The effect of powdered activated carbon and two-compartment configuration on membrane bioreactor performance

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The Effect of Powdered Activated Carbon and Two-Compartment Configuration On Membrane Bioreactor Performance

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in fulfilment of the requirement for the degree of
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COMPARISON OF MEMBRANE BIOREACTOR DESIGN CONCEPTS

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ABSTRACT

The objective of this study was to investigate alternative membrane bioreactor (MBR) designs in an attempt to reduce membrane fouling in the MBR system. Several approaches were investigated including (i) modifying the mixed liquor suspended solids (MLSS) using powdered activated carbon (PAC) to form biologically activated carbon (BAC), (ii) limiting the biomass formation from contacting with the membranes using a two-compartment reactor design and (iii) using relatively large polymer beads ($\approx 600 \mu\text{m}$) to scour the membrane surface to prevent pollutant attachment and to detach fine foulants on the membrane surface. The effect of PAC addition up to 5 g/L to the activated sludge (AS) in the MBR has been investigated in detail. The benefit of PAC addition to the MBR (AS) was confirmed in a series of tests operated at sludge retention times (SRTs) of 10 and 30 days which involved the addition of 10% and 3.3% by PAC weight addition per day respectively to compensate for sludge wastage. Better performance in terms of fouling mitigation was observed for MBR (BAC) at SRT 10 days compared to 30 days. At these SRTs the MBR (BAC) experienced less fouling than the MBR (AS). However when the MBR (BAC) were operated without wastage, its performance was worse than the MBR (AS). Thus the improved performance achieved by the MBR (BAC) requires regular replenishment of aged BAC with fresh PAC.

Several membrane fouling mitigation mechanisms involved in the MBR (BAC) were identified. Firstly the PAC can support biomass and adsorb planktonic cells to gradually form BAC. Secondly BAC is equipped with simultaneous adsorption and biodegradation properties. The organics adsorbed by the PAC can be decomposed by the attached bacteria at the PAC surface into lower molecular weight (MWt) species. This helps to partially bioregenerate the saturated BAC and the lower MWt dissolved organics would be more easily adsorbed and biodegraded by fresh and aged BAC. Thirdly, the particulate PAC could scour the surface by collision and limit fouling deposits. Finally, the presence of PAC in the biomass reduces the specific cake resistance (SCR) of deposited layers of MLSS.

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It was found that a relatively small PAC size ($14.88 \pm 3.93 \mu\text{m}$) could eliminate fine foulants better than a larger PAC size ($67.60 \pm 2.70 \mu\text{m}$) at modest fluxes ($10\text{--}25 \text{ L/m}^2/\text{hr}$). However the smaller PAC size had a tendency to deposit on the membrane surface at relatively high fluxes and this caused worse fouling control than for the larger PAC. Therefore a relatively low flux would be recommended for the MBR (BAC) using smaller PAC size in order to optimize performance in fouling prevention.

A two-compartment bioreactor was operated with two 20 L tanks where Tank 2 was equipped with a submerged membrane module. The concept was to have the submerged membrane located in a low MLSS concentration aeration chamber downstream from the high MLSS concentration aeration bioreactor. This was achieved by arranging flow distribution from and back to the high MLSS aeration bioreactor to minimize solids carry-over to the membrane zone of the second chamber. However, it was observed that the membrane located in the supernatant was more rapidly fouled than the membrane in the mixed liquor. It was found that the fine foulants (i.e. TOC and polysaccharides) in the supernatant of Tank 2 were higher than that in Tank 1 (which had MLSS). The absence of biofloc in tank 2 allowed an accumulation of supernatant components that were not biodegraded and not passed by the membrane. Thus, membrane filtration in a separate supernatant tank is not beneficial, unless a substantial recycle stream can be applied.

As noted above, the MBR (BAC) only performed better in membrane fouling control provided there was a steady renewal of used PAC. The regeneration of used PAC is complicated and potentially costly. As one of the proposed fouling control mechanisms of PAC is scouring by particulate collision, test were done using the addition of large polymer beads ($\approx 600 \mu\text{m}$) for scouring and to address the disadvantage of the MBR (BAC) in terms of regeneration of used PAC. The beads in the wastage sludge could be easily separated and reused. It was shown that the polymer beads helped to suppress the fouling rate of the MBR (AS) through its collision efficiency. However, overall the MBR (BAC) performed better than the MBR (polymer bead) in membrane fouling control, presumably due to the

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simultaneous adsorption and biodegradation effect of the BAC. Further work is recommended to investigate reusable alternatives to PAC.

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LIST OF SYMBOLS AND ABBREVIATIONS

A	Membrane area (m^2)
Adsorption	
isotherm	Adsorption equilibrium test
a_p	Particle size (μm)
AS	Activated sludge
BAC	Biologically activated carbon sludge
BET	Brunauer, Emmett, and Teller
BGAC	Biological granular activated carbon sludge
BOD	Biological oxygen demand (ppm)
BSA	Bovine serum albumin
C_B	Bulk concentration (mg/L)
C_b	The concentration of the SS of the feedwater (mg/L)
c/n	Continuous
c/p	Compartment
C_{ss}	Concentration of solutes (mg/L)
C_w	Wall concentration (mg/L)
Conc.	Concentration (mg/L)
COD	Chemical oxygen demand (mg/L)
D_{50}	Mean size of particle (μm)
Da	Dalton
DO	Dissolved oxygen (ppm)
DOTM	Direct observation through the membrane
d_p	Particle diameter (μm)
EDTA	Ethylenediaminetetraacetic acid
EPS	Extracellular polymeric substances
F/M	Food to microorganism ratio
GAC	Granular activated carbon
GB	Glass bead
HRT	Hydraulic retention time: volume of bioreactor/permeate flowrate (hr/day)
HPSEC	High performance size exclusion chromatography
IC	Inorganic carbon

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i/t	Intermittent
J	Flux ($\text{L}/\text{m}^2/\text{hr}$)
L	Litre or membrane length
Lumen	Inner side of fiber
M	Mass of the filter cake (g)
M	Effective particle diameter (μm)
MBR	Membrane bioreactor
MLSS	Mixed liquor suspended solids (g/L)
MLVSS	Mixed liquor volatile suspended solids (g/L)
MF	Microfiltration
MWCO	Molecular weight cut off
MWt	Molecular weight
N/A	Not applicable
NISS	Non-ionogenic surfactants
nm	Nanometer
OLR	Organic loading rate (g/day)
PAC	Powdered activated carbon
PE	Polyethylene
P/i	Pseudo infinity
p/y or P/y	polymer
PVDF	Polyvinylidene
P_h	Hydraulic permeability
R_c	Total cake resistance (m^{-1})
R_{if}	Irreversible fouling (m^{-1})
R_f	Fouling resistance (m^{-1})
R_{rf}	Reversible fouling (m^{-1})
R_m	Intrinsic membrane resistance (m^{-1})
Rpm	Round per minute
R_t	Total resistance (m^{-1})
s/b	Submerged
s/d	Sidestream
SBR	Sequencing batch reactor
SCR	Specific cake resistance (m/kg)
SEM	Scanning electron microscope
SMPs	Soluble microbial products

LIST OF SYMBOLS AND ABBREVIATIONS

A	Membrane area (m ²)
Adsorption	
isotherm	Adsorption equilibrium test
a _p	Particle size (μm)
AS	Activated sludge
BAC	Biologically activated carbon sludge
BET	Brunauer, Emmett, and Teller
BGAC	Biological granular activated carbon sludge
BOD	Biological oxygen demand (ppm)
BSA	Bovine serum albumin
C _B	Bulk concentration (mg/L)
C _b	The concentration of the SS of the feedwater (mg/L)
c/n	Continuous
c/p	Compartment
C _{ss}	Concentration of solutes (mg/L)
C _w	Wall concentration (mg/L)
Conc.	Concentration (mg/L)
COD	Chemical oxygen demand (mg/L)
D ₅₀	Mean size of particle (μm)
Da	Dalton
DO	Dissolved oxygen (ppm)
DOTM	Direct observation through the membrane
d _p	Particle diameter (μm)
EDTA	Ethylenediaminetetraacetic acid
EPS	Extracellular polymeric substances
F/M	Food to microorganism ratio
GAC	Granular activated carbon
GB	Glass bead
HRT	Hydraulic retention time: volume of bioreactor/permeate flowrate (hr/day)
HPSEC	High performance size exclusion chromatography
IC	Inorganic carbon

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i/t	Intermittent
J	Flux ($\text{L}/\text{m}^2/\text{hr}$)
L	Litre or membrane length
Lumen	Inner side of fiber
M	Mass of the filter cake (g)
M	Effective particle diameter (μm)
MBR	Membrane bioreactor
MLSS	Mixed liquor suspended solids (g/L)
MLVSS	Mixed liquor volatile suspended solids (g/L)
MF	Microfiltration
MWCO	Molecular weight cut off
MWt	Molecular weight
N/A	Not applicable
NISS	Non-ionic surfactants
nm	Nanometer
OLR	Organic loading rate (g/day)
PAC	Powdered activated carbon
PE	Polyethylene
P/i	Pseudo infinity
p/y or P/y	polymer
PVDF	Polyvinylidene
P_h	Hydraulic permeability
R_c	Total cake resistance (m^{-1})
R_{if}	Irreversible fouling (m^{-1})
R_f	Fouling resistance (m^{-1})
R_{rf}	Reversible fouling (m^{-1})
R_m	Intrinsic membrane resistance (m^{-1})
Rpm	Round per minute
R_t	Total resistance (m^{-1})
s/b	Submerged
s/d	Sidestream
SBR	Sequencing batch reactor
SCR	Specific cake resistance (m/kg)
SEM	Scanning electron microscope
SMPs	Soluble microbial products

COMPARISON OF MEMBRANE BIOREACTOR DESIGN CONCEPTS

S/N	Supernatant
SRT	Sludge retention time: volume of bioreactor/sludge (MLSS) wastage per day (days)
SS	Suspended solids
s/s	Suspension
t	Time
TC	Total carbon (mg/L)
TMP	Transmembrane pressure (bar)
TOC	Total organic carbon (mg/L)
UF	Ultrafiltration
UF-BAC	Ultrafiltration-biological activated carbon
UV	Ultraviolet
V	Volume
V _s	Sample volume
VCR	Volume concentration ratio
vs	Versus
w/w	Wastewater
Y	Weight of biomass in the sample
μm	Micrometer
α	Specific cake resistance (m/kg)
ρ	Particle density (kg/m ³)
ε	Porosity of membrane
μ	Viscosity (mPa.s)
ΔP	Transmembrane pressure (bar)
γ	Shear rate (sec ⁻¹)

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1.1 Background

Membrane bioreactors (MBRs) have received increasing attention in recent years for their advantages in wastewater treatment. Over the years, numerous studies have been carried out to evaluate both submerged and sidestream configurations of the MBR and their overall performance. In the last few years, the MBR has become a promising alternative to the well known conventional activated sludge process in wastewater treatment (Huang, *et al.* 2001; Ben Aim *et al.*, 2001; Melin *et al.*, 2006). It can be distinctively characterized by complete retention of microorganisms within the bioreactor achieved by membrane separation. The submerged MBR integrates membrane modules (ultra or microfiltration membranes) into the biological reactor to separate biomass and extract clean water.

The MBR system has been developed to avoid the need for secondary clarification and to achieve a total retention of suspended matter without prior biomass flocculation, which limits the nutrient transfer and therefore reduces the biokinetics (Wisniewski and Grasmick, 1998). The overall treatment plant size is greatly reduced due to the absence of large-sized sedimentation tanks and the reduction in bioreactor size. In addition, smaller reactor volumes can be realized as this process allows higher mixed liquor suspended solids (MLSS) concentrations (up to 30 g/L, or more typically 10 to 15 g/L) to be developed in the bioreactor without detrimental effects unlike the conventional activated sludge process. Additionally, the high level of MLSS can effectively achieve nitrification and denitrification without the need for extended aeration (Davies *et al.*, 1998). Another advantage of the use of membrane filtration is the relative indifference to hydraulic fluctuations (Rosenberger *et al.*, 2002). Since the membrane filtration does not rely on the settling of suspended solids (SS) as in a conventional activated sludge process, much higher solids retention times (SRTs) can be maintained. This leads to reduced excess sludge production and an increase in the organic removal efficiency.

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The main disadvantages of the submerged MBR system are that (i) the membranes have direct contact with concentrated biomass and (ii) it is difficult to provide effective hydrodynamic control of fouling. These disadvantages can cause sludge cake deposition, including biofouling, on the membrane surface in the bioreactor. In order to avoid serious fouling problems, submerged MBRs are normally only operated at fluxes less than about 20 L/m²/hr.

One of the limiting factors of the ‘conventional’ MBR is that the concentrated biomass results in high viscosity liquor in the bioreactor which limits the function of the air bubbles from the aerator at the bottom of the membrane unit for the removal of the cake layer on the membrane surface and constrains the movement of the membrane fibres. One way to tackle this problem faced by the submerged MBR is to combine the features of a submerged and a sidestream MBR system. In this approach a two-compartment MBR system can be set up in which the first compartment contains high MLSS and the second compartment, has a low MLSS content and houses the membrane module. The challenge is to devise a method for retaining the major biomass in the first tank (bioreactor) and allowing the supernatant to flow to the next membrane tank. To achieve this, one approach is to manipulate the flow to and from the second tank so that major foulants can be retained in the first tank.

The other approach is to modify the mixed liquor by an additive. Same success has been reported for a hybrid MBR with added PAC to form Biological Activated Carbon (BAC) sludge (Liu *et al.*, 2005; Fang *et al.*, 2006). The role of the PAC in improving fouling control is of interest and has been studied in this thesis. For example the mechanisms include the BAC combining the biomass and PAC into floc particles so that the specific cake resistance (SCR) of BAC can be lower than for conventional AS (Kim *et al.*, 1998). In addition, the adsorptive property of BAC could help (i) to reduce fine colloids deposited on the membrane surface through adsorption and (ii) to decompose or breakdown the adsorbed high molecular weight organics (100k-1000k daltons) into a lower range by the species attached on the PAC surface. The lower range molecular weight organics (<100k daltons) would be

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more biodegradable and adsorbed by the PAC and their tendency to block and plug the membrane micropores would be decreased. In addition, the PAC may have a 'collision' effect able to detach the fine particles on the membrane surface and enhance transport back to the bulk liquid. These three mechanisms (adsorption, simultaneous adsorption and biodegradation and 'collision' effects) have been evaluated and compared in this thesis.

In this study, the membrane modules used were an in-house fabricated hollow fibre U-shaped modules ($\approx 7\text{-}10$ cm in diameter) housed in the small floor riser area of a cylindrical shaped ($\approx 8\text{-}16$ cm in diameter) bioreactors of one compartment or two-compartment MBRs to enhance the surface velocity and scouring effect of the air. The two-compartment MBR was also run in a parallel arrangement to compare the performance of a 'conventional' MBR and a hybrid MBR with added PAC in terms of membrane fouling control.

1.2 Aims and objectives of the study

Potential limitation in the conventional MBR is that MLSS at high concentration is in contact with the membranes and tends to foul them because it is comprised of bacterial floc which includes extracellular polymeric substances (EPSs), TOC and fine colloids. This could limit the sustainable flux operation.

Three identified approaches as described below were studied in this project to tackle this limitation of the conventional MBR:

Approach 1

Reducing the filtration resistance and fouling tendency caused by EPS, TOC and fine colloids in the MLSS by combining the MLSS with an adsorptive support, such as PAC.

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Approach 2

Locating the submerged membranes in a separate chamber from the aeration tank to reduce the MLSS loading on the membranes. This can be assisted by arranging flow distribution from and back to the aeration chamber to minimize solids carry-over to the membrane chamber, and by incorporating approach 1, i.e. supporting the MLSS on media, such as PAC, in the first chamber.

Approach 3

Increasing surface shear by creating a great 'collision' impact on the membrane surface by using relatively large particle beads (e.g. polymer beads: 600 μ m) to detach the fine foulants attached on the membrane surface to recover membrane permeability.

The objectives and aims of the study are:

- (i) To examine the use of PAC as a support of biomass in the bioreactor and assess the effect of this on the membrane filtration characteristics of the MLSS.
- (ii) To investigate the fundamental mechanisms involved in improving the filtration performance of MBR (BAC).
- (iii) To optimise the filtration performance of MBR (BAC) as a function of SRT, PAC size and concentrations etc.
- (iv) To establish a suitable design and operating protocol to allow stable operation of a two compartment bioreactor, with low SS or supernatant in the second compartment.
- (v) To compare the filtration performance of the submerged membranes with MLSS and lower SS or supernatant.
- (vi) Provide guidelines for the design a high filtration efficiency, reliable and economic alternative MBR.

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1.3 The structure of the report

This report consists of nine chapters. The first chapter introduces the background of the study and its aims and objectives. Chapter 2 provides a literature review pertinent to the MBR system in terms of membrane fouling, fouling control, previous applications of PAC in activated sludge and alternative MBR designs. The following Chapter 3 describes the materials and methodologies used in this study. Chapter 4 and 8 present and discuss the results obtained and the last chapter gives conclusions from the past years study and outlines proposed recommendation work as shown in Table 1.1.

Table 1.1: Description of the chapters in this thesis

Chapter	Description
4	Effect of MBR (BAC) cultivated at SRT 30 days and effective infinity was evaluated.
5	Performance of four 2L batch MBR (BAC)s with different PAC concentrations at SRT 30 days were compared. The short term predictors were also identified to predict the long-term performance of MBR (BAC).
6	Mechanisms involved in improving filtration performance in MBR (BAC) were identified and compared.
7	The best operating conditions of MBR (BAC) in terms of SRT and PAC size were identified.
8	The alternative designs to MBR (AS) and MBR (BAC) consist of two-compartment MBR and MBR (polymer bead) were evaluated. Large polymers were found have a good potential to improve the performance of MBR (AS) in terms of fouling control and operating cost.
9	Conclusions from this study were made based on the discussion of experimental results obtained. In addition, suggestions about further investigation of this study were also recommended.

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2.1 Introduction to MBRs

MBRs are now widely used for the treatment of domestic and industrial wastewater. They consist of a bioreactor coupled with membranes. MBRs have several advantages (i) they can retain high biomass concentrations and operate at long SRT if required (Kishino *et al.*, 1996), (ii) they require a small foot print and (iii) they have the ability to treat high strength wastewater (Guttau *et al.*, 2002; Rosenberger *et al.*, 2002). Apart from keeping the biomass in the bioreactor, the membrane may also retain some of the organic carbons from escaping for subsequent biodegradation (depending on their molecular weight). For example, Xing *et al.*, (2000) have reported that approximately 12 % of pollutants removal could be attributed to the membrane compared to 85 % by the bioreactor (although this would depend on both membrane type [UF vs MF] and nature of the organics).

A feature of characteristic of the MBR is membrane fouling which leads to transmembrane pressure (TMP) increase or permeate flux decrease and shortens the life of the membrane. In order to prolong the life span and reduce the maintenance cost, fouling control is important. Energy input is required to hydrodynamically reduce fouling and chemical cleaning of membrane is employed when the operation of the system is unacceptable. However, the methods used are not always efficient due to the required system shutdown or loss of production. These problems tend to obstruct the MBR from reaching its optimum potential. In this section, pertinent studies are reviewed to obtain insight into alternative designs for MBRs with especial focus on modifying specific cake resistance, irreversible membrane fouling control and hybrid MBRs with supported biomass (i.e. PAC, polymer beads etc) and supernatant rather than mixed liquor filtration concepts.

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2.2 Factors affecting fouling in MBRs

The fundamental function of the membrane in an MBR system is for solid-liquid separation and production of particulate free clean water. However, the efficiency of the process is dependent on the choice of membrane and its characteristics. The following section will discuss the interactions between membrane pore size, membrane materials and the feed solution.

2.2.1. Membrane pore size

Usually, two types of membrane are used in MBR systems. The characteristics of these membranes are summarized in Table 2.1 along with a third option, nanofiltration. It is clear that microfiltration (MF) and ultrafiltration (UF) membranes are popularly used in MBR systems because of the disadvantage of high pressures requirement for nanofiltration (NF). Choi *et al.*, (2007) noted that the high pressure required is due to the intrinsic tightness and high salt rejection of the NF. However, they found that the NF MBR could produce high quality permeate water. Comparing the two low pressure processes, it has been observed that MF membranes tend to foul more easily than UF membranes. The cause is most probably due to internal fouling of MF membrane pores by high molecular weight (100k-1000k dalton) organics rather than the surface deposition achieved by UF membrane (Albasi *et al.*, 2002; Hong *et al.*, 2002).

Table 2.1: Types of membrane process used in MBRs (Fane & Chang, 2002)

Membrane	Pore size	Removal performance
Microfiltration	0.1–0.2 μm	(a) high SS removal
		(b) high bacteria and partial removal of viruses
Ultrafiltration	0.1μm down to less than 5 nm	(a) high virus removal
		(b) reasonable EPS removal
Nanofiltration	≈ 2nm	(a) retain most species, except for certain monovalent ions and low molecular weight organics.

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However, in MBRs, MF is often able to produce equivalent good quality of permeate to UF. Thus MF has the ability to prevent the transmission of fine particles in the permeate, which indicates that the biofilm formed on the membrane in MBRs can play an important role in helping the membrane eliminate pollutants.

2.2.2. Membrane materials and feed solution

Membrane material and feed solution interaction can have significant effects on the performance of MBRs, particularly on permeate flux and membrane fouling (Jiang *et al.*, 1995; Jönsson & Jönsson, 1995; Lindau *et al.*, 1998; Wisniewski & Grasmick, 1998). For example, in one study it was found that polyethylene (PE) membranes tend to have irreversible fouling, whereas for polyvinylidene (PVDF) membranes fouling was mostly caused by reversible components. As a result, PVDF membrane had a better fouling control than that of PE membrane in the MBR (Yamato *et al.*, 2006). It also has been reported that flux reduction appeared to be less with hydrophilic compared to hydrophobic membranes (Jönsson & Jönsson, 1995; Srijaroonrate *et al.*, 1999; Choi *et al.*, 2002a). With the same pore structure and characteristics, hydrophilic membranes also have a higher and more stable flux than hydrophobic membranes. Although hydrophobic membranes foul significantly, their initial flux may be higher than that of hydrophilic membranes (Choi *et al.*, 2002a). A good membrane should be able to produce high and stable flux throughout the operation period. This shows that the choice of membrane is very important as it is the initial step in countering membrane fouling problems (Jiang *et al.*, 1995).

Wastewater composition is also an important parameter to be considered before choosing the most appropriate membrane for operation. According to Sponza (2003), the wastewater composition rather than the operational conditions of an MBR system appears to have a strong influence on EPS production and the physiochemical properties of flocs (Sponza, 2003). EPS analysis also shows that the protein content in the activated sludge is greatly influenced by the substrate composition in wastewater (Sponza, 2003). However, extreme aeration conditions could cause floc breakage. Release of foulants such as EPS and fine colloids from

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the broken floc would occur. These foulants have the tendency to block and plug the pores of membranes during filtration. Table 2.1a shows the effect of organic fines on membrane fouling.

Table 2.1a: Type of foulants and its effect on membrane filtration

Type of membrane foulant	Effect of foulant on the membrane	References
1. Biopolymer clusters (BPC): Organic substances mainly consist of SMP and colloidal organic substances	a. BPC is normally found within the pores of the sludge cake formed on the membrane surface which cause the high filtration resistance of the cake sludge.	Wang et al., 2007; Wang & Li, 2007
2. Colloidal/EPS: Proteins, polysaccharide matters and lipids.	a. The breakage of microbial floc promotes the release of colloidal and soluble components and thus increase the accumulation of the sticky foulants on the membrane surface and cause membrane fouling.	Meng et al., 2007
3. Colloidal TOC and/or soluble EPS (soluble carbohydrates and humic substances)	a. The attachment of the sticky foulants on the membrane surface responsible for membrane fouling and the reduction of the critical flux.	Fan et al., 2006
4. Polysaccharides and other biopolymers	a. Accumulation of the foulants on the membrane decreases the membrane permeability. b. Suspension form biopolymers increases the viscosity of the MLSS and reduces membrane filterability. c. Biopolymers on the membrane surface encourage biomass	Chu & Li, 2004

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	attachment and aggravate the deposition of the sludge cake on the membrane surface.	
5. Biofilm formation (fouling layer made up of microbial cells covered with EPS)	<p>a. Biofouling is the formation of mature biofilm layer and/or the accumulation of EPS on the membrane surface.</p> <p>b. SMP would provide organic substrate to the growth of biofilm on the membrane surface.</p> <p>c. EPS would also permit microbial cells to adsorb onto the membrane surface to promote biofilm growth.</p>	Ng et al., 2006
6. Biofilm and EPS	<p>a. The growth of biofilm on the membrane was the main factor of membrane fouling.</p> <p>b. EPS in biofilm is the key membrane foulants.</p>	Yun et al., 2006
7. EPS	<p>a. . Accumulation of the EPS on the membrane surface was the major component to the increase of the membrane resistance.</p>	Chang and Lee, 1998; Naogaka, et al., 1996

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There is also evidence that inefficient aeration leading to low dissolved oxygen (DO) can cause more fouling. Jin *et al.*, (2006) found that a low DO (<0.1 mg/L) MBR had a fouling rate 7.5 times more than a high DO (>3.0 mg/L) MBR and the small particle in the biofilm and specific cake resistance (SCR) of the low DO MBR was higher than that of high DO MBR. They also reported that the porosity of the biofilm on the membrane surface was low for low DO MBR compared to high DO MBR. If the low DO was due to insufficient intensity of aeration, this would also accelerate the sludge cake deposition rate on the membrane surface. Thus, the biological activities, nature of feed solution and operating parameters should be carefully analysed and controlled when using MBRs. These factors are discussed in more detail below.

2.3 MBR hydraulic characteristics

The formation of a sludge cake on the membrane surface can lead to the development of significantly greater hydraulic resistance than that without cake formation (Tardieu *et al.*, 1998). The hydraulic characteristics in the MBR are known to be important for retarding membrane fouling or cake deposition on the membrane surface (Tardieu *et al.*, 1999). The relationship between permeate flux, TMP, superficial gas velocity, MLSS concentration and different operating modes will be discussed in the following sections. Special focus will be on the cake accumulation on the membrane surface and internal pore restriction of the membrane.

2.3.1. Flux and transmembrane pressure (TMP)

There are two operational modes in MBRs which are fixed flux or fixed pressure. Under constant flux operation, TMP change plays an important role as an indicator of fouling or stable operation of the MBR system (Rosenberger *et al.*, 2002). In constant pressure MBRs, the reduction of the flux over the time shows that membrane fouling has taken place.

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The governing equation is based on,

$$J = \frac{\Delta P}{\mu(R_m + R_f)} \quad (2.1)$$

where R_f is the fouling resistance, which could have both reversible (R_{rf}) or irreversible (R_{ir}) components. In terms of ΔP equation (2.1) can be written,

$$\Delta P = \mu (R_m + R_f) \quad (2.2)$$

It should be noted that for fixed flux operation equation (2.2) (Figure 2.1 (a)) applies and ΔP increases linearly with R_f . For fixed pressure equation (2.1) (Figure 2.1 (b)) applies and J decreases inversely with R_f (so the rate of decline of J may appear to slow down even if R_f is steady increasing).

Figure 2.1: Fixed flux (a) and fixed pressure (b) illustration graphs

In fixed flux mode, the permeate flux from MBRs is controlled at a constant flow which is normally lower than critical flux (see 2.5.1) or typically lower than a flux of 20 L/m²/hr. In the findings of Li and Wang, (2006), operating flux is the most important factor controlling fouling of their MBR. The filtration flux applied should not cause cake formation in a long-term run. Fixed pressure MBRs may produce higher initial flux compared to fixed flux MBRs. However, the initial high flux induces higher fouling rates. This indicates that operating MBRs at fixed flux with low or modest flux (< 20L/m²/hr) could be more beneficial than at fixed pressure with high constant pressure (eg. 60kPa). This is due to (i) lower initial permeate flux or constant flux has better membrane fouling control and achieves better

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production over the long term operation (Hong *et al.*, 2002; Gui *et al.*, 2002) and (ii) it can avoid overfouling during the initial stages of filtration (Defrance & Jaffrin, 1999).

Choo & Lee, (1998); Chan *et al.*, (2002) have reported that the increase in TMP in long term continuous operation could be attributed to the formation of a dynamic layer of retained microbial and particulate species. This indicates that prevention of the cake to form on the membrane surface can prolong the life of the membrane in MBRs. An effective method is to have a good aeration distribution to create shear forces on the membrane surface to prevent cake formation (Li and Wang, 2006). Equal importantly, the control of pore blocking and plugging can also extend the life span of the membrane significantly.

Without proper membrane protection in MBRs, fixed flux operation could still lead to gradual membrane fouling and ultimately cause a rapid increase in TMP, or TMP jump (Cho and Fane, 2002). Cicek *et al.*, (1997) pointed out that a sudden sharp increase in TMP would be an indication of severe membrane fouling. There are several possible reasons that could lead to a TMP jump. One of the reasons may be the slow accumulation of foulant that leads to a flux distribution that eventually reaches a condition where (in order to meet the average imposed flux) parts of the membrane have a flux raised to above the 'critical flux' of the dominant foulant (biofloc) (Cho and Fane, 2002). Another factor that could cause the TMP jump may be due to irreversible pore blocking and plugging, again on a local scale. There are indications that sudden and rapid increase of TOC in the MBRs bioreactor caused by floc breakage or bioactivity has a high possibility to accelerate the TMP jump. Wisniewski and Grasmick, (1998) have found that half of the membrane fouling could be due to the soluble elements such as polysaccharides, phospholipids, protein and etc.

To prolong the period of membrane filtration without the TMP jump in MBRs, it is advisable (i) to operate the MBR only when the biomass in the bioreactor becomes stable, to avoid filtering of large quantities of fine pollutants (TOC, EPSs) produced

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by biomass in unstable conditions, (ii) to counter the sudden higher loading rate of wastewater or bioactivity which may produce TOC in large amounts by adding adsorbent (i.e. PAC) in the bioreactor, (iii) to operate the MBR at the lower flux ($< 20 \text{ L/m}^2/\text{hr}$) and (iv) to ensure the bioreactor is equipped with good air distribution which is able to supply sufficient DO for bioactivity and create good shear force on the membrane surface (in many design this requires both fine aeration (for oxygen transfer) and coarse aeration (for surface shear)).

2.4 Biological characteristics

Biological activity plays a major and very important role in MBRs. The biomass in the bioreactor can biodegrade the organic pollutants in the wastewater up to 80-90% (Xing *et al.*, 2000) with the condition that the biomass is stable. For example, the biomass that contains excess growth of filamentous bacteria would produce large quantity of EPSs (Meng *et al.*, 2006). This means that an unstable biomass state tends to cause a mass production of the soluble microbial products (SMPs) or EPSs. SMPs in the bioreactor increase the colloidal TOC concentration in the supernatant and this will deteriorate the performance of MBRs, as SMPs can cause serious irreversible fouling to the membrane during filtration (Orantes *et al.*, 2006). The combination of these two fractions of biomass and supernatant containing colloids and solutes form the mixed liquor in MBRs (Lee *et al.*, 2003). The colloids are polymers (submicron particles) not connected to the cells at all (Laspidou & Rittmann, 2002) and the solute fraction consists of micro and macrosolutes that are mostly biopolymers and EPSs. Biomass in the mixed liquor includes bacterial floc cells, which are surrounded by EPSs or single (planktonic) organisms. EPSs, fine colloids, fine particles or SMPs in the mixed liquor have been recognized as foulants of membrane. Recently, Meng *et al.*, (2006) observed that the quantity of filamentous bacteria in the mixed liquor affected the filtration performance. They found that the sludge flocs became smaller if the quantity of filament in the mixed liquor was negligible. Whereas, the present of the large concentration of filament would cause the flocs to form a non-porous layer on the membrane surface. This denotes that the concentration and composition of the mixed liquor affects the

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performance of membranes in MBRs (Bouhabila *et al.*, 1998; Tardieu *et al.*, 1999; Bouhabila *et al.*, 2001; Lee *et al.*, 2001). Sludge and hydraulic residence times (SRT & HRT) are the two parameters that can be used in controlling mixed liquor concentration and composition. Both of these parameters affect the growth, function and production of net sludge and SMPs in mixed liquor. Therefore, good control of SRT and HRT will enhance the performance of MBRs.

2.4.1. Mixed liquor suspended solids and membrane foulants

As discussed above, mixed liquor consists of biomass and supernatant. In addition, mixed liquor in suspension form is known as mixed liquor suspended solids (MLSS) or mixed liquor volatile suspended solids (MLVSS). MLVSS is the biomass contribution to the wastewater biological treatment (Hasar *et al.*, 2002). It forms a large portion (52-75%) of MLSS with SRT ranging from 10 to >200 days whereby the higher SRT the lower the MLVSS fraction (Innocenti *et al.*, 2002). MLSS can also contain 97 % of MLVSS under suitable conditions (Shin & Kang, 2003). MLSS has been used as an indicator for the stability of MBRs, where stable MBRs will have a constant MLSS concentration throughout the operation period. Besides, stable MBRs will also have a constant and lower concentration of foulants such as SMPs, fine colloids or particles, EPSs and TOC in the bioreactor. An unstable MLSS concentration and mass production of fine foulants by unstable bioactivity will undermine the performance of the MBRs. Trussell *et al.*, (2006) observed that the membrane fouling rate could be correlated with the total SMP (quantified by polysaccharide and protein) concentrations. It was reported by Innocenti *et al.*, (2002) that 8-10 g/L of biomass is probably the optimal concentration in MBRs which will produce a good quality of effluent. However, Meng *et al.*, (2006a), reported that MLSS concentrations had an exponential relationship with the membrane fouling resistance. They argued that the higher MLSS concentration the higher the foulant concentrations in the mixed liquor. However, it could also be argued that the effect of MLSS concentration on fouling will depend on the imposed flux.

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The reported relative contributions to the resistance of membrane filtration by suspended solids (SS), colloids and dissolved molecules tend to vary depending on the type of MBR studied. In one study, the values were found to be 65%, 30% and 5% respectively (Defrance *et al.*, 2000). Bae and Tak, (2005) also found that the suspended solids were the major foulants, followed by colloids and solutes. However, they observed that solutes were the initial foulant that formed the cake layer on the membrane surface. Psoch *et al.*, (2006) also agree that cake resistance is a major cause of membrane fouling. Another finding showed that sludge layer resistance built up to about 60% of total resistance of the MBR (Chae *et al.*, 2006). Li and Wang, (2006) found that apart from filtration flux, sludge stickiness and concentration were the major problem causing membrane fouling. These results suggest that the filtration of supernatant should be more beneficial compared to mixed liquor. It has also been shown by Lee *et al.*, (2003) that no remarkable membrane fouling occurred during filtration of a supernatant fraction.

However, another group of researchers (Li *et al.*, 2005) found that the colloids and solutes in the supernatant of mixed liquor were the dominant foulants instead of the suspended sludge. Another finding revealed that colloidal particles are a predominant foulants affecting critical flux (Fan *et al.*, 2006). Whereas, they noticed that MLSS and organic solutes had little effect on the critical flux. Grelier *et al.*, (2006) identified the colloidal and soluble polysaccharide as the predominant parameters affecting membrane fouling. Chang and Kim, (2005) also reported that the filtration of supernatant containing small particles was not as beneficial compared to mixed liquor. Therefore the literature is divided on the benefit of filtration of the supernatant.

If single cells are present in supernatant, they are likely to get to the membrane. If flocs are also present, they could 'mop up' the single cells. These findings may in fact show that MLSS at stable state can actually protect the membrane. In Orantes *et al.*, (2006) opinion, a dynamic biomass layer on the membrane surface was able to act as prefilter to prevent penetration of soluble matter into the pores of the membrane to reduce fouling rate. The three possibilities whereby the MLSS can

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protect the membrane are (i) MLSS forms a dynamic protective layer to prevent the foulants from blocking and plugging the pores of the membrane, (ii) the collision between the MLSS floc and the membrane helps to detach the fine foulants on the membrane surface back to the bulk liquid and (iii) the MLSS floc 'mop up' planktonic bacteria by adsorption or enmeshment, thereby reducing their population in suspension. Though these are potentially beneficial roles of the SS, the fixed flux MBR system is not advised to be operated at high flux ($> 20 \text{ L/m}^2/\text{hr}$) or above critical flux (see section 2.5.1) to prevent premature serious fouling due to the cake formation of the SS on the membrane surface. This means that the discussed function of MLSS as a membrane protective layer may not be a significant or convincing role. Orantes *et al.*, (2006) found that the MLSS layer on the membrane surface may be beneficial to protect the membrane in the initial stage. However, they observed that if the cake layer became compacted it would lead to fouling problems for the membrane. Instead, the filtration of the supernatant would avoid the cake layer problem created by of the SS. This indicates that the alternative way to enhance the MBR performance in terms of high flux operation ($> 20 \text{ L/m}^2/\text{hr}$) is the filtration of the supernatant. However, to optimise the filtration of supernatant performance, the SMPs, colloids and solutes should be minimised or eliminated. PAC with its high adsorption capacity can play a role here effectively. Besides, the prevention of the single cells from being lysed in the separated supernatant is important too. This needs to be examined further.

The above discussion implies that there are many factors which can cause membrane fouling and it happens with both supernatant and mixed liquor filtration. The factors that cause membrane fouling may be due to the elements in the supernatant, concentration of activated sludge (MLSS) or characteristics of the feed stream to the MBR. It is important to understand the effect of the fouling components to prevent excessive membrane fouling.

