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Use of microbubbles to reduce membrane fouling during water filtration

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ABSTRACT

The present study explores how microbubbles can improve membrane filtration by preventing membrane fouling. Microbubbles (MBs) 10–50 μm in diameter were generated by pumping the feed solution with air bubbles through a slit under high shear stress. Filtration of raw river water at pilot-plant scale confirmed that adding MBs to the feed solution does indeed reduce membrane fouling. The effect of MBs on the flux was examined under various conditions of filtration flux and experimental duration. MBs reduced fouling over both short and long (>1 month) time frames, and were also effective for cleaning the membrane. Thus, incorporating MBs into water for membrane filtration may reduce the costs and energy consumption associated with water treatment.

Keywords: Microbubble; Ultrafiltration; Hollow fiber membrane; Filtration flux; Membrane fouling; Cellulose acetate

1. Introduction

Membrane filtration has been increasingly applied for treating drinking water, industrial water, and wastewater [1,2]. Membrane filtration methods such as ultrafiltration and microfiltration, which can almost completely remove fine particles, colloids, and bacteria, can yield high-quality treated water with relatively low energy consumption. One drawback of membrane filtration, on the other hand, is the difficulty in maintaining the membrane filtration flux over long-time periods due to membrane fouling. In drinking water treatment, membrane fouling involves the adsorption of substances in raw water, including suspended inorganic particles, bacteria, viruses, and organic molecules (e.g. humic substances). These foulants may either coat the inner surface of the membrane, or plug the pores altogether [3–8]. The deposited substances eventually form a gel or cake layer on the membrane, providing the dominant resistance toward water filtration. To avoid a decline in membrane flux, it is necessary to periodically clean the membrane, usually by harsh physical processes such as high cross-flow velocity, strong membrane backwashing, and air scrubbing, and also to perform frequent chemical cleaning of the membrane. Membrane fouling increases energy consumption of the filtration operation, and increases the cost of the treatment because in addition to the energy consumption of the filtration itself, chemical cleaning and membrane replacement also introduce costs.

In this work, we introduce a novel water treatment technology incorporating microbubbles (MBs) in a

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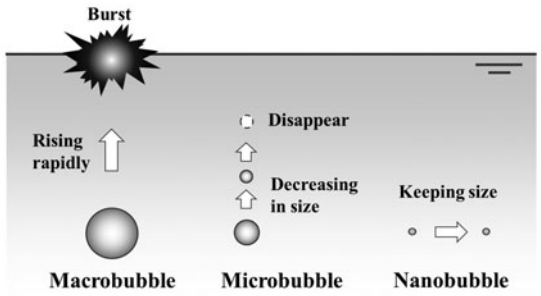

Fig. 1. Differences in the behavior of MBs and macrobubbles [10].

membrane filtration system to reduce membrane fouling. MBs are very fine bubbles whose diameters are about 10–50 μm. Takahashi et al. [9–11] have reported differences in the behavior of normal macrobubbles and MBs. While the former rapidly rise to the water surface and burst, the latter are stable for longer time periods underwater (Fig. 1). The MBs gradually decrease in size because of the dissolution of interior gases into the surrounding water, and the ζ potential of MBs in distilled water increases from −35 to −60 mV as the MBs shrink.

Recently several groups have reported on how MBs influence the cleaning of metals, plastics, and human skin, as well as biological water treatment [12–14]. Ozone MBs have been investigated for wastewater treatment under acidic conditions in the absence of dynamic stimulus [15], where treating polyvinyl alcohol solutions caused the total organic content of the solutions to decrease with treatment time. This result suggested that collapsing MBs can induce chemical reactions which can decompose organic molecules.

While MBs have attracted recent research attention, to the best of our knowledge no research has yet been done on their applications in membrane filtration. In this paper, we first introduce MB into a membrane filtration system, and investigate the fouling behavior of the system with and without MBs during the long-term filtration of river water in a pilot plant.

2. Experimental

2.1. MB generation

The MB generator (Pumpaerator, Teikoku Electric Mfg Co., Ltd), a stainless steel disc of 11 cm in diameter, was set at the outlet of the filtration pump. Air was supplied at the inlet of the pump by suction, and by controlling the volume with a valve. The MBs were generated by pumping water containing air bubbles into the accurately controlled slit of 0.2 mm in width under high shear stress.

2.2. Monitoring production and size of MBs

The MBs were generated by pumping pure water (114 L/min), along with air (1 L/min), into the MB generator. The pure water containing MBs, in an acrylic resin tank, was then flowed through a glass tube at a rate of 20 mL/min and the flow was observed by a CCD camera. The MB sizes were measured by an optical particle counter (LiQuilaz-S02, Particle Measuring Systems Co., Tokyo, Japan).

