FEATURE ARTICLE

Recent advances in membrane bioreactor technology for wastewater treatment in China

Xia HUANG (✉), Kang XIAO, Yuexiao SHEN

State Key Joint Laboratory of Environmental Simulation and Pollution Control, Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, China

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Abstract Since the introduction of the membrane bioreactor (MBR) in China in the early 1990s, remarkable progress has been achieved on the research and application of this technology. China has now become one of the most active fields in the world in this regard. This review outlines the development of MBR-based processes in China and their performance of treating municipal and industrial wastewaters. Since membrane fouling is a critical operational problem with MBR processes, this paper also proposes updated understanding of fouling mechanisms and strategies of fouling control, which are mainly compiled from publications of Chinese researchers. As for the commercial application of MBR in the country, the latest statistics of large-scale MBR plants $($ > 10000 m³ \cdot d⁻¹) are provided, and the growth trend of total treatment capacity as well as its driving force is analyzed.

Keywords membrane bioreactor (MBR), municipal wastewater treatment, industrial wastewater treatment, membrane fouling, commercial application, China

1 Introduction

The membrane bioreactor (MBR), an innovative combination of membrane technology and biological process for wastewater treatment, has been exhibiting great advantages over the conventional activated sludge (CAS) process owing to its preponderantly higher efficiency for solid-liquid separation than that of a secondary sedimentation tank. The representative advantages of MBR include [\[1](#page-21-0)]: (1) high efficiency for pollutant removal, producing stable and directly reclaimable effluent water; (2) complete separation of sludge from water providing a more stable

Received May 2, 2010; accepted June 5, 2010

and flexible operation mode attributed to the separation of hydraulic retention time (HRT) and sludge retention time (SRT); (3) high concentration of mixed liquor suspended solids (MLSS) giving rise to a higher volumetric load that enables minifying the reactor scale; (4) low sludge production favoring cost saving of subsequent excess sludge disposal; and (5) possible enhancement of contaminant removal ability of microbes in the reactor.

In light of its excellence, MBR has been attracting increasing attention in its research-development and promising application in wastewater treatment and reclamation since its introduction to China at the beginning of the 1990s. To date China has become one of the world's most active and attractive fields for both academic research and commercial application of MBR. Herein we will give an overview of recent advances in both research and application of MBR in China.

2 Research progress

Since MBR was introduced into China in the early 1990s, researches on MBR have been undertaken at an accelerating rate. Figures 1 (a) and (b) show the rough trend of intensified efforts in MBR research in recent years, taking the amount of published papers as an indicator. As can be seen in Fig. 1 (a), the annual papers increased tremendously, especially in the recent 5 years. Figure 1 (b) demonstrates the increasing contribution of China to the world's research progress. Up to the end of 2009, about one fifth of Science Citation Index (SCI) papers and one fourth of Engineering Index (EI) papers on MBR treating wastewater can be credited to China. Therefore, China has become a vital force in the world's MBR research, and will play a more and more important role in the foreseeable future.

The objectives of research on MBR may lie in two aspects: optimization and utilization. The optimization

E-mail: xhuang@tsinghua.edu.cn

Fig. 1 Trend of SCI and EI publications from Chinese institutes on MBR treating wastewater. (a) Trend of the amount of annually published papers from Chinese institutes; (b) trend of the proportion of annually published papers contributed by Chinese institutes (Search conditions: (TS = (membrane bioreactor AND wastewater) AND CU = (China)) and (TS = (membrane bioreactor AND wastewater)) in the database of SCI-EXPANDED for SCI contributed by China and by the whole world, respectively, while ((membrane bioreactor AND wastewater) wn KY AND China wn CO) and ((membrane bioreactor AND wastewater) wn KY) in the database of Compendex for EI contributed by China and by the whole world, respectively. $CO =$ country of origin, $CU =$ country, $KY =$ subject/title/ abstract, $TS = topic$, wn = within)

aims at, for instance, better performance for target pollutants removal or required water production (e.g., better water quality), higher operational efficiency in the time aspect (e.g., higher flux) or spatial aspect (e.g., smaller footprint), lower constructional and operational cost (e.g., enhanced membrane tolerance and reduced membrane replacement), and lower energy consumption during operation (e.g., weaker membrane fouling propensity). Research on the utilization of MBR includes the exploration of new functions, pragmatization, scaling up and operation maintenance.

Considering the above objectives, the recent research

progress in China will be addressed as three parts: the composition of an MBR system, the functionality of MBR in terms of pollutant removal, and membrane fouling falling into the category of operational problems of MBR.

2.1 Composition of MBR systems

2.1.1 MBR configurations

MBR systems can be configured in two ways according to the position of membrane module: submerged and sidestream MBRs. Side-stream MBR was introduced into

China prior to submerged MBR, which is in accord with the course of the world's MBR research. Submerged MBR (SMBR) was first introduced in 1989 by Yamamoto et al. [\[2](#page-21-0)]. After that, such configuration was recognized as an alternative for MBR systems featured by more compact configuration and less energy consumption due to waiver of external recirculation. This energetic advantage allows the submerged configuration to be more favorable in MBR application and thus in MBR research over the past decade. Meanwhile, side-stream MBR mainly pertains to industrial or specific wastewater treatment [\[3,4](#page-21-0)] or is related to fundamental aspects, such as cross-flow membrane filtration [\[5](#page-21-0)–[7\]](#page-21-0).

2.1.2 Membrane materials and modules

A number of membrane materials and modules that have been used in MBR studies are summarized in Table 1. The prevailing materials employed so far are halogenated hydrocarbon polymers such as polyvinylidene fluoride (PVDF), with considerations given to mechanical strength, chemical tolerance, biological stability, fabrication difficulty, etc. As a variant within the halogenated polyvinyl series, polyvinyl chloride (PVC) endowed with chlorine is also used in MBR. Meanwhile, polymeric hydrocarbons without any heteroatom, such as polyethylene (PE) and polypropylene (PP), are still popular. Apart from the above, polyether sulfone (PES), a common ultrafiltration membrane material showing a relatively hard-brittle nature, is also used for some cases of microfiltration.

Most of the studies have been focused on the utilization of hollow fiber membranes while a few are on flat sheet membranes, with metal flat sheet membranes included. The domestic hollow fiber membranes are mainly

produced by Tianjin Motimo (PVDF) and Zheda Hyflux Hualv (PP). The domestic flat sheet membranes are mainly produced by Shanghai Institute of Applied Physics (or SINAP). Ceramic membranes were also studied in MBR process due to their special qualities. Domestic membrane fabrication is expected to accelerate promisingly in the near future.

Since the relatively high cost of the membrane is one of the considerable obstacles to broader application of MBR, many efforts have been made concerning the fabrication of membranes with the results showing continuous cost decline. Meanwhile, self-forming dynamic membrane bioreactor (SFDMBR) ——a process even without manufactured membrane ——has been considered, with cheap coarse pore-sized materials such as Dacron mesh [[8,9\]](#page-21-0), nonwovens [[10,11\]](#page-22-0), stainless steel mesh [[12](#page-22-0)], etc., employed as filtration media. The sludge cake layer and gel layer dynamically formed on the filtration medium were found effective in enhancing the solid-liquid separation, and the effluent quality could be kept at a stable level with undetectable suspended solid (SS) concentration [\[8](#page-21-0)], suggesting the future potential of this kind of material. Apart from microbial flocs and colloids, some inorganic particles such as diatomite [[12](#page-22-0)] and powder activated carbon (PAC) [[13](#page-22-0)] have also been utilized with the assistance of organic substances from sludge to form a dynamic membrane.

2.2 Pollutant removal performance of MBR-based processes

Up to now, MBR has exhibited outstanding versatility for both municipal and industrial wastewater treatment. Moreover, the capability of an MBR for pollutant removal

Table 1 Main membrane materials and modules used in MBR studies in China

membrane material	module	nominal pore size/µm	manufacturer
PE	hollow fiber	$0.1 - 0.4$ 0.4	Mitsubishi Rayon, Japan Korea Membrane Separation, Korea
PP	hollow fiber	$0.1 - 0.2$	Zheda Hyflux Hualv, China
PVC	hollow fiber	0.01	Litree, China
PVDF	hollow fiber	0.4 0.1 0.04 0.04 0.1 0.22 0.01	Mitsubishi Rayon, Japan Asahi Kasei, Japan Siemens Memcor, Germany GE, USA Origin Water, China Tianjin Motimo, China Litree, China
chlorinated PE	flat sheet	0.4	Kubota corporation, Japan
PVDF	flat sheet	0.08 0.2	Toray, Japan SINAP, China
metal	flat sheet	0.2	Hitachi Metals, Japan
PVDF	tubular	150 kDa	Norit X-Flow, Netherlands
ceramic	tubular ^{a)}	0.2	Nanjing Univ. of Technol., China

Note: a) pressurized side-stream MBR

can be enhanced and/or expanded via flexible combination with other units yielding integrated processes. Herein the pollutant removal performance of MBR-based processes is overviewed.

