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Influence of Dissolved Organic Carbon and Suspension Viscosity on Membrane Fouling in Submerged Membrane Bioreactor

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Abstract: This paper deals with the membrane fouling in membrane bioreactor (MBR). Based on the experimental data obtained in the MBR pilot plant study, the influence of F/M ratio on the irreversible and reversible fouling was discussed in the wide range of MLSS concentration. In the case of lower MLSS concentration (2,000–3,000 mg/L), irreversible fouling rate of membrane increased with increasing F/M ratio because of the accumulation of DOC in the mixed liquor. It seems that soluble microbial products with the similar size of the membrane pore will be most responsible for the irreversible fouling. In the case of higher MLSS concentration (8,000–12,000 mg/L), reversible fouling rate of membrane increased with increasing F/M ratio because of the increased suspension viscosity caused by the increased activated sludge size or volume even in the same MLSS concentration.

Keywords: Membrane bioreactor, fouling, F/M ratio, dissolved organic carbon, suspension viscosity

INTRODUCTION

Application of submerged or internal membrane bioreactor (MBR) for the wastewater treatment in which biological treatment and solid-liquid

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separation are simultaneously achieved in a single reaction chamber has been much attention recently (1, 2). This is partly because higher quality of treated water and smaller foot print can be expected in an MBR, compared with other existing biological wastewater treatment processes such as the activated sludge process. The main obstacle for wider application of the MBR is, however, deterioration of membrane permeability (membrane fouling) with operation time.

Membrane fouling in MBR is caused by various types of physicochemical interactions between suspension containing biomass and the membrane itself. Various definitions of membrane fouling have been proposed by many researchers; for example, reversible fouling and irreversible fouling (3). Reversible fouling is defined as the fouling that can be tackled by a physical washing protocol (e.g. backwashing and air scrubbing). On the other hand, irreversible fouling is defined as the fouling that needs chemical membrane washing to be cancelled. The assembly of an external computerized literature database on MBRs by the Water Environment Research Foundation (WERF) provided us the opportunity to address questions about the role of MLSS in MBR operation (4). They, however, conclude that additional research is needed to further understand and characterize the relationships between the flux and MLSS concentration for submerged MBR.

In our previous research (5, 6), it was suggested that the degree of reversible fouling in a submerged MBR used for municipal wastewater treatment was related to the mixed liquor suspended solid (MLSS) kept in the MBR, while irreversible fouling might be caused by adsorption of dissolved organic carbon (DOC) on the membrane. Soluble microbial products (SMP) produced by the biomass and poorly biodegradable DOC contained in the raw wastewater has been thought to cause such irreversible fouling. Many researchers have reported the influence of SMP on the membrane fouling in the MBR operation (7, 8). However, these studies were based on laboratory-scale and short-term experiments and used synthetic wastewater. The authors (9, 10) have also carried out a series of long term pilot MBR experiments using the municipal wastewater and investigated the removal efficiency of micro-pollutants such as pharmaceuticals and estrogenic substances, biodegradability of the permeate. This paper summarizes the authors' previous research on the membrane fouling in submerged MBR, especially the influence of dissolved organic carbon and suspension viscosity on the irreversible and reversible fouling.

MATERIALS AND METHODS

MBR Pilot Plant

The MBR plant combining pre-coagulation/sedimentation with submerged membrane bioreactor is described in Fig. 1. It has been located in a

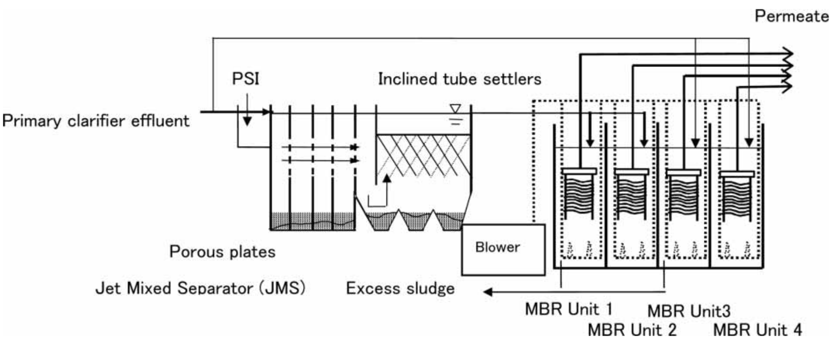

Figure 1. MBR pilot plant with pre-coagulation/sedimentation process.

