A comprehensive study on membrane fouling by different sludge fractions of a membrane bioreactor with long solid retention time

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**A COMPREHENSIVE STUDY ON MEMBRANE FOULING
BY DIFFERENT SLUDGE FRACTIONS OF A MEMBRANE
BIOREACTOR WITH LONG SOLID RETENTION TIME**

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LIST OF PUBLICATIONS

Shushu Liu, Xiwang Zhang, Darren Delai Sun and Zhenming Xu, Study on membrane fouling caused by activated sludge from a membrane bioreactor with long solid retention time, Chinese Journal of Environmental Engineering, 2008, accepted

Shushu Liu, Xiwang Zhang and Darren Delai Sun, Study on membrane fouling by various sludge fractions in membrane bioreactor with long sludge retention time, under preparation

SUMMARY

Membrane fouling and the scarce knowledge on microorganisms' behaviour limit the industrial application of MBRs with long SRT. Moreover, the mixed liquor composition varies with inflow quality and operating conditions, the contribution of each sludge fraction to membrane fouling is changed along with different inflow quality and operating conditions. Therefore, it is necessary to investigate the influences of MLSS concentration, trans-membrane pressure and cross-flow velocity on membrane fouling to have a better understanding of the fouling behaviours in MBRs with long SRT conditions.

A 300-day SRT MBR and 10-day SRT SBR were set up for the study. Activated sludge taken from these systems was fractioned into suspended solids, colloids and solutes; and their relative contributions to membrane fouling during crossflow-filtration were calculated. Furthermore, sub-divided filtration resistances were analysed using resistance-in-series model.

Results from this study indicated that the suspended solid was the main fouling fraction, with the relative contribution to the total resistance of 70% and 85% in MBR and SBR respectively. On the other hand, the importance of colloids and solutes could not be neglected. Moreover, sub-divided resistances were analysed to have a deep understanding about the fouling mechanism. It was demonstrated that the resistances of concentration polarization (R_{cp}) and cake layer (R_c) mainly contributed to the total resistance, while the internal fouling resistance (R_{if}) and membrane resistance (R_m) were negligible during the filtration of activated sludge.

Furthermore, the membrane fouling increases with the increasing MLSS concentration, rising trans-membrane pressure (TMP) and reducing cross-flow velocity (CFV) respectively. Suspended solids remained the main contributor to membrane fouling while the dominant filtration resistance varied according to different MLSS concentration, TMP and CFV. Further studies can be focused on the characteristics of biofoulants and effects of other parameters on membrane fouling caused by various sludge fractions.

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LIST OF SYMBOLS

C_b	bulk concentration (mg/L)
C_w	the solute concentration adjacent to the membrane (mg/L)
CAS	conventional activated sludge
CFV	cross-flow velocity (m/s)
COD	chemical oxygen demand (mg/L)
D_B	Brownian diffusion coefficient (m^2/s)
d_p	particle radius (m)
ε	cake porosity
F/M	food to microorganisms ratio (kg COD / kg MLVSS.d)
HRT	hydraulic retention time (h)
J	membrane flux (m^3/m^2s or L/m^2h)
J_{AS}	flux of the activated sludge
J_{sup}	flux of the supernatant
J_{sol}	flux of the solutes
J_0	initial membrane flux (m^3/m^2s or L/m^2h)
J_v	membrane flux during the filtration process (m^3/m^2s or L/m^2h)
J_{w1}	the deionized water flux(m^3/m^2s or L/m^2h)
J_{w2}	the deionized water flux after removing the concentration polarization layer (m^3/m^2s or L/m^2h)
J_{w3}	the deionized water flux after removing the cake layer (m^3/m^2s or L/m^2h)
MBR	membrane bioreactor
MF	microfiltration
MLSS	mixed liquor suspended solids (mg/L)
MLVSS	mixed liquor volatile suspended solids (mg/L)
ρ	particle density (kg/m^3)
ρ_o	fluid density (kg/m^3)
Δp /TMP	transmembrane pressure (N/m^2 or kPa)
R	filtration resistance (1/m)
R_{AS}	resistance of activated sludge (1/m)
R_{ss}	resistance of suspended solids (1/m)

R_{col}	resistance of colloids (1/m)
R_{sol}	resistance of solutes (1/m)
R_m	resistance of the membrane (1/m)
R_t	total filtration resistance (1/m)
R_c	cake resistance (1/m)
R_{cp}	resistance of concentration polarization (1/m)
R_{if}	internal fouling resistance (1/m)
δ_m	membrane thickness (m)
SBR	sequencing batch reactor
SEM	scanning electron microscope
SMP	soluble microbial product
SRT	solids retention time (d)
TOC	total organic carbon (mg/L)
μ	fluid viscosity (Ns/m ²)
UF	ultrafiltration
V	reactor volume (L)
VLR	volumetric loading rate (kg COD/m ³ d)

CHAPTER 1 INTRODUCTION

1.1 Background

Although more than 70% area of the earth is covered with water, only an estimated 3% of the total amount of water on Earth is fresh water. Of this fresh water, over two-thirds are frozen in the polar icecaps and glaciers, which means that the amount of readily available fresh water covers less than 1% of the earth's areas. Therefore, it is necessary to find sustainable solutions to rectify the current situation of inadequate fresh water for the ever-increasing global population.

Conventional activated sludge (CAS) process is the most commonly used method for wastewater treatment. It usually consists of three parts: aeration tank, where air (or oxygen) is injected into the mixed liquor; settling tank, which allows the biological flocs to settle, thus separating the biological sludge from the clear treated water; and, a reclamation system to return part of the activated sludge from the settling tank to the aeration tank. CAS has been widely used for the treatment of sewage and industrial wastewater because of its competitive operating costs as compared with other processes, such as chemical treatment processes. However, the large amount of activated sludge produced by CAS, amounts to significant costs for sludge treatment and disposal, and takes up about 60% of the total plant operating costs (Defrance and Jaffrin, 1999). In addition, the CAS treatment is unable to provide a stable and high quality treated wastewater because the stability of the CAS process depends mainly on the gravitational settling of mixed aggregated mixed microbial flocs.

The membrane bioreactor (MBR) process was first introduced to wastewater treatment in the late 1960s, as soon as commercial scale ultrafiltration (UF) and microfiltration (MF) membranes were available (Judd, 2006). It combines the biological degradation process by activated sludge with a direct solid-liquid separation by membrane filtration. By using UF or MF membranes, MBR systems allow complete physical retention of bacterial flocs and virtually all suspended solids within the bioreactor, henceforth resulting in high biomass concentration up

to 30g/L (Yamamoto, et al., 1989; Le-Clech, et al., 2006). A higher biomass concentration would lead to a more stable reactor performance even at fluctuating organic loading conditions. Besides, the MBR process has other advantages over conventional processes, including small footprint, easy retrofit and upgrade of old wastewater treatment plants (Judd, 2006). As a result, the MBR process has now become an attractive option for the treatment and reuse of industrial and municipal wastewaters. Recently, MBR was operated with a long sludge retention time (SRT) ranging from 25 to 300 days to maintain high biomass concentrations, reduce solids production and minimize reactor volume. Furthermore, long SRTs lead to endogenous respiration within the biomass and the growth of specialised bacteria, e.g. nitrifiers, and enhanced breakdown of large macromolecules. Some studies on long SRT MBR with no sludge discharge or little sludge discharge have been reported (Muller, et al., 1995; Rosenberger, 2002; Liu, et al., 2005; Pollice, et al., 2006; Pollice, et al., 2007).

However, membrane fouling, which is the result of deposition of organic and inorganic materials onto and into membrane, represents the main obstacle to the practical application of MBRs. It results in reduced performance, severe flux decline, high energy consumption and frequent membrane cleaning or replacement, which directly lead to increases in maintenance and operating costs. In the early period, the mixed liquor suspended solids were pumped at high cross-flow velocity at significant energy penalty (of the order 10 kWh/m³ product) to reduce fouling. For the current generation of MBRs, the energy input is typically ≤ 1 kWh/m³ product to ensure the economic viability.

Another limitation of the industrial application of MBRs with long SRTs is the limited knowledge on microorganisms' behaviour, such as consistency of biological degradation, effect of soluble microbial products (SMP) and their microbial activity (Ross, et al., 1998; Han, et al., 2005; Liu, et al., 2005), and system stability in a long SRT condition. More dead or inactive microorganisms were found in MBR as compared with CAS and the specific activities of nitrification, denitrification and organic removal were lower than those in the CAS process (Zhang and Yamamoto, 1996; Han, et al., 2005). Those studies revealed that long SRT operation of MBR

results in changing microorganisms composition of the mixed liquor and that may reduce its biological capability. Up to date, information on the microbial viability in long SRT condition is still insufficient.

Therefore, further study on the effects of different operating conditions on membrane fouling in a long SRT (300-day) MBR is necessary to achieve a better understanding on the biological performance of the membrane bioreactor.

1.2 Objectives

This study aimed (1) to investigate the contributions of different fractions of the activated sludge from a long SRT (300-day) MBR, namely, suspended solids (SS), colloids and solutes, in membrane fouling, and (2) to evaluate the effects of MLSS concentrations and operating conditions, i.e., trans-membrane pressure (TMP) and cross-flow velocity (CFV) on membrane fouling in cross-flow filtration of activated sludge taken from a 300-day SRT MBR and a 10-day SRT SBR (sequencing batch reactor) respectively.

1.3 Scopes of work

In this study, the fouling contributions of different fractions from two types of activated sludge were estimated. One was circulated in a 300-day SRT membrane bioreactor and the other was the conventional activated sludge, which had been cultured in an SBR for more than one month with SRT of 10 days. These activated sludge were fed with the same synthesized influent and their concentrations were adjusted to an equal level by dilution techniques, to avoid any concentration effect in the membrane fouling.

Then, the various fouling contributions of different sludge fractions from the activated sludge with SRT of 300 days and 10 days were tested, according to different mixed liquor suspended solid (MLSS) concentrations, TMPs and CFVs. In each experiment, the degree of membrane fouling was quantitatively calculated using the resistance-in-series model; and resistances of different fractions were also calculated. Furthermore, observations of fouled membranes were also conducted using the field emission scanning electron microscope (FESEM).

1.4 Structure of thesis

This thesis is divided into 6 Chapters. Chapter 1 gives background information of MBRs used in wastewater treatment and their advantages compared to the CAS process, including the specific objectives, scopes and structure of this study. In Chapter 2, reviews of the relative literature are presented and composed in three parts: MBRs, membrane mass transport mechanisms and the different parameters affecting membrane fouling. Chapter 3 is about the experimental plans, including the methodology, design, experimental procedure and analysis methods that were used in this study. Results about fouling behaviors of activated sludge, taken from 300-day SRT MBR and 10-day SRT SBR, are stated in Chapter 4. The performances of MBR and SBR, characteristics of activated sludge, contributions of different sludge components to membrane fouling are discussed. Chapter 5 is about effects of MLSS concentration, TMP and CFV on membrane fouling by various sludge fractions. The flux decline behaviors, contributions of sludge fractions to membrane fouling and fouling resistances are analyzed. Chapter 6 presents the conclusions based upon the results from this research study. The recommendations for future works are also proposed in this chapter.

CHAPTER 2 LITERATURE REVIEW

2.1 Membrane bioreactor

The MBR technology combines the biological degradation process by activated sludge with a direct solid-liquid separation by membrane filtration, ensuring effective removal of organic and inorganic contaminants and biological material from municipal and/or industrial wastewaters. Compared with the CAS process, advantages of MBR techniques include smaller footprint, higher loading rate capabilities, and modularity (Stephenson, et al., 2000). In addition, these systems are not affected by the settling properties of activated sludge as in conventional process, allowing operation at very high biomass concentrations and at prolonged SRT. However, the cost of membranes and loss of membrane filtration capacity due to membrane fouling limit the industrial application of MBRs in wastewater treatment.

2.1.1 MBR configurations

There are two main MBR process configurations: side-stream and submerged (Fig. 2.1 (Judd, 2006)). In the side-stream MBR unit, high pressures and volumetric flows are imposed to transfer the mixed liquor out from the bioreactor towards the membrane module at high cross-flow velocity and return the retained biomass to the bioreactor (Mallevalle, et al., 1996). While, in the submerged MBR unit, membrane module is submerged into the bioreactor and only the filtrate is withdrawn from the bioreactor by a pump (Yamamoto, et al., 1989; Wisniewski, 2007). Hence, submerged MBRs are generally less energy-consuming than side-stream MBRs. As a result, submerged configuration is preferred in majority of the studies dealing with domestic wastewater treatment. However, similar fouling behaviour was obtained when the submerged and side-stream MBRs were operated at superficial gas velocity (U_G) of 0.07–0.11 m/s and superficial liquid velocity (U_L) of 0.25–0.55 m/s respectively (Le-Chen, et al., 2005).

