

Membranes in water and wastewater disinfection – review

Michał Bodzek^{1*}, Krystyna Konieczny^{2,3}, Mariola Rajca²

¹Institute of Environmental Engineering Polish Academy of Sciences, Poland

²Silesian University of Technology, Poland

Institute of Water and Wastewater Engineering

³Cardinal Stefan Wyszyński University in Warsaw, Poland

*Corresponding author's e-mail: michal.bodzek@ipis.zabrze.pl

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Abstract: Production of sanitary safe water of high quality with membrane technology is an alternative for conventional disinfection methods, as UF and MF membranes are found to be an effective barrier for pathogenic protozoa cysts, bacteria, and partially, viruses. The application of membranes in water treatment enables the reduction of chlorine consumption during final disinfection, what is especially recommended for long water distribution systems, in which microbiological quality of water needs to be effectively maintained. Membrane filtration, especially ultrafiltration and microfiltration, can be applied to enhance and improve disinfection of water and biologically treated wastewater, as ultrafiltration act as a barrier for viruses, bacteria and protozoa, but microfiltration does not remove viruses. As an example of direct application of UF/MF to wastewater treatment, including disinfection, membrane bioreactors can be mentioned. Additionally, membrane techniques are used in removal of disinfection byproducts from water. For this purpose, high pressure driven membrane processes, i.e. reverse osmosis and nanofiltration are mainly applied, however, in the case of inorganic DBPs, electro dialysis or Donnan dialysis can also be considered.

Introduction

Microbiological condition of water plays a significant role for humans. The accidental appearance or permanent presence of pathogenic microorganisms in water dedicated for potable purposes may result in spreading of many diseases, thus, it is important to perform proper water treatment and disinfection processes. The latter operation is the crucial one, as it enables elimination of microorganisms, including pathogens, which may lead to epidemic effects. Microorganisms can be mainly found in soil, soil waters, shallow ground- and surface waters. In the case of groundwater, it can be generalized that the deeper the water intake is, the fewer bacteria are present. However, not all of those bacteria are harmful to humans. Moreover, in specific cases, they can even enhance the removal of particular contaminants from water. Another important topic is presence of microorganisms in wastewater, especially municipal one.

Disinfection is found to be a principle technological action of every water treatment system. It is also said to be one of the most difficult and complicated operations, regardless of the scale of water treatment plant (Collivignarelli et al. 2018). In the case of treatment of water dedicated to potable purposes, disinfection should assure both, the production of microbiologically safe water and maintenance of its quality during transport, including

prevention of secondary biological contamination of water in pipelines (Nawrocki 2010). In order to assure its biological safety, potable water is disinfected, usually by means of chlorination, which is the most common technique applied for this purpose. On the other hand, disinfection byproducts (DBPs) generated during water chlorination are found to be mutagenic and carcinogenic (Collivignarelli et al. 2018, Nawrocki 2010). Another issue related with conventional disinfection is that some microorganisms may become resistant to chlorine or require its high dosage for inactivation. The relatively high concentration of residual chlorine may in turn affect the wrong taste and smell of potable water intended for human consumption. Concerns related with water chlorination have resulted in the development of many alternative disinfection processes dedicated for potable water purposes. Among them one can find processes such as ozonation, chlorine dioxide addition, ultraviolet (UV) radiation and advanced oxidation processes (Bogacki and Al-Hazmi 2017, Collivignarelli et al. 2018, Nawrocki 2010). These techniques are found to be very efficient, but most of them require the use of either expensive chemicals or expensive devices for on-site disinfectants generation, e.g. in the case of chlorine dioxide or ozone. Moreover, many chemical disinfectants may lead to the formation of other harmful disinfection byproducts like bromates and brominated DBPs in the case of waters with

elevated bromides content (Bodzek and Konieczny 2011, Nawrocki 2010).

One of the alternatives for water disinfection is membrane filtration, especially ultrafiltration (UF) and microfiltration (MF) with polymer or ceramic membranes (Kwasny et al. 2018), while for removal of DBPs high pressure driven membrane processes, i.e. reverse osmosis (RO) and nanofiltration (NF) can be applied (Bodzek 2013, 2015).

Membranes in water disinfection

Water, which contains biologically active components, i.e. viruses, bacteria and protozoa as well as other microorganisms (fungi, algae, snails, worms and crustaceans), if dedicated to potable purposes, may seriously harm human health. It is also valid to treated and untreated wastewater discharged to natural water collectors and sewage systems. In Poland, in regulations on potable water quality one can find *Escherichia coli* and *Enterococci*, which cannot appear in a water sample of volume 100 mL. In additional water quality parameter, permissible standards on coli population, total number of microorganisms as well as *Clostridium perfringens* are defined (Ann. 2017). Due to regulation on surface water treatment, minimum removal/deactivation of *Giardia* microorganism should be at least 3 log, while viruses and bacteria at least 4 log (Zhua et al. 2005). Biological contamination of water source may appear naturally, during its intake, its treatment or in water transport in pipelines. As mentioned, there exist many methods, which can be used to water disinfection, and each of them reveals a number of advantages and disadvantages.

The application of membranes to water disinfection has already been known for many years. Membrane filters were used during the 2nd World War by German soldiers to control microbiological contamination of water after bombarding (Koltuniewicz and Drioli 2008). Membranes can be used either directly at consumers' site and/or as a part of water treatment system. Membrane filtration, especially UF and MF, can enhance and improve conventional water disinfection processes. The size of viruses' cells is of a range from 20 to 80 nm, while pore size of UF membranes is <10 nm, hence, theoretically those cells should be completely rejected. Bacteria (0.5–10 µm), cysts and oocysts (3–15 µm) are larger, thus their complete removal is practically possible with the use of UF and MF, as pore size of commercially available MF membranes is

below 0.1 µm (Bodzek 2013, 2015). Moreover, comparison of membrane pore size with microorganisms cells size indicates that UF should assure complete disinfection of water (Fig. 1).

Among microorganisms, which appear in water, the proper removal of viruses requires the most attention due to the possibility of infection at low chlorine doses, long lifespan and poor removal efficiency during conventional water or wastewater treatment. Throughout the last two decades, virus membrane filtration has become a mature standard unit operation for virus removal in the water treatment and biopharmaceuticals derived from human or animal origin (Chen and Chen 2016, Kosiol et al. 2017). It is assumed that MF membranes pore size range (0.1–1 µm) allows to reject bacteria and protozoa, while it is not suitable for viruses removal, as their cells are significantly smaller. The rejection rate of viruses cannot be predicted only on the basis of nominal pore size or molecular weight cut-off (MWCO) of UF/MF membranes. It has been confirmed by a range of studies on the application of MF to viruses removal (Bodzek 2013, 2015). In the case of UF, the membranes of MWCO ranging from 10 to 100 kDa are not always capable to reject all viruses. Hence, smaller species require the use of NF membranes, pores of which are usually below 1 nm. In Table 1, the results of the studies on a selected virus, i.e. bacteriophage MS2, removal with the use of UF and MF are shown (Bodzek 2013, 2015). UF membranes removed the virus with higher efficiency (4–7 log) than MF ones (<1–3 log). In some cases, relatively high rejection rate with MF was explained by adsorption on membrane surface, formation of filtration cake and deposition within other organic compounds, which naturally occurred in water.

