INFLUENCE OF BACTERIAL EXTRACELLULAR POLYMERS ON THE MEMBRANE SEPARATION ACTIVATED SLUDGE PROCESS

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ABSTRACT

The objective of this study is to elucidate influence of bacterial extracellular polymers (EPS) on the performance of the submerged membrane separation activated sludge process (SMAS). Hollow fiber membrane modules were submerged in laboratory-scale reactors and the permeate was pulled out from the modules by suction pumps. Acetic acid was used as a carbon source and peptone or ammonium nitrogen was used as a nitrogen source in the substrate. The change of EPS amount both in the mixed liquor and on the membrane was measured and its influence on the permeability of the membrane was studied. EPS was accumulated in the aeration tanks and also on the membrane, which caused an increase of viscosity of the mixed liquor and an increase in the filtration resistance. Specific resistance of EPS was calculated to be of the order of 10^{16} to 10^{17} m kg^{-1}. Change of the filtration resistance was explained as a function of viscosity of the mixed liquor, which is caused by rapid attachment of the suspended EPS and rapid detachment of the attached EPS. Detachment rate of the attached EPS was calculated to be nearly constant during the operation. Copyright © 1996 IAWQ. Published by Elsevier Scince Ltd.

KEYWORDS

Bacterial extracellular polymers; clogging of membrane; filtration resistance; hollow fiber membrane; membrane separation activated sludge process; viscosity of activated sludge.

INTRODUCTION

The membrane separation activated sludge process (MAS) is an activated sludge process which incorporates a membrane separation process instead of a settling process for liquid-solid separation. Suspended solids can be completely removed from the treated water in the membrane separation process thereby producing bacteria-free water and/or virus-free water. Its maintenance is very easy because bulking phenomena cause no problems in the liquid-solid separation process and the production rate of excess sludge is very low. It has therefore been widely used in water reuse systems in buildings since 1978 in Japan (Aya, 1994). Cross-flow type membrane modules have been applied in the plant because its high cross-flow velocity can remove the accumulated sludge from the surface thereby keeping good permeability for longer periods. However, it has

not been applied to municipal wastewater treatment plants and/or small-scale on-site treatment plants partly because of its high energy consumption rate.

The submerged membrane separation activated sludge process (SMAS) is a membrane separation activated sludge system with membrane modules submerged in an aeration tank, the permeate of which is sucked out of the aeration tank by a pump (Yamamoto *et al.*,1989). It requires no circulation pumps thereby making it an energy conserving system. Arai *et al.* (1995) evaluated the effect of the hollow fiber membrane module's configuration and intermittent operation cycle on the SMAS system. However, permeate flow rate is reduced by clogging of the membrane, especially by the accumulation of bacterial cells and/or bacterial productive substances, the mechanism of which should be well understood for the application of the system to small-scale wastewater treatment plants.

The objective of this study is to elucidate the influence of bacterial extracellular polymers (EPS) which are produced by activated sludge and accumulated in an aeration tank on the performance of the membrane separation activated sludge process. The change of EPS amount both in the mixed liquor and on the membrane was measured in laboratory-scale experimental reactors and its influence on the permeability of the membrane was studied.

