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Mechanisms of Fouling Control in Membrane Bioreactors by the Addition of Powdered Activated Carbon

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This paper describes the experiments and observations that examine the mechanisms by which the addition of powdered activated carbon (PAC), in the form of biologically activated carbon (BAC), improves the filtration performance of a submerged membrane bioreactor (MBR). The membrane performance was observed to increase significantly with steady state PAC concentration. It is necessary to steadily replenish the PAC, to match that which is lost in sludge wastage. The enhancement mechanisms identified are, first, the role of PAC as an adsorbent of organics and planktonic bacteria, second, the effect of PAC as a scouring agent that limits foulant deposition, and third, the effect of the combined adsorption and biodegradation of BAC on the foulant components. The effectiveness of each mechanism in decreasing the fouling rate has been carefully evaluated. All three mechanisms play a role and the most significant appears to be the combined adsorption and biodegradation effect. The properties and filtration characteristics of activated sludge, with and without BAC have been measured and compared in both short-term tests and long-term continuous operation runs. The results of the short-term (cross flow mode) tests are in qualitative agreement with long-term performance.

Keywords adsorption and biodegradation; biologically activated carbon; membrane bioreactor; powdered activated carbon; scouring

INTRODUCTION

Membrane bioreactors (MBRs) are recognized as an effective alternative to the conventional activated sludge treatment process. MBRs are able to produce higher effluent quality, less excess sludge, and have a smaller footprint. However, membrane fouling is still an issue in MBRs. The fouling consists of reversible and irreversible components which are caused by cake formation (1,2) and pore blocking or restriction (1,3–6) respectively. Reported methods used to improve fouling control, include

i. intermittent filtration (7–9) and backwashing (10–14),
ii. fixing the flux below the "sustainable" flux (15–17),
iii. good hydrodynamic design to prevent cake accumulation on the membrane surface (3,7,8,10,11,18–21),
iv. physical and chemical cleaning (22,23),
v. sidestream operation with two-phase flow applied to the lumen of the hollow fiber module (24–28), and
vi. hybrid MBRs with porous and flexible suspended carriers (29).

In addition, the modification of the characteristics of the mixed liquor suspension by additives, such as powdered activated carbon (PAC) in the MBR to improve removal efficiency and fouling control has attracted attention (30–42). In our previous work we reported on the beneficial use of PAC for MBR fouling control (38). However, it was observed that steady PAC replenishment was required, and that without fresh PAC the fouling was worse than for an MBR without PAC. In this study we examine the mechanisms by which PAC may improve fouling control of MBRs. The mechanisms may involve foulant removal through adsorption, hydrodynamic scouring effects on fouling, and modification of the MLSS floc characteristics and activity.

Conventional activated sludge (AS) wastewater treatment processes with the addition of PAC have been found able to treat wastewater containing

i. inhibitory materials (43)
ii. landfill leachate (44)
iii. phenol or aniline (45)
iv. high salinity oil-field brine (46) and
v. color from the textile industry (47) and industrial wastewater (48) effectively.

This may be due to the stable microbial film that tends to form on the PAC surface to transform it into "biologically activated carbon" (BAC) sludge (33,49) that would enhance the bioactivity in pollutants removal (31,33,47). The natural ecosystem of BAC could lead to simultaneous adsorption and biodegradation processes rather than

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the biological process alone (30,33,36,37,44,45). The microorganisms in the biofilm may be able to biodegrade the pollutants previously adsorbed by the PAC. Thus, a long-term operation could see a potential advantage of BAC in partly bioregenerating the saturated BAC (32,50) by the immobilized (46,48), acclimatized, and succession (51) bacteria in the biofilm ecosystem. Other claimed advantages of BAC include

i. increased the efficiency of substrate removal (48)
ii. improved activated sludge filterability (30,32–33,35,49)
iii. reducing the adverse effects of heavy metal ions on biomass through adsorption (52) and better performance in withstanding loading shock (35,42).

A few studies have reported that the addition of PAC can improve fouling control of the MBR system. Several researchers agree that the improved performance of the MBR with the addition of PAC (referred to hereafter as the MBR (BAC)) was due to the adsorption effect that reduces extracellular polymeric substances (EPSs) in the floc (49) and the bulk liquid (32,34) and other fine foulants such as TOC (30,31,36), DOC (37), fine colloids (32), soluble metabolic products (SMPs) (33), refractory organic matter (31), COD (35,42) and trace organics (53) in the supernatant. Seo et al., (36) 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 (30,32,33,49) 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 noted that the BAC could act as a "precoat" permeable layer on the membrane surface to prevent membrane pore restriction (30–33). Again, this requires the flux to be fixed above the critical flux so that deposition of BAC occurs, and this itself could cause fouling issues. However, under appropriate conditions, 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 (30,32,35,53). High loadings of PAC may be needed if PAC is used as a "scouring agent" and this needs to be optimized.

In summary, there are several reported functions of PAC in enhancing the performance in MBRs but it is still ambiguous as to which mechanism plays the primary role in fouling control. Therefore, the objective of our study has been to investigate the possible mechanisms involved in PAC 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 the 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.

EXPERIMENTAL

Materials

PAC (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 Fig. 1, was measured by a Malvern Mastersizer particle size analyzer. The BET surface area of the fresh PAC was about 488 m^2/g. Powdered hollow glass beads (GB) used in scouring tests were obtained from Dantec Dynamic with three different sizes of 5, 20, and 50 μm respectively.

