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Permeation Characteristics of Wastewater Containing Si Fine Particles through Ultrafiltration

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Abstract: The permeation characteristics of the wastewater containing Si fine particles were examined by ultrafiltration using the polyolefin tubular membrane module. Flux with time was due to the growth of Si cake deposited on the membrane surface and the pore plugging by fine particles. The rate of flux decline in the initial stage increased with the trans-membrane pressure. The pore blocking resistance was the dominant resistance at the initial period of filtration and the cake resistance began to dominate with the initial pore blocking resistance. The larger pores compared with the fine particles, the more the membrane pores could be blocked by the fine particles. Before and after treatment, the distribution of particle size was shifted toward to the left. Then, the average size of fine particles in the permeate was 20 nm.

Keywords: *ultrafiltration, polyolefin, colloidal solution, tubular module, cake filtration model*

1. Introduction

The ultrafiltration process are widely used in many industries such as food industry, pharmaceuticals, wastewater treatment and semiconductors. One of the major bottlenecks in ultrafiltration of colloidal solution such as that of Si is fouling, which exhibits sudden flux decline. The ultrafiltration separation technique is a very effective method for separation of the colloidal fine particles from colloidal solutions. Their technologies has been applied for the ultrapure water and the wastewater recycling system[1,2].

The ultrafiltration system is definitely superior for separation and purification performances of the colloids or the colloidal solid particles, but the disadvantage is the higher deviation of permeation flux decline. The phenomena on the flux decline result from the complicated effect on resistances of the membrane, osmosis, concentration polarization, cake-layer, and adsorption layer, etc. Particularly, the main causes on the flux decline in the colloidal solution are the concentration polarization in according to solute particles and the membrane fouling. The concentration polarization occurs rapidly within a short time, but the membrane fouling makes an appearance continuously for the operating time.[3] The major causes of the membrane fouling are the interaction between the separable objectives and a membrane material, the relations of the permeate particle size and the membrane pore size, the membrane types, and the operating condition. As the membrane fouling decreases the durability and the treating efficiency of membrane, it is important to analyse and control the affecting factors on fouling[4,5].

Fouling can be classified into two types according to its location; one is the surface fouling which occurs on the surface of membrane (the build up of cake layer) and the other is the internal fouling which occurs on the surface of membrane pores (pore plugging and adsorption). These membrane fouling is possible to decrease by the

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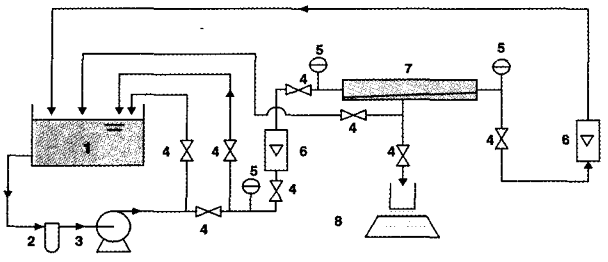

Fig. 1. Schematic diagram of the experimental apparatus.

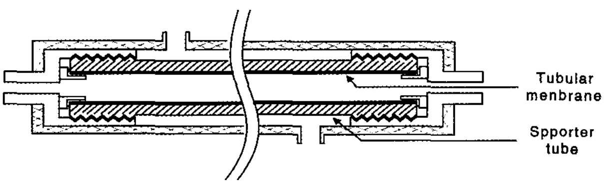

Fig. 2. Schematic diagram of tubular membrane module.

change of the membrane formation, module types, the feeding solution, and the operating types[6-8].

This work, therefore, was focussed on the investigation of the permeation characteristics of a colloidal solution containing Si fine particles through ultrafiltration using the tubular membrane module and to analyze the fouling mechanism.

2. Experimental

The experimental apparatus and the membrane module used in this study are illustrated in Figure 1 and Figure 2, respectively. The ultrafiltration system consists of the raw wastewater tank, feeding pump, and tubular membrane module. The operation was performed at 25°C, the feed flow rate of 0~4 L/min and the applied pressure of 0~3 kg_f/cm^2. The flow pattern in membrane module adopted 2 types of cross and dead-end flow. The permeate flux was measured on every 5 min, and the concentration of feed and permeate measured on every 10 min. The Si concentration of feed and permeate was determined by AA spectrophotometer (Shimadzu 6701, Japan) and turbidimeter (Oberco 965, USA), and the size distribution of Si fine particles in wastewater was measured by particle size

Table 1. Analytical data of wastewater containing Si fine particles

Analytical item	Range
pH	6.8 ~ 7.2
Turbidity(NTU)	~ 450
Si fine particles(mg/L)	~ 5.6
Average particle size(μm)	0.196
Suspended solid(mg/L)	5.8 ~ 6.4