MATERIALS AND METHODS

Experimental set-up. Figure 1 shows a diagram of the experimental set-up used in the research. Three rectangular tanks (200mm x 200mm x 700mm) made of transparent PVC were used as aeration tanks. Membrane modules were directly submerged in the aeration tanks. Influent substrate was fed by tube pumps into the reactors at a constant flow rate. Tap water was also supplied into the reactors being controlled through level sensors set in the tanks so that the water level of the tanks was kept constant. Air was supplied to just below the membrane modules so that the rising air bubbles touched the modules, which might be effective for removing attached sludge from the membrane. Permeate from the membrane module was pulled out of the modules by tube pumps which were intermittently operated with a cycle of 10-minutes-on and 5-minutes-off.
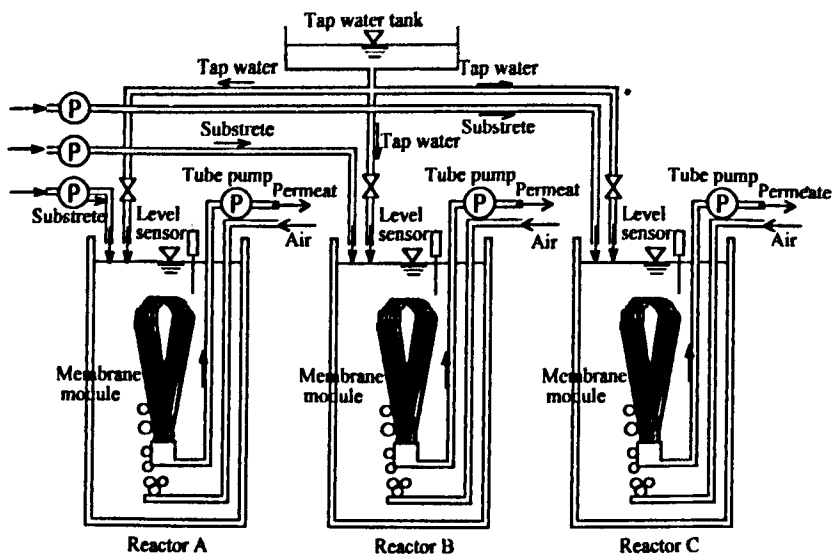

Figure 1. Schematic diagram of the experimental set-up.

Membrane module. Loop-type hollow fiber membrane modules were used for the experiment. 608 hollow fibers with length of 40cm were bundled to make a loop whose ends were fixed together in a valve socket, which provided $0.30m^2$ of total surface area of membrane. The hollow fiber membrane used was a Micro-

Filtration membrane of polyethylene with pore size of 0.1μm. The outer and inner diameter of the membrane was 270μm and 410μm respectively.

Substrates. Composition of substrates used for each reactor is shown in Table 1. Acetic acid was used as a carbon source for all reactors, while peptone was used as a nitrogen source for reactor A and ammonium chloride was used for reactor C. Reactor B is a intermediate condition between reactor A and reactor C. Loading rate of TOC and Nitrogen was set at the same value for all three reactors.

Table 1. Substrate composition (gl^{-1})

	Reactor A	Reactor B	Reactor C
Acetic Acid	13.50	22.54	31.57
Peptone	16.25	8.12	0
NH$_4$Cl	0	4.32	8.63
KH$_2$PO$_4$	1.25	1.25	1.25
FeCl$_3 \cdot$6H$_2$O	0.09	0.09	0.09
CaCl$_3$	0.18	0.18	0.18
MgSO$_4$	0.18	0.18	0.18
KCl	0.18	0.18	0.18
NaCl	0.18	0.18	0.18
NaHCO$_3$	25.2	30.4	37.8

Experimental methods. Activate sludge taken from a small-scale wastewater treatment plant was used as seeding sludge for the experiment. Permeate flow rate and suction pressure from membrane modules were monitored together with temperature in the mixed liquor. Viscosity of the mixed liquor was measured using a rotating torque cylinder, while viscosity of the permeate was measured using a glass capillary. Concentration of suspended EPS in the mixed liquor was measured approximately every 20 days. When permeate flow rate was considerably decreased, membrane modules were pulled out of the reactors for cleaning and the amount of EPS attached on the membrane was measured thereafter.

Analysis of bacterial extracellular polymers in mixed liquor. (Brown *et al.*, 1980; Sutherland *et al.*, 1971) Mixed liquor was sampled and centrifuged at 8000 rpm for 10 minutes. The sediment portion was then mixed well with distilled water and centrifuged again in the same manner. The sediment portion was mixed with the same volume of 0.1N NaOH solution and then put in a refrigerator being continuously stirred for about 40 minutes. The mixture was thereafter centrifuged at 13000 rpm for 15 minutes and the supernatant was moved to a dialysis tube after being neutralized. The tube was put in pure water for about half a day to remove low-molecular-weight organics out of the sample. The remaining TOC amount in the sample was then measured, which was regarded as high-molecular-weight bacterial extracellular polymers.