During other studies (Koltuniewicz and Drioli 2008) the removal rate of MS2 virus from surface water, using MF membranes of pore size 0.2 µm, reached 1.7–2.9 log. The number of MS2 virus cells in raw water was in the range from 1.3×10^6 do 3×10^7 , while in the permeate it was from 2.2×10^4 do 3.4×10^5 . Frohnert et al. (2015) performed experiments to determine the removal of viruses (human adenoviruses (HAdVs), murine norovirus (MNV), and the bacteriophages MS2), in different types of water (surface water from reservoirs for drinking water treatment, treated groundwater and groundwater contaminated with either 5 or 30% of wastewater) by UF using a semi-technical unit. Bacteria were not detected in the permeate, but in the case of viruses and bacteriophages the permeate still contains them: log removal

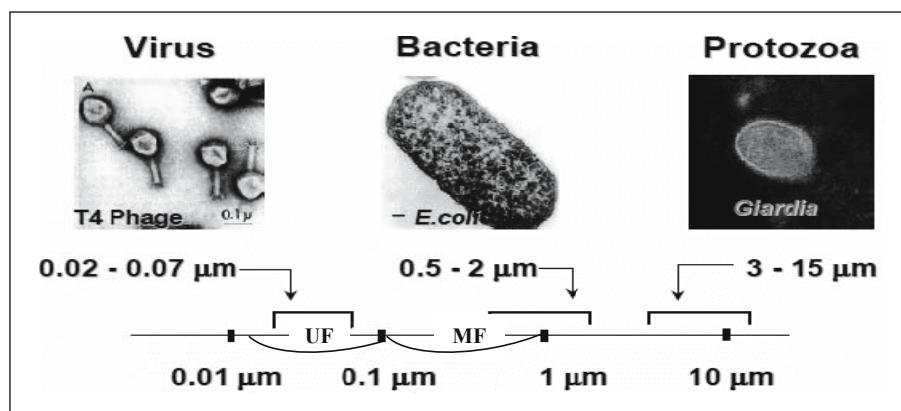


Fig. 1. Comparison of micro-organisms sizes with MF and UF membrane pore size

Table 1. The results of the virus MS-2 removal for MF and UF

Water	Membrane (module)	Average concentration in raw water, cfu/100 mL	Removal, log
Aqueduct San Diego, USA	UF	8×10^7 – 6×10^9	4.0–5.6
	Hydranautics UF	2.8×10^9 – 1.7×10^{10}	3.9–4.7
	Ionics UF	7.4×10^8 – 2.8×10^9	4.0–5.7
	UF Zee-Weed 500	3.5×10^{10} – 5.9×10^{10}	>5.5–5.8
Lake Yssel, The Netherlands	X-Flow UF	18000	4.9
	MF	1.0 – 1.1×10^5	0.7–2.3
	UF	2.2 – 2.5×10^4	>5.4
Laboratory-pure	MF	140–745	>1.5
	Koch-Lab 5UF	2.4×10^3 – 1.4×10^4	2.0–6.3
Bull Run Reservoirs, USA Lake Elsman, USA	MF	10^5 – 10^{12}	0.5–2.0
	UF		3–>7
Colorado River, USA	MF Memcor	1.3×10^9 – 1.6×10^{10}	1.7–2.9

cfu – colony forming units

values were in the range of 1.4–6.3, depending on virus sizes and water quality. The removal of polio virus was <2 log for 0.2 μm pore size membrane, while its complete rejection >6 log, accompanied with total removal of MS2 virus (Bodzek 2013), was reached with the use of UF membranes of cut off 30 and 100 kDa, respectively.

While the first commercial virus filters were intended to remove larger viruses with diameters >50 nm, like retroviruses, nowadays such filters have to ensure efficient removal (at least 4 log) also of small viruses like parvo-viruses with 18–24 nm in diameter (Cameron and Smith 2014). So, the major challenge that virus filters have to overcome is to increase their selectivity (Rayfield et al. 2015). Recent research found that the increased virus removal was accomplished by both the decrease of pore size and the increasing repulsive forces exerted by foulants (Lu et al. 2013). Other studies suggest that the virus membrane-interaction forces are significant in determining the virus removal efficacy in membrane filtration (Huang et al., 2012). Virus transport through the membrane is also influenced by the hydrodynamic forces. Due to the slow diffusion of viruses compared to the convective forces, the viruses rejected by the membrane accumulate on the membrane surface, leading to an increase of the local concentration of viruses. As a result, the virus concentration in the permeate also increases.

Based on the above-mentioned mechanisms, virus removal by membrane filtration can be improved by inducing repulsive virus-membrane interaction forces to prevent viruses to be deposited on the membrane surface. One of solutions is “Zwitterionic hydrogels”, which have been commonly used to exert repulsive forces onto a commercially available UF membrane (Lu et al. 2016, Werber et al. 2016). Lu et al. (2017) grafted the zwitterionic hydrogel, which repels the viruses from the membrane surface. It contains both positive and negative charges and improves efficiency by weakening virus accumulation on the modified filter surface. The result was a significantly higher rate of the removal of waterborne viruses, including human norovirus and adenovirus. Since hydrogel may have a minor influence on the water flux through the membrane, the virus removal would be improved without decreasing the membrane permeability. Bacteriophage MS2 and human adenovirus type 2 (HAdV-2) were used to check the

new membrane. About 18% loss in membrane permeability and increase of the removal HAdV-2 (4 log₁₀) and MS2 (3 log₁₀) were obtained. The simple graft-polymerization functionalization of commercialized membrane achieving enhanced virus removal efficiency highlights the promise of membrane filtration for pathogen control in potable water reuse.

Another way for virus reduction in water is polymeric membrane modification with cationic polymers (Sinclair et al. 2018). The poly-cationic chains can damage virus layer on membrane surface and furthermore, they can also damage the capsids of the more resistant non-deposited waterborne viruses. Specific polymers like polyethyleneimine (PEI) have been found to be good compounds for imparting antibacterial and antiviral properties onto surfaces (Larson et al. 2011). The membrane modification resulted in ~22% loss of the membrane permeability while an increase of ≥ 3 log₁₀-units ($\geq 99.9\%$) in MS2 reduction was observed.

Hence, viruses of small cell size are able to permeate through MF and UF membranes and the observed removal efficiencies are in the range from 2 to 6 log. In order to prevent the incomplete removal of viruses during UF/MF, integrated processes are applied. Among them, coagulation-membrane filtration system is the most popular (Zhua et al. 2005, Fiksdal and Leiknes 2006). Zhua et al. (2005) performed studies on the removal of MS2 bacteriophages (cell size ca. 25 nm) using coagulation with FeCl₃ proceeded with MF. In the case of coagulant doses from 0 to <2 mg Fe/L, the removal efficiency was below 0.5 log, while for doses from 5 to 10 mg Fe/L it increased to >4 log. The experimental data showed that negatively charged MS2 virus cells were firstly adsorbed on positively charged iron hydroxide particles (FeOOH), and next those were separated by MF membranes. Fiksdal and Leiknes (2006) carried out studies on the removal of MS2 virus from potable water by means of integrated coagulation-membrane filtration (MF and UF) system and with the use of aluminum coagulants (ALG and PAX). When direct membrane filtration was applied, poor removal of the virus was observed. In the case of primary coagulation with Al dose 5 mg/L (regardless of coagulant type) high rejection of the virus (>7.4 log) was obtained after membrane filtration and the decrease of the dose to 3 mg Al/L insignificantly affected the removal rate, which

decreased to >7.1 log. Only in the case of application of PAX (3 mg Al/L) followed by MF, the rejection was lower and equal to 6.7 log. Additionally, the treatment enabled the significant reduction of water colour. Meyn et al. (2012) investigated MS2 bacteriophages removal from surface water, with high natural organic matter (NOM) content, by inline coagulation/flocculation pretreatment followed by ceramic microfiltration. MS2 and DOC removal increased with lower pH and higher coagulant. Both investigated coagulants showed virus inactivation about two log units after 60 min contact time, which is equivalent to a virus inactivation of 99%. This inactivation was only reversible to a small extent by chemical or physical floc destruction. The investigated process combination can comply with modern hygienic barrier standards.

In Table 2, the results of MF and UF removal of coli group bacteria, fecal coliform and *Pseudomonas* are shown (Bodzek 2013, 2015). The retention coefficients are in the range from 0.7 to 9.8 log and the lowest rejection is observed for the lowest (at the limit of detection) concentration of microorganisms in raw water.

Hassan (2017) et al. used palm fruit stalks cellulose nanofibers (CNF), oxidized CNF (OCNF) and activated carbon (AC) to make thin film membranes for the removal of

E. coli bacteria from water. Two types of layered membranes were produced: a single layer setup of crosslinked CNF and a two-layer setup of AC/OCNF (bottom) and crosslinked CNF (up) on hardened filter paper. The two-layer AC/OCNF/CNF membrane had much higher water flux than the single layer CNF due to higher porosity on the surface of the former. Both types of membranes showed high capability of removing *E. coli* bacteria (rejection ~96–99%) with slightly higher efficiency for the AC/OCNF/CNF membrane than CNF membrane. AC/OCNF/CNF membrane also showed resistance against growth of *E. coli* and *S. aureus* bacteria on the upper CNF surface while the single layer CNF membrane did not show resistance against growth of the aforementioned bacteria.

