



Review

Membrane bioreactor: Applications and limitations in treating high strength industrial wastewater



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HIGHLIGHTS

- The state of the art of membrane bioreactor (MBR) technology for application to high strength wastewater is reviewed.
- Treatment in MBRs can be very effective.
- The major impediment to more widespread application is membrane fouling.
- Mitigation of fouling can use physical or chemical methods, or combination of both.

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ABSTRACT

This paper reviewed the application of membrane bioreactor in treating high strength industrial wastewater by analysing operational parameters, limitations and mitigations of MBR for industrial wastewater. High strength industrial wastewater is difficult to classify but by its characteristics can be predicted using biodegradability criteria (BOD₅/COD). Several factors need to be taken into consideration to find suitable operating parameters such as hydraulic retention time (HRT), solid retention time (SRT), mixed liquor suspended solid (MLSS), food to microorganism (F/M), transmembrane pressure (TMP) and Flux (J) to obtain good quality effluent and reduce the fouling effect. Fouling factors by membrane, biomass and MBR operation need to be taken seriously because they are the major problems affecting the performance of the MBR and quality of the effluent. There are specific methods to reduce and clean the clogging membrane depending on the level of severity of the fouling. The mitigation covers physical cleaning such as membrane relaxing, backwashing, a combination of both and chemical cleaning which is used for irreversible fouling. In some cases, modification of MBR is needed to improve the performance and to achieve high quality of effluent.

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Contents

1. Introduction	110
2. High strength industrial wastewater behaviour	110
3. A general MBR configuration	111
3.1. Membrane behaviour	112

Abbreviations: BAP, biomass-associated products; BOD₅, biological oxygen demand; CAS, conventional activated sludge; EMBR, extractive MBR; EPS, extra cellular polymeric substance; Floc, suspended solid particle of the mixed liquor; Flux, volumetric flow rate per unit membrane area (LMH or m/d); F/M, food per microorganism; FS, flat sheet; HF, hollow fibre; HRT, hydraulic retention time (the time taken for the liquid phase to pass through a tank); iMBR, immersed MBR; MABR, membrane aerated biofilm reactor; MBR, membrane bioreactor; MF, microfiltration; Mixed liquor, the material formed in the bioreactor, containing biomass and other solids; MLSS, mixed liquor suspended solids; NF, nanofiltration; PAC, powdered activated carbon; Permeability, flux per unit TMP; PE, polyethylene; PES, polyethylsulphone; PP, polypropylene; PVDF, polyvinylidene; sMBR, sidestream MBR; SMBR, submerged MBR; SMP, soluble microbial product; SND, simultaneous nitrification denitrification; SRT, solids retention time: the time taken for the solid (particulate) phase to pass through a tank; TKN, total Kjeldahl nitrogen; TMP, transmembrane pressure (Pa); TOC, total organic compound; TSS, total suspended solid (mg/L); UAP, substrate utilisation-associated products; UF, ultrafiltration.

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3.2. Biomass behaviour	113
4. MBR application in industrial wastewater treatment	114
4.1. Textile industries	114
4.2. Food industries	115
5. MBR: limitation and mitigation.	115
5.1. Membrane fouling	115
5.2. Fouling mitigation	117
6. Conclusion	118
References	118

1. Introduction

Membrane bioreactor (MBR) is not a new technology in water and wastewater treatment but the capability in treating wastewater cannot be disputed. Researches on MBR are ongoing to make more effective and robust towards the varieties of strength ranges and compounds of wastewater. This system is a hybrid of biological treatment and filtration but in some cases, chemicals are introduced to enhance its performance. In the industrial sector, the performance of MBR has been widely studied since the early 1990s when the first large installation of MBR was carried out in the United States by General Motors at its plant in Mansfield, Ohio [1]. In 1998, the first large scale internal MBR system to treat wastewater from the food industry was installed in North America [2].

The research on MBR has declined due to the difficulties in obtaining the membrane and the high capital and maintenance costs of the system. In the 1990s, submerged MBR was commercialised and it was found to have low operational cost [3] when compared with other types of MBR. In MBR, biological processes play major roles than filtration processes [4] where particulates in wastewater are converted into end products before filtration is carried out by the membrane.

MBR is also known as an alternative for conventional activated sludge (CAS) treatment where clarifier is removed and substituted with membrane to overcome settleability problem when undesired biomass is formed. MBR also can give high performance in treating water besides having small footprints compared to conventional activated sludge where clarifiers are eliminated. MBR also produces high quality effluent [5], is good in removing organic and inorganic contaminants, capable of resisting high organic loading [6] and generates less sludge [3]. With all the advantages of MBR, some industries install the MBR to minimise the cost of water by reusing the treated water for other processes. For instance, the treated water can be used in industrial sanitary and landscape purposes. High-quality treated water from MBR is reused for heat integration and processing by ensuring the treated water have small amounts of contaminants to prevent the breakdown of sensitive equipment or pipes [7].

Besides the high costs of MBR, membrane fouling is a major factor and investigation on how to reduce this fouling is still ongoing [8–11]. Other constraints of concern are the limitations in pH, temperature, pressure and some corrosive chemicals [3,7,12]. Fouling contaminates not only the microbes in the reactor but also destroys the membrane. However, through some researches, the membrane life can be prolonged [13–16]. Recently, some researchers have carried out modifications and integration of MBR to minimise the constraints [15–17]. The most common manufacturers of MBR are Kubota from Japan, Zenon from Canada and Mitsubishi. Kubota has dominated the installation meanwhile Zenon has installed more capacity water treated through their membrane [7]. In industries, the advantages during handling MBR are to create the optimum condition to operate MBR for high strength industrial wastewater and shock loading rate and also to control the biofouling effect and reduce energy consumption during operational [3].

Recently, fouling effect by EPS, SMP and inorganic compound dominate in MBR research. Unfortunately, less information research on this fouling effect applied for high strength industrial wastewater. This information is necessary to improve the performance of MBR in industries. This review paper is to discuss the general application of MBR in treating high strength industrial wastewater which covers membrane and biomass behaviours, MBR limitation and its mitigation.

2. High strength industrial wastewater behaviour

Nowadays, industries have to facing the critical issues towards achieving the stringent water discharge requirement. Generally in industries, the wastewater comes from a variety of streams for instance from production line, cooling tower or boiler that content diverse substances (i.e. organic and inorganic compounds, virus, bacteria and toxic compound). The diverse substances lead to reduce the ability of the conventional biological treatment due to low biomass body strength.