Analysis of bacterial extracellular polymers attached on the membrane. Attached sludge on the membrane was taken out of the modules physically using hands, rinsing the modules with tap water, and its weight was measured thereafter. The sampled sludge was mixed with the same volume of 0.1N NaOH solution and then subjected to the EPS analysis in the same manner as described above.

RESULTS

Flux. Figure 2 shows the change of permeate flux (permeate flow rate per unit membrane area expressed as m^3/d/m^2). The modules were pulled up and cleaned on the 27th, the 47th, the 72nd and the 142nd day. Although the flux was initially set at about 0.12 m d^{-1}, it gradually decreased to around 0.02 m d^{-1} because of the clogging of the membranes.
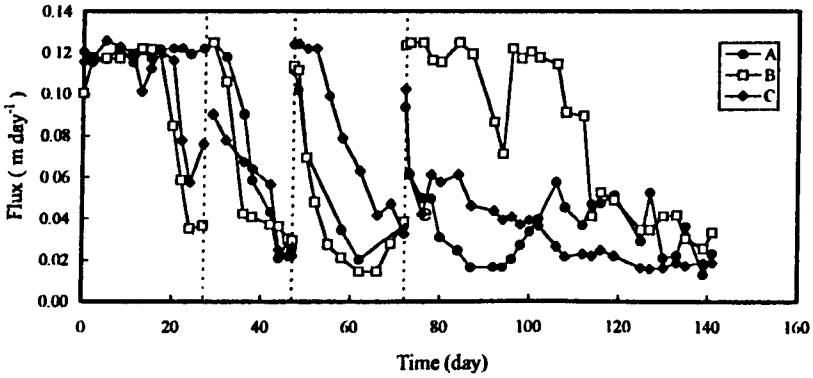
Figure 2. Change of permeate flux from the membrane modules. A: reactor A (peptone), B:reactor B (peptone and ammonium chloride), C: reactor C (ammonium chloride). The modules were pulled up and cleaned on the 27th, 47th, 72nd and 142nd day.

MLSS. Figure 3 shows the change of MLSS concentration in the aeration tanks. Removal of excess sludge was conducted on the 47th and the 67th day to keep the MLSS concentration at around 10,000 mg l^{-1}. Although no sludge removal was conducted after the 67th day, MLSS concentration in reactor A decreased between the 60th day and the 100th day partly because of foaming in the aeration tank.
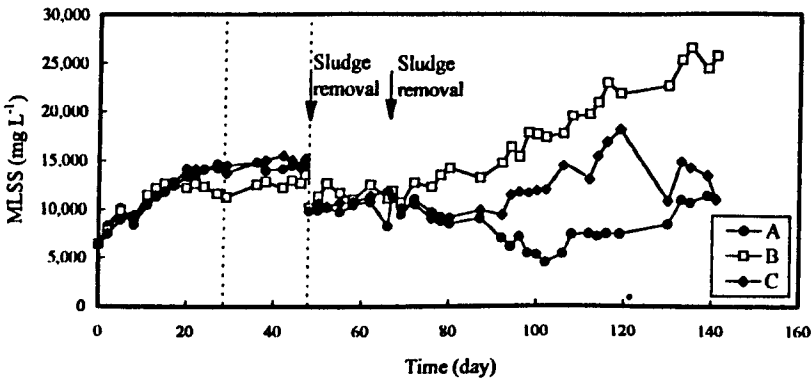

Figure 3. Change of MLSS concentration. Removal of excess sludge was conducted on the 47th and 67th day.

Filtration resistance. Filtration resistance was calculated from the data of the flux and the suction pressure according to the following equation:

