学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（工学）　氏名　ZHAO Yuanjun

学 位 論 文 題 名

Application of submicron super-fine powdered activated carbon on mitigating membrane fouling in microfiltration systems

(精密ろ過における膜ファウリング抑制へのサブミクロン超微粉砕活性炭の応用)

Membrane fouling is still a major problem in water treatment that uses membrane separation. In this study, commercially available powdered activated carbon (PAC) with a median diameter of 12–42 μm was ground into 1 μm sized superfine PAC (SPAC) and 200 nm sized submicron SPAC (SSPAC), and SSPAC was investigated as a pretreatment material for the prevention of hydraulically irreversible membrane fouling during microfiltration (MF) process. The findings are summarized as follows:

Compared with PAC and SPAC, SSPAC has a high capacity for selective biopolymer adsorption, which is a characteristic found in natural organic matter and is commonly considered to be a major contributor to membrane fouling. Precoating the membrane surface with SSPAC during batch filtration further removes the biopolymers by straining them out. In lab-scale membrane filtration experiments, an increase in the transmembrane pressure (TMP) was almost completely prevented through a precoating with SSPAC based on its pulse dose after coagulation pretreatment. The precoated SSPAC formed a dense layer on the membrane preventing biopolymers from attaching to the membrane. Coagulation pretreatment enabled the precoated activated carbon to be rinsed off during hydraulic backwashing. The functionality of the membrane was thereby retained for a long-term operation.

Furthermore, SSPAC was dosed in a pulse after coagulation pretreatment to investigate the effects of the filtration flux during precoating, the timing of coagulant dosing, and the types of coagulants on the suppression of TMP rise in a submerged filtration system by a hollow fiber polymeric (Poly Vinylidene DiFluoride [PVDF]) membrane. The TMP rise was insensitive to the filtration flux during precoating. However, the efficacy of the SSPAC was sensitive to the use of the coagulant polyaluminum-chloride (PACl). Unlike pulse dosing of SSPAC, pulse dosing of coagulant led to excessive aggregation and settling of SSPAC and insufficient formation of a precoating layer. The best filterability was achieved when the coagulant was dosed continuously, not only during the filtration period but also during the time water was being fed to the membrane tank after drainage. For sulfated PACl, high basicity (70 percent), with its high charge neutralization capacity, was preferred to medium basicity (50 percent). Meanwhile, the TMP rise was more effectively suppressed by non-sulfated PACl with a low-to-medium basicity (20–40 percent) than zero or high basicity (70 percent). This behavior was explained by the fact that, as the basicity increases, hydrolysis of the coagulants is lessened, but the charge neutralization effect becomes stronger.

On the other hand, SSPAC was applied in a pulse/continuous dose to ceramic-membrane MFs of two very different types: a submerged tubular membrane system that enabled vacuum-driven, outside-in filtration and a monolithic membrane system that enabled pressure-driven, inside-out filtration.

Constant-flux filtration with repeated hydraulic backwashing was performed using diluted secondary-treated municipal wastewater and natural river water, and the effect and mechanism of membrane fouling mitigation were investigated. Pulse dosing with SSPAC alleviated TMP rise more than continuous dosing in the submerged tubular membrane system because of the effects, inter alia, associated with precoating the SSPAC layer as well as adsorption, by which soluble membrane foulants were effectively removed. Moreover, pulse dosed SSPAC combined with a coagulant formed a precoat layer on the membrane that was easy to peel off with hydraulic backwashing. Continuous dosing of SSPAC was inferior to pulse dosing of SSPAC in terms of water quality, and some SSPAC was left on the membrane surface after hydraulic backwashing. In the monolithic membrane system, however, the superiority of pulse dosing of SSPAC was not observed: pulse and continuous dosing of SSPAC led to similar reductions of TMP rise. The SSPAC layer was unevenly distributed along the length of a channel and between channels in the monolithic membrane.

Overall, this study proved that precoating the membranes with SSPAC after coagulation is a promising way to control membrane fouling, and efficiently prevents an increase in the TMP because of the straining effect of the SSPAC and the high capacity of the SSPAC to adsorb any existing biopolymers. On the other hand, this study also enabled us to consider the possibility that there were local differences in the permeation rate of the membrane, and those differences may have affected the success or failure of the precoating method. This possibility is important to consider when apply pulse dosing of SSPAC to a full-scale membrane filtration system.