

This article was downloaded by:

On: 25 January 2011

Access details: *Access Details: Free Access*

Publisher *Taylor & Francis*

Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37-41 Mortimer Street, London W1T 3JH, UK



Separation Science and Technology

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713708471>

Operation of Membrane Bioreactor with Powdered Activated Carbon Addition

Choon Aun Ng^a; Darren Sun^a; Anthony G. Fane^a

^a School of Civil and Environmental Engineering, Nanyang Technological University, Singapore

To cite this Article Ng, Choon Aun , Sun, Darren and Fane, Anthony G.(2006) 'Operation of Membrane Bioreactor with Powdered Activated Carbon Addition', *Separation Science and Technology*, 41: 7, 1447 — 1466

To link to this Article: DOI: 10.1080/01496390600634632

URL: <http://dx.doi.org/10.1080/01496390600634632>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Operation of Membrane Bioreactor with Powdered Activated Carbon Addition

Choon Aun Ng, Darren Sun, and Anthony G. Fane

School of Civil and Environmental Engineering,
Nanyang Technological University, Singapore

Abstract: The effect of powdered activated carbon (PAC) addition to the activated sludge (AS) in a membrane bioreactor (MBR) has been investigated. The long term nature of the tests allowed the PAC to gradually incorporate into the biofloc forming biologically activated carbon (BAC). One series of tests involved 4 bench scale (2 L) MBRs operated at sludge retention times (SRTs) of 30 days with PAC inventories of 0, 1, 3 and 5 g/L and steady state biomass concentrations of 12.0 ± 1.0 g/L. The characteristics of the mixed liquors (MLSS) from the 4 reactors were compared. Short term filtration tests, including measurement of specific cake resistance (SCR), flux decline profile, and irreversible fouling resistance in an unstirred cell and “sustainable” flux (by monitoring transmembrane pressure (TMP) rise) in a crossflow cell all showed better filtration performance for the MLSS with BAC compared with the AS alone. In terms of SCR and flux decline profile the 1 g/L PAC addition performed best, but in terms of minimizing irreversible membrane fouling and maximizing “sustainable” flux the 5 g/L PAC was best. All 4 systems showed lower total organic carbon (TOC) in the permeate compared to the bioreactors, but the lowest permeate TOC (and the best removal) was for the highest PAC loading.

The benefit of PAC addition was confirmed in a second series of tests with two 20 L MBRs with submerged hollow fibers, one operated without PAC, the MBR(AS), and the other with 5 g/L PAC, the MBR(BAC). For an SRT of 30 days (which involved 3.3% sludge wastage per day and 3.3% new PAC addition per day) and a fixed flux of $21 \text{ L/m}^2\text{hr}$ the MBR(AS) showed a TMP rise of about 2.4 kPa/day whereas the MBR(BAC) showed a rise of only 0.8 kPa/day. However when the MBRs were operated without wastage the performance of the MBR(BAC) was worse than the

Received 4 January 2006, Accepted 3 February 2006

Address correspondence to Anthony G. Fane, School of Civil and Environmental Engineering, Nanyang Technological University, 637723, Singapore. Tel.: (65) 6794 3801; Fax: (65) 6792 1291; E-mail: agfane@ntu.edu.sg

MBR(AS). Thus the improved performance of the MBR(BAC) requires regular replenishment of aged BAC with fresh PAC.

Keywords: Membrane bioreactor, powdered activated carbon, specific cake resistance, irreversible fouling, sustainable flux, biologically activated carbon

INTRODUCTION

Membrane bioreactors (MBRs) are increasingly used as alternatives to the conventional activated sludge treatment process. The distinguishing characteristics of MBRs include

- i. better quality treated effluent that is suspended-solids free and with less residual organics,
- ii. the systems are compact with small footprint and
- iii. there is potentially less excess sludge production.

However, the positive characteristics of MBRs have been hampered by membrane fouling due to cake accumulation on the membrane surface (1, 2) and/or membrane pore plugging/blocking (1, 3–6).

Several strategies are used to reduce membrane fouling such as

- i. applying vigorous aeration to scour the membrane in submerged membrane systems (3, 7–14)
- ii. applying two-phase flow to the lumen of sidestream hollow fiber modules (15–19)
- iii. physical and chemical cleaning (20, 21)
- iv. intermittent suction operation (8, 11, 22)
- v. operating at fixed (moderate) flux or below the critical (sustainable) flux (23–25) and
- vi. intermittent backwashing operation (10, 14, 26–28).

An alternative approach is to modify the characteristics of the mixed liquor suspension by additives. In this study we examine the addition of powdered activated carbon (PAC) which could modify the MLSS floc and provide potentially beneficial adsorptive properties. A limited number of previous studies (29–31) have used PAC to improve removal efficiency and to control fouling in MBRs.

PAC has been used with the conventional activated sludge (AS) wastewater treatment process to treat wastewater containing

- i. inhibitory materials (32)
- ii. landfill leachate (33)
- iii. phenol or aniline (34)

- iv. high salinity oil-field brine (35) and
- v. color from the textile industry (36).

In the activated sludge environment, a layer of biofilm tends to form on the PAC surface to form “biological activated carbon” (BAC) sludge. The biofilm should be able to biodegrade the pollutants previously adsorbed by the PAC, leading to simultaneous adsorption and biodegradation processes rather than the biological process alone (33, 34, 37). Thus the potential advantage of BAC in wastewater treatment is that the biofilm on the PAC consists of immobilized (35) and acclimatized bacteria (38) that can also partly bioregenerate the saturated BAC (39). In addition, the succession of bacteria in the biofilm ecosystem can also enhance the performance of BAC in pollutant removal (38, 40). Claimed advantages of BAC include,

- i. increasing the efficiency of substrate removal,
- ii. improved mixed liquor filterability and
- iii. reducing the adverse effect of heavy metal ions on biomass through adsorption (40). However, in long-term operation, the characteristics of simultaneous adsorption and biodegradation of organics by BAC would probably be reduced due to the loss of the adsorption capacity of the BAC.