2.2.1 Municipal wastewater treatment

2.2.1.1 Regular contaminant removal

Water shortage is a significant problem in China, which is particularly the case in the northeastern and northwestern areas. MBR is an attractive technology for wastewater reclamation since it can provide stable and excellent effluent. Very extensive works have demonstrated that MBR is effective in removing turbidity, SS and chemical oxygen demand (COD) indicative of organic matter. The typical removal efficiency of turbidity, SS and COD is > 99% [[14](#page-22-0)–[16](#page-22-0)], > 99% [\[14,17](#page-22-0)] and > 90% [\[14,15,17](#page-22-0)– [19](#page-22-0)], respectively, according to data from pilot-scale MBRs. The major mechanism of pollutant retention by membrane in MBR is sieve effect. According to the typical pore size on the order of $0.05-0.5 \mu m$ (Table 1), there is no doubt that membrane can readily reject suspended particles and most colloids. In addition, the retention of dissolved organic matters (DOM) has also been observed. Wang and Wu [[17](#page-22-0)] found that the typical bimodal distribution of molecular weight (MW) of DOM in MBR supernatant was 6.1–382000 kDa while that in effluent was 10.9–1870 kDa. Moreover, continuous decrease of total organic carbon (TOC) in membrane effluent was observed, especially for gelling substances [\[20](#page-22-0)], which further confirms the enhancement of retention efficiency via pore narrowing or gel layer formation, in a manner similar to dynamic membrane formation.

2.2.1.2 Nutrient removal

With respect to the stringent anti-eutrophication regulations, MBR is required to have the qualities for nitrogen and phosphorus removal. For nitrogen removal, since MBR can easily achieve an ammonium (NH_3-N) removal efficiency higher than 90% [[14](#page-22-0),[18,21](#page-22-0)], recent studies have been focused more on total nitrogen (TN) removal. In this regard, alternate mixed liquor conditions (e.g., dissolved oxygen (DO)) are needed to facilitate nitrification and denitrification processes. The most popular process for this purpose is anoxic/oxic-MBR $(A_2/O\text{-}MBR)$ (or any other process containing this part). In Cao et al.'s pilot study [\[22\]](#page-22-0) of an anaerobic/anoxic/oxic-MBR $(A_1/A_2/O\text{-}MBR)$ where the ratio of recirculation from membrane (aerobic) zone to anoxic zone was 300%, TN removal was found to be 74.4%, close to the theoretical value. Other processes have also been proposed, such as the up-flow anaerobic sludge blanket (UASB) and aerobic MBR (i.e., UASB-MBR) [\[23\]](#page-22-0) enabling simultaneous methanogenesis and nitrogen

removal. The TN removal increased from 48.1% to 82.8% with the sludge recirculation ratio (from MBR to UASB) increasing from 50% to 800%, with the assistance of the realization of shortcut biological nitrogen removal (SBNR) process during which the ammonium-nitrogen is oxidized to the form of nitrite rather than nitrate, thereby consuming less COD. SBNR is considered particularly favorable for nitrogen removal from wastewaters with a low C/N ratio. Besides conditions of spatial alternation of nitrification and denitrification, TN can also be removed under time-alternate conditions. Zhang et al. [[24](#page-22-0)] observed a TN removal $> 65\%$ in a sequencing batch MBR (SBMBR) when COD/TN ratio was 6.3 and TN load was $0.22 \text{ kg} \cdot \text{m}^{-3} \cdot \text{d}^{-1}.$

Apart from the spatial-alternate or time-sequencing processes, even a single-tank MBR has been found to be capable of removing TN effectively, with the assistance of simultaneous nitrification and denitrification (SND), which is enabled by the heterogeneous environment (of, for instance, DO, substrate concentration and microbiology) within a sludge floc along its thickness direction (see Fig. 2). He et al. [\[25\]](#page-22-0) achieved a TN removal around 86.6% in a single-tank MBR at $DO = 0.8$ mg·L⁻¹. The effectiveness of SND could be influenced by food/microorganism (F/M) ratio, COD/TN, DO, pH as well as steric factors such as size and compactness of sludge flocs [[25](#page-22-0)]. It has been found in MBR that higher COD/TN and F/M ratios are advantageous to SND [\[25,26](#page-22-0)] on condition that nitrification is not inhibited; while for each of DO and pH, there exists an optimum located in moderate range (around $DO = 1 mg \cdot L^{-1}$ [[25](#page-22-0),[26](#page-22-0)] and pH = 7 [25]). SND could be facilitated by addition of carriers to form biofilm [[27](#page-22-0)] or utilization of aerobic granular sludge [[28](#page-22-0)].

Fig. 2 Scheme of heterogeneous environment in a sludge floc for SND (obtained from Ref. [\[25](#page-22-0)])

With respect to phosphorus removal, particular attention has been paid to MBR functionalized with enhanced biological phosphorus removal (EBPR) [[22,29](#page-22-0)–[35](#page-22-0)]. In

Zhang et al.'s study [[32\]](#page-22-0), an EBPR-MBR system composed of one anaerobic zone, two anoxic zones and the membrane (aerobic) zone was constructed and fed with synthetic domestic wastewater continuously. In order to investigate the effect of SRT on phosphorus removal, four comparative runs were operated at SRTs of 20, 30, 40 and 50 d. It can be seen in Fig. 3 that high total phosphorus (TP) removal could be achieved even when SRT was prolonged to 40 d. This was due to increased phosphorus content in the sludge (expressed as $kg-P \cdot kg^{-1}$ -MLSS) with increased SRT, which allowed for reduction of excess sludge discharge while sufficient phosphorus removal was ensured [\[32](#page-22-0)]. In addition to oxygen, $NO₃⁻-N$ in anoxic environment has also been found to be an electro-acceptor during phosphorus uptake, with this process referred to as denitrifying phosphorus removal (DPR) ascribed to

Fig. 3 Average phosphorus removal in the EBPR-MBR at different SRTs [[32](#page-22-0)]

denitrifying phosphorus removing bacteria (DPB). According to the phosphorus balance in the EBPR-MBR system performed by Zhang et al. [\[31\]](#page-22-0), 66.7%–78.8% of TP removal was due to biological phosphorus accumulation, 52% of which was attributable to DPR. It should be additionally noted that, not only microorganisms but also extracellular polymeric substances (EPS) surrounding them can play a role in phosphorus removal. Zhang et al. [[36](#page-22-0)] analyzed the various phosphorus forms in EPS from different processes using ³¹P-nuclear magnetic resonance (NMR) spectroscopy (spectra shown in Fig. 4). It can be seen that compared to EPS from CAS of poor TP removal ability, EPS from EPBR-MBR contained a significant amount of polyphosphate (characterized as "end poly-P" and "middle poly-P" peaks in Fig. 4). In the EPS from EPBR-MBR, polyphosphate accounted for ca. 55% of total phosphorus [\[36\]](#page-22-0), suggesting biological phosphorus accumulation could occur in EPS that is also contributive to total TP removal via sludge discharge.

The concentration of phosphorus in aqueous phase can be further lowered via membrane rejection. The phosphorus in aqueous phase includes inorganic phosphates of small molecular size and colloidal phosphorus typically larger than $0.025 \mu m$ on membrane partitioning [\[37\]](#page-22-0). Colloidal phosphorus is recognized as in the form of phosphorus chemically bonding with macromolecules such as protein-like substances (according to threedimensional excitation-emission matrix (EEM) fluorescence spectroscopic analysis [\[38\]](#page-22-0)) from soluble microbial products (SMP). Figure 5 shows the obvious existence of colloidal phosphorus (accounting for ca. 50% of TP in supernatant) and that this fraction was well intercepted during membrane filtration. It should be noted, however, that a new microfiltration membrane (with the pore size

Fig. 4 Typical ³¹P-NMR spectra from the investigated EPS of three sludge samples (modified from that provided by Zhang et al. [\[36\]](#page-22-0))

Fig. 5 Molecular weight distribution and TP distribution in MBR supernatant and effluent (modified from that provided by Zhang [[38](#page-22-0)]). Molecular weight distribution was measured by a gel filtration chromatography (GFC) instrument

usually on the order of $0.05-0.5 \mu m$, c.f. Table 1) is never effective in rejecting colloidal phosphorus. Only after the membrane fouling layer grows to some sufficient extent could colloidal phosphorus be well retained. The fouling layer-enhanced phosphorus retention could be confirmed by Zhang's observation [\[38\]](#page-22-0) that the retention rate of colloidal phosphorus increased rapidly from 2% to 60– 70% in accord with trans-membrane pressure (TMP) elevation during one month's operation ("2%" corresponding to freshly cleaned 0.4 µm-membrane), and the retention rate reached 87.3% when the membrane was severely fouled (TMP $>$ 50 kPa at a constant flux of \sim 12 L·m⁻²·h⁻¹ or LMH).

