

## Effect of Ultrasound on Membrane Cleaning Processes for Enhancement of its Permeability

Kyaing Kyaing Latt, Tsuyoshi Kobayashi, Takaomi Kobayashi

Department of Chemistry, Nagaoka University of Technology, 1603-1 Kamitomioka, Nagaoka, Niigata, Japan.

Fax: 81-258-47-9300, e-mail: takaomi@nagaokaut.ac.jp

In the present study, polyethylene hollow fiber microfiltration (MF) membrane with 0.4  $\mu\text{m}$  pore size was fouled by 1 wt % aqueous milk solution. The decreased permeation rate due to plugging and adsorption of rejected milk particles on the membrane was removed by the ultrasonic irradiation and the permeate flux was enhanced by various ultrasonic cleaning processes. Ultrasound effect on the fouled membrane was varied at different frequencies of 28, 45 and 100 kHz. It was found that the direction of the ultrasound irradiation was one of important factors in the efficient cleaning process by sonochemical effect.

Key words: Membrane filtration, Microfiltration membrane, Ultrasound, Fouling

### 1. INTRODUCTION

Membrane technology is an emerging technology because of its multi disciplinary characters and is used in a wide range of application in all industrial areas. Among separation and purification processes such as adsorption, evaporation, distillation and ion-exchanging etc., membrane separation process could progress the purified quality of permeate and separate the undesirable impurities based on membrane property used. But one of the disadvantages in ultrafiltration and MF processes using porous membrane is membrane fouling which leads to flux reduction. Fouling depends on physical and chemical parameters such as concentration of effluent, temperature, pH, ionic strength and specific interactions between foulants and membrane [1-4]. Most of research and literatures focused on cleaning the fouled membrane rather than to control fouling. The cleaning techniques in membrane separation process are by hydraulic, mechanical, chemical and electrical methods.

Moreover, ultrasound has also been widely used as a method for cleaning material because of its cavitation phenomenon. Cavitation is due to formation and growth implosion collapse of bubbles. Based on the cavitation effect, many studies were carried out for cleaning fouled membrane and enhancement of permeability in ultrasonic field. When ultrasound is transmitted through the liquid medium, alternate compression and expansion cycles occurs and causes the bubbles to collapse and remove the foulant from membrane surface[2]. The typical cleaning methods for tubular membrane as like hollow fiber membranes are backwashing with water and forward flushing under ultrasound. We found that irradiation of ultrasound was easy to remove the foulants from the membrane and the ultrasound intensity and frequency influenced the increasing in permeate flux of membrane [8]. In the present study, ultrasound was irradiated at frequencies of 28, 45 and 100 kHz with power intensity range of 2.5-3.3  $\text{W}/\text{cm}^2$ , when 1 wt% aqueous milk solution was permeated through polyethylene hollow fiber membrane with 0.4  $\mu\text{m}$  pore size. Ultrasound effect on the MF

membrane was examined.

### 2. EXPERIMENTS

#### 2.1 Materials and instruments

The membrane modules used in this experiment were hollow fiber MF membranes made by polyethylene (PE) and products of Mitsubishi Rayon. The diameter and length of each fiber of the membrane were 0.8 mm and 80 mm, respectively. The total surface area of the hollow fiber membrane contained bundle of 512 fibers was 0.1  $\text{m}^2$  and the pore size of membrane was 0.4  $\mu\text{m}$  diameter. Membrane filtration process was done in a stainless steel tank (0.2  $\times$  0.2  $\times$  0.25  $\text{m}^3$ ) mounted seven numbers of piezo electric ultrasonic transducer with 3cm diameter. These transducers were connected with ultrasonic multi-cleaner having 23  $\text{W}/\text{cm}^2$  output and 28, 45, 100 kHz frequency (W-115, Honda electronics Co., Ltd.). The sound pressure emitted from the transducers was estimated by using pulse receiver (Model 5058PR) that was connected with immersion transducer probe (PANAMETRICS V301). The pump used for filtration process was circular pump that was product of Millipore Company (Model XXX80) and operated under 60 kPa with 100 ml/min.