2.3. Membrane and membrane module

Cellulose acetate ultrafiltration hollow fiber membranes (Daicen Membrane-Systems Ltd., Tokyo, Japan) were used for filtration. The membrane (inner/outer fiber diameters: 0.80/1.30 mm) is hydrophilic, displaying a low tendency toward adsorption of substances in raw river water [16]. The nominal molecular weight cut-off (as determined by protein rejection) was 150,000 Da. A membrane module with a filtration area of 5.0 m^2 was used for the filtration experiments. The hydraulic permeability of the module, determined by using pure water, was 660 L/(m^2 h 0.1 MPa).

2.4. Membrane filtration measurement

The effect of MBs on membrane filtration flux was examined as follows. Raw river water sampled from the Ibo River (Hyogo, Japan) was used as the feed solution. Its turbidity was in the range of 1–60 NTU, and concentrations of dissolved organic carbon was 2–12 mg/L. UV absorbance at 260 nm (E260, an index of humic substances concentration) was 0.03–0.18 Abs/cm. Total bacteria was in the range of 500–4,200/mL.

Fig. 2 shows a schematic diagram of the pilot-plant-scale filtration equipment with the MB generator. The filtration system consisted of two loops: a recirculation loop for cross-flow filtration, and a loop for membrane backwashing. The recirculation loop comprised a membrane module, a MB generator, an air suction valve, and a recirculation pump. The raw water was introduced into the pressurized recirculation loop after passing through a coarse strainer. The feed water was then delivered to the hollow fiber membrane module, in which the water flowed through the lumen side of hollow fibers 0.8 mm in diameter. Part of the water (the permeate) was filtered through the membrane, and the remaining water (the concentrate) was recirculated to mix with the feed water. Cross-flow ultrafiltration was conducted at a flow velocity of 0.16 m/s and a constant pressure of 50 kPa. For filtration experiments of short duration, backwashing was not performed. For filtration

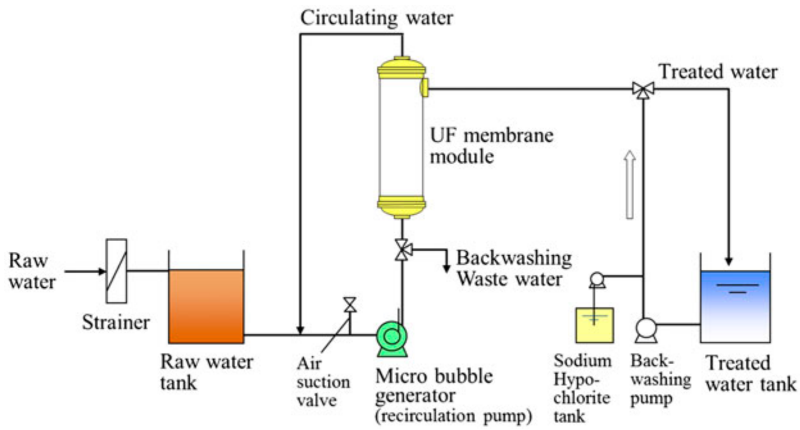

Fig. 2. Schematic diagram of the membrane filtration equipment in the pilot plant with microbubble generator.

experiments of longer duration, the membrane was backwashed every 1 h for 1 min with the permeated water, into which had been injected sodium hypochlorite at a concentration of 3–5 ppm. Filtration was tested repeatedly in two modes: with MBs (by opening the air suction valve), and without MBs (by closing the air valve).

Ultrafiltration membrane flux was determined by measuring the volume of permeate collected over the measured time intervals. The flux measurements were performed within a few minutes after periodical backwashing was finished. The flux was corrected to its value at 25℃. The fluxes J were calculated by the equation $J = Q/A$, where Q is the volumetric flow rate (m^3/d) and A is the effective area of the hollow fiber membrane (m^2).

2.5. Membrane cleaning

The effect of MBs on membrane cleaning was examined with hollow fiber membrane modules which had become fouled during filtration of the Ibo River water. The membrane modules were washed by flushing clean water with or without MBs into the lumen side (filtration side) of the hollow fiber membrane modules. The flushing was carried out at a flow velocity of 0.5 m/s in the hollow fibers for 10 min. The water flux recovery was calculated as the ratio of the water flux after cleaning to initial flux.