Factors causing reduction in the bioregeneration of BAC include,

- i. limited access to the interior of the PAC particles,
- ii. filling of the mesopores with the products of microbial biodegradation and,
- iii. strongly adsorbed recalcitrant organics (41, 42).

This suggests that replacement of aging BAC with reduced capacity may be important to maintain the properties of simultaneous adsorption and biodegradation of BAC.

A few studies of the hybrid PAC MBR (referred to hereafter as the MBR(BAC)) have been reported and the results show that the addition of PAC can enhance the performance of the MBR system. It was suggested by Kim et al., (43) that the improved performance of the MBR(BAC) was due to

1. a reduction in the extracellular polymeric substances (EPS) in the floc and
2. the formation of BAC with high porosity and low compressibility.

However the benefit of high porosity and low compressibility may only be evident if MLSS cake formation is allowed to occur, and this may not be typical for MBRs. It should also be noted that Kim et al. (43), used an MBR with an unusually low SRT of 6–8 days and a biomass level of only approximately 3 g/L (more typical values would be SRTs of 15 to 30 days and MLSS of 8 to 12 g/L). Others have suggested that the BAC would form a permeable

particulate layer on the membrane surface to act as a “precoat” filter layer for pollutants removal (29, 30). This may indicate that the particulate filter layer could also protect the membrane from pore blocking and plugging by filtering the pollutants during filtration. However, as noted above, to achieve this the flux needs to be adjusted to above the critical flux (for cake formation) of the floc, which itself could cause fouling issues.

Pibazari et al., (29) suggested that BAC floc in a MBR could produce added fluid turbulence in the presence of bubbling. The improved fluid turbulence in the bioreactor could help to depolarize particle accumulation on the membrane surface. However to use the PAC as a “scouring agent” may require a high PAC loading and this needs to be optimized. In summary, from the literature, it is evident that PAC is beneficial for use with activated sludge to achieve a better performance in wastewater treatment. However, the literature suggests that the optimal conditions in the MBR(BAC) need to be identified to obtain the improved performance.

The objective of this paper is to investigate how PAC can improve the MBR filtration under conditions not previously reported. We have attempted to apply a protocol to give a fair comparison of the effects of different PAC concentrations on the characteristics of AS at high MLSS concentrations (10.0 ± 2.0 g/L), typical of current generation MBRs. The properties and filtration characteristics of BACs have been measured in both short-term tests and in long-term continuous operation comparing a MBR(AS), (i.e., no PAC) and a MBR(BAC) run in parallel at SRTs of 30 days and ‘infinity’ (without wastage).

EXPERIMENTAL

Materials

PAC (Hydrotarco 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 Fig. 1, was measured by a Malvern Mastersizer particle size analyzer. The BET surface area of the fresh PAC was about $488 \text{ m}^2/\text{g}$.

The filtration characteristics of the BAC were measured in short-term tests in a dead-end filtration cell fitted with Millipore ultrafiltration Polyethersulfone membranes with a molecular weight cut-off of 50,000 Da. Some short-term crossflow tests were also performed using flat sheet $0.2 \mu\text{m}$ pore size microfiltration membranes. For the long-term submerged MBR trials, hollow fiber microfiltration Polyacrylonitrile membranes from Singapore Cleanseas Ltd were used for filtration comparison.

Operation of MBRs

Four 2 L (batch-continuous) and two 20 L (continuous operation) MBRs were set-up as shown in Fig. 2(a) and (d) respectively. The activated sludge used

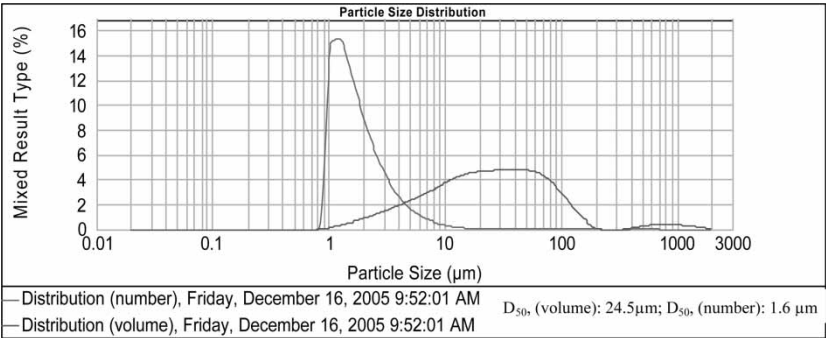


Figure 1. The particle size distributions of PAC used in MBRs.