#### 2.2 Experimental procedure

Filtration process was carried out in the absence and presence of ultrasound irradiation with different frequencies. We examined the effect of cleaning methods, position of membrane and the distance between membrane and ultrasound transducers on the cleaning efficient. Aqueous milk solution with 1 wt% concentration (8 liter) was prepared in stainless steel tank and the PE membrane was immersed in the milk solution. The permeated flux of the membrane was measured by using volume flow meter, which was detecting the solution volume permeated through the membrane per unit area and connected recorder recorded by time dependence. The unit of volume flux was expressed as  $\text{m}^3/\text{m}^2\text{s}$ . During the filtration experiments, permeate was recycled to the feed tank as shown in Fig.1.

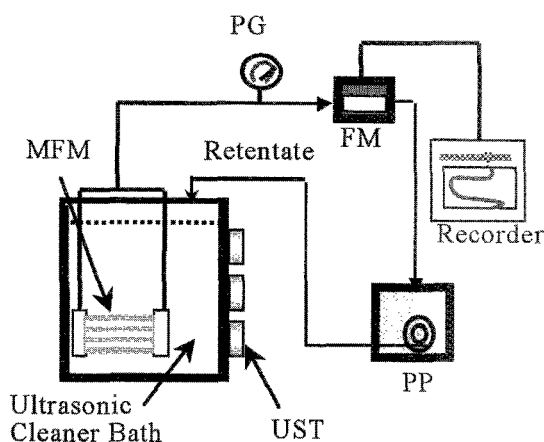


Fig.1 Schematic flow diagram for ultrasound filtration process of 1 wt% aqueous milk solution.

PG : Pressure gauge, FM : Flow meter, MFM : Microfiltration membrane, PP : Peristaltic pump, UST : Ultrasound transducers

1 wt% aqueous milk solution was filtrated by passing through PE membrane in the absence of ultrasound irradiation. The volume flux of permeate was gradually decreased and occurred fouling the membrane due to the rejected milk particles within 30 min. When the membrane was fouled after 30 min, continuous milk filtration was carried under ultrasound irradiation with different frequencies of 28, 45, 100 kHz. For the experiments of water cleaning process, we changed the feed water instead of milk solution after 30 min of fouling and cleaning process was operated under ultrasound. The fouled membrane was cleaned by backwashing with and without ultrasound and also forward flushing under ultrasound. Then, we observed the permeate flux by changing the ultrasound direction onto fouled membrane that was placed horizontal (parallel) and vertical position with ultrasound irradiation.

### 3. RESULTS AND DISCUSSION

#### 3.1 Ultrasound enhanced permeability by varying frequency

Before we started the ultrasound associated experiments of milk solution, water permeation was performed through the PE hollow fiber membrane in the absence and presence of ultrasound. We confirmed that, there was no change in water flux and was constant at  $2.3 \times 10^{-4} \text{ m}^3/\text{m}^2\text{s}$  under ultrasound. For the permeation of milk solution, the volume flow rate of permeate was gradually declined from starting flow rate,  $2.2 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$  to  $4 \times 10^{-6} \text{ m}^3/\text{m}^2\text{s}$  in the range of the 30 min filtration period, because membrane was fouled. Then, we irradiated ultrasound on the fouled membrane with different frequencies of 28, 45, 100 kHz and operated continuous filtration of milk effluent for additional 60 min. The changes of permeate volume flux under ultrasound field with various frequencies were compared with that of without ultrasound (Fig.2). The volume flux was slowly decreased and became nearly constant at  $3.5 \times 10^{-6} \text{ m}^3/\text{m}^2\text{s}$  through the whole process when filtrated without ultrasound. For cases of

permeation under ultrasound, the increase permeate flux occurred within 20 min after the ultrasound irradiation. In 28 kHz case, the volume flux was  $1.8 \times 10^{-4} \text{ m}^3/\text{m}^2\text{s}$  at 90 min. We observed that 45 kHz frequency also increased the permeability, but the volume flux was lower than that of 28 kHz case. In 100 kHz case, volume flux changes were not so much difference with the permeation without ultrasound and fouling was still occurred.