municipal wastewater treatment plant in Sapporo city. The jet mixed separator (JMS) with inclined tube settlers has been used as a unit of pre-coagulation/sedimentation, where simultaneous flocculation and sedimentation occurs in the part of porous plates and residual small particles are removed in the inclined tube settlers. The treatment capacity of the JMS is 50 m^3/day, corresponding to its hydraulic retention time of 90 min. A new coagulant, Poly-Silicato-Iron (PSI) was used in pre-coagulation/sedimentation process (11). The PSI, an inorganic polymerized coagulant, has molar ratio of Fe to Si of 1 : 1 to 1 : 5 and its molecular weight is 200 to 500 kDa. The PSI with molar ratio of Fe to Si of 1 : 1 was used in the experiment. Hollow fiber MF membranes with a nominal pore size of 0.4 μm have been used for the MBR, which is made of polyethylene and filtration area of 3 m^2.

Experiments

Four MBRs (Units 1 to 4) were operated in parallel. The primary clarifier effluent was directly fed into Units 3 and 4 (called Runs 3 and 4, respectively). The JMS effluent was fed into Units 1 and 2 (called Runs 1 and 2, respectively). Dosage of PSI was fixed at 0.19 mmol Fe/L (10 mg Fe/L). The optimum coagulation pH of PSI is around 6.5 but no pH adjustment was made in the experiment. The pH of JMS effluent was between 6.5 and 7.0. The feed water temperature varied between 15 and 20°C. Table 1 describes the average water quality of the primary clarifier and JMS effluent. Fig. 2 shows the particle size distribution of TOC in the primary clarifier and JMS effluent. In the pilot plant study, not only the F/M ratio but also biodegradability of the organics for the feed water into MBR was changed by pre-coagulation/sedimentation. The same MBR operation condition was applied in Runs 1, 2, 3, and 4 as follows; hydraulic detention time = 5.3 hrs, membrane permeate flux = 0.4 m/day, suction time = 15 min., stoppage time = 1 min. and air

Table 1. Average water quality if primary clarifier and JMS effluent

	Primary clarifier effluent	JMS effluent
Turbidity (TU)	54.7	9.5
TOC (mg/L)	47.9	18.8
DOC (mg/L)	23.3	11.8
E260 (1/cm)	0.23	0.19
Total-N (mg/L)	31.2	20.2
NH4 + -N (mg/L)	14.9	10.4
Total-P (mg/L)	3.2	0.5
Alkalinity (mg/L)	143.3	95.3

scrubbing time = 3 min. MLSS concentration in Runs 1 and 3 was kept at 2,000 to 3,000 during the long term operation. MLSS concentration in Runs 2 and 4 gradually increased from 2,000 to 12,000 mg/L during the long term operation where no sludge was withdrawn. SRT changed between 12 and 32 days in Runs 1 and 3.

Two additional experiments (Runs 5 and 6) were carried out to investigate the effect of DOC on the irreversible fouling. The primary clarifier and JMS effluent were fed into the MBR in Runs 5 and 6, respectively. The operational conditions of Runs 5 and 6 are as follows: Hydraulic detention time = 3.6–6.0 hrs, permeate flux = 0.3–0.5 m/day, suction time = 12 min., stoppage time = 3 min. The intensity and mode of the air scrubbing were changed to control the reversible fouling in Runs 5 and 6. In Runs 5 and 6, no sludge was withdrawn until the operation time of 150 days, therefore, MLSS concentration increased from about 3,000 to 25,000 mg/L. MLSS concentration was reduced after that and maintained at around 10,000 mg/L.