Figure 2.1 Configurations of a membrane bioreactor: (a) side-stream and (b) submerged

In submerged MBR processes, the membrane can be configured as flat plates, hollow fine fibres (filtration from out-to-in) or, more rarely as tubes (filtration from in-to-out) as shown in Fig. 2.2 (Judd, 2006).

Figure 2.1 Flow through membrane configured as: (a) flat plate (b) tube (c) hollow fibre

The effect of membrane configuration was also assessed, using hollow fibre and flat plate MBRs (featuring similar pore size of 0.4µm) for high-strength wastewater treatment (Yamamoto, et al., 2005). It was found that the flat sheet MBR fouled slightly more and could not recover its original performance after water cleaning when the two systems were operated at similar flux

2.1.2 Long SRT MBRs

In long SRT MBRs, the limited substrate provided for the maintenance of bacterial vital functions leads to extremely low biomass growth rates and the consequent low biomass production, thus reducing the operational costs in sludge management and disposal (Suwa, et al., 1992). If sludge is completely retained, that is, SRT tends to be infinite, no sludge would be produced, leading to less energy consumption and lower maintenance cost. Therefore, long SRT MBRs are preferred by many researchers in order to maintain high biomass concentrations, reduce biomass production and minimize reactor volume.

Muller et al. (1995) found that when sludge is completely retained, aerobic treatment of domestic wastewater at high loading rates can be applied because of its stable treatment performance and a very low sludge production. The steady and better performances of long SRT MBRs compared with CAS processes were confirmed by many other studies (Rosenberger 2002; Liu, et al., 2005). Recently, similar dewatering properties of the sludge in a complete retention MBR with respect to conventional waste activated sludge were also observed by Pollice et al. (2007).

However, the accumulation of MLSS, partially because of the production of EPS (Extracellular polymeric substances), may lead to high fouling potential. Moreover, the properties of mixed liquor, such as viscosity, concentration and composition of biomass were changed at longer SRT (Chang, et al., 1999). Therefore, detailed studies are needed to achieve a better understanding of fouling behaviours in long SRT MBRs.

2.2 Membrane mass transport mechanisms

Applying different drive forces on membranes, such as pressure, concentration gradient and temperature allow different components to pass through the membrane at different rates depending on physical and/or chemical properties of the components and membranes. The permeate flux through porous membrane can be described by Darcy's law (Ho and Sirkar, 2001):

$$J = \frac{1}{A} \frac{dV}{dt} = \frac{\Delta p}{\mu R_t}$$

where J is the flux ($\text{m}^3/\text{m}^2\text{s}$), Δp is the trans-membrane pressure (N/m^2), μ is the viscosity of permeate ($\text{N}\cdot\text{s}/\text{m}^2$) and R_t is the total resistance (m^{-1}).

2.2.1 Resistance-in-series model

Various models were developed to express mass transport mechanism in membrane processes. The simplest model to describe membrane filtration behaviour is resistance-in-series model (Lee, et al., 2001)

$$J = \frac{\Delta p}{\mu(R_m + R_c + R_f)}$$

where R_m is the intrinsic membrane resistance; R_c is the (reversible) cake resistance caused by the cake layer deposited over the membrane surface; R_f is the (irreversible) fouling resistance produced by pore plugging and/or adsorption of dissolved matter onto the membrane pore wall or surface (Bae and Tak, 2005).

By further investigation of the sub-divided resistances, many various models have been developed based on this simple model of membrane fouling.

2.2.2 Concentration polarization model

Beside the three parts of resistance that we have mentioned in the resistance-in-series model, there are other parameters also contribute to membrane fouling. The jellification or crystallization of solution, which results in a gel layer on the membrane surface, and the concentration of rejected solute near the membrane surface, cannot be ignored. Both of them are promoted by concentration polarization.

Concentration polarization describes the accumulation of the solute at membrane:solution interface during cross-flow operation, within a concentration boundary layer or liquid film (Fig. 2.3 (Judd, 2006)). The extent to which this occurs depends on the propensity of the rejected species to diffuse, the notional thickness of the stagnant region, and the rate. The latter two factors are determined

mainly by the operating conditions themselves. The thickness of the stagnant region, normally denoted by δ , can be determined from the rheological properties of the liquid, the prevailing hydrodynamic conditions and the dimensions of the flow channel (Stephenson, et al., 2000).

Figure 2.2 Concentration polarization

Early researchers believed that near the membrane, the only mode of transport within this layer is diffusion, which is around two orders of magnitude slower than convective transport in the bulk liquid region. However, Romero and Davis (Romero and Davis, 1991) discovered that transport away from the membrane surface is much greater than that governed by Brownian diffusion and is actually determined by the amount of shear imparted at the boundary layer; such transport is defined as “shear-induced diffusion”. Shear-induced diffusion is related to particle size like Brownian diffusion. While Brownian diffusion decreases with increasing particle size, shear-induced diffusion increases with increasing particle size. In addition, larger particles tend to produce less resistance to permeation once deposited as a cake, for the same mass deposited. Thus, the performance of membrane facilities is likely to depend on the characteristics of particle size distribution in the feed water.

The film theory (Michaels, 1968) assumes all particles are rejected by the membrane. When equilibrium is reached between the convection and diffusion movements, the Fick’s law can be expressed as:

$$J_f = \frac{D}{\delta} \ln \frac{C_e}{C_o} = k \ln \frac{C_e}{C_o}$$

where D is the Brownian diffusion coefficient (m^2s^{-1}), δ is the thickness of concentration boundary layer and C_e and C_o are the concentrations in the deposit and in the bulk solution respectively. This reveals how the polarization layer resistance (R_p) increases with the concentration in the solution (C_o) and decreases with the concentration in the deposit (C_e).

By integrating the Stokes Einstein equation into the film theory, we can get the Leveque solution for laminar flow and Brownian diffusive transport (Stephenson, et al., 2000). Then, the averaged flux can be given by:

$$J = 0.807 \left[\frac{\gamma_0 D_B^2}{L} \right]^{1/3} \ln \frac{C_e}{C_o}$$

where J is permeate flux (m/s), D_B is Brownian diffusivity (m^2/s), L is length of channel (m), and γ_0 is nominal shear rate (s^{-1}).

To attain a good agreement to the performance of colloid filtration, back transport models such as shear-induced diffusion and surface transport have been developed. For dilute monodisperse suspensions ($\phi_b < 0.1$) with a maximum $\phi_m = 0.6$ (Davis and Sherwood, 1990):

$$J = 0.072 \gamma_0 \left[\frac{C_e d_p^4}{C_o L} \right]^{1/3}$$

where γ_0 is the maximum shear rate (s^{-1}), d_p is the particle radius (m). In addition, a more comprehensive model was also developed (Romero and Davis, 1988) based on the shear-induced hydrodynamic diffusion phenomenon.

2.2.3 Membrane blocking models

Main mechanisms for flux decline associated with particle deposition during membrane filtration can be identified as complete blocking, standard blocking, intermediate blocking and cake filtration (see Fig. 2.4 (Judd, 2006)).

Figure 2.3 Fouling mechanisms: (a) complete blocking, (b) standard blocking, (c) Intermediate blocking, (d) cake filtration

These models were first introduced by Hermans and Bredbe (1935) when studying the clarification of dilute suspensions (<100 ppm of solids). In their original work, they considered two extreme cases of constant pressure filtration in the dead-end mode: (a) complete blocking: each particle entering the membrane completely plugs one pore on the surface and (b) standard blocking: small particles form an internal deposit on the walls of the pores, which causes a progressive restriction of the free pore volume. The intermediate blocking was introduced empirically, as a compromise between the complete and standard laws. It presumed that each particle can settle on other particle previously arrived and already blocking some pores or it can also directly block some membrane area.

Then, Hermia (1982) found a conclusive interpretation of the intermediate law, as well as the equations for non-newtonian fluids. Grace (1956) applied these laws to various filter media as a result of the intuitive concept that the standard law holds at the beginning of the filtration process when big pores get filled by small particles while as filtration proceeds and the filter becomes less open, a cake starts to build-up. Table 2.1 gives a summary of constant flux membrane blocking models (Hlavacek and Bouchet, 1993; Agustin Suarez and Veza, 2000).

Table 2.1 Summary of constant flux membrane blocking models

Model	Equation	Linearized form
Pore blockage/ Complete blocking	$\frac{1}{\Delta P} = \frac{1}{\Delta P_0} - \frac{\pi k_p r_p^4}{8 \mu L} t$	$\frac{1}{\Delta P} = a + bt$
Pore constriction/ Standard blocking	$\frac{1}{\sqrt{\Delta P}} = \frac{1}{\sqrt{\Delta P_0}} - \frac{c_1 V}{\sqrt{8 \pi N_p \mu L^3 Q}} t$	$\frac{1}{\sqrt{\Delta P}} = c + dt$
Intermediate blocking	$\Delta P = \Delta P_0 \exp K_3 t$	$\ln(\Delta P) = e + ft$
Cake formation	$\Delta P = \Delta P_0 + \frac{Q_0^2 \mu \alpha C_b}{A^2} t$	$\Delta P = g + ht$

Classical dead-end filtration laws can also be adapted for cross-flow operation if the proportion of undeposited solute material are calculated. Nataraj et al. (2008) and many others have applied these laws to cross-flow microfiltration.

2.3 Parameters affecting membrane fouling

MBRs have been widely used in wastewater treatment. However, membrane fouling represents the main limitation to membrane process operation. In MBRs, backwashing is normally used to clean the membrane surface and reduce the fouling. However, it can only remove gross solids attached to the membrane surface, generally termed “reversible” fouling (Judd, 2006), and those can not be removed are termed “irreversible” fouling.

At a constant driving force, fouling on the membrane will cause the flux, which is defined as the flow rate across the membrane over the area of the membrane, to decline with time. Otherwise, there is a need to continuously increase the driving force across the membrane to obtain a constant flux. This would undoubtedly lead to a higher energy use requirement. Besides, a higher cleaning frequency and a shorter lifespan of the membrane will also manifest, thus resulting in higher operating costs, which is the main limitation to the application of MBRs.

Fouling is defined as the attachment or adsorption of retained substances to the membrane surface or within the membrane pores (Bacchin, et al., 2006). It is the result of complex interactions between activated sludge components and the membrane involving different parameters, and can be classified under physical, inorganic or organic fouling.

Physical fouling refers to the fouling caused by suspended and colloidal matter, which adsorbs to the membrane, causing a proportion of the membrane surface blocked (Belfort, et al., 1994). Inorganic fouling, also known as scaling, can be defined as the deposition of particles on a membrane, causing it to plug (Cicek, et al., 1999). The precipitation of salts on membrane surface can be prevented by adding acids or anti-scalants to the wastewater treatment system.

Organic fouling is a major setback to membrane performance. It was caused by organic components which initiate from feed water such as natural organic matter (NOM) or microbial products, such as EPS. They perform both adhesion of microorganisms to inanimate surfaces and cohesion of the biofouling, thus forming a slime matrix that "glues" the cells to the surface and hold the biofilm together (Flemming, et al., 1997). The term "EPS" is a general and comprehensive concept for different classes of macro-molecules such as polysaccharides, proteins and other compounds which have been found at or outside the cell surface and in the intercellular space of microbial aggregates (Flemming and Wingender, 2000). It can be classed into the EPS extracted artificially from the biological cell floc (eEPS) and the soluble EPS present in the activated sludge supernatant (Le-Clech, et al., 2006).

All the parameters involved in the design and procedure of MBR operation would affect membrane fouling, as shown in Fig. 2.5 (Le-Clech, et al., 2006). While some of the parameters, e.g. operating conditions, have direct influence on membrane fouling, many other factors can only influence membrane fouling subsequently. Furthermore, complex interactions exist between these parameters (Judd, 2006; Le-Clech, et al., 2006). For example, SRT determines the concentration of the mixed liquor which impacts both on the biological properties, and also on the physical properties such as viscosity and oxygen transfer. As the mixed liquor is generated

from the feed water, its chemistry presents the greatest challenge on MBR operation. It is therefore crucial to have a full understanding of the membrane fouling phenomenon to access the fouling mechanism.

Figure 2.5 Factors affecting fouling in submerged MBR

2.3.1 Membrane characteristics

2.3.1.1 Physical parameters

Pore size and distribution, porosity and roughness of membranes are suggested relating to the effect of membrane fouling.

The effects of pore size on membrane fouling are strongly related to the feed solution characteristics, especially the particle size distribution. Granger et al. (1985) studied the relation between the membrane pore size and the particle size and found that the worst situation occurred in the case of particles and pores having the same diameter. Because of the complex and changing nature of the biological suspension in MBR systems and other operating conditions like CFV and operating modes of constant flux or constant pressure, results reported in literature have shown somewhat conflicting trends.