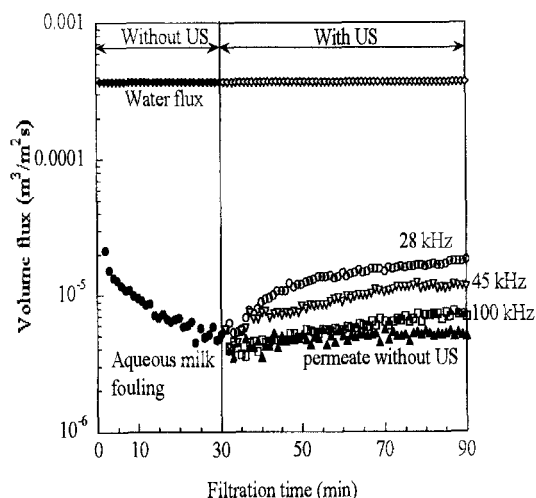


Fig.2 Volume flux changes of 1 wt% milk solution in the absence and presence of US irradiation with different frequencies of 28, 45 and 100 kHz. Permeated water flux was also plotted.

By comparing the resultant values of volume flux, low frequency ultrasound such as 28 kHz was able to enhance permeate flux. That was due to cavitation phenomenon occurred in ultrasonic water bath. It was reported that the formation of cavity bubble grows during compression and expansion cycles of ultrasound wave. In high frequency, ultrasound effect in acoustic streaming is generated by destruction of a large number of cavity bubbles, because of sufficient energy to overcome the interaction between foulant and membrane. Therefore, the permeate volume flux was decreased in high frequency.

To measure the milk solid contents in the permeate water after the ultrasonic operation, we measured absorbance by using UV spectrophotometer in the range of 250-400 nm. The values of the absorbance before and after permeation were about 2 and 0.002. This confirmed that there was no included milk particles in permeate solution.

#### 3.2 Ultrasound enhanced permeability by changing ultrasound directions and positions on to fouled membrane

In order to explain the ultrasound effect, the following water cleaning experiments were carried out with different directions of ultrasound on fouled membrane. The fouling process was the same in both cases. After fouling, the feed solution was changed with water instead of milk solution and then started the cleaning

process under ultrasound irradiation with 28 kHz frequency for 60 min. The volume flux of the permeate solution was determined by irradiating ultrasound in perpendicular and parallel direction on the fouled membrane (Fig.3). The MF module was placed at 2 cm and 6 cm from ultrasound transducers (UST) as shown in Fig.3 (a) and (b). Fig.3 (c) is for ultrasound irradiation on fouled membrane in parallel direction at 6 cm position.

After 60 min water filtration, the permeate flux increased to  $9.8 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$  for (a),  $6.6 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$  for (b) and  $4 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$  for (c) configuration. Then, the value of permeate flux were gradually declined and became constant as  $4.4 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$ ,  $2.8 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$  and  $2.0 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$  respectively.

