

3. Applications of Membrane Processes to The Treatment of Wastewaters in Japan

(Yamamoto 교수, 일본 동경대학교)

APPLICATIONS OF MEMBRANE PROCESSES TO THE TREATMENT OF WASTEWATERS IN JAPAN

Kazuo Yamamoto

Department of Urban Engineering, The University of Tokyo
7-3-1 Hongo, Bunkyo-ku, Tokyo 113, Japan

INTRODUCTION

The membrane processes that are commonly used in water and wastewater treatment are reverse osmosis (RO), ultrafiltration (UF) and microfiltration (MF), which utilize pressure differentials. There is also nano-filtration (NF), or low-pressure reverse osmosis, which is positioned midway between conventional reverse osmosis and ultrafiltration. Reverse osmosis membranes reject dissolved ions, while ultrafiltration can be used to reject relatively larger molecules, such as protein, polysaccharides and so on. Microfiltration is capable of eliminating particles at submicron level.

Wastewaters usually include dissolved organic matters, a large part of which is biodegradable. As biological treatment is the best available technology for the organic wastewaters in most cases, the membrane separation in combination with biological treatment is reasonably used for them. The activated sludge process that is the most typical suspended-growth biological wastewater treatment process normally achieves solid-liquid separation through gravity sedimentation. MSAS (Membrane Separation Activated Sludge) is the name given to processes in which this role is fulfilled by membrane separation. Since the main purpose of using membrane separation is to separate biologically treated water from suspended solids, the use of reverse osmosis would be meaningless. In fact, reverse osmosis would lead to the accumulation of salts and would inhibit microbial activities of the sludges. For this reason, membrane separation is carried out by means of ultrafiltration or microfiltration, which do not cause the accumulation of salts or organic substances that have been converted into low molecular weight ones as the final metabolic products of microorganisms.

Some industrial wastewaters such as metal plating ones require no biological treatment, where direct membrane separation is possible but other physico-chemical processes like coagulation-sedimentation are usually applied prior to the membrane separation. Direct ultrafiltration with simple pre-screening has been applied to graywater treatment in some individual building reuse systems¹⁾. However, biological filter is preferably added before the membrane separation in order to remove odorous substances.

This paper summarizes the characteristics of MSAS process first, as it is the main membrane process applied to wastewater treatment. Two successful examples of the applications, the cases of individual building reuse system and nightsoil treatment, are then shown. The latest trend of new membrane applications, i.e., immersed-type MSAS is also introduced.

CHARACTERISTICS OF MSAS PROCESS

From the viewpoint of biological treatment, a number of advantages can be gained through combination with membrane separation. First, since suspended solids are totally eliminated through membrane separation, the settleability of the sludge has absolutely no effect on the quality of the

treated water. Second, a long enough sludge retention time (SRT) can be provided, facilitating the proliferation of microorganisms with low growth rate, such as nitrifying bacteria. Third, the overall activity level can be raised, since it is possible to maintain high microbial concentrations in bioreactors while keeping the microorganisms dispersed as much as possible. Fourth, high concentrations of the sludges create a favorable environment for "endogenous" denitrification, thereby ensuring the efficient removal of nitrogen. Fifth, treatment efficiency is also improved in the sense that it is possible to prevent leakage of undecomposed polymer substances. Sixth, the method itself gives high enough removal of bacteria and viruses, although ultrafiltration membranes are not complete barrier for virus rejection²⁾.

From the perspective of membrane separation, the possible advantages of combination with biological treatment include the followings. First, dissolved organic substances with low molecular weights, which cannot be eliminated by membrane separation alone, can be taken up, broken down and gasified by microorganisms or converted into polymers as constituents of bacterial cells, thereby raising the quality of treated water. Second, polymer substances retained by means of membranes can be broken down if they are biodegradable, which means that there will be no endless accumulation of the substances within the treatment process. This, however, requires the balance between the production and degradation rates. A high organic loading might give a higher production rate of intermediate polymer metabolites than that of the degradation, resulting their accumulation. The accumulation of intermediate metabolites may decrease the microbial activities³⁾.

As mentioned above, MSAS leads naturally to operation with high concentration of activated sludges, which can easily absorb shock loadings and give very stable treatability. The performance of MSAS may be affected by irreversible changes in the membranes themselves, or by the fouling of membrane interiors or surfaces. In the membrane separation of high concentration of activated sludges, there is a particularly conspicuous tendency for permeability to decline or separation characteristics to change due to the formation of cake and gel-like layers. The actual rejection performance of ultrafiltration membranes and microfiltration membranes is often similar⁴⁾, since separation characteristics are determined by the nature of the cake and gel-like layers formed.

