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Direct measurement of cake fouling potentials by powdered activated carbon during microfiltration of surface water

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ABSTRACT

Powdered activated carbon (PAC) addition is one of the promising options to improve the natural organic matter removal efficiency in microfiltration system. However, there have been controversial research results with impacts of PAC addition on membrane fouling. Direct observation technique through membranes was applied in this study to quantitatively analyze membrane fouling by PAC. Particle deposition rates and fouling potential were measured at various PAC concentrations. Membrane filtration tests revealed that the formation of PAC cake layers on the membrane surfaces could increase membrane permeate flux due to the increased porosity of the cake layers. Increased concentrations of PAC in feed water could increase the thickness of cake layers, yet no significant decline of permeate flux was observed. The proposed direct observation technique, which showed good agreement with filtration resistance measurement, could be applied for the monitoring of activated carbon deposition on membrane surface as well as selecting proper chemical cleaning agents for fouled membranes in a relatively short time.

Keywords: Membrane filtration; Fouling; Direct observation; Powdered activated carbon; Microfiltration

1. Introduction

Low-pressure-driven membrane technologies such as microfiltration (MF) and ultrafiltration have been widely applied to remove specific pollutants, which are not normally removed by conventional processes. However, they cannot readily remove color, dissolved natural organic matters, and synthetic organic compounds. Hybrid systems of the membrane filtration coupled with adsorption and/or coagulation process could be alternative ways to remove those compounds in a cost-effective manner [1].

Powdered activated carbon (PAC) adsorption has been successfully applied in low-pressure membrane

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processes to remove trace organic compounds in drinking water treatment [2–4]. However, there were controversial research results on membrane fouling by activated carbon particles. Suzuki et al. [3] reported that a decrease in membrane permeate flux was slower in the PAC–MF system which might be resulted from the reduction of the organic loading to the membrane due to the adsorption of organics on PAC. Other researchers [4] found that there was no further decrease in the permeate flux by PAC addition to the solution containing humic acids. On the contrary, Lin et al. [5] reported that flux decline in PAC-pretreated streams were higher than that in the original humic substances solutions. Thus, further studies are required to assess the effect of PAC addition on the membrane permeability and fouling in PAC–MF hybrid processes because membrane fouling has been a major obstacle to the wide application of membrane technology [6,7].

Direct observation through the membrane technique (DOTM) is a non-invasive technique that visualizes the deposition behavior of particles during membrane filtration [8]. This technique is mainly composed of a membrane filtration cell equipped with a membrane and microscope. DOTM have been widely applied for various purposes such as solute concentration profile measurement in the polarization layer, cake layer growth measurement, and quantification of foulant detachment [8–11]. However, there is no report on application of DOTM in characterizing the PAC particle adsorption characteristics during MF process.

In this study, we used DOTM technique for the real-time monitoring of PAC particle deposition on the membrane surface. We evaluate the effect of PAC concentration on the particle deposition and membrane fouling during the MF of water taken from water treatment plant intake basin. Membrane filtration resistance and particle size distribution was also analyzed to further examine and discuss on the membrane fouling during MF.

2. Materials and methods

2.1. Direct observation flow cell

The membrane cell consisted of polycarbonate and glass. The dimensions of the flow channel were 1 mm height by 25.4 mm width by 76.2 mm length. The cell was mounted on the stage of a phase contrast microscope (Olympus, BX-51) to allow direct observation of particle deposition by light and fluorescence microscopy (Fig. 1). A CCD camera (five times magnification) was mounted on the microscope and images were downloaded in real-time to a computer for post-processing and image analysis. In order to operate at common applied pressures for MF and UF membranes, two “interchangeable parallel plates” were fused to the top and bottom cell plates and reinforced as needed. The membrane sample was placed between the two plates with a permeate spacer beneath it. Applied pressure was maintained constantly by connecting the flow cell to a pressure vessel (the feed tank) through a closed line loop. Peristaltic pumps were used for both retentate and permeate flows, which allowed for accurate control of the cross-flow and permeation velocities. The retentate and permeate flows were merged and sent back to the feed tank. Such a configuration enabled extremely stable feed pressure and flow rates within the cross-flow filtration cell. The membrane used in this study was made by