Zimer et al. (2016) present the optimization of porous anodic alumina membranes for ultrafiltration prepared by anodically oxidized aluminum foils. *Escherichia coli*, a common bacterial contamination of drinking water, was removed using these membranes with 100% of efficiency to obtain bacteria-free water.

In Table 3 the summary of bacteriological parameters of raw surface water as well as permeates obtained during filtration with the use of polymeric and ceramic membranes arranged in different modes are presented (Bodzek and

Table 2. Bacteria removal results for MF and UF

Bacteria	Water	Membrane (module)	Average concentration in raw water, cfu/100 mL	Removal, log
Coli group bacteria	Saine River, France	UF Aquasource	1800–1.0×10 ⁵	>4.3
<i>E. coli</i>	Laboratory-pure	MF and UF	6.6×10 ⁷ –9.6×10 ⁸	5.6→9.0
<i>Pseudomonas</i>	Laboratory-pure	MF and UF	1.5×10 ⁸ –5.3×10 ⁸	>8.2→8.7
Coli group bacteria	Lake Elzman, USA Bull Run Reservoir, USA	Two MF module Two UF module	11–972 6–160	>0.7→3.0 >0.7→2.2
Coli group bacteria <i>E. coli</i>	Colorado River, USA	MF Memcor	(14–240)60 9.8×10 ⁷ –2.7×10 ⁸	>1.7 >6.0→6.4

cfu – colony forming units

Table 3. Microbiological analysis of raw surface water (Kozłowa Góra water intake, south part of Poland) and permeates obtained during filtration with polymeric and ceramic membranes in different module system

Membrane	Number of <i>E. coli</i> bacteria in 100 mL			The number of mesophilic bacteria, in 1 ml at 37°C/24h		
	Raw water	Permeate	R	Raw water	Permeate	R
Polymeric flat membrane						
PAN-13	63 (240)	0 (<5)	100	36	3	91.7
PAN-15	60 (240)	0 (<5)	100	205	1	99.5
PSf-13	45 (240)	0 (<5)	100	250	0	100
PSf-15	60 (240)	0 (<5)	100	320	4	98.75
PAN/PSf-15	30 (20)	6 (6)	80	60	6	90.0
Ceramic membrane						
MF – 0.1 µm	60 (240)	0 (<5)	100	205	1	99.5
MF – 0.2 µm	60 (240)	0 (<5)	100	220	2	88.9
UF – 15 kDa	30 (23)	0 (<5)	100	60	4	93.3
UF – 300 kDa	63 (240)	0 (<5)	100	36	4	88.9
Capillary membrane						
Polypropylene	86 (62)	0	100	24	0	100
Polysulfone	28 (23)	0	96.4	23	4	82.6

R – retention coefficient, PAN – polyacrylonitrile, PSf – polysulfone

Konieczny 1998). The efficiency of disinfection performed with the use of particular membranes is high in reference to *E.Coli* as well as to mesophilic bacteria. The former ones are effectively removed from both, surface and well waters and the observed rejection is almost 100% for all applied membranes. The removal of mesophilic bacteria is in the range from 89 to 100% for surface water and from 92 to 95.5% for well water.

Polyacrylonitrile, capillary membranes impregnated with chitosan containing iron oxide nanoparticles were used for the removal of Gram positive and Gram negative bacteria of *Pseudomonas aeruginosa* (length: 1.5 μm , thickness: 0.8 μm) and *Staphylococcus aureus* (diameter: $1 \pm 0.2 \mu\text{m}$) types (Mukherjee and De 2017). The introduction of nanoparticles improved permeability, mechanical strength and hydrophilicity of membranes. Biofilm on a membrane surface caused a damage to cells' membranes, what was directly confirmed by intracellular fluid analysis carried out at UV 260 nm and by direct SEM observations. The damages of bacteria cells were probably caused by electrostatic interactions between NH_3^+ groups of nanoparticles and anionic components of phosphoryl groups of bacteria. The applied membranes revealed promising results on biofouling resistance during long time operation. The study on the impact of process conditions on retention and flux profile during long term experiments showed only 5% decrease in permeate flux.

Intestine protozoa (*Giardia* and *Cryptosporidium parvum*), which may appear in potable water, are responsible for infectious diseases. *Cryptosporidium parvum* oocysts are widely spread in surface waters, treatment of which does not always prevent the spread of diseases, especially that the harmful dose of oocysts is very low (132 oocysts), moreover, those oocysts are resistant to chlorine disinfection (Koltuniewicz and Drioli 2008). The effectiveness of *Cryptosporidium parvum* oocysts removal on sand filters reaches 2–3 log and does not guarantee their complete removal (Bodzek 2013, 2015). Thus, if the raw water is contaminated with *Cryptosporidium* oocysts at the level of >3 cells, conventional filtration process has to be replaced with an alternative technique, which guarantees their sufficient removal. MF membranes of pore size 0.2 μm seem to be suitable barrier for *Cryptosporidium* and *Giardia* as well as for other protozoa of cell size 3–14 μm (Table 4).

It has been generally accepted that MF and UF, in most cases, can provide complete removal of all protozoan cysts, in this *Cryptosporidium* and *Giardia*, with efficiency above 4.5 log and meets the limits established within water quality standards, what has been confirmed by many pilot and industrial scale studies carried out at various water treatment plants (Bodzek 2013).

It should be noted that UF membranes are not always able to assure the complete elimination of microorganisms from water. It results mainly from imperfection of membranes and membrane modules as well as from secondary growth of bacteria in water after filtration. The discontinuous structure of skin layer met in commercial membranes enables the permeation of microorganisms to permeate, while the construction of membrane modules does not always assure the complete separation of feed water from permeate. Additionally, it has been found that microorganisms' cells are able to penetrate membrane pores even though their size is much smaller than cell size. It is mainly due to pressure deformation accompanied with filtration of intracellular fluid and maintenance of cells' membrane tonus (Fig. 1). Additionally, it has been shown that cells' shape is a key factor determining membrane retention of particular microorganisms. For example, bacteria and viruses of slender, elongated shape are rejected more effectively than ones of more compact shape (Wang et al. 2008).

Capillary membrane modules have been found to be the most effective for water disinfection, as the separation of raw water from permeate is easier than in spiral wound or hollow fiber modules, what has been confirmed by studies results (Bodzek 2015, 2013, Mukherjee and De 2017).

The example of commercial use of MF for the removal of turbidity and microorganisms from surface water is the Water Treatment Plant in Sucha Beskidzka, Poland, which intakes raw water from mountain river. It is based on MF system supplied by Pall (Bodzek 2013). During the treatment, the raw water passes through clarifier and grit trap to collecting well, into which aluminum sulphate is dosed. Next, the water is pumped to post coagulation sedimentation tank, next to sand filter, and finally to clean water tank. The scheme of the water treatment plant including MF of Pall Aria, type AP, is shown in Fig. 2 (Bodzek 2013). The filtration membrane system PALL Aria™ comprises 40 membrane modules (USV-6203 type)

Table 4. Results of oocysts *Cryptosporidium* and cysts *Giardia* removal for MF and UF

Water	Membrane (module)	cysts <i>Giardia</i>		oocysts <i>Cryptosporidium</i>	
		Raw water, cfu/100mL	Removal, log	Raw water, cfu/100mL	Removal, log
Highland reservoir	MF Microza	11.8×10^6	>5.8	1.01×10^8	>6.8
	UF Aquasource	10.4×10^6	>5.5	8.2×10^7	>6.5
	UF Zee-Weed	8.6×10^6	>5.3	1.1×10^7	6.4
Laboratory-clean	MF and UF	5.4×10^4 – 1.5×10^5	4.6–5.2	2.6×10^4 – 8.2×10^4	4.2–4.9
Elsman Lake, USA Seine River, Paris	Three MF	2.8×10^4 – 1.3×10^5	>6.4–>7.0	1.1×10^4 – 7.4×10^4	>6.0–>6.9
	Three UF	2.6×10^4 – 1.0×10^5	>6.4–>7.0	2.41×10^4 – 9.1×10^4	>6.3–>7.0
Guyardotte River, USA	MF Memcor	1.0×10^7	>7.0	No	No
Colorado River, USA	MF Memcor	2.8×10^4	>4.4	No	No
Surface water	MF Fibrotex	No	No	1000	2–3

placed in single block together with the additional equipment. The recovery rate of permeate is very high and reaches 99%, in dependence of feed water parameters, and it is operated at capacity 130 m³/h. The application of such system was very important due to the appearance of harmful organisms in raw water, which were found to be resistant to conventional chlorination.