The filtration characteristics of the BAC were measured in short-term tests in a dead-end filtration cell fitted with

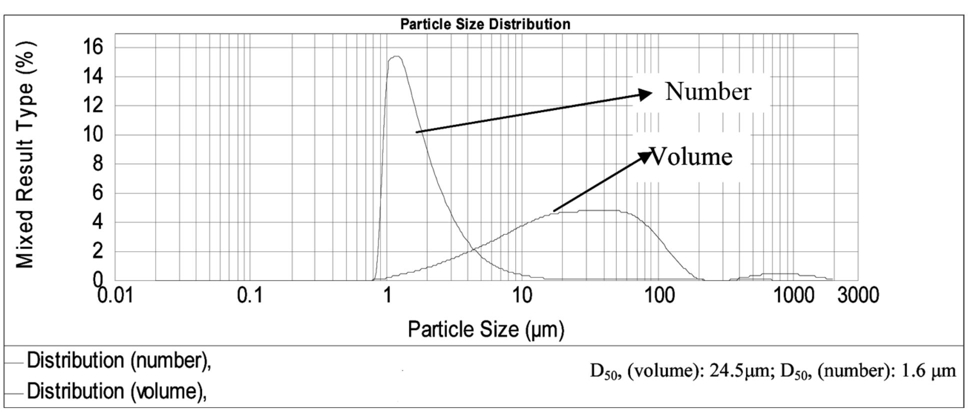

FIG. 1. The particle size distributions of PAC used in MBRs.

Millipore ultrafiltration Polyethersulfone membranes with a molecular weight cut-off of 50,000Da. For the long-term submerged MBR trials and some short-term "sustainable" flux tests, hollow fiber microfiltration Polyacrylonitrile membranes (nominal pore size: 0.5 μm) from China Blue Star Membrane Technology Co., Ltd were used for filtration comparison.

Operation of MBRs

Four 2 L (batch-continuous) MBRs were set-up and operated with 0, 1, 3, and 5 g/L PAC. The activated sludge used in the MBRs was sampled 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 12 months. 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). 500 mL from each 2 L MBR was filtered out daily and replaced by 500 mL of concentrated feed to provide an average feed of 370 ± 10.0 mg/L TOC concentration to the MBRs. The values of SRT, HRT, and superficial gas velocity (SGV) for the four 2 L MBRs were 10 days, 2.85 days, and 8.0 mm/s respectively. After the four 2 L MBRs had been cultivating for 108 days, four membrane modules were submerged into each MBR for filtration performance comparison. After approximately eight days of filtration tests, the modules were taken out and washed with 100 mL of Milli-Q water to detach the sludge accumulated on the membrane surface. The collected sludge from the membrane surface was characterized. The parameters involved in the characterization were

i. the particle size distribution
ii. suspended solids (SS)
iii. extracellular polymeric substances (EPSs) and
iv. TOC concentrations (mg/100 mL).

Cultivation of the Pure Culture Bacteria

Adsorption experiments (see 2.4) were done on pure culture bacteria isolated from an MBR. The use of pure culture facilitated observation. Cells were picked up from the R2A agar plates and inoculated in screwcapped test tubes with a 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 room temperature of about 25°C for 5 days.

Adsorption Experiments

Adsorption experiments were done to assess the efficacy of PAC in adsorbing planktonik bacteria. In addition, adsorption isotherm studies were done to evaluate the PAC adsorption capabilities on the TOC and the polysaccharide 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 TOC adsorption tests were for durations of 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.

Membrane Cell and "Sustainable" Flux 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 same reactors. 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) = 1.26×10^{-3} m^2). In addition, short-term "flux stepping" tests (see below) were carried out on the 4×2 L MBRs equipped with a data logging system. 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).

Short-term tests in the 2 L MBRs measured the "sustainable" flux (with air-bubbled crossflow). In our tests 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 and identified by flux-stepping and measuring TMP. 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 2 L MBR experiments respectively. The 2 L MBRs set-up was also used for the long-term filtration performance comparison for the MBRs with different PAC concentrations.

Resistances were estimated from the Darcy equation,

$$\text{Resistance,}\ R = \frac{\Delta P}{\mu J} \tag{1}$$

Where J is the flux, ΔP is the transmembrane pressure (TMP), and μ is the 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 reversible cake resistance, R_c, was obtained from Eq. (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,

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

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

Membrane Autopsy

The fouled membranes from the 2 L MBRs were cleaned with 0.5 L 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 Eq. (4).

$$(C) = \frac{C_1 \times 0.5\,L\ (amount\ of\ water\ used\ to\ \text{det}ach\ the\ sludge\ on\ the\ membrane\ surface)}{2\,L\ (Working\ volume\ of\ MBR)} \quad (4)$$

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

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 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 plantonik 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 (54) and Bradford reagent for protein with bovine serum albumin (BSA) as standard (55) respectively.