High strength industrial wastewater is difficult to classify as there is no specific range to differentiate between low, medium and high strength of wastewater in the industries. The strength can be different from one industry to another due to the different chemicals used during the main process i.e. food industries and chemical industries. Summer stated that high strength wastewater is defined as ‘the wastewater that contains fats, oil and grease or other organic compounds in great amount according to the types of sources that take part’ [18]. Basically it is called high strength because the elements in the wastewater are in large quantities such as high amount of COD [19], ammonia, suspended solid or heavy metal [20] and sometimes shock loading will happen. Commercially, industrial and institutional wastewaters are considered as high strength wastewater. In areas of biodegradable wastewater for instance, COD is deemed as low strength level when it is less than 1000 mg/L [21]. For example, even though petrochemical effluent have 1000 mg/L COD, it is considered high strength level whereas in the food industries, 1000 mg/L COD is considered as medium strength. This is because chemical industries contain ‘hard’ COD with high content non-biodegradable compounds such as heavy metals. Wastewater from the food industries mostly contain high biodegradable compounds such as nitrogen or phosphorus elements [19,22].

The wastewater COD “hardness” is based on the biodegradable and non-biodegradable elements contained in the wastewater. High wastewater BOD₅/COD ratio indicates ready biodegradability while low BOD₅/COD ratio indicates slow biodegradability or that the wastewater contains part of non-biodegradable elements. Slow biodegradability is caused from diverse unknown constituents in industrial wastewater that lead to fouling problems [22,23]. Durai and Rajasimman (2011) stated that 0.3 BOD₅/COD ratio for tannery wastewater is low when compared with domestic wastewater ratio of 0.5 because the latter contains BOD₅ inhibitors [24]. Samudro and Mangkoedihardjo stated that biodegradability is a measurement of allowable level of organic matter that can be used as

Table 1
High strength wastewater characteristic.

Industry	COD (mg/L)	BOD ₅ (mg/L)	BOD ₅ /COD	NH ₄ -N (mg/L)	TSS (mg/L)	SO ₄ ²⁻ (mg/L)	PO ₄ ³⁻ (mg/L)	Oil (mg/L)
Tannery [19]	16,000	5000	0.313	450	–	–	–	–
Textile [28]	6000	700	0.117	20	–	–	120	–
Textile [29]	4000	500	0.125	4.8	–	200	2	–
Dyeing [30]	1300	250	0.192	100	200	–	–	40
Textile [31]	1500	500	0.333	50	140	–	7	–
Wheat starch [2]	35,000	16,000	0.457	–	13,300	–	–	–
Dairy [19]	3500	2200	0.629	120	–	–	–	–
Beverage [19]	1800	1000	0.556	–	–	–	–	–
Palm oil [16]	67,000	34,000	0.507	50	24,000	–	–	100,000
Pet food [32]	21,000	10,000	0.476	110	54,000	–	200	–
Dairy product [33]	880	680	0.773	–	2480	–	–	–
Pharmaceutical [34]	6300	3225	0.512	–	1679	–	–	–

indicator to know the level of wastewater [25]. This ratio describes biodegradability level of materials by which organic matter containing wastewater is readily broken down in the environment. Besides that, this ratio shows the level of allowable organic matter to be degraded by biomass [25]. Generally, BOD₅/COD ratio of 0.5 is considered as readily biodegradable [22]. Kumfer et al. for example, showed the biodegradability as greater than 0.5 for spent caustic wastewater after treatment by wet air oxidation (WAO) [26]. If the ratio value is less than 0.5, the wastewater needs to have physical or chemical treatment before biological treatment takes place [25,26]. Treating high strength spent caustic with biodegradability value 0.08–0.1 show the ability of biomass to remove COD and sulphide [27]. Table 1 shows the characteristics of high strength wastewater for different industries. The biodegradability ratios for textile and tannery industries are low when compared with food industries due to the high content of 'hard' COD and other diverse pollutants.

High strength industrial wastewaters have big impacts on pipelines and equipment for boiler, cooling water, irrigation, maintenance and landscape [35]. The contaminants lead to clogging, corrosion, scale formation, biological growth and foaming in the pipelines and equipment and require high maintenance cost.

The treated wastewater would then be discharged into the environment while some industries reuse treated water for landscaping or housekeeping. However, the treated water must meet the standards that have been set in the environment regulation to avoid environmental pollution [36].

3. A general MBR configuration

In CAS treatment, large clarifying basins are needed to make sure the flocs are completely settled. High power for diffuser is used in aeration basin to make sure the nutrients are totally converted to the end products. The difference when using MBR is that there are no more settling processes needed and the area used for clarifier can be eliminated besides acting as a separator [37].

In industries, MBR is used as secondary treatment so as to reduce biodegradable and non-biodegradable matter in the end product or in advance treatment to remove residual nutrients which are not fully removed during secondary treatment [38]. The basic view of MBR configuration is important before the MBR modification for enhancement is made. Fig. 1 shows the basic schematic diagram of MBR configuration. Fig. 1a shows a side-stream or external membrane module while Fig. 1b shows an immersed membrane bioreactor (iMBR) or submerged membrane bioreactor (sMBR) module [37,39]. For sMBR system, the feed wastewater is directly in contact with biomass. Wastewater and biomass are both pumped through the recirculation loop consisting of membranes. The concentrated sludge is recycled back to the reactor while the water effluent is discharged. The idea of separating the membrane and bioreactor is to ease the membrane maintenance but it will increase the operational cost due to recirculation loop installation [40].

The iMBR system has less operational cost because there is no recirculation loop compared to the sMBR system and a biological process occurs around the membrane in iMBR. Both iMBR and sMBR need to pump out the excess sludge to maintain sludge age. The mode of membrane transportation could be pressure driven or vacuum driven. Radjenovic et al. stated that pressure-driven filtration is used in sMBR and vacuum-driven is used for iMBR, which operates in dead-end mode [7]. The air bubbles are supplied to both systems for aeration besides scouring, especially for the immersed system to reduce membrane fouling in cross-flow effect across the membrane surface [5,10]. There are also aerobic and anaerobic MBR where oxygen acts as an important medium for the microbial growth in the aerobic process whilst anaerobic is done without oxygen. Anaerobic MBR is less efficient in removing COD and takes a long time for start-up. Usually, anaerobic treatment is used for treating high strength wastewater at low temperature which is suitable for microbial growth. Moreover, it is difficult to adjust low temperature for the waste feed and it causes high fouling compared with aerobic at low flux [39].