MF membrane has a larger pore size and thus a higher permeate flux than UF under the same operating condition. Nevertheless, the permeate flux for MF declines more quickly than UF. This was due to greater internal fouling for larger membrane pore size (Chang, et al., 1994; Hong, et al., 2002). However, Chang et al. (2001) observed that MF membranes with smaller pores had the highest initial fouling while MF membranes with the larger pores had the greater long-term fouling, after experimenting MF membrane with pore size ranging from 1.5 to 5 μm at a constant TMP of 6 kPa. Another study conducted by Lee et al (2005) operated MBRs with four membranes of different pore sizes ranging from 0.1 to 0.8 μm . Then, the molecular weight distributions of the different components in the supernatants of these MBRs were presented. Though the 0.8 μm pore size MBR featured a slightly higher concentration of macromolecules in the bioreactor supernatant, the small differences in molecular weight distribution do not seem the main cause of the diverse fouling rates of these four different MBRs. Mochizuki and Zydney (1993) also discovered similar results when examining the effect of different pore size distributions on the asymptotic membrane sieving coefficient, the hindered solute diffusivity, and the membrane hydraulic permeability. They found that although the calculated sieving coefficients and hindered diffusivities both increased significantly with an increase in the breadth of the distribution, the relationship between these two membrane transport parameters was relatively unaffected by the pore size distribution. In addition, Choi et al. (2006) found that the fouling behaviours of the MF and UF membranes were different when CFV changed.

Membrane porosity and roughness were also reported as causes of differing fouling behaviours. In one study conducted by Fang et al. (2005), the track-etched membrane provided the lowest resistance as a result of its high surface isoporosity as compared to three other membranes. Surface roughness has also been mentioned as promoting fouling from studies on an anaerobic MBR system (He, et al., 2005).

2.3.1.2 Chemical parameters

Membrane chemical properties like hydrophobicity and material composition, can also influence the membrane fouling directly. Because of the hydrophobic interactions between solutes, microbial cells and membrane material, hydrophilic

membranes are expected to yield lower fluxes, i.e., more severe membrane fouling (Chang, et al., 1999; Madaeni, et al., 1999; Yu, et al., 2005; Yu, et al., 2005). Lee et al. (2007) discovered the major mechanism in membrane fouling for hydrophilic and negatively charged membranes was cake formation while internal fouling also played a role for hydrophobic and positively charged membranes. Also, it has been demonstrated that change in membrane hydrophobicity often occurs with other membrane modifications such as pore size and morphology (He, et al., 2005). A research conducted by Chang et al. (2001), who compared two UF membranes of different hydrophobicity and otherwise similar characteristics, indicated greater solute rejection and fouling and higher cake resistance for the hydrophobic membrane. It has also been suggested (Fang and Shi, 2005) that membranes of greater hydrophilicity are more vulnerable to deposition of foulants of hydrophilic nature.

Besides the hydrophobicity, membrane materials can also affect membrane fouling. Fouling behaviours are obviously different in ceramic membranes and polymeric-based membranes. Because of the high cost, ceramic membranes are not widely used for MBR applications although they feature superior chemical, thermal and hydraulic resistances. A direct comparison study between PE and PVDF membranes clearly revealed that PVDF membrane lead to a better prevention of irreversible fouling and that PE membrane fouled more quickly (Yamato, et al., 2006). Another observation by Choi et al. (2008) indicated that the permeate flux profiles of the PCTE and PTFE membranes were almost similar while the PETE membrane had the most rapid flux decline rate.

However, these parameters only play significant roles during the early stage of membrane fouling. After the initial, the membrane's chemical characteristics would become secondary to those of the sludge materials depositing on the membrane surface (Le-Clech, et al., 2006).

2.3.2 Feed-biomass characteristics

2.3.2.1 Feed characteristics

As the membrane fouling depends mainly on the interactions between membranes and the biological suspension rather than the wastewater (Choi, et al., 2005), the feed characteristics can only influence membrane fouling indirectly. The most notable effect of the wastewater nature is on the physic-chemical changes in the biological suspensions (Le-Clech, et al., 2003), resulting in the changes of membrane fouling performance. For example, compared with biomass in the MBR fed with real sewage (COD of 139mg/L), the MBR fed with synthetic feed (COD of 460 mg/L) had a lower protein fraction of EPS extracted artificially from the biological cell floc (eEPS), which was distinguished from the soluble EPS (sEPS) present in the supernatant and unassociated with the cell. Simultaneously, the MBR fed with synthetic feed had a higher fouling rate (Le-Clech, et al., 2003). In conclusion, the feed characteristics can affect the membrane fouling behavior by changing the characteristics of mixed liquor which interacts with the membrane directly.

2.3.2.2 Sludge fractionation

Activated sludge biomass can be fractionated into three idealized components according to its size, namely, suspended solids, colloids and solutes. This has been applied to account for the relative contribution of each biomass fraction on MBR fouling.

As separation methodology varies from one study to another, it remains a crucial step in the definitions of different biomass fractions and the interpretation of results. However, no standard method of separating different biomass fractions has been set up to date. A typical process to determine hydraulic resistances for suspended solids (R_{ss}), colloids (R_{col}) and soluble species (R_{sol}) is shown in Fig. 2.6 (Bae and Tak, 2005). The sample is centrifuged. Then, one portion of the supernatant is filtered with a dead-end membrane cell, and the calculated hydraulic resistance (R_{sup}) is attributed to colloids and soluble species. Another portion of the supernatant is then filtered by a microfilter with nominal pore size of 0.5 μ m to separate soluble matters

from the colloids and get the resistance R_{sol} . According to the process, resistances of different fractions in activated sludge can be calculated. The total resistance (R_t) is assumed to be the sum of each fraction resistance, hence the percentage of each fraction resistance to the total resistance can be obtained.

Figure 2.4 Experimental method for the determination of the relative fouling propensity for the three biomass fractions

The initial research work of membrane fouling contribution of different biomass fractions was conducted by Fane et al. (1991). They separated suspended solids and dissolved matter in biomass suspensions using a Whatman paper filter and found that dissolved matter accounted for 60% of the total resistance to the filtration caused by fouling as the largest contributor.

Wisniewski and Grasmick (1998), who studied the effect of recirculation, divided the biological suspensions into three parts, the SS were eliminated by settling for 2 h, the supracolloidal–colloidal fraction and the soluble fraction were recovered in the permeate after filtration through a membrane of $0.05\mu\text{m}$ pore size. The results revealed the major role of the soluble fraction in membrane fouling, which accounted for 52% of the total resistance while colloidal fraction accounted for 25% and SS accounted for 23% respectively. Another study suggested that the colloid fraction was an important factor in membrane fouling as it could cause polarization or pore clogging. It was confirmed by the experiment results observed by Itonaga et al. (2004). They operated two identical MBRs: the conventional MBR without pre-treatment and the hybrid MBR with pre-coagulation/sedimentation. The greatest

contributor to membrane fouling of the suspension collected from hybrid MBR was discovered to be colloidal matter while soluble matter accounted for the highest fouling potential in conventional MBR. However, it is noteworthy that in these studies, the interactions between different fractions were ignored.

Different phenomena were observed subsequently. In one study about the treatment of synthetic wastewater by a Membrane Granular-Sludge BioReactor (MGSBR), the SS appeared to be the main contributor and the colloids and solutes were only partially responsible for the membrane fouling (Li, et al., 2005). Defrance et al (2000) tested the contributions of various particle fractions present in urban waste sludge and investigated how fouling varied with individual concentration of each constituent (bacteria, colloids, dissolved matters and SS). They found out that SS caused majority of the fouling during the membrane filtration. Furthermore, they discovered that the total resistance was much lower than the sum of the resistances of each constituent at proper concentration, which demonstrated the interactions present between different fractions.

Although many attempts have been made about the contributions of different fractions, the results do not seem to have good agreement with one another. The relative contribution of the biomass supernatant (soluble and colloids, generally defined as soluble microbial products or SMP) to the total membrane fouling ranges from 17% (Bae and Tak, 2005) to 81% (Itonaga, et al., 2004). This is probably due to the different operating conditions and biological states of the suspension. In terms of fouling mechanisms, solute and colloidal materials are assumed to be responsible for the pore blockage of the membrane, while suspended solids account mainly for the cake layer resistance (Itonaga, et al., 2004).

2.3.2.3 Biomass (bulk) characteristics

MLSS concentration is considered as the main factor affecting membrane fouling. Although many researchers have studied the effects of MLSS concentration on membrane fouling, different results were reported (Wu, et al., 2007). If the other biomass factors are not accounted for, it seems that the increase of the MLSS concentration causes lower flux or higher TMP. However, some researchers

(Brookes, et al., 2006) found the positive influence of the MLSS concentration increase, while others observed insignificant impact (Hong, et al., 2002; Lesjean, et al., 2005). Rosenberger et al. (2005) reported that at low MLSS concentration (< 6 g/L), a rise in MLSS seems to lessen fouling; between 8 and 12 g/L, the MLSS concentration does not appear to have significant effect on membrane fouling; and at concentrations above 15 g/L, greater fouling is expected as the MLSS concentration increases. These contradictory results may arise from the different flux value at which these experiments were carried out, the test duration, the mode of filtration, i.e. constant flux or constant TMP, and the methods used in changing MLSS concentration. Therefore, the relationship between MLSS and fouling behaviour would require more elaborate studies.

In addition, biomass viscosity, temperature, dissolved oxygen and other bulk characteristics affect the membrane fouling as well. The MLSS viscosity can modify bubble size and depress the movement of hollow fibers in submerged bundles (Wicaksana, et al., 2006). Increased viscosity also reduces the efficiency of mass transfer of oxygen and therefore effects dissolved oxygen (DO) content (Germain and Stephenson, 2005). Temperature impacts on membrane filtration indirectly by influencing the permeate fluid viscosity and greater deposition of materials on the membrane surface will be observed at lower temperatures (Rosenberger, et al., 2006). DO has multiple effects on membrane fouling as it can change the biofilm structure, SMP levels, and floc size distribution (Lee, et al., 2005). Generally, higher DO concentrations tend to lead to lower fouling rate (Kim, et al., 2006).

2.3.2.4 Floc characteristics

Evidence suggests that the physical nature, specifically the size of the foulant has the greatest impact on its fouling propensity. According to the Carmen-Kozeny equation, the smaller the floc size, the greater the cake resistance will be. In general, the floc size in MBR includes a wide distribution, which ranges significantly from one study to another. Floc sizes ranging from 10 to 40 μm were reported with a mean size of 25 μm (Bae and Tak, 2005) while Zhang et al. (1997) found that the floc size in submerged MBR was 20-40 μm . For three MBRs operated at different

SRTs (from 20 to 60 days), floc size distributions reported were similar, although the mean floc size increased slightly from 5.2 to 6.6 μm (Lee, et al., 2003).

Given the large size of the floc particles compared to the pore size of the membrane generally used in MBR, pore plugging by the flocs is impossible. Flocs are also prevented from depositing on the membrane surface by drag forces and shear-induced diffusion. However, independent of their size, biological flocs play a major role in the formation of the fouling cake on the membrane surface.

Hydrophobicity and surface charge of flocs affect membrane fouling to a certain extent as well. In general, hydrophobic flocs lead to high flocculation propensity and low interaction with the hydrophilic membrane. However, a number of reports can be found in literature, providing evidence of highly hydrophobic flocs fouling MBR membranes. Relative floc hydrophobicity can be directly measured by bacterial adhesion/partition using hydrocarbons such as hexane (Jang, et al., 2005), or estimated by contact angle determination (Yu, et al., 2005). Due to the negative charges from ionization of the anionic functional groups, flocs (and EPS) of most activated sludge feature zeta potential and surface charges ranging from -0.2 to -0.6 mequiv./g VSS and from -20 to -30mV , respectively (Liu and Fang, 2003). Increasing SRT has been shown to produce an increase in both contact angle and surface charge, with an apparently strong correlation with fouling propensity (Lee, et al., 2003).

2.3.3 Operating conditions

2.3.3.1 Aeration, cross-flow velocity

In submerged membrane systems, aeration is used to provide oxygen to the biomass, maintain the activated sludge in suspension and induce the tangential flow to scour the membrane surface (Dufrèsne, et al., 1997).

Basically, the bubbles flowing near the membrane surface create local shear transients and liquid flow fluctuations, increasing back transport phenomenon. The tangential shear at the membrane surface prevents large particle deposition on the membrane surface and thus reduces the hydraulic resistance of the fouling layer.

