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Influence of feed concentration and transmembrane pressure on membrane fouling and effect of hydraulic flushing on the performance of ultrafiltration

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HIGHLIGHTS

- A diaphragm pump was dual-used in this experimental setup.
- Membrane fouling varies with feed SDS concentration and transmembrane pressure.
- Suitable flushing time is important for periodic hydraulic flushing.
- Compared with backwashing and forward flushing, the combined flushing is more effective to alleviate membrane fouling.

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ABSTRACT

Micellar-enhanced ultrafiltration (MEUF) is a promising technology developed for treating the wastewater containing metal ions or organic pollutants. One of the greatest problems in MEUF is membrane fouling which is mainly caused by concentration polarization, gel layer or cake formation caused by the deposition of surfactant micelles on the membrane surface and surfactant adsorption in the membrane interior. In this study, surfactant sodium dodecyl sulfate (SDS), which was used in membrane separation as colloidal particles, caused the flux decline. The transmembrane pressure (TMP) and feed concentration of SDS had significant influences on the flux. This paper presented that the lower TMP had a smaller effect on membrane fouling, and when SDS concentration was around the critical micelle concentration (CMC), lower permeate flux and higher additional membrane fouling resistance were obtained. The effects of three kinds of hydraulic flushing methods on membrane permeate flux were investigated, including periodic forward flushing, periodic backwashing and forward flushing followed by backwashing. It was found that when the periodic combined flushing interval was 10 min, forward flushing and backwashing phase times were 150 s and 90 s, respectively, and that combined flushing was more conductive to permeate flux recovery in this study.

1. Introduction

Micellar-enhanced ultrafiltration (MEUF) is a new technology developed for treating the wastewater containing metal ions and organic matters. In MEUF, surfactants are added to wastewater at levels equal to or higher than their critical micelle concentrations (CMCs) and surfactant monomers will aggregate to form micelles, then the micelles are able to solubilize organic solutes or bind ions on the surface of the opposite charged micelle via electrostatic interactions [1,2] which aims to promote the removal of metal ions or organic matters. MEUF has its own advantages such as simple operation, high removal efficiency, economical and practical, and small space requirement [3]. During the ultrafiltration, however, the flux decreases dramatically due to the existence of membrane fouling which results in increased operating costs, decreased membrane lifetime and reduced removal efficiency. There are many factors that lead to membrane fouling, such as the adsorption of solids onto the membrane, gel formation, pore blocking, and concentration polarization [4–6]. Particulate matter which is larger than the pores in commercial MF and UF membranes, forms a cake at the membrane surface; dissolved matter which can penetrate pores forms a surface cake, penetrates and clogs pores and reduces the pore diameter due to adsorption within pores [7]. The formation mechanism

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Nomenclature

List of symbols

J_w	the pure distilled water flux of clean membrane ($L/m^2 \cdot h$)
J_f	the pure water flux of membrane after flushing ($L/m^2 \cdot h$)
ΔP	the transmembrane operating pressure (Pa)
μ_m	the viscosity of distilled water (10^{-3} Pa·s)
R_m	the hydraulic resistance of the membrane (m^{-1})
$R_{f,}$	the additional fouling resistance (m^{-1})
J	the synthetic wastewater permeate flux ($L/m^2 \cdot h$)
A	the area of membrane (m^2)
V	the permeate volume (L)
t	the filtration time (h)
μ	the dynamic viscosity of the solution (Pa s)

of the gel layer during MEUF can be summarized as follows: adsorption of the surfactant monomer and aggregates on the membrane surface and within the pores; blockage of the membrane pores and partial constriction of membrane pores by surfactant adsorption [8,9]. JÖnsson and JÖnsson [10] investigated the impact of hydrophilic and hydrophobic membrane materials on the membrane flux reduction and fouling. They found that flux decline caused by low-molecular weight hydrophobic solutes is generally because of adsorption. Broeckmann et al. [11] reported that bulk particles can be separated into two fractions by including small enough to enter the membrane pores and the deposit on the membrane surface. One remained on the outside of the membrane. The other entered the membrane pores and contributes to pore blocking. Membrane fouling can be categorized into reversible fouling and irreversible fouling. Reversible fouling which is due to accumulation of the particles and build-up of a cake on the membrane can be mitigated by physical cleaning such as backwash and rinsing. Irreversible fouling is caused by internal accumulation of particles during the penetration of small particles through the membrane [12]. Actually, reversible fouling can transform into irreversible fouling if the formation of fouling layer with the solute during continuous filtration process was not removed timely. Irreversible fouling is normally caused by the strong attachment of particles, which leads to progressive deterioration of membrane performance and cannot be removed by physical cleaning methods [13]. Various methods have been proposed to alleviate membrane fouling [14–18] such as pretreatment of feed water (coagulation), physical methods (forward flushing, backwashing, ultrasonic), chemical methods (chemical reagents: NaOH, HCl, EDTA and NaClO) and modification of the membrane surface. However, these methods cannot completely eliminate fouling. Therefore, to maintain an efficient process, we generally preferred physical methods to control membrane fouling, because it does not cause pollution again and the operation is simple. Niina Laitinen et al. [19] analyzed in their study the effects of different backflushing parameters (backflushing frequency, length of the backflush, and the backflushing pressure) on flux and retentions. They concluded that the highest permeate flux was obtained when the backflush of 1 s was made every 2 min with a 4 bar backflushing pressure.