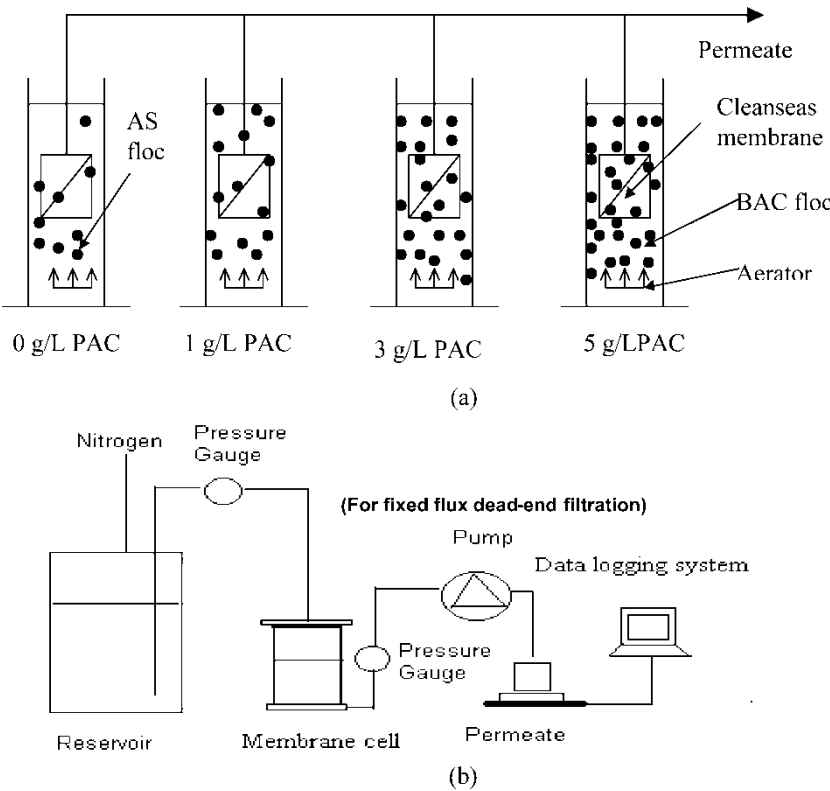
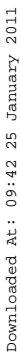
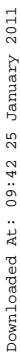


Figure 2. Schematic diagram of (a) 2 L MBRs (b) unstirred membrane cell unit (c) crossflow module cell unit for membrane filtration and (d) 20 L MBRs.

(continued)



Downloaded At: 09:42 25 January 2011



Downloaded At: 09:42 25 January 2011

Downloaded At: 09:42 25 January 2011

Downloaded At: 09:42 25 January 2011

Table 1. Concentration of the MLSS and PAC in MBRs

Name of MBRs	MLSS concentrations (g/L)	PAC concentrations (g/L)	Working volume (L)
0 g/L PAC	12.0 ± 1.0	0.0	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
MBR(AS)	9.5 ± 1.0	0.0	20.0
MBR(BAC)	14.5 ± 1.0	5.0	20.0

Membrane Cell and Crossflow Module Cell Filtration Tests

The effect of the PAC on the fouling tendency of the AS was examined in both short-term filtration tests on mixed liquor samples from the 4 × 2 L MBRs and by monitoring the long-term membrane performance in the two 20 L MBRs. 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) = $1.26 \times 10^{-3} \text{ m}^2$) and a crossflow cell ($A = 0.8 \times 10^{-3} \text{ m}^2$). The parameters measured as characteristic of the fouling tendency of the AS and the BAC were the specific cake resistance (SCR), the flux decline profile (flux vs concentration factor in dead-end), the irreversible fouling resistance R_{if} (dead-end) and the “sustainable” flux (crossflow). The unstirred dead-end cell and the crossflow cell set-ups are shown in Figs. 2(b) and 2 (c) respectively. The crossflow cell as shown in Fig. 2(c) was used to filter samples from each 2 L MBR at increasing fluxes at a crossflow of 0.2 m/s for determination of the “sustainable flux”. 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 (25) of the dominant foulant. The SCR was measured at both fixed pressure (100 kPa) and at modest (fixed) flux (= 20 L/m²hr). Flux was measured by weighing the permeate mass with an electronic balance interfaced to a personal computer. The Labview program was used to data-log the values of feed and permeate pressure and flux during the dead-end and crossflow experiments.

Resistances were estimated from the Darcy equation,

$$\text{Resistance, } R = \frac{\Delta P}{\mu J} \quad (1)$$

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,

$$R_t = R_m + R_c + R_{if} \quad (2)$$

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 cake resistance, R_c , was obtained from equation (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 [44],

$$\frac{t}{V} = \frac{\mu R_m}{A \Delta P} + \frac{\mu C_b \alpha}{2A^2 \Delta P} V \quad (3)$$

The SCR (α) was obtained from the slope of the plot. The SCR at constant flux was estimated from the measured cake resistance using,

$$R_c = \alpha \frac{M}{A} \quad (4)$$

Where R_c (m^{-1}) is the total cake resistance, α (m/kg) is the specific cake resistance and C_b (mg/L) is the feed MLSS concentration. M and A are mass of the filter cake (kg) and membrane area (m^2), respectively. The cake mass (M) was estimated from the sample volume and its concentration ($M = V_s \times C_b$); where V_s (L) was the sample volume.

Analytical Methods

TOC was measured by a Shimadzu VCSH analyzer; samples were prefiltered at $0.45 \mu\text{m}$ prior to analysis. The suspended solids (SS) were measured according to Standard Methods using an Edwards air vacuum and a GC-50 glass fiber filter ($1.2 \mu\text{m}$). 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. An optical microscope (KEYENCE VH-Z450) was used to observe images of the AS and BAC floc.

RESULTS AND DISCUSSION

AS and BAC Cultivation

The PAC used in this study had the particle size distributions shown in Fig. 1. The mean particle size of the PAC in terms of volume and number distribution was $24.5 \mu\text{m}$ and $1.6 \mu\text{m}$ respectively. This means that, the addition of the PAC would tend to increase the small particle population of the AS and

shift the BAC floc sizes to lower values. As shown below, it was found that the size of the BAC floc formation depended on the PAC concentration.