There is also a correlation between sludge concentration and permeation flux. When sludge concentration exceeds a certain limit, the permeation flux rapidly decline due to a dramatic rise in viscosity of the sludge mixture. The limit for the filtration of the activated sludge concentration is typically 30,000 to 40,000 mg/l^{5),6)}. The correlation between sludge concentration and the permeation flux will not always be negative below this threshold concentration. There is no uniform tendency and there may be either no relationship or even a positive correlation. It appears that this situation largely attributable to the influence of colloidal or soluble substances, which cannot be measured in terms of sludge concentration within the sludge mixture.

Unlike the filtration of pure water, there is no guarantee that there will be linear relationship between operating pressure and permeation flux. An increase in pressure sometimes causes a decline in the permeation flux, and it is possible that the compaction of the cake layer is a key factor here⁷⁾.

Control of the cake layer plays an important role in the reduction of fouling, since the filtration is dominated by the cake layer on the surface of the membrane. One well-known approach is to improve the scouring effect by increasing the tangential flow velocity, but this method has the drawback of increasing energy consumption. It is also important to reduce compaction of the cake layer. Low-pressure filtration and intermittent filtration appear to offer an effective means of achieving this through the manipulation of system operation⁶⁾.

REUSE OF WASTEWATER IN INDIVIDUAL BUILDING

In Japan, membrane separation technology has been used on a significant scale in wastewater reuse systems in individual buildings for the past 20 years. Rapid increase in water demand especially in big cities in 1970's due to growing activities in the business area has made the national and local governments have policies to regulate big consumers for saving water. For example, Hukuoka City often suffers from water shortage. The city government set the administrative outlines, since 1979, in which big buildings having total floor area of greater than 5000 m² or having a service water pipe of its size larger than 50 mm should install a water saving facility that means reuse of treated wastewater or rainwater. Tokyo metropolitan government also set the guidelines, since 1984, in which buildings having total floor area of more than 30,000 m² or possible amount of reused water of more than 100 m³/d should have a reuse facility.

There are three types of water reuse system: individual building reuse, district-wide reuse and large scale recycling. The report of Japan Land Agency⁸⁾ in 1990 shows that there are 840 individual reuse facilities covering about 127,000 m³/d, while 42 district-wide reuse systems that supply reclaimed water to 127 buildings that make about 56,000 m³/d and 27 large scale recycling works supplying 402 buildings and about 45,000 m³/d. Two industrial water supply works whose raw water is treated sewage also distribute the nonpotable water to buildings. Among the building reuse facilities, 35% of them are cited in Tokyo and 22% of them are in Hukuoka.

Most of large-scale recycling and district-wide reuse systems employ sand filtration as a main tertiary treatment process, some of which use ozonation as a polishing process. On the other hand, MSAS processes are popularly used in individual building reuse systems.

Figure 1 shows number of the yearly installations of individual building reuse systems calculated from the individual data listed in the Zosui Gijutsu Handbook¹⁾. Most of all membrane facilities adopt UF with few exception that two facilities utilize RO. About 40% of them adopt UF systems (two RO systems included). Figure 2 shows that 84% of UF systems are employed in combination with biological treatment, most of which are MSAS processes. Figure 1 Yearly installation of individual building reuse systems