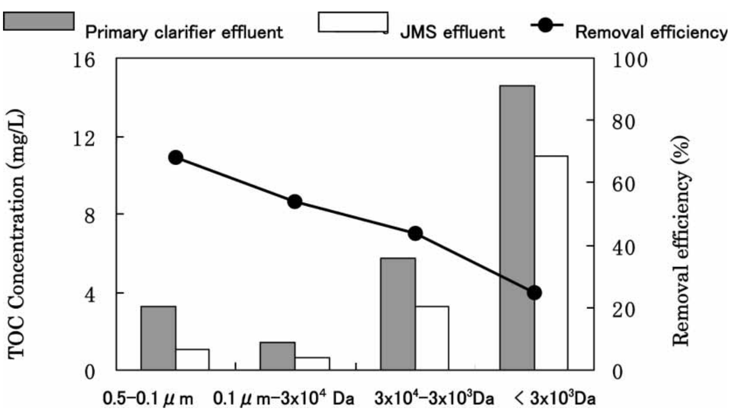

Figure 2. Particle size distribution of TOC in primary clarifier and JMS effluent.

RESULTS AND DISCUSSION

Experimental Results

Fig. 3 shows the relationship between the suspension viscosity and MLSS concentration obtained in Runs 1, 2, 3, and 4 during the long term operation. Fig. 4 shows the variation of trans-membrane pressure (TMP) during the long term operation in Runs 1 and 3. It also includes the average DOC concentration of mixed liquor and membrane permeate in the MBR for the operation time of 0 to 68 days and 70 days to 120 days. The mixed liquor in each MBR was centrifuged with 3000 rpm for 5 min., and the supernatant was filtered through a membrane with the average pore size of 0.45 μm, then DOC concentration was measured. Fig. 5 shows the similar data in Runs 2 and 4. Fig. 6 shows the changes in the total membrane filtration resistance (R_t) with increasing operation time in Runs 5 and 6. The resistance-in-series model was applied to determine the membrane filtration resistances (12). Based on this model, permeate flux on the applied TMP can be described as follows:

$$J(t) = \frac{1}{A}\frac{dV}{dt} = \frac{\Delta P}{\mu(R_t)} = \frac{\Delta P}{\mu(R_m + R_f)}$$

Where J is the permeate flux ($m^3/m^2/s$), V is the total volume of permeate (m^3), A is the membrane area, ΔP is the TMP (Pa), μ is the permeate viscosity (Pa · s), R_t is the total membrane resistance (m^{-1}), R_m is the intrinsic membrane filtration resistance (m^{-1}), and R_f is the filtration resistance due to membrane fouling (m^{-1}). R_f is calculated for each filtration experiment based on the quasi-steady state flux. Fig. 7 shows the relationship

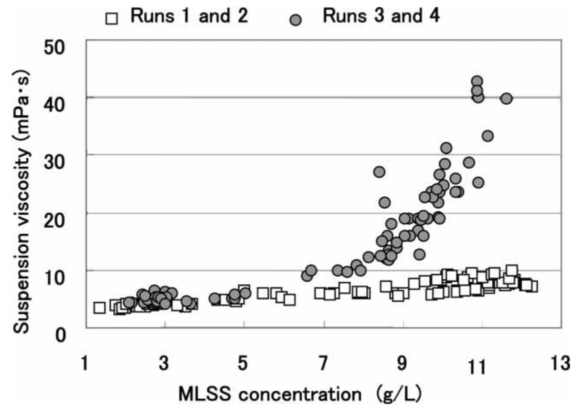

Figure 3. Relationship between suspension viscosity and MISS concentration in Runs 1, 2, 3, and 4.