The MLSS profiles for the four 2 L MBRs are shown in Fig. 3. The 2 L 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 2 L 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).

The 2 L 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 3.35 wt% 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. A mass balance check was made to confirm that the PAC contents were maintained at steady state. From Fig. 3, it can be seen that for all MBRs the MLSS contents became stable after approximately 100 days (about $3 \times \text{SRT}$). The steady-state concentrations are given in Table 1 and it is evident that the 4 MBRs all achieved a similar biomass concentration ($[\text{MLSS} - \text{PAC}] = 12.0 \pm 1.0 \text{ g/L}$). A series of experimental analyses to compare the various characteristics of the MLSS in the 2 L MBRs was started on day 109. MLSS samples were withdrawn from each of the bioreactors and tested in terms of

- i. specific cake resistance SCR
- ii. flux decline profile at unstirred fixed pressure
- iii. the mean floc size (D_{50})
- iv. TOC concentration in the supernatant and permeate
- v. the irreversible fouling resistance R_{if} and
- vi. the “sustainable” flux.

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 Fig. 4. From Fig. 4, it can be

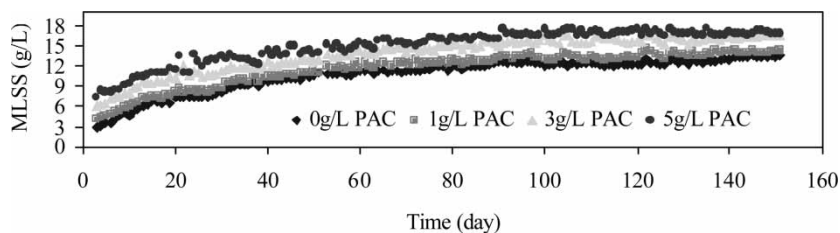


Figure 3. MLSS development in 2 L MBRs.

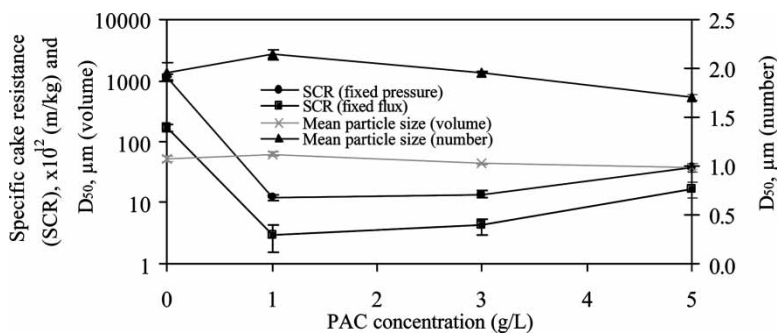


Figure 4. Relationship of D_{50} to the specific cake resistance in fixed pressure and flux.

seen that the SCR measured by modest flux ($\leq 20 \text{ L}/\text{m}^2 \text{ hr}$) was lower than that measured by fixed pressure (100 kPa) dead-end filtration. 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 ($> 100 \text{ L}/\text{m}^2 \text{ hr}$). Our observations agree with the fixed flux vs fixed pressure comparison of Defrance and Jaffrin (45). 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 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 $> 350 \text{ L}/\text{m}^2 \text{ hr}$. One conclusion from this study is that characterization of the SCR of MLSS in MBRs is best done at constant flux, although the trends (Fig. 4) are qualitatively similar for SCR (constant pressure).

The results in Figs. 4 and 5 clearly show that PAC within the AS had the ability to reduce the SCR and lift the flux decline profile significantly.

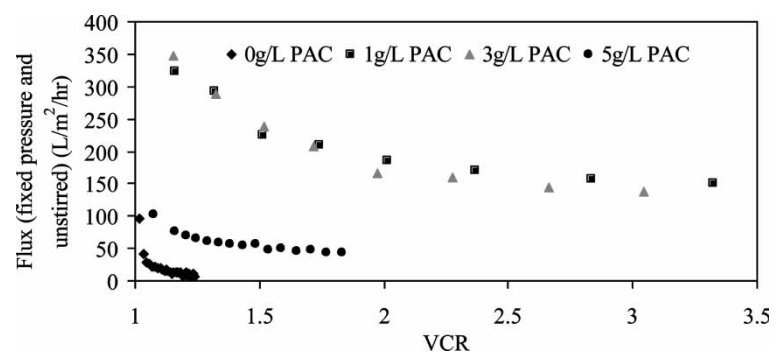


Figure 5. Flux decline profile for fixed pressure and unstirred dead-end filtration.

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 causes.

- 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 (30, 42). According to the Carman–Kozeny equation, the specific cake resistance (SCR) α , can be expressed by equation (5)

$$\alpha = \frac{180(1 - \varepsilon)}{\rho \cdot d_p^2 \cdot \varepsilon^3} \quad (5)$$

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), the effective particle size (d_p) and therefore the floc size distribution is another important factor that can affect the reduction of the SCR. Figure 4 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. (43) This trend was probably due to the fact that PAC was in powder form with relatively smaller size (see size distribution data in Fig. 1 and images in Fig. 6) 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 Fig. 4, 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). Thus D_{50} was in the decreasing order, BAC with