In addition to spatial-alternate MBR processes such as $A_1/A_2/O$ -MBR, some time-sequencing MBR processes are also effective in phosphorus removal. The sequencing batch membrane bioreactor (SBMBR) developed by Zhang et al. [\[24\]](#page-22-0) exhibited a stable TP removal around 90%. Dai et al. [[34](#page-22-0)] investigated the denitrifying phosphorus removal progress in an SBMBR and found that the proportion of denitrifying phosphorus removing bacteria increased from 19.4% to 69.6% of total phosphate accumulating organisms due to condition improvement. Yuan et al. [[39](#page-22-0)] developed an alternating of anoxic and anaerobic membrane bioreactor (AAAM) processsystem operated in a manner combining both spatial and time alternation——to enhance simultaneous nitrogen and phosphorus removal. In this process composed of three zones, the mixed liquor in the aerobic zone was recycled at a rate of 200% to either of the other two zones alternately, thereby yielding alternate switches between anoxic/anaerobic/aerobic-MBR and anaerobic/anoxic/aerobic-MBR modes. The resultant TN and TP removal successfully attained 67.4% (near theoretical value) and 94.1%, respectively.

Apart from biological means, physicochemical methods have also been employed in MBRs to elevate phosphorus removal. For instance, Cui et al. [\[40](#page-23-0)] developed an electrocoagulation-MBR (EC-MBR) process by installing iron-anode plate and titanium-cathode mesh into the downflow zone of an SMBR. By controlling the electric current intensity and current-on time thereby adjusting iron ion release, the TP removal could be maintained over 95% with the effluent $TP < 0.5$ mg $\cdot L^{-1}$.

2.2.1.3 Pathogen removal

Concerning the safety of reclaimed wastewater used for lavatory flushing, vehicle washing, urban greening, etc., the feasibility and effectiveness of pathogen removal by MBR should be well investigated. Since microfiltration mostly applied in MBR can readily reject protozoa, helminths (as well as their eggs), fungal spores and bacteria due to their sizes being larger than membrane pores, more attention has to be paid to the retention of viruses. The virus removal performance by an SMBR fed with real municipal wastewater was investigated by Wu et al. [\[41\]](#page-23-0), using indigenous somatic coliphages (SC) as an indicator of viruses. The feed SC concentration was $(2.81 \pm 1.51) \times 10^4$ PFU \cdot mL⁻¹. It was found that in the

SMBR system, biomass played the most important role in SC removal, achieving a high removal efficiency of over 98%. Membrane rejection was an essential supplement to biomass effect, which is particularly the case when the membrane was fouled, allowing the effluent SC to be less than 10 $PFU \cdot mL^{-1}$. The fouling layer-enhanced virus retention has also been reported by a few other researchers, using phage T4 [[42](#page-23-0)–[44\]](#page-23-0), phage f2 [[44](#page-23-0)], etc., as indicators. Further study by Wu et al. [\[41\]](#page-23-0) revealed that gel layer played a more important role in SC rejection than cake layer, especially at higher filtration fluxes (Fig. 6), whereas, Lv et al. [\[42\]](#page-23-0) found that cake layer prevailed in phage T4 retention. This may be because the SMBR in the latter study was operated in the constant-pressure mode with only 8.5 kPa of TMP and the flux averaged just about 3 LMH (estimated from Ref. [\[42\]](#page-23-0)) which is much smaller than those in Fig. 6, such that the gel layer might be looser with wider channels [[45](#page-23-0),[46](#page-23-0)] thereby rendering lower gel layer contribution to virus rejection than presented in Fig. 6.

Fig. 6 SC rejection contribution of different fouling layers at three fluxes when the membrane was severely fouled [\[41\]](#page-23-0) (LRVM denotes the log_{10} removal value by membrane (LRVM = log_{10}) (C_s/C_e) , where C_s and C_e stand for SC concentrations in the supernatant and effluent, respectively)

2.2.1.4 Micro-pollutant removal

Micro-pollutants, such as endocrine disrupting compounds (EDCs), pharmaceutical and personal care products (PPCPs) and persistent organic pollutants (POPs), are regarded as toxic even at trace concentrations. EDCs may be grouped into: 1) phenolic compounds including bisphenol A (BPA), alkylphenols (such as 4-n-octylphenol $(4-n-OP)$, 4-n-nonylphenol $(4-n-NP)$ and 4-tert-octylphenol (4-tert-OP)) and alkylphenol polyethoxylates (such as nonylphenol polyethoxylates (NPnEO)), and 2) steroidal estrogens including estrone (E1), 17β -estradiol (E2), 17α estradiol (17α-E2), estriol (E3), 17α-ethynylestradiol (EE2), etc. PPCPs may be grouped into: 1) antiinflammatory and analgesic drugs such as aspirin (ASA), diclofenac (DCF), ibuprofen (IBP) and ketoprofen (KTP),

2) antibiotics such as sulfamethoxazole (SMZ), norfloxacin (NOR) and cefmetazole (CMZ), 3) lipid regulating drugs such as clofibric acid (CFA), 4) developers such as diatrizoate (DTZ), 5) musks such as tonalide (AHTN) and gallaxolide (HHCB), and 6) other drugs such as caffeine (CAF), carbamazepine (CBZ) and sulpiride (SLP).

The typical (average) concentrations of EDCs and musks range from $71.3 \text{ ng} \cdot \text{L}^{-1}$ of 4-n-NP to 1968.1 ng $\cdot \text{L}^{-1}$ of HHCB in municipal wastewater and their residual concentrations from 20.4 ng \cdot L⁻¹ of E3 to 827.0 ng \cdot L⁻¹ of HHCB after conventional activated sludge (CAS) treatment, according to Zhou et al.'s statistics based on wastewater treatment plants (WWTPs) in Beijing [[47](#page-23-0),[48](#page-23-0)]. It has been reported that the hormone system of adult trout and/or roach representative of fish could be obviously disrupted when the concentration of 4-tert-OP reached the level of $10 \mu g \cdot L^{-1}$, or when that of E1 or E2 exceeded the magnitude of $10 \text{ ng} \cdot L^{-1}$ [[49](#page-23-0)]. And for EE2 which is highly endocrine-disruptive, concentrations even as low as 0.1 ng $\cdot L^{-1}$ may induce estrogenic abnormality [[49](#page-23-0)], while the concentration of residual EE2 in WWTP effluent was found to be on the order of ~ 100 ng \cdot L⁻¹ [\[48\]](#page-23-0). In consideration of the environmental risk of these micropollutants to public health and the ecosystem, wastewater containing these substances should be satisfactorily treated before discharge or reuse. While most CAS processes are not effective enough in this respect, the utilization of MBR should be worth trying in view of its possible advantages.

Chen et al. [[50](#page-23-0)] made a comparison between an MBR and a CAS reactor with respect to BPA removal. For the influent concentration of BPA ranging from 0.05– $20 \text{ mg} \cdot L^{-1}$, a stable removal of over 93.7% was attained in the MBR while that in the CAS reactor was less stable despite that their average efficiency was similar. Moreover, the MBR could bear much higher volume loadings than the CAS reactor while the BPA removal efficiency was maintained at a high level. Similar results were obtained with regard to $NPhEO$ removal also by these two reactors [[51](#page-23-0)]. Zhou et al. [[52](#page-23-0)] conducted an investigation into the removal of eight typical EDCs by an MBR in comparison with a sequencing batch reactor (SBR). The eight target EDCs selected were 4-n-OP, 4-n-NP, NPnEO, BPA, E1, E2, E3 and EE2, with their removal rates shown in Fig. 7. As can be seen, for substances that are relatively easily biodegradable such as BPA, E2 and E3, both reactors showed excellent removal efficiency of more than 95%. However, with regard to those more refractory compounds such as E1 and EE2, MBR demonstrated much better removal than SBR. Compared to the concentration of specific target compounds, the total estrogenicity, which is evaluated as E2 equivalent quantity (EEQ) determined via yeast estrogen screen (YES) assay (c.f. Ref. [[53](#page-23-0)] for details), is considered to be an essential index of the overall occurrence and risk of EDCs. It was found that the EEQ removal efficiency of around 90% in MBR was much higher than that of around 50% in SBR [[52](#page-23-0)].

Fig. 7 Removal rates of target EDCs in MBR and SBR [[52](#page-23-0)]

The EDCs removal behavior of full-scale MBR combined with anaerobic-anoxic-oxic process (i.e., A_1 / $A₂/O-MBR$) was also investigated [[54\]](#page-23-0). It was found that more than 97% of BPA, E1, E2, 17α -E2 and E3, 87% of EE2, and over 70% of 4-n-OP and 4-n-NP could be removed by this combined process. The estrogenicity was markedly reduced from 72.1 ng-EEQ \cdot L⁻¹ in the influent to 4.9 ng-EEQ \cdot L⁻¹ in the effluent.