The difficulty with MBRs treating raw municipal wastewater is to maintain a constant concentration of foulants entering the bioreactor, because the substrate concentration in raw municipal wastewater vary throughout the day. Higher loading

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rates of wastewater normally happen in the morning or afternoon and before and after working or school hours. Trussel *et al.*, (2006) noticed that increase of the food to microorganism (F/M) ratio would increase the membrane fouling rate. Nonconstant addition of wastewater loading rate has a high possibility of causing instability of bioactivity. Unstable bioactivity tends to cause a problem regarding mass production of SMPs or TOC such as macromolecules or dissolve molecules which are adverse to membrane filtration. Gui *et al.*, (2002) and Ognier *et al.*, (2002) have reported that macromolecule adsorption on the membrane during filtration can cause irreversible fouling in MBRs.

TOC accumulation in the supernatant of settled activated sludge has also been found to inhibit the metabolic activity of the activated sludge and thus contributes to the poor membrane permeability of the mixed liquor (Huang *et al.*, 2001). Wang *et al.*, (2007) also discovered that accumulation of biopolymer clusters (measured as TOC) would cause the porosity of the sludge cake to decrease and lead to increase of the cake sludge filtration resistance. This demonstrates that an effective activated sludge biomass population is required to sustain good performance of the MBRs during high wastewater loading rates. The MLSS must have the capability to withstand sudden high concentrations of loading rate of wastewater and high TOC production due to unstable bioactivity caused by inconsistent wastewater feed. In addition, MLSS should have the effect of membrane protection to prevent the fine foulants from blocking and plugging the membrane pores. To achieve this, one potentially feasible method is to combine the activated sludge with PAC to form biologically activated carbon (BAC). It has been also found that BAC was more adaptable to the sudden wastewater loading rate condition (Park *et al.*, 1999).

From the above discussion, MLSS has a very dominant role in maintaining adequate performance of MBRs. The general consensus is that the higher the MLSS concentration, the higher the tendency of membrane fouling. Li and Wang, (2006) agree that sludge concentration is one of the factors affecting filtration performance. However, Hong *et al.*, (2002) observed that the membrane fouling was independent of MLSS concentration for the range between 3.6 to 8.4 g/L. At any concentration,

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it has been agreed that to maintain good conditions of MLSS, excess sludge should be wasted at regular intervals. Without excess sludge wastage at regular intervals, the MLSS concentration in the MBR system can rise to values of between 20-60 g/L with complete retention of biomass in the MBRs being reported (Witzig *et al.*, 2002). However, it is suggested that at the high concentrations of MLSS, a very small amount of sludge has to be wasted in order (i) to get rid of accumulated inorganic substances which are detrimental to the filtration in the reactor (Rosenberger *et al.*, 2002) and (ii) to maintain low sludge viscosity, suction pressure and MLSS concentration at the designed concentration (Ueda *et al.*, 1996, Hasar *et al.*, 2002). Chae *et al.*, (2006) also agree that high viscosity of mixed liquor may cause membrane fouling. However, an advantage of high concentrations of biomass (≈ 20 g/L) in the MBRs is the ability to treat higher loading rates more effectively than low concentrations of biomass (Holler and Trösch, 2001, Rosenberger *et al.*, 2002). Thus the MBR should be operated at optimal MLSS concentration in order to achieve high permeability, low fouling rate and high quality effluent. MLSS of good quality and optimal concentration can be controlled by fixing sludge retention time (SRT) and hydraulic retention time (HRT).

The methods which can be employed to reduce membrane fouling include pretreatment of the feed stream, modification of the system geometry and use of supported biomass with adsorptive effects in MBRs. In this study, MBR (BAC) performance had been thoroughly investigated. In addition, an alternative two-compartment MBR design with PAC as biomass support which can presumably help to operate the system more efficiently in terms of (i) optimising the MLSS concentration with higher density floc, (ii) reducing fine colloids, TOC and EPS and (iii) avoid direct contact of the membrane with the mixed liquor was also studied.

2.4.2. Extracellular polymeric substances (EPSs)

The definition of EPSs can be summarised as a matrix of large polymeric molecules produced from the bioactivity that appears in bound (at cell surface) and soluble (outside cell surface) forms (Laspidou & Rittmann, 2002). Shin & Kang, (2003)

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have also defined suspended (soluble) EPSs in the supernatant as the organic compounds that result from substrate metabolism for biomass growth and decayed cells. EPSs have been recognized as having some beneficial functions in the activated sludge which cannot be denied such as (i) EPSs in activated sludge occurs as a capsule to provide large surface area per unit volume for microorganism binding or simply to provide the 'glue' for the bacteria floc which enhances flocculation (Rosenberger & Kraume, 2002; Sponza, 2003) and (ii) EPSs allow microorganisms to live continuously at high-cell densities in stable mixed population communities and allows cooperation and communication among cells in microbial aggregates (Lapidou & Rittmann, 2002).

Despite the functional importance of EPSs in the activated sludge process, EPSs appears to be one of the major components that fouls the membrane (Wisniewski & Grasmick, 1998; Cho and Fane, 2002; Meng *et al.*, 2006a; Chae *et al.*, 2006; Wang *et al.*, 2006; Meng *et al.*, 2006; Fan *et al.*, 2006). However, many researchers have reported that only soluble (suspended) rather than bound (extractable) EPSs affect the membrane fouling (Tardieu *et al.*, 1999; Rosenberger & Kraume, 2002; Rosenberger *et al.*, 2006; Yamato *et al.*, 2006; Fan *et al.*, 2006). Tardieu *et al.*, (1999) has reported that membrane fouling is only caused by suspended EPSs and colloidal components in the biological suspension if under adequate conditions of recirculation. Rosenberger & Kraume, (2002) have also found the major influence is the concentration of soluble EPSs in the supernatant, the greater the suspended EPSs concentration, the higher the membrane fouling tendency.

However, the results of other investigations show that the composition and properties of EPSs are the key parameters relating to membrane fouling (Lee *et al.*, 2003). Fan *et al.*, (2006) have identified that both soluble carbohydrate (polysaccharide) and humic substances are two major colloidal TOC components responsible for fouling. They even strongly suggested that colloidal TOC could become an indicator to predict the sludge fouling tendency. Meng *et al.*, (2006, 2006a) observed that total EPS, including protein and polysaccharide and SMPs (included in EPS) were the major components of foulants affecting membrane

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permeability. They found that an increase in the total EPSs also caused an increase in the mixed liquor viscosity and induced more polymers and sludge particles to accumulate on the membrane surface. Kim and Jang, (2006) agree that proteins and carbohydrates are the major foulant components causing membrane fouling. In summary, only certain components of EPS, rather than the total quantity of EPS, in the bioreactor appears to foul the membrane (Lee *et al.*, 2003). The above discussion indicates that the EPS in the supernatant probably fouls the membrane more significantly than the bound EPS in the pellet (bound to the cells on the floc). However, some researchers found that there is no direct correlation to link the EPS to membrane fouling (Zhang *et al.*, 2006). Apart from EPS, the presence of filamentous bacteria maybe an important ‘culprit’ affecting filtration performance as discovered by Meng *et al.*, (2006). In this study, it was found that the polysaccharide in the supernatant were one of the most significant foulants. Several researchers have made similar observations (Fan *et al.*, 2006; Rosenberger *et al.*, 2006; Wang *et al.*, 2006; Zhang *et al.*, 2006b; Grellet *et al.*, 2006; Drews *et al.*, 2006). In this study, PAC has been used to uptake the polysaccharide to reduce the concentration in the supernatant.

2.4.3. Sludge retention time (SRT) and hydraulic retention time (HRT)

As discussed in a preceding section (section 2.4.1), MLSS in its stable state and optimal concentration can enhance the performance of the MBRs. SRT is one of the operating parameters that can stabilize and optimize the MLSS condition to biodegrade the wastewater effectively. This agrees with the research findings from Innocenti *et al.*, (2002) and Lee & Kim, (2002), who reported that the ability of the biomass in MBRs to biodegrade pollutants is affected by the SRT.

Longer SRTs may be advantageous for an MBR system in order to establish a more acclimatised biomass which has high efficiency in treating wastewater (Xing *et al.*, 2000; Rosenberger *et al.*, 2002a; Shin and Kang, 2003). However, other claimed advantages of a longer SRT are also reported such as (i) a relatively high SRT induces less fouling on the membrane (Bouhabila *et al.*, 1998; Innocenti *et al.*, 2002) and (ii) due to high SRT, the concentration of suspended solids and volatile

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suspended solids in the bioreactor are increased accordingly and the net sludge production will decrease due to the resulting low food to microorganism (F/M) ratio (Xing *et al.*, 2000; Bouhabila *et al.*, 2001; Huang *et al.*, 2001; Rosenberger *et al.*, 2002; Han *et al.*, 2005). Most of the researchers quoted above recommended that a SRT of approximately thirty days is the most appropriate value to be used in operating MBRs. However, there is evidence that commercial practice is to use lower SRTs of 20 days (Qin *et al.*, 2006, 2007).

According to the reports by Lee *et al.*, (2003) and Han *et al.*, (2005), a prolonged SRT of more than 30 days is detrimental to the performance of the MBR system. This may be because (i) prolonged SRT encourages higher MLSS and viscosity and this causes the effectiveness of air scouring in membrane fouling control to be reduced (Rosenberger *et al.*, 2002; Han *et al.*, 2005), (ii) a decrease in quality of the permeate was observed for very long SRT (> 200 days) (Innocenti *et al.*, 2002), (iii) for infinite SRT, inorganic compounds tend to accumulate on the bottom of the bioreactor (Rosenberger *et al.*, 2002) and (iv) specific biological activities such as specific oxygen uptake rate, specific nitrification rate and specific denitrification rate would be decreased at prolonged SRT (>30 days) (Han *et al.*, 2005). However, Grelier *et al.*, (2006), reported that a MBR operated at SRT 40 days performed the best compared to SRT 8 and 15 days in terms of (i) polysaccharide concentration in the mixed liquor, (ii) biodegradation process and (iii) fouling rate control. Another investigation showed that SRT lower than 40-20 days would see an increase in SMP concentration in the mixed liquor and the sludge concentration and sludge filterability decrease (Holakoo *et al.*, 2007).

However, it was also reported that at an extra long SRT ($\approx$ infinity SRT), the acclimatised biomass could decompose high molecular weight organics to a lower size range which can be more easily biodegraded (Shin and Kang, 2003). At SRT > 1000 days, Laera *et al.*, (2005) found that the MBR still had (i) a complete nitrification and good efficiency in COD removal, (ii) good filtration performance at a modest flux of 16-18 L/m²/hr and (iii) no inorganic material accumulated in the bioreactor once it had achieved equilibrium. Xing *et al.*, (2006) also reported the

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long-term stable operation of an MBR without excess wastage. To minimise sludge production, Sridang *et al.*, (2006) operated an MBR without sludge extraction to treat seafood wastewater. Massé *et al.*, (2006) also noticed that the sludge production decreased as SRT increased from 9 to 110 days. The range of opinions in the literature shows that, the effect of SRT on the performance of MBRs still requires further investigation.

Apart from SRT, HRT is also one of the MBR operating parameters and HRT can be simply controlled by varying reactor volume and flow rates (Rosenberger *et al.*, 2002). Short HRT can help to stimulate the growth of biomass rather than a long HRT in the initial stage of biomass cultivation. However, the removal efficiency of chemical oxygen demand (COD) tends to be low at relatively low HRT during the MBR operation (Bouhabila *et al.*, 1998). Another finding reported that a decrease in the HRT from 10 to 4 hours led to an increase of EPS concentration and average sludge floc size, and caused the reduction in the settling of the sludge and more membrane fouling (Chae *et al.*, 2006). It was also found that shorter HRTs (12; 18 hours) required more frequent membrane cleaning and chemical cleaning compared to longer HRT (24; 30 hours) (Chang *et al.*, 2006). This supports the view that good control of SRT and HRT help to improve MBR performance. Table 2.2 shows a summary of selected MBRs reported by researchers.

Table 2.2: HRT and SRT used in MBR system

HRT (h)	SRT (d)	Tank working volume (L)	Type of w/w*	Type of system process	Membrane surface area (m ²)	MLSS concentration (g/L)	References
24	60	12	Synthetic w/w	Aerobic, s/b*	11.7	3.6	Choi, J. H <i>et al.</i> , 2002
24	60	600	Domestic and industrial	Aerobic, s/d*	0.25	10.0	Defrance <i>et al.</i> , 1999
24	60	600	From	Aerobic,	0.25	10.0	Defrance &

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			Compiègne treatment plant	s/d			Jaffrin, 1999
8-24	30- 100	50	Domestic w/w	Aerobic, s/b	0.3	0.76-1.77	Hasar <i>et al.</i> , 2002
*17	20	1000	Municipal w/w	Aerobic- anoxic, s/d	1.7	5.84 gCOD/l	Silva <i>et al.</i> , 1998
13- 16	125	21400	Domestic w/w	Aerobic, s/b	168.0	8.0-12.0	Ueda <i>et al.</i> , 1996
12- 16	20- 30	21-26	Synthetic w/w	Aerobic, s/b	0.1	8.0-16.0	Shim <i>et al.</i> , 2002
10.4- 15.6		3900	Municipal w/w	Aerobic- anoxic, s/b	13.9	18.0-20.0	Rosenberger <i>et al.</i> , 2002a
14	10, 190, >200	Pilot scale	Industrial w/w	Aerobic- anoxic	24	4.0-9.0	Innocenti <i>et al.</i> , 2002
6.7- 13.3	High	9	Oily w/w	Aerobic, s/d	0.23	48.0	Scholz & Fuchs, 2000
12, 6, 3	Infin- -ity	80	Domestic w/w and glucose	Aerobic, s/b	2.0	4.80-5.30	Visvanathan <i>et al.</i> , 1997
12		450	Domestic w/w	i/t* aeration, s/b	8.0	8.0	Shimizu <i>et al.</i> , 1996
11.5		2400 and 1863 (tank volume	Domestic w/w	Aerobic, s/b	44.0	4.0	Liu <i>et al.</i> , 2000
2-9	10, 50	Pilot plant	Municipal	Aerobic, s/b	1.83 long with	5.0, 15.0	Cote <i>et al.</i> , 1997

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200000 Dalton							
8	Vary	5	Synthetic w/w	Attached and s/s* growth, aerobic, s/b	0.0673 and 4.37	2.0-5.0 (suspension), 0.1 - 2.0 (attached)	Lee <i>et al.</i> , 2001
5	5, 10, 20, 40	1.2 (tank volume)	Domestic w/w	Aerobic, s/b	4.0	0.75-7.0	Huang <i>et al.</i> , 2001
5	5, 15, 30	30	Urban w/w	Aerobic, s/d	0.04	23.1	Xing <i>et al.</i> , 2000
1.3- 1.9		14	Synthetic w/w	Aerobic- anaerobic, s/d	3.8	10.0-22.0	Holler & Trosch, 2001
5	100	3200 (pilot scale)	Traditional Chinese medicine organic w/w	Aerobic, s/b	12.5	7.5-13.7	Ren <i>et al.</i> , 2006
0.62 5, 1.25, 2.5		0.45	Synthetic w/w	Anoxic, s/d	0.032	Pure culture	Barreiros <i>et al.</i> , 1998

w/w*: wastewater; s/b*: submerged; s/d*: sidestream; i/t*: intermitten; s/s*: suspension

Table 2.2 reveals that, most of the MBR studies have an HRT in the range between 2 to 24 hours and high SRT is more popular. The pollutant removal efficiencies shown in Table 2.2 are high. This shows that MBRs can be high efficiency wastewater treatment plants which can treat wastewater effectively, and this may be

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due to the MBR having a short HRT (compact system) but a long SRT which can retain high biomass concentrations (less waste sludge).

2.5 Fouling control in MBRs

The main problem of operating MBRs is membrane fouling as fouling shortens the life span of the membrane. Seriously fouled membranes require the shut down of the system and for it to be cleaned chemically. This disrupts the production of the MBRs. In addition, fouling limits the sustainable flux in the MBR and this influence required membrane area and capital cost. Thus, the reduction of membrane fouling leading to more cost effective operation of MBRs becomes a major research interest.

The fouling caused by cake accumulation is known as reversible fouling and the removal of the cake accumulation on the membrane surface can be done with simple flushing of the membrane with the tap water. However, membrane fouling caused by pore blocking or internal pore deposition is irreversible and cannot be easily removed by rinsing with tap water. Chemical agents are required to eliminate the foulants which have blocked and plugged the membrane pores. The definition of irreversible fouling from Choi *et al.*, (2005), is that irreversible fouling is initially formed by pore blocking of small particles followed by strong interaction of fouling layers with mainly dissolved materials and by fouling layer compaction due to permeation drag. Liu *et al.*, (2005) even found that microbial growth could occur within the membrane structure which would lead to pore plugging.

The cake accumulation on the membrane surface will happen only after operating the MBRs for a certain period of time. It depends on the condition of aeration distribution in the bioreactor, MLSS condition and concentration and operating flux. Good control of the reversible fouling of the membrane is not enough to sustain the high performance of membrane filtration in the MBRs. Only the simultaneous control of reversible and irreversible membrane fouling will enhance the performance of the MBRs.

However membrane fouling varies from one case to another. This is due to different contact times with the membrane and the compounds deposited on it (Ognier *et al.*, 2002a). If there is no cake deposition on the membrane, the soluble fraction may be the source of irreversible membrane fouling (Tardieu *et al.*, 1999). Irreversible fouling of membranes may take place in the very initial period of long-term experiments. This is due to the filtration of the mixed liquor which contains the SMPs, fine colloids and particles. These fine foulants block and plug the membrane pores during filtration and it is recognized that the irreversible fouling causes higher membrane resistance (Kim *et al.*, 1998; Lee *et al.*, 2001). This indicates that without proper control, irreversible fouling may lead to a greater flux reduction than expected (Choo & Lee, 1998) and thus demonstrates that the success of prevention of irreversible and reversible fouling at the beginning of the filtration process will be a great help to prolong the filtration period without cleaning. According to Cicek *et al.*, 1997, in order to successfully achieve the aim of the irreversible and reversible membrane fouling control, it is critical to know the relationships between the operating parameters and biological and physical performance of the MBR systems.

As a result, many methods such as (i) MBRs operating at a flux lower than the critical flux (see 2.5.1), (ii) optimization of the aeration intensity, (iii) intermittent suction and backwash and (iv) support of biomass with adsorbent, have been used to tackle membrane fouling. The discussion of the membrane fouling control methods will be discussed in this section.

2.5.1. Critical flux

The original definition of critical flux from Field *et al.*, (1995), is the flux below which a decline of flux with time does not occur. According to the definition, in principle, when the permeate flux is set below the critical flux, the TMP remains stable. This implies that membrane fouling will not occur or can be negligible below critical flux (Yu *et al.*, 2003). Figure 2.2 shows the forms of critical flux as originally defined by Field *et al.*, (1995). If the critical flux is exceeded, the TMP

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steadily increases and does not stabilize, as in dead-end filtration. The critical flux concept can therefore be used in predicting optimal operation conditions, which will prevent biomass deposition onto the membrane surface (Choo & Lee, 1998).

Figure 2.2: Forms of critical flux (Bacchin *et al.*, 2006)

However, critical flux depends on the hydrodynamics of removing cake deposited on the membrane and other variables (Field *et al.*, 1994; Gui *et al.*, 2002). If any of the variables are changed, the critical flux value obtained initially will change as well. The concept is based on a balance of forces due to convection (flux) and back transport. The back transport depends on hydrodynamic shear due to crossflow. When convection and back transport are equal the flux is ‘critical’. When flux exceeds this value, deposition can occur. For particles (supra to submicron) and large colloids, the critical flux is found to increase with particle size. This is because back transport due to inertial lift or shear-induced diffusion increases with particle size. For latex and yeast particles (sizes 3-10 μm), Li *et al.*, 2000 found their measured data could be correlated by a modified shear-induced diffusion equation,

$$J_{\text{crit}} = 0.06\gamma(a_p^4/L)^{1/3} \ln(C_w/C_B) \tag{2.3}$$

Where γ is shear rate (function of crossflow velocity) and a_p is the particle size. The relevance of this relationship for MBRs is that J_{crit} can be raised by increasing γ (for

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example increasing bubbling) and by using larger particles (for example avoiding floc breakup or the presence of small colloids).

However, in its strictest definition for MBR systems, there appears to be no critical flux, since a zero rate of TMP increase can never be attained (Le Clech *et al.*, 2003). Hence, when flux is fixed below the 'nominal' critical flux of the dominant foulants, there will also be a slow resulting TMP increase (Cho & Fane, 2002). Nevertheless, the critical flux concept is still as a useful guide for selecting the operating flux of the MBR system. Many researchers such as Chen *et al.*, (1997); Defrance & Jaffrin, (1999) and Cho & Fane, (2002) have agreed that constant flux processing below the critical flux of the 'dominant foulant' (such as biomass) should be more advantageous for MBR operation, as it should prevent premature serious membrane fouling. Wang *et al.*, (2006) noticed that the sustainable operating flux was substantially lower than the critical flux in their MBR. They identified that 56% of the critical flux was the proper operational flux to maintain good fouling control.

Several methods are available for estimation of critical flux, including Direct Observation Through the Membrane (DOTM) (Li *et al.*, 2000), and flux-stepping (Le Clech *et al.*, 2003). The flux-stepping method involves maintaining the TMP history of flux is slowly incremented. Once $dTMP/dt$ becomes measurable the critical flux has been exceeded. The critical flux concept has been widely used in this study as a short term predictor of the long-term comparative performance of the MBR and MBR (AS) (BAC). Most of the time, the concept works well, although, the test can only be used as an approximate predictor and reference.

2.5.2. Aeration in submerged MBRs

In aerobic submerged MBRs, aeration is required to supply oxygen for activated sludge growth (Kishino *et al.*, 1996; Xing *et al.*, 2000). Also, aeration is used to scour the membrane surface and with a higher aeration rate, flux decline is reduced in submerged MBRs (Bouhabila *et al.*, 1998; Hong *et al.*, 2002; Orantes *et al.*, 2006; Zheng and Liu, 2006; Fan *et al.*, 2006). This is due to the fact that aeration is

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able to reduce the formation of the cake layer by inducing surface shear (Choo & Lee, 1996; Kishino *et al.*, 1996; Ueda *et al.*, 1996; Cicek *et al.*, 1997; Cote *et al.*, 1997; Gui *et al.*, 2002; Hasar *et al.*, 2002; Innocenti *et al.*, 2002; Rosenberger *et al.*, 2002; Jeison and Lier, 2006; Li and Wang, 2006).

The prevention of the cake deposition on the membrane surface in aerobic submerged MBRs is due mainly to turbulence and shear force on the membrane surface created by bubbles. However, to enhance the function of the aeration in preventing the cake accumulation, the type and the position of the membrane in the bioreactor are also important. For example, it was found by Chang *et al.*, (2002), that the use of axially aligned hollow fibres given better fouling control than that of transverse type hollow fibre membrane modules in a bubbly flow. This means that axial flow may be better than that of transverse flow for membrane fibres during filtration with aeration and this is probably due to lesser cake deposition on the membrane surface of axial as compared to that of transverse type membranes.

Airflow rate beyond a critical value is found to have negligible effect on the cake removal efficiency (Ueda *et al.*, 1996; Bouhabila *et al.*, 1998; Wisniewski & Grasmik, 1998; Gui *et al.*, 2002; Hong *et al.*, 2002). The optimum aeration rate should be established to avoid over supply of the air which may increase the cost of the MBRs operation. It should also be noted that increasing the aeration rate for fouling control could also result in an oversupply of dissolved oxygen (DO) and create problems to the system in the case when they are also used for denitrification (Hong *et al.*, 2002).

A good design of aeration distribution should able to (i) supply enough oxygen to sustain the bioactivity at the stable state, (ii) prevent the cake accumulation on the membrane surface, (deposition of sludge floc particles as a cake layer on the membrane can cause a very quick increase in hydraulic resistance (Tardieu *et al.*, 1998)) and (iii) maintain the floc at its biggest size and without breakage. It was found that the smaller the floc size or the higher the percentage of the submicron particles in the mixed liquor, the greater the fouling tendency. This means that floc

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size may be an important factor affecting filtration performance (Meng *et al.*, 2006; Chae *et al.*, 2006; Leiknes *et al.*, 2006). The fine particles such as EPSs or individual bacterial will be released from the broken biomass floc. The released of material from broken floc has been noticed by Wisniewski & Grasmik, (1998) during the running of a sidestream MBR by recirculation without aeration in the tubular tube. According to them intensive recirculation of the mixed liquor stream in the sidestream MBR can cause floc breakage resulting in floc size reduction and the release of bacterial polymers, which can lead to membrane fouling and poor settleability of suspension floc.

As a result, to maximize the benefit of aeration distribution in the bioreactor, in many MBR designs, the air is provided in two streams, they are (i) as coarse bubbles for fouling control which can be in high intensity and (ii) as fine bubbles for oxygen supply at low aeration rate to maintain the stability of bioactivity and ignoring the scouring effect on the membrane surface.

2.5.3. Intermittent suction operation

Operating the MBR at a flux below the critical flux and optimising the aeration rate could help to decrease the fouling rate. The intermittent relaxation of the membrane filtration process is another method employed to reduce the cake deposition on the membrane surface. This is caused by the removal of particles on the membrane surface previously held by TMP during the non-suction period. This indicates that non-suction time prevents or slows the formation of consolidated cake layers on the membrane surface (Gui *et al.*, 2002). Ueda *et al.*, (1996) and Chua *et al.*, (2002) have also agreed that intermittent suction is an effective technique for long-term sustainability of membrane filtration which would slow down the build up of the membrane resistance caused by membrane fouling. Due to the advantages of the intermittent suction in membrane fouling control in submerged MBRs as stated, many researchers such as Cote *et al.*, (1997); Bouhabila *et al.*, (2001); Chua *et al.*, (2002); Gui *et al.*, (2002); Shim *et al.*, (2002); Fan *et al.*, (2006) suggested that the bioreactor design of an aerobic submerged MBR system is to immerse the membranes above the air spargers with the periodic suspension of permeation to

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control membrane fouling. Table 2.3 shows the typical intermittent suction and relaxation periods used in MBRs in membrane fouling control.

Table 2.3: Intermittent suction in MBR process

Intermittent Suction		References
On (min)	Off (min)	
8	2	Chua <i>et al.</i> , 2002; Ueda, <i>et al.</i> , 1996
10	2	Yeom, <i>et al.</i> , 1999
13	2	Huang <i>et al.</i> , 2001
10	5	Albasi <i>et al.</i> , 2002
15	15	Choi <i>et al.</i> , 2002a
1440	30	Reid <i>et al.</i> , 2006

Hong *et al.*, (2002) also agree that intermittent suction may offer an effective means of fouling control for small-scale MBR processes treating wastewater with high fouling potential. However, in their opinion, the use of intermittent suction operation may not be economically feasible for large-scale systems in terms of power usage. Apart from the cost of power usage, intermittent suction may reduce the production compared to continuous suction MBRs. It was also found that for the same on/off ratio, the longer ‘on’ period was less effective in reversible fouling layer removal (Zhang *et al.*, 2004). This leads to the idea that, if the control of the membrane fouling can be efficiently enhanced in continuous suction MBRs, it will maximize the use of MBRs in terms of cost and productivity. Alternative methods, instead of operating MBRs with intermittent suction, flux below critical flux and good aeration design could include use of hybrid MBRs with easily floated or suspended adsorbent, or particles such as polymer beads in bigger size (300-600 µm). The adsorbent will help to reduce the fine pollutants in the supernatant. Whereas, the big polymer beads could act as effective scouring agents to detach the fine foulants on the membrane surface. Large beads can also be easily separated from the wastage sludge and reused again. The other approach is to design a bioreactor whereby every area of the membrane can be accessed by the air bubbles through aeration distribution. This will make sure that all the surface area of the

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membrane used in the MBRs can be scoured by the air bubbles and if hollow fiber membrane modules are used in filtration in MBRs, the module should be designed in a way that it will not be blocked easily with the biomass and can easily be scoured. These bubble flow and module design issues appear to be well-developed by the MBR industry.

2.5.4. Membrane cleaning methods

As discussed below, membrane fouling can be divided into reversible fouling (cake deposition on the membrane surface) and irreversible fouling (pore restriction). To recover a severely fouled membrane, many methods have been used. These methods include physical and chemical cleaning. Physical cleaning methods aim for reversible fouling removal but they are not efficient in regeneration of fouled membranes caused by irreversible fouling. Chemical cleaning is the only method which can get rid of the irreversible fouling effectively.

The physical clean methods are discussed below, and include,

- (i) Rinsing the membrane with clean water.
- (ii) Backwashing techniques that have been applied to reduce the formation of a gel polarization layer and membrane fouling (Côté *et al.*, 1997; Bouhabila *et al.*, 1998; Srijaroonrate *et al.*, 1999; Bouhabila *et al.*, 2001; Rosenberger *et al.*, 2002; Psoch *et al.*, 2006). Normally, intermittent filtration and backwashing are applied in MBR operation. The typical intermittent periods are as shown in Table 2.4. The basic principal for this intermittent filtration and backwashing is to alternate periods of filtration at normal TMP and relatively low velocity, with periods of membrane backwashing performed with high crossflow velocity (Defrance & Jaffrin, 1999a). It has been found that MF rather than UF requires backflushing. A static period is sufficient for fouling limitation of UF filtration (Albasi *et al.*, 2002),
- (iii) By alternating 15 minutes of filtration and air flushing respectively. According to Visvanathan *et al.*, (1997), this method can produce a stable flux and create an essential critical erosion stress for cake

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removal and also minimize energy consumption. Chae *et al.*, (2006) found that air backwashing was more effective than air scouring in terms of irreversible fouling control,

- (iv) The backshock technique can also be used in membrane fouling control (Barreiros *et al.*, 1998). The backshock is a membrane cleaning technique with an effective backflush of less than 0.1 second with backflush intervals between 1 to 5 seconds and
- (v) Increasing the crossflow and pH or washing with clean water can also help to accelerate the cake deplORIZATION rate to recover fouled membrane (Chen *et al.*, 1997).

Table 2.4: Proposed intermittent suction and backwash

Intermittent suction and backwashing		References
Suction (min)	Backwashing (sec)	
16	30	Côtê <i>et al.</i> , 1997
5	15	Côtê <i>et al.</i> , 1997
5	15	Bouhabila <i>et al.</i> , 2001
10	35	Rosenberger <i>et al.</i> , 2002
30	15	Psoch <i>et al.</i> , 2006
8.3	15	Jeison and Lier, 2006

The above physical cleaning methods are only applied to submerged MBR systems. As for sidestream operation, methods such as depressurization and intermittent filtration can be used to remove the attached biomass on the membrane surface. Both cleaning efficiencies would decrease with operating time. This can be attributed not only to the compaction of the attached biomass but also to the precipitation of less soluble inorganic species at the membrane surface (Choo & Lee, 1996). Alternatively, aeration can be used to help to reduce the fouling in the external hollow fibre membrane module if the feed stream goes through the inner lumen as in the case of an “inside out” operation. Bubbles can be used to create a scouring effect in the lumen, which can reduce cake deposition on the membrane surface. This will be discussed more thoroughly in one of the below sections.

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In the case where the membrane cannot be recovered by physical cleaning methods such as backwashing, a chemical cleaning stage can be performed as an alternative option (Xu *et al.*, 1995; Wisniewski & Grasmick, 1998; Shim *et al.*, 2002; Rosenberger *et al.*, 2002). A combination of physical or mechanical and chemical cleaning could achieve almost complete regeneration of the fouled membrane (Maartens *et al.*, 2002; Lim and Bai, 2003). These methods include (i) clean water rinsing follow by an alkaline solution, (ii) water rinsing and cleaning with an acid in solution (Ognier *et al.*, 2002a) and (iii) chemical (acidic detergent) cleaning, followed by a backwash (Rahman *et al.*, 2006). The application of chemical cleaning would only be suggested as a last option if physical cleaning is futile in recovering the permeability of the membrane. The usage of chemicals would increase the operational cost of the MBR and create the additional chemical waste. The chemicals may harmful to the biomass and cause instability to the biomass growth. Some of the membrane materials are sensitive to the certain chemical solution; the inappropriate use of chemicals as cleaning agents may damage or reduce the life-span of the membrane.

2.5.5. Geometric design of the bioreactors

The discussions above are regarding using aeration intensities, intermitten filtration, backwash and membrane cleaning methods to control membrane fouling. Apart from that, an effective design of the bioreactor of MBR will have the ability to prolong the life of the membrane. Four basic aspects which are needed to be taken into consideration in designing the bioreactor include (i) the shape of the bioreactor, (ii) the location and design of the aerator, (iii) the location where the membrane can get the best aeration distribution and (iv) type of membrane/module used in the MBR (e.g. hollow fiber, flat sheet, tubular).

The bioreactor is normally rectangular or cylindrical. The shape of the bioreactor used will determine the type of the aerator to be used. One function of the aerator is to produce an aeration to provide oxygen to the biomass. The other function (often a different aerator) is to provide bubbles that create a shear force on the membrane

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surface to prevent cake accumulation. A few researchers have studied this and found that the relationship between the riser and downcomer area in the bioreactor of the MBR with flat sheet membranes is important (the same would apply to hollow fibre bundles). A higher ratio of down-comer to riser area in the bioreactor has shown better filtration over a long-term operation (Liu *et al.*, 2000; Shim *et al.*, 2002). This is because the membrane modules, which are positioned over a smaller floor riser area, can experience intensified air-flow and induced liquid flow and subsequently improved cake removal efficiency without increasing the air sparging rate (Ueda *et al.*, 1996; Shim *et al.*, 2002).

Apart from protecting the membrane from cake accumulation on the surface, the aeration distribution should have the capability to ensure the mixed liquor in the bioreactor is well mixed. Dead zones should be avoided to prevent the accumulation of anoxic sludge in the bioreactor. It was noted earlier (section 2.2.2) that low D.O condition can cause an increase in smaller particles. Failing to prevent the dead zones in the bioreactor may cause instability of the MBRs system because the anoxic sludge may contribute more fine foulants (SMPs, fine colloids and particles) to the supernatant. Investigations have revealed that the fouling rate for the anoxic MBR was five times higher compared to the aerobic MBR (Yun *et al.*, 2006). As a result, for a good bioreactor geometry, the bubbles produced from the aerator should be easily accessible.

2.6 Biologically activated carbon sludge (BAC)

Powder activated carbon (PAC) is an absorbent in powder form. PAC is porous and has strong adsorption characteristics and these properties have made PAC suitable for use in wastewater treatment to produce high quality permeate through adsorption of the pollutants. One example is that PAC has been added into the activated sludge wastewater treatment process (Dalmacija *et al.*, 1995; Orsharsky and Narkis, 1997; Pala and Tokat, 2002; Cecen *et al.*, 2003). Apart from PAC, granular activated carbon (GAC) is also widely applied in activated sludge systems (Mochidzuki *et al.*, 1998; Mochidzuki *et al.*, 1999), such as (i) biological granular

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activated carbon (BGAC) formed by the combination of GAC and activated sludge, (ii) carbon beds (Scholz *et al.*, 1997), (iii) the BGAC aerated stirred tank (Walker *et al.*, 1999), (iv) GAC with biofilm system (Zhao *et al.*, 1999), (v) BGAC lifted moving bed (Lin *et al.*, 2000) and (iv) the BGAC column (Sirotkin *et al.*, 2001). Applications of PAC and GAC to biological wastewater treatment are summarized in Table 2.5, which includes both membrane and non-membrane systems.

Table 2.5: Hybrid system consists of biodegradation and adsorption processes

Type of hybrid system	PAC concentration	Type of wastewater for treatment	References
PAC + activated sludge + microfiltration	20 g/L	Wastewater contains refractory organic compounds	Seo <i>et al.</i> , 1996
PAC + activated sludge + ultrafiltration	1% (by weight, approximately 10g/L)) of total volume of wastewater in the bioreactor	Wastewater contains high strength landfill leachate	Pirbazari <i>et al.</i> , 1996
PAC + anaerobic sludge + microfiltration	5 g/L	Synthetic wastewater	Park <i>et al.</i> , 1999
PAC + activated sludge + microfiltration/ ultrafiltration	5 g/L	Tap water contains trace organics	Dosoretz and Bøddeker, 2004
PAC + activated sludge + microfiltration	20 – 40 g/L	Drinking water contains disinfection by products	Seo <i>et al.</i> , 2004
PAC + activated sludge +	80 g/L	Domestic wastewater	Seo <i>et al.</i> ,

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microfiltration			2004
PAC + activated sludge + microfiltration	1.2 g/L	Synthetic wastewater	Li <i>et al.</i> , 2005
PAC + activated sludge + ultrafiltration	500 mg/L	Municipal wastewater	Liu <i>et al.</i> , 2005a
PAC + activated sludge + microfiltration	2 g/L	EPS solution	Fang <i>et al.</i> , 2006
PAC + activated sludge (SBR system)	143 mg/L	Wastewater contains Cu (II) and Cd (II)	Lim <i>et al.</i> , 2002
PAC + activated sludge	200 mg/L	Wastewater contains high- salinity oil-field brine	Dalmacija <i>et al.</i> , 1995
PAC + activated sludge	(0.74 – 3.68) g/L	Wastewater contains phenol or aniline	Orshansky and Narkis, 1997
Biological granular activated carbon (BGAC) carbon bed	Filtrisorb 400 (particle diameter:0.85-1.2 mm) used to fill up the carbon bed	Wastewater contains glycerol	Scholz <i>et al.</i> , 1997
Granular activated carbon (GAC) + activated sludge, Zeolite + activated sludge	Not specified	Plating industrial wastewater	Mochidzuki <i>et al.</i> , 1998
PAC + activated	400 mg/L	Wastewater contains color	Pala and

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sludge		from textile industry	Tokat, 2002
GAC + activated sludge	15 g GAC used to fill carbon bed	Ethylenediaminetetraacetic acid (EDTA) wastewater	Mochidzuki <i>et al.</i> , 1999
BGAC aerated stirred tank	Filtrisorb 400, 1.67 g/L	Industrial wastewater contains dye	Walker <i>et al.</i> , 1999
GAC + biofilm	Filtrisorb 400 filled up 14.7 L fluidized bed reactor	Wastewater contains toluene	Zhao <i>et al.</i> , 1999
Lifted moving bed biological activated carbon	PAC : 1.5 g/L GAC: 1530 mg used for fixed bed	Petrochemical wastewater and synthetic wastewater	Lin <i>et al.</i> , 2000
BGAC column	GAC (SKT-3): 2.5 g/L	Non-ionogenic surfactants (NISS) wastewater	Sirotkin <i>et al.</i> , 2001
PAC + activated sludge, Zeolite + activated sludge	Varying from 10- 60 g	Inhibitory materials wastewater	Park <i>et al.</i> , 2003
PAC + activated sludge	1.0 – 3.0 g/L	Landfill leachate wastewater	Cecen <i>et al.</i> , 2003

In the activated sludge environment, a layer of biofilm should form on the PAC surface to form biologically activated carbon (BAC) sludge (Liu *et al.*, 2005a; Fang *et al.*, 2006). The biofilm would biodegrade the pollutants previously adsorbed by the PAC, leading to the simultaneous adsorption and biodegradation processes in the activated sludge with added PAC treatment rather than just the biological means (Pirbazari *et al.*, 1996; Orshanky and Narkis, 1997; Scholz *et al.*, 1997; Lim *et al.*, 2002; Çeçen *et al.*, 2003; Seo, *et al.*, 2004; Seo, *et al.*, 2004a; Liu *et al.*, 2005).