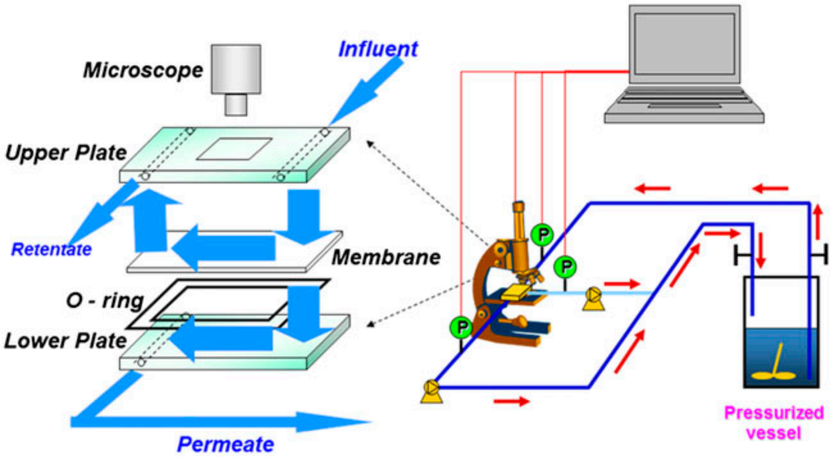
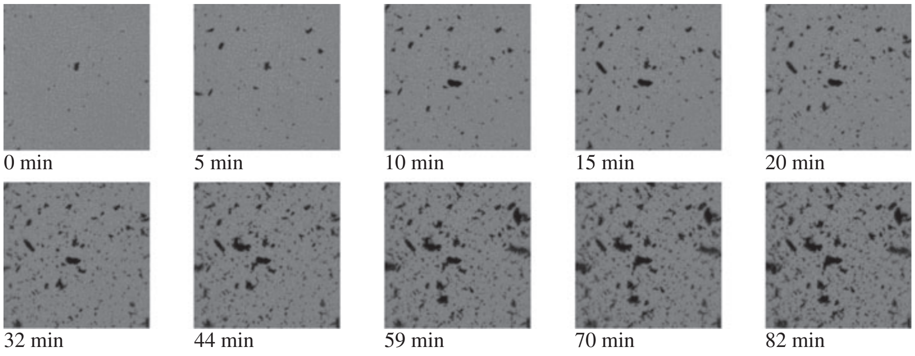
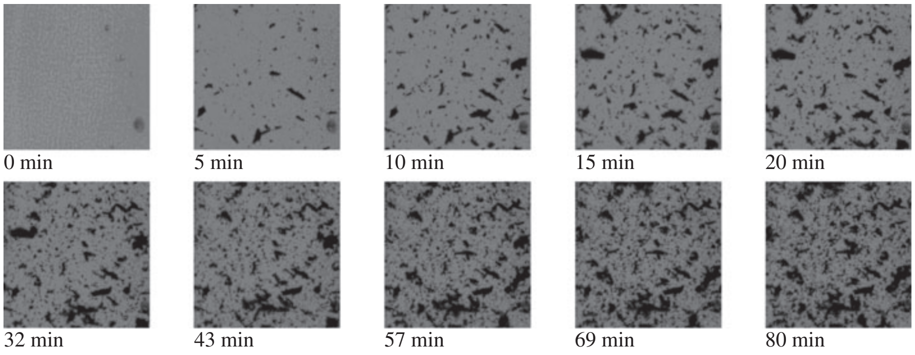


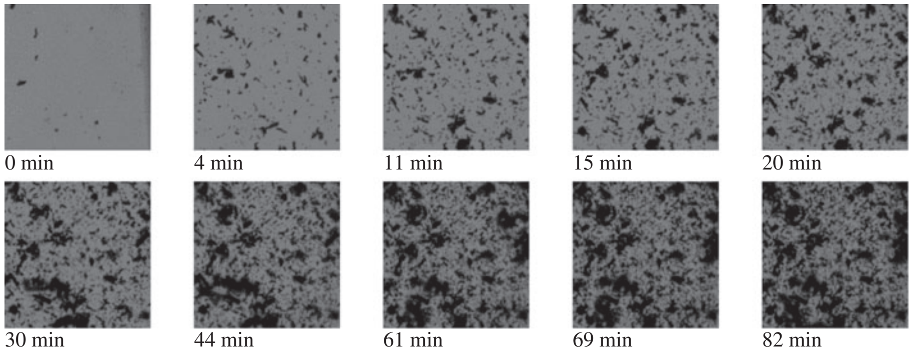
Fig. 1. Schematic diagram of direct observation technique setup.



(a) PAC 10 mg/L



(b) PAC 20 mg/L



(c) PAC 30 mg/L

Fig. 2. Particle deposition on membrane surface under various PAC concentrations.

polytetrafluoroethylene having pore diameter of 0.1 μm (Sumitomo Electric Fine Polymer, Inc, Japan). The raw water tested in this study was taken from a Daejeon Wolpyeong water treatment plant. Raw water had DOC of 3.8 ± 0.3 mg/L, UV at 254 nm of 0.045 cm^{-1}, pH of 7.5, and turbidity of 3.7 nephelometric turbidity units (NTU).

