

## 〈研究発表〉

# Development of MBR–RO Integrated Membrane System for Wastewater Reclamation

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### Abstract

The severe water shortage in the world enhances the interest in wastewater reclamation systems. One of them is the membrane bioreactor (MBR) – reverse osmosis (RO) integrated membrane system, which has a great potential for the reclamation of both municipal and industrial wastewater to produce reclaimable water. The authors developed the advanced PVDF flat sheet MBR membrane has numerous pores with small pore size and narrow pore size distribution, making the membrane highly resistant to clogging. Additionally, the low-fouling RO membrane, which has low-fouling property with keeping water permeability against chemical and biological fouling during the operation, was developed for wastewater reclamation. By using the developed membranes, MBR–RO pilot tests were carried out for reclamation of various wastewater in China. Very excellent water quality from MBR–RO system was observed both for municipal sewage and dyeing wastewater reclamation. Also both big economic benefit and environmental benefit could be obtained through the cost analysis, highly supporting the developed MBR–RO system to be a very alternative for wastewater reclamation. Moreover, the novel MBR simulation technology was developed originally and it was proved highly reliable for predicting the actual membrane performance.

Keywords: Wastewater reclamation, MBR, RO, Integrated membrane system

## 1. Introduction

Water shortage is becoming a more and more serious problem in the world with the development of global population and industry. To conserve sustainable water resources, it is required to develop more efficient technology for wastewater treatment and reclamation. Membrane technology is considered as the most prospective technology in the 21st century for developing the wastewater treatment and reclamation system. One of the promising wastewater reclamation systems is the combination of membrane bio-reactor (MBR) followed by the reverse osmosis (RO), called MBR–RO integrated membrane system because it has many advantages compared with the conventional systems, such as better treated water quality, smaller footprint, lower sludge production, more convenient operation and maintenance<sup>1-4)</sup>.

In this study, the authors firstly developed the advanced MBR and RO membrane specialized for wastewater treatment and reclamation. Secondly, MBR–RO pilot tests were carried out by using the

developed MBR and RO membrane for the municipal sewage reclamation and dyeing wastewater reclamation. Moreover, the authors developed the novel MBR simulation technology for optimizing the MBR process design and validated the reliability of the simulation by the pilot test for the dyeing wastewater.

## 2. Advanced MBR and RO membranes

### 2.1 Features of the flat sheet MBR membrane

The flat sheet membrane construction was developed for submerged MBR because its membrane surface can be cleaned up more effectively by air scouring to avoid sludge adherence compared with hollow-fiber membrane. The flat sheet membrane consisted of polyvinylidene fluoride (PVDF) functional layer and the base layer of Polyester (PET) non-woven fabric, making this membrane highly superior in physical strength and chemical stability. Numerous small-diameter pores at the average pore size of 0.08  $\mu\text{m}$  were distributed evenly over the membrane surface with a very narrow pore size distribution, shown as Fig.1 and

Fig.2. The structure resulted in excellent water permeability and high treated water quality, making the membrane highly resistant to clogging.

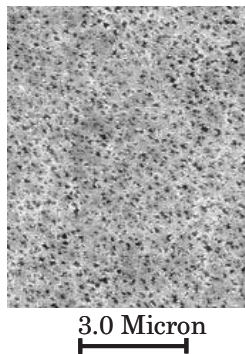


Fig.1 SEM photograph of PVDF membrane surface

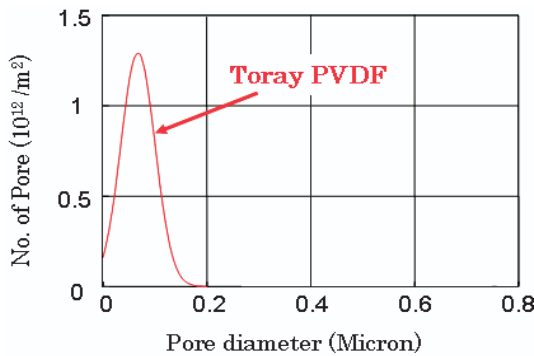


Fig.2 Pore diameter distribution

## 2.2 Features of the Low-fouling RO membrane

As a result of R&D activities, Toray has developed the low-fouling RO membrane for wastewater reclamation. The low-fouling RO membrane has the same level of pure water permeability as conventional RO membranes and also has low-fouling property with keeping water permeability against chemical and biological fouling during the operation<sup>5</sup>. To evaluate the fouling property against microbes, adsorption property of a certain hydrophobic microbe and other hydrophilic microbes were measured, shown as Fig.3.