3. Results and discussion

3.1. Monitoring the production and size of MBs

Fig. 3 shows the MBs generated in pure water. After stopping the MB generation, the MB-infused water retained its milky white color for about 3 min and the solution gradually became transparent.

The MBs were imaged with a CCD camera while flowing through a glass tube (Fig. 4(a)), and their diameters were measured from the image. The MBs had a narrow size distribution, and their diameters were several tens of micrometers, mainly 10–20 μm in

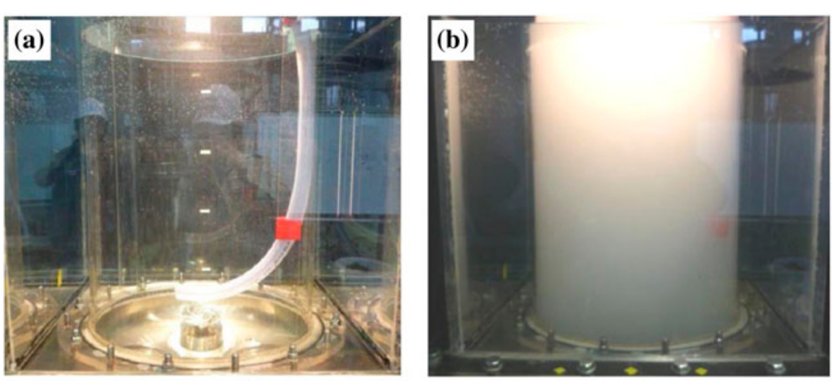

Fig. 3. MBs generated in pure water: (a) Before generation and (b) During generation.

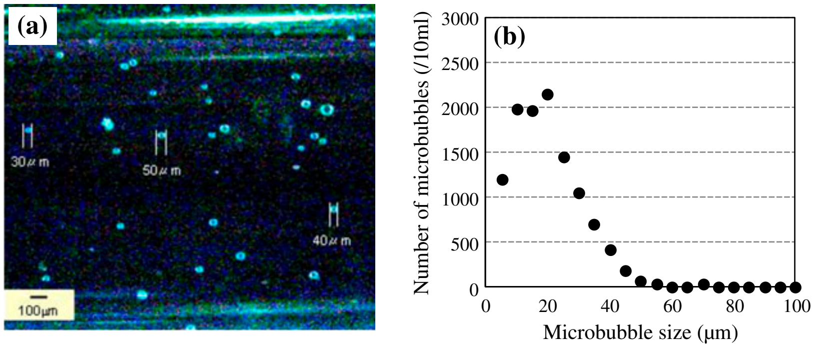

Fig. 4. (a) Optical image of MBs, taken via CCD camera. (b) Distribution of MB size measured by optical particle counter.

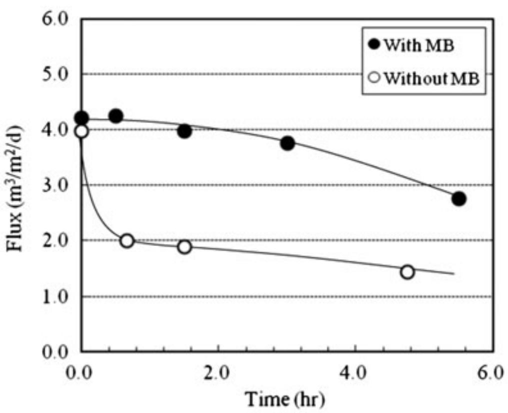

Fig. 5. Effect of MB addition on filtration flux during filtration over a short-time period. Flow velocity: 0.16 m/s.

pure water, with the largest MB about 50 μm in diameter (Fig. 4(b)).

3.2. Using MBs to reduce membrane fouling

Fig. 5 shows how the filtration flux changes during a short period of filtration without backwashing. The flow velocity was 0.16 m/s. The flux decreases immediately during filtration without MB (○), while it stays more uniformly high during filtration with MB (●). This indicates that MBs apparently enhance the filtration flux by reducing membrane fouling. This is the first report to demonstrate the use of MBs to reduce membrane fouling during filtration.

Possible mechanisms by which MBs reduce fouling include detaching the cake layer, adsorbing foulants on the MB surface, reducing the cake layer resistance by being present in the cake layer, and generating radicals, which can decompose organic matter, when they collapse. These mechanisms are schematically shown in Fig. 6 and described as follows.

3.2.1. Detachment of cake layer by MBs

MBs which contact the foulant on the membrane may remove the cake layer. Derradji et al. [17] and Hwang and Wu [18] reported that filtration flux increased when the shear stress was increased by injecting air onto the membrane surface. Although air macrobubbles were used in these experiments, similar effects can be expected for MBs in water. The MBs effect may be larger than macrobubble effect due to the longer existence life time.