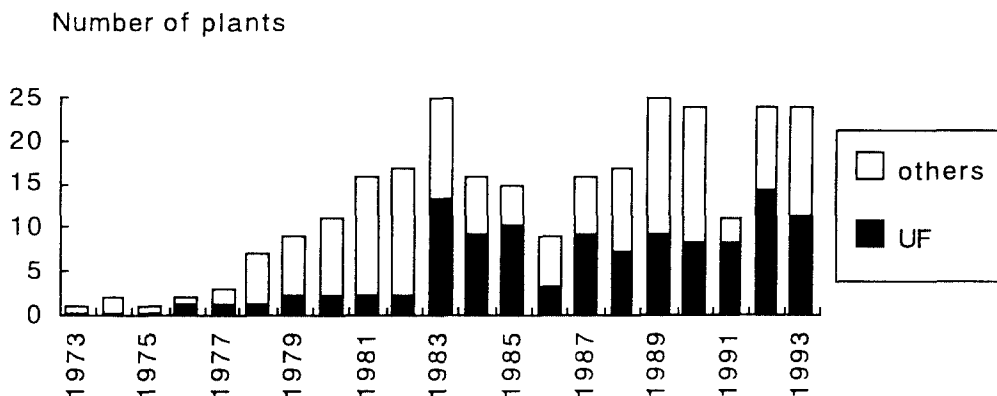


Figure 1 Numer of yearly installations of individual building reuse systems

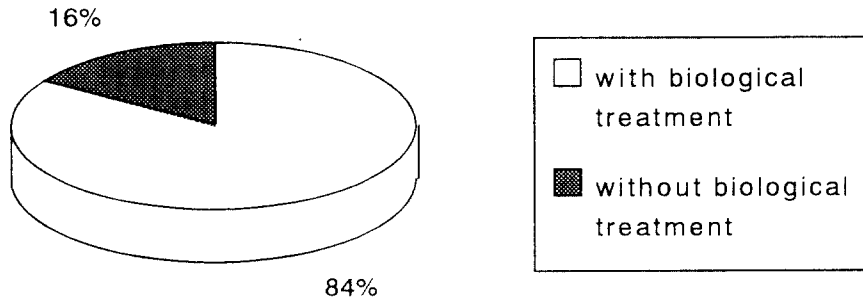


Figure 2 Type of UF facilities in individual building reuse system

MSAS processes as a building reuse facilities gain popularity because of some advantages as below:

- (1) compact and easily installed at the basement of buildings,
- (2) simple and very easy to be maintained,
- (3) hygienically reliable, and
- (4) excellent water quality compare to that from conventional tertiary treatment.

Figure 3 shows the size distribution of individual building reuse systems (the data source are the same as in Fig. 1). All of UF systems have sizes less than 500 m³/d except one case of district-wide reuse system of 700 m³/d that is to be expanded to 1,700 m³/d, using MSAS in Tama New Towns located in the western suburbs of Tokyo.

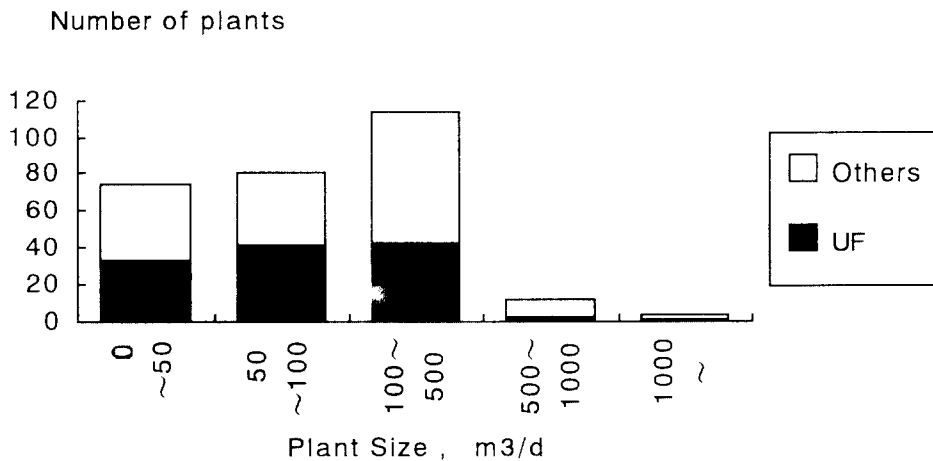


Figure 3 Size distribution of individual building reuse systems

Plate and frame modules and tubular modules are mostly used under cross-flow condition, giving disadvantages of generally high energy consumption and high cost of treatment. Recent development of new modules such as a rotating disk membrane module, an immersed-type ceramic tubular module and so on, gives a few exception.

These disadvantages are not so seriously recognized by the users in Japan, because the maintenance energy for big buildings is already very large. In addition, a very special situation in Japan that water works and public sewage works adopt an increasing block tariff gives big consumers incentives to introduce a water reuse system in their buildings. Table 1 shows examples of the water charges in some large cities in Japan. The cost for tap water service and discharge to sewerage often exceeds 600 Yen/m³, although the average production cost of drinking water in Japan is 159 Yen/m³ and the average sewage treatment cost is 154 Yen/m³ as of 1991, giving the sum of only 313 Yen/m³.