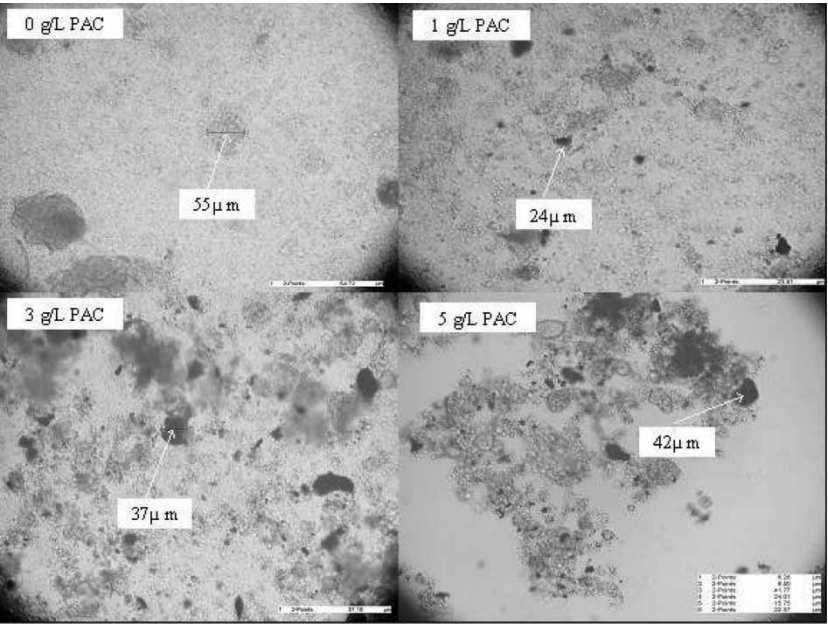


Figure 6. Images of flocs from 2L MBRs with different PAC concentrations.

1 > 3 > 5 g/L of PAC and the SCR was in the increasing order, BAC with 1 < 3 < 5 g/L of PAC.

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 (46). 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 Fig. 7 (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 (Fig. 5) 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.

It is of interest to note from Fig. 7 that there were significant differences in the TOC values in the MBRs compared 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, and 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 were

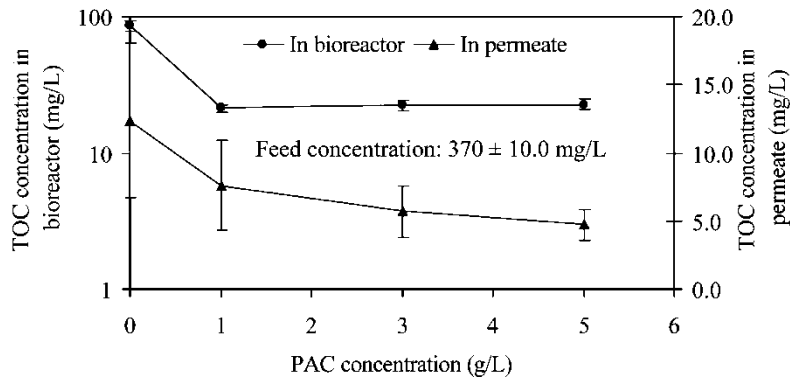


Figure 7. TOC in the bioreactors and permeate with different PAC concentrations.

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 (39) and Mochidzuki et al. (40) 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 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 Fig. 8 (the y axis represents the % increase in membrane resistance due to R_{if} after the filtration test). This may be due to the higher concentration of BAC forming a protective layer and preventing the foulants, such as TOC including EPS or fine colloids, from entering, blocking, or plugging the pores of the membrane. In addition, crossflow tests were performed on samples (50 mL MLSS diluted

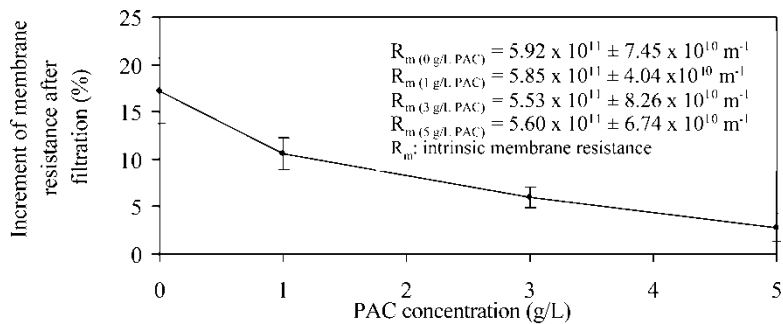


Figure 8. Membrane resistance increment due to irreversible fouling.

with 1950 mL of Milli-Q water) from each 2 L 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 Fig. 9. It was found that AS alone, without PAC, had the lowest sustainable flux ($<36\text{ L/m}^2\text{ hr}$), MLSS with 1 and 3 g/L of PAC had higher sustainable fluxes at about $36\text{ L/m}^2\text{ hr}$ and MLSS with 5 g/L of PAC had the highest sustainable flux at about $43\text{ L/m}^2\text{ hr}$.

In summary, the results from the $4 \times 2\text{ L}$ reactors showed the following;

- 1. 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,
- 2. 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 minimized 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 2 L MBRs point to the use of 5 g/L PAC as the better option.

Performance Comparison of MBR (AS) and MBR (BAC) at SRT 30 days

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 30 days and 5 g/L of PAC was added to one of them. Constant top-up of lost PAC (3.35 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

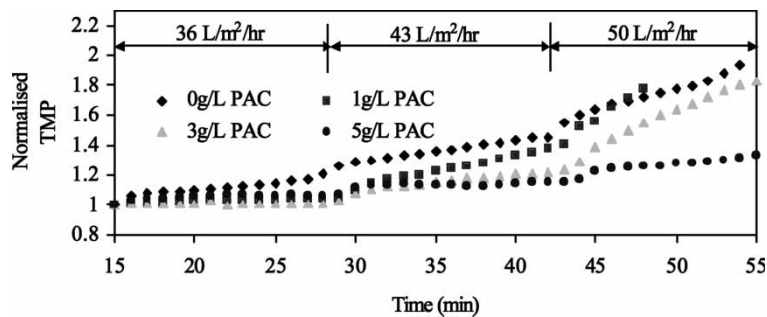


Figure 9. Sustainable flux tests for MLSS with different PAC concentrations.

for these tests, both MBRs were operated in parallel at a fixed permeate flowrate of 40 mL/min (flux = 21 L/m² hr, HRT = 8.3 hrs) to achieve a steady state. At day 177, new membrane modules with an area of 0.114 m² were submerged into each MBR and run at a fixed flux of 21.0 L/m²/hr. The key parameter used to characterise fouling was the TMP. The results are shown in Fig. 10 which clearly indicates 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 27 days.