RESULTS AND DISCUSSION

Fouling Rates for MBR (AS) and MBR (BAC)

The four small (2 L) MBRs with different PAC concentrations (0, 1, 3, 5 g/L) were operated at SRT of 10 days. 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 characterize the membrane fouling intensity and act as an indicator for the filtration performance of the MBRs. Figure 2 plots TMP history and shows that the MBR with 5 g/L PAC and without PAC had the best and worse performances in terms of fouling control respectively. Figure 2a shows data for 0 g/L PAC. At a flux of 21.0 L/m^2/hr, an immediate TMP "jump" was observed and reducing the flux from 21.0 to 10.5 L/m^2/hr helped to delay the "jump" but the effect was not significant. To avoid the immediate "jump," the MBR was run at a low flux of 5.3 L/m^2/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/m^2/hr instead of 21.0 L/m^2/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/m^2/hr could 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 2 (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 1 g/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 709.7 kPa/day, 8.6 kPa/day, 6.7 kPa/day, and 5.1 kpa/day for the MBRs with 0, 1, 3, and 5 g/L of PAC respectively for the

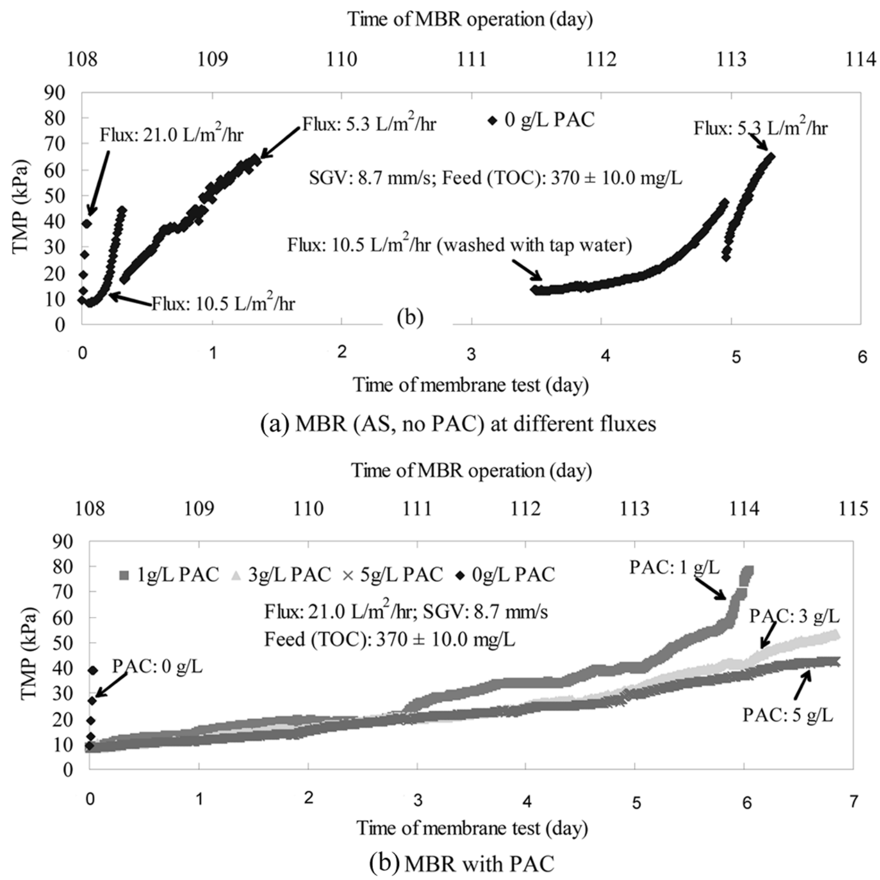

FIG. 2. Performance comparison of (a) MBR (AS) with different fluxes and (b) MBR with different PAC concentrations at SRT 10d.

7 days of operation as shown in Table 1. It should be noted that these TMP rises would be specific to the conditions chosen (SRT, the membrane/module characteristics, the aeration rate, the 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).

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 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). PAC appears to encourage biomass development, possibly by providing a sink for the substrate that would otherwise pass out of the MBR. This issue is

TABLE 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).

TABLE 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.

discussed in more detail in sections titled "PAC as an adsorbent" and "PAC as scouring agent" 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 are reported to have other advantages compared to the suspended growth biomass such as

i. more capability of withstanding shock loading (35) and
ii. a more effective substrate biodegradation capacity (48).

Effect of PAC on Planktonic Bacteria

PAC could also play a role in controlling the population 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. This is because single cells tend to have a much lower critical flux than the larger biofloc. 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.

To check the effectiveness of fresh PAC in reducing the planktonic bacteria in the supernatant, two different methods were used. The methods are summarized below. First, after 24 hours of the 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 Fig. 3. The second method involved the 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 Fig. 3 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 Fig. 3 reveal that the PAC was able to reduce significantly the planktonic bacteria concentration in the supernatant. The best result was for the highest loading of PAC. This was presumably caused by the adsorptive effect of the PAC, which may also involve the 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 (available on request) support this view by clearly showing the attachment of bacteria on the PAC surface. Live/dead staining also showed 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 Fig. 3). 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 background of PAC fines not readily removed by settling or even centrifugation. However, the results 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 biofouling rate.