Table 1 Water charges for tap water usage and discharge to sewerage in large cities in Japan (based on 200 m³/d of usage) (yen/m³) (after Ohtaka⁹⁾)

City	tap water	sewerage	total
Tokyo	382	295	677
Yokohama	325	215	540
Kawasaki	298	125	423
Nagoya	284	203	487
Osaka	298	130	428
Hukuoka	453	173	626
Kitakyushu	290	137	427

Note: 1 Yen is equivalent to 9 Won as of 1995.6

For example, the total cost for water usage in the case of the Shinjuku Station Building where a plate & frame type UF MSAS process of its design capacity of 250 m³/d has been operated since 1985 is 676.5 Yen/m³, while the total cost of reclaimed water production is 458.8 Yen/m³ (as of 1988)¹⁰⁾. The balance of 676.5 - 458.8 = 217.7 yen/m³ shows that this system gives 32% reduction of the water and sewerage charges. Table 2 summarizes some water quality of the process.

Table 2 water quality of UF MSAS at Shinjuku Station Building¹¹⁾

	raw wastewater			treated water		
	max.	min.	ave.	max.	min.	ave.
pH	6.9	5.4	6.1	7.9	7.5	7.7
SS, mg/l	264	122	184	nd	nd	nd
BOD, mg/l	800	230	453	3.6	0.3	1.3
COD(Mn), mg/l	494	153	272	16.1	6.2	9.2

Another example is the case of Nihonbashi Takashimaya Department Store located at the center of Tokyo, where a UF tubular MSAS process of its capacity of 450 m³/d has been operated since 1987. The initial cost including the facilities expanded is about 200 million Yen and the yearly running cost is about 20 million Yen, while yearly saved water is equivalent to about 90 million Yen¹²⁾. This shows that the user made profits after three year of the operation.

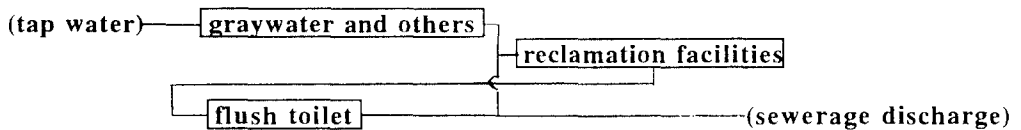
However, there are also examples where the construction plan overestimated the design capacity and resulted in too large size of the reuse facilities, giving a very high cost of reclaimed water production,

e.g., sometimes more than 1500 Yen/m³.

The UF processes receive various types of wastewater such as toilet flush, restaurant wastewater, cooling water and so on. Figure 4 shows the typical flow diagrams of individual building reuse systems according to classification whether toilet flushing is included or not. The former type includes toilet flush as raw wastewater, this means that the water is partially recycled between the toilet and MSAS process. In case of sudden stop of water supply due to accidents or disaster, this type may work as a closed system in the building, where people can use the toilets. However, the treatment cost is generally higher than the others due to higher organic loading and color problem that usually needs additional process like activated carbon filter to remove color.

The latter type gives a cascade flow that can reduce organic loading and color problem, but it requires a back up water system in case of imbalance among each water usage. As shown in Fig. 5, 73% of UF systems receives no toilet flush by employing a cascade type flow system, because any user wants to further reduce the treatment cost.

(A) partially recycle flow type



(B) cascade flow type

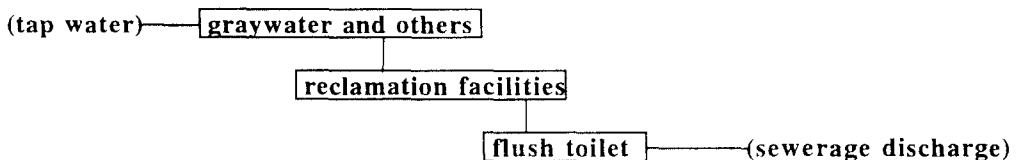


Figure 4 Typical flow diagrams of individual building reuse systems

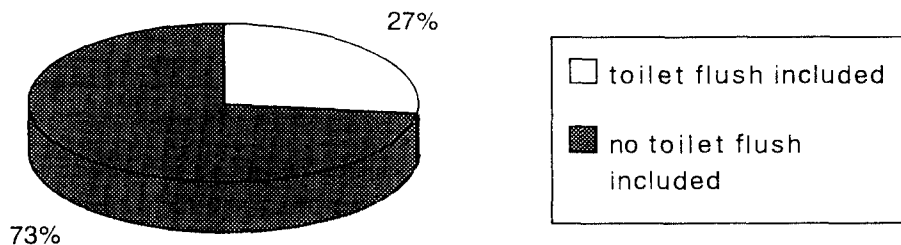


Figure 5 Type of raw wastewaters of UF reclamation facilities.