One of the advantages of BAC in wastewater treatment is that the biofilm on the PAC consists of immobilized and acclimatized bacteria (Dalmacija *et al.*, 1995). The

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biofilm can help to bioregenerate the PAC which is saturated with pollutants and decayed cells (Scholz and Martin, 1997; Li *et al.*, 2005). Apart from that, the succession of bacteria in the biofilm ecosystem can also enhance the performance of BAC in pollutant removal (Scholz *et al.*, 1997; Mochidzuki *et al.*, 1998; Lin *et al.*, 2000). The other advantages of using BAC sludge in wastewater treatment are listed in Table 2.6.

Table 2.6: Advantages of using BAC sludge in wastewater treatment

Advantages	Reference
<ul style="list-style-type: none">- Positive impact on substrate removal and sludge dewaterability.- PAC can act as a nucleus to help floc formation.	Cecen <i>et al.</i> , 2003
<ul style="list-style-type: none">- BAC is better than GAC alone in terms of system operation time and organic matter removal efficiency.- BAC bed clogging can be avoided.- Produces low turbidity and suspended solids permeate.	Scholz <i>et al.</i> , 1997
<ul style="list-style-type: none">- BAC system has the ability to reduce the negative effect of heavy metal ions by the activated carbon adsorption.	Mochidzuki <i>et al.</i> , 1998
<ul style="list-style-type: none">- BAC has better performance in dye removal rate compared to conventional wastewater system and combination of individual conventional GAC and biological processes.	Walker <i>et al.</i> , 1999
<ul style="list-style-type: none">- Improve activated sludge filterability	Pirbazari <i>et al.</i> , 1996; Kim <i>et al.</i> , 1998; Park <i>et al.</i> , 1999; Li <i>et al.</i> , 2005; Liu <i>et al.</i> , 2005; Ng <i>et al.</i> , 2006
<ul style="list-style-type: none">- BAC system can last longer than that of GAC system in terms of adsorption cycle.	Walker and Weatherley, 1999; Lin <i>et al.</i> , 2000

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However in long-term operation, the characteristic of simultaneous adsorption and biodegradation of pollutant of BAC would probably be reduced. This is due to the loss of the adsorption capacity of the BAC. Factors causing the reduction in the ability of bioregeneration of PAC adsorption capacity are the difficulty of bacteria to desorb the adsorbed species (pollutants or products of microbial biodegradation) and the fillings of mesopores with the products of dead microbial cells (Sirotkin *et al.*, 2001). Zhao *et al.*, (1999) also agree that the loss of adsorption capacity of PAC is due to the bioactivity. This seems to indicate that the replacement of aging PAC having lost adsorption capacity is important to keep the properties of simultaneous adsorption and biodegradation of BAC.

Several studies of the hybrid MBR with PAC have reported that the addition of PAC could enhance performance of the MBR system (Seo *et al.*, 1996; Pirbazari *et al.*, 1996; Park *et al.*, 1999; Seo *et al.*, 2004; Seo *et al.*, 2004a; Li *et al.*, 2005; Fang *et al.*, 2006; Fan *et al.*, 2006). Researchers tend to agree that the improved performance of the MBR with the addition of PAC was due to the adsorptive effect that reduces extracellular polymeric substances (EPSs) in the floc (Kim *et al.*, 1988) and the bulk liquid (Fang *et al.*, 2006; Li *et al.*, 2005) and other fine foulants such as TOC (Pirbazari *et al.*, 1996; Seo *et al.*, 1996; Seo *et al.*, 2004), DOC (Seo *et al.*, 2004a), fine colloids (Li *et al.*, 2005), soluble metabolic products (SMPs) (Liu *et al.*, 2005a), refractory organic matter (Seo *et al.*, 1996), COD (Park *et al.*, 1999) and trace organic (Dosoretz, *et al.*, 2004) in the supernatant. Seo *et al.*, (2004) found that most of the substances with molecular weight cut off < 1000 could be eliminated by adsorption and biodegradation and those above 1000 were gradually degraded by microorganisms of BAC during extended contact. Some researchers (Pirbazari *et al.*, 1996; Kim *et al.*, 1998; Li *et al.*, 2005; Liu *et al.*, 2005a) suggested that the formation of BAC with high porosity and low compressibility was another reason that PAC could improve fouling control. However the advantage of high porosity and low compressibility may only be significant if cake formation is allowed to occur which may not be a typical scenario for MBRs. Others noticed that the BAC could act as a 'precoat' permeable layer on the membrane surface to

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prevent membrane pore restriction (Pirbazari *et al.*, 1996; Seo *et al.*, 1996; Li *et al.*, 2005; Liu *et al.*, 2005a). However, only if the flux was fixed above the critical flux of the BAC, would deposition of BAC occur, and this itself could cause other fouling issues.

Apart from adsorption and protective layer formation, Several authors have suggested that PAC in a MBR could depolarize and remove fine particles accumulated on the membrane surface through scouring effects or enhanced fluid turbulence in the presence of bubbling (Pirbazari *et al.*, 1996; Park *et al.*, 1999; Dosoretz *et al.*, 2004; Li *et al.*, 2005). None of these authors performed specific experiments to test this hypothesis, and this has been examined in chapter 6 of this thesis. However, high loadings of PAC may be needed if PAC is used as a ‘scouring agent’ and this needs to be optimized. In summary, from the literature, the reported functions of PAC in enhancing MBR performance could include (i) adsorption, (ii) simultaneous adsorption, decomposition and biodegradation, (iii) protective permeable layer formation with low specific cake resistance (SCR) and (iv) scouring effects. However, it is still ambiguous which mechanism plays the primary role in fouling control. This suggests that further work needs to be carried out to classify the mechanisms involved in the MBR (BAC), and to optimize the enhancement effect.

One of the objectives of this study is to investigate the functions of PAC and the mechanisms involved in improving fouling control of MBRs. We have attempted to apply a protocol to give a fair comparison of those functional mechanisms. The effectiveness of each mechanism in decreasing fouling rate has been carefully evaluated. The properties and filtration characteristics of AS and BACs have also been measured and compared in both short-term tests and in long-term continuous operation runs in parallel at SRTs of 10days, 30 days and long SRT.

2.7 Comparison of MBR design concepts

This section discusses some of the alternative design concepts of the MBR process. For the conventional MBR, the designs consist of submerged and sidestream modes (popular in the few early MBRs, in the 1970s and 80s). One alternative design of sidestream MBR is to aerate the lumen (inner part) of the fibres of an external membrane module as a so called ‘two-phase’ operation process. There is also an attempt to use membranes in sequencing batch reactors (SBR) design to mimic the MBR process. The most recent trend involves using adsorbents with MBRs as discussed above and it was found that under some conditions the design performed well in membrane fouling control. The above design concepts, except for the previously discussed MBR (BAC), are described below.

2.7.1. Submerged and sidestream MBRs operation modes

MBRs operation can be divided into sidestream and submerged operations as shown in Figure 2.3. In a sidestream MBR, the membrane is situated externally from the bioreactor and traditionally the MLSS is pumped at relatively high crossflow to minimize fouling. In a submerged MBR, the membrane is directly placed into the bioreactor. For membrane maintenance, the position of the membrane module in the sidestream MBR is relatively easy compared to the submerged MBR. Also, the external membrane module may be more compact than that of an immersed membrane module (Heran *et al.*, 2006). However, sludge filterability is better in submerged MBRs than that of high crossflow sidestream systems, presumably because of floc breakup under shear. It is also more suitable to be used in high concentration MLSS and lower food to microorganism (F/M) ratio applications (Rosenberger & Kraume, 2002). Submerged systems tend to be more economic than sidestream membrane systems in terms of energy consumption (Rosenberger *et al.*, 2002).

The introduction of air injection into the sidestream membrane module helps to prevent cake formation on the membrane surface to have better membrane fouling control (Heran *et al.*, 2006). These authors have developed a new sidestream MBR

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without using the recycling pump where the suspension circulation was naturally induced by air injection at the bottom of the membrane module. This should be more economic and may avoid the breakage of the biomass floc due to high shear stress of the recycling pump.

Figure 2.3: MBRs operation modes

2.7.2. Two-phase flow in membrane processes

From the preceding sections, submerged MBRs with bubbling seem to be the preferred systems due to lower energy consumption. However, as shown by Heran *et al.*, (2006), it may be possible to operate a sidestream MBR system with energy consumption similar to that of a submerged MBR when bubbling is used to control concentration polarisation of the membrane. This can be done by flowing the mixed liquor feed through the inner lumen of hollow fibres with the injection of air bubbles or so-called two-phase flow inside the lumen of hollow fibres. Two-phase flow inside tubes and fibres can control cake deposition on membrane surfaces and subsequently slow down the TMP increase (Cabassud *et al.*, 1997; Laborie *et al.*, 1998; Laborie *et al.*, 1999; Cabassud *et al.*, 2001; Mahmud *et al.*, 2004; Psoch and Schiewer, 2006). Air sparging in the membrane lumen can remove concentration polarization effects to prevent membrane fouling (Cabassud *et al.*, 2001; Laborie *et al.*, 1998; Laborie *et al.*, 1999). Mixing and turbulence created by the bubbles in the

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liquid phase is the main flux enhancement factor (Cabassud *et al.*, 2001). The membrane surface is subjected to alternating positive and negative shear stresses, which are expected to prevent filtered particles from settling and can improve the membrane filtration mass transfer (Cabassud *et al.*, 1997). In other words, two-phase flow is a good method for flux enhancement in sidestream membrane filtration. Even at low superficial air velocity (0.1 m/s), flux enhancement can be significant (Cabassud *et al.*, 1997). Energy consumption reduction can also be achieved at low superficial air velocities (Laborie *et al.*, 1998). Indeed, at the point where superficial air velocity exceeds the optimal level, the flux would not be further enhanced but decreases slowly and the system becomes uneconomic from the energy point of view (Laborie *et al.*, 1998). In a two-phase process, slug flow plays an important role to induce high wall shear stresses which can reduce the particle deposition on the membrane surface (Cabassud *et al.*, 1997; Psoch and Schiewer, 2006). In addition, applying backflushing to control internal fouling together with air slug flow was found to be very effective in reversible and irreversible fouling control (Psoch and Schiewer, 2006). These authors also noted that though backflushing may cause the loss of productivity this loss could be minimal.

In two-phase flow, a pump and aerator are needed to recirculate the mixed liquor and produce air bubbles through the inner lumens of the hollow fibres. This means that the operation cost of running MBRs is increased due to the extra pump and aerator. To reduce the cost, the operating of the two-phase flow must be at an optimum flow ratio of mixed liquor to the air bubble. The new sidestream MBR with natural recycling flow without using a recycling pump designed by Heran *et al.*, (2006) may help to reduce the cost induced by extra pump.

2.7.3. Comparison of supernatant and mixed liquor filtration performance

In MBRs, the direct contact of the membrane with mixed liquor causes a high possibility of cake accumulation on the membrane surface. In order to prevent such a problem, the filtration of the supernatant only rather than the mixed liquor could be a benefit to MBRs in terms of reversible membrane fouling control. A limited

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number of researchers have been looking into this area such as the filtration of the effluent produced from the sequencing batch reactor (SBR). The operation of the SBR consists of five stages which are fill, react, settle, decant and idle processes. However, the SBR has a potential problem as poor clarification may cause high turbidity in the effluent but it was found that, this problem can be solved if a membrane is used in SBR system (Parameshwaran, *et al.*, 2001; Kang *et al.*, 2003). Bae *et al.*, (2003) have observed that a combination of the SBR and membranes has a high performance such as (i) suspended solids free effluent can be achieved using membrane filtration, (ii) the effluent quality in terms of BOD removal was as high as 97 to 98 % and (iii) it was found that a diffuser-attached module design in SBR system could be operating for 110 days with only one membrane wash without stopping to the system operation. This seems to indicate that in this case the filtration of the supernatant was more beneficial compared to the mixed liquor filtration. The filtration of the supernatant could avoid the problem of cake deposition on the membrane surface. However, the concern in the filtration of supernatant is the potential for pore blocking or plugging caused by fine colloids or particles in soluble form.

It is possible that failure to eliminate the fine foulants will cause the performance of the supernatant filtration be worse than that of the mixed liquor filtration. Chang *et al.*, (2005), have observed this phenomenon, moreover, they also believe that the worse performance of the supernatant filtration compared to mixed liquor is due to the small particles in the supernatant. The success in minimising the fine foulants or small particles from the supernatant before going through the membrane filtration is believed to prolong the life of the membrane significantly. One objective of this project is to look into the possibility of separating the MBR into two compartments. The first compartment is used to retain the mixed liquor and the adjacent compartment for retaining the supernatant in which the membrane is submerged. The purpose of this design concept is to avoid direct contact of the mixed liquor with the membrane in order to reduce the membrane fouling problem. Therefore one aspect of this study is to examine this two compartment concept and to establish the conditions where it may be beneficial. For example, incorporation of the BAC

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concept in the first tank to reduce the level of microfoulants to be carried over to the supernatant chamber has been evaluated.

2.7.4. Alternative designs for MBRs

Apart from the methods discussed above, others alternative MBR designs have been investigated to improve the performance of conventional MBR. For example,

- (i) Li *et al.*, (2005) found that the aerobic granule in the MBR was able to improve the membrane permeability up to 50%.
- (ii) Sequencing batch membrane bioreactor (SBMBR) had a better fouling control compared to a conventional MBR (Zhang *et al.*, 2006). However, Laitinen *et al.*, (2006) found that MBR performed better than that of SBMBR in terms of SS retention and retention of BOD, ammonia nitrogen and phosphorus.
- (iii) Recently, Yoon and Collins, (2006) have found that the addition of cationic polymer could enhance the fouling control.
- (iv) The combination of ozone pretreatment with MBR was found effective in producing excellent permeate quality and had a small sludge yield (He *et al.*, 2006).
- (v) Hybrid MBRs with porous and flexible suspended carriers showed improved filtration performance (Yang *et al.*, 2006).

2.8 Conclusions

From the literature review, it is evident that the current MBR design processes are not optimized or perfected. There are still many limitations in the conventional MBR that can lead to severe membrane fouling, such as direct contact of the membrane with high concentrations of MLSS, TOC and EPSs. High concentrations

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of MLSS will also have a high viscosity, thus limiting the air sparging in removal of cake deposition on the surface of the membrane. Aeration used in the submerged MBR is also not particularly effective in distributing the bubbles into the fibre bundle to reach each membrane fibre. However, high recirculation is needed in a sidestream MBR process which is not economic and can easily cause floc breakage.

For the conventional practice, to maintain the performance of MBRs, sludge wastage is also needed from time to time though the MBR can be operated at a high SRT. All the above limitations will incur significant maintenance costs. If all of the above limitations can be eliminated or minimised, then the reduction of MBR operating costs at a large scale could be possible. As a result, some of the recent research directions aim to address these issues, for example, to solve the problems of the sidestream MBR, a new design of the sidestream MBR has avoided the use of a recycling pump (Heran *et al.*, 2006). Hopefully, the new design can be tested at larger scale to treat real wastewater to evaluate its performance. Several researchers have also investigated the performance of MBRs with prolonged SRT as reported in section 2.4.3. Apart from that, some attention has been focused on membrane fouling control such as (i) identifying the main foulant components causing membrane fouling, (ii) optimization of the MBR operating conditions (SRT, HRT, flux) to limit these components and (iii) hybrid MBRs such as the MBR (BAC), SBMBR, granulation MBR, suspended carrier MBR and cationic polymer MBR.

In this project, MBR with addition of PAC and a two-compartment MBR have been carefully investigated. The two-compartment MBR design has been evaluated because it combines features of the submerged and external loop systems as well as the SBR hybrid membrane process to counter the above limitations. This involves submerged membranes in a separate chamber with supernatant only adjacent to the mixed liquor bioreactor. The MLSS flow from the bioreactor to the supernatant chamber at the bottom of the bioreactor is recirculated back to the bioreactor by a return sludge pump which can be operated intermittently by air.

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With this arrangement a two-compartment MBR design with mixed liquor in the first chamber and supernatant in the adjacent MBR chamber can be realized. This avoids direct contact of the membrane with MLSS. However, as discussed in detail in Chapter 8, comparison of the supernatant and mixed liquor filtration run in parallel showed that the supernatant filtration is not necessarily beneficial compared to the mixed liquor filtration in terms of membrane fouling. This may be due to fine particles in the supernatant causing the supernatant filtration to be worse than that of mixed liquor filtration (see chapter 8). In addition, the MLSS biomass floc in the bioreactor may help to protect the membrane and to detach the fine foulants on the membrane surface back to the bulk liquid through the collision. However, further comparison of the two compartment system with the supernatant and mixed liquor is still worthy of further investigations. In addition, using PAC to retain the fine foulants (EPSs, TOC, SMP) in the bulk liquid of the biomass was also carried out. It was believed that PAC can support the biomass to forming dense flocs thus reducing the EPS and TOC flowing into adjacent chamber but the results were not as anticipated (see chapter 8).

The addition of adsorbent such as PAC into the single compartment MBR may provide a number of benefits. Most importantly, the PAC may act as a sink for organic species, including substrate, EPSs and SMPs that may be further degraded and simultaneously can be limited in their interaction with the membrane. It is also possible that PAC will form a dynamic protective layer which consists of BAC on the membrane surface. The BAC layer may protect the membrane by preventing the fine foulants (macromolecules and colloids) from blocking and plugging the membrane during filtration through adsorption. Further, it is also possible that the BAC floc in suspension will have greater density than conventional AS and that shear-induced collision between the BAC flocs and membrane surface could aid in the removal of foulants. Obviously the role of BAC as a protective layer or scouring agent will depend on the imposed flux, the freshness of the BAC and aeration intensity. The study of the MBR (BAC) has been the major of this thesis. It includes (i) to investigate when does PAC help to improve the fouling control, (ii) to evaluate the

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mechanisms involved in enhancing filtration performance by PAC in the MBR and (iii) optimization of the MBR (BAC).

Thus, the work in this thesis is based on the hypothesis that the addition of adsorbent into MBRs should be beneficial in terms of membrane fouling control. However, the cost of the adsorbent should be taken into consideration. The saturated adsorbent in the MBRs may need to be replaced to keep the adsorption effect of the supported biomass. In order to maintain a low operation cost of MBRs added adsorbent, the ageing adsorbent should be recycled. Thus, while the performance of MBRs with added adsorbent is likely to be better than that of conventional MBRs, the successful recycling of adsorbent will incur an added operation cost. This needs further investigation.

CHAPTER 3 MATERIALS AND METHODOLOGIES

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3.1 Introduction

The purpose of this chapter is to present the detailed experimental design, methods and materials used for the performance comparison between (i) the conventional MBR and the MBR with added PAC (hereafter known as MBR (AS) and MBR (BAC) respectively), (ii) MBR (AS)s with different SRT, (iii) MLSS or supernatant filtration in the two-compartment MBR, (iii) MBR (AS) with the MBR incorporating polymer beads (hereafter known as MBR (polymer bead)) and (iv) MBR (BAC)s with different SRT and PAC size distributions. The information on the reactors, operational conditions and the type of membrane from the different membrane manufacturing companies involved in the results and discussions chapters (chapter 4 to 8) are listed in Tables 3.1-3.5.

the broken floc would occur. These foulants have the tendency to block and plug the pores of membranes during filtration. Table 2.1a shows the effect of organic fines on membrane fouling.

Table 2.1a: Type of foulants and its effect on membrane filtration

Type of membrane foulant	Effect of foulant on the membrane	References
1. Biopolymer clusters (BPC): Organic substances mainly consist of SMP and colloidal organic substances	a. BPC is normally found within the pores of the sludge cake formed on the membrane surface which cause the high filtration resistance of the cake sludge.	Wang et al., 2007; Wang & Li, 2007
2. Colloidal/EPS: Proteins, polysaccharide matters and lipids.	a. The breakage of microbial floc promotes the release of colloidal and soluble components and thus increase the accumulation of the sticky foulants on the membrane surface and cause membrane fouling.	Meng et al., 2007
3. Colloidal TOC and/or soluble EPS (soluble carbohydrates and humic substances)	a. The attachment of the sticky foulants on the membrane surface responsible for membrane fouling and the reduction of the critical flux.	Fan et al., 2006
4. Polysaccharides and other biopolymers	a. Accumulation of the foulants on the membrane decreases the membrane permeability. b. Suspension form biopolymers increases the viscosity of the MLSS and reduces membrane filterability. c. Biopolymers on the membrane surface encourage biomass	Chu & Li, 2004

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	attachment and aggravate the deposition of the sludge cake on the membrane surface.	
5. Biofilm formation (fouling layer made up of microbial cells covered with EPS)	<p>a. Biofouling is the formation of mature biofilm layer and/or the accumulation of EPS on the membrane surface.</p> <p>b. SMP would provide organic substrate to the growth of biofilm on the membrane surface.</p> <p>c. EPS would also permit microbial cells to adsorb onto the membrane surface to promote biofilm growth.</p>	Ng et al., 2006
6. Biofilm and EPS	<p>a. The growth of biofilm on the membrane was the main factor of membrane fouling.</p> <p>b. EPS in biofilm is the key membrane foulants.</p>	Yun et al., 2006
7. EPS	<p>a. . Accumulation of the EPS on the membrane surface was the major component to the increase of the membrane resistance.</p>	Chang and Lee, 1998; Naogaka, et al., 1996

Table 3.1: Information on the reactors involved in the study in Chapter 4

Chapter / Section	Name of MBR	Operation mode (c/n or batch feed)	Working volume (L)	Type of MLSS	Organic loading rate (g/L/d)	SRT (day)	HRT (hr/day)	Type of membrane
Chapter 4 (section 4.1)	MBR (AS)	c/n	20	AS	0.45 ± 0.02	30	8.3 hr	Cleanseas
Chapter 4 (section 4.1)	MBR (BAC)	c/n	20	AS+PAC	0.45 ± 0.02	30	8.3 hr	Cleanseas
Chapter 4 (section 4.2)	MBR (aged AS)	Batch	20	AS	0.10 ± 0.01	p/i*	15 days	Blue Star
Chapter 4 (section 4.2)	MBR (aged BAC)	Batch	20	AS+PAC	0.10 ± 0.01	p/i	15 days	Blue Star
Chapter 4 (section 4.2)	MBR (aging AS)	c/n	20	AS	0.45 ± 0.02	p/i	8.3 hr	Cleanseas
Chapter 4 (section 4.2)	MBR (aging BAC)	c/n	20	AS+PAC	0.45 ± 0.02	p/i	8.3 hr	Cleanseas

p/i*: (Pseudo infinity) no sludge wastage except for samples; c/n*: continuous

Table 3.2: Information on the reactors involved in the study in Chapter 5

Chapter / Section	Name of MBR	Operation mode (c/n or batch feed)	Working volume (L)	Type of MLSS	Organic loading rate (g/L/d)	SRT (day)	HRT (hr/day)	Type of membrane
Chapter 5	MBR (AS)	Batch	2	AS	0.37 ± 0.01	30	4 days	Cleanseas
Chapter 5	MBR (BAC)	Batch	2	AS + 1 g/L PAC	0.37 ± 0.01	30	4 days	Cleanseas
Chapter 5	MBR (BAC)	Batch	2	AS + 3 g/L PAC	0.37 ± 0.01	30	4 days	Cleanseas
Chapter 5	MBR (BAC)	Batch	2	AS + 5 g/L PAC	0.37 ± 0.01	30	4 days	Cleanseas

c/n*: continuous

Table 3.3: Information on the reactors involved in the study in Chapter 6

Chapter / Section	Name of MBR	Operation mode (c/n* or batch feed)	Working volume (L)	Type of MLSS	Organic loading rate (g/L/d)	SRT (day)	HRT (hr / day)	Type of membrane
Chapter 6	MBR (AS)	Batch	2	AS	0.53 ± 0.02	10	2.8 days	Blue Star
Chapter 6	MBR (BAC)	Batch	2	AS + 1 g/L PAC	0.53 ± 0.02	10	2.8 days	Blue Star
Chapter 6	MBR (BAC)	Batch	2	AS + 3 g/L PAC	0.53 ± 0.02	10	2.8 days	Blue Star
Chapter 6	MBR (BAC)	Batch	2	AS + 5 g/L PAC	0.53 ± 0.02	10	2.8 days	Blue Star
Chapter 6	MBR (AS)	c/n	20	AS	0.45 ± 0.02	10	8.3 hr	Blue Star
Chapter 6	MBR (BAC)	c/n	20	AS+PAC	0.45 ± 0.02	10	8.3 hr	Blue Star

c/n*: continuous

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Table 3.4: Information on the reactors involved in the study in Chapter 7

Chapter / Section	Name of MBR	Operation mode (c/n* or batch feed)	Working volume (L)	Type of MLSS	Organic loading rate (g/L/d)	SRT (day)	HRT (hr/day)	Type of membrane
Chapter 7 (section 7.1)	MBR (AS)	Batch	2	AS	0.26 ± 0.01	10	5.7 days	Blue Star
Chapter 7 (section 7.1)	MBR (fine)	Batch	2	AS+PAC	0.26 ± 0.01	10	5.7 days	Blue Star
Chapter 7 (section 7.1)	MBR (medium)	Batch	2	AS+PAC	0.26 ± 0.01	10	5.7 days	Blue Star
Chapter 7 (section 7.2)	MBR (10d)	Batch	3.5	AS	0.42 ± 0.01	10	3.5 days	Blue Star
Chapter 7 (section 7.2)	MBR (30d)	Batch	3.5	AS	0.42 ± 0.01	30	3.5 days	Blue Star
Chapter 7 (section 7.3)	MBR (BAC _{10d})	Batch	3.5	AS+PAC	0.42 ± 0.01	10	3.5 days	Blue Star
Chapter 7 (section 7.3)	MBR (BAC _{30d})	Batch	3.5	AS+PAC	0.42 ± 0.01	30	3.5 days	Blue Star
Chapter 7 (section 7.3)	MBR (BAC _{long})	Batch	3.5	AS+PAC	0.42 ± 0.01	p/i*	3.5 days	Blue Star

c/n*: continuous; p/i*: Pseudo infinity; fine: PAC with relatively smaller size; medium: PAC with relatively medium size

Table 3.5: Information on the reactors involved in the study in Chapter 8

Chapter / Section	Name of MBR	Operation mode (c/n* or batch feed)	Working volume (L)	Type of MLSS	Organic loading rate (g/L/d)	SRT (day)	HRT (hr/day)	Type of membrane
Chapter 8 (section 8.1)	Two-c/p* MBR (MLSS vs S/N)	c/n	20L each for AS sludge chamber and S/N chamber	AS sludge and low SS or S/N	0.45 ± 0.02	p/i*	11.1 hr	Blue Star
Chapter 8 (section 8.2)	Two-c/p MBR (BAC vs S/N*)	c/n	23.5 L each for BAC sludge chamber and S/N chamber	BAC sludge and low SS or S/N	0.45 ± 0.02	10	8.3 hr	Blue Star
Chapter 8 (section 8.3)	MBR (AS)	c/n	20	AS	0.45 ± 0.02	10	8.3 hr	Blue Star
Chapter 8 (section 8.3)	MBR (p/y* bead)	c/n	20	AS + p/y* bead	0.45 ± 0.02	10	8.3 hr	Blue Star
Chapter 8 (section 8.4)	MBR (BAC)	Batch	3.5	AS + PAC	0.42 ± 0.01	10	3.5 days	Blue Star
Chapter 8 (section 8.4)	MBR (P/y* bead)	Batch	3.5	AS + P/y* bead	0.42 ± 0.01	10	3.5 days	Blue Star

* S/N: supernatant; c/n*: continuous; c/p*: compartment; p/y or P/y*: polymer; p/i*: Pseudo infinity

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3.2 Experimental

3.2.1 Materials

PAC (name of brand: Hydrodarco C) was provided by the Norit Company and had about 70-75% organic content and 25-30% ash content. The particle size distribution, shown in Figure 3.1, was measured by a Malvern Mastersizer particle size analyzer. The BET total surface area and micropore volume of the fresh PAC were about 488 m²/g (micropore area: 227.36 m²/g; external surface area; 260.677 m²/g) and 0.104 cm³/g respectively. The pore diameter and volume distribution of fresh PAC is as shown in Table 3.6 and Figure 3.1a. GAC, which was used to produce two different PAC sizes by grinding, was supplied by Kimic Chemitech (S) Pte Ltd. The powdered hollow glass beads (GB) were bought from Dantec Dynamic with three different sizes (D₅₀) of 5, 20 and 50 μm respectively. The polymer bead (styrene-divinylbenzene copolymer) (600 μm; 1.18-1.27 g/cm²) was from Kimic Chemitech (S) Pte Ltd. The filtration characteristics of the AS and BAC were measured in short-term tests in dead-end and crossflow filtration cells (Figure 3.2) using flat-sheet Millipore ultrafiltration Polyethersulfone membranes with a molecular weight cut-off of 50,000Da and 0.2 μm pore size microfiltration membrane respectively. For the long-term submerged MBR trials and some short-term ‘sustainable’ flux tests, Polymen ultrafiltration (UF) hollow fibres (300,000 Da. MWCO), hollow fibre microfiltration Polyacrylonitrile membranes (nominal pore size: 0.5 μm) from China Blue Star Membrane Technology Co., Ltd and hollow fibre microfiltration Polyacrylonitrile membranes from Singaport Cleanseas Ltd were used for filtration comparison.

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Table 3.6: Pore diameter and volume distribution of fresh PAC

Mass percentage distribution of incremental pore volume (%)	Pore diameter range (μm)	Cumulative Mass percentage distribution of incremental pore volume (%)
1.9	0.17209-0.13188	1.9
4.6	0.13188-0.08281	6.5
1.9	0.08281-0.07097	8.4
13.0	0.07097-0.03625	21.4
9.8	0.03625-0.0260	31.2
8.6	0.0260-0.01986	39.8
6.6	0.01986-0.01589	46.4
4.2	0.01589-0.01366	50.6
3.9	0.01366-0.01177	54.4
3.1	0.01177-0.01037	57.5
4.9	0.01037-0.00839	62.4
4.0	0.00839-0.00702	66.4
3.4	0.00702-0.00601	69.7
3.0	0.00601-0.00524	72.8
2.8	0.00524-0.00462	75.6
2.7	0.00462-0.00411	78.3
2.6	0.00411-0.00369	80.9
2.7	0.00369-0.00332	83.6
1.8	0.00332-0.0031	85.4
3.4	0.0031-0.00274	88.8
3.4	0.00274-0.00245	92.2
2.7	0.00245-0.00225	94.9
1.5	0.00225-0.00215	96.4
1.7	0.00215-0.00205	98.1
1.9	0.00205-0.00194	100.0

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Figure 3.1a: The distribution of the surface pore diameter of PAC used in MBRs

Figure 3.2: Schematic diagram of (a) unstirred membrane cell unit (b) crossflow module cell unit for membrane filtration

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3.2.2 Operation of MBRs

The activated sludge used in the MBRs in this study was obtained from a laboratory scale MBR originally seeded from an industrial scale activated sludge plant on Jurong Island, Singapore and acclimatized on synthetic wastewater for about 1-2 years. The composition (wt %) of the synthetic wastewater used as feed to all the MBRs was as follows: peptone (10.5), meat extract (6.6), glucose (52.6), Sodium acetate (26.3), FeSO_4 (2.0) and KH_2SO_4 (2.0) would provide a reasonable C/N/P ratio for bacterial growth had been proved by the (i) high MLSS amounts for the MBRs cultivated at SRT 30 and 10 days and (ii) constant high TOC removal rate. The synthetic substrate was mixed with tap water. The trace nutrients contained in the tap water would also help to avoid nutrient deficiency of biomass and bad carbon removal. The result of measurement of the total carbon contents in the supernatant by a Shimadzu VCSH analyzer also showed that almost 100% of carbon is organic type. In addition, supernatant TOC and permeate TOC for the MBRs cultivated at SRT 10 and 30 days (chapter 7) were found to relatively approximate each other compared to the TOC in the feed solution which indicates most of the TOC is removed due to the biodegradability rather than membrane separation. The comparative filterability results between substrate and mixture of substrate and PAC are as shown in Figure 3.3. Direct contact between the PAC and the substrate had reduced the substrate concentration from 1.82 to 1.67 g/L.

Figure 3.3: Performance comparison of substrate and mixture of substrate and PAC in short-term dead-end filtration

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Two types of operation mode were carried out in this MBR study namely continuous and batch. In the continuous mode MBR, the synthetic wastewater was fed into the bioreactor continuously (non-stop) and the treated permeate was drained out to water drainage over the operation period. For batch mode MBR, the bioreactor was only fed once a day with high concentration (300-400 mg/L) synthetic wastewater. Thus, in order to maintain the working volume of the batch MBR, the permeate was recirculated back to the bioreactor. However, some of the permeate was taken out to control the HRT (hydraulic retention time) of batch MBR and its amount is depend on the HRT and SRT. Three types of different reactor size were used in this MBR study and the dimensions of them are as shown in Table 3.7.

Table 3.7: Dimensions of the reactors used in the MBR studies

Reactor (L)	Diameter (cm)	Length (cm)
2.0	8.0	53.0
3.5	12.5	40.0
20.0	16.0	100.0

Some of the MBRs in the study was purposely cultivated in the ‘stress’ condition such as at long SRT (pseudo infinity) and relatively low organic loading rate (e.g 0.09 ± 0.01 g/d). In the ‘stress’ condition, relatively higher TOC was expected to be produced in the supernatant due to cells lysis and the membranes were inclined to be fouled. In the ‘stress’ condition, the effect of PAC in the membrane fouling control could be observable and tested.

In addition, the difficulty in getting persistent values of TOC in the supernatant was becuase the MBRs in the study were run at different operating conditions as listed below:

- (a) The MBRs were fed continuously or batch fed.
- (b) The MBRs were added with different PAC concentrations.
- (c) The MBRs were cultivated in two-compartment bioreactor.

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- (d) The MBRs were added with bead or polymer bead.
- (e) The MBRs were added with different PAC size and controlled at different SRTs.

To cultivate all the MBRs involved in this study to have consistent TOC in the supernatant which operated at the different conditions would be difficult.

3.2.3 Cultivation of the pure culture bacteria

Adsorption experiments (3.2.4) were done on pure culture bacteria isolated from an MBR. The used of pure culture facilitated observation. Cells were picked up from the R2A agar plates and inoculated in screwcapped test tubes with sterile R2A broth. The composition of R2A was as follows: yeast extract 0.5 g/L, proteose peptone 0.5 g/L, casein hydrolysate 0.5 g/L, glucose 0.5 g/L, soluble starch 0.5 g/L, sodium pyruvate 0.3 g/L, dipotassium hydrogenphate 0.3 g/L and magnesium sulphate 0.05 g/L. The tubes were gently shaken at approximately 150 rpm at a room temperature of about 25°C for 5 days.

3.2.4 Adsorption experiments

Adsorption experiments to assess the performance of PAC in adsorbing planktonic were carried out. In addition, adsorption isotherm studies were also done to evaluate the PAC adsorption capabilities for the TOC and the polysaccharides in the supernatant. The conical flasks containing the supernatant (200 mL) and the different PAC concentrations (0-12 g/L) were made sterile by autoclaving. The durations for the adsorption tests with TOC were 0.5 hour, 1 and 3 days, and for polysaccharides for 90 minutes. The Freundlich capacity constant, K_f , and the Freundlich intensity constant, $1/n$, were determined by analysis of the isotherm data.

