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Can membrane bioreactor be a smart option for water treatment?

Mohd Atiqueuzzaman Khan^a, Huu Hao Ngo^{a*}, Wenshan Guo^a, Yiwen Liu^a, Soon Woong Chang^b, Dinh Duc Nguyen^b, Long Duc Nghiem^a, Heng Liang^c

^a*Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney, Sydney, NSW 2007, Australia*

^b*Department of Environmental Energy & Engineering, Kyonggi University, 442-760, Republic of Korea*

^c*National Engineering Research Center of Urban Water Resources, Harbin Institute of Technology, Harbin, 150090, China*

* Corresponding author: School of Civil and Environmental Engineering, University of Technology, Sydney (UTS), P.O. Box 123, Broadway, NSW 2007, Australia. Tel.: +61 2 9514 2745; E-mail address: ngohuuhao121@gmail.com

Abstract

The gradual increase of organic and inorganic micropollutants in natural and drinking watercourses has posed a greater challenge for current water treatment technologies. Currently established water treatment processes such as activated sludge, microfiltration, reverse osmosis, adsorption, carbon nanotube etc. have a limited range of application, low energy recovery, and cost-intensive operation. Membrane bioreactor has already been utilized as a useful option to remove soluble organics, nutrients, and micropollutants from wastewater. Although currently established Membrane Bioreactors have few limitations, recent developments on this technology have improved its energy efficiency and reduced the operating and maintenance cost. Implementing these research findings in full-scale operation can make this process a favorable option in industrial wastewater treatment.

Keywords Membrane bioreactor, micropollutants, nutrients, fouling, water treatment

1. Introduction

The rapid growth of population and extensive industrialization have accelerated the use of various persistent organic pollutants, surfactants, industrial chemicals and pesticides (Han et al., 2018; Kim et al., 2017; Luo et al., 2014). Removal of these pollutants from wastewater is particularly important as they have adverse effects on human health like bioaccumulation, carcinogenicity, and toxicity. Most of these toxic pollutants are synthetic, ubiquitous, long-range transport potent and can bio-accumulate in the human body (Grisoni et al., 2018; Mangano et al., 2017). Currently, different industries such as textile, chemical, and pharmaceuticals have become the major sources of water pollution. Organic contaminants such as Polycyclic Aromatic Hydrocarbons (PHAs), organochloride

pesticides (OCPs), polychlorinated biphenyls (PCBs), and synthetic chlorinated hydrocarbons are major organic pollutants present in water (Mangano et al., 2017). Even at lower exposure levels, they can pose serious adverse effects on aquatic organisms.

Bisphenol A (2,2-bis 4-hydroxyphenyl propane) is one of the typical endocrine disrupting chemicals; bioaccumulation of 4-chlorophenol in the food chain could have a long-term adverse health effect due to its toxicity (Yu et al., 2017). The conventional water treatment processes such as activated sludge, adsorption, forward osmosis and advanced oxidation have been effective only in removing the common biodegradable compounds or a certain type of pollutant (Chen et al., 2017; Praveen et al., 2016; Zhang et al., 2017). In contrast, membrane bioreactors (MBRs) have been proven effective in removing a wide range of organic and inorganic pollutants including biodegradable organic compounds, antibiotics, pesticides, industrial chemicals and nutrients (Fu et al., 2009; Niwa et al., 2016; Qiu et al., 2013). MBR technology has already been established as a worthy alternative over conventional water treatment processes due to developments in manufacturing low-cost membranes (Chung & Hong, 2017), reduction in operating and maintenance cost (Khan et al., 2016b; Li et al., 2017) and fouling control (Jiang et al., 2017; Tan et al., 2017; Zheng et al., 2018).

The removal efficiency of major biodegradable organics and nutrients is significantly high for MBRs compared to the conventional water treatment processes (Qin et al., 2017; Vergine et al., 2018). However, the removal of minor pollutants like antibiotics, toxicants, pesticides and hydrocarbons have become an important issue as the concentration of these emerging pollutants in different wastewater streams are on the rise. As a result, the applicability of a water treatment process has been equally important besides optimum

performance in common organics removal. In this connection, non-conventional hybrid MBRs could be a potential wastewater treatment technology in the future.

The objective of this review study is to evaluate the performance of conventional water treatment processes in removing organic hydrocarbons, micropollutants, and nutrients. Additionally, the removing efficiency of different emerging pollutants has been compared between conventional bioreactors and common MBR processes. Finally, the future perspective of membrane bioreactor has been discussed based on their current drawbacks in process operation and energy efficiency.