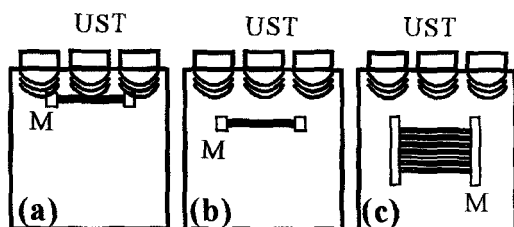


Fig.3 Top sectional view of ultrasonic water bath with different membrane position. Membrane was placed vertical position at (a) 2 cm (b) 6cm from UST and was irradiated 28 kHz frequency ultrasound in perpendicular direction. In (c) configuration, the membrane was placed horizontal position at 6 cm from UST, when irradiated ultrasound in parallel direction. M: hollow fiber membrane

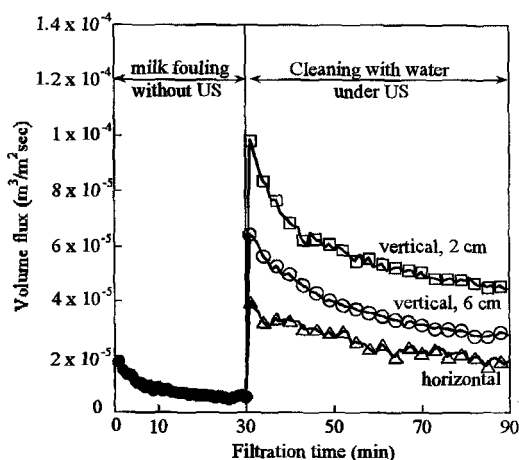


Fig.4 Changes of volume flux in water cleaning process of fouled MF membrane with different directions of 28 kHz frequency ultrasound irradiation.

Through these experiments, the 2 cm configuration of MF membrane was the highest in the enhanced permeability. The case of horizontal positioned membrane was not efficient in the enhanced permeability. Therefore, it was suggested that ultrasound irradiation effected on vertical positioned configuration. That is, the contacted area with

ultrasound was large in perpendicular direction. In the distance between membrane and the transducers, the permeate flux of the 2 cm case was higher than that of 6 cm one because of strong acoustic pressure of ultrasound. In near transducer rather than the far position, as an evidence for permeability, the ultrasound intensity at both 2 cm and 6 cm was measured at 28 kHz frequency when operated the water cleaning process on fouled membrane (Fig.5). The transducer probe was immersed in water bath and received the emitted US sound pressure by this probe. We observed that the ultrasound intensity at 2 cm was higher than 6 cm and the amplitude differences between two cases. That is, the results suggested that ultrasound pressure measured at 2cm was higher than that at 6 cm. This was reasonable for the explanation of the distance effect on the enhanced permeability.

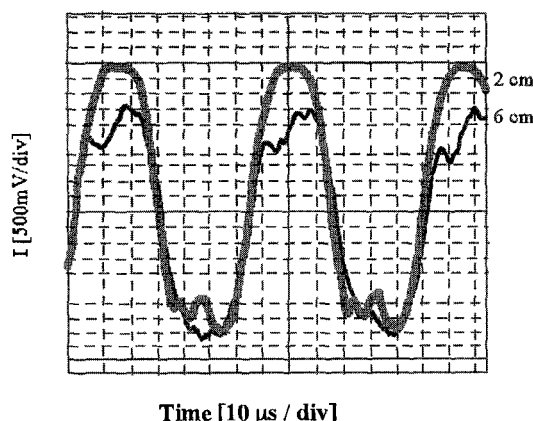


Fig.5 Time profiles of sound pressure observed in ultrasonic water cleaning process at 2 cm and 6 cm.

### 3.3 Recovery of permeate flux by different cleaning methods

Effect of membrane cleaning was investigated by three cleaning methods such as backwashing with and without US and forward flushing under US. These cleaning methods are illustrated in Fig.6. Fig.7 shows changes of volume flux observed in backwash and forward wash of the fouled membrane. The membrane fouled by 1 wt% aqueous milk solution was cleaned in water by using above three methods for 10 min cleaning period. After each cleaning process was applied, the membrane was used to filter water again and then, the permeate flux was determined for following 60 min.