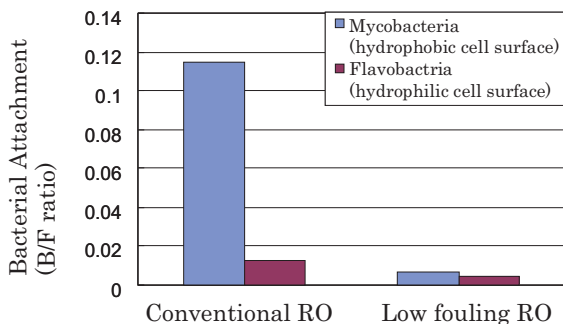


Fig.3 Fouling property against microbes for RO membranes

The hydrophobic microbe was severely adsorbed to conventional RO membranes and caused biological fouling of RO membranes. In case of

low-fouling RO membranes, the adsorption property of the hydrophobic microbe was quite low, which was less than one tenth of conventional RO membranes.

## 3. MBR-RO pilot study

### 3.1 Municipal sewage reclamation

#### (1) Objectives

The amount of municipal sewage discharge was around 30 billion Ton per year in China, meaning very great market capacity of sewage treatment and reclamation. In the pilot study, the water quality treatment efficiency as well as the membrane performance was investigated to establish the stable MBR-RO system for sewage reclamation.

#### (2) Materials and methods

The MBR-RO pilot plant, shown as Fig.4, was set and operated at a municipal sewage treatment plant in Beijing, China. The raw water was fed into the biological tanks and MBR basin and then the MBR effluent was suctioned and flowed into RO membrane for further treatment. The combined system of anaerobic – anoxic – aerobic – MBR process was designed to obtain simultaneous biological removal of carbon, nitrogen, phosphorus. The MBR unit used was the newly developed flat-sheet MBR module (TMR140-050S) with the effective membrane surface area of 70m<sup>2</sup>. RO membrane was the newly developed low-fouling RO membrane (SUL-10) with the effective filtration surface of 7m<sup>2</sup>.

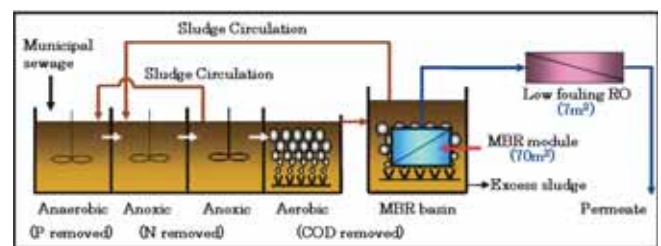


Fig.4 Schematic diagram of pilot plant for sewage reclamation

#### (3) Results and discussion

Table 1 and Table 2 showed the water quality of MBR-RO system. The MBR effluent met the Water Quality Standard for Scenic Environment Use (GB/T18921-2002), and the RO permeate sufficiently met the wastewater reclamation criteria of cooling water specified in the Code for Design of Wastewater Reclamation and Reuse of China (GB50335-2002).