2. Emerging pollutants (EPs) in natural watercourses and drinking water

Over the past few decades, little attention has been given to control the level of emerging pollutants as the regulatory lists of environmental pollutants do not include them.

Different analytical methods have been developed to identify different groups of emerging contaminants. Emerging pollutants could be classified into two major categories: The common biodegradable organic pollutants and nutrients make the first group whereas organic and inorganic micropollutants make the second group of emerging pollutants.

2.1 Organic pollutants and nutrients

The presence of biodegradable hydrocarbons, nitrates (NO_3^-), ammonia (NH_4^+) and phosphates (PO_4^{3-}) has been studied for several decades and the current water treatment technologies have already been optimized to achieve maximum removal efficiency for

these pollutants. Still, there are signs of severe organic and nutrient pollution around the world.

In China, due to eutrophication, annual occurrence rates of red tide (discoloration of seawater caused by a bloom of toxic red dinoflagellates) has increased up to 250 times recently (Xu et al., 2011). Sidek et al. (2016) studied the performance of gross pollutant trap for water quality preservation at Klang River basin, Malaysia. The study found out BOD₅ and COD in the downstream of the river were 23-170 mg/L and 62-304 mg/l respectively. The values indicated the water quality between “polluted and average” where supply-extensive treatment is required. Sagbo et al. (2008) carried out a research study that stated 2/3 of the total water in China are not acceptable for drinking purposes due to a high concentration of COD (e.g. COD ~ 10 mg/L) and nutrients (e.g. NO³⁻ > 100 mg/L). Current statistics have shown that more than 50% lake in China is classified as Class IV (eutrophic polluted water by nutrients, not suitable for drinking or bathing) or VI (not suitable for human contact) (Maryna et al., 2016).

The concentration of nutrients in water courses are also on the increase in different parts of the world. Research study from Suwarno et al. (2014) quantified inputs of nutrients from human excrements to 19 rivers in Indonesia and calculated an increase in N and P inputs to Indonesian rivers with a factor of 17–40 between 2000 and 2050. According to Global Orchestration (GO) scenario of the Millennium Ecosystem Assessment (MA), river export of N and P from human waste in South Asia are projected to increase by 20% and 33% between 2000 and 2050. However, the current projection in estimating nutrient concentration in wastewater clearly underestimates the associated level of water pollution as open defecation has not been accounted for most of the studies; for example, only in

India about 80% of the rural population practice open defecation that increases the level of nutrient pollution in water (Amin et al., 2017).

Therefore, in certain areas of the world, there is an alarming increase of COD, BOD, total nitrogen (TN) and total phosphorus (TP) in water. To combat this situation, up-gradation and modification of current water treatment processes are required through process optimization, improvement in removal efficiency and reduction in the cost of operation and maintenance.

2.2 Micropollutants

Micropollutants are mainly synthetic chemicals that are not commonly monitored in the environment but have the potential to enter into the air, water and/or soil and cause adverse effects to human health or the environment. Most emerging micropollutants have not been included into international monitoring programs and therefore the safe limit of exposure and eco-toxicological effects are yet to be identified (Geissen et al., 2015).

Micropollutants can be classified into Polycyclic aromatic hydrocarbons (PAHs), and Polychlorinated biphenyls (PCBs), Organochlorine pesticides (OCPs), Perfluorinated compounds (PFCs) and pharmaceutical compounds.

2.2.1. Polycyclic aromatic hydrocarbons (PAH)

Polycyclic aromatic hydrocarbons (PAHs) can be particularly damaging to the environment for their high toxicity. It is also suspected to cause damage to living organisms by acute toxicity (Han & Currell, 2017). The generation of PAH involves combustion of fossil fuels in vehicles, combustion of coal and wood, power generation

from fuel oil, coal etc. (Deblonde et al., 2011). According to US EPA, the threshold limit for PAH is 200 ng/l (National Primary Drinking Water Regulations) and EU standard for maximum safe PAH concentration is 100 ng/L (Drinking Water Directive 98/83/EC) (Han & Currell, 2017).