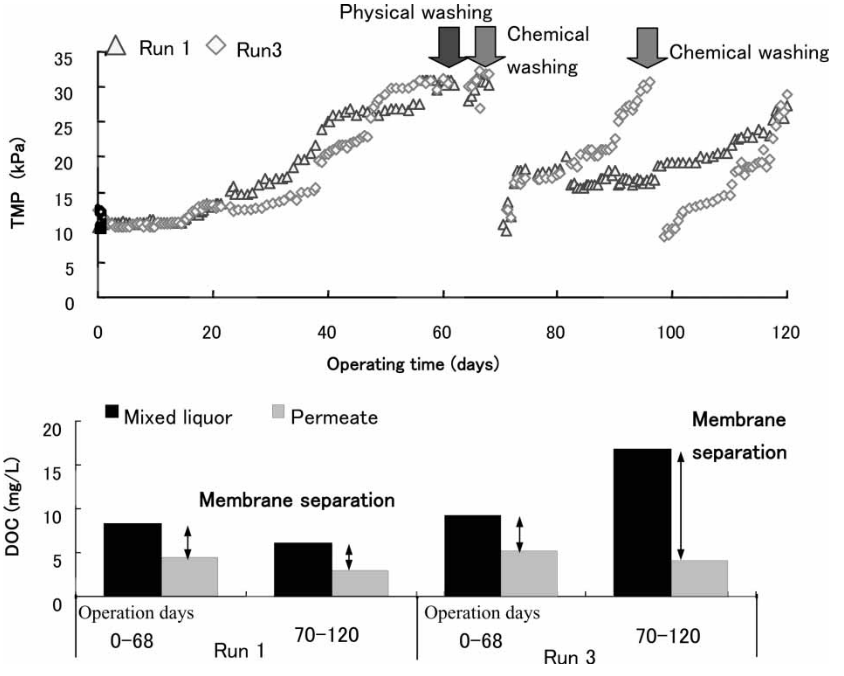

Figure 4. Variation of Trans-membrane-pressure and DOC concentration in Runs 1 and 3.

between the suspension viscosity and MLSS concentration obtained in Runs 5 and 6 during the long term operation.

Influence of Dissolved Organic Carbon (DOC) on Membrane Fouling

The TMP variation in Runs 1 and 3 was very similar until the operation time of 68 days. By comparing DOC concentration of the mixed liquor and permeate in Runs 1 and 3 during the operation time of 0 to 68 days, it is clear that the difference in DOC concentration between the mixed liquor and permeate is almost the same. It means that the amount of DOC removed by the membrane is almost the same. In Runs 1 and 3 where MLSS concentration was kept between 2,000 and 3,000 mg/l, suspension viscosity was kept in a low level. Therefore, TMP increased in a similar pattern in Runs 1 and 3. At the operation time of 58 days, membrane module was taken out from the MBR for the intensive physical washing by pressurized water.

However, TMP did not decrease by intensive physical washing. At the operation time of 68 days, fouled membranes were soaked in hydrochloric solution (pH = 2.0) and subsequently in a solution of sodium hypochlorite

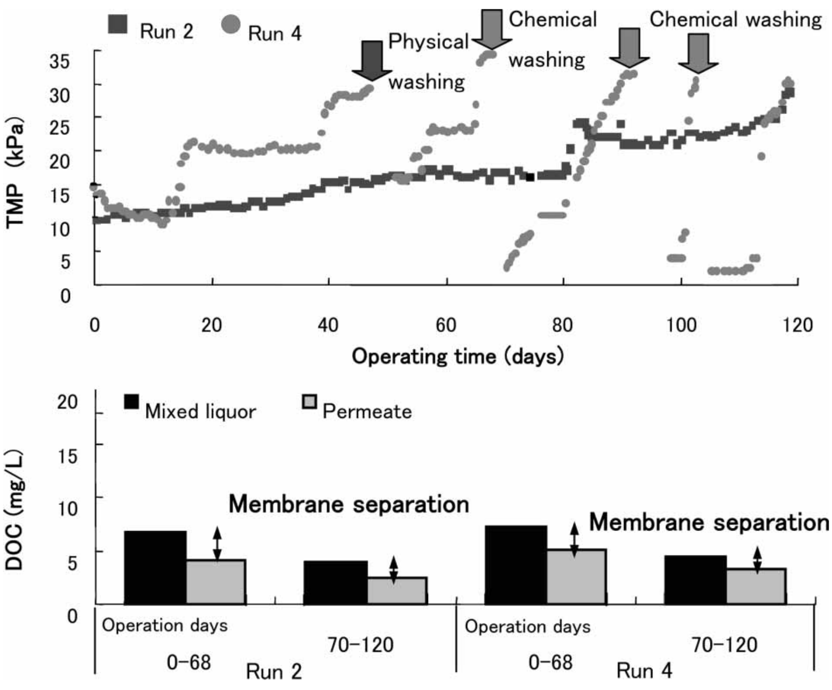

Figure 5. Variation of trans-membrane-pressure and DOC in Runs 2 and 4.