In order to better understand the mechanisms of EDCs and PPCPs removal in an MBR-based process, it is important that their transportation and transformation be clarified. Figure 8 illustrates the fate of EDCs and PPCPs in an MBR system. The major fate is usually considered to be physical adsorption by sludge or biodegradation. According to Wu et al.'s observation [\[54\]](#page-23-0), the adsorption by sludge played a prominent role in the removal of most of the selected hydrophobic EDCs, with the solid-water distribution coefficients (the ratio of solid-phase concentration to aqueous-phase concentration) found to be mostly above $10000 \mathrm{L} \cdot \mathrm{kg}^{-1}$ -SS. The *n*-octanol/water partition coefficient (K_{ow}) of the target compound has been recognized as a primary parameter predicting the propensity for adsorption. An empirical linear relationship between K_{ow} and adsorption equilibrium constant was

established based on statistical analysis of adsorption of various nonionic organic chemicals on similar-sourced organic absorbents [\[55\]](#page-23-0). On the other hand, biodegradation has also been found to be very significant in some cases, such as when Chen et al. used an MBR to remove BPA [[50](#page-23-0)] or NPnEO ($n = 1-4$) [\[56\]](#page-23-0). Zhou et al. [\[52\]](#page-23-0) provided an insight into the effect of SRT on EDCs removal in an MBR, with the results shown in Fig. 9. As can be seen, for 4-n-NP, NPnEO, E1, EE2 and the total estrogenicity expressed as EEQ, their removal was effectively improved by increasing SRT, suggesting that the enhancement of biodegradation overweighed the diminishing of adsorptional contribution due to decreased excess sludge. Further research is needed to reveal the detailed mechanisms.

2.2.2 Industrial wastewater treatment

In the industrial sector, MBR has also become more and more active in the treatment of both high strength wastewaters (such as brewery wastewater, sauce wastewater and food processing wastewater) and refractory wastewaters (such as coke plant wastewater, petrochemical wastewater and dyeing wastewater). Table 2 exemplifies recent researches on the application of MBR to industrial

Fig. 8 Scheme of possible fate of EDCs and PPCPs in an MBR system

anaerobic packed-bed biofilm reactor, SMBR = submerged MBR, TPAD = two-phase anaerobic digestion system composed of a continuous stirred tank reactor (CSTR) and an upflow anaerobic sludge blanket-anaerobic
filter (UASB-AF anaerobic packed-bed biofilm reactor, SMBR = submerged MBR, TPAD = two-phase anaerobic digestion system composed of a continuous stirred tank reactor (CSTR) and an upflow anaerobic sludge blanket-anaerobic filter (UASB-AF); c) 4-NP = 4-nonylphenol, OCPs = organochlorine pesticides, PAHs = polycyclic aromatic hydrocarbons, SCOD = soluble COD; the concentration unit is mg L^{–1} if not specified; removal efficiency is presented in parentheses; acute toxicity is expressed as equivalent concentration of Zn^{2+} in mg·L⁻¹ presented in parentheses; acute toxicity is expressed as equivalent concentration of Zn^{2+} in mg $\cdot L^{-1}$

Fig. 9 Effect of SRT on EDCs removal by MBR [\[52\]](#page-23-0)

wastewater treatment. For the high strength wastewaters, MBR can mitigate problems such as sludge bulking that are commonly troublesome for the CAS processes, thus enhancing treatment efficiency and permitting stable operation. Moreover, the relatively small footprint should be also favorable for the practical application of MBR in the industrial sector. For the refractory industrial wastewaters, membrane can retain all the biomass as well as part of the less biodegradable pollutants in the bioreactor, thereby probably improving the pollutant removal both physically (via direct membrane rejection) and biologically (via enhancement of biodegradation favored by lower F/M ratios).

Compared to municipal wastewater, industrial wastewater demonstrates characteristically high load and/or low biodegradability; moreover, there are usually remarkable disparities among wastewaters from different industrial sources. Therefore, elaborate consideration should be given to the design of MBR processes in the industrial sector to suit the specific feed conditions. For example, with respect to high strength organic wastewaters, anaerobic digestion is often applied prior to aerobic MBR process; for refractory wastewaters, anaerobic treatment governed by hydrolysis and acidification is usually proposed to ameliorate the biodegradability. Additional attention should be sometimes paid to the pre-treatment such as: regulation of acidity or alkalinity, or flotation to remove oily substances which are troublesome for subsequent biological treatment and membrane performance.

Additionally, in order to better the treatment of some specific wastewaters which are highly refractory, MBR could be further reinforced by novel technologies (such as bio-enhancement via genetic engineering). Liu et al. [\[71](#page-24-0)– [73](#page-24-0)] used a laboratory-scale MBR to treat synthetic pesticide wastewater containing a high concentration $(15-20 \text{ mg} \cdot \text{L}^{-1})$ of atrazine, which is in the POPs family and demonstrates eco-toxicity even at ppb level. The MBR was bio-augmented with a genetically engineered microorganism (GEM) and realized a high and stable removal efficiency of above 90% after a start-up period of around 10 days [[72](#page-24-0)]. Moreover, the MBR equipped with 0.4 µm PE membrane exhibited excellent retention of GEM, with the effluent GEM less than 10^2 CFU \cdot mL⁻¹ in the initial stage of filtration and undetectable with the development of fouling layer [[71](#page-24-0)]. In comparison, the CAS system operated in parallel showed severe GEM leakage especially with the occurrence of sludge bulking, leading to relatively high potential ecological risk to the environment, despite the natural decay of GEM [\[73\]](#page-24-0). This would also add to the advantages of MBR over CAS.

2.3 Membrane fouling

Membrane fouling has been recognized as a major obstacle to the wider application of membrane technology to wastewater treatment. Considerable energy consumption and engineering cost could be raised by membrane fouling through reduction of filtration efficiency, increase in frequency and intensity of cleaning, and shortening of membrane lifetime, etc. Membrane fouling is considered to be spontaneous during the microfiltration or ultrafiltration process in MBR and seems inevitable in spite of all antifouling measures taken so far. Nevertheless, it should be greatly important that cost-effective efforts be made in order to alleviate/decelerate membrane fouling as much as possible. For this purpose, it is essential to understand the mechanisms of membrane fouling comprehensively and to develop robust anti-fouling strategies.

2.3.1 Membrane fouling formation

2.3.1.1 Basic understandings

Membrane fouling is considered to result essentially from the interaction between membrane and foulant under certain conditions. Thus, membrane fouling can be influenced by properties of the fouling participants (i.e., membrane and foulant) as well as operating conditions. Important fouling-related membrane properties may

include material composition [[74\]](#page-24-0), physicochemical properties such as hydrophobicity [\[74](#page-24-0)–[77](#page-24-0)] and surface charge [\[78,79\]](#page-24-0), and steric properties such as pore size, porosity and surface roughness [\[74,77\]](#page-24-0). Likewise, foulant may be characterized from perspectives of substance types, physicochemical properties and steric properties.

Foulant can be roughly divided into organic and inorganic species. Major attention has been paid to organic foulants, whereas inorganic foulants have also been found significant in some cases [\[80\]](#page-24-0), probably in the form of inorganic scales or combined with organic skeletons. Inorganic fouling-inducing elements are usually abundant in wastewaters and are represented, apart from O, C and H, by multivalent metallic elements such as Ca, Mg, Al and Fe, and nonmetallic elements with low electronegativity such as Si and P. The grouping of organic foulants is much more complicated. In the macromolecular category, organic foulants are mostly grouped into polysaccharides, proteins and humic substances. It should be noted, however, that this grouping of organic foulants is somewhat empirical and superficial, and that the fouling behaviors should be more essentially governed by the physicochemical and steric characteristics exhibited by these foulants.

The most often mentioned physicochemical properties of foulant under consideration are hydrophobicity and surface charge. Both of the properties could be qualitatively determined via column chromatography method based on differential adsorption and elution [[81](#page-24-0)–[82](#page-24-0)], yielding hydrophilic/hydrophobic fractions and acid/base/ neutral fractions. Another useful method for determining hydrophobicity is the bacterial adherence to hydrocarbons (BATH) test based on the partition of the target (bacteria) between aqueous phase and hydrocarbon droplet surface [\[83\]](#page-24-0); this method has recently been applied to the case of sludge flocs with the result termed relative hydrophobicity (RH) [[84](#page-24-0)]. Additionally, surface charge could be otherwise quantified as zeta potential [\[84,85\]](#page-24-0), though sensitive to the ionic condition applied in the measurement [[86](#page-24-0)]. As for the steric properties of foulant, most attention has been focused on foulant size (or molecular weight at the macromolecular level) [\[82,87,88\]](#page-24-0), with which the major fouling mechanism may vary substantially, as will be discussed later. Moreover, the concentration of foulant in the form of flocs, colloids or macromolecules can also play a role in fouling [\[85,89,90\]](#page-24-0).

The operating conditions that could impact on membrane fouling may be supposedly divided into local conditions (specialized in the vicinity of the membrane, which are usually hydraulic conditions) and bulk environment. Local conditions include filtration mode (of constant-flux or constant-pressure), flux [[90](#page-24-0)–[93](#page-24-0)], TMP [\[46\]](#page-23-0), etc., conceptually perpendicular to membrane surface, and cross-flow velocity (CFV) [[90](#page-24-0)], cross-flow pattern [[77,94](#page-24-0)], shear rate [[45,](#page-23-0)[95](#page-24-0)], etc., parallel to membrane surface. Additionally, the manner of the

periodic alternation of these hydraulic conditions, for instance, suction/relaxation proportion [\[91,92,96\]](#page-24-0) or online backwash interval and duration [\[97,98\]](#page-24-0), could also affect fouling. The fouling-related bulk environment may include temperature [\[98,99\]](#page-24-0), pH [[100](#page-24-0)], viscosity [[85](#page-24-0)], etc. Moreover, there are some typically indirect operating factors, such as HRT [\[101\]](#page-24-0), SRT [[81](#page-24-0),[102](#page-24-0)], membrane configuration (e.g., membrane fiber length [[46](#page-23-0),[103,](#page-24-0)[104\]](#page-25-0)) and mixed liquor conditioning (e.g., dosage of coagulants [[105](#page-25-0),[106\]](#page-25-0), adsorbents [[107](#page-25-0)] or carriers [[95](#page-24-0)[,108](#page-25-0)–[110](#page-25-0)]), which play roles in fouling via affecting those direct factors from foulant properties or operating conditions mentioned above.