The current value of PAH concentration in some river is on the increase, in some part of Asia the value is already alarming. Minjiang and Hangzhou river Estuary in China has a mean concentration of 72,400 ng/L and 52,200 ng/L whereas in Gumti River in India the mean concentration is about 10,330 ng/L. PAH concentration at this level can be responsible to cause acute toxicity for certain organisms (Han & Currell, 2017; Lofthus et al., 2018)

2.2.2. Polychlorobiphenyls (PCBs)

Polychlorobiphenyls (PCBs) are chlorinated aromatic compounds with properties of long-range transport capability, bioaccumulation, and persistence in the air, water, and soil. They also can be a potential threat to human health through bioaccumulation (Deblonde et al., 2011). Different chemicals and industrial byproducts such as paints, plastics, heat exchange fluids, dyed paper, sealants contain PCB (Han & Currell, 2017).

Once PCBs are released into the environment, it remains stable and enters into the food chain in many parts of the world. US EPA-National Recommended Water Quality specifies the safe limit of PCB as 14 ng/L whereas average concentrations of 3110 ng/L and 3161 ng/L have been found for in surface water and seawater in China. The same study reported that seawater from Kallrigafjarden Harbor (Switzerland), Marmara Sea

(Izmit Bay, Turkey) also have PCB concentrations above 14 ng/L (Deblonde et al., 2011; Han & Currell, 2017).

2.2.3. Organochlorine pesticides (OCPs), Perfluorinated compounds (PFCs)

Dichlorodiphenyltrichloroethane (DDT) and hexachloro cyclohexane (HCH) are the two major components of organochlorine pesticides (OCPs) that are toxic, carcinogenic and harmful to the environment. They were extensively used as pesticide until 1983. DDTs generally have higher residual levels in water systems due to a low rate of degradation. Concentrations in surface water in Asia and the Middle East are high such as such as Hanoi BDL – 324 ng/L in Hanoi (Vietnam), 388 ng/L at Yamuna (India), 128–239 ng/L in El-Rahawy area (Egypt) against the safe limit of 25 ng/L by EU and UK EA (Han & Currell, 2017).

Perfluorinated compounds (PFCs) mainly includes erfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) pollution. US EPA included PFOS and PFOA as persistent organic pollutants in 2012 and specified safe threshold limit of 200 ng/L (EPA, 2012). PFC concentrations around the world are on the increase too. Review study from Han & Currell (2017) shows Ectobicoke Creek in Canada (mean concentration 106302 ng/L), Moehne River in Germany (mean concentration 6639 ng/L), seawater near Japan (range = 2 – 448 ng/L) and South Korea (range = 0.2 – 320 ng/L) have high level of PFC concentrations. Table 1 lists some common micropollutants and their concentration in the aquatic environment in different countries of the world.

Table 1: Common micropollutants and their concentration in aquatic environment in different countries.

2.2.4. Pharmaceutical Compounds

The major risk involved in pharmaceutical compounds is all of them have not been added to the list of micropollutants; therefore, their safe exposure limit and potential impact on human health and environment are yet to be identified. The pharmaceutical compounds are grouped into several classes such as antibiotics, analgesics, anti-inflammatory, anti-epileptic, statins, antidepressants, anti-cancer agents, hormones; disinfectants etc. (Deblonde et al., 2011). Currently, a few wastewater treatment processes have been optimized to remove different types of pharmaceutical compounds from wastewater (Alvarino et al., 2016; Löwenberg et al., 2014; Zhang et al., 2015). Table 2 lists occurrence and concentration data of various pharmaceuticals compounds in different areas of the world.

Table 2. Occurrence and concentration data of various pharmaceuticals compounds
(Modified from (Pal et al., 2010))

3. Limitations of conventional water treatment technologies to tackle EPs

The performance of conventional water treatment technologies could be evaluated based on the cost involved in process operation and maintenance, removal efficiency, and the range of contaminants that can be effectively removed. The following section contains discussion about a few conventional waste treatment processes and their performance in removing the emerging pollutants from water streams.

Conventional Activated Sludge (CAS) technology has been widely applied for treating both domestic and industrial wastewater for both COD and nutrient removal. However, CAS has technical limitations such as low COD removal (Araneda et al., 2017), long retention time and most importantly the dependence on active microorganisms to perform the biodegradation (Guo et al., 2017). Additionally, poor sludge settling and carryover of biological solids are common operating problems that are evident due to sludge bulking in CAS (excessive growth of filamentous microorganisms) (Zhang et al., 2017). The application of CAS process for treating different types of wastewater treatment has been limited to the case where the pollutants are easily biodegradable. Compared to other conventional water treatment process, activated sludge method has not been applied in removing different types of micropollutants from wastewater.