The exploitation of the device confirmed possibility of production of high quality water of turbidity much below 0.1 NTU and deprived of any microorganisms (Bodzek 2013). During the exploitation period, temperature and turbidity of

raw water fluctuated significantly, and during heavy rain falls the turbidity could reach more than 800 NTU. In Table 5, physico-chemical and microbiological characteristic of water treated at installation in Sucha Beskidzka is presented (Bodzek 2013). Operation costs are also a very important factor, which has a direct impact on final water price. In this case, they are compensated by significant decrease of chlorine dioxide and coagulant consumptions as well as by decrease of operational costs of sand filters.

To sum up, UF and MF membranes are an effective barrier for pathogenic protozoa cysts and bacteria. Additionally, they

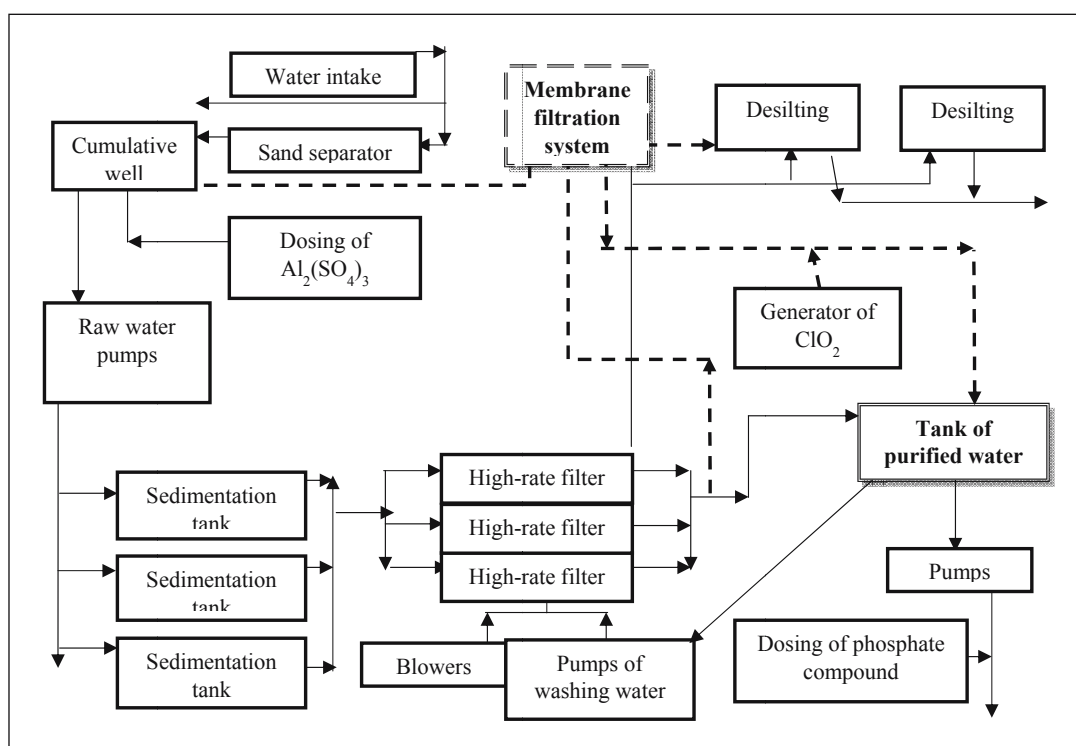


Fig. 2. Technological scheme of drinking water treatment plant for in Sucha Beskidzka, Poland

Table 5. The physicochemical and microbiological parameters of drinking water obtained in Sucha Beskidzka, Poland, plant and normative values for drinking water in Poland

Parameters	Data	Normative values
Turbidity, NTU	0.08	1
Colour, mg Pt/L	5	acceptable
pH	7.5	6.5–9.5
Conductivity, μS/cm	250	2500
Nitrates, mg/L	3.2	50
Total hardness, mg/L	96	60–500
Chloride, mg/L	6.0	250
Coliform bacteria w 100 mL water	0	0/100 mL
Coliform fecal type bacteria/100 mL water	0	0/100 mL
Fecal streptococci in 100 mL water	0	0/100 mL
Clostridia reducing sulphite w 100 mL water	0	0/100 mL
The number of colony – forming bacteria in the 37° after 24 h in 1 mL	0	20/1 mL
The number of colony – forming bacteria in the 22° after 72 h in 1 mL	2	100/1 mL

assure the reduction of chlorine consumption for treated water disinfection, which is performed for maintenance of biological water quality in a pipeline system.

Membranes in disinfection byproducts (DBPs) removal

Byproducts of disinfection (DBPs) and oxidation are undesired groups of substances formed during reaction of disinfecting agents or other strong oxidizers with admixtures and contaminants present in water (Zazouli and Kalankesh 2017, Nawrocki 2010). The group of DBPs mostly comprises organic compounds, but some of inorganic substances are also included (bromates, chlorites and chlorates). In Table 6, a series of organic DBPs is shown, among which trihalomethanes (THMs) and haloacetic acids (HAA) are ones of the highest concern (Bodzek et al. 2011, 2015, Nawrocki 2010). Most of them appear in water at very low concentration of ppb (mg/m^3) level or even lower. Hence, they are regarded as water or wastewater micropollutants.

During reaction of chlorine with organic compounds, many DBPs are formed, mainly trihalomethanes and haloacetic acids. In order to decrease DBPs concentration in water a range of methods can be applied (Bodzek 2013, 2015), such as: use of other oxidants like ozone or chlorine dioxide, removal of DBP precursors from water before oxidation, and removal of DBPs by various techniques. The best recognized chlorination byproducts are **trihalomethanes (THMs)**. Precursors of THMs are humic acids, chlorophyll

“a”, metabolites of aquatic organisms, aliphatic hydroxyl acids, mono-, di- and tricarboxylic acids and aromatic carboxylic acids (Zazouli and Kalankesh 2017, Nawrocki 2010). The main identified halogenated organic compounds are chloroform (CHCl_3), bromodichloromethane (CHBrCl_2), dibromochloromethane (CHBr_2Cl) and bromoform (CHBr_3). Among them, chloroform usually appears at highest concentration. Brominated derivatives of organic compounds are formed during disinfection of water of elevated bromides content. All THMs are highly toxic and hardly biodegradable. Due to their ability to accumulate in living cells they reveal carcinogenic, mutagenic and teratogenic effects (Wang et al. 2007). Membrane techniques, especially RO and NF, can be used to remove THMs from waters (Bodzek 2013, 2015). The studies on the removal of THMs from water with the use of RO and NF membranes by Osmonics (SS10 and MQ16) revealed that retention coefficient depended mainly on membrane flux (Table 7) (Bodzek 2015, 2013, Waniek et al. 2002). It was also found that the retention coefficient increased with THMs molecular weight increase according to a series: $\text{CHCl}_3 < \text{CHBrCl}_2 < \text{CHBr}_3 < \text{CHBr}_2\text{Cl}$ (Table 7).

During other studies on the use of nanofiltration to THMs removal by means of NF200 and DS5 (Uyak et al. 2008) membranes it was found that the increase of transmembrane pressure resulted in the increase of membrane flux, while removal rate of THMs was insignificantly affected (Fig. 3). Moreover, NF200 membrane was found to be more suitable for THMs removal than DS5 membrane. It was also shown that THMs of higher molecular mass were rejected more effectively

Table 6. Organic DBPs and oxidation of impurities and admixtures present in natural waters

Disinfectant	Organic DBPs
Chlorine	Trihalomethanes, haloacetic acids, halocetonitriles, haloaldehydes, haloketones, halopicrates, nitroso-dimethylamine, 3-chloro-4-(dichloromethyl)-5-hydroxy-2(5H)-furanone (MX)
Chlorine dioxide	Aldehydes, carboxylic acids
Ozone	Aldehydes, carboxylic acids, aldo- and ketoacids

Table 7. Retention coefficients of THMs for the RO and NF processes

Osmonics membranes	Concentration in raw water [$\mu\text{g}/\text{L}$]	CHCl_3	CHBrCl_2	CHBr_2Cl	CHBr_3
NF MQ16	10–100	83–87	88.5–96.5	90.5	92
RO SS10	10–100	67–81	65–81	57–65	61–80