The toughness of membrane fouling could be further described as removable fouling, irremovable but reversible fouling, and irreversible fouling, judging by results of physical cleaning (for removability) and chemical cleaning (for reversibility) [[111\]](#page-25-0); however, this division is not strict and is strongly dependent upon the cleaning conditions, such as cleaning mode, intensity and duration.

The process of membrane fouling typically consists of, in sequence, initial stage involving membrane pore blocking and narrowing (also known as internal fouling), gel layer stage, and cake layer stage. The fouling mechanisms of each stage as well as the roles of foulants with different sizes will be further discussed later.

2.3.1.2 Foulant identification

Herein only organic foulants are overviewed with regard to their general prevalence in fouling. The overall significance of major components of organic foulants (including suspended solids and supernatant organics) was statistically studied by Wu and Huang using Pearson correlation analyses [\[85\]](#page-24-0). MLSS ($> 0.45 \mu m$) was found to be related significantly to the apparent rate of increase in filtration resistance (i.e., $k_R = \Delta R_f / \Delta t$, where R_f is filtration resistance; say also fouling rate) when MLSS was higher than $10 \text{ g} \cdot \text{L}^{-1}$, whereas no definite correlation was observed when it was lower than $10 \text{ g} \cdot \text{L}^{-1}$. On the other hand, supernatant organics, expressed as supernatant TOC $(< 0.45 \,\mathrm{\mu m})$, were found to play an even major role in overall fouling. These substances could be subdivided into colloidal organic carbon (COC) and soluble organic carbon (SOC), the latter of which was defined as the remainder after $\text{Al}_2(\text{SO}_4)$ ₃-flocculation removing COC from supernatant TOC. The correlation between the concentration of these substances and fouling rate is given in Fig. 10. Further insight into the roles of supernatant TOC in fouling evolution indicates that, apart from membrane pore blocking and gel layer formation, these substances could also participate in cake layer building through blocking the pores and spaces between particles in the cake layer and considerably increase the specific filtration resistance of the cake layer [\[112](#page-25-0)].

Supernatant organics are composed of SMP and

Fig. 10 Relationship between apparent fouling rate (k_R) and (a) TOC, (b) COC, (c) SOC, (d) polysaccharide content and (e) protein content in the supernatant based on Pearson correlation analyses [[85](#page-24-0)] $(k_R = \Delta R_f/\Delta t$, where R_f is filtration resistance; COC = colloidal organic carbon, $SOC =$ soluble organic carbon; supernatant $TOC = COC + SOC$)

nonbiodegradable organic matters derived from feed water [\[113](#page-25-0)]. SMP are considered to be the most important contributor to supernatant TOC [[85](#page-24-0)], and can be classified into: utilization-associated products (UAP) derived from original substrate, and biomass-associated products (BAP) generated by hydrolysis of extracellular polymeric substances (EPS) [[114](#page-25-0)] that are describable as tightly-bound EPS (TB-EPS) and loosely-bound EPS (LB-EPS) [[115\]](#page-25-0).

Supernatant organics are primarily grouped into polysaccharides, proteins and humic substances, among which it has been remarked that polysaccharides (c.f. Fig. 10 (d)) and/or proteins are more significantly related to fouling [\[85,](#page-24-0)[116](#page-25-0)–[118](#page-25-0)]. Moreover, it can be inferred from many studies [[81](#page-24-0),[82](#page-24-0)] that polysaccharides and/or proteins are usually the most abundant organic species in supernatant of MBR treating municipal wastewater, whereas the case is more complicated for industrial wastewater treatment. The

molecular weight (MW) distributions of polysaccharides, proteins and humic substances are illustrated in Fig. 11. It can be seen that, all of these distributions exhibited wide ranges from < 1 kDa to > 100 kDa (or crudely, from < 1 nm to > 10 nm) approximately in a bimodal pattern, with similar findings reported by many other researchers [[113,119\]](#page-25-0). The peak zone of larger MW is more commonly concerned with fouling of microfiltration membranes as will be explained later. The average sizes of the major groupings typically follow the order: polysaccharides > proteins > humic substances. Additionally, the MW distribution of supernatant organics could vary according to operating conditions; for instance, the average MW was found to shift downwards (with the proportion of polysaccharides decreased and the peak of larger MW weakened correspondingly) during prolonged operation at $SRT = 20 d$ [\[120\]](#page-25-0). As for the hydrophobicity of supernatant

Fig. 11 Molecular weight distribution of TOC and typical foulants in MBR supernatant based on membrane partitioning (data extracted from Zhao et al.'s work [[88](#page-24-0)])

organics, it can be inferred from Fig. 12 that hydrophilic substances (HIS) and hydrophobic acids (HOA) are the main contributors to supernatant TOC, and that polysaccharides and proteins demonstrate an almost hydrophilic nature. It is worthy of note, however, that the classical division of supernatant organics into polysaccharides, proteins and humic substances is not strict; there is always to some extent an overlap among these groupings (i.e., a single macromolecule, e.g., a proteo-polysaccharide, might behave in multiple ways according to common analytical methods at present) [\[88\]](#page-24-0).

2.3.1.3 Fouling mechanisms

As has been mentioned in Section 2.3.1.1, the major mechanism(s) can vary greatly with foulant size, the range of which in this regard could be roughly divided into three zones where: 1) foulant size \gg membrane pore size, 2) foulant size is comparable with membrane pore size, and 3) foulant size≪membrane pore size. According to the microfiltration membrane pore size usually in the range of 0.05–0.5 μ m (c.f. Table 1), the foulant size in these three zones may be typically on the order of $> 1 \mu m$, 0.01–1 μ m and $< 0.01 \mu m$, respectively. Herein we will give an overview of the major mechanism(s) for each zone, on the basis of findings mainly proposed by Chinese researchers.

(1) For foulant size much larger than membrane pore size

Foulants lying in this size range are described as suspended solid particles or sludge flocs that can build a cake layer on the membrane surface. The driving force for transport of a foulant particle to the membrane surface is the drag force of filtration, whilst those for back transport include Brownian diffusion, shear-induced diffusion and inertial lift, among which Brownian diffusion is usually considered less significant with respect to this foulant size range

Fig. 12 Hydrophilic/hydrophobic fraction distribution of TOC and typical foulants in MBR supernatant based on the method using nonionic macroporous chromatographic resin column (SupeliteTM DAX-8 resin) (data extracted from Shen et al.'s work [[82](#page-24-0)]) (HIS = hydrophilic substances, HOA = hydrophobic acids, HOB = hydrophobic bases, HON = hydrophobic neutral substances)

[[121](#page-25-0)]. Moreover, when the particle arrives at the membrane or cake layer surface (providing pivot positions), there is an opposition between the filtration drag torque and the shear torque, the latter of which gives rise to surface transport that is anti-fouling [[121](#page-25-0)].

The key parameters related to these fouling processes include filtration flux, shear rate, particle size and particle concentration. Since filtration drag force as well as its torque is directly related to filtration flux, it is understandable that flux should be a crucial fouling-inducing factor. The significance has been confirmed by extensive

works [[90](#page-24-0)–[93\]](#page-24-0). Moreover, there exists a critical flux over which fouling will deteriorate sharply [[91](#page-24-0)], possibly corresponding to the filtration drag effect overwhelming the back-transport effects.

Modeling works suggest that higher shear rate is favorable for all the back transport and surface transport processes mentioned above [\[121\]](#page-25-0). Shear rate can be otherwise parameterized as cross-flow velocity, aeration intensity, etc. The significance of the anti-fouling effect of aeration intensity was confirmed by Gui et al. [\[91\]](#page-24-0), with the effect found to be more pronounced at higher MLSS concentrations. The existence of a critical aeration intensity as well as a critical cross-flow velocity was validated by Liu et al.'s modeling work [\[90](#page-24-0)].

As for the effect of particle size, it was found that most of the back transport effects (except for Brownian diffusion) as well as filtration drag force are favored by larger particle size, and that the overall back transport is more sensitive to particle size than is filtration drag [\[121](#page-25-0)]. Thus, larger particle size should be preferable for the purpose of anti-fouling, and there should be a "critical foulant size" in response to the force balance. This is in accord with the finding that fouling caused by particles with mean diameter smaller than 80 μ m was significantly more severe than by those larger than 80 μ m, based on statistical analysis of mixed liquor samples from various MBRs [\[85\]](#page-24-0).