(500 mg/L) for 24 hours for chemical washing. By the chemical washing, permeability of membrane was completely recovered. However, as seen in Fig. 4, increasing pattern of the TMP in Runs 1 and 3 was very different after the operation time of 70 days. It may be because that the amount of accumulated DOC in the mixed liquor and removed DOC by the membrane were much larger in Run 3 comparing with in Run 1. The experimental results shown in Fig. 4 demonstrate that main foulant in MBR operated in a low MLSS concentration of 2,000 to 3,000 mg/l will be dissolved DOC which can be removable by the chemical washing. In Runs 5 and 6, R_f changed depending on the flux and the intensity and mode of air scrubbing, which influenced the reversible membrane fouling caused by the cake formation. In Run 5 with higher F/M ratio, rapid increase in R_f was frequently observed. A less frequency of intensive physical washing was required in Run 6 with lower F/M ratio. Recorded values of R_f just after intensive physical washing may represent the magnitude of irreversible fouling since intensive physical washing was supposed to remove reversible fouling. Based on this assumption, changes of irreversible resistance in Runs 5 and 6 are traced by the dotted line in Fig. 6. Accumulation rate of irreversible resistance observed in Run 6 was about 40% lower than that in Run 5.

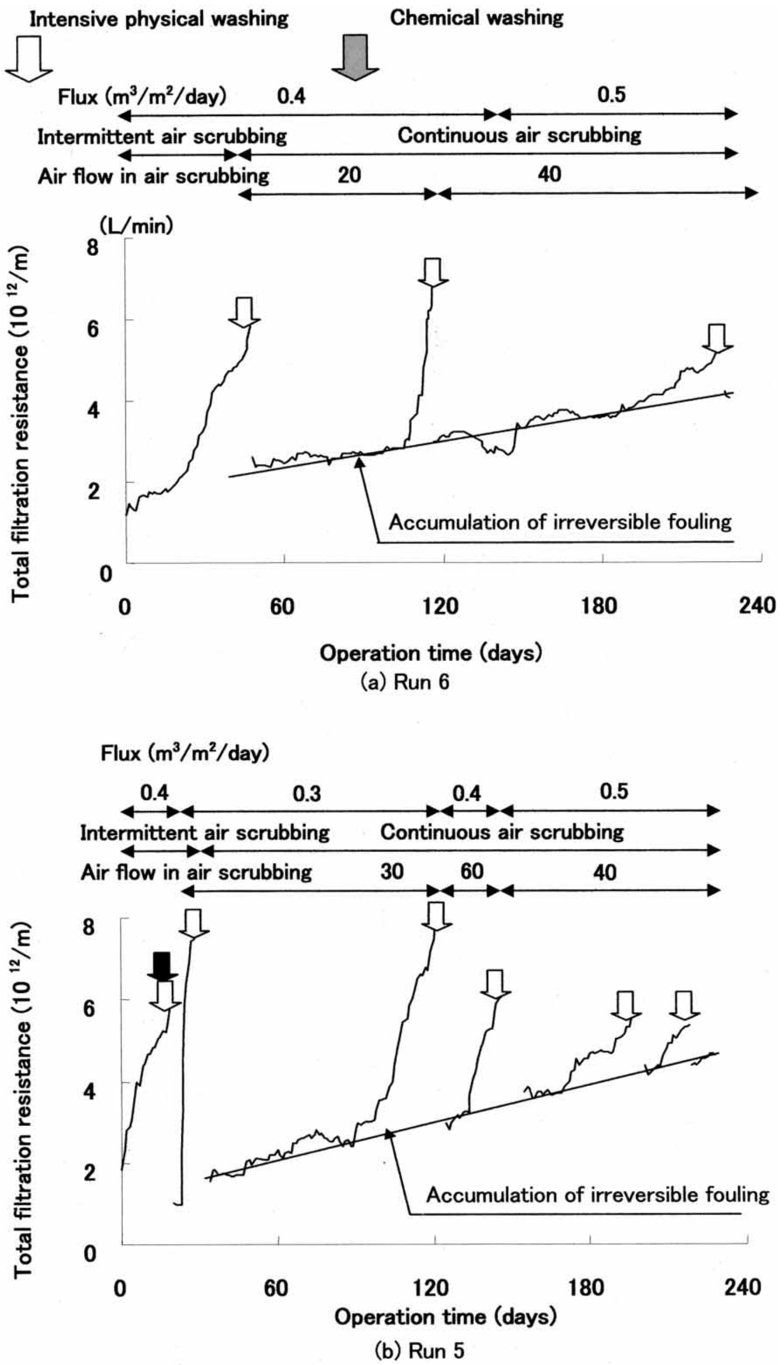

Figure 6. Change in total filtration resistance in Runs 5 and 6 with operation time.

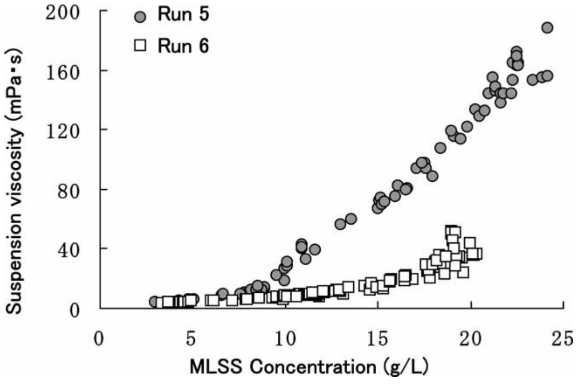

Figure 7. Relationship between suspension viscosity and MLSS concentration in Runs 5 and 6.