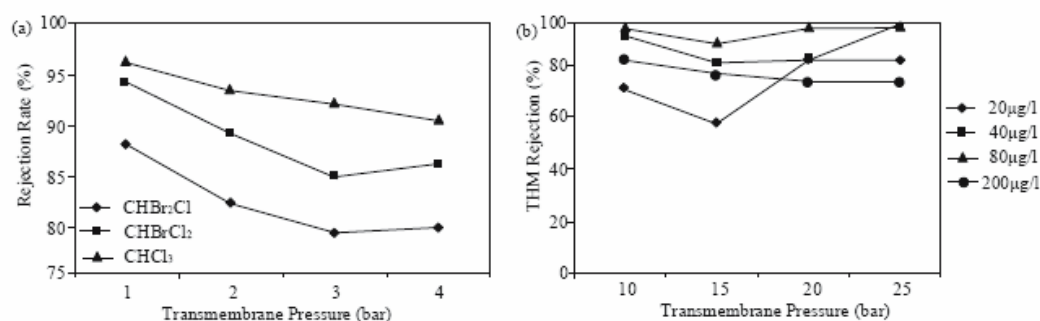


Fig. 3. Influence of transmembrane pressure, concentration and THM type on retention coefficient of THMs, (a) NF200 membrane and (b) DS5 membrane

according to a series: $\text{CHCl}_3 < \text{CHBrCl}_2 < \text{CHBr}_2\text{Cl}$ (Uyak et al. 2008). The highest retention rate observed for CHBr_2Cl resulted from the higher molecular mass of bromine than chlorine and thus, of higher molecular mass of the CHBr_2Cl than other THMs.

Xu et al. (2005) performed studies on the retention of chloroform and CHBr_3 using NF-90, XLE and TFC-HR membranes (Koch Membrane Systems). At the beginning of the process the retention of both substances was similar for all investigated membranes and reached 90% for CHBr_3 (253 Da) and 80% for CHCl_3 (119 Da). The difference in retention resulted from the fact that CHBr_3 was more hydrophobic ($\log K_{\text{ow}} = 2.40$) than CHCl_3 ($\log K_{\text{ow}} = 1.97$) and its removal was additionally improved by hydrophobic-hydrophilic interactions with membrane surface. After ca. 3 h of filtration, the retention of all membranes significantly decreased and reached values ranging from 20 to 35% for CHCl_3 and from 35 to 45% for CHBr_3 . TFC-HR membrane characterized with lowest retention rate, whereas XLE membrane, as the more hydrophobic one, revealed the highest efficiency. It resulted from the fact that hydrophobic XLE membrane enabled the adsorption of hydrophobic contaminants on its surface, hence the overall efficiency was better than in the case of other membranes. Yaman and Çakmakçı (2016) in order to remove the organic matter and THMs, ozone and membrane process were performed. The comparison of the treatment methods used during the study showed that the highest removal efficiency of 76% THMFP, 21% UV and 44% DOC was possible with the combination of ozone+ membrane system.

Except for THMs, water chlorination can lead to formation of **haloacetic acids – HAA**. Main representatives of this group of contaminants are: chloroacetic acid (CH_2ClCOOH), bromoacetic acid (CH_2BrCOOH), dichloroacetic acid (CHCl_2COOH), trichloroacetic acid (CCl_3COOH) and dibromoacetic acid (CHBr_2COOH). Additionally, the presence of tribromoacetic acid (CBr_3COOH), bromochloroacetic acid (CHBrClCOOH), dibromochloroacetic acid ($\text{CBr}_2\text{ClCOOH}$) and dichlorobromoacetic acid ($\text{CCl}_2\text{BrCOOH}$) has also been confirmed (Kowalska et al. 2011). Moreover, for example, dichloroacetic acid and trichloroacetic acid are found to be carcinogenic. According to the EPA, increased risk of cancer is a result of long-term consumption of water with levels of HAA's that exceeds 0.06 mg/L in water. Similarly as in the case of THMs, the removal of HAAs from water can be performed by RO and NF. The studies on the removal of five HAA by means of NF revealed that membranes of compact, negatively charged structures (e.g. aromatic polyamide ES10 membrane) were more efficient than more open membranes of negative/neutral surface charge (Chalatip et al. 2009). It was caused by both, higher repulsing forces (Donnan exclusion) and sieving effect. Very high efficiency, ranging from 90 – 100% was already achieved at low transmembrane pressure at a level of 0.1 MPa, and the increase in acids concentration resulted in retention decrease due to more intensive concentration polarization (Chalatip et al. 2009). Yang et al. (2017) investigated the removal of 9 HAAs by four commercial RO and NF membranes. Under typical conditions (pH 7.5 and 50 mM ionic strength), HAA rejections were >60% for NF270 with molecular weight cut-off (MWCO) equal to 266 Da and equal or higher than 90% for XLE, NF90 and SB50 with MWCOs of

96, 118 and 152 Da, respectively, as a result of the combined effects of size exclusion and charge repulsion. A range of studies on the removal of HAA from water in membrane bioreactor with enzymes immobilized on UF membranes was also carried out (Kowalska et al. 2011). Polyamide, flat sheet membrane modified with glutaraldehyde, was used as a support for enzymes. The modification was applied in order to assure the formation of durable covalent bonds between membrane material and a protein. Enzymes used during the process were isolated from species of bacteria present in active sludge. The study with the use of five HAAs (CH_2ClCOOH , CHCl_2COOH , CCl_3COOH , CH_2BrCOOH , CHBr_2COOH) mixture of concentration 1 mg/L each showed that the use of optimal process parameters assured the complete removal of contaminants within 6 hours (Kowalska et al. 2011).

Contamination of water with **bromates (BrO_3^-)** is usually caused by the formation of disinfection byproducts (DBPs) during ozonation of water containing bromides (Br^-), which are firstly oxidized to hypobromites (BrO^-) and then to bromates (BrO_3^-) (Bodzek et al. 2011, Bodzek and Konieczny 2011, Wisniewski et al. 2013). Their concentration in fresh water usually varies from 15 to 200 $\mu\text{g/L}$ and it is higher in ground waters and brackish waters. Bromates (BrO_3^-) have been classified by International Agency for Research on Cancer to 2B group, i.e. compounds possibly carcinogenic to humans (Butler et al. 2005). It indicates the necessity of bromides (DBPs precursors) removal and other bromooxy ions from potable water. The decrease of bromates concentration in water can be achieved by three main methods (Bodzek and Konieczny 2011):

- removal of bromates precursors, i.e. bromides and natural organic matter before ozonation,
- monitoring of bromates formation during ozonation by pH adjustment at low ranges, ammonia or hydrogen peroxide addition and technological modification of ozonation process,
- removal of bromates after ozonation.

Among methods dedicated to bromates removal from water one may distinguish UV radiation (100–400 nm), photocatalysis, coagulation and application of anion-exchange resins (Butler et al. 2005). Biological process with denitrification bacteria may also be used (Butler et al. 2005). Activated carbon adsorption (Huang and Cheng 2008) and membrane processes (Bodzek and Konieczny 2011) can also be successfully applied.

Reverse osmosis and nanofiltration are the most popular membrane techniques involved in the removal of bromates from water. The removal rate of contaminant observed for NF membranes varies from 75–100% at initial concentration 285 $\mu\text{g/L}$, while for RO it is usually at the level of 97% (Butler et al. 2005, Bodzek and Konieczny 2011, Bodzek 2013, 2015). It has been found that NF is more cost effective than RO, mainly due to the lower transmembrane pressure required. For surface water treatment, membrane processes should be applied before ozonation (Bodzek and Konieczny 2011). Moreover, such an arrangement enables minimization of bromates and bromo-derivatives formation during further water treatment. The removal of bromide and bromate anions by hybrid coagulation-NF technique was systematically investigated by Listiarini et al. (2010). Two types of membranes (NF-270

and NF-90) and two types of coagulants (alum and ferrous sulphate) were investigated with regard to humic acid, bromide and bromate removals. It was found that bromide could not be effectively removed by NF, coagulation, or hybrid coagulation-NF, whereas bromate was reduced to bromide when ferrous sulphate was used. Moslemi et al. (2012) investigated effects of pH and the addition of calcium chloride (CaCl_2) on bromate (BrO_3^-) and bromide (Br^-) rejection by a ceramic membrane. Rejection of both ions increased together with pH. At pH 8, the rejection of BrO_3^- and Br^- was 68% and 63%, respectively. Donnan exclusion appears to play an important role in determining rejection of BrO_3^- and Br^- . In the presence of CaCl_2 , rejection of BrO_3^- and Br^- ions was greatly reduced, confirming the importance of electrostatic interactions in determining rejection of BrO_3^- and Br^- . The effect of Ca^{2+} is so pronounced that in most natural waters, rejection of both BrO_3^- and Br^- by the membrane would be extremely small.