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 and is derived from the "critical" flux concept and in this study is taken as the highest flux for which dTMP/dt is negligible. Figure 4 shows that the MBR (AS) without PAC had a "sustainable" flux of about 10.4 $L/m^2/hr$. For the MBR (BAC) with 1 g/L of PAC, a significant increase in the "sustainable" flux to about 31.0 $L/m^2/hr$ was obtained. The "sustainable" fluxes for the MBR (BAC)s with 3 and 5 g/L of PAC were about 36.0 $L/m^2/hr$ and 41.0 $L/m^2/hr$ respectively. It is interesting to note that the results from

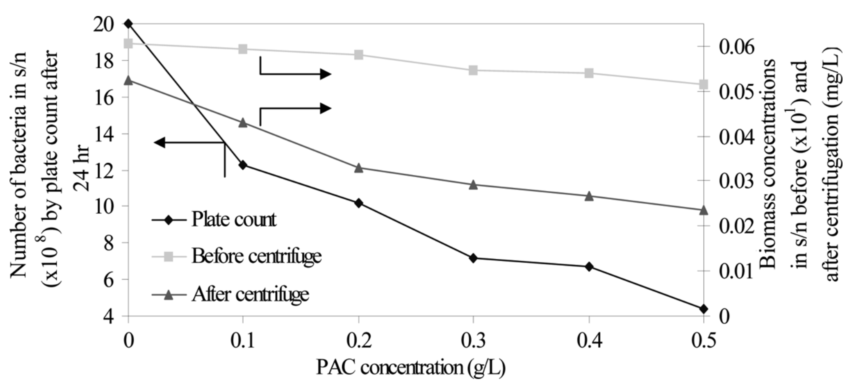

FIG. 3. Effect of different PAC concentrations in suspended bacteria reduction.

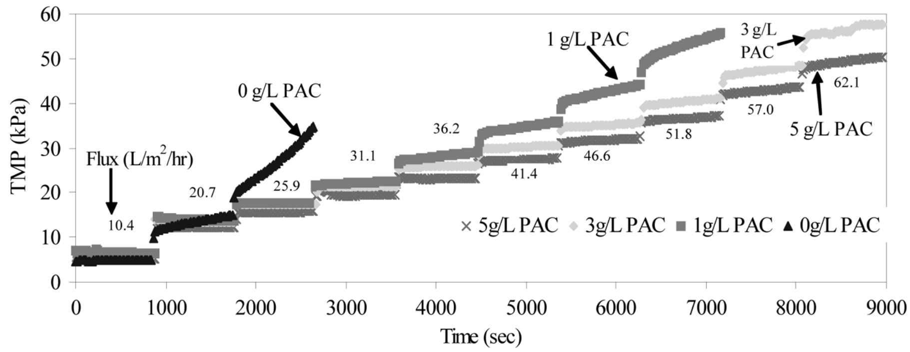

FIG. 4. Sustainable flux tests for MLSS with different PAC concentrations, measured in the individual MBRs.

the short-term flux-stepping tests to identify flux mirror qualitatively the results of the long-term trials (Fig. 2).

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 2 L MBRs. Table 3 and Fig. 5 show that all the MBR (BAC)s with PAC had better results compared to the MBR (AS) in terms of specific cake resistance (SCR), flux decline profile, and irreversible fouling resistance. 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 other researchers (49). Figure 6 includes the D_{50} data (both number and volume based), and it is evident that the BAC floc were smaller. This decrease in size relate to the presence of small PAC particles in the BAC. However, 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 3) and flux decline profiles (Fig. 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 Fig. 6. 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 Fig. 7. TOC includes not only unused substrate but also extracellular polymeric substances (EPSs), such as polysaccharides and proteins, believed to foul membranes (56). The TOC would foul the membrane by plugging or blocking the pores and also cause the deposited cake formed on the membrane surface in the SCR tests to be less porous by filling the interstitial gaps between the flocs. This could account for the worse filtration performance at 0 g/L PAC which had the highest TOC level.

From the results of the short-term tests it is probable that both the floc size and the 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 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 3, resistance increment (%)).

TABLE 3
Irreversible fouling (% increase in R_m) and specific cake resistance for MBRs with different PAC loadings

PAC (g/L)	7 day tests on 2 L 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): $[(R_{fouled}/R_{initial})-1] \times 100$.

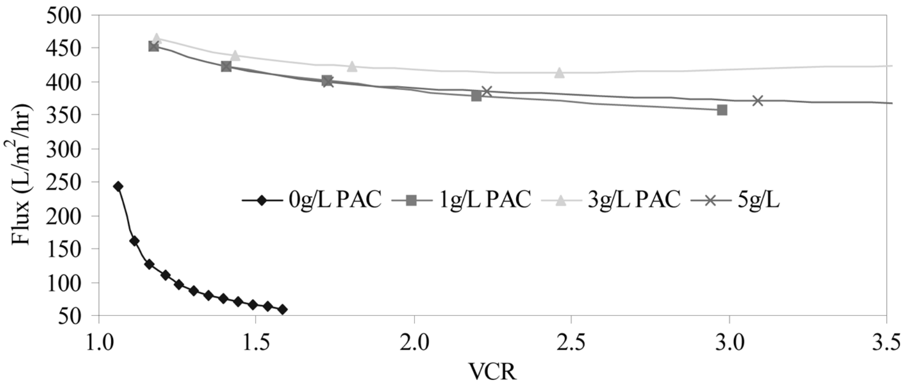

FIG. 5. Flux decline profiles for fixed pressure (100 kPa) and unstirred dead-end filtration, using Millipore UF Polyethersulfone membrane.