3.2.5 Membrane cell and 'sustainable' flux filtration tests

The effects of the BAC and AS were examined in short-term filtration tests by monitoring the long-term membrane performance. The purpose of this was to establish if short-term tests were qualitatively useful predictors of long-term performance in this system. The short-term tests involved an unstirred dead-end cell (volume: 140 mL; membrane area (A) = $1.26 \times 10^{-3} \text{ m}^2$) (Figure 3.2 (a)). In addition

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short-term ‘flux stepping’ tests (see below) were carried out by using a crossflow module cell unit without aeration (Figure 3.2 (b)) and the bioreactor itself (with air-bubbles) with the help of data logging system to measure the ‘sustainable’ flux. Only the ‘sustainable’ flux results in chapter 4 (section 4.13) and chapter 5 (section 5.2) were tested using the crossflow module cell unit. The rest of the ‘sustainable’ flux short-term tests in this thesis were done by using the bioreactor itself. The term ‘sustainable’ flux describes the maximum flux at which the transmembrane pressure (TMP) does not noticeable rise over a period of 15 minutes. It is an approximation to the critical flux (section 2.5.1) of the dominant foulant and is identified by flux-stepping and measuring TMP. The parameters measured in the unstirred cell as characteristic of the fouling tendency of the AS and the BAC were, (i) the specific cake resistance (SCR), (ii) the flux decline profile (flux vs concentration factor), (iii) the irreversible fouling resistance R_{if} (dead-end). The SCR was measured at a fixed pressure of 100 kPa. Flux was measured by weighing the permeate mass with an electronic balance interfaced to a personal computer. The Labview and i-Fix programs were used to data-log the values of feed and permeate pressure and TMP during the unstirred cell and ‘sustainable’ experiments respectively.

Resistances were estimated from the Darcy equation,

$$\text{Resistance, } R = \frac{\Delta P}{\mu J} \quad (3.1)$$

Where J is flux, ΔP is transmembrane pressure (TMP) and μ is permeate (water) viscosity.

The membrane resistances, R_m , were obtained at 100 kPa with Milli-Q water. The total resistance, R_t , was obtained from the filtration flux and the TMP, where,

$$R_t = R_m + R_c + R_{if} \quad (3.2)$$

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The resistance, R_{if} , is the irreversible fouling resistance (caused by pore plugging and restriction) and was obtained at the end of the filtration tests by water washing to remove the cake and then repeating the Milli-Q water test. The reversible cake resistance, R_c , was obtained from equation (3.2) knowing R_m and R_{if} . Estimation of specific cake resistance (SCR) at constant pressure was obtained by filtration at 100 kPa, collecting the data for permeate volume (V) as a function of time (t), and plotting according to the classic cake filtration equation,

$$\frac{t}{V} = \frac{\mu R_m}{A \Delta P} + \frac{\mu C_b \alpha}{2 A^2 \Delta P} V \quad (3.3)$$

The SCR (α) was obtained from the slope of the plot.

3.2.6 SEM observation

The mixed liquor samples were centrifuged at 4,000 rpm for 10 minutes to remove the water. Then the samples were fixed by adding 2% glutaraldehyde. After 2 hours, the samples were washed in 0.10 M sodium cacodylate buffer for 20 minutes three times. Then the samples were dehydrated in series of 10 minute washes in 50%, 70%, 85% and 95% ethanol. After filtering with a 0.2 μ m membrane, the samples were dried with freeze-drier equipment (Christ, Germany). The samples were gold-coated using a sputter gun (SPI supplies, USA). Scanning Electron Microscope (Jeol JSM-5310LV) was used to take SEM images of the membrane surface.

3.2.7 Membrane autopsy

The fouled membranes from 2L MBRs (see section 6.5.2) were cleaned with 0.5L Milli-q water to detach the reversible foulants on the membrane surface. The foulants were analyzed in terms of sludge concentration, EPSs and particle size. The concentrations (C) of the foulants (sludge and EPSs) were calculated according to equation (3.4).

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$$(C) = \frac{C_1 \times 0.5L \text{ (amount of water used to detach the sludge on the membrane surface)}}{2L \text{ (Working volume of MBR)}} \quad (3.)$$

$$C_1 \text{ (g / L)} = \text{Foulants (on membrane surface) in 0.5L (amount of clean water used to detach foulants)}$$

3.2.8 Analytical Methods

TOC was measured by a Shimadzu VCSH analyzer; samples were prefiltered at 0.45 μm prior to analysis. The suspended solids (SS) were measured according to Standard Methods using a GC-50 glass fiber filter (1.2 μm) and an Edwards air vacuum pump. The ROCKLAB grinder was used to grind the GAC into the smaller sizes. The particle sizes of the biomass floc and PAC were measured using a particle size analyzer (Malvern Mastersizer). A BET surface area analyzer (Micromeritics ASAP 2010) was used to measure the surface area and pore size distribution of the PAC.

A Confocal Laser Scanning Microscope (CLSM) (OLYMPUS) (FLUOVIEW) and a live/dead staining kit (Oncogene Research Products) were used for the determination of the dead and live bacteria of AS floc and on the PAC surface. An optical microscope (KEYENCE VH-Z450) was used to observe images of the AS and BAC floc. Scanning Electron Microscope (Jeol JSM-5310LV) was used to take SEM images of the membrane surface. Spectrophotometer (UV/VIS) Jasco (V-550) with ultraviolet (UV) absorbance at a wavelength of 420 nm and total plate count methods were used to determine the planktonic bacteria concentration in the supernatant. The molecular weight distribution of the dissolved solids was determined by using High Performance Size Exclusion Chromatography (HPSEC) (Waters, USA). EPSs (polysaccharide and protein) concentrations (mg/100 mL) detached from the membranes surface were measured with the methods of phenol-sulfuric acid (Dubois *et al.*, 1956) and Bradford reagent for protein with bovine

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serum albumin (BSA) as standard (Bradford, 1976) respectively. A method to determine the PAC concentration in the MBR (BAC) is discussed in Appendix A.

CHAPTER 4 PRELIMINARY COMPARISON OF MBRs WITH AND WITHOUT PAC

CHAPTER 4 PRELIMINARY COMPARISON OF MBRs WITH AND WITHOUT PAC

Synopsis:

The purpose of this chapter is to present some preliminary observations and comparisons of two submerged membrane MBRs, one without added powdered activated carbon (PAC) and one with PAC (note: because the PAC tends to incorporate into the biofloc, it is referred to as biological activated carbon (BAC)). The observations include the relative performances of the two MBRs during start up and approaching steady state as well as operation at an SRT of 30 days and infinity. An important difference between these SRT conditions is that at SRT infinity there was no PAC replacement whereas at SRT 30 there was daily replenishment of a small amount of PAC. Parameters used for comparison of the MBRs included MLSS, TOC loads, floc size, transmembrane pressure (TMP) (the key indicator of fouling) and filtration characteristics of the mixed liquor measured ex-situ. Three parallel samples were collected for the samples. The average result from the three parallel samples was used as data. If the results from the three parallel samples deviated from each other by more than 10%, the samples were tested again to verify the results. In this chapter, a number of mechanisms are proposed to explain the performance differences. These mechanisms are explored more deeply in subsequent chapters (chapters 6, 7 and 8).

4.1 Comparison of 20L MBRs at SRT of 30 days

Performance comparison of the MBR (AS) (conventional MBR) and the MBR (BAC) was obtained in terms of fouling control. The PAC used in this study had the particle size distributions as shown in Figure 3.1 (in chapter 3). The mean particle sizes (D_{50}) of the PAC in terms of volume and number distributions were 24.5 μm and 1.6 μm respectively. Both MBR (AS) and MBR (BAC) were set up in continuous feed mode and operated at a fixed SRT of 30 days, which provided a 3.35wt% PAC top-up daily to the MBR (BAC). The MLSS concentration was 5.5-

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6.5 g/L for the MBR (AS) and 10.50-11.50 g/L for MBR (BAC) (due to the extra 5 g/L of PAC added) at the early stage of filtration as shown in Figure 4.1.

Figure 4.1: MLSS development trend

After running both MBRs for about 3 months, the MLSS concentration increased from 5.5-6.5 g/L to 8.0-10.0 g/L for the MBR (AS) and 10.5-11.5 to 13.0-14.0 g/L for the MBR (BAC). It can be noticed that the growth rate of biomass in the MBR (AS) with suspended growth and biomass in the MBR (BAC) with attached growth were almost similar at SRT 30 days. Effectively the biomass concentrations in both MBRs were the same [Note: in a later chapter (Chapter 6), it is shown that this was not the case for a shorter SRT of 10 days]. In this work, it was found that the concentration of MLSS per se (itself) did not affect the filtration performance of the membranes. From Figure 4.2, it was observed that both MBRs had a better filtration performance (the key parameter used to characterise fouling was the TMP rise) at the higher MLSS concentration (day 80-220) compared to the lower MLSS concentrations during the initial period of filtration (from day 0-80). Furthermore, the MBR (BAC) with approximately 5 g/L more MLSS compared to the MBR (AS) also had the better filtration performance as evidence by the slower TMP rises. This indicates that the performance of the well-aerated MBR (AS) in terms of filtration was mainly affected by the foulants (TOC, EPSs and colloids) in the supernatant rather than the MLSS biomass. However, higher MLSS concentrations tend to have a higher viscosity that may affect the aeration distribution.

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rather than the MLSS biomass. However, higher MLSS concentrations tend to have a higher viscosity that may affect the aeration distribution.

Figure 4.2: Membranes filtration performance in MBR and hybrid PAC MBR

4.1.1 Operation of 20L MBRs at steady and unsteady states

Figure 4.2 (long terms operation test) shows that the operation of both MBRs was more beneficial at steady state rather than unsteady state (based on the great fluctuation of TOC concentration in the supernatant of the mixed liquor shown in Figure 4.3).

Figure 4.3: TOC concentration in the supernatant

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(a)

(b)

Figure 4.2.1: Membranes filtration performance in MBR and hybrid PAC MBR (derived from Figure 4.2) in (a) unsteady state and low MLSS concentration and (b) steady state and high MLSS concentration

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The TMP rise during the unsteady state was faster than that during steady state for both MBRs (Figure 4.2). The jump in TMP (a rapid TMP rise (Cho and Fane, 2001)) could also be observed for both MBRs during unsteady state conditions. The jump happened at day 39 and 80 for MBR (AS) and MBR (BAC) respectively (Figure 4.2.1a). It occurred when the concentration of TOC in both reactors was at its peak (Figure 4.3). The sudden increase of TOC concentration in the unsteady state may explain the TMP jump scenario for the MBR (BAC). TOC with molecular weights (MW) in the range between 100k-1000k daltons (particle size: ≈ 0.05 - $0.5 \mu\text{m}$) would more likely to foul the microporous membrane by restricting and plugging the pores compared to lower molecular weight ($<100\text{k}$) (particle size: $\approx < 0.05 \mu\text{m}$) due to the relatively large membrane micro pore size (nominally $0.5 \mu\text{m}$) usually used in the MBR system. Chapter 6 provides analysis of TOC molecular weights and more discussion of organic fouling. In summary, it was found that approximately 72 % of the TOC in the supernatant of the MBR (AS) in the range of $100\text{k} < \text{MW} < 1000\text{k}$ could be retained by the membrane (nominally $0.5 \mu\text{m}$) whereas, only about 50 % of the TOC in the range of $10\text{k} < \text{MW} < 100\text{k}$ was retained by the membrane (chapter 6: Table 6.3 and 6.4). It is proposed that, TOC may foul the membrane by (i) filling the voids of the thin biofilm layer formed on the membrane surface and (ii) forming a non-porous thin layer on the membrane surface if the operating flux exceeds the 'sustainable' flux. Loss of pores due to fouling could change the initial average flux to give local fluxes which might be above critical flux and cause TMP 'jump' (Ognier *et al.*, 2004; Cho and Fane, 2002). The 'jump' process would be accelerated if the initial operating flux was relatively high. The preliminary conclusion is that the TOC concentration, particular in the MBR (BAC), during unsteady state was mainly in the range of 100k-1000k dalton or above. This is suggested by the low concentration of TOC in the permeate (≈ 1 - 8 mg/L) (Figure 4.4) compared to the TOC concentration in the supernatant of the MBR (BAC) (≈ 20 - 140 mg/L) (Figure 4.3) from day 40-80. If the majority of the TOC in the supernatant of the MBR (BAC) was in the lower range MW ($<100\text{k}$), a higher TOC concentration should be found in the permeate.

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Figure 4.4: TOC concentration in permeate

Several aspects, of the sudden increase of TOC in the supernatant of MBR (BAC) are discussed as follows:

- i) It was found that the sudden increase in the TOC concentration of the MBR (BAC) was not caused by impurities introduced with the PAC since the phenomenon was a temporary trend and occurred at day 40 and ended at day 110. To confirm this, an experimental test using conical flasks filled with different concentrations of fresh PAC (1, 3, 5 mg/L) and clean water were stirred for 24 hours. The aim was to evaluate the contribution of fresh PAC to the TOC concentration in clean water. The results in Table 4.1 show that the TOC concentration increment in the clean water due to fresh PAC was negligible. The high TOC concentration of the MBR (BAC) may be contributed by the EPSs in the supernatant of the mixed liquor. However, the EPSs (polysaccharide and protein) only occupied less than 50 % of the TOC because of its low concentration in the supernatant as shown in Appendix B (Figure 4.1.B and Figure 4.3.B). The balance of TOC would have been SMPs not reporting as protein or polysaccharide. The TOC measurement involves

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prefiltration by 0.45 μm filter so it could include finely colloidal matter present in the supernatant.

Table 4.1: Contribution of fresh PAC with different concentrations to the TOC increment in clean water after mixing for 24 hours

PAC concentration (g/L)	TOC concentration increment (mg/L)
1	0.30 ± 0.02
3	0.62 ± 0.01
5	1.20 ± 0.08

- ii) According to Seo *et al.*, (1996), the sudden increase in TOC in a bioreactor with added PAC may be caused by more stabilized organics such as excreted (by products of bacteria) low MW intermediates or high MW components such as enzymes produced by microorganisms.
- iii) TOC fluctuations in the MBR (BAC) were also not due to the changes in the operating flux which was increased from 10.5 to 21.0 L/m²/hr because the feed flowrates (40.0 mL/min) of both MBRs were the same throughout the operating period. The flux change from 10.5 to 21.0 L/m²/hr was achieved by changing the membrane area from 0.228 to 0.114 m² respectively.
- iv) Dosoretz and Bøddeker, (2004), found that shear forces generated by pumping may cause attrition of the BAC flocs and lead to re-suspension of the bacteria from the flocs. They found this phenomenon accelerated fouling rate. This indicates that another possibility for the sudden high TOC increase scenario may due to attrition of the BAC surface due to bubble-induced shear forces. The ‘peeled off’ small

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particles ($< 0.45 \mu\text{m}$) from the BAC surface could be transferred to the bulk liquid. The small particles may contribute to the TOC concentration (from day 40-80) (Figure 4.3) in the supernatant of the mixed liquor. The attrition of the BAC flocs would mean a reduction in the BAC size. This is supported by the smaller D_{50} (number based) of the BAC from day 40 to 60 as shown in Figure 4.5. However, the D_{50} started to increase again from day 65-70 which may due to the coagulation or flocculation of the detached small particles. A relatively smaller TOC fluctuation occurred again from day 160 to 220 and the same reduction of the D_{50} of the BAC was observed.

Figure 4.5: Particle size (D_{50} , in number) development and distribution

Having discussed the significant increase in the TOC of the supernatant of the BAC mixed liquor, the reduction in the TOC concentrations from day 80 till 105 is also an interesting phenomenon. The reduction of the TOC in the bioreactor could be due to several factors such as (i) biodegradation of the detached small particles by the biomass, (ii) the attachment of the TOC onto the new membrane surface and (iii) escape of the TOC from the bioreactor to the permeate. However, factors (ii)

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and (iii) are unlikely. For example, a rapid TMP rise was not observed (Figure 4.2.1b) as would be expected for (ii). Also TOC concentrations in the permeate were also relatively low from day 80-105 as compared to the other operational days as shown in Figure 4.4.

The most probable reason for the reduction in the TOC was biodegradation. As can be seen from Figure 4.1, the MLSS from both the MBRs started to increase from around day 75. This may be because after approximately 2.5 cycles of SRT (30 days) operation period (75 days), the biomass was more stable and acclimatized. These conditions would encourage biomass production rate. The MLSS increase for the MBR (BAC) and MBR (AS) was from about 11.0 to 14.0 and 7.5 to 10.0 g/L respectively. The excess 3 g/L (14.0-11.0 g/L) biomass in the MBR (BAC) would have improved the biodegradation efficiency of the TOC in the supernatant of the mixed liquor and help to reduce the high TOC concentration into the lower range.

As discussed (please refer Figure 4.2.1a), operating the MBRs at the unsteady state could accelerate the TMP rise during filtration. However, the real MBR plant always tends to be operating at unsteady state since the feed wastewater concentration is dependent on the production from the municipal activity. For example, the production rate of the wastewater would be higher in the morning and evening compared to the rest of the day. This means that it is unavoidable to operate MBRs at unsteady state (due to variation of the TOC load). An effective MBR should have the ability to sustain the effectiveness of the MBR without causing serious fouling to the membrane during operation at a higher wastewater load. Figure 4.2.1a and 4.2.1b show that the MBR (BAC) was more stable compared to the MBR (AS) at unsteady state especially at the lower flux (period a1, 10.5 L/m²/hr). However, Figure 4.2.1b shows that the MBR (BAC) at the higher flux of 21.0 L/m²/hr was also tolerant to the relatively low range TOC (20-60 mg/L) fluctuation (period b2, day 180-220). However, a lower flux would be recommended due to its slower TMP rise and ability to delay the TMP 'jump' when the TOC fluctuation was high (such as periods a1, b1, day 30-75) (Figure 4.2.1a). It

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is probable that intermittent operation and/or backwash could significantly slow this rise. Nevertheless the benefit of PAC addition is clearly evident.

4.1.2 Effect of BAC floc sizes on membrane filtration

In these preliminary runs it was observed that the particle sizes (D_{50}), both in number (Figure 4.5) and volume (Figure 4.6) of the BAC and AS fluctuated during the initial days of operation. The particle sizes became more stable when the stabilization of MLSS in both MBRs was achieved (from about day 100 to 110 in Figure 4.1). It has been suggested that the larger BAC floc could be a factor for improved filtration performance of the MBR (BAC) (Li *et al.*, 2005).

Figure 4.6: Particle size (D_{50} , in volume) development

However, in this study it was observed that the particle sizes of the BAC both in number (Figure 4.5) and volume (Figure 4.6) were smaller than that of AS from day 90-120, even though the filtration performance of the MBR (BAC) was still better than that of the MBR (AS) in the period. This indicates that the particle size of the MBR (BAC) is not a major factor in improving the filtration performance. The particle size would play a role when the floc starts to attach on the membrane surface and provide a deposited cake resistance that determines the filtration performance. However, gross sludge accumulation on the membrane surface is usually avoided by control of flux and having an effective aeration distribution.

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In brief, the mean size of the single PAC used in the study was at about 1.6 μm (please refer to Figure 3.1; chapter 3). Due to the adsorption effect, fine foulants, substrate and bacteria cells (0.5-1.0 μm) would be adsorbed by the PAC and would move the fine particle size distribution (number) to a higher range. This could explain the cause of the floc size increase in the MLSS with addition of the PAC.

The size of the BAC would decrease if the bacteria cells detached from the PAC due to factors such as:

- (i) long SRT (≥ 30 days): At long SRT, the adsorbed substrate by PAC would be fully biodegraded by the attached bacteria. Without continuous substrate supply or sufficient food, the attached bacteria especially inner layer bacteria of the BAC would become dead cells and be detached.
- (ii) Acclimatisation period: the suspended bacteria cells need a certain period to acclimatise into attached growth condition. During acclimatisation period, the attached bacteria cell may easily be detached from the PAC especially under hydraulic turbulence conditions. This may explain the idea of BAC floc breakage as shown in Figure 4.5.

Figure 4.6 does not show the trend of floc breakage because it is based on the volume distribution. The distribution is biased to large particles and does not show detailed change in the smaller particle size range.

4.1.3 Mechanisms involved in membrane fouling control

In sections 4.1.1 and 4.1.2, it was shown that BAC had the better fouling control during the higher TOC concentrations in the supernatant of the mixed liquor even though the BAC had smaller floc size. The possible beneficial effects of BAC are,

- (i) adsorption of foulants,
- (ii) enhanced biological decomposition and biodegradation (please refer to chapter 6 for detail)

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- (iii) additional scouring of the membrane surface to detach or backtransport foulants.

These effects could occur simultaneously.

The adsorptive properties of PAC are well-known, and this mechanism is explored in more detail in Chapter 6. The loss of adsorptive capacity in aged BAC is shown to be detrimental in section 4.2 of this chapter. The second mechanism is linked to the first and is based on the idea that adsorbed substrate on PAC becomes available to the biomass and encourages growth; this is essentially the BAC concept. More consideration of this mechanism is provided in Chapter 6. Thirdly, fresh PAC is a non-compressible solid which could collide with the membrane surface with the help of the air bubbles to detach the fine foulants on the membrane surface back to the bulk liquid. The particulate PAC could also augment boundary layer backtransport of foulants and prevent them from depositing on the membrane. These 'hydrodynamic' mechanisms are considered in Chapter 6, 7 and 8. For example, in chapter 6, glass beads (D_{50} : 5, 20, 50 μm) are shown to be able to reduce the membrane fouling rate better with its 'scouring' effect compared to AS floc. In chapter 7, the 'scouring' effect of AS floc was also examined and the results indicate that AS floc has some scouring effect. The bigger AS floc size has a better effect. The interesting and challenging aspect of the MBR (BAC) is that both scouring and adsorption mechanisms can work in parallel. For example, the adsorptive property of AS floc would not be comparable to fresh BAC floc. Thus, the smaller BAC floc with better adsorptive properties may outperform bigger AS floc in limiting the amount of foulants attached on the membrane surface. Moreover, smaller size fresh PAC has also been found better than larger size polymer beads (D_{50} : 600 μm) in terms of foulant detachment from the membrane surface (see chapter 8 (Figure 8.9)). Others have suggested that the BAC in the vicinity of the membrane may also act as an effective buffer zone to minimise the contact of the fine foulants with the membrane surface. It is of interest to note that Pirbazari *et al.*, (1996) and Seo *et al.*, (1996) reported that PAC added to an MBR could form a dynamic particulate layer on the membrane surface to act as a precoat filter layer to protect the membrane from foulants. However, this effect would

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require the flux to be sufficiently high to deposit a cake layer. In this study, elevated fluxes have been found to be detrimental.

A comparison of filtration of AS and BAC MLSS was also made in a series of short-term tests (see 3.2.5 for protocols). Based on fixed pressure dead-end unstirred filtration, the BAC floc was found to have better performance in controlling R_{if} (irreversible membrane fouling) for samples taken at steady and unsteady state compared to AS floc. The R_f values (the values are in % which represents the irreversible fouling increment after filtering the samples of AS and BAC from the MBR (AS) and the MBR (BAC) respectively) of AS and BAC are shown in Table 4.2. BAC was found to decrease irreversible fouling better even during the period when the TOC concentration in the supernatant of the MBR (BAC) was higher than the MBR (AS) (days 98-107, 156-177 and 178-220, as shown in Figure 4.3). This demonstrates that BAC in the mixed liquor can protect the membranes better than AS. The measured performance of the MBR (AS) and MBR (BAC) in terms of R_{if} control increased with time. The longer the operation of both the MBRs, the better the R_{if} (irreversible membrane fouling) control. This is supported by the data in Table 4.2. Table 4.2 shows the trend of the longer the operation of both the MBRs, the better the R_{if} control. The probable reason is that the longer the operation of the MBRs, the better the condition in terms of bioactivity.

Table 4.2: Membrane resistance increment (%) after filtration between day 100-210

MBR	Day 98-107	108-155	156-177	178-220
AS	86.50 ± 16.25	75.00 ± 35.09	34.79 ± 5.88	18.54 ± 7.03 (%)
BAC	35.20 ± 10.66	36.64 ± 13.58	19.42 ± 5.55	18.02 ± 7.25 (%)

* Resistance increment (%) = $[(R_{fouled} / R_{initial}) - 1] \times 100$

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Figure 4.2.1c: Membranes filtration performance in MBR and hybrid PAC MBR

The above series of short-term tests was done under unstirred conditions. A question arises as to whether this short-term test can be used to predict MBR performance. Considering Figure 4.2.1c (a section of Figure 4.2) (day 177-201) where at day 177, new membrane modules with an area of 0.114 m² were submerged into each MBR and run at a fixed permeate flowrate of 40 mL/min (flux = 21 L/m²hr, HRT = 8.3 hrs). The results as shown in Figure 4.2.1c (from day 177-201) have clearly indicated that the MBR (BAC) with 5 g/L of PAC performed better. The average rate of TMP rise was 2.4 kPa/day for the MBR (AS) and only 0.8 kPa/day for the MBR (BAC) throughout the operating period of about 25 days. Inspection of the hollow fibres modules removed from the MBRs at day 201 revealed negligible cake deposition or accumulation, as shown in the photo images (Figure 4.7).

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Figure 4.7: Images of the membrane modules from the MBR (AS) (a) and MBR (BAC) (b) at day 201

However at that point of time the MBR (AS) membranes had a significant fouling resistance with a required TMP of about 70 kPa compared to about 25 kPa for the MBR(BAC) membranes and < 10 kPa for new membranes (see Figure 4.2.1c). After water washing, the MBR (AS) membranes had a required TMP of 50 kPa. Following rinsing, the MBR (AS) membranes were used again to continue the filtration process and it was observed that the TMP increased rapidly from 50 kPa back to 70 kPa (Figure 4.2.1c). These observations and Figure 4.7 (no serious biomass cake attachment) suggest that the membranes in the MBR (AS) had significant pore plugging and/or restriction due to components in the MLSS. The much lower TMP rise for the MBR (BAC) implies less irreversible fouling resistance.

From the short-term tests reported in Table 4.2, it was found that the R_{if} control of AS and BAC between day 178-220 was almost similar, whereas the real filtration test (i.e MBR profile for TMP) showed that the MBR (BAC) performed better compared to the MBR (AS). This indicates that, under bubbled crossflow conditions, the BAC flocs have better filtration results. One reason may be the better ‘scouring’ impact of BAC to detach the fine foulants attached on the membrane surface through simultaneous ‘collision’ and adsorption effects.

Apart from enhancing performance by protecting the membrane pores from blocking or plugging by ‘scouring’ effect, it was also reported that PAC could reduce the specific cake resistance (SCR) of the AS (Kim *et al.*, 1998). However,

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the results from the unstirred fixed-pressure dead-end filtration tests (protocol in section 3.2.5) show that the SCR and flux decline profile of BAC were not always better than that of MBR (AS). The SCR was higher (Figure 4.8) and flux decline profile (Figure 4.9) was lower for the BAC as compared to AS for the period day 48 to 107 when the fluctuation of TOC was high.

Figure 4.8: Specific cake resistance of MLSS samples

Figure 4.9: Flux decline profiles of 20L MBR MLSS samples for unstirred fixed pressured dead-end filtration (example from period day 48-107)

This indicates that the TOC concentration in the MBR (BAC) affected the SCR and flux decline profiles in unstirred dead-end filtration. The TOC may have reduced the porosity of the cake layer by filling up the gaps between the biomass floc. Eventually, the lower porosity of the cake layer caused the high SCR and lowered

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the flux decline profile. The TOC concentration in the MBR (BAC) dropped to a level lower than the TOC concentration in the MBR (AS) after day 107 to 140 before another two small scale TOC fluctuation happened (day 190-220; 160-175). The lower TOC concentration in the MBR (BAC) produced better performance of the BAC compared to the AS in terms of SCR and flux decline profile as shown in Figure 4.8 and Figure 4.10 respectively. However, the short-term tests were not mirrored in the TMP profiles in Figure 4.2 and the filtration performance of MBR (BAC) was always better than that of MBR (AS). This indicates that cake accumulation on the membrane surface in both MBRs was not significant.

Figure 4.10: Flux decline profiles of 20L MBR MLSS samples for unstirred fixed pressured dead-end filtration (example form period day 107-140)

In summary, the performance of the MBR (BAC) was overall better than the MBR (AS) suggesting that the SCR and flux decline profile characteristics are not a strong indicator of filtration performance of the larger MBRs (except possibly when local flux starts to exceed the critical flux of the MLSS that is when cake is allowed to deposit in the MBR).

To further examine if the filtration performance of the BAC was better than that of AS even under higher TOC concentrations, the filtration behaviour using a fixed flux mode with crossflow (0.2 m/s; without aeration) (protocol see section 3.2.5) was assessed at day 196 (TOC in supernatant: MBR (BAC) > MBR (AS)). Figure 4.11 shows that under these conditions the filtration of the BAC was always better

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than that of AS in terms of stability and the TMP rise. This indicates that BAC was still capable to raise the ‘critical’ flux of the MBR (BAC) under relatively unstable conditions when compared to the MBR (AS). The probable mechanisms of simultaneous adsorption, decomposition, biodegradation and scouring as mentioned above may help to prevent the foulants such as TOC, EPSs or even planktonic cells in the supernatant from (i) blocking and plugging the pores of the membrane and (ii) depositing on the membrane surface. The above mentioned mechanisms are further discussed in chapter 6.

Figure 4.11: Sustainable flux tests for AS and BAC at day 196

4.2 Performance comparison of MBR (AS) and MBR (BAC) at ‘infinite’ SRT

To further investigate the effect of BAC in mitigating membrane fouling in MBR, the two batch 20L MBRs (the MBRs were only fed once a day) were operated without sludge wastage (effectively infinite SRT). This operating strategy meant that the 5 g/L of PAC inventory in the MBR was not gradually replenished but remained ‘aged’ in the MBR (hereafter known as MBR (aged)). A filtration performance comparison of the two batch MBRs running at low fluxes of 7.0 L/m²hr was done after cultivating the two batch MBRs for 61 days, when membranes were introduced. The results in Figure 4.12 show that the MBR (aged

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BAC) had a slightly greater fouling rate than the MBR (aged AS). This indicates that the MBR (aged BAC) with aged BAC could not improve the performance of the MBR. The poor performance of the MBR (aged BAC) compared with the MBR (aged AS) at infinite SRT contrasts dramatically with the much better performance at the SRT of 30 days as discussed above.

Figure 4.12: Performance comparison of 20L batch MBR and MBR (BAC) at pseudo infinity SRT

It was observed that the TOC concentration in the supernatant for both the prolonged SRT MBRs was higher than that of the TOC of the feed solution. One reason for this could be the presence of significant quantities of lysed cell products. Figure 4.13 shows confocal images of AS and BAC floc with evidence of dead cells. However, the TOC analysis involves prefiltration through a 0.45 µm membrane so that some of the cell debris was probably not analysed as TOC. The other reason for the high TOC may be the accumulation of high MWt species due to organic retention. Although the MBR membranes are MF pore size, they would rapidly close up to give solute retention. This phenomenon is discussed in Chapter 6. The long SRT conditions (no sludge wastage and presence of significant dead cells) may contribute to the high concentration of the TOC and polysaccharide in the mixed liquor.

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Figure 4.13: Confocal images of AS (a) and BAC (b) flocs at pseudo infinity SRTs
[Note: live bacteria show green, dead show red and combination of live and dead bacteria show yellow]

From the results (Figure 4.12; insert), it was found that both MBRs had quite similar foulant concentrations in the supernatant consisting of TOC and polysaccharide. Under the prolonged SRT conditions, the TOC and polysaccharide concentrations in the supernatant for the MBR (aged BAC) were 219.1 ± 31.96 mg/L and 152.88 ± 14.87 mg/L and for the MBR (aged AS) were 186.9 ± 47.42 mg/L and 179.70 ± 7.76 mg/L as shown in Figure 4.12 (insert). This indicates that the BAC had lost most of its adsorptive effect. However, the MBR (aged BAC) was still able to produce higher quality permeate with lower TOC concentration (Figure 4.12; insert) and less yellowish permeate color (Figure 4.14) as compared to the permeate from the MBR (aged AS).

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Figure 4.14: Color quality comparison of permeate of 20L batch MBR and MBR (BAC) at pseudo infinity SRT

The reason that the aged BAC had lost most of its adsorptive capability may have been due to blocking of the PAC pores by (a) fine foulants, (b) products of microbial biodegradation and (c) dead microbial cells (Sirotkin *et al.*, 2001). As a result, the aged BAC would not do its job effectively in ‘picking’ up and assisting in decomposition and biodegradation of fine species such as TOC, polysaccharides and planktonic cells in the supernatant or on the membrane surface that could foul the membrane.

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Figure 4.15: Particle size distribution and D_{50} (in number and volume) of BAC and AS

It was also observed that the MBR (aged BAC) had a smaller floc particle size (D_{50}) than that of MBR (aged AS) as shown in Figure 4.15. The shift of D_{50} to somewhat lower values may be due to the contribution of the powdered PAC having formed smaller average BAC flocs compared to long SRT AS floc.

The smaller D_{50} of aged BAC may be one of the reasons that caused the slightly higher SCR and the lower flux decline profile as compared to AS as shown in Figures 4.16 and 4.17 respectively (note: on the other hand, fresh BAC with ‘adsorptive effect’ was found can improve membrane fouling control though it also formed smaller BAC floc compared to AS floc).

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Figure 4.16: Specific cake resistance and resistance increment (irreversible fouling) for MLSS with different SRTs

Figure 4.17: Flux decline profile for fixed pressure and unstirred dead-end filtration of aging BAC and AS

Since the aged BAC lost most of its ability to adsorb the fine pollutants, it was expected that the aged BAC would not help to reduce irreversible fouling resistance, R_{if} . A short term unstirred fixed pressure test to evaluate the performance of aged BAC in reducing irreversible fouling is shown in Figure 4.16 (the y axis represents the % increase in membrane resistance due to R_{if} after the

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filtration test). The result supports the claim that aged BAC did not help to enhance the irreversible fouling control.

In addition, short term ‘sustainable’ flux tests by flux stepping were performed with clean membranes submerged directly into both MBRs to estimate their relative sustainable fluxes and compare their performance in fouling resistance control. The tests included flux stepping and TMP measurement as an indicator of the membrane fouling tendency. The data are shown in Figure 4.18. It was found that the AS had a sustainable flux <18.0 L/m²hr, but the aged BAC had a sustainable flux at only about 6.0 L/m²hr. From the above discussions, it can be concluded that the aged BAC was not be able to raise the performance of the MBR in terms of fouling control.

Figure 4.18: Sustainable flux tests for batch AS and BAC at pseudo infinity SRT

A series of separate tests were performed using the same 20 L reactors with continuous feed rather than batch mode in order to simulate real operating MBR system. The MLSS concentrations in the continuous feed MBR (aging AS) and MBR (aging BAC) increased from 1-8 g/L and 6-12 g/L respectively over the experimental duration of 20 days at a flux 10.5 L/m²hr without sludge wastage. The membrane in the MBR (aging AS) was cleaned by flushing the surface with tap

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water at approximately day 18 due to a sudden increase in the TMP ‘jump’ (Figure 4.19).

Figure 4.19: Performance comparison of MBR and MBR (BAC) at pseudo infinity SRTs (flux: 10.5 L/m²/hr)

The TMP dropped from approximately 40 kPa to 15 kPa. Comparing the TMP profiles of both MBRs, the TMP rises were almost similar, suggesting no improvement of filtration with the addition of 5 g/L PAC without sludge wastage and renewal of aged BAC.

After comparing the performance of both MBRs at a flux 10.5 L/m²/hr, the MBRs were tested again at flux of 21.0 L/m²/hr from day 35, when new membranes were introduced. The results in Figure 4.20 show that the MBR (aging BAC) had a much greater fouling rate than the MBR (aging AS).

Cleaning the membranes from the MBR (aging BAC) after 3 days of filtration lowered the TMP a little, but it was then followed by a rapid increase in the TMP. Even though the membrane was repeatedly cleaned with tap water, it failed to recover its permeability. This indicates that the MBR with the aging BAC had a lower sustainable flux than the MBR (aging AS).

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Figure 4.20: Performance comparison of MBR and MBR (BAC) at pseudo infinity SRT (flux: 21.0 L/m²/hr)

The worse performance of the MBR (aging BAC) compared with the MBR (aging AS) at infinite SRT contrast with the much better performance at the SRT of 30 days, as shown in Figure 4.2.1 (a, b, c). It is important to note that this behaviour was qualitatively reproducible. Thus the benefit of the PAC inventory was lost as the BAC aged. One effect of ‘aged BAC’ is the loss of adsorptive capacity which is clearly important.

If a key role of the PAC was as a scouring agent this should be sustained even with aging, but it was not. This either means that the ‘scouring’ mechanism is not significant or that when PAC is embedded over time within the bioflocs (Figure 4.21) and it loses its impact; in other words only fresh PAC particles have significant scouring effects. The aged BAC would simply act as ineffective particles with smaller D₅₀ and also increase the MLSS concentration. A higher MLSS may be an obstacle to membrane movement in the bioreactor and obstruct the air bubbles from easily contacting the membrane surface. These factors may explain the poor performance of the MBR (aged BAC) as compared to the MBR (aging AS).

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Figure 4.21: Optical image of PAC is embedded over time within the biofloc

4.3 Conclusions

The effects of the 5 g/L of PAC on AS for membrane filtration were studied in the 20L MBRs at SRT 30 days and long SRT. The filtration performance of the MBR (BAC) operated at SRT 30 days was always better than that of MBR (AS) at steady and unsteady state throughout the operation period. The preliminary conclusions are that the BAC has the simultaneous roles of adsorption, decomposition, biodegradation and scouring that minimise the fouling rate.