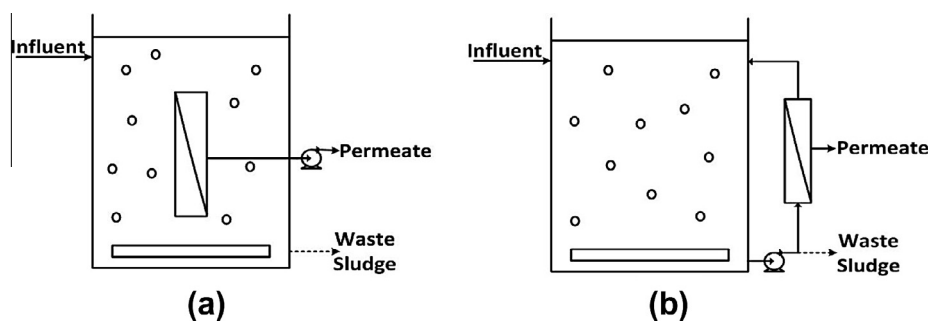


Fig. 1. Basic schematic of MBR. (a) Immersed MBR, and (b) side-stream MBR.

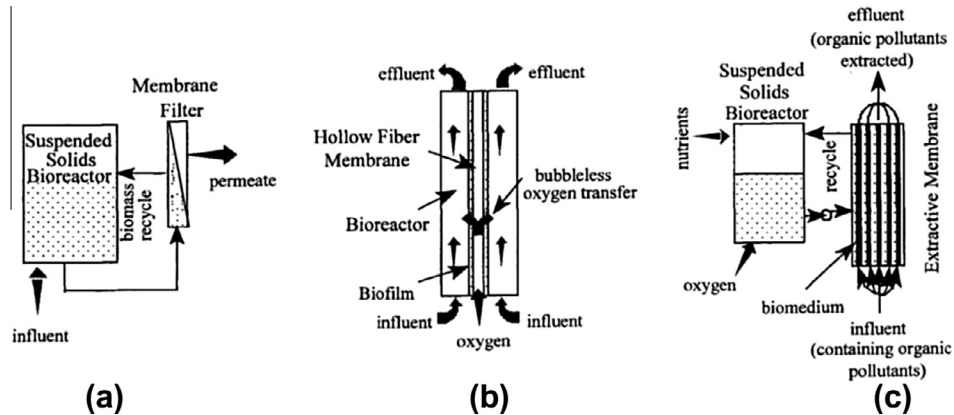


Fig. 2. Types of MBR processes. (a) MBR biomass separation, (b) membrane aeration bioreactor, and (c) extraction MBR [42].

Fig. 2 shows three different processes of membrane bioreactor that have been modified according to the types of wastewater. Fig. 2a MBR shows biomass separation, Fig. 2b is a membrane aeration bioreactor (also called membrane aerated biofilm reactor (MABR)) and Fig. 2c is an extractive MBR (EMBR). However MABR and EMBR were applied in a pilot scale for industrial wastewater. MABR directly used purified oxygen without bubble formation for biofilm growth on the external side of the membrane. The organic matters were biodegraded within the biofilm under aerobic condition and the oxygen supplied was almost 100% utilised for biodegradation. It is difficult in maintaining the optimum biofilm thickness to get enough oxidation, otherwise excessive biofilm can cause liquid flow problem [41]. EMBR is normally operated for high concentration of inorganic compounds such as high salinity and extreme pH value that might inhibit the biodegradation process. EMBR selectively extracts specific organic pollutants (i.e. phenol, hydrogen sulphide and some inorganic) that can be degraded in separated bioreactor [42].

3.1. Membrane behaviour

The importance of studying membrane behaviour is to select good quality membrane in treating high strength industrial wastewater. High strength wastewater consists of diverse contaminants that could possibly corrode the membrane and lead to operational failure. The efficiency of the membrane also depends on the size of pores, types of materials, types of wastewater to be treated, solubility and retention time. Retention is observed due to the MLSS concentration change between the retentate (a part of solution that cannot cross over the membrane) and permeate (solution after filtration). Permeability, flux, pressure (TMP) and resistance are the parameters that also need to be considered. Permeability is flux per pressure ($J/\Delta P$) or LMH/ ΔkPa . Flux (LMH) is the flow of permeate per unit of membrane (component accessibility to the membrane) and it is related to hydraulic resistance, thickness of the membrane or cake layer and driven force. Driven force is the gradients of membrane potential area (unit area of the membrane) of mass transport that involve pressure and concentration of particles. The mass transport mechanism for the membrane depends on the structure and materials of the membrane. Membrane structure plays an important role in transport mechanism whether the structure is parallel or in series. Diffusion and solubility of the component are related to the kinetic ability of mass transport for membrane. For the membrane itself, pore-size membrane participates in kinetic mass transport. [43].

The types of membranes used are different depending on the size of contaminants contacting during the treatment process.

Basically contaminants with the size of a particle from 100 to 1000 nm use microfiltration (MF) for removing suspended particles, ultrafiltration (UF) for particle size 5–100 nm (bacteria and virus), and nanofiltration, (NF) for particles with size 1–5 nm for dissolved particles. In treating high strength industrial wastewater with shock loading of matter, microfiltration is chosen among the others in order to prolong membrane usage. Most of the treatment plants use MF or UF instead of NF with regard to fouling and cost factors [3,7,10,39].

Two types of materials are used to construct membranes – polymeric and ceramic. Ceramic membrane is usually used for industrial wastewaters and has a good performance in filtration compared to polymer due to its high chemical resistance, is inert and easy to clean [44–46]. Chemical stability does not only depend on the materials used but also on the size of the pore where it reduces the stability of the membrane when the structure is too fine. Ceramic also has higher hydrophilic ability due to the water contact angle. However, the main setback of ceramic membrane is it is very expensive to fabricate and it is easily breakable [45].

However, in recent membrane technology development, polymers have been used commercially in the form of PVDF, PES, PE and PP because of good physical and chemical resistance. Polymer membrane (porous membrane) has its own weaknesses where it can foul easily because of its hydrophobic characteristic. The hydrophobic membrane is used because the pore size can easily be fabricated. Hydrophobic membrane weakness can be improved by coating the membrane with hydrophilic polymer [47]. PE is more quickly fouled compared to PVDF [3]. Membrane configuration also play an important part since every configuration has its own advantages and disadvantages based on the cost, capability to withstand turbulence and back-flushing (normally suitable for HF membrane) [39].

In membrane application, there are two types of membrane operations – dead-end and cross-flow operations and are shown in Fig. 3a and b. Both are pressure driven (TMP) with the dead ends filtered perpendicularly to the membrane surface. The solids from the feed that are greater than pore size are easier to deposit on the membrane surface. Most dead-end processes are in batch process [48]. Cross-flow is liquid flow parallel to a filter surface and suspended particles are transported across membrane surface by permeate flow due to pressure drop. Basically this type of filtration is carried out by using hollow fibre (HF), flat sheet (FS) or multi tubular (MT) elements. Cross-flow filtration can reduce formation of cake layer on the surface of the membrane [39,48].