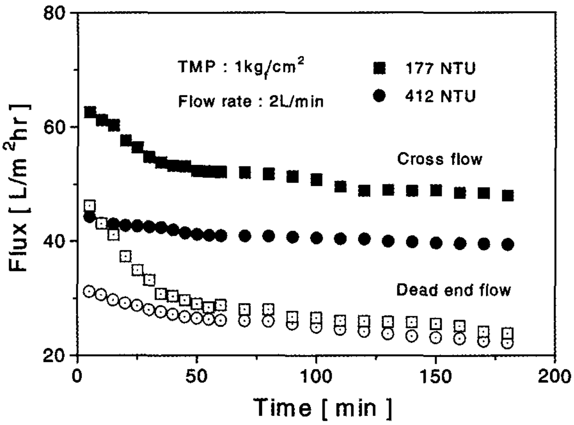

Fig. 3. Effect of permeate flux with operating time and flow patterns.

analyzer(Melburn, Zetasizer-1000, UK).

The membrane used in this work was a commercial poly olefin ultrafiltration tubular membrane(ID : 1.15 cm, MWCO : 20,000) manufactured by Nitto Denko, Japan. The constituents of wastewater used was presented in Table 1. The concentration of the ionic substances or the metallic constituents, such as Na, K, Ca, etc., was below the level of ppb in wastewater used. The COD was below 1 ppm and 10 ppb as the TOC.

3. Results and Discussion

The flux decline through ultrafiltration is an important factor. The main causes of the flux decline are the performances of treated water, the membrane characteristics, the interaction of membrane and treated water, and the operating condition, etc., and yet it is affected with the flow patterns to be introduced in process system. As shown in Figure 3, the flux decline with time was observed for both the dead-end

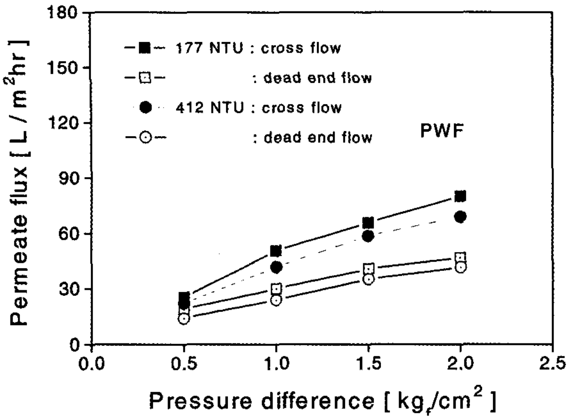

Fig. 4. The variation of relative permeate flux with the trans-membrane pressure.

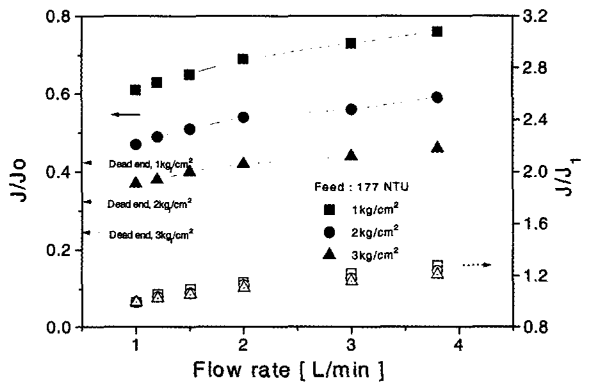

Fig. 5. Effect of feed flow rate on the relative flux. (J_0 : pure water flux, J_1 : permeate flux at 1 L/min feed)

and the cross flow. The phenomena of flux decline is similar for both patterns, but the rate of decline is more significant for the dead-end flow type. This is regarded as due to accumulation of the fine particles both rejected on the membrane surface and adsorbed in membrane pores. For the initial turbidity of 177 NTU, the flux decreased to about 75% in cross flow and 50% in dead-end flow. In Figure 4, the variation of relative permeate flux with the trans-membrane pressure is presented and the initial turbidity of wastewater and the flow patterns are compared. The flux of pure water is normally increased with the trans-membrane pressure. For wastewater containing Si fine particles, the increasing rate of the permeate flux grows smaller compared with pure water. Also, it can be seen that on the initial concentration of wastewater,

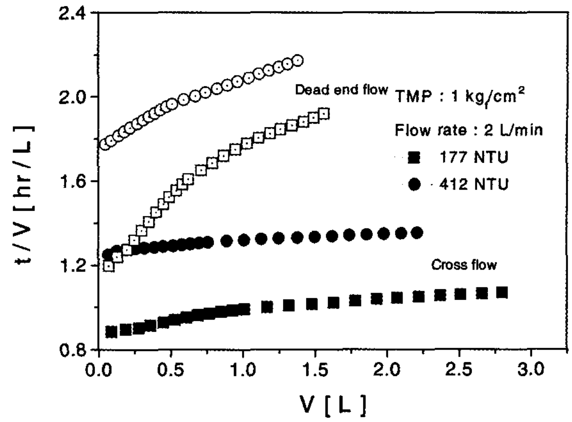

Fig. 6. The model analysis of cross and dead-end flow with the different NTU of feed.

the permeate flux depend on trans-membrane pressure. Figure 5 shows the effect of feed flow rate on the relative flux compared with the pure water flux and the permeate flux.