We have been conducting the research using the same MBR pilot plant about the effect of DOC on membrane fouling (13). We have recently found that organic particles with the size of 0.1–0.45 μm, which may be polysaccharides produced by microorganisms, significantly contribute irreversible fouling (14). More detailed study is needed to understand what kinds of DOC are the main foulants.

Influence of Suspension Viscosity on Membrane Fouling

F/M ratio has a significant effect on the membrane fouling. As discussed above, the higher the F/M ratio, the more serious the irreversible fouling in a lower range of MLSS concentration because of the accumulation of DOC in the mixed liquor. F/M ratio also has a significant effect on the reversible fouling, as seen in Fig.5. In Runs 2 and 4 where MLSS concentration increased from about 2,000 to 12,000 mg/L, TMP changed differently. TMP increasing rate was very low in Run 2 comparing with that in Run 4. The difference in TMP increase was very significant especially after the operation time of 70 days. In Run 4, membrane fouling was completely removed by chemical washing carried out at the operation time of 68 days. However, it increased very rapidly after that. The same phenomenon occurred after the operation time of 100 days. TMP increased very slowly in Run 2, although no physical and chemical washing of membranes was carried out during the long time operation. The amount of DOC separated by the membrane was almost the same in Runs 2 and 4, and irreversible fouling was completely removed in Run 4, as seen in Fig. 5. Therefore, difference in TMP increasing rate may come from the difference in the suspension viscosity as shown in Fig. 3.

Suspension viscosity seems to be determined mainly by the sludge volume. We measured the relationship between the suspension viscosity

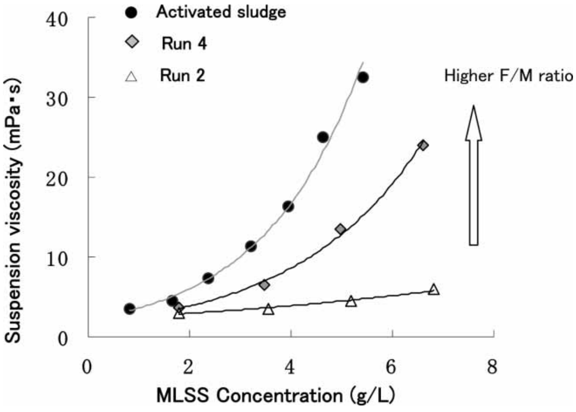

Figure 8. Relationship between suspension viscosity and MLSS concentration in AS, Runs 2 and 4.