Critical flux is an important parameter that needs to be considered during MBR operation. It is a value of flux that exists as irreversible deposit. There is no fouling below the critical flux. Critical

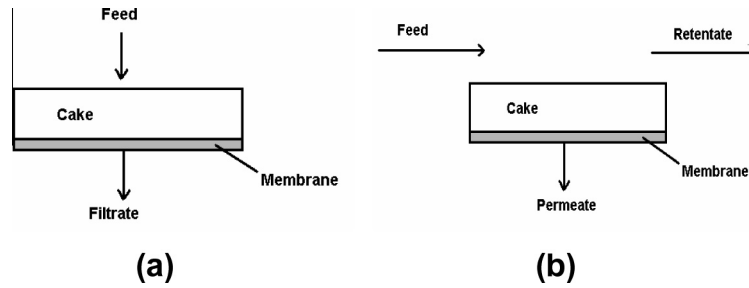


Fig. 3. (a) Dead-end filtration, and (b) cross-flow filtration [48].

flux happens when a thick cake layer forms on the membrane. Fouling could happen when the flux is above critical flux. From critical flux, the suitable flux for the operation can be defined based on the TMP sustainability. There is no standard method to find the critical flux due to the difficulties during reporting data. One practical method that can be used, which is flux-step method, is shown in Fig. 4. This method is relevant for short term critical flux operation and not relevant for long term operation. There are two concepts of flux, strong and weak. In the strong concept the flux obtained during sub-critical is equal to clean water flux but this concept is not relevant with MBR due to high sludge found in the reactor. In the weak concept, the flux is obtained during operation start-up and is maintained for period of time but is not necessarily equal to clean water flux [49]. The highest flux can be determined when the flux is increased and there is no TMP increment or less permeate. It is shown when fouling is about to happen. Eqs. (1)–(4) are used to define the fouling performance for each flux-step [50].

$$\text{Initial TMP increase : } \Delta P_0 = \text{TMP}_i^n - \text{TMP}_f^{n-1} \quad (1)$$

$$\text{Rate of TMP increase : } \frac{dP}{dt} = \frac{\text{TMP}_f^n - \text{TMP}_i^n}{t_f^n - t_i^n} \quad (2)$$

$$\text{Average TMP : } \frac{\text{TMP}_f^n - \text{TMP}_i^n}{2} \quad (3)$$

$$\text{Permeability of the system : } K = \frac{1}{P_{\text{avg}}} \quad (4)$$

From the ensuing calculation, the relationship between the fouling rate (dP/dt), permeability (J/dP) and flux is plotted. The critical flux is the intersection between permeability and fouling rate

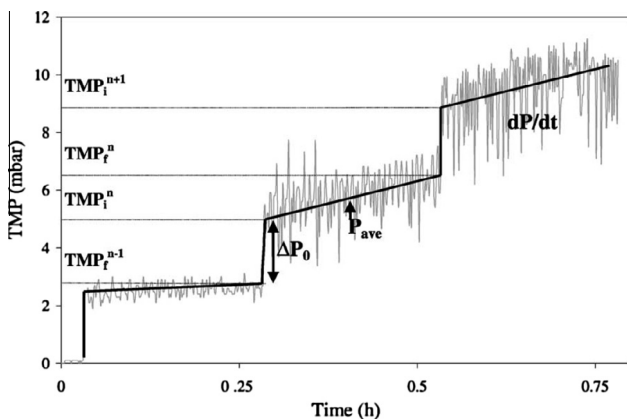


Fig. 4. Flux-step method [50].

line [51] with a typical one shown in Fig. 5. Xu et al. [53] shows the natural flux mode method to identify critical flux at Fig. 6 with the listing of critical flux identification method shown in Table 2.

3.2. Biomass behaviour

Biomass in activated sludge from industries is heterogenous with the basic nutrients being glucose, nitrogen and phosphorus in the ratio 100:5:1 by weight. The domination of biomass can occur through the acclimation process and this depends on the major constituent of feed wastewater. CAS has been used for a long time but with respect to high strength wastewater, this method is not able to cope with the high content of organic loading and inorganic matter because of low biodegradability and inhibition and in some cases it can destroy the microbes because of shock loading of matters [56]. This is because microbes take a long time to biodegrade the inorganic matters and need high concentration of biomass (MLSS) to ensure all the organics are totally biodegraded.

Basically, SRT is operated coupled with HRT for CAS. SRT is the solids or flocs growth in many sizes that need to be retained in the plant before it settles down for a period of time while HRT is the time taken for the organic matters to pass through the plant. This means that CAS relies on both to ensure the flocs are really settled before going for other treatments [22,39]. Similarly, CAS needs biomass with fast growth and flocs formation species. If the biomass has low growth rate, it will lead to washing-out together with the excess sludge because of the shortage of SRT. Consequently, the production of sludge and F/M ratio is high and will end up with high excess sludge for disposal which will increase the total cost of wastewater treatment by about 50–60% [7].

To get the best performance in treating high strength wastewater, the MLSS must be high to increase the process of degradation. One of the treatment problems with MBR is that the increased MLSS hastens the membrane fouling due to high suspended solid [54,57]. During acclimation, long biomass adaptation is needed to degrade complex pollutants in high strength industrial wastewater

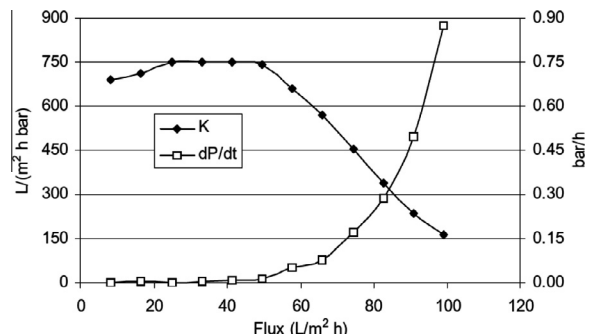


Fig. 5. Relationship between permeability, fouling rate and flux to critical flux determination [49,52].