Sodium dodecyl sulfate (SDS), an anionic surfactant, is often chosen for the effective removal of zinc, copper, nickel, cadmium and methylene blue [20–23]. Meanwhile, the influence of SDS on colloidal interactions in the cake layer on the ultrafiltration membrane surface was investigated. The relationship between fouling potential and SDS depends on the concentration of SDS [24]. Some researchers have pointed out that the low SDS concentration did not benefit the rejection of SDS because of the small number of micelles, and with the increasing feed SDS concentration, the SDS rejection increased dramatically, for the reason that the SDS concentration at the vicinity of the membrane surface reached the CMC of SDS, leading to the formation of more micelles. However, when the feed SDS concentration was 10 times the CMC of SDS (80 mM), the SDS rejection decreased, this was attributed to the change of micelle shape from spherical to cylindrical or plate like and then the micelles could easily pass through the membrane pores causing considerable drop in the rejection of SDS [24–26]. In this study, the anionic surfactant sodium dodecyl sulfate (SDS) was added into the aqueous solution to form micelles when the SDS concentration reaches the critical micelle concentration and can be retained by an ultrafiltration membrane with pore sizes smaller than the micelle size. During MEUF, micelles bind ions on the surface of the opposite-charged micelles via electrostatic interaction. Therefore, metal ions associated with micelles are removed effectively. In fact, UF membrane cannot reject free ions at any transmembrane pressure [3]. So our study only considers SDS as membrane pollutant.

In previous studies, we reported the effects of different conditions (pH, TMP, temperature, feed concentration) on removal efficiency, fouling mechanisms and flux decline [27–29].

The purpose of this study is an attempt to investigate the effects of feed concentration and TMP on membrane fouling which was mainly reflected by permeate flux, and the influence of rinsing time on permeate flux, including the effects of forward flushing and backwashing on permeate flux.

2. Experimental

2.1. Chemicals

SDS ($C_{12}H_{25}NaS0_4$) was obtained from Tianjin Kermel Chemical Reagents Development Center, China. Phosphoric acid (H_3PO_4), potassium hydrogen phthalate ($KHC_8H_4O_4$), Sodium bicarbonate ($NaHCO_3$) and Hydrochloric acid (HCl) used in these experiments were obtained from Guoyao Chemical reagent Plant, China and analytically pure. In all the experiments distilled water was used as solvents.

2.2. Membrane

The spiral-wound membrane module used in this study was supplied by Dalian Yidong Membrane Engineering Equipment Co., Ltd., Dalian, China. The membrane material was polyethersulfone (PES) which was hydrophobic in nature. The permeate flux of the membrane was measured under standard test conditions and was found to be 44 $L/m^2 \cdot h$ at 0.05 MPa. The properties of the membrane are given in Table 1.

2.3. Performance of the experiments

All the experiments were conducted at temperature 25 ± 5 °C. According to the experiment design, the synthetic wastewater was made by adding a pre-determined amount of SDS into distilled water. Before the experiments, the solutions were stirred fully using a magnetic stirrer for about 10 min to provide highly efficient mixing and settled for 45 min to ensure the formation of micelles of constant size. The schematic diagram of the experimental setup is shown in Fig. 1.

In MEUF experiments, 40 L of feed solution was filtered through the membrane by a diaphragm pump under different pressures (0.05 MPa, 0.10 MPa, 0.15 MPa, 0.20 MPa) and the retentate was recirculated to

Table 1
Characteristics of the used spiral-wound ultrafiltration membrane module.

Type	JU1812-41
Membrane material	Polyethersulfone
MWCO (Da)	10 k
Contour size φ × L (m)	0.046 × 0.305
Effective membrane area (m^2)	0.4
Operating pressure (MPa)	<0.3
Operating temperature (°C)	5–50
pH	1–13