The combination of microfiltration and reverse osmosis system has been proven effective Pharmaceuticals and pesticides in reclaimed water. But the removal rates reached up to 85-100% only for a few selected analgesics and anti-inflammatories (Rodriguez-Mozaz et al., 2015). The same study also added that pollutants like propyphenazone, propranolol, sotalol, carbamazepine, and diclofenac were poorly or not removed at all by the MF/RO process. Another study by Al-Rifai et al. (2011) was carried out to investigate the occurrence, persistence, and range of micropollutants at different processing points at a full-scale water recycling plant (WRP) in Queensland, Australia. The research findings indicated removal efficiency ranged from 97 to 74% but a complete non-removal of Bisphenol A from the wastewater stream.

It is particularly challenging to evaluate the cost assessment and technical advantage of MF on the RO operation, since the cost associated with RO fouling is site-specific and largely depends on the concentration of organic pollutants present in the wastewater. Garcia et al. (2013) performed a Scale-up economic assessment and experimental analysis of MF–RO integrated membrane systems and found out an operating cost of 0.17–0.19 GBP/m³ for municipal wastewater reuse using MF-RO process. Regardless the operating and maintenance cost involved in MF/RO process is quite evident that the process could not be applicable for removing certain compounds like Bisphenol A, propyphenazone, propranolol, carbamazepine, and diclofenac.

Chemical adsorption process has the advantage to remove both biodegradable and non-biodegradable pollutants from wastewater (Álvarez-Torrellas et al., 2017; Chang et al., 2016; Natarajan et al., 2018) where the removal efficiency for total organic carbon, total nitrogen, and carbonates (CO_3^{2-}) can go up to 98.8%. But the major drawback of the conventional adsorption process lies in the design process of adsorbents. Different type of adsorbent materials can only remove one particular group of pollutants. Carbon adsorbents have been particularly suitable for removing pharmaceutical pollutants like carbamazepine, ciprofloxacin and organic carbon (Álvarez-Torrellas et al., 2017). Additionally, Macroporous adsorption resin (MAR) has been utilized to remove adsorbing soluble microbial products (SMP) (Chen et al., 2017) whereas, hydrogel-based adsorption process has been developed to remove high-concentration heavy metals from industrial wastewater.

In case of nutrient removal from wastewater, Kim et al. (2016) revealed that NO_3^- is adsorbed after primary adsorption by PO_4^{3-} due to the fact that it has higher ionization degree compared to NO_3^- . Their experiment used an amine-grafted adsorbent for simultaneous PO_4^{3-} and NO_3^- and found out that the synthesized adsorbent recovered 78% nitrate and 93% phosphate from wastewater. Therefore, it is particularly challenging to design an adsorbent that can completely remove both PO_4^{3-} and NO_3^- from wastewater. Although a large number of studies has been performed to figure out the efficiency in adsorption by removing organic and inorganic pollutants there has not been any single report that provides a full comparative analysis between different adsorption processes. As a result, the adsorption process can be referred to be effective but the application is subjected only to remove a specific type of pollutants. In this connection, designing a hybrid adsorbent to remove a wide range of micropollutants can be potential area of future research.

Electrochemical oxidation process has been applied to remove persistent organic pollutants like dyes, pesticides, pharmaceuticals etc. and it can be applied as standalone treatment process or coupled with biological treatment, electrocoagulation or membrane filtration (Moreira et al., 2017). In this connection, Boczkaj & Fernandes (2017) calculated the total cost for operation and maintenance in advanced oxidation processes. The results showed that the cheapest value was 90 \$ to treat 10 m^3 wastewater referring a value of \$7 million per year to treat around 14400 m^3 annually. (Ioannou-Ttofa et al., 2017) performed an economic evaluation for OMW wastewater using advanced oxidation process. The experiment was carried out in three separate bioreactor arrangements and the lowest value in initial installation and cost indicated 238,000 € (for treatment capacity of

180 m³ OMW/d), operation and maintenance cost for the same plant was 751,000 € (for treatment capacity 180 m³/d and duration: 42 days). Hence, no valid conclusion could be made since direct comparison of capital, operation and maintenance were not available from these studies. Still, the costs available from individual experiments could be utilized in future case of comparison.