**Table 1** Results of water quality treatment efficiency for MBR

Parameter	MBR influent	MBR effluent	Scenic environment use
BOD <sub>5</sub> (mg/L)	130	2.4	≤ 6
NH <sub>4</sub> <sup>+</sup> -N(mg/L)	46.9	1.2	≤ 5
TN (mg/L)	56.0	14.3	≤ 15
Turbidity (NTU)	25.1	0.21	≤ 5
LAS (mg/L)	2.1	ND	≤ 0.5

**Table 2** Results of water quality treatment efficiency for RO

Parameter	RO permeate	Reclamation criteria
Turbidity (NTU)	< 0.1	≤ 5
COD (mg/L)	< 5	≤ 60
Cl <sup>-</sup> (mg/L)	0.62	≤ 250
Hardness(mg/L)	< 10	≤ 450
TDS (mg/L)	16	≤ 1000

During the MBR continuous operation of more than 10 months, the membrane required temporal chemical cleaning in-place (CIP) by using the chemicals of 0.3% NaClO with contact time of 2 hr several times, but confirmed successful MBR operation under the very high flux at 0.8m/d even under low temperature of 10-15 degree C.

For the RO operation, the RO permeability was very stable without any chemical cleaning under the flux at 21 L/m<sup>2</sup>/h and recovery ratio at 40% with the intermitted sulfuric acid addition, suggesting neither chemical fouling nor bio-fouling occurred on the low fouling RO membrane.

### 3.2 Dyeing wastewater reclamation

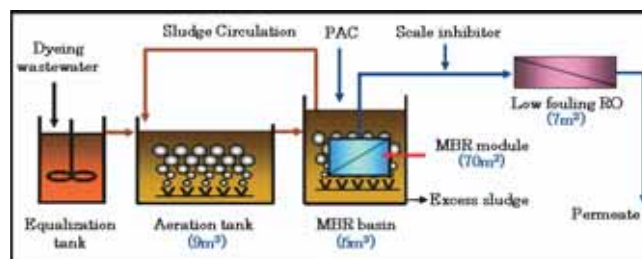
#### (1) Objectives

The textile industry is one of the most important industries in China. More than 80% textile wastewater was discharged by dyeing manufacturing facilities. Generally, the dyeing wastewater has a very low ratio of BOD<sub>5</sub>/COD and heavy color, and becomes one of the most difficult

wastewaters to be treated in China<sup>6)</sup>. Moreover, the discharge standards for dyeing wastewater treatment are more and more strict recently. Therefore, the authors tried to apply MBR-RO system for the dyeing wastewater reclamation through the pilot test in Nantong, China.

#### (2) Materials and methods

The schematic process of the pilot plant for dyeing wastewater reclamation was shown as Fig.5. The raw water used in this pilot test was taken from the neutralization tank of existed wastewater treatment plant. Poly Aluminum Chloride (PAC) was dosed to MBR basin to increase water treatment efficiency during the operation period. And scale inhibitor was added to the RO feed water for protecting RO membrane from scaling. The types of MBR unit and RO membrane used were TMR140-050S and SUL-10, respectively.



**Fig.5** Schematic diagram for dyeing wastewater reclamation

#### (3) Results and discussion

Table 3 showed the water quality of MBR-RO system. The water quality of MBR treated water could undoubtedly meet the updated Discharge Standards of Pollutants for Dyeing and Finishing of Textile Industry. Throughout the RO operation, the water quality of RO permeate was very excellent and sufficiently met the standard of water for dyeing production recommended by Dyeing Association in China. The Toray low-fouling RO membrane achieved greater than 99.0% of salt rejection in conductivity for all samples.

**Table 3** Results of water quality treatment efficiency for MBR-RO system

Parameters	Influent	MBR effluent		RO permeate	
		Measurement	Discharge criteria	Measurement	Reclamation criteria
SS (mg/L)	100-300	Not determined	≤ 70	Not determined	≤ 10
COD <sub>Cr</sub> (mg/L)	600-1500	35-65	≤ 80	< 5.0	--
BOD <sub>5</sub> (mg/L)	80-250	< 5	≤ 20	Not determined	--
Color	40-120	10-20	≤ 60	1	≤ 10
Total Hardness (mg/L)	50-300	50-300	--	< 10	≤ 100
Conductivity (μS/cm)	700-2000	700-2000	--	< 50	≤ 25

The cost analysis was made to know the benefits for performing dyeing wastewater. The industrial water consumption of the dyeing manufacturer was

6000m<sup>3</sup>/d. And the wastewater recovery of the MBR-RO system was set at 70%. As a result, not only big economic benefit with around 3.5 million

RMB/year total cost-cutting but also great environmental benefit with 70% reduction of total pollution discharge can be obtained.