2.2. Analytical methods

Organic carbon was determined on a Dohrmann DC-180 TOC Analyzer (Rosemount, USA) after filtration through 0.45 μm filter. Ultraviolet absorbance at 254 nm (UVA_{254}) was measured on a Beckman DU-65 spectrophotometer (Beckman-Coulter Inc., Fullerton, USA). Turbidity was measured by a portable meter (2100P Portable Turbidimeter, Hach Company, Loveland, CO, USA) and reported in NTU.

To test the fouling characteristics of the samples, a stirred cell having an effective volume of 200 mL (Amicon, USA) was used. Total resistance (R_t) was obtained by filtration of the raw water with PAC. For cake and pore blocking resistance (R_{a+p}), the cake layer was first removed then permeate flow was measured.

R_c was estimated from R_t to R_{a+p} values. To avoid temperature effects, the resistances were normalized to 25°C.

3. Results and discussion

3.1. PAC particle deposition on membrane surface

Fig. 2 shows particle deposition images during MF of raw water with different PAC concentrations. The direct microscopic images show increasing extents of surface coverage with increasing feed PAC concentration. The particle deposited on membrane surface randomly in the beginning of the filtration test (<20–30 min), however, some agglomerated particles were observed with prolonged filtration time. There was change in the size of some agglomerated particle as well as their location. A similar phenomenon has been observed in other study for loosely deposited yeast cell on membrane surface [8].

For the quantitative comparison of particle deposition rate, surface coverage was further analyzed using the imaging software. Fractional surface coverage increased in proportion to the feed PAC concentration as shown in Fig. 3. Surface coverage increased linearly in all experiments up to fractional values of 20–40%, and then tapered off with continued particle deposition. As explained above, the PAC particles deposited on membrane surface very loosely, therefore, some portion of the deposited particles might be detached from the membrane surface.

Generally, the initial deposition is most important step among the various stage of membrane fouling [12]. Moreover, it was observed that particle detachment took place simultaneously with particle deposition after 20 min of filtration. Therefore, only the first 20 min data, which is linear portion of particle deposition profile (Fig. 3), was used for the deposition rate calculation. The deposition rate jumped up from 0.37 to 1.38%/min with the increase of PAC concentration from 10 to 20 mg/L, however, additional 10 mg/L of PAC concentration increase (20 mg/L) did not increase particle deposition rate much (Table 1). This result implies that there might be some additional factors which accelerate the particle deposition on membrane surface at low PAC concentration. Particle deposition rate was 1.38–1.77%/min for PAC concentration of 20–30 mg/L, respectively.

Table 1
Particle deposition rates at different filtration time with various PAC concentrations

Item	10 (mg/L)	20 (mg/L)	30 (mg/L)
Deposition at 20 min (%)	7.2	25.1	34.8
Deposition at 80 min (%)	25.2	54.6	75.8
Deposition rate (%/min)	0.37	1.38	1.77

Above a certain PAC concentration, the PAC particle inside the feed water stream might collide with pre-deposited particle on membrane surface and it might increase shear force acting on the particle which could retard the particle deposition on membrane surface. Also, it might be caused by scouring actions of PAC particles [13]. The particle size distribution with different PAC concentrations was analyzed and will be discussed in the next section.

It was expected that permeate flux will decrease as the membrane fouling progressed. But from the

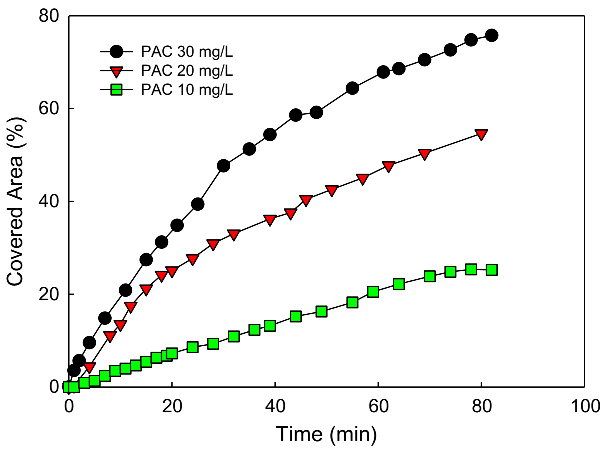


Fig. 3. Membrane surface coverage by PAC particle with different PAC concentrations.