Carbon nanotube (CNT) is one of the most promising water purification technologies that have been applied in removing organic pollutants (Qu et al., 2016; Un & Temel, 2018), heavy metals, (Luzardo et al., 2017) and antibiotics from wastewater (Ncibi & Sillanpää, 2015). Through a number of experiments, it has identified that nanomaterials can be effective in desalination, removing a various type of dyes and halogenated compounds (Das et al., 2014). Again, like the previously discussed technologies, the major limitation for carbon nanotube technology is a limited range of application; one type of nanomaterial can only be effective in removing one particular type of pollutant from wastewater. Apart from technical feasibility, the economic assessment also needs to be performed when it is applied in full-scale operation for treating industrial and domestic wastewater. Additional limitations of this technology include thermal instability, high operating pressure, precipitation, fouling, low rate of influx, slow rate of reaction and formation of toxic intermediates (Das et al., 2014). Additionally, disposal of nanomaterial and relevant effects on the environment are also important issues for carbon nanotube technology.

4. MBRs in removing EPs from water

Over the past few years, Membrane Bioreactors (MBRs) with different design and configurations have been well-established in full -scale industrial and municipal wastewater treatment plants (Huang & Lee, 2015; Krzeminski et al., 2017; Li et al., 2018). The technology has already come into the matured stage through process optimization (Di

Bella et al., 2013; Khan et al., 2016a), novel configurations, energy reduction and fouling control (Krzeminski et al., 2017; Yang et al., 2017). This section of this review study includes the performance highlights and comparison of MBR performance with some other conventional methods in wastewater treatment.

4.1 Application of submerged MBR

Submerged membrane bioreactors have the advantages of small footprint, high removal efficiency, simple flow configuration and the ability to handle a high biomass concentration without the potential of sludge settling (Li & Chu, 2003). As a single stage bioreactor, the initial cost of investment is relatively low compared to the two-stage bioreactors as the capital cost for the membrane, and the additional stage is less compared to the multiple-stage assembly (Khan et al., 2016b).

Different submerged MBR models have achieved high removal efficiency for both organic and inorganic pollutants with issues in process operation such as severe membrane fouling, reactor acidity, low organic the loading rate etc. (Garcia et al., 2013; Krzeminski et al., 2017; Li & Chu, 2003). These operating issues in submerged MBRs have been resolved due to the recent developments in fouling control, cost reduction, and process optimization. (Huang et al., 2013; Martinez-Sosa et al., 2011; Qin et al., 2015). Table 3 lists the removal efficiency of submerged MBR in contrast to the CAS process.

Table 3: Comparison of different pollutant removal efficiency between submerged MBR and conventional CAS process

4.2 Performance of MBRs in removing nitrate and perchlorate

Different membrane bioreactor systems have been utilized to remove anions like nitrate and perchlorate from wastewater (Van Ginkel et al., 2010). Pressure-driven MBRs can produce the highest production rate of the effluent per unit membrane area whereas ion exchange membrane bioreactors have been utilized for highest removal efficiency (Crespo et al., 2004). Most recently, Sulphur-based mixotrophic MBRs and hybrid membrane bioreactors have been employed for high anion removal and treated water production rate (McAdam & Judd, 2006; Sahinkaya et al., 2017). Table 4 lists the performance of nitrate and perchlorate removal of some hybrid MBR systems.

Table 4: Performance of membrane bioreactors for removing of anions from water (Crespo et al., 2004; McAdam & Judd, 2006; Sahinkaya et al., 2017)

^aDetection limit $\sim 0.01 \text{ mg NO}_3^- \text{ L}^{-1}$; ^bTOC, total organic carbon;

^cMeasured as dissolved organic carbon; ^ddetection limit $\sim 0.5 \text{ mg carbon L}^{-1}$.

4.3 MBR + PAC process for drinking and micro-polluted surface water treatment

Non-conventional membrane bioreactors have been proven effective as they offer the removal of both biodegradable and non-biodegradable pollutants through a combination of biological and physical/chemical treatment processes. MBRs having Powdered Activated Carbon in treating micro-polluted surface water can be utilized to uptake soluble organics and colloids. Different research studies have shown the high removal efficiency of MBR+PAC in treating a wide range of micropollutants from different wastewater streams (Du et al., 2017; Gao et al., 2016; Hu et al., 2014; Zhang et al., 2015).

Addition of PAC with the conventional MBR processes can be beneficial in two different ways. Firstly, it increases the COD, TOC and nutrient removal from wastewater. Sagbo et al. (2008) reported an increase in COD removal from about 51% to 67% when PAC was added for drinking water treatment. According to the study from Zhang et al. (2015), PAC addition increased COD, UV₂₅₄ removal efficiency from 40.5 to 66.6 and 26.4 to 69.5% respectively during surface water treatment. Secondly, organics that do not go through biodegradation can be adsorbed by the powdered activated carbon. Different PAC+MBR systems that have been proven successful in removing micropollutants have been listed in Table 5.