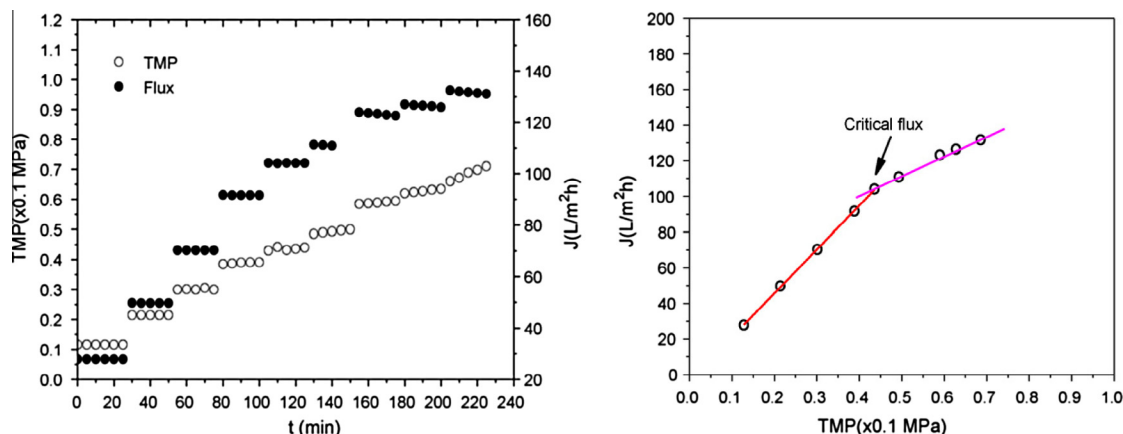


Fig. 6. Natural flux mode method [53].

Table 2
Criteria for critical flux identification.

Reference	Criteria of critical flux
Le-Clech et al. [49]	$dp/dt = 0$ (strong concept) $dp/dt < 0.1$
Bottino et al. and Damayanti et al. [54,55]	$dp/dt \geq 0.5$
Xu et al. [53]	Natural flux mode method (flux versus TMP)
Le-Clech et al. and Pollice et al. [49,52]	Mean permeability versus flux

and achieve high quality effluent [23]. On the other hand, in MBR system, SRT and HRT do not rely on each other because MBR is more on the membrane filtration rather than settling by gravity and this system does not consider the flocs growth but still maintains the minimum sludge production with low F/M ratio (less substrate is presented per unit of biomass) [7,37] and retaining the biomass in the reactor and sludge age [4,39,58,59]. Besides that, the formation of flocs makes it easier to filter. However, if F/M is too low, the biomass in the activated sludge could not grow well [4], or else if MBR has very high MLSS it will lead to clogging, low efficiency of aeration and will need a large bioreactor (increase the initial capital cost) [7]. Low HRT will increase organic loading rate (OLR) with reactor volume reduction and reduced performance of MBR whereas for a high HRT, MBR has a good performance [23,60,61]. Diverse pollutants and complex components leading to slow biodegradation have led to the use of MBR in treating industrial wastewater. HRT is closely related with quality of effluent water and does not influence nutrient removal in treating high readily biodegradable pollutants. However, in industry, wastewater contains plenty slowly biodegradable pollutants and small variation on HRT affects the nutrients removal efficiency to achieve high quality effluent [23]. Dominguez et al. reported that biomass growth was closely related to OLR and in this study, the steady state OLR for MF MBR and UF MBR was 0.15 kg COD kg MLVSS⁻¹ d⁻¹, where the biomass growth rate was 5–8 times faster at higher OLR [62].

Other studies showed the correlation between SRT and formation of SMP and EPS. Increased SRT will decrease SMP and EPS whereby the biomass will stay longer in the reactor and prolong the biological degradation process whereas for lower SRT, it will increase the level of SMP and fouling [60,63,64]. Other studies showed reverse results [64]. High SRT also create starvation condition (low F/M ratio) which can reduce SMP formation, good for nitrification and less sludge production [39]. Nevertheless, if SRT

takes too long, it tends to foul the membrane with the accumulation of matters and high sludge viscosity [60]. In anaerobic membrane bioreactor, HRT and SRT are independent and also produce methane as a by-product and odour [39] whereas they do not use any aeration process and energy saving. Besides that, methane can be collected for energy generation [65]. There are several advantages of applying high SRT in MBR which include (i) slow biomass growth responsible for the biodegradable of organic and inorganic pollutants; (ii) higher MLSS can be operated in MBR that prepare starvation condition to achieve good quality of effluent; and (iii) high SRT create low F/M ratio that reduces SMP production and lead to lower membrane fouling [39,66].

4. MBR application in industrial wastewater treatment

During MBR operation, there are different operating conditions depending on the level of constituents of high strength wastewater. The operating conditions cover the sludge behaviours (e.g. MLSS, DO, SRT and HRT) and membrane behaviours (e.g. membrane configuration and pore size). Table 3 shows the operational parameters according to the types of industries. These two types of industries are chosen because of the difference in biodegradability ratio. Table 1 shows the categorising of both industries' effluent as high strength wastewaters but they are different in terms of biodegradability. Textile industries have low biodegradability compared to food industries due to the slow biodegradable organic or toxic matters present [25]. Food industries are known as high strength organic wastewater and the level of biodegradability is high due to the high content of readily biodegradable or organic matters [67].

4.1. Textile industries

In textile industries, the primary source of wastewater is spent dye and rinsing water that contain low biodegradability, toxicity and colour issues. Most textile industries apply biological treatment, chemical precipitation, adsorption and membrane technology [31].

Hai et al. applied hollow fibre (HF) and flat sheet (FS) membranes at the same condition with 50–200 μm pore of membrane for treating high strength wastewater from synthetic textile industries consisting of various chemicals, organic loading and colour. In this study, white-rot fungi *C. versicolor*, NBRC 9791 was used to remove specifically colour and other nutrients. The reactor had a volume of 12.5 L and each membrane had a surface area of 0.2 m² and 0.4 μm (Mitsubishi Rayon). The two membranes performances

Table 3
MBR operational parameters for industrial wastewater.

	Textile			Food		
	Textile [28]	Textile [29]	Textile [31]	Pet food [32]	Palm oil [16]	Dairy product [33]
Reactor volume (L)	500	20	230	20	20	20
Reactor type	Aerobic, side-stream	Aerobic, side-stream	Aerobic, submerged	Aerobic, submerged	Aerobic, submerged	Aerobic, submerged
Membrane configuration	UF, (7 tubular modules), PVDF	UF, external tubular cross-flow, PVDF	HF	2 modules	FS, 1 module	MF, 34 strands of a HF
Membrane surface area (m ²)	–	0.28	–	0.047	0.1	0.00162
Pore size (µm)	0.025	–	0.04	0.04	0.4	0.4
Flux (L/m ² h)	–	30	20	–	10	Horizontal: 5.03 Vertical: 2.27
MLSS (mg/L)	5000–15,000	–	13,900±	–	5000±	4000–10,000
MLVSS (mg/L)	–	–	–	47,000±	–	–
DO (mg/L)	1–3±	2–3±	–	3±	8±	–
HRT (day)	2	0.7–4	0.58	2.9±	0.8±	–
SRT (day)	–	11	25	50	–	–
COD removal (%)	97±	90±	97	97±	94±	–
Colour removal (%)	70±	98±	98	–	–	–
TSS removal (%)	–	–	99	–	–	99

were compared under different operating fluxes (0.05–0.3 m/d) while maintaining other operating conditions (e. g. 8000 mg MLSS/L and 0.01 m/s of aeration) of the colour and the TOC removal was 97–99% respectively. HF membrane shows a slightly good performance when compared with FS membrane [8].