However, it can also break large particles into small ones, resulting in more severe irreversible fouling. An optimum value of air flow rate was identified beyond which further increases had no effect by Ueda et al. (1997).

Many studies were focused on the shear rate caused by the tangential flow at the surface of a membrane. During cross-flow filtration, the rate of deposition is balanced by permeate drag force, back diffusion from membrane surface and the convection towards the filter exit by tangential flow. It is believed that higher shear rate produced by higher CFV will increase the back diffusion and the convection towards the filter exit. The striking influence of the shear rate on the membrane flux in systems containing colloidal substances was reported by Ann-Sofi Jönsson (1993). It was observed that if the shear rate was decreased, flux would decrease as a result of the increase in thickness of the boundary layer of retained solutes at the membrane surface. And flux would increase again if the shear rate was increased for low molecular solutes. However, for large solutes, like polymeric and colloidal substances, the flux cannot be recovered to its original value sometimes because of irreversible fouling. Another study confirmed that tangential flow caused slightly higher irreversible fouling due to higher permeation drag, as compared with the case of absent tangential flow (Choi, et al., 2005). In another study, the researchers also gathered results that permeate flux increased linearly with increasing cross-flow velocity and a high CFV was more effective in reducing fouling of MF membranes than that of UF membranes. Furthermore, the formation of a reversible fouling layer was actually prevented by a CFV of 3.0 m/s for MF membrane and 2.0 m/s for UF membrane (Choi, et al., 2005). In a series of flux-stepping experiments conducted by Ye et al (2005), who investigated the evolution of fouling of sodium alginate for a better understanding of the fouling mechanisms of EPS, the rates of TMP increase in each flux step (dP/dt) were found to decrease with increasing CFV for the similar permeate flux; This demonstrated that fouling during the subcritical filtration was not only caused by adsorption, but also by small amount of deposition. However, Beaubien et al. (1996) reported that fouling resistance during low-pressure operation was independent of CFV. The different results between the two studies may be due to different operation modes.

2.3.3.2 Flux, Trans-membrane pressure

There are two different operation modes of the MBRs, constant flux vs. constant TMP. In general, constant pressure experiments can be recommended when working with suspensions, containing components that show little tendency to adsorb onto the membrane, as steady state permeate fluxes will be achieved. On the other hand, constant flux experiments are well adapted to reveal fouling phenomena with complex suspensions as steady state fluxes may not be achievable.

For the constant flux operation mode, flux selection is an important factor in determining fouling rate. Generally speaking, membrane fouling increases with increasing imposed flux, whereas no fouling was observed when imposed flux was maintained below the critical flux (Chang, et al., 2002; Judd, 2004). Theoretically, the critical flux can be defined as the flux at which the hydrodynamic force transporting the particle towards the membrane pore and the opposing back transport forces reach an equilibrium (Bowen, et al., 1999). However, conditions that are believed to prevent the average particle from depositing on membrane surface will not guarantee that all particles of a particular size and density will remain detached from a surface. Thus, critical flux should be considered to be the operating condition that separates slow fouling from rapid fouling rather than the flux at which there will be no fouling at all. The critical flux value is dependent on the membrane characteristics, mixed liquor composition and operating conditions, therefore critical flux has to be determined experimentally for each case.

For the constant TMP operation mode, a rapid flux decline occurs initially, then the rate of fouling decreases until it reaches a plateau. It is generally considered that higher TMP leads to higher filtrate flux at the beginning of the experiment. However, more solutes are driven to the membrane. Some are able to penetrate through the porous medium and cause fouling. Simultaneously, pressure causes settling of the deposits and decreases its porosity. In addition, some substances were able to precipitate or form a gel layer by localized over-concentration (Ousman and Bennisar, 1995). As a result, higher TMP causes higher fouling resistances, as confirmed by Ugur Salgın and Sema Salgın (2007). Furthermore, they observed that the TMP was more effective than the CFV in the initial stage of the cross-flow

filtration. In another study, it was found that for a given CFV, an increase in TMP reduced the concentration of both the BSA protein and dextrin components of the model beer in the permeate and owed the phenomenon to the increase of fouling layer thickness, coupled with a decrease in fouling layer voidage at higher TMPs (Thomassen, et al., 2005).

However, Li et al. (2007), who examined the variation of different resistances according to different operating conditions during MF of raw soy sauce, found that while the concentration polarization layer resistance R_{cp} was proportional to the TMP, the cake resistance R_c grew gradually with increasing TMP from 0.05 to 0.1 MPa, but decreased slightly when TMP was higher. This can be explained that some fine particles in the cake layer might be transported into the membrane pores, resulting in more severe internal membrane fouling R_{if} .

2.3.3.3 *Solid retention time*

Because SRT influences the mixed liquor characteristics (MLSS concentration, particle size, EPS content, etc.), which can then be directly related to fouling rates (Chang and Judd, 2002), it is probably recognized as the most important operating factor affecting the fouling propensity in MBRs.

It was reported that fouling rate increased nearly 10 times when SRT was lowered from 10 to 2 days (Trussell, et al., 2006). The similar tendency was also observed by Bouhabila et al. (1998), who studied the MF MBR system and found that 30 days SRT condition provided a slower fouling rate compared with a 10 or 20 days SRT system. Basically, given the small amount of excess sludge and the low fouling rate, a long SRT condition is preferred. Nevertheless, higher MLSS concentration related with long SRT may also result in higher fouling propensity. Experiments conducted by Han et al. (2005) revealed an increase in MLSS levels when the SRT was increased from 30 to 100 days, resulting in increased membrane fouling even when the aeration was intensified significantly. Long SRT conditions also lead to high viscosity which could attenuate the effect of aeration.

SRT plays a critical role in influencing biomass characteristics, like MLSS, SMP, eEPS concentrations, thus selection of it must be precise and meticulous in order to optimize MBR operation.

2.4 Summary

As discussed before, membrane fouling is a result of complex interactions between membrane and mixed liquor. Numerous researches have been devoted to the study of membrane fouling in MBRs. Since biomass characteristics vary significantly from one plant to another, the results are not in good agreement with one another. SRT, which is considered as one of the main operating parameters during the characterization of the biomass suspension and its fouling propensity, can then be directly related to fouling rates.

Long SRT has been reported to be an effective way to minimize the sludge production in MBRs. Some studies were focused on infinite SRT, i.e. no waste sludge produced (Orantes, et al., 2004; Ito, et al., 2005). However, industrial application of long SRT MBRs is still limited because of insufficient knowledge about biomass activity and system stability in long SRT conditions. Furthermore, there is not much information available with regards to the effects of long SRT condition on membrane filtration performance.

Therefore, detailed studies on the effects of different parameters and contributions of different sludge components to membrane fouling under long SRT condition are required to achieve a better understanding of fouling behaviour under such SRT condition.

CHAPTER 3 EXPERIMENTAL SETUP AND PROCEDURE

3.1 Introduction

Membrane fouling is a result of deposition of organic and inorganic materials onto and into the membrane, attributed to the interactions between activated sludge components and the membrane itself. As the mixed liquor composition varies with different operating conditions, the contribution of each fraction is changed along with varying operating conditions. In this study, the fouling behaviors of activated sludge from MBR system with SRT of 300 days and SBR system with SRT of 10 days, fed with the same synthesized wastewater, were compared using the Cross-flow filtration membrane unit. In the first stage, the fouling contributions of different fractions from these two types of sludge were tested. The MLSS concentrations were adjusted to the same level with deionized water to avoid the concentration effect of membrane fouling. Analysis in the second phase focused on the effect of MLSS concentration, TMP and CFV on the contributions to membrane fouling by different activated sludge's components, in terms of resistance. Then, filtration resistances were calculated according to the resistance-in-series model. Observations of fouled membranes were also conducted using field scanning electron microscope (FESEM).

3.2 Materials

One type of activated sludge used in this study was taken from a submerged MBR with 300-day SRT and the other was cultured in a SBR with SRT of 10 days for more than one month, until the performance became stable. Fig 3.1 presents the schematic diagram of the MBR and SBR systems. The characteristics of the synthetic wastewater used in this study and the operating conditions of the MBR and SBR systems are listed in Table 3.1 and Table 3.2 respectively. The feed water was synthesized using milk-based powder (high calcium low fat, Anlene) to represent the milk industrial wastewater, and the concentration is around 1g/L. MF polypropylene membranes with effective pore size of 0.2 μm (GHP-200, Pall

Corporation) were employed in the cross-flow filtration experiments. The new membrane was first rinsed by letting it float in deionized water for 24 hours. Before each experiment, filtration using deionized water was performed for 30 minutes in order to compact the new membrane surface (i.e., compaction between membrane surface and supporting layer), as well as to stabilize permeate flux and operating conditions.

Figure 3.1 Schematic diagram for (a) MBR (b) SBR

Table 3. 1 Characteristics of the synthetic industrial wastewater

Component	Concentration
COD (mg/L)	1000
Soluble COD (mg/L)	813.4
TOC (mg/L)	377.2
NH ₄ ⁺ -N (mg/L)	12.6
NO ₃ ⁻ -N (mg/L)	5.64
NO ₂ ⁻ -N (mg/L)	0.8
pH	6.10

Table 3. 2 Operating conditions of MBR and SBR systems

Parameters	300-day MBR	10-day SBR
Working volume (L)	20	5
HRT (hr)	24	25
SRT (d)	300	10
Flux (L/m ² ·hr)	5.78	
VLR (kgCOD/m ³ ·d)	1	1.44
Airflow rate (L/min)	6	1.5
Temperature (°C)	25	25

3.3 Experimental system and analysis

Flux decline behavior was measured using a cross-flow filtration membrane unit, Model C10-T (Nitto Denko Co., Japan), with an effective membrane surface of 60cm². The rotary pump (MOD. DPS.68MMTTIRM0000, SER. 906944, Tuthill Pump Co., USA) used in the unit is driven by a motor (41K60A-BF, Okiental Motor Co., LTD, Japan). The schematic diagram is shown in Fig. 3.2. The feed activated sludge was pumped across the membrane module and recirculated back to the feed tank, which was placed on the magnetic stirrer to ensure complete homogenization. Constant TMP was established by in/outlet valves. The permeate was collected in a beaker placed on a digital balance and connected to a computer. Both retentate and permeate were returned to the feed tank to keep concentration constant.

In this study, the activated sludge samples were diluted with deionized water to different MLSS concentrations. The TMPs used in this study were 0.5, 1, 1.5, and 2 bars. The CFVs were 0.438m/s, 0.279m/s and 0.156m/s.

Figure 3.2 Schematic diagram of cross-flow membrane unit

MLSS and MLVSS (mixed liquor volatile suspended solids) concentration were measured based on Method 2540D and 2540E (AWWA, 1998) respectively. COD was measured based on Method 5220D (AWWA, 1998), via a closed reflux, colorimetric method. The particle size distribution of activated sludge and its supernatant were analysed using Malvern Master Sizer Model Hydro 2000S. The molecular weight distribution was measured by the HPSEC (Perkinelmer 200 with a Shodex KW 802.5 SEC column), under the wave length of 224nm. The FESEM imaging analysis was conducted using JEOL, JSM-6340F.

3.4 Fouling contributions of sludge fractions

It was assumed that the activated sludge used in this study consisted of three components, i.e., suspended solids, colloids and solutes. Any interactions between these fractions were neglected, which means that the overall resistance of the activated sludge equals to the sum of the resistances of each fraction. It can be expressed as:

$$R_{AS} = R_{ss} + R_{col} + R_{sol} \quad (3.1)$$

where R_{AS} is the resistance of the activated sludge, R_{ss} is the resistance of the suspended solids, R_{col} is the resistance of colloids and R_{sol} is the resistance of the solutes.

In order to separate these fractions from one another, a series of pretreatment were operated. One portion of the activated sludge were centrifuged at 3000 rpm for 10 minutes, then the supernatant to about 1.5~ 2 cm depth from the liquid surface in the centrifuging tube were collected. It was assumed that the supernatant contained only the colloids and the solutes. Another portion of the activated sludge were filtrated using a MF membrane (Nylon) with pore size of 0.45 μm to obtain the solutes.

The resistances of each fraction can be measured from the filtrations of different feeds:

for activated sludge,

$$R_{AS} = R_t - R_m = \frac{\Delta P}{\mu J_{AS}} - R_m \quad (3.2)$$

for supernatant containing the colloids and the solutes,

$$R_{sup} = R_{col} + R_{sol} = \frac{\Delta P}{\mu J_{sup}} - R_m \quad (3.3)$$

for the solutes,

$$R_{sol} = \frac{\Delta P}{\mu J_{sol}} - R_m \quad (3.4)$$

where R_t is the total resistance (m^{-1}), R_m is the intrinsic membrane resistance (m^{-1}), R_{sup} is the resistance of supernatant (m^{-1}), ΔP is the trans-membrane pressure (Pa), μ is the viscosity of permeate ($\text{N}\cdot\text{s}/\text{m}^2$), J_{AS} , J_{sup} , and J_{sol} are the fluxes of the activated sludge, the supernatant and the the solutes at steady state ($\text{m}^3/\text{m}^2\cdot\text{s}$) respectively.