The comprehensive effect of flux $(J, \text{ in LMH})$, crossflow velocity (U, in $m \cdot s^{-1}$) and SS concentration (X, in $g \cdot L^{-1}$) on the increase rate of TMP (k_R , in $m^{-1} \cdot h^{-1}$) has been quantitatively described by a multiplied power function expressed as:

$$
k_{\rm R} = \lambda X^{\alpha} J^{\beta} U^{\gamma}.
$$
 (1)

This relationship, though somewhat empirical, was found to be statistically valid and parameterizable (e.g., λ) = 8.933 \times 10⁷, α = 0.532, β = 0.376, and γ = -3.047 in the case of Liu et al.'s study) [\[90\]](#page-24-0).

(2) For foulant size comparable with membrane pore size

Colloidal and macromolecular foulants sized about $0.01-1$ µm may participate in all fouling stages, as has been mentioned in Section 2.3.1.2. Herein particularly concerned are their detailed roles in the initial stage and gel layer stage of fouling. For distinguishing these stages, the classical blocking laws for constant-pressure filtration [\[122,123](#page-25-0)] or constant-flux filtration [[124](#page-25-0)] could be useful, describing the mechanisms as standard blocking, complete blocking, cake filtration (or bridging filtration), and intermediate blocking falling empirically between the standard type and the cake type. Cake blocking law is applicable to description of gel layer as well as cake layer for constant-flux filtration, as has been validated by Wang and Waite's work showing that gel layer could be

instantaneously consolidated to acquire a stable depthaverage specific resistance fitting the cake type, despite that the gel layer is highly compressible and far from uniform over its depth [[46](#page-23-0)]. On the other hand, standard blocking, complete blocking and intermediate blocking laws pertain to description of the initial stage of fouling.

For the initial stage of fouling, it was found that HIS was the most responsible for the rapid flux decline in the initial stage of microfiltration using hydrophilically enhanced PVDF membrane, and that the role of the fraction of HIS with $MW > 100$ kDa was particularly prominent despite its relatively low abundance [[82](#page-24-0)]. This should be ascribed to steric effect rather than hydrophobic affinity between membrane and foulant. A considerable overlap between size distributions of membrane pores and foulant with $MW > 100$ kDa was observed [\[88\]](#page-24-0), indicating the possibility of effective size exclusion, which could be further confirmed by the predominance of complete blocking revealed by modeling analysis coupled with atomic force microscopy (AFM) examination [[82](#page-24-0)]. The course of size exclusion may be anatomized into steps as follows: 1) the foulant molecule enters the membrane volume a bit easily as long as at least one dimension of the molecule is smaller than the pore size; and 2) the foulant molecule directly plugs the pore when it rotates with its larger side applied to the pore channel or when it goes to a bottleneck of the passage; or 3) the foulant molecule gets entrapped and "locked" in the membrane matrix if the molecule is flexibly long-chained (which is particularly the case for polysaccharide-like substances) and the pore channels are tortuous and interconnected (which is usually the case for most polymeric membranes) [\[88\]](#page-24-0). These size exclusion processes are illustrated by Fig. 13. In comparison with size exclusion, hydrophobic interaction as well as electrostatic interaction was found insignificant in the initial stage with regard to flux decline (i.e., *apparent* fouling phenomenon) [[82](#page-24-0)]. However, we cannot exclude the possibility that they have profound impacts on following fouling evolution.

For the gel layer stage of fouling, the organics associated are termed gelling soluble and colloidal microbial products (gSCMP), which are mainly composed of polysaccharides (as an example, polysaccharides, proteins and humic substances were shown to be in the ratio of $6.8:1.7:1$ within a gel layer in Wang and Waite's study [\[20\]](#page-22-0)). It is recognized that the gel layer is structurally supported by a network of polysaccharides while other SCMP components are trapped on their passage through the assemblage (and have minor structural contribution) [[20\]](#page-22-0). As a representative polysaccharidic foulant, alginate was widely applied to studies on gel-related fouling mechanisms. A honeycomb-like structure of gel layer formed by Caalginate was observed [\[45\]](#page-23-0). Over the range of the applied pressures of 11.7–135 kPa, the alginate gel layer was found to be very porous with a water content of more than 96% but very low Darcy permeability of less than 1×10^{-17} m²,

Fig. 13 Scheme of fouling-inducing size exclusion processes

possibly due to lack of wide-through channels [[46](#page-23-0)]. The apparent channel size was estimated to be on the order of 5–30 nm when $TMP \le 100$ kPa according to Hagen-Poiseuille relationship [[45](#page-23-0)]. The firm bridging between polysaccharide chains to form a network is principally attributed to covalent interactions, such as that with the assistance of complexation with multivalent metal cations. Calcium ion was found to be a key player in this regard for most cases rather than magnesium ion (which shows slightly weaker complexation than calcium ion) and ferric ion (which is usually less abundant); while carboxylic (and presumably phosphoric) groups were most likely the binding sites. The binding stability constant of CagSCMP was estimated to be ca. 4.9×10^3 M⁻¹, assuming a 1∶1 complexation [\[20\]](#page-22-0).

For the colloidal and macromolecular foulants mentioned in this section, there might also exist a critical flux for the initial stage or a limiting flux (for an observational pseudo fouling-ceasing state) for the following fouling layer stages, as may be inferred from Tang and Leckie's analyses of force balance for the case of nanofiltration (NF) and reverse osmosis (RO) [\[125\]](#page-25-0); but the hypothesis for microfiltration or ultrafiltration is still unproven as far as we know.

(3) For foulant size much smaller than membrane pore size

For foulants with size much smaller than membrane pores, size exclusion should be negligible and adsorptive fouling in the membrane matrix should prevail. The propensity and rate of this kind of fouling could be quantitatively described by adsorption equilibrium and rate constants, determinable via Thomas' dynamic adsorption model based on Langmuir kinetics [\[126,127](#page-25-0)]. It was found that bovine serum albumin (BSA, representative of prot-eins) and Aldrich humic acid (filtered to be $< 0.05 \,\mu m$) showed higher propensity of adsorptive fouling than dextran T70 (DEX, representative of polysaccharides) [[89](#page-24-0)], despite that DEX is almost non-charged exhibiting minor electrostatic repulsion. The driving force is thus considered to be hydrophobic attraction which may very likely overweigh electrostatic repulsion. The membrane-binding affinity (or area-divided interaction energy, associated with adsorption equilibrium constant) is not only dependent upon

hydrophobicity, but should also be positively correlated to the foulant size as suggested by the extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory [[128\]](#page-25-0). Moreover, it was shown that the adsorbed amount (i.e., adsorptive fouling extent) is a function of not only the membrane-binding affinity but also the concentration of foulant in the solution [[89\]](#page-24-0).

2.3.2 Membrane fouling control

2.3.2.1 Membrane and module optimization

A great deal of attention has been paid to hydrophilic enhancement of membrane via, e.g., surface modification, which aims to implant polar organic functional groups onto membrane surface. Surface modification could be conducted by means of plasma treatment, surface grafting, surface coating, surface blending, etc. UV photo-induced graft polymerization of acrylic acid and acrylamide on a PP membrane surface was successfully performed by Yu et al. [[75,76,78](#page-24-0)], with decreased water-contact angle and increased zeta potential (absolute value) observed with increased grafting degree; however, too much grafting amount could in reverse lead to considerable porosity loss. Plasma treatment was found efficient in creating hydrophilic functional groups on membrane surface, with the plasmas in use including air plasma $[129]$ $[129]$ $[129]$, $O₂$ plasma [[130](#page-25-0)], N₂ plasma [[131](#page-25-0)], CO₂ plasma [\[132\]](#page-25-0), H₂O plasma [[133](#page-25-0)], NH_3 plasma [\[134\]](#page-25-0), etc.; whereas, the damage rendered by plasma treatment as well as the instability of plasma-induced hydrophilicity needs to be concerned. Surface coating via adsorption of surfactants was also investigated. Xie et al. [\[135\]](#page-25-0) used polysorbate 20 (also known as Tween 20) to form a monolayer on PP membrane surface, such that the polar ends of Tween 20 molecules were oriented outwards uniformly thereby hydrophilizing the membrane surface. However, the physical tolerance (to, e.g., desorption, cross-flow and aeration) and the biochemical stability of the coating layer under SMBR conditions are also worthy of note. In order to harmonize the requirements of performance (e.g., hydrophilicity), fabrication ease and durability, some researchers suggested that membrane be blended with copolymers comprising various functional segments. As an example, Li et al. [[136](#page-26-0)]

blended PES membrane with a terpolymer of poly (acrylonitrile-acrylic acid-N-vinyl pyrrolidinone) (P(AN-AA-NVP)), in which AN provided water insolubility, AN hydrophilicity and negative charge, and NVP hydrophilicity and miscibility with PES.

Apart from the simple-patterned hydrophilic modification, a number of studies have been devoted to morphological improvement such as, covering membrane outer surface or pore surface with star copolymers [[137](#page-26-0)], or brush or comb copolymers [[138](#page-26-0)–[140\]](#page-26-0) to form a hydrophilic fine-hair layer (as illustrated in Fig. 14) preventing foulant from depositing. Polyethylene glycol (PEG) or PEG-containing copolymers have been usually adopted owing to the flexibly long-chained structure and outstanding hydrophilicity (PEG known as the "most" hydrophilic synthetic polymer [\[128\]](#page-25-0)). The integration of hydrophilic repulsion and steric hindrance could provide membrane with excellent anti-fouling nature. It should be noted however, that excessive modification could be adverse to membrane's mechanical properties [[141](#page-26-0)].