Electrodialysis (ED), especially its reversal mode (EDR), is also proposed for bromates removal from water (Wisniewski and Kliber 2010). Studies on ED with anion exchange membrane (Neosepta AMX) revealed efficient removal of bromates at a level of 86–87%, while in the case of monoanion selective membrane (Neosepta ACS) even 99% retention was obtained at current density 20 A/m². The effectiveness of the process obtained for bromates indicates that ED of water of initial contaminant concentration 100 µg BrO_3^- /L results in the production of water of final bromates concentration 1 µg BrO_3^- /L, which is far below the permissible level (10 µg BrO_3^- /L). For bromates removal, Donnan dialysis (DD) is also proposed with the use of anion exchange membrane. The membrane separates feed solution (which contains anions that need to be removed) and receiving solution (which is usually solution of NaCl at concentration up to 1 mol/L) (Wisniewski et al. 2013, Bodzek and Konieczny 2011). Bromates present in feed solution are substituted with neutral anions from receiving solutions, in this particular case with chlorides. The method can be applied to remove anions (bromates, nitrates) harmful to human health, but also the ones which bring difficulties during water desalination (carbonates, sulphates). DD enables the efficient decrease of bromates even from high initial concentrations 500 µg/L (after ozonation concentration <100 µg/L) to 18 µg/L (Wisniewski and Kliber 2009). In such a case, the retention rate of bromates is 96%, what indicates the suitability of the process to the treatment of water contaminated with this substance.

Microorganisms in wastewater

It is commonly known that raw municipal wastewater contains many pathogenic and opportunistic microorganisms, including those resistant to antibiotics, mainly of fecal origin. The presence of pathogenic microorganisms in wastewater is dangerous, as they may lead to epidemic effects, allergic reactions, toxic or immunotoxic health interactions in humans, animals, and other environmental species. Despite very high, reaching 99% reduction of bacteria in wastewater during its treatment, wastewater treatment plant effluent may still contain from 10⁵ to 10⁷/mL of indicator fecal coliform (Campos et al. 2016, Olanczuk-Neyman 2001). Among all waterborne pathogens, viruses have the smallest size and therefore are the hardest to be removed by sedimentation and filtration.

Though disinfection has been adopted for pathogen removal, neither UV nor chlorine achieved satisfactory virus removal in wastewater treatment (Simmons and Xagorarakis 2011). This observed insufficient virus removal could pose a threat to public health (Vergara et al. 2016).

On the basis of the literature data (Michalkiewicz et al. 2011) one may find that mechanical treatment processes enable the decrease of total number of bacteria by 20% and of *Salmonella* species and tuberculosis mycobacteria by 90%, whereas for biological wastewater treatment by means of activated sludge, removal efficiencies reach 90–98% for total number of bacteria, 55–98% for *Salmonella* species and 45% for tuberculosis mycobacteria. Conventional wastewater disinfection, including decomposition of byproducts which are formed during the treatment, is known as specific disinfection. It can be run using physical and chemical methods. Among former techniques one can distinguish: pasteurization, UV irradiation, thermal drying, ionization radiation, ultrasonication and membrane filtration (UF/MF). Chemical methods of disinfection rely mainly on the addition of oxidizing compounds, such as chlorocompounds (chlorine, sodium hypochlorite, bleaching powder, calcium hypochlorite, chlorine dioxide, calcium hydroxide or oxide), ozone, peracetic acid or on the application of alternative techniques. The issue related with the presence of pathogenic microorganisms in biologically treated wastewater has become important due to their reuse or water reclamation in industry and agriculture (Li et al. 2013, Zanetti et al. 2010).

Similarly as in the case of water treatment, MF and UF are mentioned to be suitable physical wastewater disinfection technique, as they guarantee high removal rate of viruses and microorganisms, colloids, suspensions and high molecular weight organic compounds (Bodzek 2013, 2015, Michalkiewicz et al. 2011). In the case of wastewater disinfection, membranes of pore size 0.2 µm are sufficient enough to assure the effective process performance. Such a disinfection method is especially recommended for existing wastewater treatment plants. Quant et al. (2009) have run studies on the use of UF (200 kDa) to remove bacteria from biologically treated wastewater. The rate of bacteria rejection was found to decrease during the process run and at the final stage single bacteria cells (4–5) appeared in the permeate (filtrate). The decrease in process efficiency was probably caused by physical changes of membrane surface occurring during the filtration. Jastrzębski and Ilnicki (2016) performed a series of pilot studies on membrane filtration at one of wastewater treatment plant localized in Southern Poland. The pilot installation was fed with treated wastewater which characterized with significant microbiological contamination. Three types of capillary membrane modules were tested: two MF modules of pore size 0.1 µm (USV and UNA), and one UF module (LOV) of molecular weight cut-off 80 kDa. The summary of obtained results is shown in Table 8. In the case of USV module, commercially dedicated to potable water disinfection, complete removal of all analyzed pathogenic microorganisms was obtained despite their high content in feed stream (Table 8). The significant reduction of total number of microorganisms, especially those determined at 37°C, was also reached. As it was assumed, the application of UF LOV module resulted in better effect concerning the removal of microbiological contaminants. MF UNA module,

commercially applied to filtration of water for industrial purposes, also enabled complete removal of pathogens and satisfactory reduction of total number of microorganisms.

A study on the removal of fecal bacteria, *E. Coli* and *Enterococci* from municipal wastewater with the use of MF membranes of pore size range from 0.2 to 0.8 μm (Osmonics, Pall and Millipore) was also carried out (Modise 2003). It was found that membranes of pores size below 0.45 μm were suitable to remove examined bacteria to the level that met standards established in proper regulations. Another study (Koltuniewicz and Drioli 2008) proved significant removal of coli bacteria from municipal wastewater after primary and secondary treatment with the use of MF membranes of pore size 0.45 μm and 1.2 μm . The removal rate equal to 4.8 log was obtained for wastewater after primary treatment and 4.1 log for wastewater after secondary treatment when 0.45 μm membrane was used, while for more open 1.2 μm membrane those rates were equal 2.3 log and 3.3 log, respectively, for wastewater after primary and secondary treatment. Such high rejection observed for 1.2 μm membrane was explained by formation of filtration layer on the membrane surface, which acted as a secondary skin layer and improved the removal efficiency. Similar studies carried out for municipal wastewater with polypropylene membranes of pore size range from 0.2–1.2 μm indicated a significant decrease in removal efficiency of coli bacteria in the case of membranes of pore size above 0.67 μm (Koltuniewicz and Drioli 2008).

Membrane bioreactors (MBR) can be an example of direct use of MF/UF membrane to wastewater treatment, including disinfection. MBR integrates biological reaction/transformation processes with membrane separation (Noworyta and Trusek-

-Holownia 2006, Trusek-Holownia 2009, Szewczyk 2009, Deowan et al. 2015). Two main configurations of membrane bioreactors are available for industrial scale systems: devices in which membrane module is immersed in the reactor chamber and devices in which membrane module is separated from the reactor (Fig. 4) (Noworyta and Trusek-Holownia 2006, Deowan et al. 2015). Such solutions are applied at municipal wastewater treatment plants as well as at industrial wastewater treatment plants (Noworyta and Trusek-Holownia 2006). A range of advantages of MBR in refer to conventional systems can be mentioned, and among them the most important are: higher biomass concentration, higher solid retention time (SRT), and higher purity of treated wastewaters (Szewczyk 2009, Deowan et al. 2015). When well designed and operated, MBRs can consistently achieve efficient removals of suspended solids, protozoa and coliform bacteria. Under optimal conditions, MBR systems can also significantly remove various viruses and phages (Hai et al. 2014). Virus removal in water reuse should not solely rely on disinfection. In full-scale wastewater treatment plants, the contribution of secondary treatments on virus removal is much larger than that of disinfection, probably due to the high concentration of nutrients in wastewater increasing the consumption of disinfectants (Simmons and Xagorarakis 2011). Unlike disinfection, the improvement of virus removal in the secondary treatment does not rely on augmenting the disinfectant dosage. Hence, effective disinfection of wastewater is assured.