It was noted that backwashing and forward flushing were useful to cleaning especially for the MF membrane. In these experiments, all of cleaning methods could remove fouling occurred on the membrane. We found that by using ultrasound associated cleaning processes were more effective than that operated in the absence of ultrasound. As seen, the efficiency of membrane cleaned by forward flushing and backwashing with US was about 1-1.5 times higher than that of backwashing without ultrasound, when water was permeated for 60 min. Volume flux of cleaned membrane increased by backwashing without ultrasound to  $1.1 \times 10^{-4} \text{ m}^3/\text{m}^2\text{s}$  and then declined to  $7 \times 10^{-5} \text{ m}^3/\text{m}^2\text{s}$  within 20 min as water permeated. For the membrane cleaned under

ultrasound, the value was increased to  $2.3 \times 10^{-4} \text{ m}^3/\text{m}^2\text{s}$  and the permeate flux of water was maintained until to the end of 60 min.

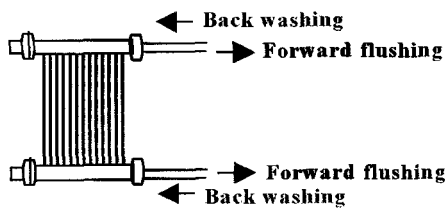


Fig.6 Illustration of water cleaning processes of fouled polyethylene hollow fiber MF membrane with  $0.4 \mu\text{m}$  pore size.

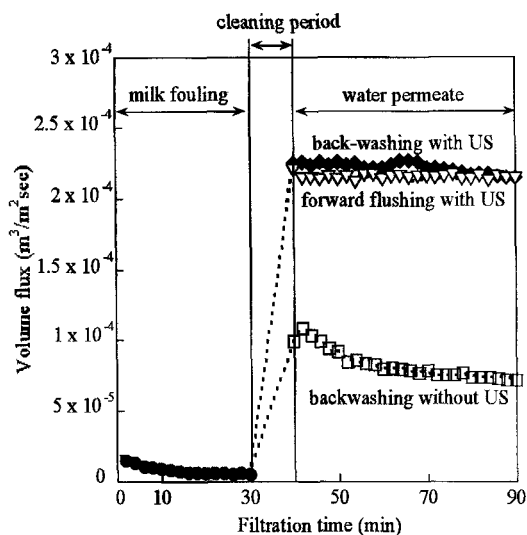


Fig.7 Changes of volume flux of water permeation after cleaning the fouled membrane with different methods for 10 min.

#### 4. CONCLUSION

We observed that 28 kHz frequency was able to enhance permeability for hollow fiber MF membrane fouled by 1 wt% aqueous milk solution. Because ultrasound caused remarkable cavitation phenomenon with large amount of cavity bubbles in the frequency field to be effective in preventing fouling and enhanced permeability was observed. Enhanced permeate flux was also dependent of direction of ultrasound irradiation. Than, cleaning process of fouled membrane operated under ultrasound was more effective than without ultrasound.

#### REFERENCES

- [1] M. Mulder, "Basic Principles of Membrane Technology", Kluwer Academic Publishers, Dordrecht, (1996) pp. 154-60.
- [2] Jianxin Li, R.D.Sanderson, E.P. Jacobs, J. Membr. Sci., 205, 247-57 (2002).
- [3] T.Kobayashi, T.Kobayashi, N.Fujii, Jp. J. Appl. Phys., 39, 2980-81 (2000)
- [4] X. Chai, T. Kobayashi, N. Fujii, Separation and Purification Technology, 15, 139-46 (1999).

[5] T.Kobayashi, T.Kobayashi, Y.Hosaka, N.Fujii, Ultrasonic, 41, 185-90 (2003).

[6] T.Kobayashi, Y.Hosaka, J. Appl. Phys., 42, 2954-2955 (2003).

[7] R.D. Noble, S.A. Stern, Membrane Separation Technology Principles and Applications, Elsevier, Amsterdam, (1995) pp. 70.

[8] T.Kobayashi, X.Chai, N.Fujii, Separation and Purification Technology, 17, 31-40 (1999).

(Received October 13 2003; Accepted March 31, 2004)