Demonstrative Study on Membrane Separation Technology for Night Soil Treatment (Part1)

— Results of Pilot Plant Study —

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Abstract

Membrane separation technology has been applied for the solid-liquid separation of activated sludge in high-rate denitrification processes employed for the treatment of night soil. It renders the system capable of maintaining high concentration of microorganisms in activated sludge process. A parameter of great importance in the membrane separation is to maintain the designed flux for discharging the expected daily volume of effluent. Performance of a tubular type polyolefin membrane was evaluated in this study. The test results based on the flux data in the bench scale and pilot plant tests reveal that low-pressure operation was more favourable than the high-pressure operation for separating activated sludge. After extended periods of operation, the membrane was coated with scale composed of calcium carbonate, phosphate and similar materials. Types of coagulants used for coagulating the sludge had an important effect on the flux. Alkaline sodium hypochlorite solution and citric acid were remarkably effective as cleaning chemicals.

Key words :

Introduction

Since collected night soil and sludge from on-site treatment tanks (johkaso sludge) sent to night soil treatment plants contain nitrogen in high concentration, they are often treated by a biological denitrification process. Most of the night soil treatment plants have recently been constructed using high-load denitrification process which can meet the requirements for reducing the plant size. Night soil can be treated under high volumetric load by maintaining high concentration of activated sludge in this high-load denitrification process.^{1,2)}

Although the solid-liquid separation by

gravity, centrifugal concentrator, and some others has been used for maintaining the activated sludge at high concentration so far, the solid-liquid separation process using an ultrafiltration membrane has recently been developed. Forty-four night soil treatment plants using the membrane separation process have been constructed till 1992 in Japan.^{3,4,5)}

Since it is unnecessary to control the sludge in settling tanks in the ultrafiltration membrane separation process, clear effluent can always be discharged. The application of the ultrafiltration membrane to the night soil treatment process has been derived from the findings of a joint research of government schools and private sectors. The

Table 1. The specifications of the main equipment

Equipments	Types of equipments	Specifications
Pretreatment screen Night soil storage tank Feed pump	Rotary drum wedge wire Made of FRP Peristaltic pump	Screen opening ; 1.2mm 2.87m ϕ $\times$ 3.47mH ; 20m ³ 1.12m ³ /hr $\times$ 1.0kg/cm ² $\times$ 0.4kW
No.1 Reactor Denitrification tank Nitrification tank	Made of steel plate Made of steel plate Made of steel plate	2.2m ϕ $\times$ 11mH $\times$ 37.5m ³ 2.8m ϕ $\times$ 5mH $\times$ 3/8 ; 9.2m ³ 2.8m ϕ $\times$ 5mH $\times$ 3/8 ; 9.2m ³
Aerator(No.1 Reactor) Aerator (Nitrification tank)	Submerged axial down-flow aerator Draft tube Aeration blower Aeration blower	25m ³ -H ₂ O/min $\times$ 0.2kgf/cm ² $\times$ 22kW 550mm ϕ $\times$ 5.0mH 2.3m ³ -air/min $\times$ 0.65kgf/cm ² $\times$ 5.5kW 0.37m ³ -air/min $\times$ 0.2kgf/cm ² $\times$ 0.75kW
Heat exchanger Defoaming equipment	Spiral wound type Defoaming by turbine fan	2.5m ³ 1500rpm $\times$ 0.4kW
Activated sludge-separating membrane Circulation pump	Polyolefin tubular type Submerged pump	Molecular cut off ; 20,000 2.3m ³ $\times$ 6Modules 22m ³ /hr $\times$ 6kgf/cm ² $\times$ 22kW
Coagulated sludge-separating membrane Circulation pump	Polyolefin tubular type Peristaltic pump	Molecular cut off ; 100,000 1.56m ³ $\times$ 4Modules 6m ³ /hr $\times$ 4kgf/cm ² $\times$ 5.5kW

Fig.1 Flow chart of the pilot plant

specifications of membrane vary with the membrane module, membrane material and molecular weight to be fractionated. Membrane separation has been used not only for the separation of activated sludge but also for the separation of coagulated sludge in night soil treatment.

This paper describes the results obtained from the pilot plant study particularly using a tubular type polyolefin membrane and discusses the treatment characteristics. Part 2 of this study will summarize the operational data from a commercial plant constructed on the basic of test results of the pilot plant and discuss discrepancies in the data between the pilot plant and the commercial plant.

Outline of pilot plant

The flow chart of the pilot plant is illustrated in Fig.1. The specifications of the main equipments are listed in Table 1.