The improved membrane performance of the MBR (BAC) with added PAC could be due to a number of factors which we discuss below.

- i. The PAC provides a sink for some of the fouling components such as EPS and fine colloids. Evidence in favour of this is provided in Figs. 8, 9, and 10, for the 2L MBRs, where the TOC for MBR(AS) was significantly higher than for MBR(BAC) and the MBR(AS) showed greater fouling.
- 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 (47) 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 next section (3.3) cast some doubt on this mechanism in the MBR (BAC).

Inspection of the hollow fibers removed from the MBRs at day 201 revealed negligible cake deposit or accumulation, as shown in the photo images (Fig. 10, inserts). However at that 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

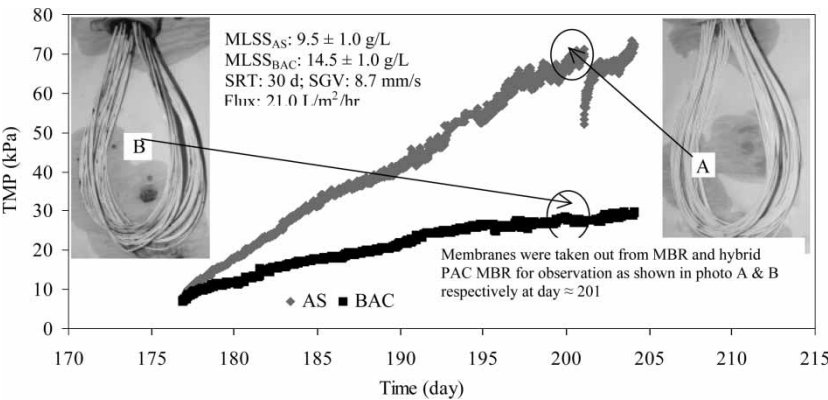


Figure 10. Performance comparison of MBR(AS) and MBR(BAC) at SRT 30 d.

new membranes (see Fig. 10). After water washing the MBR(AS) membranes had a required TMP of 50 kPa. Following rinsing, the 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.

These observations 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 reversible fouling resistance, which is in agreement with the short term test results showing lower R_{if} with BAC (Fig. 8).

**Performance Comparison of MBR(AS) and MBR(BAC)
At “infinite” SRT**

To further investigate the role of PAC in mitigation of MBR fouling the two 20 L MBRs were operated without sludge wastage (effectively infinite SRT). This operating strategy meant that the 5 g/L PAC inventory in the MBR(BAC) was not gradually replenished but remained “aged” in the MBR.

The performance comparison of the two MBRs running at fluxes of 21.0 L/m²hr was done after operating the two MBRs for 35 days, when new membranes were introduced. The results in Fig. 11 show that the MBR (aging BAC) had a greater fouling rate than the MBR(AS). Cleaning the membranes from the MBR(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(AS).

The worse performance of the MBR(BAC) compared with the MBR(AS) at infinite SRT contrasts dramatically with the much better performance at the SRT of 30 days. It is important to note that this behavior was qualitatively reproducible. Thus the benefit of the PAC inventory was lost as the BAC aged.

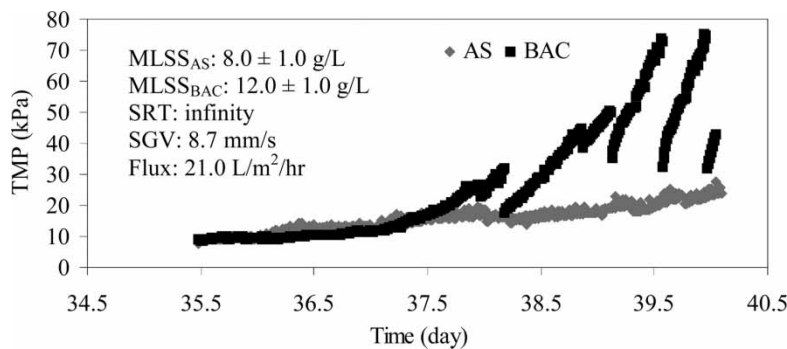


Figure 11. Performance comparison of MBR(AS) and MBR(BAC) at infinite SRT.

If the major 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 biofloc it loses its impact; in other words, only fresh PAC particles have scouring effects. We plan to report on scouring studies in a later paper.

The other role of PAC is adsorptive. The aged BAC would lose this capability due to pore blocking by

- a. fine foulants,
- b. products of microbial biodegradation and
- c. dead microbial cells (41).

As a result the BAC would not be able to pick up TOC or fine colloids that could foul the membrane. Instead acting as a scouring agent and adsorbent, the aging BAC would add to the fouling potential by

- i. increasing the MLSS concentration in contact with the membrane and
- ii. restricting the movement of the fiber bundle (due to raised suspension viscosity).