Badani et al. showed the best performance by MLSS was at 15,000 mg/L with average colour removal of 70%. The TMP was increased and the flux decreased due to membrane fouling. The HRT of 2 days was sufficient to degrade COD up to 97% of removal in 15,000 mg/L of MLSS for a long term performance [28]. Brik et al. stated that at the beginning of the process, the sludge 5000 mg/L of MLSS was treated with municipal wastewater before being acclimatised with textile wastewater. The MLSS increased from 5000 to 10,000 mg/L and continued to increase up to 15,000 mg/L before sludge withdrawal. In this study, it was also observed that the effect of nutrient addition did not improve COD removal. However, adding the nutrient contributes to conductivity because of the inorganic content in the additive nutrient and resulted in severe fouling [29]. A study by Yigit et al. investigated highly concentrated mixed textile wastewater and found that the influent BOD₅/COD ratio was 0.32 which was dominated by slowly biodegradable and biorecalcitrant organics. The result also showed a successful performance of SMBR and it could be concluded that colonies of biomass with a wide spectrum of degradation capability were able to degrade as readily biodegradable, slow biodegradable and biorecalcitrant. The mechanisms of colour removal was basically by using biodegradation and adsorption onto solid [31].

4.2. Food industries

Majority of food processing facilities are characterised as very high organic strength wastewater generators. Major pollutant loadings are BOD, COD, TSS, fat, oil, grease and nutrients. Most food industries employ on-site primary treatment prior to sending their wastewater to municipal wastewater treatment plants [67].

According to Acharya et al. [32], the wastewater had biodegradability below 0.5 and removal of organic high strength pet food wastewater by crossing the membrane for the first stage was 7–37% and is 20–37% for the second stage. This was due to the retention of particles larger than the membrane pore size and it is supported by Huang et al. (2001) as (10–20%) of COD removal across the membrane [68]. In addition, this study showed the mechanism of nitrogen removal which is divided into two stages for MBR treatment where nitrification–denitrification (SND) mod-

ules develop simultaneously. The model indicated that 21% of influent nitrogen is removed by SND, 31% by cell synthesis (N_{cell}) and 13% by stripped ($N_{stripped}$) at the first stage of MBR treatment.

Katayon et al. studied different configurations of membrane (horizontal and vertical) at different concentrations of MLSS and their effect on the performance of MBR. Though the COD and BOD values were low, the amount of TSS was too high. The result showed slow horizontal flux decline when compared with vertical flux and by increasing the MLSS concentration, the permeate flux was decreased. TSS and turbidity removal had good performance results at low MLSS concentration. This study also showed pH performance in low and high MLSS where both results show influent with pH of 3.8 which increased to 7–9. At low MLSS, turbidity and suspended solid removed were higher than high MLSS [33]. Palm oil is categorised as agro-industry but it is also described as a food industry. Yejian et al. reported that the treatment of palm oil mill by using anaerobic expanded granular sludge bed (EGSB) and aerobic biofilm reactor (ABR) integrated with UF and reverse osmosis (RO) gave successful result. For the first EGSB, COD removal was 93% and 43% of the organic matter was converted into biogas as recovered energy. However, in ABR, the COD removal was only 27% from the effluent of EGSB and UF removed 10% of COD from ABR. In RO, there were no COD detected. All suspended solids were captured by UF and the RO filtered dissolved solids and inorganic salts. It can be concluded that EGSB and ABR were purposely used to reduce TSS and oil to lower concentrations in order to prolong the lives of UF and RO membranes. However, EGSB operation has problem where scum was created and blocked the gas outlet, clogging the line and leading to high pressure in the reactor [17].

5. MBR: limitation and mitigation

Judd stated that cost is a major constraint to MBR technology in the 1990s because of high cost of membrane which leads to the increase of maintenance and operational costs. Membrane cost covers replacing of severe membrane fouling or corrupted membrane and membrane cleaning processes during maintenance [3,39].

5.1. Membrane fouling

Fouling is a major factor that needs consideration when it comes to membrane. When dealing with high strength wastewater containing high load of contaminants, it will lead to high clogging

of the membrane due to the membrane characteristics, biomass and operating conditions. Factors that influence membrane fouling during MBR operation covers membrane (membrane configuration, material, hydrophobicity, porosity, pore size), biomass (MLSS, EPS, SMP, floc structure and size, dissolved matter) and operating condition (MBR configuration, cross-flow velocity, aeration, HRT, SRT, TMP) [68]. Fouling can be monitored through TMP and flux changes. Originally, flux-step method is the correlation between TMP and flux at a time interval of 15 min [39] but the time interval can vary. When flux increases, TMP also increases and hence, more wastewater can be separated until the TMP levels off when the flux continues to increase. Decreasing phase shows that membranes have a high resistance and need cleaning before they become fouled which can lead to membrane damage.

Fouling is the physicochemical interaction between the biofluid and membrane to form a cake layer and the adsorption of the dissolved particles into membrane pores, leading to flux decline. If a physical cleaning takes place, it is classified as reversible fouling. Irreversible fouling is due to the adsorption of the particles into the membrane and blocking the pore [68]. Fig. 7 shows the mechanisms of fouling dependence on particle size to the pore diameter [39]. The formation of cake which is inevitable on the membrane surface becomes one of the factors that lead to membrane fouling. In a general system, side streams of MBR have higher fouling tendency than submerged MBR. This is because the side stream of MBR needs high energy of pumping that produces high flux that will lead to repeating the fouling compared to submerged MBR [39].

Organic fouling in MBR is caused by deposition of small size of biopolymers such as proteins and polysaccharides on the surface of the membrane. These depositions are more difficult to remove than large particles like sludge floc. Deposition of inorganic elements (Ca, Mg, Al, Si, etc.) detected on the surface of the membrane may lead to severe inorganic fouling. Wang et al. shows the severe inorganic fouling that happened at high alkalinity of activated sludge [69]. There is less research information on inorganic fouling especially in treating high strength industrial wastewater since they are more concerned with fouling caused by biomass and biopolymer.