R_{AS} , R_{ss} , R_{col} and R_{sol} were calculated by equations (3.1) to (3.4).

3.5 Fouling resistances analysis

The degree of membrane fouling was quantitatively calculated according to the resistance-in-series model (Ousman and Bennisar, 1995). It can be expressed as:

$$J = \frac{\Delta p}{\mu R_t} = \frac{\Delta p}{\mu(R_m + R_{cp} + R_c + R_{if})} \quad (3.5)$$

where R_t , R_{cp} , R_c and R_{if} are the overall filtration resistance (m^{-1}), the concentration polarization layer resistance (m^{-1}), the cake resistance (m^{-1}) resulted from the cake layer formed by the deposited particles and other solutes, and the membrane internal fouling resistance (m^{-1}) resulted from membrane plugging and/or adsorption of foulants, respectively.

Among all of these resistances, R_{cp} can be removed by rinsing with deionized water; R_c can be removed by cleaning the membrane surface with a sponge while R_{if} can be removed by chemical cleaning (Li, et al., 2007). Therefore, these different resistances can be determined as follows:

$$R_m = \frac{\Delta P}{\mu J_{w1}} \quad (3.6)$$

$$R_t = R_m + R_{cp} + R_c + R_{if} = \frac{\Delta P}{\mu J_{AS}} \quad (3.7)$$

$$R_m + R_c + R_{if} = \frac{\Delta P}{\mu J_{w2}} \quad (3.8)$$

$$R_m + R_{if} = \frac{\Delta P}{\mu J_{w3}} \quad (3.9)$$

where J_{w1} , J_{w2} , J_{w3} are the deionized water flux ($m^3/m^2 \cdot s$), the deionized water flux after removing the concentration polarization layer ($m^3/m^2 \cdot s$) and the deionized water flux after removing the cake layer ($m^3/m^2 \cdot s$). Then, each resistance can be calculated using the equations above.

CHAPTER 4 MEMBRANE FOULING BY VARIOUS SLUDGE FRACTIONS FROM 300-DAY SRT MBR AND 10-DAY SRT SBR

4.1 Performances of 300-day SRT MBR and 10-day SRT SBR

The MBR with SRT of 300 days has been operated for over 2 years. While the SBR with SRT of 10 days, which was set up as the conventional activated sludge process, has been cultured for more than 1 month before the experiments. The COD/TOC removal, MLSS, MLVSS concentration and F/M ratios were measured to assess the biological performance of 300-day SRT MBR and 10-day SRT SBR separately.

Table 4.1 summarized the performances of COD/TOC removal efficiency in submerged MBR and SBR treating the high strength synthetic industrial wastewater. It was observed clearly that the permeate COD of MBR had been reduced to 11.8 mg/L from around 1000 mg/L of influent wastewater during the steady-state, leading to a overall COD removal efficiency of 98.8%. The high COD removal efficiency was attributed to the performance of both biological activities and membrane filtration. In line with the results shown in our previous work (Sun, Hay et al. 2006), the biological COD removal efficiency of MBR, which was determined from the supernatant of the activated sludge after it was centrifuged in 3000rpm for 10 mins, was similar to that for SBR. Their values are 93.9% and 94.8% respectively.

In MBRs, the membrane separation serves as an addition purification phase to retain the remaining particulate COD. Coupled with the membrane module, the bioreactor achieved the higher removal efficiency, from 93.9% to 98.8%. Such high removal efficiency may be due to the complete retention of all particulate and biomass in the bioreactor by the membrane separation, establishing the microorganism that able to remove all excess substrate and hence improve the treatment efficiency. The removal efficiency of TOC increased from 95.7% to 98.6% as well.

Table 4.1 COD/TOC performances for MBR with SRT of 300 days and SBR with SRT of 10 days

	MBR		SBR	
	COD	TOC	COD	TOC
Influent (mg/L)	1000	400	1000	400
Supernatant (mg/L)	61.1	28.9		
Effluent (mg/L)	11.8	5.6	52.4	17.1
Biological removal (%)	93.9	92.8	94.8	95.7
Total removal (%)	98.8	98.6	94.8	95.7

MLSS and MLVSS concentration in MBR remained about 30000 mg/L and 26500 mg/L respectively. Therefore, the ratio of MLVSS to MLSS stayed stable at about 88%. The relatively stable ratio of MLVSS to MLSS indicated that the amount of biomass and inorganic remainders constituted a dynamic balance. There was no obvious accumulation of inorganic compounds in the MBR system. This finding is acceptable as the synthetic wastewater contained very small amount of inorganic trace elements. This was in agreement with the results presented by other researchers (Rosenberger, 2002). They operated an MBR for 3 years and discovered no decrease in the volatile fraction with the MLSS/MLVSS ratio remaining constant at approximately 75%.

Meanwhile, the MLSS and MLVSS concentration in SBR were 3000 mg/L and 2750 mg/L respectively, resulting in a MLSS/MLVSS ratio of 92 %. This high ratio was probably derived from the high organic loading. F/M ratios of MBR and SBR were 0.067 kgCOD/kgMLSS.d and 0.48 kgCOD/kgMLSS.d respectively. Compared to the MBR, there were huge amount of excess food substrates presented in the SBR, which would contribute to the volatile suspended solids concentration. With lower biomass concentration, there might not be complete degradation of organic matters.

From results discussed above, the performances of 300-day SRT MBR and 10-day SRT SBR had already achieved stable and the characteristics of activated sludge, i.e. mixed liquor, remained constant.

4.2 Characteristics of different sludge fractions

As mentioned in Chapter 3, the activated sludge was fractionated into three constituents: suspension solids, colloids and solutes. The supernatant, containing colloids and solutes, were acquired by centrifuging at 3000 rpm for 10 minutes, while the solutes were obtained by filtration of the activated sludge with 0.45 μm MF membrane (Nylon).

(a)

(b)

Figure 4.1 Particle size distribution of activated sludge in (a) MBR; (b) SBR

Fig. 4.1 shows the particle size distributions of the activated sludge suspension in MBR and SBR. Samples taken from the MBR and SBR were diluted by 100 times and then measured. The particles in the MBR activated sludge suspension ranged from 2 to 140 μm , and the mean particle size was 54 μm . Whereas the particles in

the SBR activated sludge suspension were at 2 to 550 μm , with the mean particle size of 125 μm , relatively larger than that in the long SRT MBR. This phenomenon was confirmed by Zhang et al. (1997), who compared four MBRs with four conventional activated sludge processes and found that the size distribution of flocs were smaller in MBRs at 7 to 40 μm than 70 to 300 in conventional activated sludge. The smaller sludge particles in the bioreactor would enhance the mass transfer for both carbon and oxygen, thus enabling the system to perform more sustainable with a higher organic removal rate.

(a)

(b)

Figure 4.2 Particle size distribution of the supernatant in (a) MBR; (b) SBR

Fig. 4.2 shows the particles size distribution of the supernatant in MBR and SBR. The supernatant is assumed to contain colloids and solutes only. In the MBR supernatant, the particles ranged from 2 to 130 μm , and the mean particle size was 30 μm . Although the range did not change a lot, particles larger than 60 μm , were

almost removed by centrifugation. For supernatant from SBR, there were two peaks of particle size, one is around 40 μm and the other is 400 μm . This would possibly because that some large particles formed by bacterial cells adhered to each other did not readily settle down even after the centrifuge and were collected as part of the supernatant. Therefore, the supernatant used in this study contained some large particles called supra-colloids besides the colloids and solutes. However, they are still referred as colloids for convenience.

Figure 4.3 Molecular weight distribution of MBR and SBR solutes

Fig. 4.3 displays the molecular weight distribution of solutes separated from MBR and SBR. There are mainly two peaks for both solutes, one was at 15-20 Da, while the other was around 80 Da. It seems no obvious difference existed between the solutes from MBR and SBR, thus various membrane fouling caused by supernatant should mainly attributed to the colloids. However, it should be noted that UV is not able to measure all organic compounds in the solutes because some organic molecules do not response to UV irradiation.

4.3 Filtration characteristics of different sludge fractions

4.3.1 Flux decline behaviors of MBR and SBR activated sludge

The permeate flux declines during the membrane filtration of MBR and SBR activated sludge were studied. All filtration tests were performed for 4 hours to achieve steady-state conditions, given the biological activities as well. The MLSS concentration of each activated sludge suspension was adjusted to around 300 mg/L prior to membrane filtration in order to exclude the concentration effect on the permeate flux and obtain enough amount of influent, about 3 to 5 liters, for cross-flow filtration.

Figure 4.4 Flux declines during the filtration of diluted sludge suspension from MBR and SBR

The results of flux decline behaviors are illustrated in Fig. 4.4. As the operation conditions of these two systems varied a lot from each other, leading to different sludge characteristics, the flux decline behaviors were expected to be significantly different. However, similar fouling tendencies were observed contrary to expectation. Both activated sludge suspensions from MBR and SBR systems displayed the similar initial membrane flux at 750 L/m²h, after which the membrane flux in both cases declined remarkably, finally reached the steady-state. In addition, the stable membrane flux, i.e., flux after filtration for 4 hours, for activated sludge

suspension taken from 300-day SRT MBR was $40\text{ L/m}^2\text{h}$, while a slightly lower flux of $32\text{ L/m}^2\text{h}$ was observed for activated sludge suspension from 10-day SRT SBR. Compared with the value of initial flux, it indicated that activated sludge from 300-day SRT MBR exhibited almost the same filtration resistance with the activated sludge from SBR with SRT of 10 days.

These results demonstrated that in the same operation conditions, activated sludge with the same MLSS concentration exhibited the similar fouling tendencies. Therefore, it is hypothesized that the suspended solids is the main contributor to membrane fouling as these two types of activated sludge suspension, displaying the similar fouling tendency, were adjusted to the same MLSS concentration. However, the colloids and solutes should also been taken into account as they contributed to membrane fouling as well and caused the slight difference between flux of MBR and SBR in the steady state.

As a result, in-depth study on the role of different sludge constituents, from 300-day SRT MBR and 10-day SRT SBR, in membrane fouling should be conducted to have a detailed understanding about the fouling phenomenon.

4.3.2 Contributions of sludge fractions to membrane fouling

As mentioned in Chapter 3, activated sludge consists of three parts: suspended solids, colloids and solutes, which played different roles during membrane filtration. Fouling contributions of these various fractions from activated sludge in 300-day SRT MBR and 10-day SRT MBR may be variable. Therefore, resistances of sludge fractions were quantified using equations (3.1) to (3.4), as shown in Fig. 4.5.

Figure 4.5 Resistances of sludge fractions from MBR and SBR

Results from previous studies and the present work, summarized in Table 4.2 (Defrance, et al., 2000; Bae and Tak, 2005; Lee, et al., 2003; Wisniewski and Grasmick, 1998; Bouhabila, et al., 2001), were quite different from each other. Those differences were due to (1) the nature of the substrate (some used synthetic wastewater based on milk, some used domestic wastewater), (2) due to fractionation methods (one group used centrifugation to eliminate the suspended solids and flocculation to obtain the soluble fraction, while another group used sedimentation and filtration to separate all sludge fractions), membrane materials and biomass characteristics (Bouhabila, et al., 2001). Furthermore, different operating conditions and other factors may also contribute to these differences. Nevertheless, it could be concluded that the suspended solids, colloids and solutes are all partially responsible for the membrane fouling during the filtration.

Table 4.2 Relative fouling resistances of sludge fractions

	Our results (%)		Defrance	Bae	Lee	Wisniewski	Bouhabila
	MBR	SBR	(%)	(%)	(%)	(%)	(%)
R_{AS}	100	100	100	100	100	100	100
R_{ss}	70	85	65	72-83	63-71	24	24
R_{col}	20	7	30	4-14	29-37 ^a	24	50
R_{sol}	10	8	5	13-14	29-37 ^a	52	26

^a The contribution of supernatant to membrane fouling

(Operating conditions: MLSS = 320mg/L, TMP = 1bar, CFV = 0.438m/s)

In this study, SS was obviously the main contributor to the total membrane fouling both in MBR and SBR systems. It is in agreement with the results reported by Defrance, Bae, Lee et al.. Under the steady state condition, the suspended solids from MBR with SRT of 300 days had a filtration resistance of $2.98 \times 10^{12} \text{ m}^{-1}$, while for the SBR with SRT of 10 days, R_{ss} was $6.44 \times 10^{12} \text{ m}^{-1}$, much higher than that for MBR. And the suspended solids contributed about 70% and 85% to the total resistance during the filtration of activated sludge from the MBR with SRT of 300 days and the SBR with SRT of 10 days respectively. Both types of activated sludge were diluted with deionized water into the same MLSS concentration to exclude the influence of MLSS concentration. As a result, differences between the R_{ss} of MBR and SBR would be derived from the particle size distribution and other characteristics of these two types of sludge. As discussed above, the mean size of particles in MBR system was 53.96 μm , while in SBR it increased to 125.73 μm .