Membrane separation for activated sludge

Special features of membrane flux

The operating conditions of the pilot plant and the monthly mean values of flux are shown in Fig.2. High-pressure operation was carried out using six ultrafiltration membrane modules connected in series for the first eight months, while low-pressure operation was carried out using two trains of three modules each connected in series for the latter two months.

Since the configuration of the membrane modules is an important factor for determining the average operating pressure of the ultrafiltration, the configuration of six membrane modules connected in series was selected by investigating the data of a bench scale test conducted prior to the pilot test. Fig.3 illustrates the data of the bench scale test and flux data in the high- and low-pressure operations in the pilot test. As shown in this figure, the flux increased linearly

Fig.2 The operating conditions of the pilot plant and monthly mean values of flux

Fig.3 Relationship between the flux and the operating pressure in activated sludge separation

Table 2. Flux in the high- and low- pressure operation

	high-pressure operation	low-pressure operation
Inlet pressure kgf/cm^2	6.3	3.3
Average pressure kgf/cm^2	3.5	2.0
Outlet pressure kgf/cm^2	0.7	0.7
Linear velocity m/sec	3.4	3.1
MLSS mg/l	Ave. 18,400	Ave. 23,000
Temperature of mixed liquor $^{\circ}\text{C}$	36.3	34.4
Flux $\text{l/m}^2/\text{hr}(25^{\circ}\text{C})$	54.2	53
Configuration of modules	6 Module series	3 Module series ×2 trains

$$\text{Notes) Average pressure} = \frac{\text{Inlet pressure} + \text{Outlet pressure}}{2}$$

with the operating pressure until approximately 3 kgf/cm^2 in the bench scale test, while it reached the critical flux at the operating pressure of approximately 2 kgf/cm^2 in the pilot test. Thus, there was a difference in the critical flux at similar linear velocities between the bench scale test and the pilot test. This suggested that compaction took place at that part of the membrane exposed to high-pressure in the pilot plant operation. Table 2 lists the data obtained in the high- and low-pressure operations. This table reveals that the flux in the low-pressure operation is approximately 2% different from that of the high-pressure operation although the inlet pressure in the low-pressure operation is as low as approximately $1/2$ of that of the high-pressure operation. It is considered, therefore, that the high-pressure operation has few advantages regarding the selection of the pump and compaction of membrane.

In the pilot plant operation, critical flux occurred at a lower pressure condition than that of the bench scale test. This seems to be because of different types of gel layers formed on the surface of the membrane during the long term pilot plant operation. The composition of feed solution should be, therefore, taken into

account in determining the permeability and selecting the cleaning method.

Chemical cleaning of membrane

Fouling of the ultrafiltration membrane by activated sludge may be caused by the gel layer formed by microorganisms, colloidal substances, protein and the like. A mixed solution of sodium hypochlorite and sodium hydroxide was used as the cleaning fluid. The cleaning was carried out based on the bench scale test data in such a way that the membrane module was cleaned with water followed by circulating the cleaning fluid (pH:11.5) containing 100 mg/l of NaOCl and NaOH for approximately an hour. The recovery rate of flux by cleaning the membrane module with the mixed cleaning fluid (pH:11.5) was as low as 75% during the period when ferric chloride was added to the activated sludge tank to remove phosphate in the pilot test. It was, therefore, necessary to clean the inorganic matter with hydrochloric acid or citric acid for approaching 100% of the recovery rate.

Table 3. Background of used modules

Membrane material : Polyolefin

Molecular cut off : 20,000

Module No.	M-22	M-16
The term in use	About twenty-two months	About sixteen months
Flux at the first stage	Flux = 64.7 $\ell / \text{m}^2 / \text{hr}$ $\bar{P} = 3.1 \text{ kg/cm}^2$ LV = 3.6 m/sec By well water (21°C)	The same as the left
Flux just after the final cleaning	Flux = 66.3 $\ell / \text{m}^2 / \text{hr}$ $\bar{P} = 1.83 \text{ kg/cm}^2$ LV = 2.6 m/sec By well water (25°C)	Flux = 25.8 $\ell / \text{m}^2 / \text{hr}$ $\bar{P} = 1.85 \text{ kg/cm}^2$ LV = 2.7 m/sec By well water (24.5°C)
The final cleaning	NaOCl 100mg/ ℓ +NaOH solution (pH12) and 0.5% citric acid solution (pH2.7)	NaOCl 100mg/ ℓ +NaOH solution (pH12)