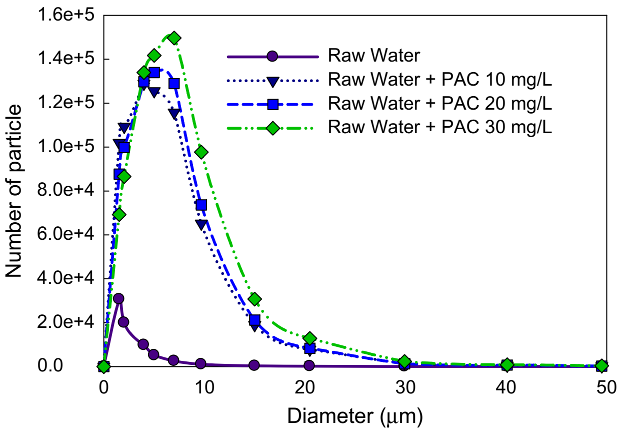
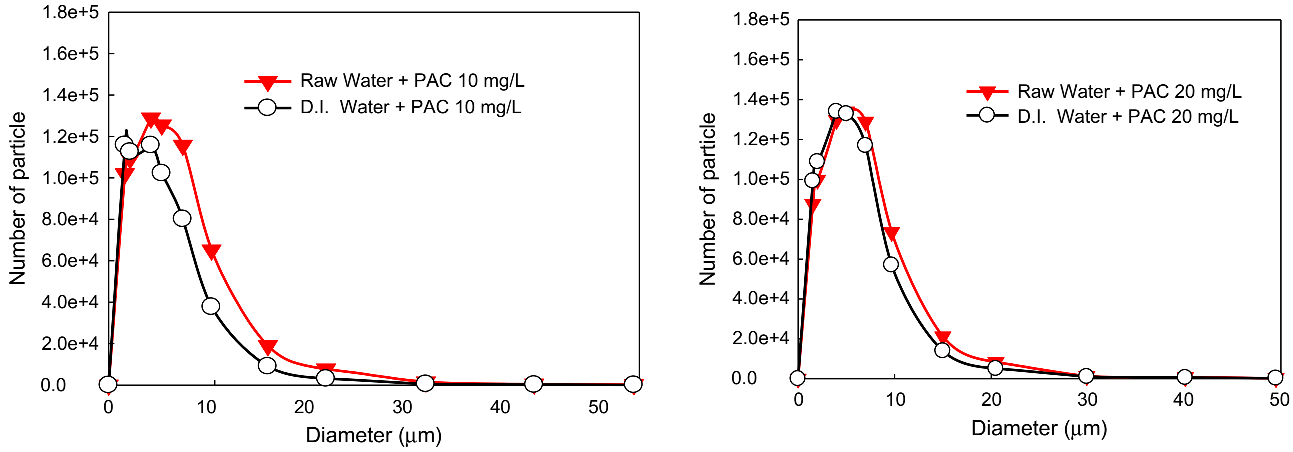
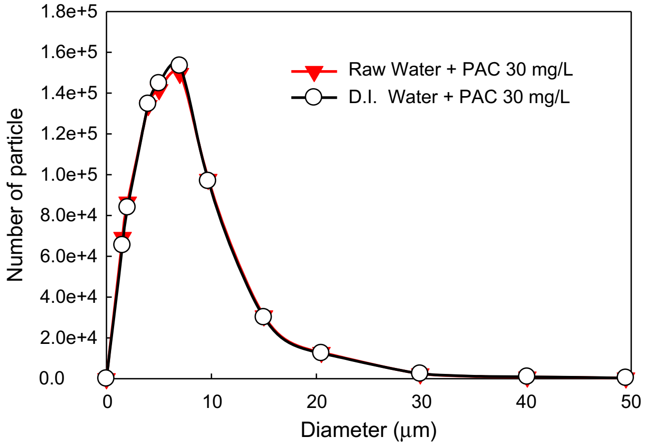


Fig. 4. Changes in particle size distribution with different PAC concentrations.



(a) PAC 10 mg/L

(b) PAC 20 mg/L



(c) PAC 30 mg/L

Fig. 5. Comparison of particle size distributions between raw and deionized water with various PAC concentrations.