Table 5: Removal efficiency of micropollutants by different PAC+MBR systems.

^a Approximate value (Data acquired from graph)

5. Future Perspective

The recent developments in membrane bioreactor technology include reduction in energy consumption, novel bioreactor arrangements, characterization of membrane fouling and control, commercial development in membrane fabrication and enhanced efficiency in removal of micropollutants. Although the recent developments in MBR are yet to be adopted by the full-scale industrial plants, the current growth of the MBR market has been promising. In 2014, the global MBR market was worth \$425.7 million which is projected to approach \$777.7 million by 2019 with a corresponding five-year compound annual growth rate (CAGR) of 12.8% (Krzeminski et al., 2017). The following section of the review study discusses the recent developments in MBR technology that could contribute to the industrial application of MBR technology.

➤ Compared to Conventional Activated Sludge (CAS) process, MBRs still have higher energy requirements. The average specific energy requirements for MBR operation is ranged from 0.6 to 2.3 KWh that can be further reduced down to 0.4 KWh through process optimization (Brepols, 2010). Utilization of hydraulic capacity such as hydraulic load close to design flow rate, compact membrane module with higher packing density, aeration control strategy can reduce the energy consumption in MBR. The study from Sun et al. (2015) reported a 20% reduction in aeration requirement by applying ammonia based aeration control strategy in full-scale MBRs. According Itokawa et al. (2014), the current annual energy consumption of full-scale MBR is averaged to 0.39 KWh/m³ that is much lower compared to currently available data for energy consumption in MBRs.

➤ Although the configuration and operation of MBR processes are mainly bound by COD removal, the removal of minor pollutants are not just a consequence with no freedom of turning. The primary design aspect of a non-conventional MBR process should be focused on simultaneous removal of soluble organics and micropollutants. Integration of an independent chemical/ physical process along with the biological water treatment process will enable the option of process optimization of these two different processes separately. Therefore, novel membrane bioreactor configurations can be a potential area of research for enhanced removal of nutrients, micropollutants, antibiotics and energy efficiency.

➤ The product spectrum of conventional anaerobic MBR systems can be changed. Recent developments in anaerobic membrane bioreactors have unlocked the technical feasibility to produce biohydrogen and volatile fatty acid in addition to methane-containing biogas production (Khan et al., 2016b). Simultaneous production of biohydrogen and

methane could contribute to improving energy recovery from anaerobic MBRs (Cheng et al., 2017; Intanoo et al., 2014; Sunyoto et al., 2016). Also, the combination of conventional MBR with microbial fuel cell (MFC) has produced cost-effective results. The research study by Wang et al. (2016) showed that MFC an MFC generated electrical field (0.114 V cm^{-1}) could be utilized to reduce the deposition rate of foulants.

➤ Recent developments in membrane fouling control have reduced energy demand and increased removal of nutrients and refractory compounds (Krzeminski et al., 2017; Liao et al., 2018). Application of low amplitude vertical vibration, electrostatic repulsion (Wang et al., 2016), dynamic shear-enhanced filtration through rotation (Wu et al., 2008), cross-flow MBR with rotating ceramic disks (Bentzen et al., 2012) have been proven effective in controlling membrane fouling. In addition to these research findings, the following strategies could be applied for fouling control in full-scale MBR operation (Judd, 2006):

- Application of suitable pre-treatment of feed water
- Permeate back-flushing
- Chemical cleaning and chemical enhanced backwash
- Membrane scouring
- Chemically modifying mixed liquor

➤ New generation membranes and membrane modules have been commercially developed with less energy requirement and higher surface area. For example, Lorain et al. (2010) has proposed a new design that eliminated the buildup of fibrous material that is responsible to block the upper end of the hollow fiber membranes. In 2011, GE WPT introduced Zee Weed membrane that reduces the energy consumption by 30%. Pentair introduced X-flow tubular membrane that is back washable, has 40% increase in

operational flux and 35% reduction in energy requirement during cross-flow MBR operation (Krzeminski et al., 2017).

➤ Results from research studies have shown that MBR can be equally effective in removing a wide spectrum of organic micropollutants. Different MBR configurations have been developed to increase the removal of organics and refractory compounds such as pharmaceuticals. Sun et al. (2013) carried out an experiment using a conventional MBR with post-denitrification process. Results from this experiment indicated OC, TN, and TP removal efficiencies equal to 94%, 85%, and 87% respectively. For antibiotics removal, a combined MBR and UV/TiO₂ photocatalysis process removed up to 95% of carbamazepine (Laera et al., 2011). As the membrane serves to separate suspended particles, more emphasis should be given on biodegradation, nitrification/denitrification, precipitation or adsorption process to increase the overall efficiency of MBR.