NIGHTSOIL TREATMENT

Recently there has been a dramatic increase in the use of membrane separation technology in nightsoil treatment. Membrane separation, as an innovative technology, seems very attracting to the people who live in the neighborhood of the treatment plant. The plant size is small and it is suitable to membrane separation. The strict requirement of the effluent quality have made the nightsoil treatment plants introduce advanced treatment including color removal and the elimination of nitrogen and phosphorus. In order to reduce the hydraulic loading on subsequent advanced treatment processes and improve denitrification efficiency, it is necessary to operate the preceding biological treatment process at high sludge concentrations with little or no dilution. This is reflected in higher biological concentrations in treatment process, with the result that it is no longer possible to separate liquids and solids reliably by conventional gravity sedimentation. MSAS has been introduced as one of means of overcoming this problem¹³. The first MSAS plants of 20 m³/d was constructed in 1986. There are 51 MSAS plants operating (as of 1993)¹⁴. The largest plant has capacity of 160 m³/d, while most of plants have their size of less than 100 m³/d. MSAS as a first stage is employed for biological nitrogen removal process as well as organic removal. Most of them have following secondary membrane separation stage in combination with chemical coagulation to remove color and phosphorus. Many plants add further polishing stage using activated carbon adsorption. About half of new plants have introduced MSAS in 1992 and 62% of them install tubular modules and most of remaining are plate and frame modules. A few of immersed type MSAS processes and rotating membrane disk modules have been introduced very recently.

Figure 6 shows the treatment flow diagram of a UF MSAS plant. It has been operating since 1989. The daily treatment capacity is 40 m³/d, including 28 m³/d of nightsoil and 12 m³/d of jokaso(septic tank) sludge. Two-stage membrane separation is employed, where the first stage is a MSAS used for organic removal coupled with biological nitrogen removal and the second stage is a coagulation-membrane separation process for color and phosphorus removal.

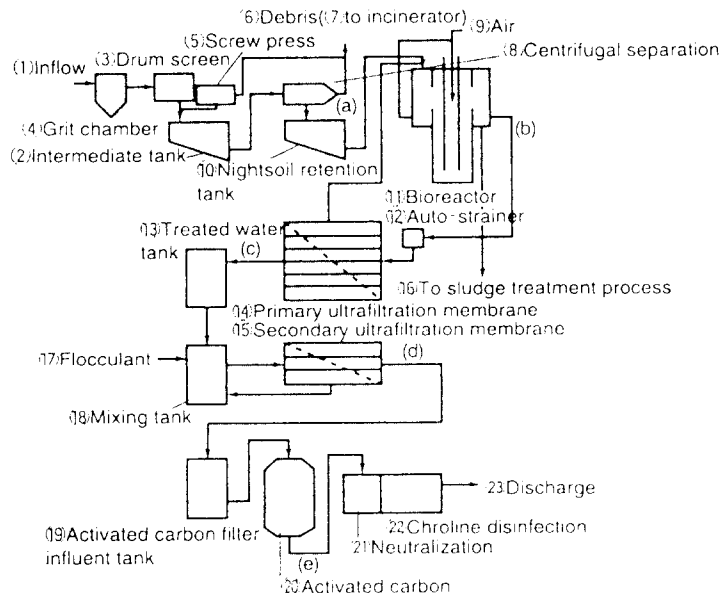


Figure 6 Schematic flow diagram of a nightsoil treatment plant

Table 3 shows membrane specifications and average performance data are summarized in Table 4.