and MLSS concentration in the conventional activated sludge process, MBR operated in Runs 5 and 6. In the case of conventional activated sludge process, mixed liquor taken from the aeration tank (SRT = 6 days) was settled, then settled sludge was diluted using MBR permeate to prepare the mixed liquor with a designed MLSS concentration. In the case of MBR, mixed liquor was taken when SRT was 38 days and 78 days in Runs 5 and 6, respectively. These samples were also diluted using the MBR permeate. Using the non–diluted samples, relationship between the sludge size and corresponding sludge volume percentage was determined. Figures 8 and 9 show the obtained relationship between the suspension viscosity and MLSS concentration and sludge size distribution in the three samples,

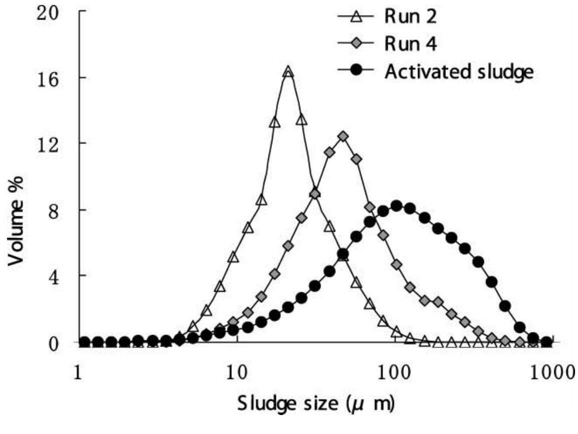

Figure 9. Sludge size distribution in AS, Runs 2 and 4.

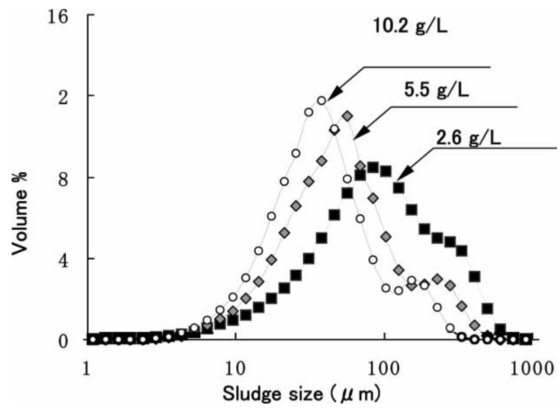

Figure 10. Sludge size distribution in Run 5.

respectively. The influence of MLSS concentration on the sludge size distribution was also investigated in Runs 5 and 6. Figures 10 and 11 show the sludge size distribution in Runs 5 and 6 with various MLSS concentration, respectively. Role of SMP or EPS should be studied to characterize the activated sludge in MBR.

CONCLUSION

This paper summarized the authors' research on the membrane fouling in the MBR. We have been conducting the MBR pilot plant study using municipal wastewater with or without pre- treatments since 1998 and published several

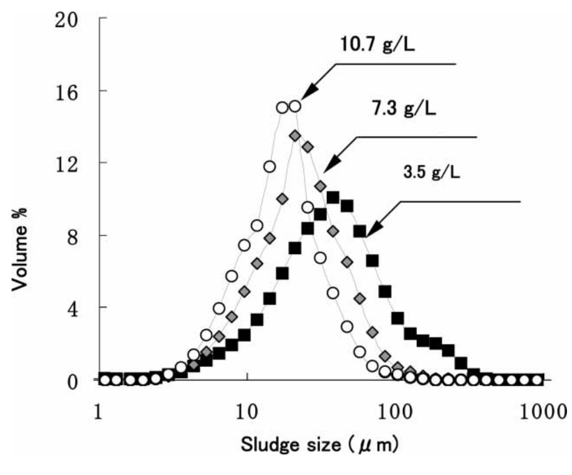

Figure 11. Sludge size distribution in Run 6.

papers. F/M ratio in the MBR was changed by the pre-coagulation/sedimentation and MLSS concentration. In the case of lower MLSS concentration (2,000–3,000 mg/L), irreversible fouling rate of membrane increased with increasing F/M ratio because of the accumulation of DOC, which seems to be soluble microbial products with the similar size of the membrane pore. In the case of higher MLSS concentration (8,000–12,000 mg/L), reversible fouling rate of membrane increased with increasing F/M ratio because of the increased suspension viscosity caused by the increased activated sludge size or volume even in same MLSS concentration.

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