondary settling tank, while, IMS included pretreatment and MBR, ultrafiltration (UF) and RO (Fig. 8). The MBR contained capillary PVDF membranes with a pore size of 0.4 μ m, operating at a capacity of 1.50 × 10⁸ m³/d, while the RO was constructed with flat membranes with a pore size of 0.0001 μ m and a capacity of 4.0 × 10⁷ m³/d. The recovered water the RO process can be reused as industrial water. The removal of MPs in the IMS system after treatment in the MBR was 93.2% and increased to 98.0% after RO. The concentration of MPs in the MBR leachate was reduced from 1.5×10^{13} MPs/d to 10.2×10^{11} MPs/d, and by the RO process to 2.7×10^{11} MPs/d. Membrane purification showed a good degree of removal of MPs of various types, sizes and shapes. The results showed that IMS is more effective in removing MPs from wastewater, but should be taken into account the possibility of passing fine MPs fibers (<200 µm) through the IMS system, even equipped with RO.

5.4. Forward osmosis

The driving force of mass transport in the Forward Osmosis (FO) process is the difference in osmotic pressures on both sides of a semi-permeable membrane (Fig. 9a).

Except for some applications, the FO process always requires a water and a draw solution recovery unit (Fig. 9b) [118,119]. Water recovery from the dilute draw solution is carried out using another membrane technique, such as RO [99,116,117]. In the FO, similarly to RO, non-porous asymmetric membranes made of hydrophilic polymers, that is, cellulose triacetate, or composite membranes containing a polyamide active layer have been used [116]. The FO process is often used for obtaining drinking water and wastewater treatment due to its high retention of contaminants and volumetric efficiency of the membrane, as well as low fouling rates [99,116,119,120].

Recently developed the fertilizer-driven FO (FDFO) process allows the regeneration of wastewater, which can then be directly used to irrigate plant crops, with low energy consumption in the water recovery from the draw solution. Such use of FDFO process in wastewater reclamation for irrigation can reduce the need for water in agriculture, which is very important from the point of view of protecting water resources in the world [121,122].

Wang et al. [122] conducted a study on MPs/NPs removal through the FDFO process, aimed at generating highquality water for hydroponic irrigation from wastewater. Experiments were conducted using a FO membrane filtration



Fig. 8. Concepts of RO process application in wastewater treatment, own compilation based on [7,115].



Fig. 9. Scheme of (a) forward osmosis (FO) process, and (b) water/wastewater treatment installation using FO process [116-119].

system with cellulose triacetate FO membranes. The mode of investigation was to circulate the feed and receive separately on both sides of the FO membrane (Fig. 9b). The FO of the FDFO process was prepared by adding different concentrations of isolated bacterial extracellular polymers (5 mL/L concentration) and plastic model compounds (1 μ m and 100 nm polystyrene at 5 mg/L concentration) in deionized water. KCl (1 mol/L) was applied as FDFO draw

solution (DS). As expected, MPs and NPs were not detected in the water recovered after the FDFO process, the results of the experiment proved that the FDFO process can produce high quality water from wastewater by eliminating all contaminants present, including MPs. With their removal, the risk of absorbing the remaining contaminants in the reclaimed wastewater into the food web through irrigation with reclaimed wastewater can be significantly reduced. An additional advantage of the FDFO process is that the solvent (fertilizer) can be specifically selected to meet the requirements of specific plants grown in hydroponic systems, due to the cut-off of FO membranes (about 200 Da, comparable to RO membranes), indicating the complete removal of plastics in the FDFO process. Good removal of MPs and NPs from wastewater can significantly reduce the risk of contaminant-related plastics entering the human food web through wastewater reuse. However, to determine whether This FDFO can be applied in practice, the investigation of longterm stability and fouling rate are needed. Similar to other membrane filtration process, membrane fouling is also the main challenge for the stability of FDFO process [123].