$$R = \frac{J}{\mu \cdot \Delta P} \tag{1}$$

where R is filtration resistance (m^{-1}), J is flux ($m\ d^{-1}$), μ is viscosity of the permeate (mPa s) and ΔP is pressure difference through the membrane (kPa). Viscosity m was calculated from the temperature of the mixed liquor assuming that the viscosity value is the same as that of pure water. Figure 4 shows the change of filtration resistance of the membrane modules. Although the resistance was decreased by the cleaning of the membrane modules on the 27th, 47th, 72nd and 142nd days, it showed a trend of a steady increase. The decrease of the resistance in reactor A between the 90th day and the 100th day was caused by the decrease of MLSS concentration in the aeration tank during the period.
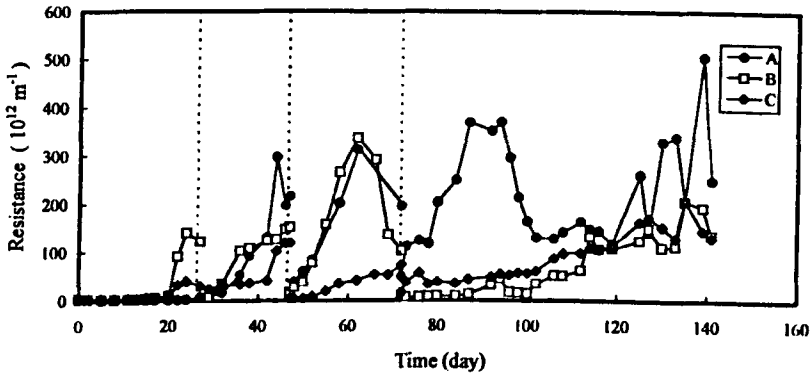
Figure 4. Change of the filtration resistance of the membrane modules. A: reactor A (peptone), B: reactor B (peptone and ammonium chloride), C: reactor C (ammonium chloride). The modules were pulled up and cleaned on the 27th, 47th, 72nd and 142nd day.

Viscosity. Figure 5 shows temporal changes of viscosity of the mixed liquor in the aeration tanks, which suggests a strong interrelationship between the viscosity and the filtration resistance of the membrane.
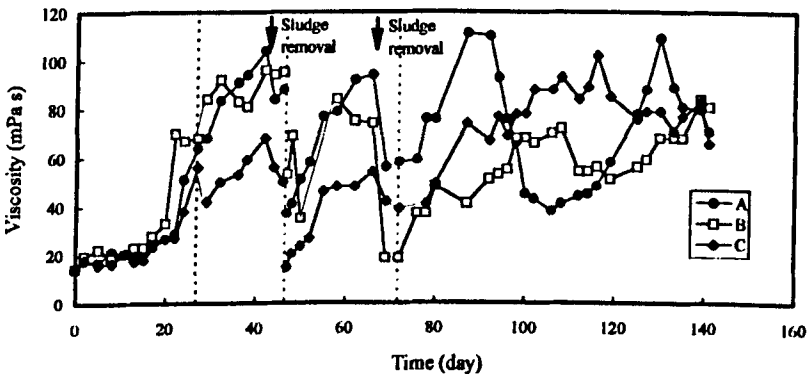

Figure 5. Change of viscosity of the mixed liquor in the aeration tanks. A: reactor A (peptone), B: reactor B (peptone and ammonium chloride), C: reactor C (ammonium chloride).

Viscosity of the permeate was almost constant at around 1 mPa s (almost similar to that of pure water) (data are not shown) even though viscosity of the mixed liquor was much higher, which means that substances which increase viscosity in the mixed liquor are removed on the surface of the membrane and accumulated in the reactors.

EPS. Figure 6 shows change of EPS concentration suspended in the mixed liquor and Fig. 7 shows change of EPS density attached on the membrane. EPS are accumulated both in the mixed liquor and on the membrane, which suggests that EPS are main substances that increase the viscosity and cause a decline in the flux.

Specific resistance of EPS was calculated according to the following equation:

$$\alpha = \frac{R}{D_p} \qquad (2)$$

where α is specific resistance of EPS (m kg^{-1}), R is resistance (m^{-1}) and D_p is EPS density on the membrane (g-TOC m^{-2}) and the calculated values are shown in Table 2.
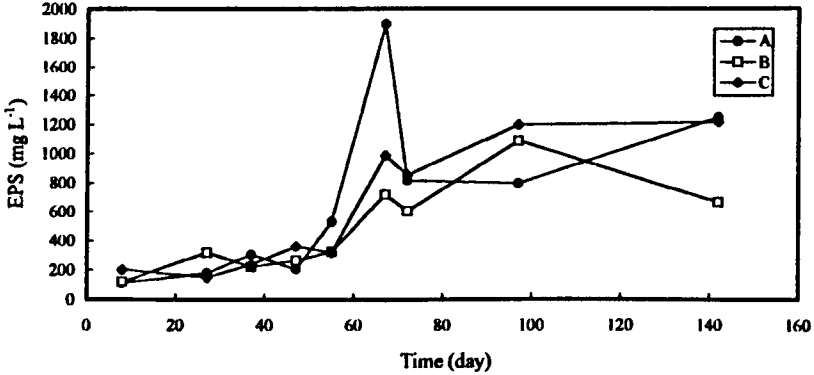