Granger et al. (1985) found that the worst fouling situation occurred when particles and pores having the same diameter. However, compared to the pore size of the membrane used in this study (0.2 μm), the particles were too large to cause pore blocking. Furthermore, they were prevented from deposition on the membrane surface by drag forces induced by cross-flow condition. Nevertheless, they played a major role in the formation of the cake layer on the membrane surface.

According to the Carmen-Kozeny equation, the smaller the floc size, the greater the cake resistance will be. However, in this study, the sludge with large particles from SBR system exhibited higher fouling resistances compared to the sludge with small particles from MBR. This contrary phenomenon may be because that the cake layer formed by large particles were incompact and porous compared with the one formed by small particles, leading to more macromolecular and soluble matters passing through the cake layer and causing severe pore blocking. Detailed information of various resistances would be discussed below.

Colloids, which mainly cause concentration polarization phenomenon, played important role during the membrane filtration as well. The values of R_{col} for MBR and SBR were $0.84 \times 10^{12} \text{ m}^{-1}$ and $0.51 \times 10^{12} \text{ m}^{-1}$, and the relative contributions to the total resistance were 19.6% and 6.8% respectively. Moreover, the importance of solutes cannot be neglected as they attribute to adsorption and polarization partially. The values of R_{sol} for MBR and SBR were $0.46 \times 10^{12} \text{ m}^{-1}$ and $0.61 \times 10^{12} \text{ m}^{-1}$, and the relative contributions were 10.7% and 8.0%.

Under long SRT condition, the microbial community will enter the phase of endogenous respiration as a result of the shortage of available substrate (Orhon and Artan, 1994). Therefore, more colloids and solutes (EPS and SMP) were considered to be produced in microbial metabolism, leading to higher fouling resistances. This may explain the higher R_{col} for the MBR with SRT of 300 days compared to the SBR with SRT of 10 days.

4.3.3 Resistances analysis

To have a deeper understanding of the fouling resistance during the membrane filtration of different feeds, sub-divided resistances, i.e., R_m , R_{cp} , R_c and R_{if} , were analysed. Fig. 4.6 presents the resistances of various influents. And their relative contributions to the total resistance were shown in Table 4.3.

It appears clearly that in both types of activated sludge from MBR with SRT of 300 days and SBR with SRT of 10 days, R_{cp} was the dominant contributor to the total filtration resistance. This is in accordance with the results obtained by Choi et al. (2005), who studied the influence of CFV on membrane performance for MF and

Figure 4.6 Filtration resistances analysis

UF membranes, that R_{cp} was dominant in all cross-flow velocities for MF. The values for 300-day SRT MBR and 10-day SRT SBR were $2.82 \times 10^{12} \text{ m}^{-1}$ and $4.65 \times 10^{12} \text{ m}^{-1}$, with 63.0% and 59.6% contributions to the total resistances respectively. Nevertheless, the suspended solids played the significant role in membrane fouling as discussed above, thus the resistance of cake layer, which mainly caused by large particles, should also dominated the filtration resistance. This contrary result may be due to the low MLSS concentration, i.e. small amount of particles for cake layer formation. Moreover, R_{cp} is defined as a filtration resistance which can be recovered by “rinsing” the membrane where only the feed solution is replaced with deionized water at the same operating condition (Cheryan, 1998). The “rinsing” may result in the derangement of the boundary layer, the increase in back-diffusion of retained materials, and the decrease in osmotic pressure (Bader and Veenstra, 1996; Sablani, et al., 2001). In this study, part of particles loosely deposited on the membrane surface may also be taken away by the cross-flow, thus greater fouling resistances were obtained. Higher R_{cp} for MBR than that for SBR rose from more colloids contained as discussed before.

Table 4.3 Relative contributions of filtration resistances in different influents

	Activated sludge		Supernatant		Solutes	
	%		%		%	
	MBR	SBR	MBR	SBR	MBR	SBR
R_{cp}/R_t	63.04	59.95	29.86	12.84	4.10	10.51
R_c/R_t	30.90	36.42	54.54	67.18	60.27	59.34
R_{if}/R_t	1.55	0.99	1.94	4.27	6.62	5.31
R_m/R_t	4.51	2.64	13.66	15.71	29.01	24.84

On the other hand, R_c also played a remarkable role to the total filtration resistance both for MBR and SBR. The values for 300-day SRT MBR and 10-day SRT SBR were $1.38 \times 10^{12} \text{ m}^{-1}$ and $2.83 \times 10^{12} \text{ m}^{-1}$, taking up 30.90% and 36.42% of the total resistances respectively. Although the MLSS concentration for these two types of activated sludge were adjusted to the same level, the particle size distributions were not changed and the organic content in the two types of sludge still differed from each other. Large particles presented in SBR led to higher cake resistance than MBR. In addition, the contribution of internal fouling was insignificant compared with the contributions of cake layer and concentration polarization for the activated sludge from MBR and SBR. Therefore, R_{if} was negligible. Furthermore, R_m remained constant values around $0.20 \times 10^{12} \text{ m}^{-1}$ because it was not affected by the characteristics of influents.

When the feed influent changed from activated sludge to supernatant and the solutes, the main contributor shifted from concentration polarization to cake layer. For the solutes, there was no significant difference between various resistances for MBR and SBR, implying the similar characteristics of soluble matters in MBR and SBR. While for the supernatant which contained colloids and solutes, the existence of more colloids in activated sludge from MBR with SRT of 300 days was confirmed again by the various R_{cp} values for MBR and SBR.

4.3.4 Deposition of sludge fractions on membrane surface

In order to estimate the performance of sludge fractions along the filtration time, the filtration resistances of sludge fractions according to the relative filtrate volume were studied and the results are shown in Fig. 4.7.

Both types of sludge display a similar pattern with increasing resistances, although the filtration resistance values were different. This result is consistent with the previous studies of other researchers (Bae and Tak, 2005), while in the present work, MF membranes were used instead of UF membranes. Solutes deposited on the membrane surface quickly and reached the steady state in a short time after the filtration started. This result is in agreement with another study about the macromolecular adsorption during filtration (Ognier, et al., 2002).

All sludge fractions are influenced by drag force and back transport together. The permeation drag increased with particle size, and back transport consists of Brownian diffusion, inertial lift and shear-induced diffusion (Chen, et al., 1997). Brownian diffusion decreases with increasing particle size, while other forces increase with increasing particle size. As the large particles were the main fractions in activated sludge of MBR and SBR, and the influence of Brown diffusion is nearly negligible, it can be concluded that the back transport velocity of the particles increased with their size (Bae and Tak, 2005). Therefore, soluble matters have the smallest back transport velocity and are strongly adsorbed onto the membrane surface by permeation drag, not readily detached by shear force induced by cross-flow. It may attribute to the rapid deposition of the solutes.

On the other hand, suspended solids, which have the largest size among all sludge fractions, spend the longest time reaching the equilibrium between the permeation drag and back transport. In the initial stage of membrane filtration, quantities of suspended solids deposited on the membrane surface rapidly by permeation drag, however, they are easily removed by back transport as well. As a result, the resistance of suspended solids continuously increased. And colloids took longer time to reach steady state.

(a)

(b)

Figure 4.7 Filtration resistances of sludge fractions according to relative filtrate volume (a)MBR; (b)SBR
(MLSS=320mg/L, TMP=1bar, CFV=0.156m/s)

4.4 FESEM imaging

Observation of fouled membrane was also conducted using FESEM. Fig. 4.8 presents the FESEM images of fouled membranes at different operation stages.

From these images, a compact cake layer, consisted of all these sludge fraction: suspended solids, colloids and solutes, was clearly observed in Figs. 4.8 (a) and (b). The existence of biomass was demonstrated obviously in Fig. 4.8 (a). No other obvious differences between the cake layer of MBR and SBR can be detected barely using FESEM. Fig. 4.8 (c), a cross-sectional image of the fouled membrane by activated sludge from MBR, gives more detailed information about the cake layer. According to the scale at the top right corner, the thickness of cake layer was estimated to around 4-6 μm . However, accurate results could not be achieved due to the limitation of the equipment and the pretreatment of membrane samples. Therefore, the observation of membrane morphology could be a possible direction for the further study.

On the other hand, the physical cleaning seemed be able to remove the cake layer absolutely. Simultaneously, the pore size was reduced compared with the new membrane, demonstrating the internal fouling.

Figure 4.8 Images of membrane morphology (a) Fouled membrane (MBR, 8000x); (b) Fouled membrane (SBR, 8000x); (c) Fouled membrane (MBR, cross section, 4500x); (d) New membrane (4000x); (e) Fouled membrane after physical cleaning (MBR, 4000x); (f) Fouled membrane after physical cleaning (SBR, 4000x) (MLSS=320mg/L, TMP=1bar, CFV=0.438m/s)

4.5 Summary

The biological performance of the 20L MBR with SRT of 300 days, operated for over 2 years, was investigated. A 5L SBR with SRT of 10 days, representing the conventional activated sludge process, was set up as a comparison with the long SRT MBR. The results are shown as follows:

- 1) The overall removal efficiencies of COD and TOC for 300-day SRT MBR were 98.8% and 98.6% respectively. MLVSS/MLSS ratio indicated that no accumulation of inorganic compounds occurred in the 300-day SRT MBR.
- 2) Particles presented in MBR activated sludge and supernatant were relative smaller than that in SBR, while the molecular weight of organic molecules in MBR solutes was similar to that in SBR. These phenomena partially contributed to different filtration resistances of each fraction from MBR and SBR sludge.
- 3) Suspended solid was the dominant contributor to membrane fouling during the filtration of activated sludge from MBR and SBR system. Colloids and solutes also played important role during the filtration. Furthermore, the relative contribution of colloids in MBR was higher than that for SBR.
- 4) According to the results of more in-depth analysis of sub-divided resistance, the contributions of R_{cp} and R_c to the total resistances of activated sludge were notable. R_{cp} took up as high as 60% of R_t both in MBR and SBR, while the relative contribution of R_c was 30% in MBR and 36% in SBR. On the other hand, R_{if} and R_m were nearly negligible.
- 5) Solutes and colloids deposited quickly in the initial stage of filtration, causing rapid decline of permeate flux. It took the longest time for suspended solids to deposit on the membrane surface and achieve the equilibrium between the permeation drag and back transport as they had the largest size.
- 6) A compact cake layer was clearly observed from FESEM images. And there were no significant differences between the fouled membrane of MBR and SBR. Physical cleaning can remove the cake layer effectively, while the internal fouling was demonstrated by smaller pore size after cleaning.

- 7) Various parameters, e.g. TMP, would affect the behaviors of sludge fractions to membrane fouling during the filtration. Therefore, detailed study about the influence of some parameters on the contributions of sludge fractions would be discussed in the next section.

CHAPTER 5 MEMBRANE FOULING BY VARIOUS SLUDGE FRACTIONS UNDER DIFFERENT OPERATING CONDITIONS

Though many researchers had studied the roles and behaviors of sludge fractions during the membrane filtration, different results were reported. Those differences were due to the nature of the substrate, fractionation methods (one group used centrifugation to eliminate the suspended solids and flocculation to obtain the soluble fraction, while another group used sedimentation and filtration to separate all sludge fractions), membrane materials and biomass characteristics (Bouhabila, et al., 2001). Bae et al (2005) insisted that fouling contributions from various sludge fractions may change according to membrane properties, hydrodynamic conditions and the physiological properties of the biomass. As the characteristics and components of activated sludge in long SRT MBR changes with time, different fouling contributions of sludge fractions were expected to be observed under different conditions. Thus, effects of MLSS concentration, TMP and CFV on membrane fouling caused by different sludge fractions were studied.

5.1 Effects of MLSS concentration on membrane fouling by various sludge constituents

5.1.1 Flux decline during membrane filtration of activated sludge

The flux decline behaviors of activated sludge, i.e. diluted mixed liquor from 300-day MBR were studied to evaluate the effects of MLSS concentration on membrane fouling. The activated sludge with different MLSS concentration were prepared by dilution with deionized water. This provided the consistency of the activated sludge properties, such as particle size distribution, EPS, SMP and etc. Hence, MLSS concentration was the only factor that could affect the membrane filtration. As mentioned before, all filtration tests were performed for 4 hours to achieve the steady-state.