This may explain the poor performance of the MBR(BAC) at “infinite” SRT. From Figs. 10 and 11, it is evident that aging BAC decreases the sustainable flux of the MLSS and that steady replacement of saturated BAC is important to extend the MBR operation before cleaning. Further work is required to identify if there is an optimum strategy in terms of PAC load, SRT, imposed flux and aeration conditions.

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). 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 in qualitative agreement with the long term runs, but could only be regarded as very approximate predictors of long term performance. The long-term runs in the 20 L MBRs with submerged hollow

fibers operated at SRT 30 days and a flux of 21 L/m²hr showed a much slower fouling rate (TMP rise) for the MBR with 5 g/L PAC (in 9.5 g/L biomass) compared with the MBR without PAC (also 9.5 g/L biomass). However when these MBRs were operated at “infinite” SRT (no wastage and no PAC replacement) the MBR without PAC performed better than the MBR with the “aged” biological activated carbon. Therefore to gain the benefit of the MBR (BAC) it is necessary to have regular replenishment of aged BAC with fresh PAC. This suggests that the primary role of the PAC is to provide adsorptive removal of TOC and fine colloids rather than providing “scouring” control of fouling. However this other role cannot be discounted.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the support of the Agency of Science, Technology and Research of Singapore (A*STAR) for funding the Temasek Professor Program in Membrane Technology for Sustainable Water.

REFERENCES

1. Lee, J., Ahn, W.Y., and Lee, C.H. (2001) Comparison of the filtration characteristic between attached and suspended growth microorganisms in submerged membrane bioreactor. *Water Res.*, 35: 2435–2445.
2. Ognier, S., Wisniewski, C., and Grasmick, A. (2002) Membrane fouling during constant flux filtration in membrane bioreactor. *Membrane Technology.*, 2002: 6–10.
3. Choo, K.H. and Lee, C.H. (1998) Hydrodynamic behavior of anaerobic biosolids during crossflow filtration in the membrane anaerobic bioreactor. *Water Res.*, 32: 3387–3397.
4. Tardieu, E., Grasmick, A., Geaugey, V., and Manem, J. (1999) Influence of hydrodynamics on fouling velocity in a recirculated MBR for wastewater treatment. *J. Membr. Sci.*, 156: 131–140.
5. Choi, H., Zhang, K., Dionysiou, D.D., Oerther, D.B., and Sorial, G.A. (2005) Effect of permeate flux and tangential flow on membrane fouling for wastewater treatment. *Separation and Purification Technology.*, 45: 68–78.
6. Liu, R., Huang, X., Chen, L., Wen, X., and Qian, Y. (2005) Operational performance of a submerged membrane bioreactor for reclamation of bath wastewater. *Process Biochemistry.*, 40: 125–130.
7. Kishino, H., Ishida, H., Iwabu, H., and Nkano, I. (1996) Domestic wastewater reuse using a submerge membrane bioreactor. *Desalination.*, 106: 115–119.
8. Ueda, T., Hata, K., and Kikuoka, Y. (1996) Treatment of domestic sewage from rural settlements by a membrane bioreactor. *Water Sci. Tech.*, 34: 189–196.
9. Cicek, N., Winnen, H., Suidan, M.T., Wrenn, B.E., Urbain, V., and Manem, J. (1997) Effectiveness of the membrane bioreactor in the biodegradation of high molecular weight compounds. *Water Res.*, 32: 1553–1564.
10. Côté, P., Buisson, H., Pound, C., and Arakaki, G. (1997) Immersed membrane activated sludge for the reuse of municipal wastewater. *Desalination.*, 113: 189–196.

11. Gui, P., Huang, X., Chen, Y., and Qian, Y. (2002) Effect of operational parameters on sludge accumulation on membrane surfaces in a submerged membrane bioreactor. *Desalination*, 151: 185–194.
12. Hasar, H., Kinaci, C., and Unlu, A. (2002) Viability of microbial mass in a submerged membrane bioreactor. *Desalination*, 150: 263–268.
13. Innocenti, L., Bolzonella, D., Pavan, P., and Cecchi, F. (2002) Effect of sludge age on the performance of a membrane bioreactor: influence on nutrient and metals removal. *Desalination*, 146: 467–474.
14. Rosenberger, S., Krüger, U., Witzig, R., Manz, W., Szewzyk, U., and Kraume, M. (2002) Performance of a bioreactor with submerged membranes for aerobic treatment of municipal waste water. *Water Res.*, 36: 413–420.
15. Cabassud, C., Laborie, S., and Laine, J.M. (1997) How slug flow can improve ultrafiltration flux in organic hollow fibers. *J. Membr. Sci.*, 128: 93–101.
16. Laborie, S., Cabassud, C., Bourlier, L.D., and Laine, J.M. (1998) Fouling control by air sparging inside hollow fiber membranes—effect on energy consumption. *Desalination*, 118: 189–196.
17. Laborie, S., Cabassud, C., Bourlier, L.D., and Laine, J.M. (1999) Characterisation of gas-liquid two-phase flow inside capillaries. *Chemical Engineering Science*, 54: 5723–5735.
18. Cabassud, C., Laborie, S., Bourlier, L.D., and Laine, J.M. (2001) Air sparging in ultrafiltration hollow fibers: relationship between flux enhancement, cake characteristics and hydrodynamic Parameters. *J. Membr. Sci.*, 181: 57–69.
19. Mahmud, H., Kumar, A., Narbaitz, R.M., and Matsuura, T. (2004) The air-phase mass transfer resistance in the lumen of a hollow fiber at low air flow. *Chemical Engineering Journal*, 97: 69–75.
20. Maartens, A., Jacobs, E.P., and Swart, P. (2002) UF of pulp and paper effluent: membrane fouling—prevention and cleaning. *J. Membr. Sci.*, 209: 81–92.
21. Lim, A.L. and Bai, R. (2003) Membrane fouling and cleaning in microfiltration of activated sludge wastewater. *J. Membr. Sci.*, 216: 279–290.
22. Chua, H.C., Arnot, T.C., and Howell, J.A. (2002) Controlling fouling in membrane bioreactor with a variable throughput. *Desalination*, 149: 225–229.
23. Chen, V., Fane, A.G., Madaeni, S., and Wenten, I.G. (1997) Particle deposition during membrane filtration of colloids: Between concentration polarization and cake formation. *J. Membr. Sci.*, 125: 109–122.
24. Defrance, L., Jaffrin, M.Y., Gupta, B., Paullier, P., and Geougey, V. (2000) Contribution of various constituents of activated sludge to membrane bioreactor fouling. *Bioresource Technology*, 73: 105–112.
25. Cho, B.D. and Fane, A.G. (2002) Fouling transients in nominally sub-critical flux operation of a membrane bioreactor. *J. Membr. Sci.*, 209: 391–403.
26. Bouhabila, E.H., Aim, R.B., and Buisson, H. (1998) Microfiltration of activated sludge using submerged membrane with air bubbling (application to wastewater treatment). *Desalination*, 118: 315–322.
27. Srijaroonrate, P., Julien, E., and Aurelle, Y. (1999) Unstable secondary oil/water emulsion treatment using ultrafiltration: fouling control by backflushing. *J. Membr. Sci.*, 159: 11–20.
28. Bouhabila, E.H., Aim, R.B., and Buisson, H. (2001) Fouling characterisation in membrane bioreactors. *Separation Purification Technology*, 22–23: 123–132.
29. Pirbazari, M., Ravindran, V., Badriyha, B.N., and Kim, S.H. (1996) Hybrid membrane filtration process for leachate treatment. *Water Res.*, 30: 2691–2706.