Apart from reducing the fine foulants in the supernatant of mixed liquor by adsorption, decomposition and biodegradation, it was also postulated that BAC which had contacted with the membrane through ‘collisions’ that could detach and then adsorb the fine foulants from the membrane surface and thereby recover the permeability of the fouled membrane. However, it was found that the MBR (aged/aging BAC) with no renewal of aged BAC with fresh PAC did not have better performance than that of MBR (AS). It is suggested that aged BAC had lost its adsorption capacity due to the pores being saturated by the adsorbed fine foulants. Aged BAC was also found embedded within the biomass flocs. These may be the reasons why the aged BAC lost its simultaneous ‘collision’ and adsorption effects. The aged BAC without its adsorptive capacity in powdered form would only have

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the ‘collision’ effect of a typical AS floc. However, the aged BAC would also increase the MLSS of the MBR (aged/aging BAC).

The higher MLSS in the MBR (aged/aging BAC) may (i) limit the distribution of the air bubbles to access to the membrane surface and (ii) constraint the movement of the membrane module. Both limitations may also explain the relatively poor performance of the MBR (aged/aging BAC). Therefore to gain the benefit of the MBR (BAC), it is necessary to have regular replenishment of aged BAC with fresh PAC. The preliminary conclusions suggest that the primary role of the BAC is to provide simultaneous adsorption, decomposition, biodegradation and possibly scouring effects in the removal of fine foulants (i.e. TOC, polysaccharide, planktonic cells, fine colloids and etc) rather than providing ‘scouring’ or adsorption effect alone in fouling control. These mechanisms are further discussed in subsequent chapters (6, 7, 8) especially in chapter 6.

CHAPTER 5 THE EFFECT OF PAC CONCENTRATION ON MBR PERFORMANCE

CHAPTER 5 THE EFFECT OF PAC CONCENTRATION ON MBR PERFORMANCE

Synopsis:

In the previous chapter, the effect of PAC on membrane fouling control in MBRs at SRT 30 days and long SRT was examined. It was confirmed that operating the MBR (BAC) at SRT 30 days shows the PAC to be more beneficial. However, whether there is an optimum PAC concentration for enhanced MBR performance is unclear. As a result, a filtration performance comparison of mixed liquor from MBRs with different PAC concentrations was investigated in this chapter. The experimental set up, detailed in section 3.1 (Table 3.2), involved use of 4 small bench-scale (2 L) MBRs operated in parallel with different PAC concentrations. The fouling tendency of the mixed liquor was measured ex-situ in small test cells. Other parameters measured were MLSS, TOC and mean floc size.

5.1 Performance comparison between four 2L batch MBRs with different PAC concentrations at SRT 30 days

The MLSS profiles for the four 2L MBRs are shown in Figure 5.1. The 2L MBRs were initially filled with AS at 3 g/L. Then PAC was added to reactors 2, 3 and 4 at a loading of 1, 3, and 5 g/L PAC respectively. Thus, all 2L MBRs started with similar amounts of AS biomass but different concentrations of MLSS due to the differences in PAC content, i.e. reactor 4 initially had a starting concentration of 8 g/L (3 g/L biomass + 5 g/L PAC). In this study, 5 g/L PAC was used as a reasonable maximum loading. The 2L MBRs were operated with sludge retention times (SRT) of 30 days which required sludge wastage of approximately 67 mL per day. This caused a loss of 3.35wt% of PAC from the bioreactor. The PAC was topped-up daily to account for this loss and to maintain constant PAC concentrations in the bioreactors. Obviously even higher PAC loading could be used but because PAC replenishment is required, the added cost and handling become a constraint.

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Figure 5.1: MLSS development in 2L MBRs

Table 5.1: Concentration of the MLSS and PAC in MBRs at Day 100

Name of MBRs	MLSS concentrations (g/L)	PAC concentrations (g/L)	Working volume (L)
0 g/L PAC	12.0 ± 1.0	No PAC added	2.0
1 g/L PAC	13.0 ± 1.0	1.0	2.0
3 g/L PAC	15.0 ± 1.0	3.0	2.0
5 g/L PAC	17.0 ± 1.0	5.0	2.0

A mass balance check was made to confirm that the PAC contents were maintained at steady state. From Figure 5.1, it can be seen that for all MBRs the MLSS contents became stable after approximately 100 days (about 3 x SRT). The steady-state concentrations are given in Table 5.1 and it is evident that the 4 MBRs all achieved a similar biomass concentration ($[MLSS - PAC] = 12.0 \pm 1.0$ g/L). A series of experimental analyses to compare the various characteristics of the MLSS in the 2L MBRs was started on day 109. MLSS samples were withdrawn from each of the bioreactors and tested in terms of (i) the mean floc size (D_{50}) (ii) TOC concentration in the supernatant and permeate (iii) specific cake resistance (SCR) (iv) flux decline profile at unstirred fixed pressure (v) the irreversible fouling resistance R_{if} and (vi)

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the ‘sustainable’ flux. Details of the filtration test equipment and protocols were given in chapter 3 (section 3.2.5).

5.2 Comparison of characteristics of AS and BACs

The incorporation of PAC into the AS reduced the SCR values measured at fixed pressure and at fixed flux as indicated in Figure 5.2. From Figure 5.2, it can be seen that the SCR measured at modest flux ($\leq 20 \text{ L/m}^2\text{hr}$) was lower than that measured by fixed pressure (100 kPa) dead-end filtration.

Figure 5.2: Relationship of D_{50} to the specific cake resistance in fixed pressure and flux

This suggests that operation of the MBR system with modest fixed flux should be more beneficial than with fixed pressure though the latter could initially produce higher fluxes ($> 100 \text{ L/m}^2\text{hr}$). Our observations agree with the fixed flux vs fixed pressure comparison of Defrance and Jaffrin (1999). The probable reason is that with fixed pressure filtration there is a high initial flux causing more pore plugging and restriction and a more compact cake layer on the membrane surface. Figure 5.3 shows the flux decline profiles for the fixed pressure (100 kPa) tests plotted against the volume concentration ratio ($\text{VCR} = [V_{\text{feed}}/V_{\text{concentration}}]$), with initial fluxes of 100 to $> 350 \text{ L/m}^2\text{hr}$. One conclusion from this study is that characterisation of the

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SCR of MLSS in MBRs is best done at constant flux, although the trends (Figure 5.2) are qualitatively similar for SCR (constant pressure).

Figure 5.3: Flux decline profile for fixed pressure and unstirred dead-end filtration

The results in Figures 5.2 and 5.3 clearly show that PAC within the AS had the ability to reduce the SCR and lift the flux decline profile significantly. It is also evident in these tests that PAC concentrations of 1 and 3 g/L gave the lowest SCR and the highest fluxes. The 0 g/L PAC sample was clearly worse than any of the PAC (BAC) samples. The lower SCR and higher fluxes of the BAC could have several explanations, (i) increase in the porosity of the activated sludge and (ii) the formation of a BAC particulate layer with lower cake compressibility which gives a lower cake resistance to the activated sludge (Seo *et al.*, 1996; Zhao *et al.*, 1999). According to the Carman–Kozeny equation, the specific cake resistance (SCR), α , can be expressed by equation (5.1)

$$\alpha = \frac{180(1 - \varepsilon)}{\rho_p d_p^2 \varepsilon^3} \quad (5.1)$$

where ε is the porosity of the cake layer, ρ is the particle density (kg/m^3) and d_p is the effective particle diameter (m). It should be noted that a relatively small change in ε can cause large changes in α . From equation (5.1), the effective particle size

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(d_p) and therefore the floc size distribution is another important factor that can affect the reduction of the SCR.

Figure 5.2 shows the D_{50} of the AS and the BACs. At the same superficial gas velocity (SGV) of 8.7 mm/s, the biomass floc was marginally bigger in the bioreactor with 1 g/L PAC as compared to the bioreactor with AS only. This suggests that suspended growth of biomass was dominant in the bioreactor with 1 g/L of PAC; so that the PAC merely attached onto/into the biomass floc to form bigger particles. However, with the addition of 3 g/L and 5 g/L of PAC into the bioreactors, the D_{50} shifted to somewhat lower values. These observations agree with those of Kim *et al.*, (1998). This trend was probably due to the fact that PAC was in powder form with relatively smaller size (see size distribution data in Figure 3.1 and images in Figure 5.4) and the biofilm could have grown onto the PAC particles to form smaller average BAC floc. The BACs in the bioreactors with 3 and 5 g/L of PAC had a smaller mean floc size as shown in Figure 5.2, which suggested that attached growth was predominant. Therefore, BAC with 1 g/L of PAC may have had the lowest SCR and the highest flux profile because of its larger floc size. Based on particle size alone the BACs with 3 and 5 g/L of PAC should have higher SCR values compared to the AS (no PAC) due to their smaller particle size. However, BACs with 3 and 5 g/L of PAC had significantly lower SCRs and better fluxes compared to AS alone. This suggests that the addition of 3 and 5 g/L of PAC into the bioreactors could have improved the porosity of the cake of the BAC though formed from smaller floc sizes. This improved porosity could be due to less EPS in the interstitial voids and/or reduced cake compressibility. In general for this test, comparison between the performances of BAC with 1, 3 and 5 g/L of PAC agree qualitatively with equation 5.1. Thus D_{50} was in the decreasing order, BAC with $1 > 3 > 5$ g/L of PAC and the SCR was in the increasing order, BAC with $1 < 3 < 5$ g/L of PAC.

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Figure 5.4: Images of flocs from 2L MBRs with different PAC concentrations

The level of TOC in the reactor was also considered to be one of the factors that could affect the filtration performance of the AS and BAC. This is because TOC includes not only unused substrate but also extracellular polymeric substances (EPS), such as polysaccharides and proteins, believed to foul membranes (Huang *et al.*, 2001). The bioreactor without PAC, where the AS had the highest SCR and the lowest flux profile also contained the highest amount of TOC concentration as shown in Figure 5.5 (note log-scale for reactor TOC). As mentioned above, the EPS could influence the cake resistance by filling up the void space between the flocs formed on the membrane surface. In addition the EPS could contribute to the flux decline (Figure 5.3) by plugging or restricting the pores. The bioreactors with 1, 3 and 5 g/L of PAC, which all had substantially lower TOC contents also had lower SCR values and lower flux decline profiles.

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Figure 5.5: TOC in the bioreactors and permeate with different PAC concentrations

It is of interest to note from Figure 5.5 that there were significant differences in the TOC values in the MBRs compare with the permeates. Average membrane retentions $\{= 100 \times [1 - (\text{TOC}_{\text{permeate}}) / (\text{TOC}_{\text{MBR}})] \%$ were 85.5, 64.2, 74.4, 79.3 % for PAC loads of 0, 1, 3, 5 g/L respectively. This shows that the membranes provided relatively good separations, possibly due to their fouling layers, coupled with the PAC adsorption effects. The best overall removals of TOC substrate across the MBRs (i.e. lowest TOC permeate) were obtained with the highest PAC loading, which provides evidence for adsorption and the improved treatment afforded by the MBR (BAC) process. In other studies, Scholz and Martin (1997) and Mochidzuki *et al.*, (1998) found that PAC enhanced the removal of TOC through biodegradation of the adsorbed TOC.

From the above discussions based on measured SCR and flux decline profiles (at constant pressure) the addition of 1 g/L of PAC appears to be sufficient. However, further tests examined the degree of irreversible fouling by measuring the irreversible fouling resistance, R_{if} (after water wash) and the ‘sustainable’ flux by flux stepping. It was found that BAC with 5 g/L of PAC was able to protect the membrane better than BAC with 0, 1 and 3 g/L of PAC as shown in Figure 5.6 (the y axis represents the % increase in membrane resistance due to R_{f} after the filtration test). This may due to the higher concentration of BAC forming a protective layer and preventing the foulants, such as TOC including EPS or fine colloids, from

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entering, blocking or plugging the pores of the membrane. In addition, crossflow tests (without aeration) were performed on samples (50 mL MLSS diluted with 1950 mL of Milli-Q water) from each 2L MBR, to estimate their relative sustainable fluxes. The tests included flux stepping and TMP measurement as an indicator of the membrane fouling tendency. The data are shown in Figure 5.7. It was found that AS alone, without PAC, had the lowest sustainable flux (<36 L/m²hr), MLSS with 1 and 3 g/L of PAC had higher sustainable fluxes at about 36 L/m²hr and MLSS with 5g/L of PAC had the highest sustainable flux at about 43 L/m²hr.

Figure 5.6: Membrane resistance increment due to irreversible fouling

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Figure 5.7: Sustainable flux tests for MLSS with different PAC concentrations

In summary, the results from the 4 x 2L reactors showed the following; (i) the results based on dead-end membrane filtration tests (SCR, flux decline profile) indicated that BAC with 1 g/L of PAC had the best performance, (ii) on the other hand, BAC with 5 g/L of PAC had the best performance in terms of R_{if} control (unstirred dead-end test) and sustainable flux (crossflow tests). Thus although the dead-end filtration tests show important differences between AS and BAC at different concentrations of PAC they relate to conditions where a filter cake is allowed to form. In an MBR, this is avoided or minimised by aeration and the use of relatively low fluxes. With this in mind the PAC load giving the best performance in the crossflow sustainable flux test would be more relevant to an MBR. Thus our short term filtration tests with the 2L MBRs point to the use of 5 g/L PAC as the better option.

The improved membrane performance of the MBR (BAC) with added PAC could include the following factors;

- i) The PAC provides a sink due to adsorption for some of the fouling components such as EPS and fine colloids. Evidence in favour of this is provided in Figures 5.5, 5.6 and 5.7, for the 2L MBRs, where the TOC for MBR (AS) was significantly higher than for MBR (BAC) and the

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MBR (AS) showed greater fouling. The adsorption mechanism is examined in more detail in chapter 6.

- ii) A second factor could be the potential 'scouring' effect of the BAC at the membrane surface as the bubbled suspension is carried past the membranes. There is evidence from other membrane studies (Fane, 1984) that supramicron particles reduce the rate of fouling by enhancing back-transport of fouling species. While this is an attractive explanation, the results presented in the previous chapter (4) cast some doubt on this mechanism in the MBR (BAC) especially MBR (aged BAC). As discussed, the 'scouring' impact of the BAC in membrane fouling control would only be significant if the BAC is still equipped with adsorptive effect. Only the simultaneous 'collision' and adsorption would enhance the 'scouring' effect. The scouring mechanism is examined in more details in chapters 6, 7 and 8.

5.3 Conclusions

The addition of PAC to the activated sludge in an MBR can improve the membrane performance. This conclusion was confirmed in short-term tests on mixed liquor from bench-scale MBRs (0 to 5 g/L PAC in about 12 g/L biomass) as well as long-term runs in two 20 L MBRs (0 and 5 g/L in about 9.5 g/L biomass) in chapter 4. Short-term tests to measure specific cake resistance (SCR) at both fixed pressure and fixed flux produced similar trends with the lowest SCR at a PAC addition of 1 g/L. The SCR (fixed flux) values were significantly lower. Similar trends were obtained for flux decline profiles in deadend filtration. The SCR and flux declines for 0 g/L PAC were significantly higher. Short-term tests measuring irreversible fouling resistance and 'sustainable' (low TMP rise) flux favoured 5 g/L PAC addition, and again 0 g/L PAC mixed liquor performed much worse. The short term tests were useful in comparing the relative filterability of the different MLSS, but could only be regarded as approximate predictors of long term performance.

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Synopsis:

This chapter describes experiments and observations that aim to examine the possible mechanisms by which the presence of PAC, in the form of biological activated carbon (BAC), could improve the filtration performance of the MBR. A protocol had been applied to give a fair comparison of the potential mechanisms. The examined mechanisms include (i) effect of PAC on MLSS, (ii) effect of PAC on planktonic bacteria, (iii) effect of PAC as an adsorbent, (iv) effect of PAC as scouring agent and (v) effect of PAC in the BAC form for adsorption and biodegradation. The effectiveness of each mechanism in decreasing fouling rate has been carefully evaluated. The properties and filtration characteristics of AS and BACs have also been measured and compared in both short-term tests and in long-term continuous operation runs in parallel at SRTs of 10 days. This relatively low SRT (for MBRs) was selected as it is a condition likely to experience fouling without PAC. As such it should demonstrate the effect of PAC addition.

6.1 Fouling rates for MBR (AS) and MBR (BAC)

Four small (2L) MBRs with different PAC concentrations (0, 1, 3, 5 g/L) were operated at SRT of 10 days. Details of the experimental set-up, feed composition and general protocol are given in chapter 3. At day 108, new hollow fiber membrane modules were submerged in each MBR to test their performance in terms of membrane fouling control. The transmembrane pressure (TMP) was used to characterise the membrane fouling intensity and act as an indicator for the filtration performance of the MBRs. Figure 6.1 plots TMP history and shows that the MBR (BAC) with 5 g/L PAC and without PAC had the best and worse performances in terms of fouling control respectively. Figure 6.1 (a) shows data for 0 g/L PAC. At a flux of 21.0 L/m²/hr, an immediate TMP 'jump' was observed and reducing the flux from 21.0 to 10.5 L/m²/hr helped to slow the 'jump' but the effect

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was not significant. To avoid the immediate 'jump', the MBR (AS) was run at a low flux of 5.3 L/m²/hr. After approximately one day of operation, the membrane was taken out and washed with tap water. The membrane was used again at day 111.5 with a starting flux of 10.5 L/m²/hr instead of 21.0 L/m²/hr. The TMP rise at this operating flux was much slower as compared to the previous operation. After the membrane had fouled seriously, decreasing the operating flux from 10.5 to 5.3 L/m²/hr would not reduce the TMP rise rate. In other words, the fouled membrane was not restored by simply reducing flux and in order to restore the fouled membrane back to the starting TMP, chemical cleaning was required.

Figure 6.1 (b) shows, for MBR (BAC) with 1 g/L of PAC, that the rapid TMP rise ('jump') was delayed to approximately 6 days. The MBR (BAC)s with 3 and 5 g/L of PAC had better results compared to the MBR (BAC) with 1g/L of PAC with a smaller dTMP/dt and no 'jump' observed over the period. This demonstrates that the higher the PAC concentration, the better the performance in terms of fouling control. The average rate of TMP rise was 709.68 kPa/day, 8.62 kPa/day, 6.67 kPa/day and 5.1 kPa/day for the MBRs with 0, 1, 3 and 5 g/L of PAC respectively for the 7 days of operation as shown in Table 6.1. It should be noted that these TMP rises would be specific to the conditions chosen (SRT, membrane/module characteristics, aeration rate, activated sludge condition etc). However, they clearly confirm the benefit of PAC addition with replenishment, the rate of replenishment being determined by the sludge wastage (SRT 10 days means 10% per day).

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Figure 6.1: Performance comparison of (a) MBR (AS) with different fluxes and (b) MBR with different PAC concentrations at SRT 10d

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Table 6.1: Average TMP rise of MBRs

PAC (g/L)	(dTMP/dt) Average (kPa/day)	Time to 'jump' (hr)	Flux (L/m ² /hr)
0	709.7	1.0	21.0
1	8.6	140.0	21.0
3	6.7	N.A	21.0
5	5.1	N.A	21.0

N.A: Not applicable (no obvious 'TMP' jump over the filtration period)

6.2 Effect of PAC on MLSS

At the relatively short SRT of 10 days it was noted that the PAC addition to the MBRs was able to enhance the development of the biomass. This is shown in Table 6.2 where the MBRs with 0, 1, 3, 5 g/L of PAC had MLSS concentrations of 3.5 ± 0.5 , 6.0 ± 0.5 , 9.0 ± 0.5 and 11.0 ± 0.5 g/L respectively. This means that the MBR (BAC) with 1 g/L of PAC with MLSS of 6.0 ± 0.5 g/L had an excess of about 1.5 g/L [6.0 g/L (biomass + PAC) – 1.0 g/L PAC – 3.5 g/L of MLSS of MBR] of biomass compared to the MBR (AS) without PAC. With more PAC dosage, the MBR (BAC)s with 3 and 5 g/L of PAC had about 2.5 g/L more biomass concentration than that of the MBR (AS). This effect was not observed previously at SRT 30 days (Chapter 4 and 5), which may be due to the lower proportion of fresh PAC at longer SRT. However at SRT 10 days the PAC appears to encourage biomass development, possibly by providing a sink for substrate that would otherwise pass out of the MBR. This issue is discussed in more detail in sections 6.5.1-6.5.2 where it is shown that the MBR (BAC) provides greater removal of TOC. Apart from promoting biomass production and providing more acclimatized and homogenous bacteria, the attached growth biomass also have some other advantages compared to the suspended growth biomass such as (i) more capability of withstanding shock loading (Park *et al.*, 1999) and (ii) a more effective substrate biodegradation capacity (Walker and Weatherley, 1999).

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Table 6.2: Steady-state MLSS in the MBRs

PAC (g/L)	MLSS (g/L)	'Excess' MLSS ^(a) (g/L)
0	3.5 ± 0.5	0
1	6.0 ± 0.5	1.5
3	9.0 ± 0.5	2.5
5	11.0 ± 0.5	2.5

(a): Excess MLSS = (MLSS-PAC-3.5) g/L

6.3 Effect of PAC on planktonic bacteria

PAC could also play a role in controlling the amount of planktonic bacteria in the mixed liquor. Planktonic bacteria (single suspended cells) and their by-products (i.e. EPS; TOC etc) in the supernatant could plug or block the pores of the membrane leading to fouling problems. If planktonic bacteria deposit and attach to the membrane surface, they could also colonize and start to form biofilms on the membrane. Therefore, it may help to control fouling if the population of planktonic bacteria could be reduced in the supernatant. It is also important to note that critical flux (for deposition) tends to increase with particle size (for sizes > about 0.3 µm). This means large floc are unlikely to deposit but small individual bacterial are much more likely to reach the membrane.

To check the effectiveness of fresh PAC in reducing the planktonic bacteria in the supernatant, two different methods were used as described in section 3.2.8. The methods are summarized below. Firstly, after the 24 hours of PAC contact, samples were taken and allowed 10 minutes of free settling after which bacteria were enumerated by standard plate-count protocol. The results are shown as plate-count data in Figure 6.2. The second method involved measurement of optical density by UV spectrophotometry on settled (10 minutes) samples and centrifuged (30 seconds at 13,000 rpm) samples; the results are shown in Figure 6.2 as 'before' and 'after' centrifugation. The methods aimed to quantify the bacterial concentrations in the supernatant after mixing the pure culture bacteria at a concentration of 0.6 ± 0.1 g/L with PAC inventories of 0, 0.1, 0.2, 0.3, 0.4 and 0.5 g/L for 24 hours. The results in

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Figure 6.2 reveal that the PAC was able to reduce the planktonic bacteria concentration in the supernatant. The best result was for the highest loading of PAC. This was probably caused by the adsorptive effect of the PAC.

Figure 6.2: Effect of different PAC concentrations in suspended bacteria reduction

The adsorptive process may also involve adsorption of organic solutes onto the PAC surface as a conditioning film. The adsorbed substrate would provide an attraction to the viable planktonic bacteria which would colonize the rough surface of the PAC. Confocal images in Figure 6.3 supports this view by clearly showing the attachment of the bacteria on the PAC surface. Each photo is divided into three types of observation, namely live/dead, dead and normal optical images (from left to right). The green, red and yellow colors on the photos represent the live, dead and the mixture of live and dead bacteria respectively. The dominance of the green color in each photo shows that most of the bacteria on the PAC surface were living. The predominant adsorption of living bacteria onto the PAC supports the significant drop in culturable bacteria in the supernatant (plate count data in Figure 6.2). The less significant drop in optical density of the supernatant may be due to a larger proportion of nonculturable (possibly dead) bacteria, as well as a back-ground of

Figure 6.3: Confocal images of suspended flocs (a) and attached growth bacteria (b)-(f) with different PAC concentrations [Note: live bacteria show green, dead show red and combination of live and dead bacteria show yellow]

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PAC fines not readily removed by settling or even centrifugation (30sec@13000rpm). However, the results in Figure 6.2 and 6.3 provide strong evidence that PAC in the bioreactor could decrease the population of the viable planktonic cells in the supernatant and thereby help to reduce the rate of attachment of such bacteria to the membrane surface and subsequently decrease the fouling rate.

6.4 Short-term filtration tests

Another beneficial effect of PAC in fouling control was that the PAC was able to increase the ‘sustainable’ flux of the MBR identified by flux-stepping. The ‘sustainable’ flux measurement test was made in each MBR (see section 3.2.5) and is derived from the ‘critical’ flux concept (section 2.5.1) and in this study is taken as the highest flux for which dTMP/dt is negligible. Figure 6.4 shows that the MBR (AS) without PAC had a ‘sustainable’ flux of about 10.4 L/m²/hr. For the MBR (BAC) with 1 g/L of PAC, a significant increase in the ‘sustainable’ flux to about 31.0 L/m²/hr was obtained. The ‘sustainable’ fluxes for the MBR (BAC)s with 3 and 5 g/L of PAC were about 36.0 L/m²/hr and 41.0 L/m²/hr respectively. It is interesting to note that the results from the short-term flux-stepping tests to identify flux mirror qualitatively the results of the long-term trials (Figure 6.1).

Figure 6.4: Sustainable flux tests for MLSS with different PAC concentrations, measured in the individual MBRs

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Short term unstirred tests were also carried out to obtain values for specific cake resistance (SCR), flux decline profile and irreversible fouling resistance for the 2L MBRs (the protocols are detailed in section 3.2.5). Table 6.3 and Figure 6.5 show that all the MBR (BAC)s with PAC had better results compared to the MBR (AS) in terms of SCR, flux decline profile and irreversible fouling resistance.

Table 6.3: Irreversible fouling (% increase in R_m) and specific cake resistance for MBRs with different PAC loadings

PAC (g/L)	7 day tests on 2L MBRs (% increase)*	Short-term dead-end tests on MBRs sample (% increase)*	Specific cake resistance (m/kg)
0	234.6	41.2	$237.00 \times 10^{12} \pm 39.90 \times 10^{12}$
1	178.0	8.0	$5.13 \times 10^{12} \pm 0.91 \times 10^{12}$
3	160.3	6.4	$2.19 \times 10^{12} \pm 1.21 \times 10^{12}$
5	108.9	5.6	$4.35 \times 10^{12} \pm 2.06 \times 10^{12}$

* (% increase): $[(R_{\text{fouled}} / R_{\text{initial}}) - 1] \times 100$

Figure 6.5: Flux decline profiles for fixed pressure (100 kPa) and unstirred dead-end filtration, using Milipore UF Polysthersulfone membrane

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This may be because the BAC layers formed on the membrane surface had a higher porosity and was less compressible though from smaller floc sizes; similar observations were made by Kim *et al.*, 1998. Figure 6.6 includes the D_{50} data (both number and volume based), and it is evident the BAC floc were smaller. This decrease in size relate to the presence of small PAC particles in the BAC. However specific cake resistance (SCR) is more sensitive to cake porosity than to 'particle' size. The porosity would depend on compressibility and 'void' filling with material such as EPS, fine colloids etc.

The results of both the SCR (Table 6.3) and flux decline profiles (Figure 6.5) show that the MBR (BAC) with 3 g/L of PAC was slightly better than the MBR (BAC) with 5 g/L of PAC. This may be because the BAC flocs in the MBR (BAC) with 3 g/L of PAC were the biggest as shown in Figures 6.6. Other data on floc properties are also given in Figure 6.6 which shows size distributions and BAC floc images. However, the floc size is probably not the determining factor. This is illustrated by comparing the performance of the MBR (BAC) with 5 g/L of PAC with that at 1 g/L. The MBR (BAC) with 5 g/L of PAC had the better performance in the SCR and flux decline profile tests compared to the MBR (BAC) with 1 g/L of PAC though formed from smaller BAC floc sizes (compared D_{50} number based). The reason may be that the supernatant TOC concentration in the MBR (BAC) with 1 g/L of PAC was higher (at 11.0 ± 2.5 mg/L) than that of in the MBR (BAC) with 5 g/L of PAC (at TOC 7.0 ± 2.5 mg/L) as shown in Figure 6.7. TOC includes not only unused substrate but also extracellular polymeric substances (EPSs), such as polysaccharides and proteins, believed to foul membranes (Huang *et al.*, 2001). The TOC would foul the membrane by plugging or blocking the pores and also cause the BAC cake formed on the membrane surface to be less porous by filling the gap between the flocs. This could account for the worse filtration performance at 0 g/L PAC which had the highest TOC level.

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Figure 6.6: Distribution (a) and images (b) of flocs from 2L MBRs in mixed liquor with different PAC concentrations

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From the results of the short-term tests it is probable that both floc size and fine foulants (i.e. TOC; colloids; EPS etc) play major roles affecting the results of the SCR and flux decline factor. However, cake formation on the membrane surface is not a typical scenario of the MBR, which is normally operated at modest flux and is equipped with air sparging to prevent the biofloc deposition. A greater concern is irreversible fouling caused by pore blocking/plugging by fine foulants. The PAC in the MBR (BAC) was found to be able to control the irreversible fouling significantly as compared to the MBR (BAC) in unstirred dead-end tests (see Table 6.3, resistance increment (%)). Similar results were obtained in separate longer-term MBR tests. These tests were based on the 2L MBRs.

Four clean membranes were tested for their intrinsic resistance with Milli-Q water before submerging into the 2L MBRs with different PAC concentrations. After seven days of filtration at a flux of 21.0 L/m²/hr except for MBR (AS) at the fluxes of 21.0, 10.5 and 5.3 L/m²/hr, the membranes were washed with clean water to remove the reversible fouling. The membranes were then tested again with Milli-Q water to check for their increment in resistance caused by irreversible fouling. The results are shown in Table 6.3 ('7 day tests'), which also includes the short-term dead-end data. Both tests show that the best performance of the MBR in terms of irreversible fouling control was for the MBR (BAC) with the highest PAC concentration.

In brief, the short terms test in measuring specific cake resistance (Table 6.3) and flux decline profiles (Figure 6.5) showed that 3g/L BAC was better than that of 5g/L BAC. This was due to (i) the bigger particle size (Figure 6.6) of the 3 g/L BAC than that of the 5 g/L BAC (the smaller size of the 5 g/L BAC was due to its higher PAC concentration compared to 3 g/L BAC) and (ii) the comparison was under relatively high flux (>80 L/m²/hr), unstirred, dead-end and cake formation allowed filtration conditions. These were not typical operating conditions of the MBR.

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However, in the suspension (stirred, aerated or crossflow) condition (typical operating condition of the MBR), both the short term (Figure 6.4) and long terms (Figure 6.1) filtration comparison tests showed that the 5 g/L BAC performed better than that of 3 g/L BAC. 5 g/L BAC also performed better as compared to 3 g/L BAC in respect of irreversible fouling control. It means that the comparison results obtained under suspension condition and in terms of irreversible fouling are more representative of the real performance of the MBR.

It is of interest to compare the three short term tests to evaluate which is the best predictor of long term performance of the MBR with different PAC concentrations. Both the 'sustainable' flux and irreversible fouling tests were in qualitative agreement with the long term runs. However the SCR implied that the MBR (BAC) with 3 g/L had the best performance instead of the MBR (BAC) with 5 g/L of PAC (Table 6.3). As a result, both the 'sustainable' flux (by flux-stepping) and the irreversible fouling short term tests were judged the better predictors for the long term performance of the MBRs. Both had their own advantages as a long term run predictor. The irreversible fouling test only took about 5-10 minutes to get the results of a sample. The total time was about half an hour for this test to predict the long term performance of the MBRs. The 'sustainable' flux test consumed about 2 1/2 hours (if run as simultaneous tests) to predict the long term performance. However, the flux-stepping test could give an indication of the suitable 'sustainable' operating flux for each MBR which helps to prevent premature membrane fouling or delaying the TMP 'jump'. This information could not be provided by the irreversible fouling test.

6.5 Potential flux-enhancement mechanisms

Four mechanisms were considered to explain the function of PAC in the MBR in irreversible fouling control, namely:

- (i) PAC can act as a protective layer on the membrane surface.
- (ii) PAC helps to reduce the fine foulants reaching the membrane surface through adsorption.

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- (iii) PAC has a scouring effect that prevents the fine foulants accumulation on the membrane surface.
- (iv) PAC would form BAC which is equipped with simultaneous adsorption and biodegradation effects that help to decompose the high molecular weight organics which tend to foul the membrane into a lower molecular weight range more effectively than AS flocs.

Mechanism (i) is considered to be less likely as cake formation on the membrane surface requires the MBR to operate above the critical flux of the biofloc which itself will be a fouling scenario. This suggests that the protective layer would not be the primary mechanism for PAC in irreversible fouling control. Therefore, the other three mechanisms have been considered more relevant and examined in detail.

6.5.1 PAC as an adsorbent

PAC is recognized as a good adsorbent to reduce the fine foulants in the wastewater. Section 6.3 describes the planktonic cell reduction by PAC which is an example of the good adsorptive effect of PAC. Figure 6.7 shows that TOC concentration in the supernatant was 41.0 ± 13.1 , 11.0 ± 2.5 , 8.2 ± 2.0 , 7.0 ± 2.5 mg/L for the MBR (AS) without PAC and the MBR (BAC)s with 1, 3, 5 g/L of PAC respectively. This may be partially due to the slightly higher biomass concentrations in the MBR (BAC)s (see section 6.2). The reduction of the TOC in the supernatant may also due to the adsorptive effect of fresh PAC. This explanation is supported by the results for the TOC concentrations in the permeate which were 15.5 ± 2.1 , 8.8 ± 1.6 , 7.0 ± 1.1 , 5.1 ± 0.8 mg/L for MBR (AS), MBR (BAC) with 1, 3 and 5 g/L PAC respectively. The more PAC in the MBR, the better the quality of the permeate in terms of TOC concentration (Figure 6.7).

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Figure 6.7: Effect of different PAC concentrations on the TOC in the supernatant and permeate and on the membrane surface

To check the adsorption of components of the supernatant, adsorption isotherm tests were carried out with PAC at different concentrations (0-12 g/L). The adsorbates used in the tests were TOC and polysaccharides in the supernatant. These TOC (Huang *et al.*, 2001; Wang *et al.*, 2006) and polysaccharides (Fan *et al.*, 2006; Rosenberger *et al.*, 2006; Wang *et al.*, 2006; Zhang *et al.*, 2006b; Grelrier *et al.*, 2006; Drews *et al.*, 2006) parameters were used as characteristic adsorbates because they have been identified as representing potentially the main components that foul the membranes in MBRs. The test for TOC was performed at three different times of 0.5 hour, 1 day and 3 days, whereas the test for polysaccharide was done after a contact time of 90 minutes. It was found that the adsorption of TOC achieved equilibrium concentration between day 2 and 3. The results of the adsorption isotherm tests for TOC and polysaccharide are shown in Figures 6.8 (a) and (b). The test results followed the Freundlich equation well for TOC ($r^2 = 0.89-0.91$) and polysaccharide ($r^2 = 0.86$) respectively. All of the results discussed showed that PAC could decrease the fine foulants through adsorption.

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Figure 6.8: Adsorption isotherms for (a) TOC and (b) polysaccharide in supernatant

Further evidence of the adsorption phenomena with PAC and mixed liquor is shown in Figure 6.9. This figure shows that the pore volume of the used PAC (BAC from MBR operated at SRT 30 days) was much lower than that of fresh PAC (only about 10% of fresh PAC). This means that the PAC pores were saturated with fine pollutants during the adsorption process in the AS. The results suggest that to maintain good performance by the PAC based on its adsorption mechanism in the MBR (BAC), the aged BAC should be replaced or run at shorter SRT (≤ 30 days).

Figure 6.9: Pore volume comparison of fresh and used PAC

In brief, Figure 6.9 shows that the BAC operated at the SRT 30 days had only retained about 10% of its original pore volume. This indicates that the aged BAC had lost its adsorption effect and means aged BAC also lost its beneficial characteristics in terms of simultaneous adsorption and biodegradation. Without the adsorption effect, the fine aged BAC would not help in the fine particles reduction through adsorption and instead increases the fine particles concentration in the MLSS. Higher fine particle concentration would have more tendency to deposit on the membrane surface and cause more serious fouling. The aged BAC would also increase the MLSS concentration and these would constraint the movement of the membrane fibres and the distribution of the bubbles and encourage more attachment of the fine particles on the membrane surface.

6.5.2 PAC as scouring agent

Apart from adsorption, another possibility is that PAC has a scouring effect in preventing or minimizing the sludge and fine pollutants deposition on the membrane surface to reduce membrane fouling. To examine this potential scouring role, an autopsy of the fouled membranes after use in 7 days filtration tests was carried out. 0.5L Milli-Q water was used to flush the reversible sludge and fine foulants from the membrane surface. The collected detached sludge and fine foulants in the 0.5L Milli-Q water were analyzed (see section 3.2.7) in terms of

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EPSs concentrations, sludge concentration and particle size as shown in Figure 6.10 and Table 6.4. It was found that the floc sizes in the mixed liquor and formed on the membrane surface (Table 6.4 and Figure 6.11) of the MBR (BAC)s were smaller than that of MBR (AS). However, the filtration performance of MBR (BAC)s was better than that of MBR (AS). The particle sizes (number) for the MBR (BAC)s with 5 g/L of PAC were the smallest yet the filtration performance was the best. This may be due to the adsorption effect of the PAC as discussed. It was found that the cake concentrations for the MBR (BAC)s with 1 and 3 g/L PAC were higher than that of MBR (AS) (Table 6.4). However, the percentage (%) (suspended solids on the membrane surface to MLSS in the bioreactor) of sludge attached on the membranes was less for the MBR (BAC)s compared to the MBR (AS) (Table 6.4).

Figure 6.10: Effect of different PAC dosages on the attachment of EPSs on the membrane surfaces

The least was for the MBR (BAC) with 5 g/L of PAC (1.2 %) and followed by the MBR (BAC)s with 3 g/L PAC (2.50 %) and 1 g/L PAC (3.8 %) and the worst was for the MBR (AS) (5.0 %) as shown in (Table 6.4). The EPSs (total EPS = polysaccharide + protein) concentrations accumulated on the membrane surface had similar trends where less EPSs concentrations was for the MBR (BAC)s compared with the MBR (AS) as shown in Figure 6.10.