The results are shown in Fig. 5.1. All constituents of the activated sludge with different MLSS concentrations displayed similar trends. The flux decreased sharply in the initial stage, followed by gentle decline, before reaching a steady-state. However, it took different time to reach the steady-state under various MLSS concentration. As MLSS concentration increased, shorter time was needed for influent to achieve the steady-state. In other words, flux of influents with higher MLSS concentration decreased much faster, especially for the activated sludge which consisted of all fractions, i.e. most foulants. Furthermore, different sludge fractions, separated from the sludge with the same MLSS concentration, reached different flux values at the steady state while the initial flux in all conditions were around 900 L/m²h. On the other hand, the steady state flux values of the same fractions decreased with the rising MLSS concentration. This indicated that the resistances of all constituents grew obviously with the increase of MLSS concentration of the activated sludge. From all discussed above, it can be concluded that the feed characteristics, including constituents and solids concentration, would affect the performance of sludge fractions during the filtration but not the initial flux.

Figure 5.1 Flux declines during the filtration of various sludge fractions according to MLSS concentration (TMP=1bar, CFV= 0.438m/s)
 MBR: (a₁) MLSS=320mg/L, (a₂) MLSS=685mg/L,
 (a₃) MLSS=1285mg/L
 SBR: (b₁) MLSS=320mg/L, (b₂) MLSS=1250mg/L

5.1.2 Contributions of different sludge constituents

Filtration resistances of suspended solids, colloids and solutes for all activated sludge with different MLSS concentration were calculated using equations (3.1) to (3.4). Figs. 5.2 (a₁) (a₂) show the relationship between filtration resistances of sludge constituents and the MLSS concentration of activated sludge from MBR. R_{AS} fit well to the trend line with a high correlation coefficient of 0.9969. For individual sludge fractions, R_{ss} and R_{sol} both displayed a linear relationship with the MLSS concentration, presenting a correlation coefficient of 0.9660 and 0.9664, respectively. Unlike R_{ss} and R_{sol} , R_{col} increased firstly when the MLSS concentration increased from 320mg/L to 685mg/L, and then decreased slightly when the MLSS was over 1200mg/L, resulting in a low correlation coefficient of 0.5173 of the fit line. This phenomenon might because that during the separation of supernatant from activated sludge with 685mg/L MLSS, some large particles, which should be classified into suspended solids, floated on the top of centrifugal tube and were transferred to the feed tank as part of the supernatant. Therefore, the R_{ss} and R_{col} in the MLSS of 685mg/L departed from the fit.

As the slopes of the trend lines for R_{AS} and R_{ss} were 0.0022 and 0.0013 respectively, the relative contribution of suspended solids changed a little with the increase of MLSS concentration, from 69.8% to 60.4%, as shown in Fig. 5.2 (a₂). It revealed that the suspended solids remained the predominant fraction in membrane fouling when MLSS concentration varied between 320mg/L and 1285mg/L. The higher MLSS concentration, the lower contribution of the suspended solids to membrane fouling was. Simultaneously, the significance of solutes increased according to the increasing MLSS concentration while the relative contribution of colloids first increased and then decreased slightly.

Figure 5.2 Resistances of different sludge fractions according to MLSS concentration (a₁) (a₂) MBR, (b₁) (b₂) SBR (TMP=1bar, CFV= 0.438m/s)

Results for SBR with SRT of 10 days are shown in Figs. 5.2 (b₁) (b₂). For individual fractions, the similar patterns of resistances were observed. As MLSS concentration increased, the resistance of suspended solids increased rapidly while resistance of colloids and solutes increased gradually. Nevertheless, the relative contribution of suspended solids remained constant around 85.17%-89.38%, higher than that in MBR activated sludge. Moreover, the secondary dominant fraction shifted from colloids to solutes. It can be easily explained by more colloids and solutes produced under long SRT condition (Orhon and Artan, 1994) and their complete retention by the membrane. In any case, higher MLSS concentration leads

to higher fouling resistances. Based on the results shown below, the MLSS concentration did not influence the relative contribution of sludge fractions significantly. This might be due to the small range of MLSS concentration used in this study. The fouling behaviors of sludge fractions in higher MLSS concentration should be investigated in the next period.

5.1.3 Filtration resistances of sludge constituents

To have a better understanding about the contributions of different sludge fractions, detailed investigation using resistance-in-series model is needed. Based on the equations (3.5) to (3.9), R_m , R_{cp} , R_c and R_{if} of different sludge fractions were calculated as shown in Fig. 5.3 and Fig. 5.4.

Fig. 5.3 (a₁) presents the hydraulic resistances of the activated sludge from MBR, which consists of all the fractions. It was revealed that the dominant resistance depended on MLSS concentration. As the MLSS concentration increased, the cake layer resistance rose as well while the concentration polarization resistance dropped. As a result, the dominant resistance shifted from R_{cp} to R_c with the increase of MLSS concentration. The growth of R_c can be attributed to more particles depositing on the membrane surface. However, R_{cp} , which was also supposed to increase with the MLSS concentration according to the film theory, decreased significantly. This may be due to the decline of viscosity of the activated sludge when we diluted the mixed liquor from the 300-day SRT MBR into different MLSS concentration. The MLSS concentration, above which R_c was more dominant than R_{cp} , was between 400 to 600 mg/L. On the other hand, the contribution of internal fouling was insignificant compared with the contribution of cake layer and concentration polarization for the activated sludge. Therefore, it seemed that R_{if} was negligible. As discussed above, the suspended solids played the significant role in membrane fouling, thus the resistance of cake layer, which mainly caused by large particles, also dominated the filtration resistance. The relative contribution of cake resistance varied from 30.9% to 73.2% while the relative contribution of concentration polarization decreased from 63.0% to 22.9% when MLSS concentration increased from 320 to 1,285 mg/L. The R_m and R_{if} almost stayed stable, indicating that internal fouling barely related with the MLSS concentration.

It could be concluded that influence of MLSS concentration on cake fouling is more remarkable.

The results of resistances of supernatant and solutes from MBR were also displayed in Figs. 5.3 (b₁), (b₂), (c₁) and (c₂). Similarly, R_c increased with the increasing MLSS concentration of the sludge that the supernatant and solutes separated from, while R_m and R_{if} remained constant values. Nevertheless, an increase of the R_{cp} for supernatant was observed followed by a steep decrease. A positive relationship between R_{cp} and MLSS concentration was also presented.

On the other hand, different phenomena of fouling were observed for SBR as shown in Fig. 5.4. With more foulants, a rise in MLSS from 310mg/L to 1250mg/L increased R_c and R_{cp} of activated sludge, which contributed 60% and 36% to the total resistance, respectively. Considering the mean floc size of activated sludge from MBR (54 μ m) and SBR (125 μ m), as shown in Fig. 4.1, floc size difference would be one of the factors for the higher R_c of the activated sludge from SBR than that of sludge from MBR in low MLSS concentration (320mg/L). It can be easily explained by Carman-Kozeny equation. On the other hand, small particles existed in MBR activated sludge were tend to form more compressible cake layer than that of SBR sludge. Therefore, more foulants were retented, resulting in higher R_c . Furthermore, other factors like EPS concentration, the microorganism morphology in the cake layer may also contributed to the different cake resistances. Hence, when the MLSS concentration reached 1285mg/L, the activated sludge from MBR presented higher R_c than the sludge from MBR.

In addition, the contributions of R_c were higher than R_{cp} both in colloids and solutes of SBR, taking up 60%-65% of the total resistances, while the relative contribution of R_{cp} dropped to 15% and 10% in colloids and solutes, respectively. Despite these differences, the increase of MLSS does not seem to vary the relative contributions of resistances. In other words, the relative contribution of various resistances in different sludge fractions stayed almost constant along with the growth of MLSS although the resistance values increased.

Figure 5.3 Filtration resistances in (a₁)(a₂) Activated sludge; (b₁)(b₂) Supernatant; (c₁)(c₂) Solutes from 300-day SRT MBR according to MLSS concentration (TMP=1bar, CFV=0.438m/s)

Figure 5.4 Filtration resistances in (a1)(a2) Activated sludge; (b1)(b2) Supernatant; (c1)(c2) Solutes from 10-day SRT SBR according to MLSS concentration (TMP=1bar, CFV=0.438m/s)

5.2 Effects of trans-membrane pressure on membrane fouling by various sludge constituents

5.2.1 Flux declines during membrane filtration of activated sludge

Fig. 5.5 displays the flux declines during membrane filtration of activated sludge from MBR with SRT of 300 days and SBR with SRT of 10 days. In spite of the similar pattern of flux decline, the initial flux values increase significantly along with the growing TMP. When TMP was 0.5bar, the initial flux was around 600 L/m²h; when TMP increased to 2 bar, the initial flux also increased to 1500 L/m²h, more than double. This phenomenon is induced from the higher driving force caused by higher TMP. According to Darcy's law, a rise in TMP leads to an increase in filtrate flux at the beginning of the experiment. In addition, with a pure solvent the increase in filtrate flux is directly proportional to the pressure. This was easily proved by the filtrate flux values of solutes. On the other hand, with a complex solution the effect of TMP is more difficult to predict. For activated sludge both from MBR and SBR, the flux value at the steady-state decreased significantly according to the increasing TMP. On contrary, some researchers (Bhave and Fleming, 1988; Lahiere and Goodboy, 1993) found that the filtrate flux increases with pressure until the stabilization reach a maximum. Afterwards, the filtrate flux remained constant or decreased slightly. This inconsistent phenomenon represents the negative effect of pressure dominant in this study.

Moreover, the rate of flux decline was faster at high TMP, i.e. it took shorter time to reach the steady-state of membrane fouling. On the other hand, in each group of feed water, the flux of activated sludge with the most foulants decreased the most rapidly, followed by the supernatant, which contains colloids and solutes both, and the solutes.

Figure 5.5 Flux declines during the filtration of various sludge fractions according to trans-membrane pressure
 MBR: (a₁) TMP=0.5bar, (a₂) TMP=1bar, (a₃) TMP=1.5bar, (a₄) TMP=2bar
 SBR: (b₁) TMP=1bar, (b₂) TMP=2bar
 (MLSS=320mg/L, CFV= 0.156m/s)

5.2.2 Contributions of different sludge constituents

Fig. 5.6 demonstrates contributions of different sludge fractions, i.e. suspended solids, colloids and solutes, to the membrane fouling. As shown in Fig. 5.6 (a₁), resistances of all sludge fractions were proportional to the TMPs.

TMP is the main driving force in the cross-flow filtration of Darcy's law. Accordingly, the increase in the initial filtrate flux is proportional to the TMP as observed in 5.2.1. However, this improvement of mass transfer gives a more significant foulant flow to the membrane. Some solutes readily penetrate the porous medium either causing fouling or passing as filtrate. Likewise, enhancement of the convective flow of particles towards the membrane, resulting from an increase in TMP, leads to the polarization and deposition of particles. As a result, R_{AS} of MBR increases linearly according to the increasing TMP from 0.5 bar to 2 bar, with a high correlation coefficient of 0.9954. Such high correlation was also observed for suspended solids and colloids: 0.9952 and 0.9945 respectively. Whereas, the fitting line of R_{sol} with increasing TMP had a lower correlation coefficient of 0.7476.

Moreover, the slopes of the lines in Fig. 5.6 (a₁) are 10.09, 2.05 and 0.12 for R_{ss} , R_{col} and R_{sol} , respectively. This implies that although the resistances of all sludge fractions were proportional to the TMP applied, the degree of this proportion varied among the sludge fractions. Meanwhile, the influence of TMP on membrane fouling increased with the particle size of foulants at the same CFV. Therefore, regardless of TMP, the contribution of R_{ss} to the total resistance was higher than that of any other fraction as shown in Fig. 5.6 (a₂). This because suspended solids was the main fraction of the activated sludge and played a key role in the formation of the cake layer, with the relative contribution to the total resistance varied from 65.92% to 80.01%. Simultaneously, the relative contribution of colloids and solutes ranged from 11.53 % to 18.48% and 22.55% to 3.7%, respectively.

Figure 5.6 Resistances of different sludge fractions according to trans-membrane pressure (a₁) (a₂) MBR, (b₁) (b₂) SBR (MLSS=320mg/L, CFV= 0.156m/s)

For SBR, the predominant resistance was suspended solids as well, with the relative contribution remain 90%. Colloids and solutes took up 5% of the total resistance respectively. Hence, confirming that the activated sludge from 300-day SRT MBR contains more colloids than that from 10-day SRT SBR.