30. Seo, G.T., Suzuki, Y., and Ohgaki, S. (1996) Biological powdered activated carbon (BPAC) microfiltration for wastewater reclamation and reuse. *Desalination*, 106: 39–45.
31. Li, Y.Z., He, Y.L., Liu, Y.H., Yang, S.C., and Zhang, G.J. (2005) Comparison of the filtration characteristics between biological powdered activated carbon sludge and activated sludge in submerged membrane bioreactors. *Desalination*, 174: 305–314.
32. Park, S.J., Oh, J.W., and Yoon, T.I. (2003) The role of powdered zeolite and activated carbon carriers on nitrification in activated sludge with inhibitory materials. *Process Biochemistry*, 39: 211–219.
33. Cecen, F., Erdinciler, A., and Kilic, E. (2003) Effect of powdered activated carbon addition on sludge dewaterability and substrate removal in landfill leachate treatment. *Advances in Environmental Research*, 7: 707–713.
34. Orshansky, F. and Narkis, N. (1997) Characteristic of organics removal by PACT simultaneous adsorption and biodegradation. *Water Res.*, 31: 391–398.
35. Dalmacija, B., Karlovic, E., Tamas, Z., and Miskovic, D. (1995) Purification of high-salinity wastewater by activated sludge process. *Water Res.*, 30: 295–298.
36. Pala, A. and Tokat, E. (2002) Color removal from cotton textile industry wastewater in an activated sludge system with various additives. *Water Res.*, 36: 2920–2925.
37. Lim, P.E., Ong, S.A., and Seng, C.E. (2002) Simultaneous adsorption and biodegradation processes in sequencing batch reactor (SBR) for treating copper and cadmium-containing wastewater. *Water Res.*, 36: 667–675.
38. Lin, C.K., Tsai, T.Y., Liu, J.C., and Chen, M.C. (2000) Enhanced biodegradation of petrochemical wastewater using ozonation and BAC advanced treatment system. *Water Res.*, 35: 699–704.
39. Scholz, M. and Martin, R.J. (1997) Ecological equilibrium on biological activated carbon. *Water Res.*, 31: 2959–2968.
40. Mochizuki, K. and Takeuchi, Y. (1998) The effect of some inhibitory components on biological activated carbon processes. *Water Res.*, 33: 2609–2616.
41. Sirotkin, A.S., Koshkina, L.Y., and Ippolitov, K.G. (2001) The BAC-process for treatment of waste water containing non-ionogenic synthetic surfactants. *Water Res.*, 35: 3265–3271.
42. Zhao, X., Hickey, R.F., and Voice, T.C. (1999) Long-term evaluation of adsorption capacity in a biological activated carbon fluidized bed reactor system. *Water Res.*, 33: 2983–2991.
43. Kim, J.S., Lee, C.H., and Chun, H.D. (1998) Comparison of ultrafiltration characteristics between activated sludge and BAC sludge. *Water Res.*, 32: 3443–3451.
44. Boerlage, Siobhan, F.E., Kennedy, M.D., Aniye, M.P., Abogrean, E., Tarawneh, Z.S., and Schippers, J.C. (2003) The MFI-UF as a water quality test and monitor. *J. Membr. Sci.*, 211: 271–289.
45. Defrance, L. and Jaffrin, M.Y. (1999) Reversibility of fouling formed in activated sludge filtration. *J. Membr. Sci.*, 157: 73–84.
46. Huang, X., Gui, P., and Qian, Y. (2001) Effect of sludge retention time on microbial behaviour in a submerged membrane bioreactor. *Process Biochemistry*, 36: 1001–1006.
47. Fane, A.G. (1984) Ultrafiltration of suspensions. *J. Membr. Sci.*, 20: 249–259.