 $\bar{P}$; Average pressure LV ; Linear velocity

Table 4. Deposited matter on the surface of the activated sludge-separating membrane

Sample	Analytical methods	Outline of observational results
Module M-22 surface of the membrane	SEM and EDX	•Microorganisms and compounds of Fe and Si were detected partially on the surface of the membrane. •Cl ⁻ was also detected.
	FT-IR	•Amide coming from microorganisms remained. •Compounds of Ca and Fe remained. •Calcium-carbonate or -phosphate was not detected.
Module M-16 surface of the membrane	SEM and EDX	•Calcium compounds were detected on the whole surface of the membrane.
	FT-IR	•Calcium-carbonate and -phosphate remained.
Module M-16 support tube of the membrane	SEM and EDX	•Considerable microorganisms, slime, phosphate, and calcium compounds were detected.
	FT-IR	•Considerable absorbance due to calcium carbonate and calcium phosphate was detected.

Observation of materials adhered to the surface of membrane and analysis of scale

The surfaces of two types of membranes and membrane supporters with different service conditions as shown in Table 3 were observed by scanning electron microscopy (SEM) and energy dispersive X-ray analysis (EDX), and the materials adhered to the surface of membranes were analysed by Fourier transform infrared spectroscopy (FT-IR). The results are listed in Table 4. An example of the result of FT-IR is illustrated in Fig. 4. No scale composed of calcium carbonate and phosphate can be observed on the surface of the membrane cleaned with citric acid. The citric acid cleaning is, therefore, effective.

The analytical results of the ultrafiltration membrane and the membrane supporters used for sixteen months indicate that the scale composed of calcium carbonate, calcium phosphate and similar materials causes fouling.

Membrane separation for coagulated sludge

Special features of membrane flux

Unlike the mixed solution with activated sludge, the flux is almost directly in proportion to the operating pressure generated by passing the coagulated sludge-containing solution, less depending upon the linear velocity. The flux of the membrane used for separating coagulated sludge is shown in Fig. 5. This suggests that few gel layers are formed on the surface of the membrane passing the coagulated sludge-containing solution.

The flux through the membrane used for separating activated sludge can be steadily maintained under the same operating conditions for two to three months after the cleaning, while that for separating the coagulated sludge sometimes decreases to approximately 1/3 in three weeks under the same conditions depending upon

the types of coagulants.

Chemical cleaning of membrane

The cleaning test was carried out using sodium hypochlorite and sodium hydroxide followed by hydrochloric acid or citric acid cleaning to determine the water flux of each cleaning fluid at 25°C. The results of the test are listed in Table 5. The recovery rate by sodium hypochlorite (pH:12) was approximately 86% and it reached approximately 100% by additionally using hydrochloric acid. The flux sometimes decreased during cleaning the membrane with sodium hypochlorite (pH:12) after the hydrochloric acid cleaning in the runs of ④ and ⑤. This may be due to the deposition of scale on the surface of the membrane, considering that the calcium content in the cleaning fluid is reduced by approximately 50%. Since the repeated cleaning with citric acid can be accomplished in a shorter time than that with hydrochloric acid, citric acid is considered more effective than hydrochloric acid.

Coagulants and fouling

A laboratory test was made using a tubular module, 11.5 mm in diameter and 500 mm in length, to determine the effect of coagulants on the polyolefin membrane. Ferric chloride, poly ferric sulfate and aluminum sulfate were used as coagulants. Each coagulant was added to the permeated water of activated sludge-separating membrane to adjust the concentration at 500 mg-Fe/l or 395 mg-Al/l respectively. Thereafter, it was coagulated by adjusting the pH of the solution in the range of 4.5 to 5.5. The coagulated sludge was separated through the membrane such that the flux was always maintained at approximately more than 80 l/m²/hr through a step-by-step increase in the operating pressure. About 2.5 kgf/cm² operating pressure was required at the final stage. These results are illustrated in Fig. 6. The fouling period till the time to be cleaned varied

Fig.4 Result of FT-IR spectroscopic analysis
(M-16 module, Difference spectrum to new membrane)

Fig.5 Relationship between the flux and the operating pressure in coagulated sludge separation