3.2.2. Adsorption of foulant on MB surface

Hydrophobic foulants easily adsorb on the MBs due to hydrophobic interactions. Adsorption foulants onto MBs may reduce the foulant concentration in the solution. Nguyen et al. [19] and Eftekhardadkhah et al. [20] have reported the adsorption of frothers, collectors, and crude oil components onto air bubbles.

3.2.3. MB presence in the cake layer reducing the cake layer resistance

The presence of MB in the cake layer may decrease the resistance of the cake layer. Cabassud et al. investigated the use of gas sparging to prevent particulate deposition during ultrafiltration. They reported that the gas sparging caused the cake porosity to increase and the specific cake layer resistance to decrease [21].

3.2.4. Decomposition of organic matter by radicals generated from the collapse of MBs

Takahashi et al. reported that free radicals generated from MB collapse can decompose phenols [10]. Decomposition of organic matter in this manner may reduce membrane fouling.

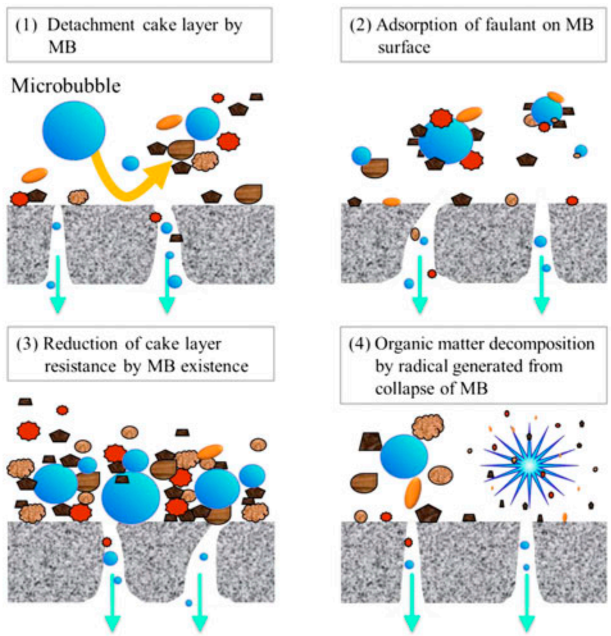

Fig. 6. Possible mechanisms by which MBs reduce fouling.

To discuss the mechanism for the reduction of membrane fouling by MBs, more fundamental approaches are necessary. These studies are now underway in our group.

Fig. 7 shows the effect of cross-flow velocity on the filtration flux for longer time intervals. Two cross-flow velocities, 0.16 and 0.01 m/s, were tested. To make the microbubble concentrations almost the same, air volumes were controlled to 200 mL/min at 0.16 m/s and approximately 10 mL/min at 0.01 m/s by regulating the suction valve. In both cases, adding MBs successfully prevented membrane fouling for about 10 h. The effect of MBs was more pronounced when the cross-flow velocity was low. The high flux was obtained at the lower cross-flow velocity of 0.01 m/s when MBs were present (●); the flux was even higher than that obtained at the higher cross-flow velocity of 0.16 m/s without MBs (△). This means that adding MBs lowers the cross-flow velocity required to produce a given flux, which causes the filtration to consume less energy consumption. After 20 h the fluxes showed almost same value. It is supposed that MBs are effective for reversible fouling, and are not effective for irreversible fouling by the natural organic matter adsorption.

Fig. 8 shows results for filtration, performed with backwashing, for a longer duration than the experiments in Fig. 7. Filtration with MBs was continued for 11 d at almost constant water flux, following the stop of MB supply by closing the air valve. The filtration flux then gradually decreased due to membrane fouling. This suggests that the foulant, which decreased the flux from 4.0 to 2.0 $m^3/m^2/d$ for 1 h filtration (in Fig. 7), was not removed completely by backwashing. Backwashing was important to achieve before irreversible fouling progressed. Subsequently, MBs were again generated by opening the air valve. The flux recovered and maintained an almost constant value during this operation. It assumed that reversible fouling was reduced by the MBs and fouled membrane was easy to be backwashed. This series of test was repeated two times. The results show that the MB enabled the flux to maintain a high value over a long-time period, while without MBs, the flux decreased rapidly due to fouling. The turbidity of the permeate was 0.1 NTU and there was no difference between operations with MB and without MBs. The current value increased

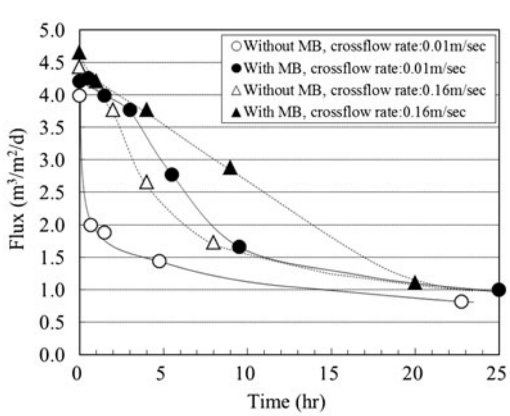

Fig. 7. Influence of flow velocity on the effectiveness of MB addition.