Table 9 shows MS2 phage removal by different membranes most frequently used in MBR. Direct MF may only achieve around one log removal of virus, while with the common UF membranes, which can be generally considered to be equivalent

Table 8. Results of microbiological tests on sewage treated biologically and after membrane filtration

Bacteria	Raw sewage	Permeate, MF UNA module	Permeate, MF USV module	Permeate, UF module
Amount of bacteria in the given wastewater volume				
Total number of bacteria 37°C, 1 mL	10 ⁵	10–500	5	1
Total number of bacteria 20°C, 1 mL	10 ⁵	30–750	10–50	2
Coli bacteria, 100 mL	10 ⁴	0	0	0
<i>E. Coli</i> , Faecal coliform 100 mL	10 ⁴	0	0	0
<i>Enterococci</i> , 100 mL	10 ⁴	0	0	0
Salmonella, 100 mL	00	0	0	0

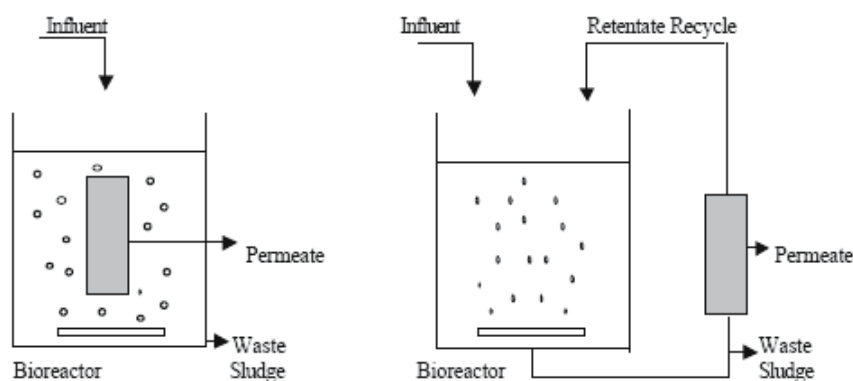


Fig. 4. Bioreactor with immersed and external membrane module

to 200 kDa, variable log removal of virus depending on factors, such as membrane pore size and material may be achieved (Hai et al. 2014).

Shang et al. (2005) and Hai et al. (2014) obtained the removal of *E. Coli*, fecal coliforms and fecal streptococci, *Salmonella* and other pathogenic indicators by MBR at levels acceptable for drinking water. Francy et al. (2012) examined the effectiveness of MBR in the removal of microorganisms from wastewater by two full scale MBR plants, each with a capacity of 12,900 m³/d, both using 0.4 µm chlorinated-polyethylene membranes. The study found that for all MBR samples there was almost complete removal of bacteria. The recorded concentrations of the indicator organism *E. Coli* and fecal coliforms in the treated wastewater were within the standards for reuse for urban and agricultural purposes, with many of the samples having values of less than 1 CFU/100 mL. In Table 10 the removal of microorganisms obtained for various MBR installations supplied by different producers and

for a number of membranes and MBR systems is presented (Hai et al. 2014, Till and Manillia 2001).

Due to the relative size of viruses to the MF and UF membranes commonly used with MBRs, there is much greater attention to virus removal and the implication this has on disinfection than the removal of bacteria or protozoa. Simmons et al. (2011) reported that removal efficiencies could reach 6.3, 6.8, and 4.8 logs for human adenoviruses, enteroviruses, and noroviruses, respectively. Kuo et al. (2010) reported 4.1–5.6 log removals for human adenoviruses, and average of 5.0 ± 0.6 log for the removal of HAdV by MBR. Also Da Silva et al. (2007) obtained high removal efficiencies for noroviruses in a full-scale MBR. Table 11 summarizes the findings of some key case studies regarding the removal of phages and other viruses by MBR (Hai et al. 2014).

The removal efficiency of pathogens from wastewater by MBR is generally higher than that of classical activated sludge (CAS) method and has even been shown to be equivalent to

Table 9. MS2 phage removal by different membranes from spiked deionized water

Membrane Specification	Virus Concentration in Feed (PFU/mL)	LRV
RO (PA-TFC)	10 ⁵ –10 ⁶	>6.5
RO (PA-TFC)	10 ⁵ –10 ⁶	5.6
RO (PA-TFC)	10 ⁵ –10 ⁶	2.7
RO (CA)	10 ⁵ –10 ⁶	>4.9
RO (CA)	10 ⁵ –10 ⁶	4.6
UF 300 kDa (PS)	na	>4
UF 100 kDa (PS)	na	>4
UF 10 kDa (PS)	na	3–4
UF 100 kDa (PES)	10 ³ –10 ⁶	3.54±0.56
UF 150 kDa (PES)	10 ³ –10 ⁶	>4.89
UF 100 kDa (CA)	10 ³ –10 ⁶	>6
MF 0.2 µm (PS)	na	<1
MF 0.1 µm (PVDF)	na	<1
MF 0.1 µm (PVDF)	10 ³ –10 ⁶	1.79±0.09

Notes: PFU = plaque forming unit; LRV = log removal value; na = not available; MF = microfiltration; UF = ultrafiltration; RO = reverse osmosis; PA = polyamide; CA = cellulose acetate; PS = polysulphone; PES = polyether sulphone; PVDF = polyvinylidene fluoride.

Table 10. Removal of microorganisms in different MBR systems

Membrane	Pore size, µm	Average reduction, log	Bacteria/viruses	
MBR:	Polysulfone	0.5	4–6	Σ coli bacteria
	Polysulfone	0.3	5	Σ coli bacteria
	Polyethylene	0.4	6.86	Fecal coliform
	Polyethylene	0.4	>5.83	Fecal streptococci
	Chlorinated polyethylene	0.4	6	Enterococci
	Polyethersulfone	0.05	5.5	Fecal coliforms
	Flat-sheet PVDF/PET	0.08	5.9	Fecal coliforms
	Hollow fibre PVDF	0.1	5.4–5.7	Fecal coliforms
	Hollow fibre PVDF	–	6.7	Total coliforms
	Polysulfone	–	6.1	Total coliforms
Memtec (raw wastewater)	0.2	–	Σ coli bacteria	
Memcor (raw wastewater)	0.2	3.8	Fecal coliform	
Renovexx (raw wastewater)	0.5–1.5	3.3	Fecal coliform	
Stork (purified wastewater)	0.05–0.2	2.5	Fecal coliform	
DOW	0.2	<7	Σ coli bacteria	

a CAS system with a tertiary treatment line (Ottoson et al. 2006). The addition of a membrane to a CAS system to form an MBR treatment system reduces the required footprint of the plant, as the “physical” removal of pathogens by the membrane complements the removal by the “biological process”, which is the only removal mechanism in a CAS operation. Table 12 provides a comprehensive comparison of removal of different viruses by full-scale wastewater treatment plants (WWTP): overall, full-scale MBR plants achieved higher virus removals (Hai et al. 2014).

In Table 13, the comparison of microbiological indicators obtained for conventional active sludge systems and membrane bioreactors is given (Konieczny 2015). The application of membrane as a biomass separator results in partial disinfection of treated wastewater. Additionally, some bacteria, including fecal species and *Enterococci*, are completely rejected by UF membranes. In the case of coli bacteria and other microorganisms, they still appear in permeates, however their concentration is much lower (1.4×10^3) in comparison with

effluent from conventional secondary settlers (1.1×10^5). The obtained results met standards established in proper regulations and the final effluent could be safely deposited to environment. Other studies (Hai et al. 2014, Harb and Hong 2017) revealed a range of advantages of aerobic MBR used to remove bacteria (e.g. *E. Coli*, coli, fecal coliform) from treated wastewater.

Despite high quality and low particulate content in aerobic MBR effluents it has been noticed that 100% rejection of bacteria cannot be obtained for such the system, especially if it is equipped with MF membranes (Konieczny 2015, Jong et al. 2010, van der Akker et al. 2014). The durability in process efficiency ($<10^4$ to $>10^6$ log) indicates the necessity of chlorination of MBR effluents.