5.5. Membrane bioreactors

Membrane bioreactor (MBR) is wastewater treatment processes where MF or UF membrane has been integrated with a biological process (bacteria, enzymes) [1,124]. The removal mechanism is dual in nature, that is, biodegradation and membrane filtration. Two MBR design solutions are used: a membrane module outside the bioreactor (sMBR) and an immersed module in the bioreactor (iMBR) (Fig. 10) [1,124,125]. The iMBR configuration requires less frequent cleaning and has lower energy consumption. The sMBR configuration, on the other hand, can use a higher concentration of MLSS (Mixed liquor suspended solids), which affects ease of system maintenance and module replacement and cleaning because the system is more compact. MBR uses modules that include flat sheet membranes, capillary membranes and multi-channel modules. The choice of module type depends on the specific MBR application. Modules containing flat-sheet membranes and capillary membranes are mainly used in iMBR, for the treatment of both industrial and municipal wastewater. In smaller installations, modules with flat-sheet membranes are preferred because of their simplicity of operation. In larger installations, MBRs containing capillary modules are usually used, due to their lower energy requirements.

In MBR, UF or MF membrane separates the solids from the biomass-containing liquid, which allows to eliminate the secondary settling tank used in the classical activated sludge method and to achieve high biomass concentration. UF/MF membranes used in MBR are characterized by high hydrophilicity, which provides high flux (efficiency) and high oxidation resistance and tensile strength (≥200 N). It fits perfectly with the principles of green chemistry, according to the logic of process intensification, which offers new and much greater opportunities for competitiveness, product quality improvement, process innovation or novel products, and environmental friendliness [125]. With the increasing number of commissioned large-scale (≥10,000 m³/d) and super large-scale (≥100,000 m³/d) wastewater treatment plants, MBR technology is taking a prominent place in the field of municipal and industrial wastewater treatment [1,118,124]. The success of MBR over traditional biological wastewater treatment methods, is mainly due to the high quality of treated wastewater, the small area occupied by the plant, the independence of hydraulic and biomass retention time, the ease of scale-up, while the main disadvantage is the significant energy consumption, mainly due to membrane fouling [1,124–126].

The use of MBR in the treatment of municipal and industrial wastewater can also contribute to increasing the removal of MPs. In MPs removal, the role of MBR is to facilitate biodegradation of organic matter, which translates into high MPs removal and further treatment. In MBR, MPs is concentrated in the retentate stream due to the presence of a membrane that separates solids from liquid.

A study by Talvitie et al [9] compared the efficiency of MPs removal using MBR (UF modules containing flat membrane sheets with a pore size of 0.4 μ m and a surface area of 8 m²) with three tertiary wastewater treatment technologies, that is, disk filter, rapid sand filtration and dissolved air flotation. The MBR treatment process achieved MPs removal of 99.9% (from 6.9 to 0.005 MPs/L), while using sand filter 97% (from 0.7 to 0.02 MPs/L), flotation 95% (from 2.0 to 0.1 MPs/L) and disk filter 40%–98.5% MPs (from 0.5–2.0 to 0.03–0.3 MPs/L). These studies have proven that by using tertiary wastewater treatment in the form



Fig. 10. Outside (a) and immersed (b) MBR configurations [1,124,125].

Method	Effluent type	Before (MPs/L)	After (MPs/L)	Removal (%)
Disc filter 10 µm	After second stage treatment	0.5	0.3	40.0
Disc filter 20 µm	After second stage treatment	2.0	0.03	98.5
Sand filter	After second stage treatment	0.7	0.02	97.1
Flotation	After second stage treatment	2.0	0.1	95.0
MBR	After first stage treatment	6.9	0.005	99.9

Table 5 MPs concentrations before and after treatment with different technologies [128]

of advanced technologies, the amount of MPs discharged from WWTPs into the natural aquatic environment can be significantly reduced.

Similarly, Lares et al. [32] studied the MPs removal from wastewater by a municipal wastewater treatment plant that operated using a conventional activated sludge process (CAS) and a pilot-scale MBR. They found that the permeate from the MBR contained only 0.4 MPs/L and effluent from the CAS process (1.0 MPs/L). The results showed, that MPs removal efficiency in amounted to 99.4% and for CAS-based process 98.3%. The study shows that both fibres and MPs particles are released from the wastewater treatment plant into the aquatic environment.