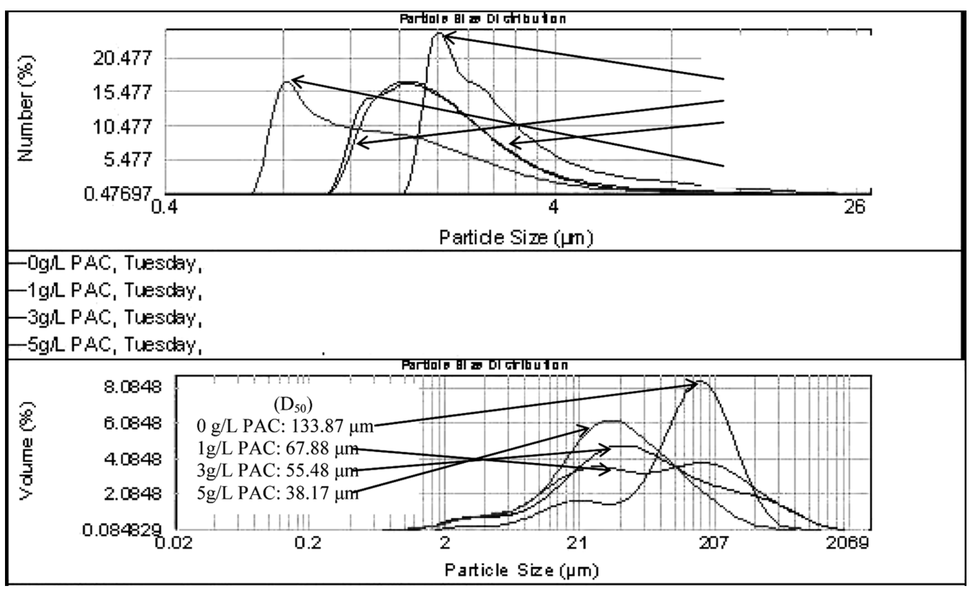

FIG. 6. Size distribution of flocs from 2 L MBRs in mixed liquor with different PAC concentrations.

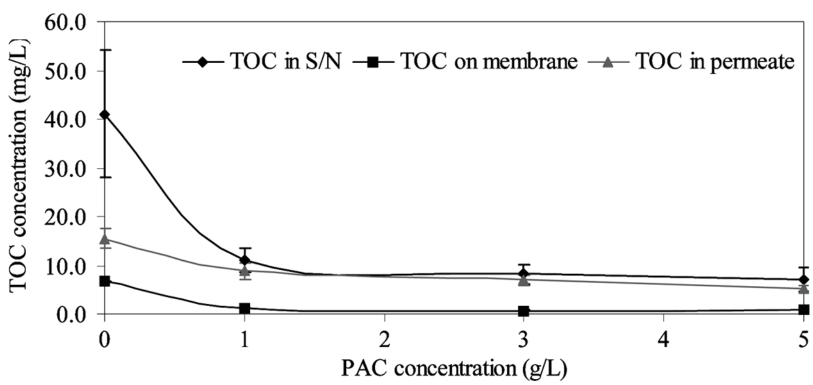

FIG. 7. Effect of different PAC concentrations on the TOC in the supernatant and permeate and on the membrane surface.

Similar results were obtained in separate longer-term MBR tests. These tests were based on the 2 L MBRs. Four clean membranes were tested for their intrinsic resistance with Milli-Q water before submerging into the 2 L MBRs with different PAC concentrations. After seven days of filtration at a flux of 21.0 $L/m^2/hr$, except for MBR (AS) at fluxes of 21.0, 10.5, and 5.3 $L/m^2/hr$, 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 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-term test in measuring specific cake resistance (Table 3) and flux decline profiles (Fig. 5) showed that 3 g/L BAC was better than that of 5 g/L BAC. This was due to

i. the bigger particle size (Fig. 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 being under relatively high flux (>80 $L/m^2/hr$), unstirred, dead-end and cake formation filtration conditions. These are not typical operating conditions for MBRs.

However, in suspension conditions (stirred, aerated, or crossflow), which is typical in operating conditions of MBRs, both the short-term (Fig. 4) and long-term (Fig. 2) comparison tests showed that the 5 g/L BAC performed better than the 3 g/L BAC. 5 g/L BAC also performed better in terms of irreversible fouling control. This suggests that the comparison results obtained under suspension conditions are more representative of the real performance of the MBR.

Potential Flux-Enhancement Mechanisms

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

i. PAC helps to reduce the fine foulants reaching the membrane surface through adsorption;
ii. PAC has a scouring effect that limits the fine foulants accumulation on the membrane surface;
iii. PAC forms BAC which is equipped with simultaneous adsorption and biodegradation effects and is more effective than AS floc in decomposing the high molecular weight organics which tend to foul the membrane;
iv. PAC can act as a protective layer on the membrane surface.

Mechanism (iv) 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. MBRs are operated below the critical flux of the biofloc. This suggests that the protective layer formation would not be the primary role for PAC in irreversible fouling control. Therefore, the other three mechanisms have been considered to be more relevant and examined in detail.

PAC as an Adsorbent

PAC is recognized as a good adsorbent for reducing contaminants in wastewater. Section titled "Effect of PAC on Planktonic Bacterin" described the planktonic cell reduction by PAC which is an example of the good adsorptive effect of PAC. Figure 7 shows that the 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 titled "Effect of PAC on MLSS") but also due to the adsorptive effect of fresh the PAC. This explanation is supported by the results for the TOC concentrations in the permeate which where 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 (Fig. 7). 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, chosen as characteristic adsorbates because they have been identified as representing potentially the main components that foul the membranes in MBRs. The batch adsorption results fitted the Freundlich equation well, having $r^2 = 0.89$ for TOC (3 days) and 0.86 for the polysaccharide data. The capacity constants, K_f, were 0.49 (TOC) and 0.16 (polysaccharide) and the intensity constants, $1/n$, were 0.36 and 0.6 respectively. These discussed confirmed that PAC could decrease the fine foulants through adsorption.