Considering limitations of aerobic MBR, anaerobic MBR (AnMBR) systems have become potential technology dedicated to municipal wastewater treatment and disinfection, mainly due to the lower biomass growth, lower energy demand and generation of effluents enriched with nutrients (Harb and Hong 2017). Harb and Hong (2017) carried

Table 11. Indicator virus removal by MBR

Patogen/Indicator	Membrane pore size, μm	Final concentration, CFU/100mL	Average reduction, log
Indigenous phage	0.4	8.8	5.9
Somatic coliphage	0.4	–	2.6–5.6
Indigenous MS2 coliphage	–	–	3.2–4.7
F-specific coliphage	0.4	0–1.26	6
Enterovirus	0.4	–	1.79
Norovirus	0.4	–	1.14
T4 coliphage	0.1 & 0.22	–	1.7–6.4
F-specific phage	0.1	–	3.3–5.7
M2 coliphage	0.4	–	0.4–2.1
Somatic coliphage	0.1	–	3.1–5.8

Table 12. Reported virus removal in full-scale wastewater treatment plants (WWTP)

Virus	Log Removal	
	Conventional WWTP	MBR
Adenovirus	1.3–2.4	3.4–5.6
Enterovirus	0.44–3.6	3.2–6.8
Norovirus I	0.2–2.7	0–5.5
Norovirus II	1.6–3.0	2.3–4.9

Table 13. The results of the microbiological analysis of purified wastewater obtained using MBR Bio-Cel installation coming from the Microdyn Nadir firm

Parameter, cfu/1 ml	Biologically purified wastewater	Purified wastewater from MRB			
		1	3	8	11
Day of the test	–	1	3	8	11
Number of microorganisms colonies at 36°C after 48 h	2×10^5	2×10^3	2.2×10^3	1.5×10^4	>300
Number of microorganisms colonies at 22°C after 72 h	12×10^7	4×10^4	10^4	–	>300
Coliform bacteria	1.1×10^5	4.6×10^3	10^2	1.2×10^2	1.4×10^3
<i>Escherichia coli</i>	0.74×10^3	0	0	0	0
<i>Enterococci</i> (fecal streptococci)	0.36×10^3	0	0	0	0

out a comparative study on application of aerobic MBR operated on industrial scale and anaerobic MBR operated on laboratory scale to municipal wastewater treatment. Both systems were equipped with polymeric MF membranes. The obtained results indicated differences in the removal of particular species of microorganisms, regardless of the MBR system applied. Effluents from both reactors still contained pathogenic, opportunistic microorganisms (e.g. *Pseudomonas*, *Acinetobacter*) in a wide concentration range from <2 log to >5.5 log (Table 14) (Harb and Hong 2017).

All kinds of microorganisms identified in municipal wastewater were also found in AnMBR effluents, whereas the rate of their removal varied from 2.7 log to 5.6 log. The highest retention, above 5 log, was reached for *Acinetobacter*, *Arcobacter*, *Aeromonas* and *Streptococcus*, while the lowest one, below log 3, was observed for *Mycobacterium* and *Legionella*. Among 13 groups of pathogens identified in wastewater feeding the bioreactor, the presence of 5 was confirmed in the effluent, whereas the appearance of remaining 8 was not confirmed (Table 14). The pathogens which were identified in the effluent were *Acinetobacter*, *Aeromonas*, *Arcobacter*, *Pseudomonas* and *Stenotrophomonas*, and their retention rates were 2.5 log, 3.9 log, 2.9 log, 2.5 log and 1.7 log, respectively.

Conclusions

Production of sanitary safe water of stable and high quality with the use of membrane technology is an excellent alternative for conventional disinfection methods, as UF and MF membranes are found to be an effective barrier for pathogenic protozoa cysts, bacteria, and, partially, viruses. The application of membranes in water treatment enables the reduction of chlorine consumption during final disinfection, what is especially recommended for long water distribution systems, in which microbiological quality of water needs to be effectively maintained. Low pressure driven membrane filtration, i.e. MF and UF, can be also applied to biologically treated wastewater disinfection. Membrane bioreactors can be mentioned as an example of direct application of MF/UF to wastewater treatment, including disinfection. Nevertheless, no

membrane system can be considered as an absolute barrier to all microorganisms, as viruses can permeate not only through MF membranes, but also through much more compact ones, due to possible deformations of their cells observed during filtration. The implementation of the membrane systems in water and wastewater disinfection is limited by the phenomenon called fouling i.e. accumulation of organic and/or inorganic substances on the surface and in pores of the membrane (Shi et al. 2014). The intensity of fouling depends on many factors, among which the properties of water, membrane type and parameters are of the greatest importance. It is caused by both, electrostatic repulsive forces between charges of foulants and membrane, and adsorptive properties of membrane material connected with its hydrophobicity and hydrophilicity. Fouling may result in an increase of operational costs, due to an increased energy demand, additional labour for maintenance, cleaning chemical costs, and shorter membrane life. It requires effective and efficient methods for its control and minimization. It may be possible to prevent fouling before its occurrence by methods such as pre-treatment of the feed streams, chemical modification to improve the anti-fouling properties of a membrane, and optimization of the operational conditions. However, periodic membrane cleaning is still currently inevitable. It is indeed an integral part of most membrane processes in modern industries, and must be regularly carried out to remove the fouled materials and restore the productivity of the operation.

Membrane techniques can also be applied to remove disinfection byproducts from aquatic environment. In such cases, high pressure driven membrane processes, i.e. RO and NF are considered, however, for elimination of inorganic DBPs from water ED or Donnan dialysis can be used. The main disadvantage of high pressure membrane processes, beside fouling, is membrane scaling (Bodzek et al. 2018). Scaling causes a decrease in both, membrane capacity and permeate quality, and the intensity of the phenomenon depends on the water recovery rate. When the water recovery rate is higher than 50%, scaling reduces the usefulness of RO for water treatment. The phenomenon may be controlled by the addition of anti-scalants such as polyphosphates or polycarboxylic acids, but even then there are inorganic substances in the water

Table 14. The estimated average number of cells per litre for various types of opportunistic pathogens in the effluents after aerobic and anaerobic MBR

Type	Raw wastewater	Effluent from aerobic MBR		Effluent from AnMBR MBR	
		Number	Log	Number	Log
<i>Mycobacterium</i>	No	1.9×10^1	2.8	No	–
<i>Treponema</i>	3.3×10^4	No	–	No	–
<i>Arcobacter</i>	1.0×10^7	2.7×10^1	5.6	1.2×10^4	2.9
<i>Neisseria</i>	3.4×10^4	3.4×10^4	–	No	–
<i>Acinetobacter</i>	1.4×10^7	1.1×10^2	5.1	4.7×10^4	2.5
<i>Pseudomonas</i>	2.4×10^5	7.7×10^1	3.5	8.1×10^2	2.5
<i>Legionella</i>	1.0×10^4	2.0×10^1	2.7	No	–
<i>Escherichia</i>	9.8×10^4	No	–	No	–
<i>Stenotrophomonas</i>	1.6×10^5	2.2×10^1	3.9	3.0×10^3	1.7
<i>Aeromonas</i>	1.6×10^6	8.3×10^0	5.3	2.3×10^2	3.9
<i>Streptococcus</i>	1.0×10^6	8.5×10^0	5.1	No	–
<i>Enterococcus</i>	No	No	–	No	–
<i>Dialister</i>	3.9×10^5	No	–	No	–

produced which cause fouling. An additional factor which can encourage scaling of the membrane can be the tendency to precipitate sulphate and silica deposits and increase feed water temperature. The use of the reverse osmosis (RO) process in water treatment often requires careful selection of the methods of pre-treatment.

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Membrany w dezynfekcji wody i ścieków – przegląd literatury

Streszczenie: Filtracja membranowa, szczególnie ultrafiltracja (UF) i mikrofiltracja (MF), może wspomóc i polepszyć proces dezynfekcji wody i ścieków oczyszczonych biologicznie, ponieważ membrana stanowi barierę dla wirusów, bakterii i pierwotniaków. Przykładem bezpośredniego zastosowania membran UF/MF do oczyszczania ścieków, w tym ich dezynfekcji, są bioreaktory membranowe. Techniki membranowe stosuje się ponadto do usuwania ze środowiska wodnego ubocznych produktów dezynfekcji (UPD). Wykorzystuje się tutaj przede wszystkim wysokociśnieniowe procesy membranowe, tj. odwróconą osmozę i nanofiltrację, chociaż w przypadku nieorganicznych UPD brane są również pod uwagę elektrodializa i dializa Donnana.