Li et al. [82] investigated the removal efficiency of a 10-particle/L PVC gel (particle size $<5 \,\mu$ m) by an MBR method containing an immersed 0.1 μ m UF membrane with a surface area of 0.1 m². No MPs were detected in the permeate of the MBR system at a 2.5-h HRT, temperature 19°C and pH. Fouling investigation of the MBR membrane showed that MPs could lead to more membrane fouling, including irreversible.

Baresel et al. [127] investigated an MBR (with UF module and a 10-h HTR) combined with a 0.3 m² granular activated carbon biofilter for micropollutants removal, of which MPs, from real wastewater (Henriksda WWTP, Stockholm). It was found that pharmaceuticals, phenolic compounds, bacteria and MPs, have been removed to below detection limits or very low concentrations. This shows that the combination of filtration, adsorption and biodegradation provides a broad and efficient removal of micropollutants and MPs.

A comparison of MPs removal by the tertiary treatments is shown in Table 5 [128], from which it follows that MBR allowed the highest reduction of MPs in the effluent, demonstrating that the membrane-based technology is the most efficient.

MBR can remove all size fractions (especially the smallest sizes, 20–100 μ m) and all shapes of MPs from wastewater compared to other advanced treatment methods [9,32]. However, thanks to the presence of membranes with a certain porosity, the efficiency of the removal of MPs MBR is not to be influenced by the shape, size and composition of MPs.

It has been observed that MPs with smaller sizes, especially fibres, are not completely removed by MBR due to their high length-to-width ratio [129,130]. MPs therefore remain in the sludge after filtration, and have to be again treated as solid waste, which increases treatment cost. Other drawbacks of wastewater treatment by means of MBR are the control of biofilm thickness, fouling, and liquid distribution, which determine effectiveness and the cost treatment [1,131]. Future MBR technology research should be directed at fouling, and the degradation and/ or transformation of MPs in MBRs. Various methods have been developed to control fouling primarily: membrane flushing assisted with aeration, intermittent permeation or relaxation, backflushing with water and/or air, cleaning with chemicals and enzymes, bacterial degradation of the fouling layer, electrically assisted mitigation of fouling, as well as the use of membranes containing nanomaterials and the application of antifouling agents [124–126].

New future challenges for MBR are related to the isolation of a bacterium (*Idonella sakaiensis*) that will use polyethylene terephthalate (PET) as a major source of energy and carbon [132]. This bacterium produces two enzymes that can convert PET into less hazardous monomers (terephthalic acid and ethylene glycol). In addition, MPs exposed to Antarctic krill (*Euphasia superba*) were found to undergo fragmentation involving a reduction in size (from 31.5 μ m to less than 1 μ m) [133]. The resulting enzymes can be integrated into the MBR to form an enzymatic membrane reactor and used to degrade MPs [134].

The studies cited above indicate that the removal rate of MPs in MBR is highly effective and relatively stable technology for wastewater treatment.

6. Final remarks

Microplastics cause pollution of the Earth's environment, which is a serious problem to be solved. It is therefore necessary to develop MPs treatment processes to reduce plastic pollution. The water and wastewater treatment industry does not currently have the technology or experience to effectively remove MPs from the aquatic environment. Research conducted to date indicates that tertiary treatment processes include advanced separation processes are needed to remove MPs from wastewater. The most promising method appears to be membrane processes, particularly MBR, which provide MPs removal of 99.9%, also offering the possibility of reducing the number of process steps in wastewater treatment plants. Integration of coagulation with UF is one of the important water treatment processes in today's water utilities, showing significant removal of organic matter, including MPs. Membrane technologies are not designed to remove MPs sufficiently, due to the problems of membrane fouling and declining performance during operation. Therefore, research should be directed toward minimizing membrane abrasion and fouling in membrane technology for MPs removal. However,

116

membrane treatment technology can be attractive for MPs removal when combined with biological processes such as MBR or chemical processes such as coagulation.

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