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Table 6.4: Cake concentration (g/L) attached on membrane surface and D₅₀ (μm) of biomass flocs in mixed liquor and attached on the membrane surface

PAC (g/L)	Cake concentration attached on the membrane surface (C) (g/L)* ^A	Concentration of MLSS attached on the membrane surface (%)* ^B	D ₅₀ (number and volume) of biomass floc attached on the membrane surface (μm)	D ₅₀ (number and volume) of biomass floc in the mixed liquor (μm)
0	0.175 ± 0.013	5.0 %	2.82 (number); 133.87 (volume)	3.10 (number); 120.10 (volume)
1	0.226 ± 0.019	3.8 %	2.10 (number); 67.88 (volume)	2.20(number); 37.70 (volume)
3	0.226 ± 0.002	2.5 %	2.10 (number); 55.48 (volume)	2.30 (number); 40.80 (volume)
5	0.129 ± 0.004	1.2 %	1.43 (number); 38.17 (volume)	0.80 (number); 38.70 (volume)

$$*^A (C) = \frac{C_1 \times 0.5L \text{ (water used to detached the sludge on the membrane surface)}}{2L \text{ (Working volume of MBR)}}$$

$$C_1 \text{ (g/L)} = \text{Sludge (on membranesurface) in 0.5L (amount of clean water used to detach sludge)}$$

$$*^B = \frac{\text{Sludge concentration attached on the membrane surface (C)} \times 100\%}{\text{MLSS in MBR}}$$

The best result was for the MBR (BAC) with 5 g/L PAC (total EPSs = 8.51 ± 1.38 mg/L) and followed by the MBR (BAC)s with 3 g/L PAC (total EPSs = 11.05 ± 1.08 mg/L) and 1 g/L PAC (total EPSs = 15.26 ± 1.07 mg/L). MBR (AS) had the most EPSs (total EPSs = 28.00 ± 0.34 mg/L) attached on its membrane. Figure 6.7 (p120) also shows that the TOC concentration on the membrane surface was relatively lower for MBR (BAC)s as compared to the MBR (AS). This indicates that PAC could create a scouring effect on the membrane surface to minimize foulants attachment.

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Figure 6.11: Distribution (a) and images (b) of biomass flocs on the membrane surface from 2L MBRs with different PAC concentrations

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In support of the scouring mechanism, it is useful to report separate experiment on a fouled membrane from the 20L bioreactor (described in chapter 4). The fouled membrane from the bioreactor (20 L) was shifted to a small vessel (3.5 L) containing only Milli-Q water and coarse aeration. Initially, the TMP dropped from approximately 49 to 41 kPa (Figure 6.12), possibly due to detachment of some fine foulants on the membrane surface into the Milli-Q water. After filtering the Milli-Q water for about five hours, it was observed that the TMP increased from about 41 to 42 kPa and became stable for the next 7 hours. This indicates that some of the detached fine pollutants in the Milli-Q water had been returned to the membrane surface during filtration.

Figure 6.12: Effect of particle scouring on the TMP reduction

After half a day of filtration when the TMP was at steady state, 5 g/L of powdered hollow glass beads (GB) of narrow size distribution of about 20 μm , with non-adsorptive properties were added into the reactor. An immediate drop of the TMP was observed from about 42 to 40 kPa after two days of filtration (Figure 6.12). This shows that the GB helped to detach some of the reversible fine foulants back to the Milli-Q water. This test supports the claim that particles, such as PAC, have scouring capabilities able to detach flocs and fine foulants accumulated on the membrane surface. There is previous evidence from other membrane studies (Fane, 1984) that supramicron particles reduce the rate of fouling by enhancing back-

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transport of fouling species. It should be noted that the reversal of fouling, shown by TMP decline was relatively small. This may be due to the degree of fouling which was rather severe (TMP approaching 50 kPa). The scouring efficiency may be greater with less consolidated fouling.

To summarize at this stage it appears that the PAC in the MBR (BAC) has potentially important roles in terms of adsorption (reducing supernatant TOC and planktonic bacteria [section 6.3]) and particulate scouring (foulant removal or increasing back transport). Furthermore, as shown in section 6.2, the PAC appears to enhance MLSS development and this could be linked to substrate adsorption. The other important factor observed in Chapter 4 is that ‘aged’ BAC does not enhance flux, but makes fouling worse. A portion of fresh PAC, added to replenish BAC removed with waste sludge, is required. This observation tends to favor an ‘adsorption’ role for PAC, although fresh PAC may have ‘scouring’ properties until incorporated into large biomass floc. Thus it is still ambiguous whether adsorption or scouring is the more important mechanism controlling fouling.

In order to further evaluate the dominant mechanism, another series of tests were performed. The 3.5L MBR (see section 3.2.2) was operated for 20 days (no sludge wastage through out the operation period) and run at a flux of 7.9 L/m²/hr. As shown in Figure 6.13, the TMP rose from 10 to 32 kPa over a period of 30 hours of operation. At day 21.2, 5 g/L of GB with mean particle size of 25 µm (consisting of 3 types of GB with sizes of 5, 20 and 50 µm in same proportion to give a size distribution) were put into the bioreactor and a change in the ‘slope’ of the TMP rise (dTMP/dt) trend was observed. The dTMP/dt reduced from 35.3 to 19.4 kPa/day. This demonstrates that the addition of GB decreased the fouling rate. The GB is not an adsorbent and this indicates that scouring/collision and not the adsorption effect took place in the MBR from day 21.20 to 21.88. The same amount (5 g/L) of PAC (D₅₀ = 25 µm) was then added to the bioreactor at day 21.88. This caused a sudden drop of the TMP from 43.7 to 40.2 kPa. The TOC in the supernatant was also reduced from 135.2 to 87.7 mg/L. The reduction of the TOC was mostly caused by adsorption by the PAC. This comparison shows that the

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performance of the PAC was better than that of the GB. The results confirm that both scouring and adsorption mechanisms could play a role, and PAC has an advantage being capable of both beneficial effects.

Figure 6.13: Performance comparison between scouring and adsorption on TMP reduction

When a similar series of ‘particle’ additions were carried out at a higher flux, a qualitatively different result was obtained. In this case, the test was carried out in one of the 2L MBRs which had been cultivated for 10.0 days (no sludge wastage) at a flux of 16.0 L/m²/hr instead of 7.9 L/m²/hr (Figure 6.14).

After filtering the bioreactor with a new membrane for about 3 hours on day 10.12, the same addition of glass beads used in the previous experiment was put into the 2L bioreactor (Figure 6.14). This experiment had a higher flux (16.0 L/m²/hr) but a lower MLSS concentration (3.3 g/L) and a higher superficial gas velocity (SGV) (9.0 mm/s) compared to the previous experiment where the MLSS and SGV were 7.5 g/L and 6.0 mm/s respectively. It was expected that the TMP rise would be slowed down with the help of the GB. On the contrary, a more rapid TMP rise was observed as shown in Figure 6.14. The dTMP/dt changed from 95.8 to 116.6

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kPa/day which means that the GB had accelerated the fouling rate rather than reducing it as reported in Figure 6.13.

Figure 6.14: Performance comparison between collision and adsorption on TMP reduction (flux: 16.0 L/m²/hr)

This indicates that the use of fine GB to create a scouring effect to reduce fouling rate may only be significant if operated at a relatively low flux. At the higher flux, the fine particles would tend to deposit on the membrane surface and contribute to cake formation. This may undermine the scouring effect of the GB and the ability to detach fine particles. In addition, at the higher flux, the GB had a tendency to accumulate around the membrane module and obstruct the movement of the membrane and aggravate the fouling intensity. GB (or any other particles) have their own ‘critical flux’ and they also increase the solids concentration. This changes the ‘sustainable flux’ of the MBR, and therefore, preliminary determination of the ‘sustainable flux’ of the MBR with the ‘collision agent’ particle would be important to prevent premature fouling. The scouring/collision mechanism is discussed further in Chapter 7 (section 7.2) and 8 (section 8.3).

At day 10.17, 5 g/L PAC was added into the 2L bioreactor, following which dTMP/dt and TOC concentrations were reduced from 116.6 to 39.7 kPa/day and 51.5 to 16.2 mg/L respectively as shown in Figure 6.14. Thus both experiments, operated at fluxes 7.9 and 16.0 L/m²/hr respectively, showed that PAC could help to

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control the fouling rate more effectively than that of GB. This suggests that the adsorptive effect was more significant than that of the scouring effect for the MBR (PAC). However, the scouring mechanism should also be credited with a contribution to fouling control if under appropriate flux operation.

6.5.3 BAC for adsorption and biodegradation

From the discussion above and the comparison of results shown in Figures 6.13 and 6.14, the role of PAC in adsorbing the fine foulants was judged to be probably the primary mechanism in reducing fouling rate. However, the adsorption capacity of PAC was easily reduced. This is supported by Figure 6.13. At day 21.88, the reduction of TMP confirms the effect of adsorption in fouling control, however, the significant effect was only over a very short duration (≈ 8 hours) and subsequently the TMP was observed to increase again.

Nonetheless, the benefit of BAC cultivated at SRT 30, and especially 10 days in membrane fouling control is undeniable. In addition, the enhanced MLSS observed in the MBR (BAC) (section 6.2) suggests an additional role. It is possible that simultaneous adsorption and biodegradation may be important coupled mechanisms in the MBR (BAC) that helps in fouling prevention. In order to examine this hypothesis, a series of tests were carried out. Two 3.5 L reactors were filled with well acclimatized activated sludge (AS) from the lab-scale MBR which had been cultivated for about three years. 5 g/L PAC was added to one of the reactors. After aeration for about 20 hours, the supernatant and permeate samples from the both reactors were analyzed for their dissolved organics distribution in terms of molecular weight (MWt) and TOC as shown in Tables 6.5 and 6.6. The MWt distributions were obtained by HPSEC as detailed in section 3.2.8. It was found that PAC could not only reduce the TOC concentrations but also enhance the biomass capability to decompose high MWt dissolved organics into lower MWt components in the supernatant of the mixed liquor.

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Table 6.5: Mass percentage molecular weight (MWt) of dissolved organics and dissolved organic carbon (DOC) distributions in supernatant of mixed liquor

Molecular weight distributions (%) of dissolved organics	100 > MWt	100 < MWt < 1k	1k < MWt < 10k	10k < MWt < 100k	100k < MWt < 1000k	1000k < MWt
AS (total: 100 %)	4.62 ± 1.89	0	0	4.18 ± 2.96	69.12 ± 5.71	22.08 ± 6.85
AS + PAC (total: 100 %)	0.02 ± 0.03	0.58 ± 0.36	13.10 ± 2.72	71.74 ± 1.22	13.74 ± 2.45	0.83 ± 0.22
Particle size (PS) distributions (µm) of DOC	≈ 0.0004 > PS	≈ 0.0004 < PS < ≈ 0.002	≈ 0.002 < PS < ≈ 0.005	≈ 0.005 < PS < ≈ 0.05	≈ 0.05 < PS < ≈ 0.5	≈ 0.5 < PS
AS (DOC) (Total: 5.996 ± 0.28 mg/L)	0.356 ± 0.023	0	0	0.322 ± 0.021	5.320 ± 0.354	N.A
AS + PAC (DOC) (Total: 2.965 ± 0.13 mg/L)	≈ 0	≈ 0	0.392 ± 0.024	2.144 ± 0.133	0.411 ± 0.025	N.A

N.A: Not applicable (samples had been gone through 0.45 µm filter)

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Table 6.6: Mass percentage molecular weight (MWt) of dissolved organics and dissolved organic carbon (DOC) distributions in permeate

Molecular weight distributions (%) of dissolved organics	100 > MWt	100 < MWt < 1k	1k < MWt < 10k	10k < MWt < 100k	100k < MWt < 1000k	1000k < MWt
AS (total: 100 %)	4.62 ± 1.82	0	0	2.77 ± 1.12	79.57 ± 2.17	12.82 ± 2.27
AS + PAC (total: 100 %)	0.05 ± 0.12	0.03 ± 0.07	3.67 ± 1.19	61.69 ± 2.17	31.61 ± 3.24	2.95 ± 0.20
Particle size (PS) distributions (µm) of DOC	≈ 0.0004 > PS	≈ 0.0004 < PS < ≈ 0.002	≈ 0.002 < PS < ≈ 0.005	≈ 0.005 < PS < ≈ 0.05	≈ 0.05 < PS < ≈ 0.5	≈ 0.5 < PS
AS (DOC) (Total: 1.647 ± 0.18 mg/L)	0.088 ± 0.014	0	0	0.052 ± 0.008	1.507 ± 0.233	N.A
AS + PAC (DOC) (Total: 1.598 ± 0.20 mg/L)	≈ 0	≈ 0	0.061 ± 0.011	1.016 ± 0.180	0.521 ± 0.093	N.A

N.A: Not applicable (samples had been gone through 0.45 µm filter)

The analysis results (Table 6.5) revealed that more than 90% of the MWt distribution of dissolved solids in the supernatant of the MBR (AS) was above 100k, whereas, for MBR (BAC) (i.e. AS + PAC), the dissolved organics in the supernatant were mostly below 100k (≈ 85%) or in the range of 10k-100k (≈ 72%). The TOC concentration in the supernatant of AS + PAC was also lower (2.97 ± 0.13 mg/L) compared to AS (6.00 ± 0.28 mg/L). The shift in the MWt distribution of dissolved solids from higher (100k-1000k; >1000k) to lower ranges (<100k) seems to indicate that the high MWt dissolved organics were better decomposed by biomass in the presence of PAC. The possible mechanisms involved in decomposition of high MWt dissolved organics in AS + PAC are discussed below.

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- a) Porous PAC with good adsorption capacity could become a place for (i) relatively high organic substrate loading (Walker and Weatherley, 1999; Dosoretz and Bøddeker, 2004) and (ii) attachment sites for immobilized bacteria which have better substrate biodegradation ability (Walker and Weatherley, 1999). These conditions could enhance decomposition of adsorbed high MWt dissolve organics into lower MWt ranges by immobilized bacteria. The lower MWt dissolved organics could be further biodegraded and adsorbed by BAC. This may also explain the lower TOC concentration in the supernatant and higher biomass concentration of AS + PAC as compared to AS. More importantly, the lower range (<100k) dissolved solids would not easily block and plug the pores ($\approx 0.5 \mu\text{m}$) of the membrane used in this project study. However, with the formation of a conditioning biofilm on the membrane surface, the membrane pores are likely to be effectively smaller than $0.5 \mu\text{m}$. When the bulk supernatant liquid is filtered through the membrane, the conditioning biofilm may help to further biodegrade the low MWt organics and block and prevent the high MWt species from passing into the permeate (high MWt organics may need some extended time to be gradually biodegraded), but this would come with the penalty of a fouling problem. Results from Table 6.6 illustrate this picture more clearly. Almost 72 % of the high MWt (100k-1000k) TOC concentration ($5.32 \pm 0.35 \text{ mg/L}$) in the supernatant of the MBR (AS) was prevented from transmission to the permeate side (at TOC: $1.51 \pm 0.23 \text{ mg/L}$), some of the TOC retained by the membrane may create a fouling issue if not successfully biodegraded or decomposed by the attached conditioning biofilm on the membrane surface. On the other hand, about 50% of the lower MWt (10-100k) TOC concentration ($2.14 \pm 0.13 \text{ mg/L}$) in the supernatant of the MBR (BAC) freely passed out from the membrane into the permeate (at TOC: $1.02 \pm 0.18 \text{ mg/L}$). The other 50% of the TOC concentration could be blocked by the membrane, but could be expected to be biodegraded more rapidly by the conditioning biofilm compared to the high MWt organics. This difference in supernatant MWt distribution could

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be an important mechanism enable by the PAC that provides a reduction in membrane fouling for the MBR (BAC).

- b) Another potential benefit was suggested by Dosoretz and Bøddeker, (2004). They noted that below a certain substrate threshold concentration there would be difficulty to further biodegradation due to the lack of enzyme induction. To solve this problem, PAC could act as a sink for accumulation of the substrate and bacteria. Immobilized bacteria could react better with the relatively higher substrate concentrations accumulated at the surface and in the pores of the PAC. This may be another explanation for the better decomposition of high MWt dissolved organics in the MBR (BAC).

The presence of PAC in AS would form BAC that could induce simultaneous adsorption and biodegradation mechanisms. This simultaneous process could (i) improve high MWt dissolved organics decomposition and (ii) partly bioregenerate the saturated BAC (Walker and Weatherley, 1999): This would help to prolong the life-span of the BAC in membrane fouling control.

In summary, it is agreed that the ratio of PAC/biomass is important in membrane fouling control. Higher PAC/biomass ratio may be more beneficial in membrane fouling control but because PAC replenishment is required in order to see the good effect of BAC in fouling control, the added cost and management become a restriction. The parameter of PAC/biomass ratio would also become irrelevant in membrane fouling assessment if the aged BAC was not replaced by the fresh PAC.

In relative terms, the sludge retention time (10-30 days) [volume of the bioreactor/sludge wastage rate] (SRT) should be more important than that the ratio of PAC/biomass as a parameter for guidance in membrane fouling assessment. Lower SRT (higher BAC replenishment rate) would have a better performance in the MBR (BAC) system (please refer to chapter 7; section 7.3) in terms of TOC concentration in the supernatant and fouling control.

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It was found that the MBR (BAC) cultivated at relatively longer SRTs (pseudo infinity) tends to produce relatively higher TOC concentration in the supernatant (please refer to chapter 7; section 7.3), also the aged BAC would loss its good characteristics in controlling membrane fouling which is involved (i) reducing the specific cake resistance (SCR), (ii) adsorption of single cells (planktonic cell support) (iii) adsorption of TOC and polysaccharides, (iv) scouring by collision effects and (v) simultaneous adsorption and biodegradation. Therefore to gain the optimum benefit of the MBR with PAC, it is necessary to have regular replenishment of the aged BAC with fresh PAC.

In addition, good hydrodynamics (aeration distribution) of the bioreactor is necessary to prevent the attachment of the BAC to the membrane surface. The good effect of the BAC in membrane fouling control would decrease if attachment of the BAC to the membrane is allowed (please refer to chapter 7; section 7.1). Park et al., (1999) also found that PAC would have better fouling control with relatively higher shear force on the membrane surface.

6.6 Performance of 20L MBR (AS) and MBR (BAC)

To confirm the ability of 5 g/L of PAC in reducing membrane fouling, the two larger MBRs with working volumes of 20 L were set-up at SRT 10 days and 5 g/L of PAC was added to one of them. A constant top-up of the lost PAC (10.0 wt%) in the wasted sludge was provided daily to keep the PAC concentration constant in the MBR (BAC). In order to make a fair comparison for these tests, both MBRs were operated in parallel at a fixed permeate flowrate of 40 mL/min (flux = 22.00 L/m²hr, HRT = 8.3 hrs) to achieve a steady state. The MLSS for the MBR (AS) reached 3.3 ± 0.20 g/L and for the MBR (BAC) reached 10.3 ± 0.20 g/L. This again showed the ability of the BAC to encourage biomass growth with an 'excess' of about 2.0 g/L (10.3 - 5 g/L PAC - 3.3 g/L AS)

At day 52, new membrane modules with an area of 0.115 m² were submerged into each MBR and run at a fixed flux of 22.0 L/m²/hr. The key parameter used to

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characterise fouling was the TMP. The results are shown in Figure 6.15 which clearly indicates that the MBR (BAC) with 5 g/L of PAC performed better. The average rate of TMP rise was 3.2 kPa/day for the MBR (AS) and only 1.1 kPa/day for the MBR (BAC) throughout the operating period of about 14 days. At day 66, a TMP 'jump' was observed for the MBR (AS). This ceased the operation of the MBR (AS). The membrane would need to be cleaned with chemicals if it was to be used again to continue the filtration process. This phenomenon was not observed for the MBR (BAC).

Figure 6.15: Performance comparison of 20L MBR and MBR (BAC) at SRT 10d

At day 67, SEM images of the fouled membranes from the both MBRs were scrutinized. Samples of membrane were removed and prepared for SEM without water washing but following standard sample preparation protocols (see section 3.2.6). From the images (Figure 6.16), it was found that a layer of cake had formed on both the membrane surfaces. However, it is clearly evident that the layer formed on the membrane surface of the MBR (AS) was denser than that on the membrane surface from the MBR (BAC). The pores of the membrane from the MBR (AS) were no longer observable but the shape of the pores of the membrane from the MBR (BAC) was still clearly visible. This supports the conclusion that PAC helps to prevent fine foulants accumulating on the membrane surface through mechanisms of simultaneous adsorption and biodegradation as well as scouring. Following this, two new membrane modules were submerged into each reactor to

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check for their ‘sustainable’ flux by flux-stepping. It was found that the ‘sustainable’ flux for the MBR (AS) was $< 31.8 \text{ L/m}^2/\text{hr}$ and for the MBR (BAC) $> 44.7 \text{ L/m}^2/\text{hr}$ respectively as shown in Figure 6.17. This further confirms that PAC has the ability to reduce the fouling rate of the MBR (BAC).

Figure 6.16: SEM images of the membrane surface. (a) clean membrane, (b) membrane for the MBR (BAC), (c) membrane for the MBR (AS) at 3,500 x

Figure 6.17: Sustainable flux tests for MBR (AS) and MBR (BAC)

6.7 Conclusions

The efficiency of PAC in enhancing fouling control was proved again in short and long terms tests on mixed liquor from the 2L MBRs (0, 1, 3 and 5 g/L PAC in about

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3.5 g/L of biomass) and 20L MBRs (0 and 5 g/L PAC in about 3.3 g/L of biomass) at SRTs of 10 days. All the short-term tests involving the SCR, flux decline profiles, irreversible fouling and 'sustainable' flux at both fixed pressure and bubbled crossflow showed that the MBR (BAC) with PAC addition were better than that of MBR (AS). In this experiment, the lowest SCR value was for a PAC with 3 g/L instead of 1 g/L as reported in chapter 5 where the MBRs were run at SRT 30 days. Similar trends were obtained for flux decline profiles in deadend filtration. Again, the SCR and flux declines for 0 g/L PAC were the worst. As found in MBRs operated at SRT 30 days (reported in chapter 5), the best result for the short-term tests measuring irreversible fouling resistance and 'sustainable' (low TMP rise) flux was for the 5 g/L PAC addition, and again 0 g/L PAC mixed liquor performed much worse. However, the results from Chapter 5 and in this Chapter 6 should not be closely compared since the operating conditions (SGV, HRT and the source of aeration) are different. A fairer comparison between the MBR (AS)s and MBR (BAC)s at different SRTs is discussed in more detail in Chapter 7.

The long-term runs in the 2L MBRs with submerged hollow fibres operated at a flux of 21 L/m²hr showed that the slowest fouling rate (TMP rise) was for the MBR (BAC) with 5 g/L of PAC, followed by the MBR (BAC)s with 3, 1 g/L of PAC and MBR (AS). Comparing the results of the two 20L MBRs, the average rate of TMP rise was 3.2 kPa/day for the MBR (AS) and only 1.14 kPa/day for the MBR (BAC) throughout the operating period of about 14 days.

Among the short-term predictors, it was found that the 'sustainable' flux and irreversible fouling tests were better predictors than the SCR and flux decline profile. They were more in qualitative agreement with the long term runs as compared to SCR and flux decline profiles. The irreversible fouling test consumed relatively little time and was easy to measure compared to the flux-stepping for the 'sustainable flux'. The advantage of the 'sustainable' flux over the irreversible fouling test was that the 'sustainable' flux was able to give information of the 'sustainable' operating flux for each MBR to prevent a premature TMP 'jump'.

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However, both of methods should only be regarded as rather approximate predictors of long term performance.

The mechanisms of the PAC in controlling fouling involved (i) reducing the SCR, (ii) adsorption of single cells (planktonic cell support) (iii) adsorption of TOC and polysaccharides, (iv) scouring by collision effects and (v) simultaneous adsorption and biodegradation. It was found that the simultaneous adsorption and biodegradation was the primary mechanism in fouling control. Therefore to gain the optimum benefit of the MBR with PAC, it is necessary to have regular replenishment of aged BAC with fresh PAC. However the contributions of the other roles should not be neglected.

CHAPTER 7 OPTIMIZATION OF MBR (BAC)**Synopsis:**

In chapters 4, 5 and 6, it was shown that under conditions of replenishment the addition of PAC to the MBR, to develop BAC, was able to enhance filtration performance compared with a conventional MBR. This chapter addresses two issues,

- (i) Does the size of the PAC have an effect on MBR (BAC) performance; and
- (ii) How does the SRT, which influences both MBR characteristics and PAC replacement rate, influence the MBR (BAC) performance.

To assist in interpretation of the trends, various characteristics have been measured, such as particle/floc size, concentration of MLSS and TOC as well as short-term filtration properties. The long term filtration profiles, in terms of TMP versus time, have been used as the major characteristics of MBR fouling. Due to the relatively long time-scale of the experiments, particularly after changing reactor parameters, the numbers of specific conditions tested and compared has been limited.

7.1 Comparison of MBRs with different PAC particle sizes

The PAC used in this study originated from granular activated carbon (GAC) with a particle size distribution ranging from 300-600 μm . The GAC was ground into two different PAC sizes to test their relative effectiveness in fouling control of MBRs. The two PAC types, namely 'fine' and 'medium', had the particle size distributions shown in Figure 7.1. The D_{50} of the two types of PAC in terms of volume and number was $14.88 \pm 3.93 \mu\text{m}$ and $0.24 \pm 0.06 \mu\text{m}$ (fine) and $67.60 \pm 2.70 \mu\text{m}$ and $1.72 \pm 0.11 \mu\text{m}$ (medium) respectively. [Note: for comparison the PAC used in the earlier chapters had D_{50} (volume) of $24.5 \mu\text{m}$ and D_{50} (number) of $1.6 \mu\text{m}$].

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(a)

(b)

Figure 7.1: Particle size distribution of PAC in volume (a) and in number (b)

Three batch bench-scale 2.0L MBRs, namely MBR (AS), MBR (fine) and MBR (medium) were set-up and initially filled with AS at 1.0 g/L. Then 5 g/L of fine and medium PACs were added to the reactors MBR (fine) and MBR (medium) respectively. The 2.0L MBRs were operated with SRT of 10 days which required sludge wastage of approximately 200 mL per day. This caused a daily removal of 10.0wt% of PAC from the MBR (fine) and MBR (medium). The PAC was topped-up to account for this loss and to maintain constant PAC concentrations in the both MBRs. After cultivating the three MBRs for 36 days ($> 3 \times \text{SRT}$), a series of

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experimental analyses were started to compare the various characteristics of the MLSS in the 2.0L MBRs. MLSS samples were withdrawn from each of the bioreactors and tested in terms of (i) MLSS concentrations, (ii) the mean (D_{50}) and distribution of floc sizes and (iii) TOC concentration in the supernatant and permeate. The results of these tests are shown in Table 7.1 and Figure 7.2. Table 7.1 shows that both types of PAC helped to reduce the TOC in the supernatant and permeate. Figure 7.2 shows the floc size distributions of the AS and BACs from the three MBRs. However, fine PAC was able to decrease the TOC in the supernatant more significantly and produce a better quality of permeate compared to the medium size PAC. This indicates that the fine PAC with its higher surface area than that of medium size PAC was able to adsorb, and facilitate (as BAC) the decomposition and biodegradation of TOC more efficiently. In addition, it was found that the MLSS concentration in the MBR (fine) was higher than the other MBRs as shown in Table 7.1. This suggests that the fine PAC with its higher surface area also had an advantage in enhancing MLSS production. With its greater ability in TOC reduction, it was anticipated that the MBR (fine) would have better performance in fouling control as compared to the MBR (AS) and MBR (medium).

Table 7.1: Concentrations of MLSS and TOC and AS and BAC floc sizes (D_{50}) in the MBRs (between days 36-44)

MBR	MLSS (g/L)	Particle size (μm)		TOC (mg/L)	
		D_{50} (Volume)	D_{50} (Number)	Supernatant	Permeate
AS	3.44 ± 0.03	30.20 ± 0.50	0.65 ± 0.01	90.88 ± 8.55	27.14 ± 3.99
Fine	9.22 ± 0.03	38.15 ± 1.52	0.58 ± 0.01	24.70 ± 1.34	8.50 ± 1.56
Medium	7.67 ± 0.01	90.05 ± 0.52	2.92 ± 0.05	40.08 ± 4.55	10.67 ± 1.25

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(a)

(b)

Figure 7.2: Particle size distribution of AS and BAC flocs in (a) volume and (b) number

To assess the capability of the MBR (fine) in fouling control, new membrane modules with an area of 0.057 m² were submerged into each 2L MBR and run at a fixed flux of 13.5 L/m²/hr at day 36. The results are shown in Figure 7.3. The TMP increased rapidly for the MBR (AS) from 15.0 kPa to approximately 75.0 kPa in 10 hours presumably due to the high TOC concentration in the supernatant. This confirms that high TOC concentration in the supernatant can accelerate the fouling rate. Figure 7.3 also clearly shows that the MBR (medium) performed better than the MBR (fine). In about one and half days of operation, the TMP of MBR (fine)

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increased from about 15.0 kPa to 80.0 kPa whereas the increment of the TMP for MBR (medium) was only from 15.0 kPa to 35.0 kPa. At day 37.6, the membranes from the MBR (fine) and MBR (medium) were removed for inspection.

Figure 7.3: Performance comparison of MBR (AS) and MBR (BAC)s with different PAC particles sizes distribution at SRT 10d (flux: 13.5 L/m²/hr)

It was observed that a layer of BAC attached on the membrane surface for the membrane module from MBR (fine) as shown in Figure 7.4. This may be because the fine PAC contained a significant inventory of fine particles which tended to deposit on the membrane surface during the relatively higher flux filtration and with an aeration rate which was not adequate to detach them. This is supported by the data in Table 7.1 where the fine PAC have shifted the floc size (D_{50}) of the AS in terms of number from $0.65 \pm 0.01 \mu\text{m}$ to a lower range of $0.58 \pm 0.01 \mu\text{m}$. Figure 7.2 (b) also shows the floc size distribution of BAC_{fine} was lower when compared to AS and $\text{BAC}_{\text{medium}}$. The observed deposit of fine BAC on the membrane surface may explain the less impressive performance of the MBR (fine) compared to the MBR (medium). The filtration performance shown in Figure 7.3 does not reflect the lower TOC in the supernatant of the MBR (fine). It shows that the potential advantage of less organic load in the supernatant can be negated by the presence of a significant inventory of PAC fines if the conditions favour particle deposition.

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Figure 7.4: Images of the used membrane modules from the MBR (BAC)s with different PAC particles sizes distribution (a) fine and (b) medium at SRT 10d

To test this hypothesis a new set of membranes were introduced and the flux lowered to $8.5 \text{ L/m}^2/\text{hr}$. The results of the filtration performance of the three MBRs with the lower flux are as shown in Figure 7.5. The best performer was now the MBR (fine). This suggests that to optimise the filtration performance of the MBR (fine) equipped with fine BAC, which enhances TOC reduction, it is necessary to operate at a flux that minimises fine particle attachment to the membrane surface. The appropriate flux will depend on the module design and hydrodynamic conditions.

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Figure 7.5: Performance comparison of MBR (AS) and MBR (BAC)s with different PAC particles sizes distribution at SRT 10d (flux: 8.5 L/m²/hr)

7.2 Comparison of MBRs with different SRTs

Before discussion of the comparative performance of MBR (BAC)s with different SRTs (10d, 30d and long) in section 7.3, the filtration performance of two batch 3.5L MBRs at SRT 10 and 30 days without addition of PAC will be compared and discussed. The sludge was wasted from the MBR (10d) and MBR (30d) at 10.0 wt% and 3.35 wt% respectively daily. After operating the two batch bench-scale 3.5L MBRs for 47 days at an HRT of 3.5 days, new membrane modules with an area of 0.057 m² were submerged into each MBR and run at a fixed flux of 10.0 L/m²/hr. The results are shown in Figure 7.6, which shows that the MBR (30d) had a better filtration performance. Over a 4 day filtration period, the TMP increase was 9.1 kPa/day for MBR (10d) but only 3.3 kPa/day for MBR (30d). At day 4, the membrane in MBR (10d) was fouled seriously and experienced the TMP ‘jump’. Due to the batch mode of operating the MBRs, the permeate from each MBR was returned back to the reactor to keep the average working volume at a constant level. This method means that, the operation of the batch MBRs with different membrane areas and fluxes would not affect the net feed loading rate. At day 6 of the test (day 53 of the reactor operation), both membranes from the MBRs were replaced by new membranes with similar areas (0.057 m²) but operated at a flux of 15.0 instead of

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10.0 L/m²/hr. As expected, the increased flux accelerated the fouling rate. In two filtration days, the TMP increased from 10.8 to 65 kPa (22.0 kPa/day) and 11.6 to 30 (8.0 kpa/day) kPa for MBR (10d) and MBR (30d) respectively as shown in Figure 7.7. The MBR (30d) still significantly outperformed the MBR (10d) in terms of membrane fouling control.

Figure 7.6: Performance comparison of MBRs at SRTs 10 and 30 (flux: 10.0 L/m²/hr).

Figure 7.7: Performance comparison of MBRs at SRTs 10 and 30 (flux: 15.0 L/m²/hr)

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Short-term filtration tests (see section 3.2.5 for protocol) were performed on MLSS from both MBRs. It was found that AS_{30d} had a lower specific cake resistance but a higher irreversible fouling than that of AS_{10d} for short term unstirred tests as shown in Figure 7.8. The higher irreversible fouling may due to the higher polysaccharide and TOC concentrations in the supernatant of MBR (30d) than that of MBR (10d) as shown in Table 7.2.

Figure 7.8: Specific cake resistance and resistance increment (irreversible fouling) for MLSS with different SRTs

Table 7.2: Concentrations of the MLSS and TOC and AS and BAC floc sizes (D₅₀) in MBRs (between days 47-56)

MBR	MLSS (g/L)	Polysaccharide in the supernatant (mg/L)	Particle size (µm)		TOC (mg/L)	
			D ₅₀ (Volume)	D ₅₀ (Number)	Supernatant	Permeate
10d	4.14 ± 0.82	85.97 ± 5.82	23.35 ± 1.19	2.01 ± 0.10	75.30 ± 7.13	14.70 ± 1.88
30d	8.88 ± 0.96	123.58 ± 13.26	44.03 ± 4.99	2.76 ± 0.25	95.85 ± 20.04	15.30 ± 1.63

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However, the better performance of the AS_{10d} in irreversible fouling control was not mirrored in the MBR situation where the aeration was applied. The filtration performance comparisons between MBR (10d) and MBR (30d) at fluxes of 10.0 and 15.0 L/m²/hr showed that the MBR (30d) performed better than that of MBR (10d). The better performance of the MBR (30d) compared to MBR (10d) is mirrored in the smaller SCR as shown in Figure 7.8. The smaller SCR of the AS_{30d} MLSS may have been due to the D₅₀ of AS_{30d} being bigger than that of AS_{10d} as shown in Table 7.2. However, cake accumulation on the membrane surface is not a typical feature of MBR operation as bubbling creates shear forces on the membrane surface to control and minimise deposition.

Even if the flocs do not accumulate on the membrane they may still interact and scour the surface. This could explain the better performance of the AS_{30d} MBR. The D₅₀ and particle size distribution for the AS₃₀ and AS₁₀ are shown in Table 7.2 and Figure 7.9 respectively. The D₅₀ of AS_{30d} and AS_{10d} was $44.03 \pm 4.99 \mu\text{m}$ and $23.35 \pm 1.19 \mu\text{m}$ respectively. The D₅₀ of AS_{30d} was almost twice the size of the AS_{10d}. It is plausible that the bigger floc size could detach the fine foulants from the membrane surface better than the smaller floc when collision occurred between the flocs and the membrane surface.

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(a)

(b)

Figure 7.9: Particle size distribution of AS_{10d} and AS_{30d} and in volume (a) number

(b)

When the flux was raised to 23 L/m²/hr, the MBR (AS) (30d) lost its advantage. At a flux of 23.0 L/m²/hr, the TMP increased from 13.9 to 73.3 kPa within about 30 minutes of operation for MBR (30d), and from 13.4 to 66.5 kPa in about 8 hours for MBR (10d) as shown in Figure 7.10. Neither MBR had a good performance in terms of maintaining the dTMP/dt at an acceptable level.

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Figure 7.10: Performance comparison of MBRs at SRTs 10 and 30 (flux: 23.0 L/m²/hr)

Figure 7.11 attempts to summarize the TMP rise data, plotted as ‘average’ dTMP/dt versus imposed flux. For fluxes in the range 10 to 15 L/m²/hr both MBRs exhibited modest values of TMP rise; the MBR (30d) was particularly effective. This is similar to the results of Zhang *et al.*, (2006a) for a flat sheet submerged membrane. However, for fluxes in excess of about 20 L/m²/hr both MBR (10d) and MBR (30d) showed serious fouling. The implications are that the sustainable MBR fluxes are about 20 L/m²/hr. Short-term flux-stepping tests confirm this as shown in Figure 7.12; both reactors showed a slight rise in TMP at a flux of 17 and a significant rise at a flux of 22.5 L/m²/hr.