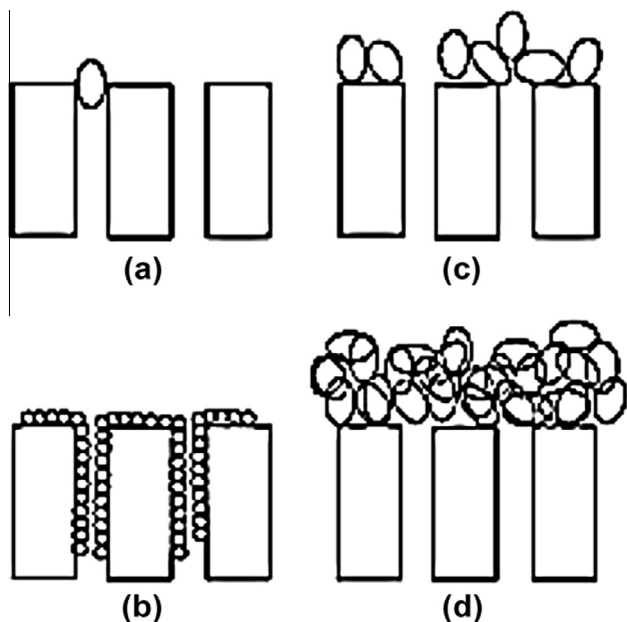


Fig. 7. Fouling mechanism (adapted from [39]) (a) complete blocking, (b) standard blocking, (c) intermediate blocking, and (d) cake filtration.

Both EPS and SMP which are bound and in soluble form can lead to membrane fouling. EPS is located outside the cell surface and SMP is the organic compound that is released from substrate metabolism (substrate utilisation-associated products, UAP) or biomass decay (biomass-associated products, BAP). Both consist of protein, polysaccharides, nucleic acids, lipid, humic acids, etc. The correlation between EPS and SMP with membrane is critically difficult. From Laspidou and Rittmann, unified theory has found that EPS and SMP overlapped each other or else cells use electrons from the electron-donor substrate to build active biomass, and produce bound EPS and UAP at the same time and in proportion to substrate utilisation. Bound EPS are hydrolysed to BAP, while active biomass undergoes endogenous decay to form residual dead cells. Finally, UAP and BAP, being biodegradable, are utilised by active biomass as recycled electron-donors substrates [70]. In addition, some SMP can be adsorbed by biomass floc to become bound EPS [71]. Wang et al. reported that two different sludge characteristics gave different fouling status. Excess growth filaments controlled by low DO concentration gave better filtration when compared with normal sludge (high DO concentration) due to large particle sludge distribution, lower hydrophobic contents in SMP and special fouling layer formed by filamentous bacteria [72].

EPS is closely related with specific cake resistance, where an increase in EPS lead to increase in specific cake resistance [73]. Filamentous bacteria create bulking problem and at the same time lead to producing high EPS concentration rather than normal bacteria while SMP tend to accumulate in MBR or deposit into membrane pores [71].

Eq. (5) shows the relationship between TMP and flux from the fundamental of Darcy's Law. This became a benchmark for measuring the resistance of membrane, driving force for each unit membrane area and fouling and the time for cleaning the membrane [39]. Eq. (6) is a resistance in membrane series model (RIS) with simple model to describe membrane fouling mechanisms [74].

$$J = \text{TMP}/(\eta \cdot R_t) \quad (5)$$

$$R_t = R_m + R_e + R_i = R_m + (R_p + R_c) + R_i \quad (6)$$

where R_m is the constant resistance of the clean membrane, R_e is the internal fouling resistance which includes R_p (resistance due to concentration polarisation, which can be neglected) and R_c (the cake layer resistance) and R_i is the resistance due to pore blocking.

Choo et al. reported that sMBR with FS membrane had 0.5% R_m , 82.8% R_p , 16.1% R_{ef} (external fouling resistance) and 0.5% R_{if} (internal fouling resistance) in treating alcohol distillery wastewater for 200 days operation. R_p gave a large contribution to fouling due to colloidal solutes and macro molecular species accumulated at membrane [9]. According to Mutamim et al., a high MLSS gave high total resistance, R_t , due to high sludge resistance, R_s [27]. Hai et al. in their study of textile industry stated that the mechanism of fouling occurs because of the formation of layer of cake fungi and sticky starch. FS membrane was vulnerable to internal pore blocking but HF happened due to the presence of cake layer in the latter case [8]. Badani et al. stated that membrane fouling depends on the extent of shear stress imposed on microbial flocs [75]. Manser et al. reported that floc size from the MBR was ten times smaller than CAS. Since there was no selection for settleable flocs in MBR system, the biomass had no physical inducement to build large flocs that could lead to membrane fouling [76].

Chang et al. showed the results from SEM to EDX spectra analysis towards the membrane before and after fouling, where cake layer deposited on the membrane surface when compared with the inner surface by biomass physiological properties (EPS and SMP). Inorganic elements (Mg, Ca, Cu, Rb, Pt and Al) were detected

at the inner and outer surfaces of the membrane [34]. Amiri et al. stated that by increasing heavy metal concentration, the permeability is reduced due to high formation of EPS [77]. Vireo et al. noted that SMP content might be considered an indicator of the fouling level since increase in SMP concentrations in MBR tend to reduce the permeability of the membrane [78]. Dominguez et al. stated that high membrane fouling occurs during low sludge age or low MLSS concentration due to high solubility of EPS [62].

5.2. Fouling mitigation

Operational parameters become a part of limitation in MBR. Therefore, the operational parameter needs to be wisely controlled to minimise its fouling effect. SRT is an important operating parameter that influences MBR performance especially in the control of fouling problem. A long SRT normally improves filtration performance and reduces EPS and SMP production by creating starvation conditions [39]. A too long SRT leads to severe fouling due to high MLSS accumulation or old sludge (filamentous) production. Similarly if the SRT is too short, there will be reduced performance of MBR due to low biomass [79]. High F/M ratio can also increase EPS concentration because of high food utilisation by biomass [79].