Further evidence of the adsorption phenomena with PAC and mixed liquor is shown in Fig. 8 which 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 ($\leq$30 days).

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

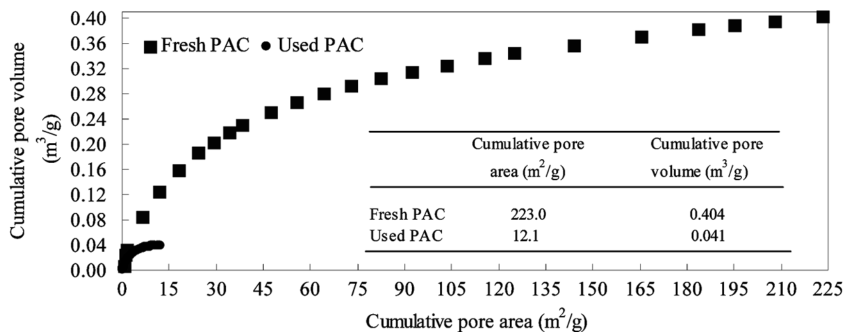

FIG. 8. Pore volume comparison of fresh and used PAC.

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. 0.5 L 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 0.5 L Milli-Q water were analyzed (see section titled "Membrane Cell and 'Sustainable' Flux Filtration Tests") in terms of sludge concentration and particle size (see Table 4) and EPSs concentration (see Fig. 9). It was found that the floc sizes in the mixed liquor and formed on the membrane surface (Table 4) 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 that that of MBR (AS) (Table 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 4). The least was for the MBR (BAC) with 5 g/L of PAC (1.2%) and the most was for the MBR (AS) (5.0%). The EPSs (total EPS = polysaccharide + protein) concentrations accumulated on the membrane surface had similar trends with lower EPSs concentrations for the MBR (BAC)s compared with the MBR (AS) as shown in Fig. 9. 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 7 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.

In support of the scouring mechanism, it is useful to report separate experiments on a fouled membrane from the larger scale bioreactor (continuously run lab-scale MBR (20 L)). The fouled membrane from the bioreactor (20 L) was shifted to a smaller vessel (3.5 L) containing only Milli-Q water and coarse aeration. The TMP data are shown in Fig. 10, where t = 0.5 days represents the start of the test. Initially, the TMP dropped from approximately 49 to 41 kPa, 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. 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 vessel. A steady drop of TMP was observed from about 42 to 40 kPa over two days of filtration. This suggests that the GB helped to detach some of the reversible fine foulants back to the Milli-Q water and 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 (57) that supramicron particles reduce the rate of fouling by enhancing back-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 titled "Effect of PAC on Planktonic Bacteria"]) and particulate scouring (foulant removal or increasing back transport). Furthermore, as shown in section titled "Effect of PAC on MLSS", the PAC appears

TABLE 4
Cake concentration (g/L) attached on membrane surface and D_{50} (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)

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

$C_1\ (g/L) = Sludge\ (on\ membrane\ surface)\ in\ 0.5\,L\ (amount\ of\ clean\ water\ used\ to\ \mathrm{det}ach\ sludge)$

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

to enhance MLSS development and this could be linked to substrate adsorption. This observation tends to favor an "adsorption" role for PAC, although fresh PAC may have "scouring" properties until incorporated into a 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 3.5 L vessel was operated as an MBR with no sludge wastage for 20 days at a flux of 7.9 $L/m^2/hr$. As shown in Fig. 11, 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 the 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 ($D_{50} = 25$ 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 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.

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 2 L MBRs which had been cultivated for 10.0 days

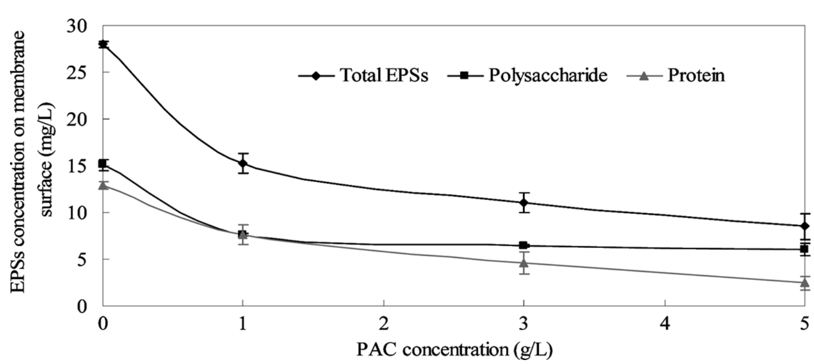

FIG. 9. Effect of different PAC dosages on the attachment of EPSs on the membrane surfaces.

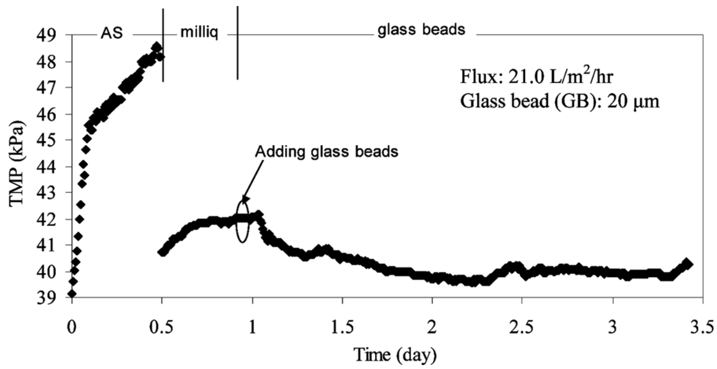

FIG. 10. Effect of particle scouring on the TMP reduction.