Figure 6. Change of suspended EPS concentration in the aeration tanks as mg of TOC of EPS per unit volume of the aeration tank. A: reactor A (peptone), B: reactor B (peptone and ammonium chloride), C: reactor C (ammonium chloride).
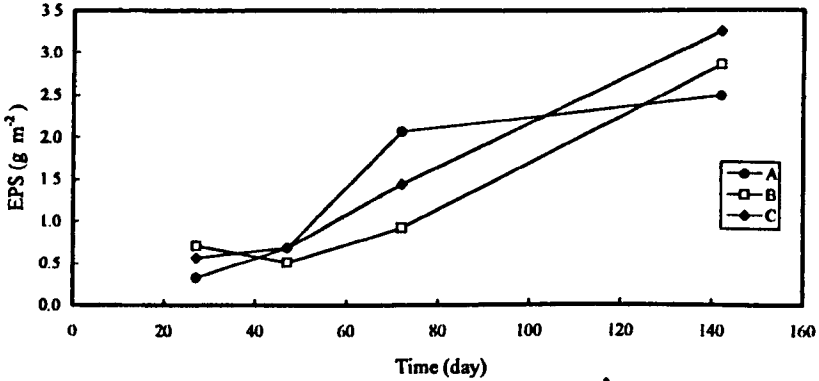

Figure 7. Change of EPS density attached on the membrane as mg of TOC of EPS per unit surface area of the membrane. A: reactor A (peptone), B: reactor B (peptone and ammonium chloride), C: reactor C (ammonium chloride).

Table 2. Specific resistance of EPS (m kg^{-1})

Day	Reactor A	Reactor B	Reactor C
27	1.7×10^{16}	1.7×10^{17}	5.2×10^{16}
47	3.2×10^{17}	3.0×10^{17}	1.8×10^{17}
72	9.5×10^{16}	1.1×10^{17}	5.0×10^{16}
143	1.0×10^{17}	4.7×10^{16}	4.0×10^{16}

DISCUSSIONS

Influence of viscosity on resistance. As Figures 4 and 5 show, viscosity of the mixed liquor influences the value of the filtration resistance. The relationship between the viscosity and the resistance is plotted in Fig. 8 on logarithmic scales, and shows a strong dependency of the resistance on the viscosity. Both the accumulation rate of EPS onto the membrane and the detachment and/or decay rate of the accumulated EPS

might be very high, which enables an equilibrium between the suspended and attached EPS to be reached rapidly.
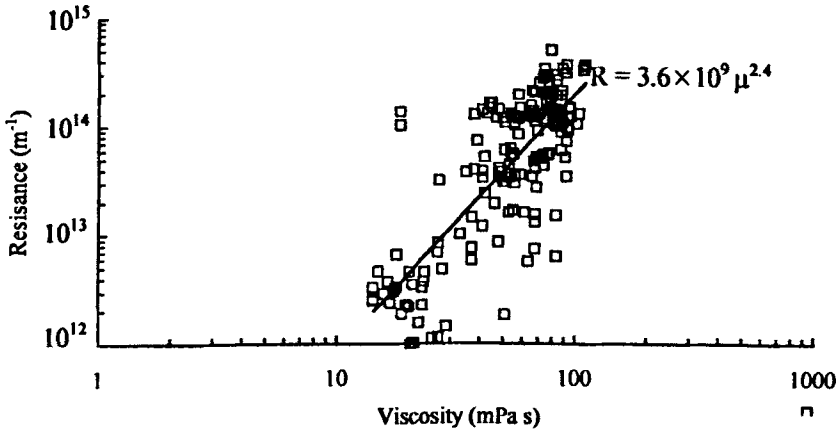

Figure 8. Relationship between viscosity of the mixed liquor and the filtration resistance of the membrane modules. Data of reactor A, reactor B and reactor C are plotted together. The equation in the figure is a regression curve to relate the viscosity μ (m Pa s) to the resistance (m^{-1}).