Figure 7.11: dTMP/dt and fluxes of MBR (AS)s

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Figure 7.12: Sustainable flux tests for AS_{10d} and AS_{30d}

It is of interest to speculate why the MBR (30d) actually performed worse than the MBR (10d) at the flux of 23.0 L/m²/hr, as shown in Figure 7.10. Firstly, it appears that for both MBRs the flux of 23 L/m²/hr was above the ‘critical’ flux for significant deposition. The rate of fouling (represented by dTMP/dt) can be related to the net flux and the concentration of potential foulants, i.e.

$$\frac{dTMP}{dt} \propto (\text{Flux} - \text{critical flux of foulants}) \times (\text{concentration of foulants}) \quad (7.1)$$

The serious fouling depicted in Figure 7.10 suggests that the foulants included biomass floc as well as supernatant species (TOC, fine colloids etc). Table 7.2 shows that MBR (30d) had twice the MLSS and 50% more polysaccharide than MBR (10d). As a result once the critical flux of these dominant foulants was exceeded the dTMP/dt would be larger for the MBR (30d). Any potential benefit from ‘floc’ scouring of the membrane would be lost if the flux was high enough to cause accumulation at the membrane surface. Thus in order to achieve the filtration benefit of the longer SRT, the imposed flux needs to avoid significant deposition. Values higher than the 23 L/m²/hr should be possible with a better module design and/or improved hydrodynamic air scouring.

7.3 Performance comparison between MBR (BAC)s with different SRTs

For the MBR (BAC), the BAC with simultaneous ‘collision’ and adsorption effects may work better in controlling foulants on the membrane surface compared to AS

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floc with only a ‘collision’ effect. This means that as long as the BAC is not saturated and embedded by the biomass floc or sticky fine pollutants, it could help to improve the filtration performance of the MBR even if the D_{50} becomes smaller.

Investigation of the filtration performance of three batch bench-scale 3.5L MBR (BAC)s with 5 g/L of PAC and at different SRT was carried out. The MBR (BAC)s were operated at 10d, 30d and long (pseudo infinity) SRT, namely MBR (BAC_{10d}), MBR (BAC_{30d}) and MBR (BAC_{long SRT}) respectively. Daily top-up of lost PAC in the wasted sludge was provided to maintain a constant PAC concentration in the MBR (BAC_{10d}) (10.0 wt%) and MBR (BAC_{30d}) (3.35 wt%). No sludge was wasted from the MBR (BAC_{long SRT}) and this meant that there was no requirement for PAC top-up. After cultivating the three batch MBR (BAC)s for 59 days, new membrane modules with an area of 0.057 m² were submerged into each MBR and run at a fixed flux of 23.0 L/m²/hr. The results are as shown in Figure 7.13. This clearly indicates that the MBR (BAC_{10d}) performed the best and the worst performer was the MBR (BAC_{long SRT}). Lowering the fluxes from 23.0 L/m²/hr to 15.0 L/m²/hr for MBR (BAC_{10d}) and MBR (BAC_{30d}) and to 10.0 L/m²/hr for MBR (BAC_{long SRT}) did slow the fouling rate (figure 7.14). However, the trend of the filtration performance was the same as the previous results.

Figure 7.13: Performance comparison of MBR (BAC)s at different SRTs (flux: 23.0 L/m²/hr)

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On day 64 (Figure 7.14) an upset caused a sudden increase in the TMP in both the MBR (BAC_{10d}) and MBR (BAC_{30d}). This was due to dysfunction of the aerator and without aeration, the prevention of cake sludge accumulation on the membrane surface failed. This led to serious membrane fouling and the classic TMP ‘jump’. The TMP jumped from about 15 to about 23 kPa and 18 to about 34 for MBR (BAC_{10d}) and MBR (BAC_{30d}) respectively. The more serious ‘jump’ for the MBR (BAC_{30d}) was probably due to the higher MLSS and fine foulants in that MBR. After the problem was fixed, the TMP of both MBR (BAC)s dropped back to a lower level. At day 64.2, the membranes from both the MBR (BAC)s were taken out and flushed with tap water to remove residual sludge and reversible foulants attached on the membrane surfaces. This further reduce the TMP level of both the MBR (BAC)s. The average rate of TMP rise was 3.8 kPa/day for the MBR (BAC_{30d}) and only 2.0 kPa/day for the MBR (BAC_{10d}) throughout the operating period of about 10 days.

Figure 7.14: Performance comparison of MBR(BAC)s at different SRTs (flux: 10.0 and 15.0 L/m²/hr)

Table 7.3 summarizes the dTMP/dt data for the series of experiments, including MBR (AS) and MBR (BAC). Recalling that these data represent the TMP trends of

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relatively long experiments (in some cases) they are not precise measures. However, the trends are clear and show the benefit of BAC over AS in the MBRs tested. In particular at a flux of 15 L/m²/hr the BAC reduces the dTMP/dt of the SRT 10 MBR by almost a factor of 10; the effect at SRT 30 is less significant but is still a fact of 2 improvement. The effect at SRT (long) was not measured at the flux of 15, but from the data at a flux of 10 and 23 L/m²/hr, it can be inferred that BAC did not provide a benefit and was detrimental. This confirms the general observation made in Chapter 4 that PAC replenishment is required.

Table 7.3: Relationship between dTMP/dt and fluxes for MBR (BAC)s and MBR (AS)s at different SRTs

MBR (AS)	10 L/m ² /hr	15 L/m ² /hr	23 L/m ² /hr
SRT: 10 days	9.0 (kPa/day)	22.0 (kPa/day)	150.0 (kPa/day)
SRT: 30 days	3.6 (kPa/day)	8.0 (kPa/day)	3538.0 (kPa/day)
MBR (BAC)	10 L/m ² /hr	15 L/m ² /hr	23 L/m ² /hr
SRT: 10 days	N.A	2.0 (kPa/day)	54.0 (kPa/day)
SRT: 30 days	N.A	3.7 (kPa/day)	258.0 (kPa/day)
SRT: Long	43.0 (kPa/day)	N.A	1059.0 (kPa/day)

N.A: No applicable (no test was carried out at this flux)

Overall, the best membrane filtration performance was by the MBR (BAC_{10d}) compared with the other two MBR (BAC)s. The possible factors causing this trend are discussed below.

- i) The MBR (BAC_{10d}) with SRT 10 days (fresh PAC replaced at 10wt%/day had a higher percentage of fresh PAC than that of MBR (BAC_{30d}) (3.35 wt%/day) and MBR (BAC_{long SRT}) (no replacement of aged BAC). The higher concentration of fresh PAC in the reactor would provide better simultaneous adsorption, decomposition, biodegradation effects for the reduction of fouling components such as EPS, fine colloids and planktonic cells in the supernatant of the mixed liquor. Evidence in favour of this is provided in Table 7.4 where the TOC for

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MBR (BAC_{10d}) was significantly lower than for MBR (BAC_{30d}) and especially the MBR (BAC_{long SRT}). With lower TOC concentration in the supernatant, MBR (BAC_{10d}) also showed a much slower fouling rate (dTMP/dt value).

Table 7.4: Concentrations of the MLSS and TOC and AS and BAC floc sizes (D₅₀) in MBRs (between days 60-74)

MBR	MLSS (g/L)	Particle size (µm)		TOC (mg/L)	
		D ₅₀ (Volume)	D ₅₀ (Number)	Supernatant	Permeate
10d	10.33 ± 0.4	15.32 ± 3.9	1.95 ± 0.09	9.26 ± 1.24	4.32 ± 1.65
30d	15.53 ± 0.6	18.01 ± 1.36	1.06 ± 0.16	18.15 ± 1.03	6.15 ± 2.54
Long	18.78 ± 0.6	34.70 ± 1.13	3.93 ± 0.15	278.18 ± 27.50	20.47 ± 3.50

- ii) A second factor could be that the ‘scouring’ of the BAC in the MBR (BAC_{10d}) was more effective. The fresh PAC which was not fully incorporated or attached with biomass would have a better ‘scouring’ impact on the membrane surface through simultaneous ‘collision’ and adsorption effects as the bubbled suspension is carried past the membranes. This explanation assumes that the biomass around PAC when it forms BAC may act as a cushion which reduces the impact of the ‘collision’ effect. MBR (BAC_{10d}) had a greater concentration of fresh PAC with more adsorptive effect and with less of its inventory incorporated into biofloc with its surface covered by the attached growth biomass compared to the MBR (BAC_{30d}).

Confocal images of sludge samples from the MBR (BAC_{10d}) and MBR (BAC_{30d}) are shown in Figure 7.15 and support the hypothesis. Figure 7.15 shows that the BAC from the MBR (BAC_{10d}) was less covered by

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the biomass than that of the MBR (BAC_{30d}). This means that the particle size (D₅₀) (volume) (only the bigger BAC was taken into the consideration in creating ‘collision’ effect) of the MBR (BAC_{10d}) would be smaller than that of MBR (BAC_{30d}) since it would be less likely to be incorporated into biofloc. Table 7.4 supports this explanation, showing that the D₅₀ for MBR (BAC_{10d}) was $15.32 \pm 3.9 \mu\text{m}$ as compared to the $18.01 \pm 1.36 \mu\text{m}$ for MBR (BAC_{30d}).

Figure 7.15: Confocal images of attached growth bacteria on PAC surface at SRT 10 and 30 days

It is possible to point out a critical SRT based on the observation of the BAC with the help of Confocal Laser Scanning Microscope. The BAC floc cultivated at a

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critical SRT (pseudo infinity SRT) observed under the Confocal Laser Scanning Microscope should show the mixture colours of green, yellow and red which indicates relatively larger portion of the biomass attached on the BAC are dead cells. For the BAC cultivated at lower SRT (≤ 30 days), which the replenishment rate of the PAC is also high, more green colour (lively bacteria) in the biofilm attached on the PAC could be observed.

- iii) The higher MLSS in the MBR (30d) would have given a higher viscosity that would dampen the effect created by the aeration that helps in moving the hollow fiber membrane to minimise the accumulation of the sludge or fine foulants on the membrane surface (Wicaksana *et al.*, (2006). The lower MLSS concentration in the MBR (BAC_{10d}) (10.33 ± 0.4 g/L) compared to MBR (BAC_{30d}) (15.53 ± 0.6 g/L) and MBR (BAC_{long}) (18.78 ± 0.6 g/L) is shown in Table 7.4.

Short term flux-stepping tests (for protocol see 3.2.5) were also carried out to compare the MBR (BAC)s with different SRT. New membranes were submerged into each of the three MBR (BAC)s to carry out the ‘sustainable’ flux tests which included flux stepping and TMP measurement as an indicator of the membrane fouling tendency. The data are shown in Figure 7.16.

Figure 7.16: Sustainable flux tests for MLSS from MBR (BAC)s with different SRTs

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It was found that the MBR (BAC_{long}) without renewal of aged BAC, had the lowest ‘sustainable’ flux (about 6 L/m²hr), MBR (BAC_{30d}) had a higher ‘sustainable’ fluxes at about 15.0 L/m²hr and MBR (BAC_{10d}) had the best ‘sustainable’ flux at about 21.0 L/m²hr. The better short-term filtration performance of the MBR (BAC_{10d}) compared to the others is also shown in Figure 7.17. The unstirred short term dead-end filtration tests at fixed pressure shows that the flux decline profile for MBR (BAC_{10d}) was the best. This may also be because the SCR (Figure 7.17 (inserts)) of the MLSS and the fine foulants (i.e. TOC) from the MBR (BAC_{10d}) was the lowest.

Figure 7.17: Flux decline profile for fixed pressure and unstirred dead-end filtration for MLSS with different SRTs

7.4 Conclusions

The comparative filtration performance of the MBRs with (i) different PAC sizes, (ii) different SRTs with and without PAC can be summarized as below.

- a) The MBR hybrid with a relatively fine PAC, which is equipped with a high adsorption capacity would only be beneficial if operated at a flux where the fine PAC tended not to attach on the membrane surface. When

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the flux is too high, the attachment of the relatively fine PAC on the membrane surface can increase the fouling rate.

- b) For experiments without BAC the, MBR (30d) was found to have a higher MLSS concentration and TOC and polysaccharide contents in the supernatant as compared to MBR (10d). However, it still performed better than the MBR (10d) at modest fluxes of 10.0 and 15.0 L/m²/hr. The probable reason for this was that the AS_{30d} had a bigger floc size than the AS_{10d}. The bigger floc size may help to detach the fine foulants (TOC and polysaccharide) attached on the membrane surface if the floc can scour, rather than deposit. However, the higher MLSS concentration and TOC and polysaccharide contents in the MBR (30d) were also a limitation when the membrane were operated at relatively high flux (23.0 L/m²/hr). The higher flux was probably above the critical flux of the dominant foulants, which were in greater concentration in the MBR (30 days). This would cause a greater fouling for MBR (30) compared with MBR (10). This observation raises the issue that choice of an optimal SRT, based on membrane fouling, depends on the desired flux (as well as module design and hydrodynamics).
- c) MBR (BAC_{10d}) had the best filtration performance compared to MBR (BAC_{30d}) and MBR (BAC_{long}) at all fluxes tested. One of the key factors would be that the fresh PAC concentration in the MBR (BAC_{10d}) was the highest due to the higher frequency of replacement of aged BAC (10.0 wt%/day) compared to MBR (BAC_{30d}) (3.35 wt%/day) and MBR (BAC_{long SRT}) (no replacement of aged BAC). More fresh PAC in the reactor would have a better function in reducing the fine foulants (i.e. EPS and TOC) through simultaneous adsorption, decomposition and biodegradation effects. A second factor may be that the 'scouring' effect of the BAC in the MBR (BAC_{10d}) was more significant. Fresh PAC which is yet to be fully surrounded by the biomass would have better simultaneous 'collision' and adsorption effects as the bubbled

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suspension are carried past the membranes. The lower MLSS concentration in the MBR (BAC_{10d}) compared to MBR (BAC_{30d}) and MBR (BAC_{long SRT}) would also be another advantage. Furthermore, the higher MLSS for MBR (BAC_{30d}) and MBR (BAC_{long SRT}) may have higher viscosity that may hamper the movement of the hollow fiber membranes and restrict the access of air bubbles to the membrane surface and this would worsen the fouling rate.

Two types of MBRs are in the current industrial. They are submerged and sidestream MBRs. The SRT for the submerged MBR and sidestream MBRs is normally controlled at between 20-30 days. In this study, the BAC at SRT 10-30 days was found better than AS in terms of membrane fouling control. This indicates that BAC at SRT 10-30 days was sufficient to have a good membrane fouling control compared to AS at similar SRT. To reduce the SRT from 30 days to 10 day would improve the performance of BAC in fouling control but requires higher replenishment BAC rate. This may increase the MBR operation cost. However, with better membrane fouling control, the rate of the chemical used in the membrane cleaning would be reduced and the permeate production would not be disrupted.

PAC in powder form normally tends to attach to the membrane surface at relatively higher flux ($>30 \text{ L/m}^2/\text{hr}$) and poor airflow distribution, however, most of the commercialised submerged MBR systems are operating at flux lower than $30 \text{ L/m}^2/\text{hr}$ (Zenon MBR: $25 \text{ L/m}^2/\text{hr}$; Mitsubishi MBR: $25 \text{ L/m}^2/\text{hr}$; Kubota MBR: $20 \text{ L/m}^2/\text{hr}$) and equipped with good aerator which can produce sufficient airflow rate to create good shear force on the membrane surface to control the attachment of the particles. Sidestream MBR may operate at relatively higher flux ($>30 \text{ L/m}^2/\text{hr}$), but the sidestream MBR also has relatively high crossflow rate ($>140\text{-}225 \text{ m}^3/\text{hr}$) and may run at two phase flow mode. High crossflow and two-phase flow may help to prevent the attachment of the BAC to the membrane surface.

The MBR (BAC)'s SRT controlled in the range between 10-30 days, would have the MLSS at the concentration range of 8-15 g/L and relatively lower TOC

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production in the supernatant (after achieving steady state) compared to the MLSS cultivated at long SRT (e.g infinity). Lower SRT (e.g 10 days) would have higher replenishment rate of the aged BAC. To optimise the beneficial effect of the PAC, (i) the hydrodynamics of the MBR system should be well designed to prevent the attachment of the PAC to the membrane surface and (ii) the MBR system should be operated below critical flux of the PAC.

The MBR (BAC) cultivated at the infinity SRT is likely to have a relatively higher MLSS concentration (>20 g/L) and most of the BAC could have lost its adsorptive capacity. The MBR (BAC) at infinity SRT also has a tendency to produce more sticky EPSs and TOC concentrations which tend to foul the membranes. This means that well control of SRT and hydrodynamic are the prerequisites to have a good performance MBR (BAC).

Overall, it is evident that the MBR (BAC) has better fouling control than the MBR (AS). The benefit can be maximized by use of fine, rather than coarse PAC and operation at relatively short SRTs (for MBRs). Careful selection of the imposed flux is required to maximise the fouling control benefit.

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Synopsis:

In chapters 4, 5, 6 and 7, the discussions were mainly focused on the MBR (BAC) compared to the MBR (AS). The better performance of the MBR (BAC) under conditions of regular replenishment PAC compared with a conventional MBR has been confirmed. This chapter discusses two alternative concepts of the MBR (AS) and MBR (BAC). One alternative involved a two-compartment MBR filtering supernatant instead of MLSS. The other concept was an MBR incorporating polymer beads to control fouling.

The purpose of designing of the two-compartment MBR was to locate the submerged membranes in a reactor separate from the aeration bioreactor to minimise the MLSS loading on the membranes. However, the results of the investigation revealed that there is no benefit in filtering supernatant compared to MLSS.

The MBR (polymer bead) was found could be a potential alternative to the conventional MBR (AS) provided the operating flux is carefully controlled.

8.1 Comparison between MBR (MLSS) and MBR (supernatant)

In a membrane bioreactor, the fouling components of the MLSS include the biomass floc, EPS, fine colloids and planktonic cells where the relative contributions of these species depend on the conditions. In this subproject, a two-compartment MBR was set up based on the concept of avoiding direct contact between the membrane and the high MLSS liquor. The first compartment acts as the bioreactor while the second compartment is the filtration chamber, with the bulk of the MLSS being retained in the first compartment. This can be achieved by manipulating the return flow to the first chamber from the second compartment. With the aerator positioned just below the membrane module in the upper part of

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the filtration chamber, the MLSS does not rise up the filtration vessel. The two-compartment MBR is illustrated in Figure 8.1. After operation for a period of 120 days (4 months), it was observed that the MLSS in the first compartment (bioreactor) and the submerged chamber were approximately 4.0 g/L and 0.0 g/L respectively. This indicated that the two-compartment MBR design was capable of retaining the high MLSS concentration in the bioreactor without the sedimentation stage and allowing only the supernatant to flow over to the adjacent submerged membrane chamber.

Figure 8.1: Two-compartment set-up used to separate the supernatant from the MLSS flocs

The fouling tendency of the mixed liquors in the first (bioreactor) and second (supernatant) compartments was compared using a submerged Polymem hollow fibre module in each compartment, operated at a constant flux of $18.0 \text{ L/m}^2\cdot\text{h}$. The aeration flow rate in both chambers was similar. The fouling tendency was characterized by measuring the TMP versus time profiles.

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Unexpectedly, a higher TMP rise was observed for the supernatant filtration than for the bioreactor liquor (4.0 g/L of MLSS) as shown in Figure 8.2. The higher TMP rise occurred in the initial stages with the supernatant and may be due to EPS and fine colloids, which could block the unprotected membrane pores and cause the filtration resistance to increase. The lower filtration resistance for the bioreactor liquor seemed to indicate that the biomass floc could help to adsorb the supernatant foulants or alternatively protect the membrane with a ‘precoat’ of floc. The flocs may also collide with the membrane and scour the fine foulants accumulated on the membrane surface back to the bulk liquid.

Figure 8.2: Performance comparison of AS and supernatant

A further study was carried out by adding PAC to the MBR chamber with the purpose of producing a better quality of supernatant that would cause less membrane fouling. The results are discussed below.

8.2 Comparison between MBR (BAC) and MBR (supernatant)

Initial design of the two-compartment MBR (Figure 8.1) was modified to the new design as shown in Figure 8.3. The new design had additional two smaller 3.5L

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reactors. The purpose was to enhance the effectiveness of the two-compartment MBR in separating the supernatant from the mixed liquor.

Figure 8.3: Two-compartment set-up used to separate the supernatant from the BAC flocs

In the previous design, the aerator in the supernatant chamber was mounted half way up the 20L reactor to prevent the rise of the mixed liquor which was bypassed at the bottom of the reactor. The design had constrained the aeration rate because a higher rate may have induced the upflow of the mixed liquor. Locating the aerator in the 3.5L vessel connected to the 20L compartment filled with supernatant (Figure 8.3) was aimed to solve this problem. The design could also allow PAC to be added together with the supernatant to enhance the filterability. This was not possible with the previous design with the aerator in the middle of the reactor because settling of the PAC would occur. To make a fair comparison of the filtration performance of the supernatant and the mixed liquor, a 3.5L vessel was also connected to the side of the 20L reactor filled with the mixed liquor. The two 3.5L reactors were linked to

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the 20L reactors beside them by means of a pump. This was to ensure the contents of the 3.5L reactors were the same as the 20L reactor located adjacent to them.

In addition, a returned accumulated sludge flow line was added at the bottom of the 20L reactor (supernatant). One of the purposes of this was to minimise the accumulation of the sludge at bottom of the vessel as shown in Figure 8.3. The second purpose was to return the separated supernatant back to the mixed liquor chamber. This aimed to minimise the retention time of the planktonic cells in the separated supernatant in the critical condition of the supernatant chamber which had insufficient substrate or oxygen. The longer the retention time the most likely the planktonic cells would go through a lysis process that could contribute more fine foulants in the separated supernatant in the supernatant chamber. Apart from the additional two 3.5L vessels and the return accumulated sludge flow line, the concept of separating the supernatant from the mixed liquor remained the same as the previous design.

For the new two-compartment MBR design, PAC was added into the mixed liquor to form BAC and run at SRT 10 days. At SRT 10 days, the replacement of the aged BAC was done daily at 10.0 %wt. After operation for 53 days, the new design was observed to have the capability of retaining 8.14 ± 0.82 g/L of BAC concentration (Table 8.1) without the sedimentation stage and allowing only the supernatant to flow over to the adjacent 20L supernatant chamber. However, it was found that the TOC and polysaccharide concentrations in the supernatant of the mixed liquor were different from the separated supernatant in the supernatant chamber as shown in Table 8.1. The TOC and the polysaccharide concentrations were 2.50 ± 1.20 and 2.10 ± 0.87 mg/L for the supernatant of the mixed liquor and 12.28 ± 1.38 and 9.82 ± 1.29 mg/L for the separated supernatant. The concentrations of the TOC and the polysaccharide in the separated supernatant in the supernatant chamber were about five times higher than that of the supernatant of the mixed liquor.

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Table 8.1: Concentrations of the MLSS and TOC and MLSS floc sizes (D_{50}) in MBR (BAC) and MBR (supernatant) (between days 53-58)

MBR	MLSS (g/L)	Particle size (μm)		Polysaccharide in the supernatant (mg/L)	TOC (mg/L)	
		D_{50} (Volume)	D_{50} (Number)		S/N	Permeate
BAC	$8.14 \pm$	$40.07 \pm$	$3.68 \pm$	2.10 ± 0.87	$2.50 \pm$	$1.61 \pm$
	0.82	2.45	0.92		1.20	0.41
S/N	$0.011 \pm$ 0.001	ND	ND	9.82 ± 1.29	12.28 ± 1.38	$3.14 \pm$ 0.61

S/N: supernatant; ND: not detectable

There are two possible reasons for this effect. Firstly, the returned accumulated sludge line was operated at a rate of 2.4 L/hr; which means that the retention time of the separated supernatant in the supernatant chamber (23.5 L) was 9.8 hours. This rather long period of retention time may cause the lysis of planktonic cells in the separated supernatant due to insufficient substrate or oxygen in the supernatant chamber. Reducing the retention time of the separated supernatant in the supernatant chamber may help to reduce the gap between the TOC and polysaccharide concentration in the supernatant of mixed liquor and separated supernatant. However, it is expected that, the TOC and polysaccharide concentrations in the separated supernatant could always be higher than that in the supernatant of the mixed liquor due to insufficient food and oxygen.

The second reason relates to the differences in retention of TOC in the two vessels as well as the possible differences in molecular weight distribution of TOC in the two systems. Unfortunately, the analytical facility for MWt distributions was not available at the time of the experiment. However, based on the study reported in chapter 6 (section 6.5.3) a rather broad distribution of molecular weight would be expected. The large species, including polysaccharides would tend to foul the membranes. In the MBR (BAC), these components would be degraded and kept in balance by the biomass. However in the supernatant vessel these components would

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either pass through the membrane or be retained and accumulated. The TOC data in Table 8.1 shows that the retention of TOC by the membranes was about 35.6 % for the MBR (BAC) and 74.4 % for the supernatant chambers. This points to a significant accumulation mechanism in the supernatant chamber. If this is the case, the concept of separating the membranes from the MLSS in the way described has an intrinsic problem. There may be ways to overcome this but it would require substantial recycle from the supernatant chamber. For example, when the supernatant circulation rate was changed from 2.4 to 6.0 L/hr, the TOC in the supernatant chamber dropped from 12.28 ± 1.38 mg/L to approximately 8.0 mg/L.

At day 53, the filtration performances of the mixed liquors of the BAC in the first chamber and the separated supernatant in the adjacent supernatant chamber were compared using a Blue Star hollow fibre module (see section 3.2.1) submerged in each 3.5L reactor, operated at a constant flux of $20.0 \text{ L/m}^2\cdot\text{h}$. A higher fouling rate was observed for the separated supernatant than the BAC mixed liquor as shown in Figure 8.4. The higher fouling rate for the filtration of the separated supernatant was most probably due to the higher concentration of foulants (i.e TOC and polysaccharides) in the separated supernatant which could plug and block the membrane pores. The lower filtration resistance of the BAC mixed liquor may also be due to the presence of the BAC flocs equipped with simultaneous adsorption, decomposition and biodegradation properties that may help to break down the organic foulants for the subsequent adsorption and biodegradation process. This would reduce the high molecular weight organics in the supernatant of mixed liquor that tend to foul the membranes. In addition, the BAC may help to detach the fine foulants attached on the membrane surface through the 'scouring' effect discussed previously.

Figure 8.4: Performance filtration comparison of BAC and supernatant at flux 20.0 L/m²/hr

One of the limitations of MBRs is not being able to be operated at relatively high flux. The typical flux of the conventional MBR is about 10-20 L/m²/hr. This is because the high MLSS concentrations (8-12 g/L) of the MBR may tend to accumulate on the membrane surface at higher flux. The accumulation of the MLSS may cause the TMP 'jump' or serious fouling. Therefore, filtration of the supernatant with very low SS could be seen as a strategy to avoid the sludge accumulation problem. To examine this, two new Blue Star hollow fibre modules were submerged in each 3.5L chamber and operated at a flux of 40.0 instead of 20 L/m²/hr. However, the filtration performance of the separated supernatant was still worse than that of the mixed liquor as shown in Figure 8.5. The rate of the TMP increase for the separated supernatant and BAC mixed liquor was 30.43 and 7.61 kPa/hr respectively as shown in Table 8.2. This indicates that the filtration of the separated supernatant is not beneficial when compared to BAC mixed liquor at either low (20.00 L/m²/hr) or high (40.00 L/m²/hr) fluxes. From the findings in the previous chapters, it was considered that the addition of PAC into the separated

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supernatant may be beneficial. The PAC may help to reduce the fouling rate through ‘collision’ and adsorption effects. To test this, 5 g/L of PAC was added into the separated supernatant in the 3.5L supernatant reactor. A new membrane was submerged in the 3.5L supernatant reactor which had added PAC.

PAC did help to reduce the filtration fouling rate of the separated supernatant as shown in Figure 8.5. It decreased the fouling rate from 30.43 to 17.75 kPa/hr as shown in Table 8.2. This may be due to the reduction of the fine foulants such as TOC in the separated supernatant through adsorption by the PAC. The TOC concentrations dropped from 14.34 to 8.25 mg/L (Figure 8.5 (inserts)). However the filtration of the separated supernatant with added PAC was still not better than that of BAC mixed liquor. One of the reasons may that the adsorption capacity of the amount of PAC added was not high enough to reduce the fine foulants (e.g. TOC) to a concentration as low as in the supernatant of the BAC mixed liquor.

Figure 8.5: Performance filtration comparison of BAC and supernatant at flux 40.0 L/m²/hr

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Table 8.2: Fouling rate of the BAC, supernatant and supernatant added with PAC at flux 40.0 L/m²/hr

MBR	Filtration period (day)	Rate of TMP increase dTMP/dt (kPa/hr)
BAC	58.00-58.18 (4.42 hours)	7.61
Separated supernatant	58.00-58.05 (1.20 hours)	30.43
Separated supernatant + PAC (1)	58.06-58.14 (1.92 hours)	17.75
Separated supernatant + PAC (2)	58.14-58.18 (1.06 hours)	29.49

After the TMP of the membrane used in the 3.5L reactor with separated supernatant and PAC had reached 50 kPa, the filtration was stopped. The membrane was replaced again with a new membrane to continue the filtration process. The second filtration performance of the separated supernatant with PAC was worse than that of the first filtration but quite similar to separated supernatant without PAC filtration. This demonstrates that the PAC had lost its adsorption effect and did not contribute to the fouling control. This view was supported by the TOC concentration in the separated supernatant (12.79 mg/L) which slowly increased back to the level before the PAC was added (14.34 mg/L) as shown in Figure 8.5 (inserts). The similar filtration TMP increase rate for the second filtration of the separated supernatant with PAC and before adding PAC indicates that the ‘collision’ effect of the PAC was not obvious. This may be the fine foulants were tightly attached on the membrane surface by the higher suction force due to the higher flux (40.0 L/m²/hr) and not easily detached by the collision of the PAC. The collision effect may be more significant if the flux was set at 10.0-20.0 instead of 40.0 L/m²/hr.

To prevent the loss of the PAC from 3.5L reactor into the 20L supernatant chamber, the circulation pump linking the contents of the 3.5L reactor with the 20L supernatant was stopped. This means that the fine foulants from the 20L supernatant chamber continued flowing in the 3.5L reactor of separated supernatant with PAC. This could have accelerated the saturation of the PAC. This situation was similar to the operating condition of the MBR (BAC) when synthetic wastewater (TOC: 155 ±

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5.0 mg/L) was fed continuously into the system. Based on the rapid rise of TMP for the supernatant filtration, it appears that most of the fine foulants in the separated supernatant were in the high molecular weight range or were fine colloids. The species would tend to block or plug the membrane pores and deposit on the membrane surface, especially at higher flux. The rapid saturation of the PAC in only two and half an hours operating time may due to (i) PAC with limited micropore volume (about 25% of the total volume based on the BET analysis) that could be easily filled up by the high and low molecular weight fine foulants and (ii) the deposition of the fine foulants on the PAC surface could also rapidly diminish the adsorption effect. The better performance of BAC in TOC reduction and membrane fouling control compared to AS or PAC alone, may be because the fine foulant with high molecular weight was adsorbed by the BAC and could be decomposed by the more acclimatised and homogenous biomass attached on the PAC surface. The resultant lower molecular weight organics would be more easily further biodegraded and adsorbed by the PAC and have less tendency to foul membrane. This sequence would enhance the membrane fouling control. The decomposition and biodegradation process could also have helped to bioregenerate the BAC and open up its pores which had been blocked by the organic foulants. Indirectly, these mechanisms may extend the life-span of BAC as a simultaneous adsorption, decomposition and biodegradation agent. This implies that, the biomass flocs and PAC worked synergistically to improve the filtration performance. The absence of one of them would make the fouling control ineffective. Filtration of the separated supernatant with and without PAC further confirmed the conclusion in chapter 6 that the major mechanisms involved in fouling control by the PAC are simultaneous adsorption, decomposition and biodegradation rather than collision or adsorption alone.

In addition to the long term filtration performance comparison, a series of short term filtration tests were carried out consisting of SCR, irreversible fouling assessment, flux decline profile (operated at fixed pressure in the unstirred cell) and flux-stepping to obtain the 'sustainable' flux in the 3.5L reactor with air crossflow. All the filtration results favored the BAC mixed liquor. As expected, irreversible

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fouling by the separated supernatant was higher than that of BAC mixed liquor as shown in Figure 8.6 due to the higher concentration of fine foulants such as TOC and polysaccharide that could block and plug the pores of the membrane.

Figure 8.6: Specific cake resistance and resistance increment (irreversible fouling) for BAC and supernatant at fixed pressure short-term unstirred test

However, it was also found that the relatively thin layer formed by the fine particles in the separated supernatant on the membrane surface had a much higher value of SCR compared to the BAC as shown in Figure 8.6. Both the higher irreversible fouling and the SCR could be factors that caused the worse performance of the separated supernatant filtration compared to the BAC mixed liquor. This conclusion was supported by the lower flux decline profile (Figure 8.7) of the separated supernatant compared to the BAC mixed liquor.

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Figure 8.7: Flux decline profile for fixed pressure and unstirred dead-end filtration for BAC and supernatant

This shows that, reducing the fine particles in the supernatant is very important but to prevent them from attaching on the membrane surface is more crucial in minimising the membrane filtration fouling rate. This was further confirmed by the ‘sustainable’ flux test done at the air crossflow condition as shown in Figure 8.8. The MLSS concentration of the BAC mixed liquor was much higher than that of the separated supernatant, however it had a higher ‘sustainable’ flux at about 40 L/m²/hr compared to the separated supernatant with only about 28.5 L/m²/hr.

Figure 8.8: Sustainable flux tests for BAC and supernatant

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In summary, the better filtration performance of the BAC mixed liquor compared to the separated supernatant could be due to the factors discussed below:

- i) The higher fine foulants (i.e TOC and polysaccharides) in the separated supernatant compared to the BAC mixed liquor. This may be due to the accumulation of species retained by the membrane and not removed by biological activity.
- ii) Apart from biodegrading and maintaining the fine foulants in low and constant concentrations, biomass may help to bioregenerate the aged PAC. This means that, the functional period of the PAC in adsorbing the fine foulants may be prolonged and more sustainable if used together with the biomass.
- iii) BAC flocs which had simultaneous adsorption, decomposition and biodegradation effects may also act as 'scouring' and adsorbent agents which help to detach and disperse the fine foulants. These include relatively fine size PAC attached to the membrane surface transported back to the mixed liquor and then able to 'pick' up the dispersed fine organic particles in the supernatant to go through the biodegradation process. This would not be possible by using PAC or biomass alone.

In these experiments, both the biomass and the BAC flocs seemed able to play the 'scouring', biodegradation, decomposition and adsorption roles well, but the BAC performed better. The PAC and biomass appear to be a good combination that provides mutual benefit to minimise the membrane fouling rate. For the conditions studied it would appear that filtration with MLSS (AS or BAC) is more beneficial than supernatant. A similar observation was reported by Chang and Kim (2005).

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8.3 Comparison between MBR (AS) and MBR (polymer bead)

From the above discussions, we can conclude that BAC can improve the performance of the MBR through adsorption, decomposition, biodegradation and scouring. However, because the PAC is in powder form, the efficiency of it in creating a scouring effect is limited. From the experimental scouring results obtained using hollow glass beads as discussed in chapter 6, the scouring effect may only be obvious when operated at low flux ($< 10 \text{ L/m}^2/\text{hr}$) to prevent the attachment of scouring agent to the surface of membrane. Otherwise, that attachment would aggravate the fouling tendency. PAC that had been saturated or embedded by the biomass would also lose its simultaneous 'collision' and adsorption effects. The major mechanism of the PAC in helping fouling control is simultaneous adsorption, decomposition, biodegradation and 'scouring' effects. However, replacement of aged BAC should be done constantly in order to maintain that good functional simultaneous mechanisms. This is a limitation of the MBR (BAC). Putting aside the simultaneous mechanism effect and focusing on enhancement of 'collision' impact agents, the replacement of PAC with larger sized beads with a density approximately similar to water (to avoid settlement) to test its 'collision' effect in membrane fouling control would be of interest. In addition, the bigger beads in the wastage sludge could be easily separated and reused which would be more cost effective. To test if the concept discussed above is practical, a comparison between larger beads (polymer beads with the D_{50} of $600 \mu\text{m}$) and PAC in fouling control was carried out. The results are shown in Figure 8.9.

After use in the 20L bioreactor for a period of time (1.5 days), the fouled membrane was shifted from the 20L bioreactor to the 3.5 L reactor filled with milli-Q water. This caused an initial drop of TMP from about 25.5 to 22.2 kPa (Figure 8.9). After filtering the milli-Q water for about half a day, the TMP increased back from 22.2 to 24.7 kPa and became stable. The TMP trend may have been due to the detached fine foulants in the milli-Q water reattaching to the membrane surface. At day 2, 5 g/L of polymer beads were added into the 3.5 L reactor, the TMP was observed to decrease gradually from 24.7 to 23.7 kPa over the 24 hours of filtration process.

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However, the TMP started to increase again at day 3 and the TMP was back to 24.7 at day 3.2. This may have been due to the fine particles detached from the membrane surface becoming smaller. The collision or ‘grinding’ between the fine particles and the polymer beads may cause this to happen. The finer particles would foul the membrane more seriously. However, the phenomena (fine particle ‘neutralising’ the scouring effect) may not necessary happen in the MBR. The fine particles would attach to the biomass flocs or the biomass flocs would help to adsorb the detached finer particle. This may help to maintain the good ‘collision’ effect created by polymer beads (the comparison of the MBR and MBR (polymer bead) is discussed below).