(no sludge wastage) at a flux of $16.0\,L/m^2/hr$ instead of $7.9\,L/m^2/hr$ (Fig. 12). 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 2 L bioreactor (Figure 12). This experiment had a higher flux ($16.0\,L/m^2/hr$) 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 Fig. 12. The dTMP/dt changed from 95.8 to 116.6 kPa/day which means that the GB had accelerated the fouling rate rather than reducing it as reported in Fig. 11. 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, a preliminary determination of the "sustainable flux" of the MBR with the "collision agent" particle would be important to prevent premature fouling.

At day 10.17, 5 g/L PAC was added into the 2 L 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 Fig. 12. Thus both experiments, operated at fluxes 7.9 and $16.0\,L/m^2/hr$ respectively, showed that PAC could help to 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.

BAC for Adsorption and Biodegradation

From the discussion above and the comparison of results shown in Figs. 11 and 12, the role of PAC in adsorbing the fine foulants was judged to be probably the primary

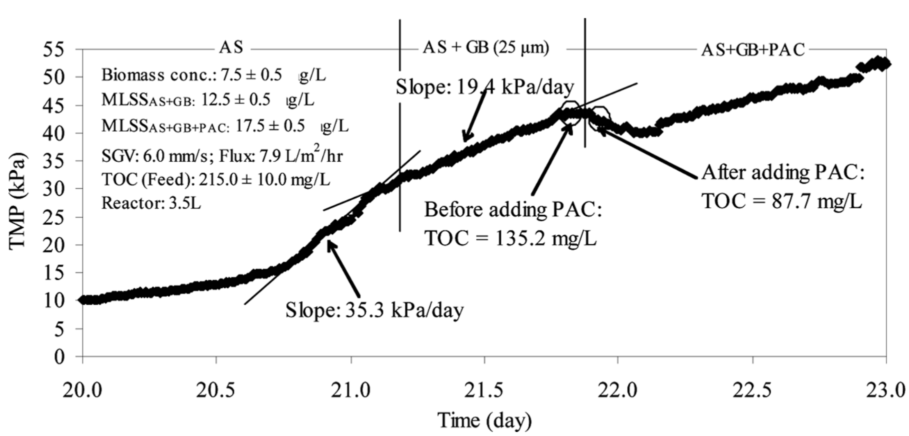

FIG. 11. Performance comparison between scouring and adsorption on TMP reduction.

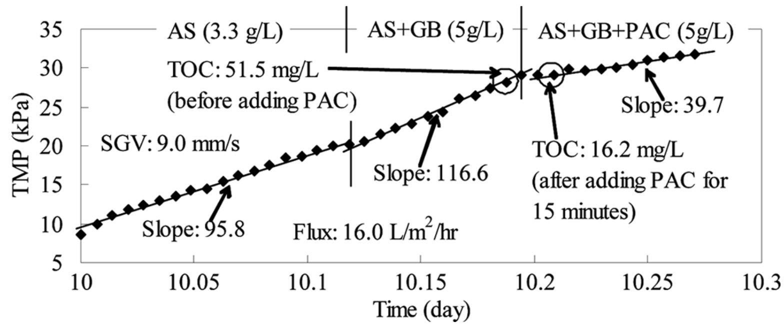

FIG. 12. Performance comparison between collision and adsorption on TMP reduction (flux: 16.0 L/m^2/hr).

mechanism in reducing fouling rate. However, the adsorption capacity of PAC was easily reduced. This is supported by Fig. 11. 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 ($\approx$8 hours) and subsequently the TMP was observed to increase again. Nonetheless, the benefit of BAC in membrane fouling control is undeniable. In addition, the enhanced MLSS observed in the MBR (BAC) (Table 2 in section titled "Effect of PAC on MLSS") 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 5 and 6. The MWt distributions were obtained by HPSEC as detailed in section 2.6. 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.

The analysis results (Table 5) revealed that more than 90% of the MWt distribution of dissolved solids in the supernatant of the MBR (AS) was above 100 k, whereas, for MBR (BAC) (i.e., AS + PAC), the dissolved organics in the supernatant were mostly below 100 k ($\approx$85%) or in the range of 10 k–100 k ($\approx$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 (100 k–1000 k; >1000 k) to lower ranges (<100 k) 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.

a. Porous PAC with good adsorption capacity could become a location for
   (i) relatively high organic substrate loading (48) and
   (ii) attachment sites for immobilized bacteria which have better substrate biodegradation abilility (48).

These conditions could enhance the decomposition of the 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 (<100 k) dissolved solids would not easily block and plug the pores ($\approx$0.5 µm) 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 µm. 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. The results from Table 6 illustrate this picture more clearly. Almost 72% of the high MWt (100 k–1000 k) TOC concentration (5.32 ± 0.35 mg/L) in the supernatant of the MBR (AS) was prevented from transmission to the permeate side (at TOC: 1.51 ± 0.23 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 (10–100 k) TOC concentration (2.14 ± 0.13 mg/L) in the supernatant of the MBR (BAC) freely passed out from the membrane into the permeate (at TOC: 1.02 ± 0.18 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

TABLE 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 < 1 k	1 k < MWt < 10 k	10 k < MWt < 100 k	100 k < MWt < 1000 k	1000 k < 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).

the conditioning biofilm compared to the high MWt organics. This difference in supernatant MWt distribution could be an important mechanism enabled by the PAC that provides a reduction in membrane fouling for the MBR (BAC).

b. Another potential benefit was suggested by other researchers (53). They noted that below a certain substrate threshold concentration there would be difficulty for further biodegradation due to the lack of enzyme induction. This limitation is overcome by the PAC which acts as a sink for accumulation of the substrate and bacteria.