Fig. 14 Scheme of (a) an amphiphilic brush-like copolymer and (b) the attachment of these copolymers to membrane surface (Poly (methyl methacrylate-r-poly(ethylene glycol) methyl ether methacrylate) (P(MMA-r-PEGMA)) blended with PVDF membrane is taken as an example and the pictures obtained from Ref. [\[140\]](#page-26-0))

Recent attention has also been paid to novel membrane modification methods using, e.g., mineral nanoparticles and zwitterionic materials. Li et al. $[142]$ $[142]$ $[142]$ prepared a TiO₂
nanoparticle self-assembly membrane based on nanoparticle self-assembly membrane based on polyvinylidene fluoride/poly(styrene-alt-maleic anhydride) blend. The as-prepared membrane, with nanosized anatase $TiO₂$ crystals observable on the surface, exhibited superior resistance to protein adsorptive fouling. The successful examples of PES membranes modified by $TiO₂$ nanoparticles have also been reported [[143,144\]](#page-26-0). The mechanism of $TiO₂$ bonding with membrane surface was supposed to be complexation or hydrogen-bond-bridged

interaction between titanium and certain functional groups (e.g., carboxyl, ether and sulfone groups) on the membrane [[142](#page-26-0),[143\]](#page-26-0). Zwitterionic polymers, bearing both cationic groups such as tertiary amine or quaternary ammonium groups and anionic groups such as sulfonic or carboxylic groups, have been applied to modification of PES ultrafiltration membranes [[145](#page-26-0)–[147](#page-26-0)]. The zwitterionicity may be expressed in different polymer segments [[145](#page-26-0)] or even in a single monomer unit such as carboxybetaine or sulfobetaine [[146,147\]](#page-26-0). However, to date this modification technology in China has been mostly limited to ultrafiltration membranes with relatively low molecular weight cutoff (MWCO), as far as we know.

As for configuration of, e.g., hollow fiber membranes, the anti-fouling effect of membrane fiber length alteration has been studied. It has been found that the local flux along a fiber is unevenly distributed due to frictional water head loss [\[46\]](#page-23-0), and involves a dynamic redistribution during membrane suction, such that part of the fiber may experience supra-critical flux even when the overall average flux is sub-critical [[104](#page-25-0)]. Thus, shorter membrane fiber length [\[103\]](#page-24-0) (as well as larger inner diameter of fiber [[46](#page-23-0)]) should be preferable for the purpose of fouling mitigation.

2.3.2.2 Mixed liquor conditioning

Dosing the mixed liquor with additives is an important strategy for fouling mitigation. The additives, such as carriers, suspensible particles, coagulants and other chemical agents, can exert their anti-fouling impacts physically or chemically.

Addition of suspended polypropylene carriers (cylindrically shaped, diameter \times length = 3 mm \times 3 mm) was found effective in inhibiting cake layer formation, the mechanism attributable to inertial lift being enhanced by increased shear rate with the addition of the carriers. However, an overdose could break sludge flocs and deteriorate fouling (c.f. Section 2.3.1.3 (1) for the effect of floc size on fouling). The optimum carrier doses were 1%, 1.3% and 2.3% (v/v) at MLSS of 5, 8 and 11 $g \cdot L^{-1}$, respectively [[95](#page-24-0)].

Addition of powdered activated carbon (PAC) sized 1– 80 μ m (median diameter = 39.0 μ m) was found to increase the median diameter of sludge flocs from 48.8 µm to over 57 µm and meanwhile reduce supernatant organics effectively through adsorption, thereby ameliorating fouling. However, an overdose of PAC could reduce critical flux because excess PAC itself could be a foulant [[107\]](#page-25-0). The effect of powdered zeolite addition was also investigated by a few researchers, finding that zeolite could modify some properties of sludge flocs and could likely form a "protection coating layer" over the membrane surface whereby benefitting filterability [[148,149\]](#page-26-0). However, like PAC, an overdose of powdered zeolite could also exacerbate fouling [[150\]](#page-26-0).

Inorganic coagulants, including aluminum sulfate, ferric chloride, polyaluminum chloride (PACl) and polyferric sulfate (PFS), were utilized for mixed liquor conditioning and their anti-fouling effect compared in Wu et al.'s study [\[105\]](#page-25-0). Through the addition of inorganic coagulants, the floc size was enlarged accompanied by the reduction of supernatant TOC, and consequently the specific filtration resistance of fouling layer could be reduced. It was found that the polymeric coagulants (especially PFS) were more effective in this regard than those monomeric ones. Moreover, considering the possibility that these coagulants might cause inorganic fouling, a long-term observation of PFS's performance in an SMBR fed with domestic wastewater was conducted. At the optimal PFS dosage of 1.0 mM-Fe-equivalent and the dosing interval of 15–30 d, no substantial deposition of the exotic Fe on the membrane surface was observed, and neither did severe inorganic fouling occur [[106](#page-25-0)]. The addition of Fe-containing coagulant could be otherwise implemented via electrolysis of iron-anode plate in an electrocoagulation-MBR (EC-MBR) whereby iron ions could be released into the mixed liquor decelerate fouling evolution [\[40\]](#page-23-0). The anti-fouling effect of organic flocculants such as polyacrylamide (PAM) and chitosan was also studied. At the dosages of \leq 20 mg·L⁻¹ and \leq 150 mg·L⁻¹ for PAM and chitosan respectively, membrane fouling was significantly alleviated, as a concomitant of increased floc size and decreased SMP concentration [\[84\]](#page-24-0).

The effect of ozone addition on mixed liquor conditioning was also investigated. It was found that ozone at a dosage of less than $0.7 \text{ mg} \cdot \text{g}^{-1}$ -SS could lead to an enhanced filterability in initial stages as well as a decelerated fouling evolution. The underlying mechanism was deduced to be the ozone-induced re-flocculation. The LB-EPS surrounding the sludge flocs may be "trimmed" off by ozone, rendering the flocs higher hydrophobicity and lower surface charge so that destabilization of the flocs was achievable. The mean diameter of the flocs was observed to grow from $49.9 \mu m$ to $69.8 \mu m$ due to the re-flocculation, thereby mitigating fouling possibly according to the above-mentioned size effects (c.f. Section 2.3.1.3 (1)). However, a dosage over $0.7 \,\text{mg} \cdot \text{g}^{-1}$ -SS could break the flocs and release colloidal and soluble organics, which could contrarily deteriorating fouling [[151\]](#page-26-0).

2.3.2.3 Operational improvement

The effect of shear rate and filtration flux on fouling has been discussed above (see Section 2.3.1.3), as has the existence of their critical values for anti-fouling. Moreover, it is also recognized that the suction/relaxation alternation could have significant impacts [\[91\]](#page-24-0). It was found that in one suction/relaxation circle, too long suction-time and too short relaxation-time rendered severe fouling [[92](#page-24-0)], while shortened suction-time and prolonged relaxation-time over an appropriate range could alleviate fouling but with the effectiveness of further adjustment limited [\[96\]](#page-24-0). It is worthy of additional note that, the suction/relaxation-time ratio did not show monotonic correlation with fouling propensity [[92](#page-24-0)], thus may create difficulties with predictive assessment of parameter optimization.

2.3.2.4 Membrane cleaning

Apart from the most well-known physical cleaning measures such as tap-water flushing, permeate rinsing, cross-flow scouring, backwashing and soaking, ultrasonication has also been found useful, especially for on-line physical cleaning. Periodic sonication was successfully applied to aerobic MBR [\[152\]](#page-26-0) as well as to anaerobic MBR [[153](#page-26-0)]. Liu et al. showed that ultrasound was particularly effective in alleviating gel-type fouling [\[152](#page-26-0)]. However, excessive ultrasonic intensity and working time could break sludge flocs and release soluble and colloidal organics thereby exacerbating fouling [[154,155\]](#page-26-0). Moreover, prolonged duration of sonication might be adverse to sludge activity (especially to nitrification activity) [\[154\]](#page-26-0). It should be also noted that, the ultrasound-associated acoustic cavitation may cause damage to the membrane. Proper selection of ultrasonic intensity and working time, and keeping a certain thickness of fouling layer on membrane surface could be effective ways to protect the membrane against damage [\[156\]](#page-26-0).