Fig.6 Operating performance of coagulated sludge separation

Table 5. Results of chemical cleaning of membrane

Processes of cleaning runs	Cleaning methods	Flux of tap water ($\ell/\text{m}^2/\text{hr}/\text{atm}(25^\circ\text{C})$)
R-0	Before the coagulated sludge-separating test.	7 6
R-1	After the coagulated sludge-separating test. (Before cleaning)	4 8
C-①	Cleaning with NaOCl 100mg/ ℓ and NaOH solution (pH12) for 1 hour	6 1
C-②	Cleaning with NaOCl 150mg/ ℓ and NaOH solution (pH12) for 1 hour	6 2
C-③	Cleaning with HCl(pH2) for 1 hour	7 4
C-④	Cleaning with NaOCl 150mg/ ℓ and NaOH solution (pH12) for 2 hours	6 2
C-⑤	Cleaning with NaOCl 100mg/ ℓ and NaOH solution (pH12) for 1 hour	6 2
C-⑥	Cleaning with HCl(pH2) for 1 hour	7 3
C-⑦	Cleaning with HCl(pH2) for 2 hours	7 4
C-⑧	Cleaning with 5W/V% citric acid (pH2.7) for 2 hours	7 8

Table 6. Recovery of water flux after the physical cleaning

Membrane No.	Used coagulants	Flux of tap water($\ell/\text{m}^2/\text{hr}, 25^\circ\text{C}$)		
		Before the separation test	After the separation test	After the physical cleaning with a test tube brush
A	Ferric chloride	2 4 4	1 3 7	2 2 5
B	Poly ferric sulfate	2 6 8	1 0 6	2 7 0
C	Aluminum sulfate	2 2 3	9 8	1 9 8

Flux was measured after 30 minutes passage point of the separating run.
Average operating pressure is 1.5 kgf/cm²

Table 7. Results of the property analysis after the physical cleaning

Membrane No.	Used coagulants	Analytical methods	Outline of observational results
A	Ferric chloride	SEM	•Few depositions were detected.
		EDX	•Fe was detected.
		FT-IR	•Absorbance due to hydrocarbon and hydroxide existed.
B	Poly ferric sulfate	SEM	•Some coagulated matter was adhered.
		EDX	•Fe, Al, and Ca were detected. S was not detected.
		FT-IR	•Absorbance due to Fe, hydroxide, and amide existed.
C	Aluminum sulfate	SEM	•Coagulated matter was deposited on the whole surface.
		EDX	•Al and P were detected. S was not detected.
		FT-IR	•Absorbance due to Al, hydrocarbon, and hydroxide existed.

according to the type of coagulant and was about 420 hours for ferric chloride, 270 hours for poly ferric sulfate and 120 hours for aluminum sulfate. The used ultrafiltration membrane was physically cleaned with a test tube brush and then the water flux through the membrane was determined. The results are shown in Table 6. These results indicate that physical cleaning is effective for recovering the water flux. The qualitative analysis of the ultrafiltration membrane cleaned with the brush was made by SEM and EDX, and the composition analysis was made by FT-IR. The results are shown in Table 7. The quantities of scale deposited on the surface of the membrane were high in the decreasing order of aluminum sulfate, poly ferric sulfate and ferric chloride. This order is inversely proportional to that of the fouling time. A sticky substance was deposited on the whole surface of the membrane when using aluminum sulfate. No hard scale composed of CaSO_4 and others derived from the anions including Cl^- and SO_4^{2-} of the coagulants was observed.

Conclusions

The results obtained from the pilot plant study carried out to investigate the application of the membrane separation technology to the night soil treatment are summarized as follows:

- (1) For separating activated sludge, there is a difference in the critical flux passing through the membrane between the bench scale test and the pilot test. This may be because compaction takes place and an unremovable gel layer or scale is formed on that part of membrane exposed to high-pressure for a long time.
- (2) The high- and low-pressure operations of the activated sludge-separating membrane in the pilot plant were carried out and the results indicated that the high-pressure operation has few advantages.
- (3) Scale composed of calcium carbonate, phosphate and the like was observed on the surface of

the membrane used for separating activated sludge for a long time. The activated sludge-separating membrane is generally cleaned with alkaline sodium hypochlorite to recover the flux. The acid cleaning is effective for the membrane used for a long time because inorganic scale is deposited on its surface.

(4) The flux depends upon the types of coagulants used for the coagulated sludge-separating membrane. The results of FT-IR of the membrane indicate that hydroxides derived from the cations of the coagulants, Ca and P are deposited on the surface of membrane to some extent along with a smaller amount of hydrocarbons.

(5) The results of the membrane cleaning test and the membrane surface observation indicate that an alkaline sodium hypochlorite solution and citric acid are remarkably effective as the cleaning fluids for membrane.

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