In summary, the presence of PAC in AS would modify the MLSS by forming 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 (44). This would help to prolong the life-span of the BAC in membrane fouling control.

However, in our previous work (38) it was found that the MBR (BAC) cultivated at relatively longer SRTs (pseudo-infinity) tends to produce relatively high TOC concentrations in the supernatant. The aged BAC also loses its good characteristics in controlling membrane fouling, namely the adsorption of TOC and polysaccharides and single planktonik cells, scouring by collision effects and

TABLE 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 < 1 k	1 k < MWt < 10 k	10 k < MWt < 100 k	100 k < MWt < 1000 k	1000 k < 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).

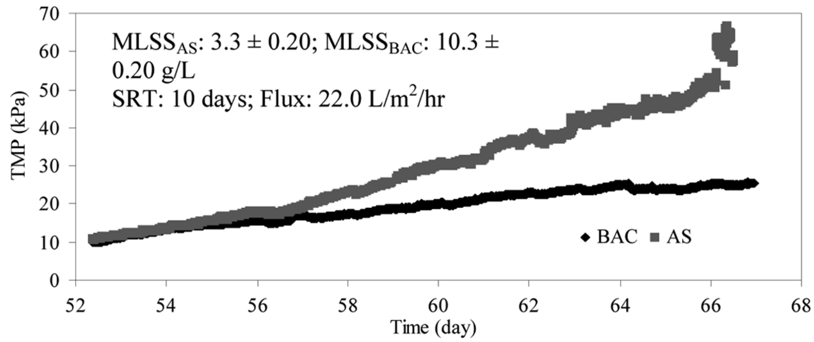

FIG. 13. Performance comparison of 20 L MBR and MBR (BAC) at SRT 10d.

simultaneous adsorption and biodegradation. Therefore, to gain the optimum benefit it is necessary to have regular replenishment of the aged BAC with fresh PAC. This implies an optimum SRT for the MBR (BAC).

Comparison of Performance of 20 L MBR (As) and MBR (BAC)

To confirm the ability of 5 g/L of PAC in reducing membrane fouling, 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/m^2 hr, 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 m^2 were submerged into each MBR and run at a fixed flux of 22.0 L/m^2/hr. The key parameter used to characterise fouling was the TMP. The results are shown in Fig. 13 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).

At day 67, SEM images of the fouled membranes from the both MBRs were scrutinized. Samples of the membrane were removed and prepared for SEM without water washing but following standard sample preparation protocols. From the images (Fig. 14), 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 check for their "sustainable" flux by flux-stepping. It was found that the "sustainable"

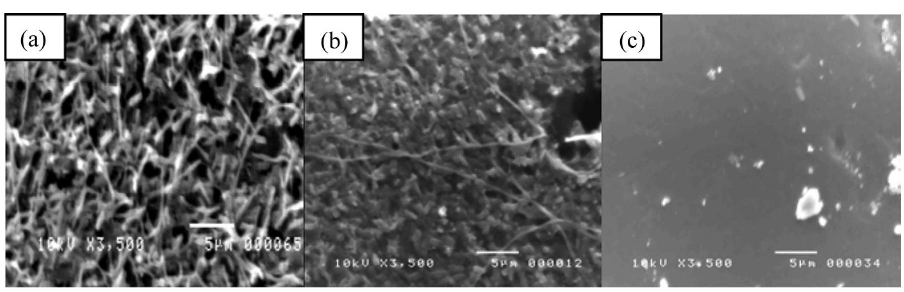

FIG. 14. SEM images of the membrane surface. (a) clean membrane, (b) membrane for the MBR (BAC), (c) membrane for the MBR (AS) at 3,500 x.

flux for the MBR (AS) was <31.8 L/m^2/hr and for the MBR (BAC) > 44.7 L/m^2/hr respectively. This further confirms that PAC has the ability to reduce the fouling rate of the MBR (BAC).

CONCLUSIONS

The efficacy of PAC in enhancing fouling control was confirmed in short and long terms tests on mixed liquor from the 2 L MBRs (0, 1, 3, and 5 g/L PAC in about 3.5 g/L of biomass) and 20 L 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 these experiments, the lowest SCR value and the best flux decline profiles were for a PAC with 3 g/L. The SCR and flux declines for 0 g/L PAC were the worst. 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.

The long-term runs in the 2 L MBRs with submerged hollow fibers operated at a flux of 21 L/m^2 hr 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 20 L 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.

The mechanisms of the PAC in controlling fouling involved

i. adsorption of foulants including single planktonik cells, TOC and polysaccharides,
ii. scouring of the membrane by collision effects and
iii. simultaneous adsorption and biodegradation to modify the amount and molecular weight distribution of the organics.

The evidence points to simultaneous adsorption and biodegradation as the primary mechanism in fouling control. To gain the optimum benefit of the MBR with PAC, it is necessary to have regular replenishment of aged BAC with fresh PAC.

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