5.2.3 Filtration resistances of sludge fractions

Figs. 5.7 and 5.8 represent the detailed analysis of sub-divided resistances. Theoretically, R_c and R_{cp} have positive relationship with TMP. This is due to an increase of TMP, which could enhance the convective flow of foulants towards the

membrane surface, subsequently enhancing the polarization and deposition of particles. In activated sludge from MBR (Fig. 5.7 (a₁)), both R_c and R_{cp} grew significantly from $0.88 \times 10^{12} \text{ m}^{-1}$ to $11.41 \times 10^{12} \text{ m}^{-1}$ and $1.87 \times 10^{12} \text{ m}^{-1}$ to $9.89 \times 10^{12} \text{ m}^{-1}$ when TMP increased from 0.5 to 2 bar. Moreover, R_c increased more rapidly than R_{cp} , thus the dominant contributor shifted from R_{cp} to R_c when the TMP increased to 2 bar. On the other hand, more fine particles may be pressured to travel through the porous medium and plugging the pores by bridging or transporting into pores, and thus create internal pore restrictions to fluid flow. These reasons may cause the slight rise of R_{if} from $0.029 \times 10^{12} \text{ m}^{-1}$ to $0.21 \times 10^{12} \text{ m}^{-1}$. In addition, R_m remained constant values around $0.20 \times 10^{12} \text{ m}^{-1}$.

As shown in Fig. 5.7 (a₂), R_{cp} (25.78% to 44.72%) and R_c (52.87% to 71.03%) were the main components of the fouling resistances at low operating pressure range (0.5 to 1.5 bar), while R_{cp} (52.41%) became the dominant fouling resistance and R_c (45.41%) became the secondary dominant resistance when the TMP increased to 2 bar. Compared to R_{cp} and R_c , R_{if} and R_m , which consisted 0.71-0.98%, 1.22- 6.47% of the total resistances, were nearly neglectable. For activated sludge from SBR (Figs. 5.7 (b₁) and (b₂)), the similar fouling phenomena were observed except that R_{cp} remained the dominant resistance all along the TMP.

For supernatant and solutes, R_{cp} and R_c increased as well, and their values became the same order of R_m and R_{if} . Instead of R_{cp} , R_c played the key role during the membrane fouling.

Figure 5.7 Filtration resistances in (a₁)(a₂) Activated sludge; (b₁)(b₂) Supernatant; (c₁)(c₂) Solutes from 300-day SRT MBR according to trans-membrane pressure (MLSS=320mg/L, CFV= 0.156m/s)

Figure 5.8 Filtration resistances in (a₁)(a₂) Activated sludge; (b₁)(b₂) Supernatant; (c₁)(c₂) Solutes from 10-day SRT SBR according to trans-membrane pressure (MLSS=320mg/L, CFV= 0.156m/s)

5.3 Effects of cross-flow velocity on membrane fouling by various sludge constituents

5.3.1 Flux declines during membrane filtration of activated sludge

The flux decline behaviors of influents with different constituents are shown in Fig. 5.9. Theoretically, the CFV does not influence performance and selectivity of the membrane when pure solvent is filtered as it does not appear in Darcy's law. Therefore, the initial filtrate flux values of all influents were around 950L/m²h. It was much higher than the applied flux in long SRT MBR as the cross-flow testing is tend to simulate the side stream MBR system. However, CFV plays a significant role during the filtration of influents containing complex constituents. The rate of flux decline was slow at high CFV, i.e. it took longer time to reach the steady-state of membrane fouling. Furthermore, the CFV used in this study is close to the superficial liquid velocity (0.25–0.55 m/s) created by bubbles in the research conducted by Le-Clech et al. (2005).

Figure 5.9 Flux declines during the filtration of various sludge fractions according to cross-flow velocity
 MBR: (a₁) CFV=0.156m/s, (a₂) CFV=0.279m/s, (a₃) CFV=0.438m/s
 SBR: (b₁) CFV=0.156m/s, (b₂) CFV=0.438m/s
 (MLSS=320mg/L, TMP=1bar)

5.3.2 Contributions of different sludge constituents

The values of various resistances of sludge fractions from 300-day SRT MBR and 10-day SRT SBR as a function of CFV are demonstrated in Fig. 5.10.

CFV is a very important operating parameter for the filtration of complex solutions. Unlike MLSS concentration and TMP, CFV has a negative effect on membrane fouling, i.e. higher CFV could prevent membrane fouling effectively. It appears clearly from Fig. 5.10 that all fraction resistances decreased with the increasing CFV. This is coincide with the result observed by Choi et al. (2005) that the permeate flux increased linearly with increasing CFV. Once again, the different slopes of these trend lines revealed that the CFV influenced fractions in various degrees. Suspended solids were most influenced by CFV, i.e. R_{ss} dropped the most steeply. Furthermore, the decrease of R_{AS} was mainly caused by the variation of R_{ss} . Colloids and solutes linearly decreased with relatively low correlation coefficient of 0.6645 and 0.4364 respectively. This low correlation implies fluctuation of resistances analysis.

According to the study conducted by Bae et al. (2005), all sludge fractions during the cross-flow filtration are influenced by drag force and the back transport. The drag force increased with operation flux, trans-membrane pressure and particle size. For back transport velocity, it is complex to predict its effect. It is consist of Brownian diffusion, inertial lift and shear-induced diffusion (Belfort, et al., 1994). All forces except for Brownian diffusion, are generated by cross-flow and tend to increase with CFV and particle size, while Brownian diffusion had an inverse relationship with particle size. Nevertheless, the influence of Brownian diffusion is extremely small because the activated sludge contains huge amount of large flocs and small amount of solutes. Hence, the back transport velocity increases with particle size. Therefore, the fraction of the largest size, i.e. suspended solids, were most influenced by CFV. As a result, fouling control for suspended solids appears to be simpler than for other fractions.

From results shown in Fig. 5.10 (a₂), it was clearly that suspended solids remained the main contributor to membrane fouling though the values of R_{ss} changed significantly. And the relative contribution of R_{ss} changed between 69.77% and 75.06%. Simultaneously, the relative contributions of colloids and solutes changed in the range of 11.37 - 19.57% and 6.46 - 10.66%, respectively.

Figure 5.10 Resistances of different sludge fractions according to cross-flow velocity
(a₁) (a₂) MBR, (b₁) (b₂) SBR
(MLSS=320mg/L, TMP=1bar)

Likewise, the same fouling phenomena were observed in the filtration of sludge fractions from SBR. R_{AS} and R_{ss} both decreased significantly with the increasing CFV, while R_{col} and R_{sol} were not affected by CFV obviously. Furthermore, their

relative contributions stayed around 86%, 5% and 9% respectively. It seems that the variation of CFV between 0.156-0.432m/s does not change the relative contributions of sludge fractions though the values of resistances decreased significantly according to CFV.

5.3.3 Filtration resistances of sludge fractions

Analysis of resistances consisted of the total resistance was presented in Fig. 5.11 and Fig. 5.12. For activated sludge from MBR, R_c dropped significantly and R_{cp} slightly increased, while R_m and R_{if} remained negligible.

Generally, higher CFV induces higher shearing stress, reducing the thickness of cake layer and diffusion layer. This can explain the decrease of R_c . However, the polarization resistance, which depends on the diffusion layer thickness according to film's theory, should reduce as well. However, the increase of CFV also has the negative influence on the filtration. Increasing CFV will break long-chain macromolecules and colloids into finer ones and cause a rise in the concentration gradient between the membrane surface and the bulk side, leading to higher Brownian diffusion, which may result in higher R_{cp} . During the low CFV range (0.156-0.438m/s) compared with other studies (Zhao, et al., 2003; Choi, et al., 2005), the negative influence of CFV was dominant, leading to increasing R_{cp} . Moreover, the cross point of CFV, above which R_{cp} was more dominant than R_c , was about 3m/s. For activated sludge from SBR, the trends of R_{cp} and R_c were reversed. R_{cp} contributed more to the total resistance than R_c . R_{cp} reduced as the result of decreasing thickness of diffusion layer. The difference may attributed to the different EPS concentration and the morphology of microorganism existing in MBR and SBR system. It had been reported that the EPS can determine the pore size and porosity of the biolayer (Zhang and Bishop, 1994). In long SRT conditions, large amount of EPS were produced. However, when samples were diluted to the same MLSS concentration, the EPS concentration was also changed and differed from each other, leading to different fouling behaviours. Furthermore, EPS and filamentous index could influence the cake resistance by changing the relative hydrophobicity and zeta potential measured in the biomass floc directly (Le-Clech, et al., 2006). The growth of filamentous bacteria may lead to higher EPS

concentration, lower zeta potential, more irregular floc shape and higher hydrophobicity (Meng, et al., 2006). Further studies on the cake layer structure and the biofilm need to be conducted.

Trends of R_{cp} and R_c for supernatant from MBR were resemble to that for activated sludge from MBR. Whereas the R_{cp} and R_c for supernatant from SBR were changeless. It seems that supernatant from MBR contains more fine particles and colloids compared with SBR, thus more affected by CFV. Likely, solutes both from MBR and SBR were slightly influenced by CFV.

Figure 5.11 Filtration resistances in (a1)(a2) Activated sludge; (b1)(b2) Supernatant; (c1)(c2) Solutes from 300-day SRT MBR according to cross-flow velocity (MLSS=320mg/L, TMP=1bar)

Figure 5.12 Filtration resistances in (a₁)(a₂) Activated sludge; (b₁)(b₂) Supernatant; (c₁)(c₂) Solutes from 10-day SRT SBR according to CFV (MLSS=320mg/L, TMP=1bar)

5.4 Summary

Effects of MLSS concentration, TMP and CFV on membrane fouling by various sludge fractions were discussed in this chapter. The flux decline behaviors, contributions of various sludge fractions and the filtration resistances were studied. The results of sludge fractions from 300-day SRT MBR were compared with that of sludge fractions from 10-day SRT SBR. The main conclusions are shown as follows:

- 1) Various sludge fractions under different operation conditions displayed the similar flux decline behaviors. The flux decreased sharply in the initial stage, and then followed by gentle decline, finally reaching the steady-state.
- 2) MLSS concentration and TMP had a positive relationship with resistances of sludge fractions while CFV were negatively related with resistances of sludge fractions. Simultaneously, the insignificant influence of MLSS concentration on the relative contribution of sludge fractions suggested further study about the fouling behaviors in higher MLSS concentration. Moreover, different slopes of fitting lines for sludge fractions presented that they were affected differently by these parameters.
- 3) Among sludge fractions, suspended solids played the key role during the filtration both in MBR and SBR activated sludge. On the other hand, the secondary predominant contributor shifted from colloids to solutes from MBR with SRT of 300 days to SBR with SRT of 10 days because of less colloids in SBR activated sludge.
- 4) R_{cp} and R_c were dominant contributors to membrane fouling while R_m and R_{if} were almost negligible. The relative contributions depended on the operation conditions applied, i.e. MLSS concentration, TMP and CFV.

CHAPTER 6 CONCLUSION AND RECOMMENDATION

In this thesis, performances of MBR with SRT of 300 days and the effects of some parameters on membrane fouling are stated, compared with that of SBR with SRT of 10 days, which were set up to represent the conventional activated sludge.

The contributions of sludge fractions to membrane fouling were studied, indicating that the suspended solid was the main fouling fraction both in MBR and SBR. On the other hand, the importance of colloids and solutes could not be neglected. Moreover, sub-divided resistances were analysed to have a deep understanding about the fouling mechanism. It was demonstrated that the resistances of concentration polarization and cake layer mainly contributed to the total resistance, whereas the internal fouling resistance and membrane resistance were negligible during the MF filtration of activated sludge. Furthermore, the membrane fouling increases with the increasing MLSS concentration, rising TMP and reducing CFV respectively. The main contributor of resistance changed according to various MLSS concentration, TMP and CFV.

However, there are a number of important areas which require additional experimental and theoretical investigation.

- 1) Characteristics of the cake layer formed during the membrane filtration are still unclear. As mentioned in chapter 4, detailed information of the cake layer, such as the mass, biomass composition and so on, were not able to be acquire from FESEM. Thus, one of the promising areas is employing microscopic techniques, e.g. confocal laser scanning microscopic (CLSM) and fluorescently labeled probes, to explore the structure and composition of the biofoulant.
- 2) The influences of operating parameters on membrane fouling were studied in small ranges. For example, CFV changed during 0.156-0.438m/s, relatively small compared with other researches. Therefore, further studies

could focus on the influences of operating parameters with high values to have a better understanding.

- 3) Interactions between sludge fractions were ignored in this study, while they do exist during the membrane filtration. Hence, another possible direction for further studies may be exploring the effects of these interactions on membrane fouling caused by different sludge fractions.

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