Besides operational control, the membrane cleaning need to be done when the flux is slightly dropped (filterability reduction) and TMP increases drastically. There are three types of membrane cleaning – physical, chemical and combination of physical and chemical. Physical cleaning includes backwashing (suitable only for HF), and where the effluent is pumped in the reverse direction but it is not suitable for FS membrane. Membrane brushing is also a method of physical cleaning that could be applied *in situ* for FS membrane. It is a quick process but is less effective than chemical cleaning. Relaxation is the intermittent cessation of permeation for flux recovery if the membrane is submerged and scoured with air when permeation stopped. The combination of relax/permeate and backflush/permeate can reduce chemical cleaning and prolong membrane life [80]. Basically, physical cleaning only removes the coarse solid or cake on the surface of the membrane, while chemical cleaning removes the flocs. It can also remove strong matters that stick on the membrane's surface. It needs to be taken into consideration that the energy consumption for physical cleaning and almost 30% of permeate (effluent) is used for back washing. Blocher et al. stated that the purpose of chemical cleaning, besides fouling elimination, is also for membrane disinfection [81]. For industrial purpose, *in situ* cleaning is usually performed if the fouling is not severe otherwise *ex-situ* cleaning takes place. Most of the studies showed that the first chemical used for membrane cleaning was sodium hypochlorite (NaOCl) [8,31,81,82]. Broeck et al. applied relax/filtrate cycle at influent to municipal wastewater to prolong the membrane life and reduce the cycle of chemical cleaning [66]. Eq. (7) as shown below:

$$Q_{in} = \frac{J_{gross} * A_{mem} * t_{fil}}{t_{fil} + t_{rel}} \quad (7)$$

where Q_{in} is the influent flow rate, A_{mem} the membrane area, t_{fil} the time of filtration and t_{rel} is the time of relaxation.

Hai et al. noted that when the flux per unit pressure dropped, the cleaning process was recovered by *in situ* membrane brushing because air bubbles from diffuser could not fully scrub the fungi off from the membrane. The worst thing was that the air bubble diffuser pushed the fungi towards the membrane. The *ex situ* cleaning and sludge withdrawal was carried out when the flux per unit pressure was almost zero. Table 4 shows the value of TMP after the membrane was cleaned by water and chemical [8]. Katayon et al. reported that with diffuser at the bottom of the reactor, the membrane configuration with horizontally placement minimised the

Table 4
TMP value (kPa) after water and chemical cleaning (flux = 0.3 m/d) [8].

	Hollow fibre	Flat-sheet
Initial	6	7
Fouling	65	86
Cleaning: water	10	86
Chemical (NaOCl)	6	7

membrane fouling when compared with the vertical one [33]. Yigit et al. reported that during operation, backwashing routine was taken as 15 s per 10 min of permeate production. The first chemical backpulse when the TMP was 60 kPa was by sodium hypochlorite. However the *ex situ* cleaning (sodium hypochlorite with hydrochloric acid) was applied when irreversible fouling took place [31]. Membrane configuration also plays an important part in reducing fouling. Katayon et al. reported that horizontal membrane configuration produced slow permeate with declining flux when compared with the vertical one [33]. Generally, chemical cleaning is applied at every 7–14 days and the maximum allowable rate of pressure change is 0.6 bar/week (0.06 kPa/week) [80].

Regular cleaning of the membrane shortens its life and membrane change is needed when the membrane can no longer be used. Therefore activated carbon (AC) is applied in MBR as a biofouling reducer (BFR) to prolong membrane life. AC has the ability to adsorb organic and other pollutants besides becoming a scrubber for membrane. Small sized and small pores of AC will have more surface area that increases the adsorption velocity. Between the granular and powder AC, the powdered version has a higher adsorption capacity [16] and is able to remove low molecular weight organic rather than granules because it has a higher surface area. Powdered activated carbon (PAC) can also be a medium for bacterial sticking and growth. As a result, biological activity would increase by sustaining the PAC in a reactor. Widjaja et al. used a set-back flushing method for 10 min to increase the performance of MBR in treating shock loading of a toxic compound [4]. Biological degradation with added PAC gives better results compared with non-PAC because of their characteristic to adsorb organic matters. Besides that, PAC can treat COD in shock loading by increasing its quantity when it will stabilize the shock loading performance [4]. Yuniarto et al. showed that by adding PAC, the performance of MBR increased up to 3.8% in removing high strength palm oil wastewater [16]. Damayanti et al. showed the performance of three types of BFRs in removing SMP in palm oil mill effluent (POME) treatment. PAC gave good performance followed by Mo and Ze. PAC existed as cationic polymers with a surface area of 3000–4000 m²/g, compared to Ze and Mo whose surface areas ranged from 600 to 800 m²/g and 713 to 744 m²/g, respectively. PAC enhanced the flux three times lower than no-BFR and successfully formed flocs by charging the neutralization mechanism from organic and inorganic components and enlarging the floc to build up porosity in the cake layer [15]. Adsorbents and coagulants also have the ability to reduce the SMP where the SMP tends to be entrapped in biofloc [79].

Lee et al. reported that the membrane fouling reducer, MFR, flocculated the activated sludge to reduce the cake layer on the surface of the membrane. The result showed that in order to achieve 30 kPa of TPM, a small amount of MFR was needed effectively in the removal of high contaminants in wastewater. MFR from cationic polymer acted as a positive charge and when it was added into the reactor, it adsorbed the negative charges from the microbial flocs and changed into a neutral charge. The neutralized sludge floc then attracted each other to form large flocs by a charged neutralization mechanism which is also called flocculation process. On the other hand, in high concentration of MFR over the

optimum concentration, the surface turned to positive charge and deflocculation began by a mechanism of electrostatic repulsion [83]. More study on flocculation, coagulation and adsorption with respect to fouling mitigation is required especially in the industrial sector to reduce the cost of membrane maintenance.

6. Conclusion

Most high strength wastewaters are successfully treated by MBR and can be shown in textile and food industries which have different types of strength. High strength industrial wastewater is difficult to be identified and biodegradable (BOD₅/COD) ratio is a method to identify the 'hardness' of wastewater. By controlling parameters such as SRT, HRT, TMP, Flux and MLSS to an optimum condition, the best performance of MBR can be produced. Besides, these controls of operation parameters have proved to be effective in reducing fouling problems. SMP and EPS are recent parameters that affect permeability performance and these parameters need more investigation in the industrial sectors. For wastewater with very high loading, it has to be treated before entering MBR to avoid membrane fouling. Physiological characteristics (MLSS concentration, EPS, SMP, organic and inorganic matters) can change according to the MBR operation condition and the changes make it difficult to control and predict the membrane fouling. However, there are some methods to reduce the fouling problems and enhance the performance of MBR to produce high quality effluents. Membrane is the heart of this system but it is also very sensitive to unusual chemicals besides pH, pressure and temperature that contribute to reduce the performance of MBR. These problems need to be of concern since the quality of wastewater treatment depends on the processes and the environment involved. Sometimes, modifications of the wastewater such as dilution for high strength and toxic wastewater or neutralization of acidic or base of wastewater need to be done to prolong the lifespan of the membrane besides maintaining the microbial growth.

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