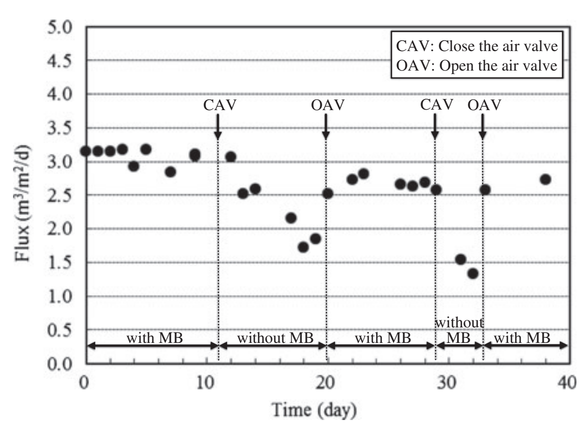

Fig. 8. Effect of MB addition during filtration over a long-time period.

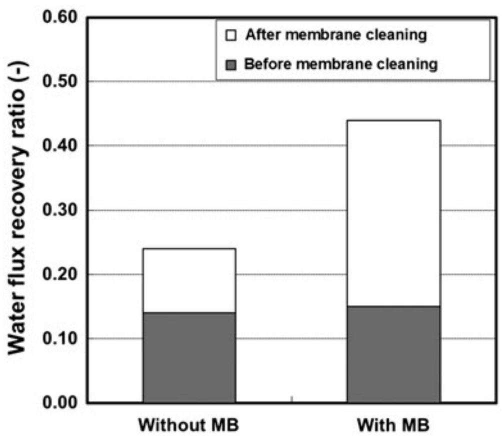

Fig. 9. Effect of MB addition on the membrane cleaning.

from 4.5 to 4.9 A when the general centrifugal pump (P121 type EBARA Co.,) was replaced by MB generator, and the power consumption was up to 1.09 times higher. Even if power consumption increased as for this MB generator, the filtration flux was enhanced almost twice. This effective system can reduce the total water treatment costs.

3.3. Effects of MB addition on membrane cleaning

The effects of MB addition on chemical-less membrane cleaning are shown in Fig. 9. In these experiments, the membranes that had been fouled to the same extent were used in two cleaning operations. The ordinate of Fig. 9 is the water flux recovery ratio, which is defined as the ratio of the water fluxes after cleaning to initial fluxes. The water recovery ratio was much higher when the fouled membranes were flushed with clean water containing MBs. However, this water recovery ratio was lower than 90% that was general recovery ratio by sodium hypochlorite. It was assumed that MBs removes only reversible fouling. MBs in water are effective for chemical-less cleaning of the membrane by removing foulants on the membrane surface. MBs in water are effective for cleaning the membrane by removing foulants on the membrane surface. This suggests that using MBs may reduce the amount of chemicals needed to clean membranes. MBs evidently can enhance the cleaning of fouled membranes, as well as reduce the incidence of fouling during filtration.

4. Conclusions

Incorporating MBs into water destined for membrane filtration was first examined as a method to reduce membrane fouling. The usefulness of the MB addition was confirmed in a pilot plant using raw river water. The cross-flow velocity influenced the effectiveness of the MBs; a larger effect was obtained in the case of lower cross-flow velocity. Long-term filtration experiments, with backwashing, were conducted for more than one month. MBs prevented membrane fouling in both the short-term and long-term experiments. MB addition also improved membrane cleaning. The fouled membrane was effectively regenerated by flushing with clean water containing MBs.

This work presents experimental evidence that MBs not only reduce the incidence of membrane fouling, but also improve the cleaning of fouled membranes. However, it is not clear whether the MBs act by detaching the cake layer, adsorbing foulants, reducing the resistance of the cake layer, producing radicals that decompose organic matter, or by another mechanism. Our next target is to clarify the mechanism by which the addition of MB improves membrane filtration for water treatment.

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