Table 3 Membrane specifications and operating conditions

	primary UF module	secondary UF module
module type	tubular	tubular
material of membrane	polyolefin	polysulphon
molecular weight cut off	20000	10000
number of modules per system	24	12
total membrane surface,,m2	55.2	27.2
crossflow velocity, m/s	3.0	1.5
inlet pressure, kPa	410	78
outlet pressure, kPa	78	20
temperature, °C	29-39	26-37
SS, kg/m3	19-26	5-8

Table 4 Water quality at each stage of treatment ((a) to (e) refer to the same in Fig. 6)

	pH	SS mg/l	BOD mg/l	COD _{Mn} mg/l	T-N mg/l	T-P mg/l	Cl- mg/l
(a)after centrifugal separation	7.6	6700	5300	4700	2500	220	1900
(b)effluent of bioreactor	6.8	18000	18	440			
(c)filtrate of the first stage UF	7.1		2.6	330	29	0.5	1400
(d)filtrate of the second stage UF	4.6		<1	70	13	<0.1	2000
(e)effluent from activated carbon	5.0		<1	<1	3.8	<0.1	1800

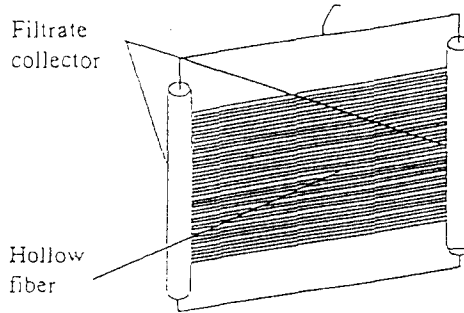
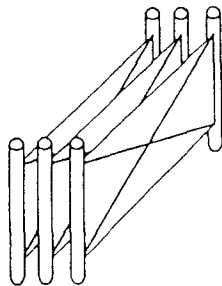
IMMERSED-TYPE MSAS FOR DOMESTIC AND INDUSTRIAL WASTEWATER TREATMENT

A new generation of MSAS is the immersed-type one where membrane modules are directly immersed in an aeration tank. This aims to tremendously reduce the energy consumption by eliminating such big circulation pumps as the conventional MSAS processes require for operation under crossflow condition that gives high energy consumption about 3 - 5 kWh/m³ of the filtrate. These values are more than ten times larger than those for sewage treatment plant operation.

A prototype using hollow fibers has been developed in laboratory scale experiments, with findings of the feasible operation of applying suction and intermittent filtration, and the potential of the process as on-site small-scale advanced domestic wastewater treatment has been shown by the author and his coworkers.^{6),15),16)} Several full scale applications using hollow fiber modules^{17),18),19)}, ceramic tubular membrane modules²⁰⁾ and plate and frame type organic membrane modules²¹⁾ have demonstrated the processes are really feasible and promising.

The following is the example of full-scale application of the immersed-type hollow fiber MSAS to industrial and domestic wastewaters¹⁸⁾. New design of hollow fiber modules have been developed and some parallel sets of the modules are vertically positioned in a aeration tank, as shown in Figure 7. Aeration is applied beneath the module sets, giving air bubbling to the membrane surfaces for washing as well as oxygen supply to microbial cells. The hollow fibers are made of polyethylene with inner and outer diameters of 0.27 and 0.41, respectively. The nominal pore size of the membrane is 0.1 μm and the membrane surface has been treated to get hydrophilic nature.

module arrangement



module sets

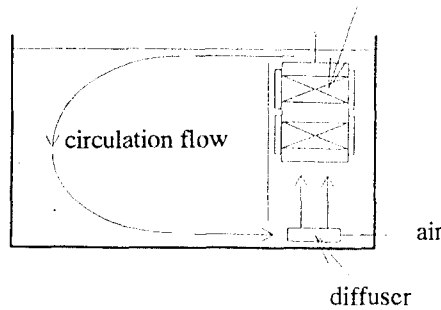


Figure 7 A newly designed immersed-type hollow fiber module and its arrangement in a aeration tank (after Futamura¹⁸⁾)

Futamura shows that his immersed-type MSAS processes only require energy consumption as low as 0.006 kWh/m³ and that it is recommended to operate the system at pressures below 30 kPa and at air to flux ratios of more than 1800, which is defined as the volumetric amount of air supply rate per unit surface of membrane divided by permeation flux¹⁸⁾. Performance of the process in treating wastewaters from food industries and household are respectively summarized in Tables 5 and 6.