#### 4. Validation of MBR simulation

The novel MBR simulation technology was developed originally to optimize the MBR process design, contributing to energy saving and cost reduction. The basic mechanisms were shown as below equations (Fig.6), of which the membrane fouling mainly induced by cake formation and pore clogging.

**< Cake formation rate >**

$$\frac{dX_c}{dt} = X \cdot J \cdot \frac{K_{\tau 1}}{K_{\tau 1} + \tau} - \gamma \cdot \tau \cdot \frac{K_{\tau 2}}{K_{\tau 2} + \Delta P} \cdot \eta \cdot X_c^2$$

Attachment                  Detachment

$$R_c = \alpha_2 \cdot (1 + \alpha_1 \cdot \Delta P) \cdot X_c$$

$\alpha_2$ : Cake resistance parameter  
 $\gamma$ : Detachment parameter  
 $\alpha_1$ : Sensitivity of pressure to resistance  
 $X$ : Amount of biomass

**< Membrane clogging rate >**

$$\frac{dX_f}{dt} = \psi \cdot (1 - \varepsilon \cdot X_f^2) \cdot \Delta P$$

Driving force is TMP

$$R_f = \beta \cdot X_f^{1.5}$$

$\beta$ : Specific resistance parameter for clogging  
 $\psi$ : Transferring rate into membrane pores  
 $\varepsilon$ : Auto-inhibition of pore clogging

**< TMP calculation >**

$$\Delta P = \frac{\Delta P_c}{J} + \frac{\Delta P_f}{J} + \frac{\Delta P_m}{J} = \mu \cdot J \cdot (R_c + R_f + R_m)$$

cake   clogging   membrane

$J$ : Flux    $P$ : Pressure    $R$ : Resistance  
 $\mu$ : Viscosity of permeate water

Fig.6 Equations of MBR simulation technology

The reliability of the simulation was validated by the MBR pilot test for treating dyeing wastewater, shown as Fig.7. MBR could be operated stably when the actual operation flux was below critical flux (the maximum flux to be able to conduct the stable operation) calculated by the simulation soft, and the Trans-membrane pressure (TMP) increased as the actual flux was higher than critical flux. Therefore, the simulation was proved very highly reliable for predicting the actual membrane performance.

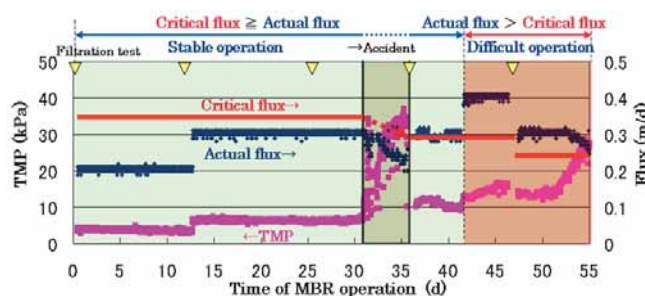


Fig.7 Validation of MBR simulation by the MBR pilot test

#### 4. Conclusions

The authors developed the advanced PVDF flat sheet MBR membrane, which has numerous pores with small pore size and narrow pore size distribution, making the membrane highly resistant to clogging. Additionally, the low-fouling RO membrane, which has low-fouling property with keeping water permeability against chemical and biological fouling during the operation, was developed for wastewater reclamation.

By using the newly developed membranes, MBR-RO pilot tests were carried out for reclamation of various wastewater in China. Very excellent water quality from MBR-RO system was observed both for municipal sewage and dyeing wastewater reclamation. Also both big economic benefit and environmental benefit could be obtained through the cost analysis, highly supporting the developed MBR-RO system to be a very alternative for wastewater reclamation. Moreover, the novel MBR simulation technology was developed originally, and it was proved highly reliable for predicting the actual membrane performance.

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