Figure 8.9: Performance comparison of collision effect in fouling control between polymer bead, glass bead and PAC

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At day 3.2, glass beads (D_{50} : 100 μm) were put into the 3.5 L reactor. Due to their high density, most of the glass beads settled to the bottom of the reactor but some remained in suspension due to bubbling. The glass beads helped to slightly reduce the TMP from 24.7 to 24.1 kPa. At day 4.2, 5 g/L of PAC was added to the reactor. An immediate rise of TMP was observed possibly due to the small fines in the PAC which tend to attach to the membrane surface and cause TMP rise. This phenomenon is also not necessarily observed in the MBR (BAC) as the fine PAC would attach to the biomass flocs. The TMP 'jump' was short-lived as the PAC is equipped with high adsorption capacity and this took effect to reduce the TMP back to the 23.7 kPa and became stable. At this point in time (day 5.2), some of the sample of the PAC in the reactor was taken out for observation under an optical microscopic. The image is shown in Figure 8.9 (picture b). Comparison between the used PAC and fresh PAC (Figure 8.9; picture a) indicates that the PAC was embedded within the sticky fine pollutants. This would cause the PAC to lose its 'scouring' effect. Addition of new PAC into the reactor would be necessary in order to further improve the TMP reduction. This was confirmed when new PAC with concentration of 5 g/L was added and further reduced the TMP from 23.7 to 22 kPa. From these comparison tests, it was concluded that the 5 g/L of polymer beads had a similar 'scouring' effect as 5 g/L of PAC in TMP reduction.

To verify the ability of polymer beads in improving membrane fouling control, the two 20 L MBRs were set-up at SRT 10 days and 5 g/L of polymer beads with a homogenous size of 600 μm was added to one of them. Constant reused of lost polymer beads (10.0 wt%) in the wasted sludge was done daily to keep the polymer beads concentration constant in the MBR (polymer bead). However, due to the relatively higher density of the polymer beads (1.18-1.27 g/cm^3), some of them settled at the bottom area of the reactor and could not be accessed by the aeration. However, there was still about 2 g/L of polymer beads well mixed in the reactor. In order to make a fair comparison for these tests, both MBRs were operated in parallel at a fixed permeate flowrate of 40 mL/min (flux = 41.4 and 20.7 $\text{L}/\text{m}^2\text{hr}$, HRT = 8.3 hrs). At day 10, new membrane modules with an area of 0.057 m^2 were submerged into each MBR and run at a fixed flux of 41.4 $\text{L}/\text{m}^2\text{hr}$. The results are

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shown in Figure 8.10 which clearly indicates that the MBR (polymer bead) performed worse. The serious TMP ‘jump’ was observed for MBR (polymer bead) after had been operated for a day. Rinsing the membrane from MBR (polymer bead) was done with tap water and was able to reduce the TMP from 72 to 42 kPa. The membranes were used again to continue the filtration process and it was observed that the TMP increased rapidly again from 42 kPa back to 88 kPa. Further rinsing the membranes with tap water did not help to restore the membrane permeability.

Though the MBR (polymer bead) performed badly at a high flux of 41.4 L/m²/hr, its performance was checked again at a lower flux. Two new membrane modules with an area of 0.114 m² were submerged into each MBR to replace the fouled membranes. Both MBRs were then operated at a flux of 20.7 instead of 41.4 L/m²/hr. Contrary to the previous filtration performance, the MBR (polymer bead) did much better than that of MBR (AS). The average rate of TMP rise was 5.45 kPa/day for the MBR (AS) and only 1.27 kPa/day for the MBR (polymer bead) throughout the operating period of about 12 days.

Figure 8.10: Performance comparison of MBR and MBR (polymer bead)

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The filtration performance of the MBR (polymer bead) with added polymer bead is summarised below.

- a) The main characteristic of MBR (polymer bead) could be the ‘collision’ effect of the polymer beads at the membrane surface as the bubbled suspension is carried past the membranes. The useful ‘collision’ effect in membrane fouling control by powdered hollow glass bead has already been discussed and proved in Chapter 6. The purpose of using relatively larger size of polymer beads instead of glass bead is to enhance and multiply the effect of ‘collision’ and make it easier to recover and reuse the scouring agent. The floc size distribution and D_{50} (number) of MLSS (AS + polymer bead) was smaller than AS flocs as shown in Figure 8.11. This suggests that the polymer beads in the MBR could have increased local shear of the biomass floc to disperse them into a smaller size. This can be argued as proof that the polymer may collide with the membrane surface to detach the fine foulants attached. Although it had smaller floc size (number), the MBR (polymer bead) still performed better than that of the MBR (AS) at the relatively lower flux ($20.7 \text{ L/m}^2/\text{hr}$). The results in Figure 8.10 are evidence that polymer beads can enhance the membrane performance.

Figure 8.11: Particle size distribution and D_{50} of AS flocs and AS + polymer beads in number

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- b) However, the MBR (polymer bead) contained floc to smaller D_{50} (number) as compared to the MBR (AS) and at higher flux ($41.4 \text{ L/m}^2/\text{hr}$) these appeared to deposit and induce higher suction pressure. The tightly attached fine particles on the membrane surface would be more difficult to be detached by the 'collision' effect of the polymer beads and this would increase the fouling rate. This phenomenon was also observed in the comparison between the MBR (10d) and MBR (30d), as reported in the section 7.2. The MBR_{30d} had the bigger floc sizes (number and volume), but the polysaccharide and TOC in the supernatant was higher compared to the MBR (10d). As a result, the MBR (30d) performed better only at fluxes of 10.0 and $15.0 \text{ L/m}^2/\text{hr}$ but not at $23.0 \text{ L/m}^2/\text{hr}$. This may be because the operating flux had exceeded the 'sustainable' flux. As a result, higher polysaccharide and TOC in the supernatant were the 'culprit' foulants at flux $23.0 \text{ L/m}^2/\text{hr}$. The 'scouring' effect of the bigger floc in the MBR (30d) would not be sufficient at this flux. For the polymer beads at high flux the greater suction force would cause the beads to accumulate around the membrane module. This could obstruct the movement of the hollow fiber membrane and inhibit the access of air bubbles into the bundle and this would also lead to more serious fouling. Similar to the case discussed in section 7.2, this limitation can be avoided or minimised by optimising the aeration rate and the use of relatively modest fluxes.
- c) The benefits of the polymer beads in the MBR consist of (i) it would create a significant scouring or 'collision' effect, (ii) it can be separated out from the waste sludge and (iii) it is easily be reutilized. However, a suitable density of particle bead is particularly important to make sure that it could be easily floated and mixed well inside the reactor.

These experimental results suggest that polymer bead addition could play a significant role in membrane fouling control improvement. The much lower TMP rise for the MBR (polymer bead) and the reusable nature of the bead imply that it can be an economic and alternative system for the conventional MBR. While this is

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an encouraging finding, the results presented in the Figure 8.10 indicate that the polymer bead would only help to improve the fouling control up to certain flux (modest flux is preferable). The operation of the MBR (polymer bead) at high flux may deteriorate membrane performance and lead to serious fouling.

8.4 Comparison between MBR (BAC) and MBR (Polymer bead)

The comparison between the conventional MBR (AS) and MBR (Polymer bead) at SRT 10 days was discussed in section 8.3. It was found that at the lower flux (21.0 L/m²/hr), the polymer bead could play a beneficial role to limit the rise in the TMP. Due to the advantages of the good ‘collision’ effect and being reusable, the MBR (Polymer bead) might be an alternative to the MBR (BAC). As noted earlier, the benefit of using PAC in the MBR to control fouling can only be seen if the saturated PAC is steadily replenished. In terms of operating cost, this would be less economic compared to the polymer bead. It is of interest to know if the fouling control performance of the MBR (Polymer bead) is comparable to the MBR (BAC) at SRT 10 days (note: MBR (BAC) at SRT 10 days is the best fouling control performer among the MBRs in this study). As a result, two batch bench-scale 3.5L MBRs with the addition of 5 g/L of PAC and polymer bead respectively were set-up and run at SRT 10 days to compare their performance.

The sludge was wasted from MBR (BAC) and MBR (Polymer bead) at 3.35 wt% daily. PAC was topped-up daily into the MBR (BAC) to account for the loss of PAC in the wastage sludge. Whereas the polymer bead (600 μ m) in the wastage sludge was separated out and return to the bioreactor. After operating the two MBRs for 14 days at HRT 3.5 days, the MLSS was 8.36 ± 0.62 and 8.58 ± 0.56 for MBR (Polymer bead) and MBR (BAC) respectively. New membrane modules with an area of 0.057 m² were then submerged into each MBR and operated at a fixed flux of 10.5 L/m²/hr. The result shows that MBR (BAC) performed better than the MBR (Polymer bead) as shown in Figure 8.12. Within the filtration period from day 14 to 22, the TMP increased was 0.15 kPa/day for MBR (BAC) and 0.63 kPa/day for MBR (Polymer bead) respectively. The good performance of the MBR (BAC)

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was mainly due to its simultaneous adsorption, decomposition and biodegradation effects as discussed repeatedly and at length in earlier sections. Though the performance of the MBR (Polymer bead) was not better than the MBR (BAC), however, it had a better performance compared to the MBR (10d) and MBR (30d) which operated under the same conditions as discussed in section 7.2. The TMP increased was 9.13 kPa/day for MBR (10d) and 3.31 kPa/day for MBR (30d) (see section 7.2) compared to 0.63 kPa/day for the MBR (Polymer bead). This indicates that, apart from MBR (BAC), the MBR (Polymer bead) could be another alternative for conventional MBR. The good performance of the MBR (Polymer bead) may be attributed to the scouring effect of polymer beads. The polymer beads which occupied only 10% of the floc (volume) (Table 8.3) had helped to detach the fine foulants attached on the membrane surface back to the mixed liquor.

Figure 8.12: Performance comparison of MBR and MBR (polymer bead) at flux 10.5 L/m²/hr

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Table 8.3: Concentrations of the MLSS and TOC and AS + Polymer bead and BAC floc sizes (D_{50}) and $D_{(0.9)}$ in MBRs (between days 14-22)

MBR	MLSS (g/L)	Particle size (μm)			TOC (mg/L)	
		$D_{(0.9)}$ (Volume)	D_{50} (Volume)	D_{50} (Number)	Supernatant	Permeate
Polymer bead	8.36 ± 0.62	665.2 ± 23.4	24.7 ± 1.4	2.2 ± 0.3	113.3 ± 7.2	21.5 ± 1.6
BAC	8.58 ± 0.56	65.6 ± 2.1	27.0 ± 3.7	2.1 ± 0.3	13.73 ± 2.2	5.6 ± 1.8

At day 22.5, the aeration supplied to the both MBRs failed and this led to a sudden TMP ‘jump’. No aeration means that no air bubbles are present to create the shear forces on the membrane surface. This encourages the attachment of gross sludge and fine foulants on the membrane surface leading to serious reversible and irreversible fouling. This may have caused the TMP ‘jump’ from 7.4 to about 20 kPa for MBR (BAC) and from 10.9 to about 100 kPa for MBR (Polymer bead). The higher TMP ‘jump’ of the MBR (Polymer bead) indicates that the AS + Polymer bead sludge had higher SCR and irreversible fouling tendency compared to BAC. This is supported by the short-term fixed pressure dead-end tests of SCR and irreversible fouling control as shown in Table 8.4. The short-term flux decline factor under unstirred filtration also showed that the flux for AS + Polymer bead sludge filtration reduced much faster than that of BAC as shown in Figure 8.13.

Table 8.4: Specific cake resistance and irreversible fouling control of AS + Polymer bead and BAC (between days 14-22)

MBR	Specific cake resistance (m/kg)	Membrane resistance increment after filtration (irreversible fouling) %
Polymer bead	$6.8 \times 10^{14} \pm 1.5 \times 10^{14}$	32.8 ± 3.1
BAC	$4.8 \times 10^{13} \pm 1.1 \times 10^{13}$	16.6 ± 3.2

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Figure 8.13: Flux decline profile for fixed pressure and unstirred dead-end filtration for BAC and AS + Polymer bead

Figure 8.14: Particle size distribution of BAC flocs and AS + polymer beads in volume (a) and number (b)

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Both the BAC and AS + Polymer bead sludge had a quite similar D_{50} (Table 7.6) and floc sizes distribution (Figure 8.14) but a great different in TOC concentration in supernatant of mixed liquor as shown in Table 8.3. The high TOC concentration (113.3 ± 7.2 mg/L) for MBR (Polymer bead) compared to lower TOC concentration (13.73 ± 2.2 mg/L) for MBR (BAC) may be the main factor caused the serious fouling for MBR (Polymer bead) during the aeration failure period. The TOC would block and plug the pores of the membrane and fill the gap between the sludge flocs to form non-porous cake layer on the membrane surface.

Figure 8.15: Performance comparison of MBR and MBR (polymer bead) at flux $19.0 \text{ L/m}^2/\text{hr}$

This indicates that the prevention of TOC attachment on the membrane surface is very important to have a good performance in the MBR (Polymer bead). The TOC may easily be detached from the membrane surface by the movement of the fibres and the collision of the polymer bead at lower flux. However, if the flux was increased to a higher value, the fine foulants (i.e. TOC or SMPs) would be more tightly attached onto the membrane surface and would not be easily detached. At day 24, both membranes from the MBRs were replaced by new membranes with a similar area (0.057 m^2) but operated at flux 19.0 instead of $10.5 \text{ L/m}^2/\text{hr}$. The higher imposed flux accelerated the TMP rise rate for both MBRs especially for MBR

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(Polymer bead). The TMP increased from 16.0 to 50 kPa and 15.8 to 21.6 for MBR (Polymer bead) and MBR (BAC) respectively as shown in Figure 8.15. The rapid TMP increase for MBR (Polymer bead) was probably due to inability of the polymer bead to detach the foulants on the membrane surface and the operating flux (19.0 L/m²/hr) being higher than the ‘sustainable’ flux as shown in Figure 8.16. This shows that beneficial of using the polymer beads in fouling control of MBR would only be significant if the operating flux is lower than the ‘sustainable’ flux.

Figure 8.16: Sustainable flux tests for BAC and AS + Polymer bead

These experiments indicate that under some conditions the polymer bead can play an effective role almost comparable to the MBR (BAC) in fouling control. However, the permeate quality was not as good as the MBR (BAC) (Table 8.3) and only a relatively low operating flux would see the benefit of MBR (Polymer bead) in fouling control. Nonetheless, MBR (Polymer bead) is still a potential alternative to the conventional MBR in terms of membrane fouling control.

In brief, the beads would impact and detach the foulants attached on the membrane surface. This process helps to improve the membrane fouling control. A few potential strategies as listed below could be used to optimise the use of the beads in MBRs to control membrane fouling:

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- (i) The beads should be easily floated by the aeration.
- (ii) The hydraulic movement should be well distributed to prevent the settlement of the beads.
- (iii) The surface of the beads should not damage the membranes during collision. Also, the beads must have a good resistance from being crushed.
- (iv) The intensity of the aeration rate used to scour the membranes should be sufficient to prevent trapping of the beads into the hollow fiber membrane bundle or the gap between the flat sheet membranes.
- (v) The size of the beads chosen to be used in tubular crossflow MBRs must smaller than the diameter of the tubular membranes. Flow velocity in the tubular tube should be sufficient to avoid the beads choking the membrane tubes.
- (vi) The size of the beads should be large enough to be separated from the wastage sludge using easy methods such as net filtration. The beads can be repeatedly used and save the cost of the operation.
- (vii) The collision may break the particles on the membrane surface into fine particles. Low flux ($< 15 \text{ L/m}^2/\text{hr}$) is preferable than that of relatively high flux.
- (viii) The SRT should be control so the MLSS is at a lower concentration. The quantity of the beads to be used should be determined to avoid the over MLSS concentration and subsequently increase the viscosity of the MLSS.

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8.5 Conclusions

The comparative filtration performance between AS vs supernatant and BAC vs supernatant is as below:

(i) It was found that the AS or BAC mixed liquor is less fouling than the supernatant separated out from the mixed liquor. The probable reasons are (i) the separated supernatant contained higher content of fine foulants (i.e. TOC and polysaccharide) which may due to lyses of the planktonic cells caused by insufficient substrate and oxygen in the supernatant chamber, (ii) the retention and accumulation of high MWt species in the chamber (in the absence of biodegradation) and (iii) the tendency for the fine particles to attach on the membrane surface without the presence of biomass or BAC flocs. The flocs could acted as a 'brush' to detach the fine foulants from the membrane surface back to the bulk liquid. In addition, the present of the relatively larger particles with adsorptive effects such as biomass or BAC flocs in the mixed liquor would prevent reattachment of the dispersed detached particle (due to 'collision' effect of the PAC) back to the membrane surface. It was found that BAC had a better performance than that of PAC or AS alone in membrane fouling control.

(ii) The comparative filtration results between MBR (AS) and MBR (polymer bead) suggest that the polymer beads could improve the fouling control. The advantages of using relatively big polymer beads (D_{50} : 600 μm) in the MBR included (i) they would create bigger impact of 'collision' effects and (ii) they could be easily separated from the waste sludge and be reused. In addition, the polymer beads may encourage the growth of attached biomass on its surface. This may help to reduce the planktonic cells in the supernatant. However, the operation of the MBR (polymer bead) at a relatively high flux may deteriorate the membrane performance and lead to more serious fouling. Higher flux tends to induce higher suction forces that cause more smaller flocs to firmly attach onto the membrane surface. The tightly attached fine particles on the membrane surface would be more difficult to detach by the 'collision' effect of the polymer beads and this may increase the

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membrane fouling rate. Also, the higher suction force may also attract polymer beads into the membrane bundle which may block the movement of the hollow fiber membrane and lead to more serious fouling problems. Overall the MBR (AS), MBR (Polymer bead) could be a potential alternative to the conventional MBR (AS) provided the operating flux is carefully controlled.

CHAPTER 9 CONCLUSIONS AND RECOMMENDATIONS

CHAPTER 9: CONCLUSIONS AND RECOMMENDATIONS**Synopsis:**

The purpose of this chapter is to summarize the results and discussions in the thesis. The results are briefly reviewed and recommendations regarding new directions in these areas of the study are proposed. The potential of the MBR (BAC) and MBR (polymer bead) in membrane fouling control is encouraging. As a result, the optimization of both systems is necessary especially in terms of concentrations of PAC (for the MBR (BAC)) and polymer beads (for the MBR (polymer bead)). Further study of the MBR (polymer bead) at different SRTs is also of interest.

9.1 Conclusions

The objectives of this study were to compare some alternative MBR concepts and operating strategies to minimize membrane fouling. Three approaches have been considered, one of which is to support the biomass on suspended PAC (adsorbent) in a MBR (BAC), the other is to attempt to operate the MBR with the membranes filtering supernatant liquor rather than concentrated MLSS and the third approach is an addition of the polymer bead in the MBR (polymer bead) to create collision effect to control membrane fouling. The major focus has been on the performance of the MBR (BAC), with some observations of supernatant filtration and the MBR (polymer bead) option. Investigation of the performance of the MBR (BAC) had been done intensively. As a result, two 20L MBRs were operated in continuous feed and filtration modes to simulate the conditions of a large-scale domestic MBR systems. One of the MBRs had added PAC and was identified as the MBR (BAC). The long term filtration performance was analyzed, evaluated and compared. In addition, 2L bench scale MBRs operated in batch-continuous mode were established to examine (i) the effect of different PAC concentrations on filtration performance improvement and mixed liquor composition, (ii) the mechanisms involved in the MBR (BAC) and (iii) the effect of different PAC sizes on membrane fouling reduction. In addition, 3.5L bench scale batch MBRs were also set-up to

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investigate the performance of (i) the MBR and MBR (BAC) at different SRTs and (ii) the MBR (polymer bead) compared to the MBR (BAC). A two x 20L compartment MBR was also commissioned that allowed the chambers to be operated in series to separate the supernatant from the SS. This was done by manipulating the recycle flow back to the first compartment from the bottom of the second compartment.

The observations and conclusions from this project are summarized below:

- a) Chapter 4: In the 20L MBR (BAC), accumulation of TOC was observed in the early stages of BAC cultivation. This was caused by the detachment of the biofilm formed on the BAC surface into the supernatant of the mixed liquor. The high TOC concentration in the supernatant of the mixed liquor in the MBR (BAC) was observed to have a relationship with the TMP profile. At high TOC (80-140 mg/L) concentration, the TMP rise was accelerated and led to a TMP 'jump' at the relatively high flux of 21.0 L/m²/hr. However, at lower flux (10.0 L/m²/hr) there was no observed early TMP 'jump' at the high TOC concentration in the MBR (BAC). The overall filtration performance for the MBR (BAC) was better than that of the MBR at steady state (constant low TOC concentration in the supernatant) and also unsteady state (sudden TOC increased in the supernatant of the mixed liquor). The better performance of the MBR (BAC) at steady and unsteady conditions may due to the BAC floc being equipped with simultaneous adsorption, decomposition and biodegradation properties. The BAC could also act as a 'scouring' tool through its simultaneous 'collisions' and adsorption effects to protect the membrane by (i) bringing the fine particles deposited on the membrane surface back to the bulk liquid, (ii) adsorbing components on the membrane surface and (iii) providing sites for foulants including planktonic cells on the membrane surface or dispersed finer foulants from the membrane surface in the supernatant to become incorporated into BAC floc. These mechanisms would help to reduce the membrane fouling even during the unsteady conditions in the bioreactor.

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The simultaneous adsorption, decomposition, biodegradation and 'scouring' mechanisms are regarded to be the major mechanisms involved in improving the filtration performance rather than that of 'collision' or adsorption effects alone.

In the MBR (BAC), the top-up of PAC to account for the loss of PAC in sludge wastage was done daily for the MBR with 10 or 30 days SRT. The newly added PAC in the hybrid MBR was able to adsorb the foulants more effectively compared to the aged (saturated) BAC in the system. The loss of the adsorption capacity of the aged BAC is probably due to pore saturation of BAC with the by-products of substrate biodegradation and decayed microbial cells. This indicates that the renewal of the BAC in the MBR (BAC) with fresh PAC is important to maintain the good function of the system. It was found that the filtration performance of MBR (aged BAC) was not better than that of MBR (AS) at low flux (7.0-10.5 L/m²/hr). At higher flux (21.0 L/m²/hr), the performance of the MBR (aged BAC) was much worse than the MBR. This may be because the aged BAC had lost its adsorption capacity and scouring effect (embedded into the biomass flocs) and also increased the MLSS concentration and shifted the particle size distribution to lower values. Instead of helping to improve the fouling control, the aged BAC may prevent the movement of the membrane fibers and inhibit the air bubbles in creating a shear force on the membrane surface. All of these limitations appear to lead to a more serious fouling for the MBR (aged BAC) especially at relatively high flux. A similar phenomenon was also observed in comparing the filtration of the MBR (10d) and MBR (30d) as well as the MBR (AS) and MBR (polymer bead) systems. The better filtration performance of the MBR (30d) and MBR (polymer bead) at lower flux became insignificant or worse at the relatively higher fluxes as compared to MBR (10d) and MBR (AS) respectively. At relatively high flux, the higher MLSS concentration for the MBR (30d) and MBR (polymer bead) became an obstruction to the membrane module

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movement and the efficient use of air bubbles in preventing accumulation of the fine particles on the membrane surface.

- b) Chapters 5 to 7: A concentration of 5 g/L of PAC in the continuous feed 20L MBR gave improved fouling control in the long term tests. The filtration results for two sets of four batch-continuous bench-scale 2L MBRs (0, 1, 3 and 5 g/L PAC in about 3.5 g/L of biomass) at SRT 30 and 10 days also showed that MBRs with any PAC concentration addition had a better performance than that of the PAC-free MBR (AS). Short-term characterization tests which involving the specific cake resistance (SCR), flux decline profiles, irreversible fouling and 'sustainable' flux at both fixed pressure and crossflow showed that the MBR with PAC addition were better than that of the MBR (AS). The SCR values were significantly lower for a PAC with 1 g/L and 3 g/L for SRT 30 and 10 days respectively due to their relatively large D_{50} compared to the others. Similar trends were obtained for flux decline profiles in dead end filtration. The SCR and flux declines for 0 g/L PAC were the worst. However, the short-term tests measuring irreversible fouling resistance and 'sustainable' (low TMP rise) flux favoured 5 g/L PAC addition for both SRT 10 and 30 days, and again 0 g/L PAC mixed liquor performed much worse. This suggests that the 'sustainable' flux and irreversible fouling tests are better predictors than that of the SCR and the flux decline profile. However, both of them could only be regarded as approximate predictors of long term performance for the MBR with PAC addition.

However, for comparison between MBRs without PAC, the SCR may be the best predictor for long term filtration performance as compared to the other short term test predictors. For example (based on section 7.2) the mixed liquor from MBR (30d) had the worse short term test results for 'sustainable' flux, irreversible fouling and flux decline profile but only better for the SCR as compared to the MBR (10d). However, the long term filtration tests showed that the MBR (30d) was better compared to the MBR

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(10d) at fluxes of 10.0 and 15.0 L/m²/hr. The better lower SCR would probably mean that the bigger biomass floc size of the AS_{30d} compared to AS_{10d} may have helped in creating a better scouring effect. The irreversible fouling and flux decline profile tests were all done at fixed pressure and this produced high flux at the initial stage. This means that these tests only reflect the performance of the MBR run at relatively high flux. The MBR (30d) performed much worse than the MBR (10d) at a flux of 23.0 L/m²/hr. This means that irreversible fouling and flux decline profile tests would only be good predictors for the MBR operated at relatively high flux. For the ‘sustainable’ flux short term test the flux steps were done at approximately 15 minute intervals. This short period may be sufficient to indicate the performance of the MBRs at higher flux (> 30.0 L/m²/hr) but possibly not enough to distinguish the performance between the MBRs operated at lower flux. Overall, the short term predictors discussed above may give sufficient information to predict the long term performance of the MBRs and MBR (BAC)s if only we can define them and properly understand the mechanisms involved in fouling control.

Based on the results in chapter 6, it was found that only the fresh BAC was equipped with simultaneous adsorption, decomposition, biodegradation and scouring effects. ‘Aged’ BAC fully incorporated within the biomass floc and with pores filled with fine foulants would lose those functional mechanisms. Therefore to optimize the beneficial effect of PAC in improving MBR performance, the aged BAC should be replaced regularly.

- c) Chapter 7: A series of experiments proven the benefit of using PAC in enhancing the performance of the MBR. The effect of PAC size on the MBR (BAC) performance was investigated. It was found that using smaller size of PAC (D_{50} : $14.88 \pm 3.93 \mu\text{m}$ (volume); $0.24 \pm 0.06 \mu\text{m}$ (number)) was more beneficial than relatively larger PAC size (D_{50} : $67.60 \pm 2.70 \mu\text{m}$ (volume); $1.72 \pm 0.11 \mu\text{m}$ (number)) in terms of fouling control but only at a relatively low flux (8.5 L/m²/hr). The benefit of the smaller PAC size may be due to

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slightly greater adsorption capacity (higher surface area) making it easier to pick up the fine foulants more effectively. However, when the flux was increased from 8.5 to 13.5 L/m²/hr the observed benefit of the smaller PAC size receded. Larger PAC size was better in controlling the fouling rate at a flux of 13.5 L/m²/hr. This was caused by the resistance at the higher flux induced by the deposition of the smaller or finer BAC on the membrane surface. The deposition caused the fouling rate to increase. This means that the choice of the correct PAC size should be based on the flux imposed on the MBR. This is an important consideration in order to prolong the MBR (BAC) operation.

In this study, an investigation of the conventional MBRs at different SRTs (10 and 30 days) was also carried out. It was found that the MBR operated at SRT of 30 days had a better filtration performance than that of the MBR with SRT 10 days at relatively low flux (10.0 and 15.0 L/m²/hr) although it had a higher MLSS concentration and more TOC and polysaccharides in the supernatant of the mixed liquor. A possible reason for the better performance may be the relatively bigger floc sizes for AS_{30d} which could create a better scouring effect than the AS_{10d}. However, the higher concentration of MLSS and fine foulants in the supernatant of the mixed liquor of the MBR (30d) was also a limitation for its operation at a higher flux (23.0 L/m²/hr). The greater flux may convect more MLSS to accumulate around the membrane module and cause (i) the fine foulants to be more tightly attached on the membrane surface, (ii) the movement of the membrane module to be impeded, (iii) the pathway of the air bubbles to the membrane surface to be blocked and (iv) the 'collision' effect to diminish. Thus, the MBR (30d) performed worse than the MBR (10d) at a flux 23.0 L/m²/hr.

For the MBR (BAC)s operated at different SRTs of 10 days, 30 days and long time, the best performer was the MBR (BAC_{10d}) and the worst was the MBR (BAC_{long}). As discussed in the section 7.3, the MBR (BAC_{10d}) at SRT

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10 days, was the system which had the most regular replenishment of the aged BAC with fresh PAC, followed by the MBR (BAC_{30d}). At long SRT, the PAC in the MBR (BAC_{long}) was not replaced during the operating period. The MBR (BAC_{10d}), with more fresh BAC and the lowest MLSS concentration would mean that (i) the adsorption of the fine foulants in the supernatant of mixed liquor and scouring effect would be more significant and (ii) the obstruction of the membrane movement would also less. These strengths helped to achieve the good performance of the MBR (BAC_{10d}) compared to the MBR (BAC_{30d}) and MBR (BAC_{long}). On the other hand, without replacement of the aged BAC and with higher MLSS and TOC concentration in the supernatant (Table 7.3), the performance of MBR (BAC_{long}) was the worst among the three MBR (BAC)s. The BAC size of the MBR (BAC_{long}) was the biggest and this meant that the PAC had been seriously embedded within the biomass flocs. This caused the BAC to lose its adsorption and scouring effects.

- d) The direct contact between the membrane and the MLSS could be considered as one of the causes of membrane fouling. This suggests that the filtration of the supernatant could be more beneficial than that of the MLSS. In this study, a two-compartment MBR was designed and operated in a way that separated the supernatant from the MLSS without the sedimentation stage. Surprisingly, it was found that, filtration of the separated supernatant from the mixed liquor with or without PAC addition was much worse than that of filtration of the MLSS. Investigation found that the supernatant separated from the mixed liquor had a higher concentration of fine foulants compared to the supernatant of the mixed liquor. This may be due to (i) the retention and accumulation of high MWt species in the chamber (in the absence of biodegradation) and (ii) the lack of food and oxygen to sustain the viability of the planktonic cells in the supernatant chamber. The starvation and depleted oxygen conditions may have caused lyses of the planktonic cells. The excretions from the cells may contribute to the concentrations of the fine foulants in the supernatant chamber. In addition,

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the absent of the large particles with adsorption properties such as biomass or BAC flocs to detach the fine particles from the membrane surface may also be factors that led to more serious fouling.

- e) Chapter 8: In addition to the MBR (BAC), the MBR (polymer bead) may also be a useful alternative design which could improve filtration performance of the MBR (AS). The relatively large polymer beads ($\approx 600 \mu\text{m}$) in the MBR had a significant scouring effect and were able to detach the fine particles from the membrane surface. Equally important, the polymer beads were easily separated from the wastage mixed liquor and could be reused. The MBR (BAC) lacked this advantage. The powdered PAC is difficult to regenerate and separate from the sludge. The few regeneration methods such as, (i) heat treatment in a furnace and (ii) chemical washing are time consuming and costly. However, overall, the filtration performance of the MBR (BAC) was significantly better than that of the MBR (polymer) in terms of (i) fouling control, (ii) permeate quality and (iii) operation at relatively higher flux.

9.2 Recommendations

It was found that BAC could help to reduce the membrane fouling tendency compared to AS in the MBR. As a result, optimization of the MBR (BAC) in terms of PAC concentration is needed and worthy of further investigation. However, renewal of the saturated BAC with fresh PAC is required to keep the fouling control mechanisms (simultaneous adsorption, decomposition, biodegradation and 'scouring') of the BAC performing. Effective and economic methods are required to regenerate the saturated PAC in order to add more value to its beneficial function in membrane fouling control. One of the recommendations is to investigate bioregeneration of the saturated PAC under anaerobic conditions which is potentially more economic and environmentally friendly compared to combustion techniques and chemical cleaning.

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In the study, there was difficulty to separate the used PAC from the biomass by using chemicals. Combustion may be a good method to recycle and regenerate the used PAC, but needs high energy and may not be economic. To use fresh and cheap adsorbent is a good alternative to replace used adsorbent. Adsorbent in the MBR system would reduce the membrane fouling rate and subsequently reduce the rate of chemical cleaning and permeate production. This indicates that by using adsorbent in the MBR system may reduce the operation cost. It is suggested that the next PhD student may consider spending some efforts in this area in terms of operation cost of the MBR (BAC) as compared to the conventional MBR.

As a result, other adsorbents (i.e. zeolite, polymeric particles, porous silica, sponge-like solids, rice husk, palm husk etc) should be tested for their effectiveness in membrane fouling control as an alternative to the relatively expensive PAC.

It was found in this study that filtration of the supernatant had no benefit compared to MLSS. This may be due to more high MWt fine foulant in the separated supernatant compared to the supernatant in the mixed liquor. We assume this was caused by the lysed planktonic cell in the supernatant chamber and the retention and accumulation of high MWt species in the chamber (in the absence of biodegradation). A preliminary test showed that continuous return of the supernatant in the supernatant chamber to the mixed liquor chamber could reduce its fine foulant concentration. Other methods to prevent the planktonic cells from going through the lysis process and accumulating in the supernatant chamber are worth investigating. In addition, the addition of large polymer beads ($\approx 600\ \mu\text{m}$) in the supernatant chamber to test its feasibility as 'scouring' agent in helping fouling reduction could be evaluated.

Further work is also recommended to understand more fully the role of the polymer beads in the MBR (polymer bead). There are some questions such as, (i) would the polymer beads act as biomass carriers for which the attached growth bacteria have the ability to decompose the high molecular weight organics as found with the BAC, (ii) would the beads perform better in a higher SRT MBR (i.e. 30 or longer

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days) compared to a SRT 10 days in terms of membrane fouling control and permeate quality and (iii) is less or higher concentration than 5 g/L perform better. All of the above suggestions are recommended for future study.

In addition, the other recommendations are as follow:

- (i) To investigate the influence of PAC concentration and particle size on the viscosity evolution of the suspension.
- (ii) To investigate the scouring effect in comparison with practice of relaxation or back-flush performances.
- (iii) To investigate the polymer beads influence by comparison of relaxation or back-flush practices according to the composition and compactness of the cake layer deposit.
- (iv) To investigate the PAC sorption capacity to evaluate the optimal PAC retention time with TOC in solution.
- (v) To investigate the biological performances in terms of apparition of very high TOC values. Is it due to a progressive accumulation of non biodegradable compounds coming from feed or SMPs or due to not optimal functioning of the biomass?
- (vi) To investigate choice of an optimal dose of added PAC according to adsorption rate and capacity linked to high TOC values.
- (vii) To investigate origin of fouling (cake deposit or irreversible fouling) that induces the type of membrane regeneration, external fouling and interest of scouring or relaxation (according to the composition and compactness of cake layer) or chemical cleaning (for irreversible fouling).
- (viii) To investigate sludge production and valorisation, energetic costs and MBR development to solve the case of specific industrial waste.

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(ix) To investigate the quantity of polymer beads needed and the frequency of hits between membrane and beads for possible scale-ups.

(x) To investigate the impact of limiting nutrients to the growth of the biomass, thus producing the suspended polysaccharide.

Finally, I hope that the MBR (BAC) and MBR (Polymer bead) can be applied in real world wastewater treatment plant to evaluate their performance.

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Appendix A: Methods for determining the PAC concentration in the MBR (BAC)

The concentrations of PAC in the bioreactors should be kept at the same level during the experimental runs. This means that the amount of PAC lost during sampling or by wastage of sludge should be replaced. The concentration of PAC in the sample can be estimated using equations (2) and (3). The equations are based on the results shown in Figures A.1 and A.2.

There is about 8-10 % of biomass from the laboratory pilot scale two compartment MBR that could not be burnt off at the temperature of 950° C as shown in Figure A.1. Figure A.2 shows that the weight of the remains for all the samples are about 24 % of the original weight of the samples after burning at the same temperature.

Figure A.1: Weight of biomass lost after burning at temperature of 950° C

APPENDIX A

Figure A.2: Weight of PAC lost after burning at temperature of 950° C

Weight of PAC and biomass mixture after drying at 105° C (R) = X + Y

$$R = X + Y \tag{2}$$

Where X is the weight (g) of PAC concentrations in the sample and Y is the weight (g) of biomass in the sample. At a temperature of 950° C, Figure A.2 shows that the remains of PAC is about 24 % of the original weight. Equation (3) can be written with this assumption.

Weight of PAC and biomass mixture after drying at 950° C (P) = 24%X + 9%Y

$$P = 0.24X + 0.09Y \tag{3}$$

By solving equations (2) and (3), the concentrations of PAC and biomass can be determined. The equations can also be used to determine the weight of PAC and biomass at low MLSS concentrations. In the future works, the equations can be applied to calculate the concentration of PAC in the wasted sludge. Then, PAC can be added back into the bioreactors. It was found that the value of the biomass after

APPENDIX A

burning remains at the temperature of 950° C changes from one bioreactor to the other bioreactor. It is recommended to repeat the biomass burning process at the temperature of 950° C to confirm and determine the biomass remains for the individual bioreactor.

Appendix B: EPSs concentration in the MBR (BAC) and MBR (AS) in chapter 4 (section 4.1)

Figure 4.1.B: Concentrations of polysaccharide in the supernatant of the MBR (BAC) and MBR (AS)

Figure 4.2.B: Concentrations of polysaccharide in the pellet of the MBR (BAC) and MBR (AS)

APPENDIX B

Figure 4.3.B: Concentrations of protein in the supernatant of the MBR (BAC) and MBR (AS)

Figure 4.4.B: Concentrations of protein in the pellet of the MBR (BAC) and MBR (AS)

APPENDIX B

Figure 4.5.B: Concentrations of total EPSs in the MBR (BAC) and MBR (AS)

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