6. Conclusion

The continuous increase of emerging pollutants in natural water courses has accelerated the research and development of different water treatment processes. Considering the range of application, removal efficiency, environmental impact and cost of application, MBR technology can be a suitable option to treat wastewater from different industrial and domestic sources. Although some current alternatives in water treatment exceed cost and energy efficiency of MBR technology, the potential developments in bioreactor design and pollutant removal efficiency can make MBR system a smart option for wastewater treatment in near future.

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Table 1. Common micropollutants and their concentration in aquatic environment in different countries.

Table 2. Occurrence and concentration data of various pharmaceuticals compounds
(Modified from (Pal et al., 2010))

Table 3. Comparison of different pollutant removal efficiency between submerged MBR and conventional CAS process

Table 4. Performance of membrane bioreactors for removing of anions from water
(Crespo et al., 2004; McAdam & Judd, 2006; Sahinkaya et al., 2017)

Table 5. Removal efficiency of micropollutants by different PAC+MBR systems.

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Table 1: Common micropollutants and their concentration in aquatic environment in different countries.

Categories	Compound s	Sampling sites	Concentrati on (µg/L)	References
<i>PCP</i>				
Musk fragrance	Galaxolide	Spain, Western Balkan Region	0.03–25	(Santos et al., 2009; Terzić et al., 2008)
Disinfectant	Triclosan	US, Greece, Korea	0.03–23.9	(Behera et al., 2011; Kumar et al., 2010; Pothitou & Voutsas, 2008)
<i>Industrial chemicals</i>				
Plasticizers	Bisphenol A	China, EU- wide, Greece, US	< 0.013– 2.14	(Martin Ruel et al., 2010; Nie et al., 2012; Pothitou & Voutsas, 2008; Yu & Chu, 2009)
Fire retardant	TCEP	EU-wide, Germany	0.06–0.50	(Loos et al., 2013; Reemtsma et al., 2008)
<i>Surfactants</i>	Nonylpheno l	China, Spain, US, Western Balkan Region	< 0.03– 101.6	(Céspedes et al., 2008; Nie et al., 2012; Terzić et al., 2008)
	Octylphenol	Spain, China	< 0.2–8.7	(Céspedes et al., 2008; Nie et al., 2012)
<i>Pesticide</i>				
Herbicide	Atrazine	EU-wide, Greece	0.02–28	(Campo et al., 2013; Loos et al., 2013; Stamatis & Konstantinou, 2013)
Insecticide	Diazinon	EU-wide, Spain	< 0.684	(Campo et al., 2013; Loos et al., 2013; Luo et

Categories	Compound s	Sampling sites	Concentrati on (µg/L)	References
				al., 2014)

Table 2. Occurrence and concentration data of various pharmaceuticals compounds

(Modified from (Pal et al., 2010))

Compounds	Concentration (ng/l) in freshwater, rivers, and canals			Lowest predicted no-effect concentration (PNEC) (ng/l)
	America	Europe	Asia and Australia	
Antibiotics				
Trimethoprim	2 – 212	0 – 78.2	4 – 150	1000
Ciprofloxacin	–	–	23 – 1300	20
Sulfamethoxazole	7 – 211	< 0.5 – 4	1.7 – 2000	20,000
Analgesics and anti-inflammatory				
Naproxen	0 – 135.2	< 0.3 – 146	11–181	37,000
Ibuprofen	0 – 34.0	14 – 44	28 – 360	5000
Ketoprofen	–	< 0.5 –14	< 0.4 – 79.6	15.6 × 10 ⁶
Diclofenac	11 – 82	21– 41	1.1 – 6.8	10,000
Salicylic acid	70 –121	< 0.3 – 302	–	–
Mefenamic acid	–	< 0.3 – 169	< 0.1 – 65.1	–
Acetaminophen	24.7 – 65.2	12 – 777	4.1 – 73	9200

Table 3: Comparison of different pollutant removal efficiency between submerged MBR and conventional CAS process