The most commonly utilized chemical cleaning reagents include sodium hypochlorite (NaClO), sodium hydroxide (NaOH), hydrochloric acid (HCl), citric acid, etc. It is widely accepted that the oxidizing agent NaClO can remove most of the microorganisms and organic foulants from membrane effectively [[157,158\]](#page-26-0). NaOH can also remove a wide variety of organic foulants [\[158\]](#page-26-0), possibly via alkaline hydrolysis or dissolution. HCl is effective in breaking metal-associated structures including metalorganic foulant complexation [[159](#page-26-0)] and inorganic scales [[157](#page-26-0)], presumably in the light of proton's activity; and it can also remove part of polysaccharide-like substances [[158](#page-26-0)]. Citric acid can remove metallic foulants as well, due to its acidity [\[160\]](#page-26-0) and/or its outcompetitive binding ability with metal ions (e.g., the binding stability constants being $5.0 \times 10^4 \text{ M}^{-1}$, $3.2 \times 10^9 \text{ M}^{-1}$ and $2.0 \times 10^{12} \text{ M}^{-1}$ for CaL (L denoting citric acid ligand), CaHL and CaH2L, respectively [\[161\]](#page-26-0), compared to 4.9×10^{3} M⁻¹ for CagSCMP [[20](#page-22-0)]). On-line chemical cleaning using NaClO was applied to a pilot-scale SMBR treating municipal wastewater and its long-term fouling behavior observed in Wei et al.'s study [[16](#page-22-0)]. The MLSS level was around $10 \text{ g} \cdot L^{-1}$ and the filtration flux was maintained at 30 LMH. When the TMP reached 15 kPa, the membrane was backwashed with NaClO solution at a dosage of $4-6$ g-Cl \cdot m⁻²membrane (or $54-81$ mg-Cl \cdot L⁻¹-reactor effective volume). During 190 days of operation, the on-line chemical cleaning

was carried out for 7 times, controlling fouling effectively without off-line chemical cleaning. Meanwhile, the on-line chemical cleaning exhibited little effect on sludge activity and removal efficiency of COD and $NH₃-N$, therefore verifying the feasibility of this anti-fouling strategy.

3 Commercial application

The application of MBR technology in China up to now may have experienced several stages as follows: (1) laboratory experiments, pilot-scale tests and a few demonstration projects from 1990 to 2000, (2) practical application on the scale of hundreds of tons per day mainly serving for small residential areas and industrial fields from 2000 to 2003, (3) practical application on the scale of thousands of tons per day for municipal and industrial wastewater treatment from 2003 to 2006, and (4) practical application of large-scale MBRs (of over $10000 \text{ m}^3 \cdot \text{d}^{-1}$) from 2006 to the present. The earliest practical MBRs in China, in the beginning of 2000s, were on the scale of only several ten tons per day; whereas by the end of 2009, there had been approximately a total of more than a hundred fullscale MBR installations in service (only taking into account those larger than $100 \text{ m}^3 \cdot \text{d}^{-1}$, providing a treatment amount of nearly $1.2 \times 10^6 \text{ m}^3 \cdot \text{d}^{-1}$. Among these installations, there are nearly 30 large-scale MBR plants each with a designed capacity greater than $10000 \text{ m}^3 \cdot \text{d}^{-1}$, as enumerated representatively in Table 3. In addition, about another 8 large-scale MBR plants with a total capacity of $3.7 \times 10^5 \text{ m}^3 \cdot \text{d}^{-1}$ have been contracted and are expected to be commissioned in 2010. Among all large-scale MBRs particularly noteworthy are Miyun MBR plant (in Beijing) and Shiyan Shending River MBR plant (in Hubei Province) (Fig.15), which are landmarks of MBR application in China. Miyun MBR was the first to reach the capacity of $10000 \text{ m}^3 \cdot \text{d}^{-1}$; and Shiyan Shending River MBR, commissioned in October 2009 for municipal wastewater treatment, is so far the largest MBR plant in China, possessing a capacity of $110000 \text{ m}^3 \cdot \text{d}^{-1}$. Now, China has become one of the most active countries in the world for MBR application. The expansion of domestic MBR market has been tremendous; it is expected that in the next 5 years it will keep on growing at an annual rate of around 50%, which should be much higher than the global average rate.

MBR technology has been commercially applied to the treatment of a variety of wastewaters in China, including municipal wastewater, industrial wastewater, landfill leachate, bathing wastewater, hospital wastewater, etc., among which municipal wastewater treatment and industrial wastewater treatment account for about 60% and 30% of the total capacity, respectively. In the industrial sector, about half the capacity is devoted to petrochemical wastewater, which is followed by chemical industrial wastewater, food processing wastewater, dyeing

 (b)

Fig. 15 View of (a) Miyun MBR plant (in Beijing) for municipal wastewater treatment (design capacity: $45000 \text{ m}^3 \cdot \text{d}^{-1}$) and (b) Shiyan Shending River MBR plant (in Hubei Province) for municipal wastewater treatment (design capacity: $110000 \text{ m}^3 \cdot \text{d}^{-1}$)

wastewater and the others.

The major membrane unit suppliers in China now are Asahi Kasei (Japan), Mitsubishi Rayon (Japan), GE (USA), Siemens Memcor (Germany), Origin Water (China), Memstar (Singapore), Tianjin Motimo (China), Norit (Netherlands), Litree (China), SINAP (China), etc. Besides overseas-funded membrane suppliers, Chinese companies have also shown noticeable contribution. Different from the European MBR market, hollow fiber membrane modules are prevailing in the China MBR market.

As for the professional companies dealing with the engineering design, equipment manufacture and operation management of MBR plants, besides some well known overseas-funded companies like GE, Siemens and NOVO (Singapore), a lot of domestic companies have grown up such as Origin Water, Tianjin Motimo, etc. Origin Water, GE, NOVO and Siemens are now leading the market as the top four, according to the summed capacity of the largeand medium-sized plants built by the end of 2009.

Of the various factors influencing the MBR markets in China, water scarcity is the most important one. This problem, particularly severe in the northeastern and northwestern areas, is further exacerbated by water pollution. In China, there is a law specially drawn up to manage the water resources of the country. It is "*The water* law of the People*'*s Republic of China" revised in 2002, where wastewater reclamation and reuse are encouraged by the 52nd item. The government further issued national standards for reclaimed water to promote wastewater reuse (GB/T 18919-2002, GB/T 18920-2002, GB/T 18921- 2002, GB/T 19772-2005, GB/T 19923-2005 and GB 20922-2007 for classification of wastewater reuse, urban miscellaneous uses, scenic environment uses, groundwater recharge, industrial use and farmland irrigation, respectively). Very extensive works have demonstrated that the MBR effluent is qualified for national reclaimed water standards; thus MBR application has earned promotion in this context. In addition, the serious eutrophication in some sensitive drainage basins (such as Tai Lake basin and Dian Lake basin) has accelerated the drawing up of more stringent discharge regulations by local governments to prevent the deterioration of water quality. As an example, the installation of four MBRs in Wuxi City (by Tai Lake) in 2009 was motivated by this need. Despite the big area of China, some large cities still do not have enough land available for construction of municipal wastewater treatment plants. The advantage of MBR with small footprint is especially attractive in these areas. Besides, remarkable decrease in MBR investment costs as well as increasing acceptance and maturity of this technology (especially those owned by domestic companies) should also be the driving force for the tremendous growth of the MBR market.

Wider and larger application of MBR in China in the future is foreseeable; however, the economic factors including higher investment and running costs compared to conventional processes are also substantial. As can be seen in Table 3, so far most large-scale MBRs treating municipal wastewater have been centered in Beijing and Jiangsu Province, both of which are more developed than most of the other provinces. In addition, standardized guidance for engineering design, equipment manufacture and operation management of MBRs should be formulated to regulate MBR application.

4 Conclusions

MBR, a newly introduced technology to China excelling in wastewater treatment, has attracted extensive attention in the recent 15 years. Great progress has been achieved on both academic research and commercial application of MBR in the country over the last few years. China has now become one of the world's most active fields in this respect. In light of the increasing requirement and

expanding market, it is expected that this technology will be further promoted, developed and applied at a tremendous speed. However, in order to support the rapid development of MBR in China, there are still critical challenges from both technological and managerial perspectives, such as: (1) further enhancement of treatment performance via process optimization in response to increasingly stringent standards, (2) extension of MBR's functionality to fulfill updating requirements of pollutant removal (such as elimination of emerging pollutants), (3) further reduction of membrane cost and improvement of membrane quality (e.g., with prolonged membrane lifetime), (4) development of robust anti-fouling strategies, and (5) regulation of the MBR market and development of standardized guidance for engineering design, equipment manufacture and operation management.

Acknowledgements This work was supported by the National Science Fund for Distinguished Young Scholars (Grant No. 50725827) and the National High Technology Research and Development Program (863 program) of China (Grant No. 2009AA062901).

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Xia Huang received her PhD degree in Environmental Chemistry and Engineering from Tokyo Institute of Technology, Japan in 1988. Then she joined the Department of Environmental Science and Engineering, Tsinghua University, and became associate professor in 1992 and got the full professor position in 1997. She worked in Department of Ecological Engineering of Toyohashi

Institute of Technology, Japan from 3/1995 to 3/1997 as an associated professor. She focuses on membrane based wastewater treatment processes, microbial fuel cell for wastewater treatment and electricity generation, and novel nitrogen and phosphorus removal processes. She has published more than 100 papers in recent 5 years, and was awarded Environ. Sci. Technol. 2009 best paper, the 2nd Class of the State Science and Technology Progress Award twice and several awards issued by Ministry of Education of China.