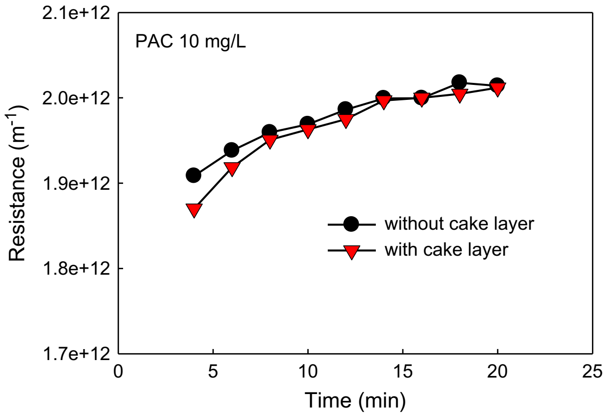


Fig. 6. Membrane filtration resistance with and without cake layer (CFV = 15 μm/s).

permeate volume measurement, there was no decrease in filtrate volume. Considering the DOTM tests for different PAC concentration and filtrate flux measurement, there might be no pore blocking effect and/or irreversible fouling by PAC particles [14]. Also the relationship between membrane surface coverage and actual membrane fouling which are manifested by flux decline should be studied with other analytical tools such as membrane resistance analysis or TMP monitoring of continuous operation system.

3.2. *Particle size distribution*

The particle size distribution of PAC particle with different PAC dosage was analyzed in an attempt to elucidate the effects of particle size on PAC deposition rate on membrane surface. PAC particles exhibited a wide range of particle size, ranging from <0.5 to 40 μm in diameter with the mean size of 6.01 μm as reported previously [14]. The mean particle size increased to 6.99 μm by adding 30 mg/L of PAC (Fig. 4). The mean particle diameter for raw water was 3.34 μm as the raw water also contains some particulate matters. Even though the size has increased by PAC addition, it was difficult to determine whether the increase in particle size is caused by the self-aggregation of PAC particle itself or adsorption of colloidal/particulate contaminants on PAC surface.

To determine how much the particle in raw water contributes to the particle size increase with PAC addition, PAC with deionized water under different PAC concentration was analyzed (Fig. 5). In case of PAC 10 mg/L, particles larger than 5 μm were increased by adding raw water than that with deionized water. But there was no difference between deionized water and raw water when the PAC concentration was 30 mg/L, as shown in Fig. 5. It might be the most of colloidal materials which can be adsorbed on PAC particles could be removed enough only with 20 mg/L of PAC concentration. Therefore, excessive addition of PAC could not give any significant benefit in membrane fouling control, or just would increase membrane fouling resistance with the increased particle deposition on membrane surface as discussed previously.

3.3. Filtration resistance with/without cake layer

Even though PAC addition before the MF increased particle deposition on membrane surface, the actual filtration flux should be investigated to know whether they are reversible or irreversible foulants. To investigate the effect of PAC cake layer deposited on membrane surface on filtration flux, the flux was measured with cake-formed membranes which were prepared by filtering PAC slurry for 1 h with different PAC concentration. Also, the filtration resistance measured after removing the cake layer with deionized water.

As shown in Fig. 6, filtration resistance increased slightly with the filtration time regardless of presence of cake layer. Interestingly, the filtration resistance slightly increased after removing PAC cake layer. This result indicates that PAC cake layer played a positive role in terms of membrane fouling. Previous researchers [5–15] also reported that addition of PAC resulted in the formation of larger particles and reduced TMP increase.

4. Conclusions

The effect of PAC addition on membrane fouling was assessed with direct observation of PAC particle deposition on membrane surface. The following conclusions have been drawn from this study:

(1) The DOTM was successfully applied to monitor and analyze PAC particle deposition on membrane surface. The addition of PAC increased membrane surface coverage and particle deposition rates, but the deposition on the membrane surface was relatively loose. The cake layer was easily detached from surface by the collision with PAC particles contained in the feed water stream.
(2) The particle size increased from 6.02 to 6.99 μm by adding 30 mg/L of PAC. The increase in particle size was mainly caused by adsorption of particulate/colloidal materials on PAC particles. However, at higher PAC concentration, there was no difference in particle size distribution compared to that measured without raw water. This result implies that excessive amount of PAC gives no positive effect in particulate matter removal and might accelerate membrane fouling by increasing particle deposition on membrane surface.
(3) Filtration resistance measurement confirmed that PAC addition does not increase the membrane fouling potential. Moreover, decreased membrane resistance after removing PAC cake layer revealed that PAC particles play a positive role in membrane fouling. The cake layer might form a more porous secondary membrane layer on top of membrane surface.
(4) Direct observation technique can be applied for the monitoring the activated carbon deposition on membrane surface as well as selecting proper chemical cleaning agent for the fouled membrane in a relatively short time.

Acknowledgments

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