Figure 6 illustrates the plot of t/V vs V on both flow patterns for showing linearity. These results indicate that cake formation on the membrane surface may be a dominant mechanism. Especially, as the filtration process starts, Si fine particles over the pore size of membrane should be rejected and accumulated on membrane surface such as the cake filtration model. An empirical equation of the cake filtration model is as followed[9];

$$\frac{t}{V} = \frac{k_c}{2} V + \frac{1}{Q_0} \tag{1}$$

where t is time, V is volume, k_c is a generalized model, and Q_0 is flow rate. Therefore, the cake of Si fine particles can grow gradually, and the cake layer is thickened. The slope variation of the t/V vs V lines for the dead-end flow and the cross flow indicates the difference in specific resistance of cake. The resistance is obtained from the slopes of these lines. The intercept of the t/V axis shows the membrane resistance to be calculated. In case of the dead-end flow, the initial deviation from linearity suggests an initial pore plugging. The larger pores compared with the fine particles, the more the membrane pores can be blocked by the fine particles. The specific resistance of the dead-end flow

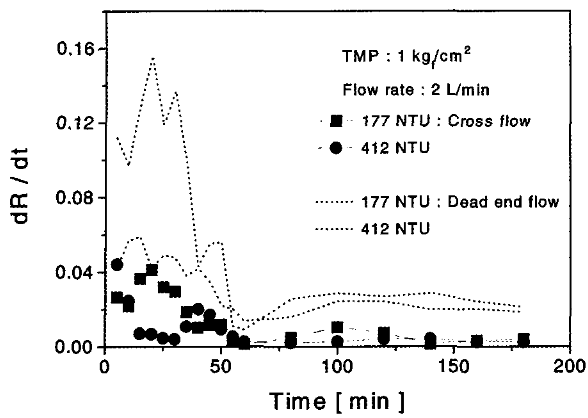

Fig. 7. Cake filtration model and resistance gradient analysis with the different NTU of feed.

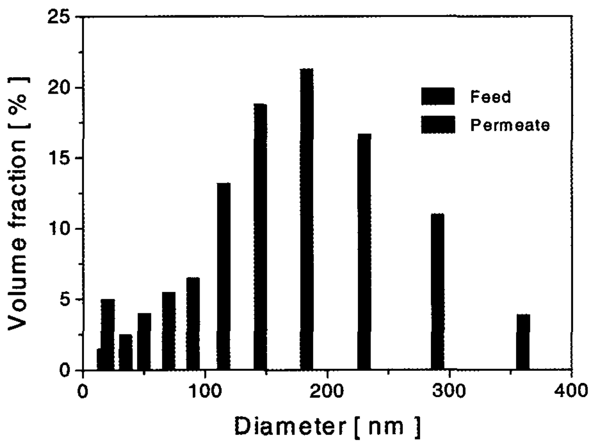

Fig. 8. Particle size distribution of the wastewater feed and the permeate.

is higher than that of the cross flow. Figure 7 presents the cake filtration model and the resistance gradient analysis with the different initial feed concentration. The cake of filtered fine particles for the dead-end flow was more compressed than that for cross flow. The thickness of filtering cake for the dead-end flow was thicker than that for the cross flow. The thickness of cake includes the depth of penetration of fine particles to the membrane pore. The rejected fine particles were accumulated on the membrane surface with blocking the entrance of membrane pores. Figure 8 shows the distribution of fine particle size in the wastewater feed and the permeate. Before and after treatment through ultrafiltration, the distribution of particle size is shifted toward to the left, and the size of fine particle in the permeate was below 20 nm. And, the fine particle was removed about 98% through ultrafiltration by tubular membrane module.

4. Conclusions

The permeation characteristics of a wastewater solution containing Si fine particles were studied to investigate and analyze the fouling mechanism through ultrafiltration using the tubular membrane module.

(1) The rate of flux decline in the initial stage through ultrafiltration increased with the trans-membrane pressure.

(2) The pore blocking resistance was the dominant resistance at the initial period of filtration, and the cake resistance began to dominate with the initial pore blocking resistance. The larger pores compared with the fine particles, the more the membrane pores can be blocked by the fine particles.

(3) Before and after treatment through ultrafiltration, the distribution of particle size is shifted toward to the left. Then, the average size of fine particles in the permeate was 20 nm.

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