Dynamic analysis of resistance. The resistance R is increased by the accumulation of EPS on the membrane, which is caused by the movement of bulk water into the membrane, and is decreased by the detachment and/or biological self-decay of the attached EPS. A simplified differential equation to describe the change of the resistance was therefore developed as follows:

$$\frac{dR}{dt} = \alpha \cdot J \cdot C_p - k \cdot R \tag{3}$$

where R is resistance (m^{-1}), t is time (d), α is specific resistance of EPS (m kg^{-1}), J is flux (m d^{-1}), C_p is concentration of EPS in the mixed liquor and k is specific decrease rate of the resistance (day^{-1}). The left-hand side of equation (3) was of the order of 10^{13} m^{-1}d^{-1} while each term of the right-hand side was of the order of 10^{15} m^{-1}d^{-1}, which can make the equation more simple as follows:

$$k \cong \frac{\alpha \cdot J \cdot C_p}{R} \tag{4}$$

The value of k was then calculated and plotted against the resistance R in Fig. 9 (the value of $\alpha = 10^{17}$ m kg^{-1} was used). The value of k is nearly proportional to the reciprocal of the resistance, which means that the detachment and/or decay rate of the attached EPS is not dependent on the EPS density itself and is nearly constant during the operations of the order of 10^{15} m^{-1}d^{-1}. It might be concluded that the detachment or the decay of the attached EPS takes place only in a surface region which is close to the mixed liquor and subjected to shear stress by the rising air bubbles.

Influence of nitrogen source. Although the variation patterns of the filtration resistance were different among the three reactors (Fig.4), EPS variations, the relationship between the mixed liquor viscosity and the resistance, and the dynamics of the resistance change were similar in the three reactors (Figs 6-9). It was concluded that there was no influence of nitrogen source on the relationship between EPS and the filtration resistance.
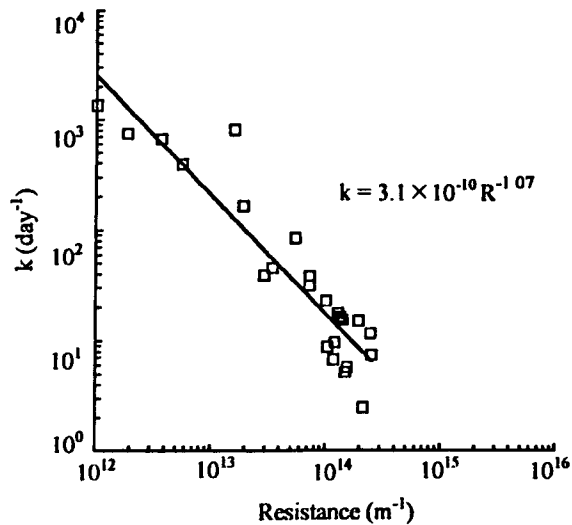
Figure 9. Dependency of the decrease coefficient of the resistance, k, on the resistance. Data of reactor A, reactor B and reactor C are plotted together. The equation in the figure is a regression curve to relate the resistance R (m^{-1}) to the coefficient k (d^{-1}).

CONCLUSIONS

In an SMAS process using hollow fiber membrane modules, EPS accumulated both in the mixed liquor and on the membrane, which might have caused an increase the viscosity of the mixed liquor and an increase in filtration resistance of the membrane. Specific resistance of EPS was calculated to be of the order of 10^{16} to 10^{17} m kg^{-1}. Detachment rate of attached EPS (decrease rate of the resistance) was nearly constant during the operation. Change of the filtration resistance was explained as a function of viscosity of the mixed liquor, which suggests the importance of the monitoring of the viscosity during the operation.

ACKNOWLEDGMENT

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