Components	Removal Efficiency (%) ^a		References
	Submerged MBR	Conventional CAS	
Tannery wastewater			
COD (mg/L)	90.08	70.5 – 82.4	(Zupančič & Jemec, 2010)
Domestic Wastewater			
COD (mg/L)	96.5	70.0	(Stazi & Tomei, 2018)
NH ₄ ⁺ -N (mg/L)	66.8	20.12	(Liu et al., 2018)
Total nitrogen (TN) (mg/L)	81.7,	56.34	
Total phosphorus (TP) (mg/L)	56.4%	-	
Polluted surface water			
TOC (mg/L)	61±12 (30–85)	-	(Habib et al., 2017; Li & Chu, 2003; Umaiyakunja ram & Shanmugam,
NH ₃ -N (mg/L)	98±1 (95–99)	-	
UV ₂₅₄ (/cm)	69±5 (54–86)	-	
THMFP (µg/L)	75±6 (59–85)	-	
CHCl ₃ FP (µg/L)	74±6 (57–86)	-	
CHBrCl ₂ FP (µg/L)	77±5 (62–88)	-	
CHBr ₂ ClFP (µg/L)	86±8 (51–99)	-	

CHBr ₃ FP (µg/L)	98±1 (96–99)	-	2016)
Turbidity (NTU)	4±1 orders (4–5 orders)	-	
Total coliforms (#/mL)	98±1 (96–99)	-	

^a Average values have been considered for multiple results

Table 4: Performance of membrane bioreactors for removing of anions from water

(Crespo et al., 2004; McAdam & Judd, 2006; Sahinkaya et al., 2017)

bioreactor type	Membrane configuration	Anion content (Polluted water) (mg L ⁻¹)	Anion removal rate (Treated water) (g m ⁻² h ⁻¹)	Treated water production rate (L m ⁻² h ⁻¹)	Secondary pollution as TOC ^b (mg L ⁻¹)
Sulfur-based mixotrophic	Polyethersulfone (PES), 0.45 µm pores	50 ± 5	0.5 ± 0.4	15–32	-
Hydrogen Hybrid	Hollow fiber membrane	330	<1	14.3–24.6	-
Pressure-driven	Cellulose hollow fibers	120	<20	11	100
Pressure-driven	Polyvinylidene difluoride	150	<2	3.1	21
Pressure-driven	Polysulfone hollow fibers	148	<1	4.5	80
Gas-transfer	Silicone-coated fibers	73	Not detected ^a	0.8	10
Gas-transfer	Composite hollow fibers	55	4	0.4	8
Gas-transfer	Composite hollow fibers	12	<0.14	0.1	10
		0.1	<0.004	0.9 × 10 ⁻³	-

Ion Exchange MBR	Dense anion- exchange; flat	150	20	1.4	11
Ion Exchange MBR	Dense anion- exchange; flat	60	<3	0.2	3
		0.1	<0.004	0.3×10^{-3}	-

^aDetection limit $\sim 0.01 \text{ mg NO}_3^- \text{ L}^{-1}$; ^bTOC, total organic carbon;

^cMeasured as dissolved organic carbon; ^ddetection limit $\sim 0.5 \text{ mg carbon L}^{-1}$.

Table 5. Removal efficiency of micropollutants by different PAC+MBR systems.

Membrane type	Operating condition	Pollutant	Maximum Removal ^a (%) by PAC-MBR	Reference
Microfiltration (MF) flat sheet membrane 0.45 µm pores	Working volume 30 L; 250 mg/l PAC HRT 24 h	Carbamazepine	98	(Alvarino et al., 2016)
		Diclofenac	85	
		Roxithromycin	>99	
		Trimethoprim	>98	
		Sulfamethoxazole	93	
		Erythromycin	100	
		Naproxen	97	
		ibuprofen	>99	
		Estrone	>99	
		Ethinyl estradiol	>98	
Ultrafiltration (PVDF) 0.08 µm pores	Working volume 4L; 1 g/L PAC	2,4,6-trichloropheno	86	(Zhang et al., 2015)
		Nitrobenzene	90	
		Trichloroethylene	79	
Cellulose membrane filters 0.45 µm pores	Batch volume 100 ml; (0.25, 0.5, 1, 2, 4, 7, 15 mg PAC/mg DOC)	Diclofenac	>95	(Zietzschmann et al., 2016)
		Benzotriazole	>98	

Ultrafiltration	Working volume	Sulfamethoxazole	68	(Löwenberg et al., 2014)
Multibore [®] 40	30 L;	Carbamazepine	97	
nm (PES)	17.1 mg/L PAC	Mecoprop	86	
20 nm pores		Diclofenac	83	
		Benzotriazole	94	

^a Approximate value (Data acquired from graph)