Table 5 Treat ability of immersed-type hollow fiber MSAS to wastewaters from food industries

industry	cooked vegetables			sweets			bean paste(miso)		
reactor volume, m ³	50			40			7		
MLSS, mg/l	8530			9540			10000		
	Influent	Effluent	%removal	Influent	Effluent	%removal	Influent	Effluent	%removal
pH	5.4	7.4		5.5	7.6		4.4	7.6	
BOD, mg/l	2150	5	99.8	1590	1	100	3630	5.0	99.9
COD _{Mn} , mg/l	852	19.4	97.7	650	7	99.0	2320	12.6	99.5
SS, mg/l	603	n.d.	100	380	n.d.	100	1660	n.d.	100
T-N, mg/l	603	5.6	93.7	87.4	2.8	97.0	209	8.6	95.9
transparency, cm		>100			>100			>100	

Table 6 Water qualities in household domestic wastewater treatment

BOD volumetric loading, kg/(m ³ d)		0.29	
BOD sludge loading, 1/d		0.03	
MLSS, mg/l		9220	
		Influent	Effluent
water temperature, °C		17.8	19.3
pH		7.3	6.7
SS, mg/l		89	0.0
BOD, mg/l		127	0.8
CODMn, mg/l		44	3.9
Kj-N, mg/l		24	0.9
NH ₃ -N, mg/l		16	0.6
NO _x -N, mg/l		1.6	11.6
T-P, mg/l		3.2	2.0
color, deg.			17.6
coliforms, MPN/100ml			0~43

CONCLUDING REMARKS

New membrane processes such as immersed-type MSAS give a direction in minimizing the energy required to operate systems. It is obviously necessary to achieve further reduction of membrane costs for wider application of membrane process to wastewater treatment. We do not have to use the membranes with sophisticated quality control such as a grade for medical use. It would be wasteful to use them in wastewater applications. Consideration should therefore be given to the development of membrane modules with quality standards and grades that are appropriate to the requirement of wastewater treatment. In this sense, there must be considerable scope for the reduction of membrane costs. It is also clear that existing membrane modules offer little scope for merits of scale. Before the MSAS processes can be expanded for large-scale application in urban sewage treatment and other fields, it will be necessary to develop large-scale modules that offer merits of scale. On the other hand, it is also anticipated that the predominant direction of development will be toward downsizing in step with advances in remote monitoring and control technology. If this is indeed the case, it is possible that membrane separation technology, which offer reverse merit of scale, will become the dominant technology in the field of wastewater treatment.

REFERENCES

- 1) ZOSUI GIJUTU HANDBOOK, Water Re-use promotion center, 1993
- 2) T. Urase, K. Yamamoto, S. Ohgaki and N. Kamiko, *Proc. of Environ. Eng. Research*, 30, 1994
- 3) K. Nishimura, Y. Ogata, M. Arano, S. Kawamura and Y. Magara, *Proc. of Annual Meeting of JSWE*, 1994
- 4) Y. Fuchigami, K. Yamamoto, K. Asami and T. Matsuo, *Proc. of Environ. & Sanitary Eng. Res.*, 23, 1987
- 5) K. Yamamoto, K. Asami and T. Matsuo, *Proc. of 42nd Annu. Meeting, JSCE, II*, 1987
- 6) K. Yamamoto, M. Hiasa, M. Mahmood and T. Matsuo, *Water Sci. & Tech.*, 21, 1989
- 7) K. Nishimura, K. Kawamura, K. Ito and Y. Magara, *Proc. of Environ. & Sanitary Eng. Res.*, 28, 1992
- 8) Water Resources in Japan, Japan Land Agency, 1994
- 9) M. Ohtaka, *Sangyo Kikai*, November, 1989
- 10) C. Kobayashi, *Kuuki Chowa Eisei Kogaku*, 62, 1988
- 11) C. Kobayashi, *Kagaku Soti*, No.10, 1991
- 12) D. Kuwabara, personal communication, 1994
- 13) Y. Magara, *Japan J. Water Pollution Res.* 13, 1990
- 14) H. Aya, Personal communication, 1993
- 15) K. Yamamoto and K.M. Win, *Water Sci. & Tech.*, 23, 1991
- 16) C. Chiemchaisri, Y.K. Wong, T. Urase and K. Yamamoto, *Water Sci. & Tech.*, 25, 1992
- 17) N. Yamamoto, *Proc. 29th Annu. Meeting on Japan Sewage Works Association*, 1992
- 18) O. Futamura, Seminar Text on latest trend in development of MSAS, Technical Information Center, 1995
- 19) O. Futamura, K. Oonishi and H. Ikeda, *MIFS '94*, MIE Academic Press, 1994
- 20) Y. Narukami, Seminar Text on latest trend in development of MSAS, Technical Information Center, 1995
- 21) H. Ishida, S. Izumi and M. Moro, *Proc. of 2nd Annu. Meeting on Japan Society of Waste management Experts*, 1992