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Theory investigation on the variation of fouling resistance during water rinsing process of the membrane fouled with sodium alginate

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ABSTRACT

Water rinsing could effectively alleviate polysaccharides fouling in membrane bioreactor. To understand water rinsing mechanism, a suitable model was developed to depict the variation of fouling resistance during water rinsing process with different microfiltration membranes and feed solution/suspensions at different operation conditions. Various parameters including contact angle, Zeta potential, the roughness and the free energy of the membrane surface were measured or calculated to assess membrane properties. The results showed that the membrane flux was more sensitive for temperature than agitation velocity and operating pressure during water rinsing process. The membrane flux initially increased before decreasing over time due to shearing effect (removing the deposits from the membrane and forcing small cells into the pores). The predictive values of the model had a good agreement with experimental observations ($R^2 > 0.94$). Further, the model allows predicting the resistance due to deposit at the membrane surface and in-pore fouling, respectively.

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1. Introduction

The inevitable operational problem of membrane bioreactor (MBR) is membrane fouling which is mainly caused by extracellular polymeric substances (EPS) in feed suspension/solution [1]. Water rinsing is a universal and important step to alleviate membrane fouling due to its environment friendly character [2–4]. However, real EPS extraction is time-consuming and inefficient [5]. Therefore, polysaccharide such as sodium alginate (SA) is frequently used to simulate real EPS to conduct membrane fouling [6,7].

Besides, various cleaning models have been developed to understand different cleaning mechanism and provide guides to the industrial cleaning processes. Cabero et al. [8] developed a first-order model including two time-dependent rates to predict membrane resistance decay. A second order model was established to depict the variation of cake resistance during sodium hydroxide cleaning process [9]. A first-order model combined with Arrhenius form was used to describe the chemical cleaning process [10]. A three parameter model was constructed to predict the irreversible fouling resistance with cleaning time and cleaning agent concentration [11]. Popović et al. [12] proposed a kinetic model to predict the cake resistance variation for alkali and detergent cleaning process. Assuming that soluble microbial products and suspended solids fouling in a MBR went through three stages, Charfi et al. [13] developed a first-order model based on mass balance to describe the fouling resistance during the periodic relaxation and backwash operation. Based on resistance-in-series model and considering the shear effect of recycled biogas, Robles [14] proposed a model to accurately reproduce the filtration performance of submerged anaerobic MBRs. These models have given useful guides for industrial cleaning process. However, the modeling for water rinsing process of the polyacrylonitrile (PAN) membrane fouled with SA has received limited attention.

Therefore, the main objective of this work is to develop a suitable model to accurately describe water rinsing behaviors of the PAN membrane fouled with SA. In addition, different feed suspensions and microfiltration membranes were used to validate the applicability of the proposed model. Furthermore, the surface free energies of membranes and foulant were calculated to statistically assess the effects of fouling state and different water rinsing conditions on membrane flux.

2. Experimental

2.1. Materials

In this study, 0.1 μm PAN and polyvinylidene (PVDF) microfiltration membranes (Ande Membrane Separation Technology &

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Table 1
Quality of active sludge suspension used in the experiment.

COD (mg/L)	NH_3-N (mg/L)	TOC (mg/L)	pH	Turbidity
226–229	46–74	86–115	7.5–8.0	20–26

Engineering (Beijing) Co., Ltd.) were used and all chemicals were of analytical grade (Beijing YILI fine Chemical Engineering, Ltd.). The filtration solution of SA was prepared by mixing SA with sodium bicarbonate as buffer [15], which consists of 100 mg/L SA and 0.5 mmol/L sodium bicarbonate (pH = 7.8). The quality of active sludge suspension from MBR in lab was shown in Table 1.

2.2. Experimental procedures

The experiments were performed in a dead-end system with magnetic stirring.

(1) The PAN membranes with an effective surface area of 37.39 cm^2 were soaked in the deionized (DI) water at 4 °C for 12 h to remove the conservation chemicals (glycerin). (2) 300 mL DI-water (20 °C) was poured into the cylindrical cell where the membrane had been loaded. Then DI-water was pressured through the membrane by compressed air under 0.05 MPa to measure the pure water flux (PWF) of virgin membrane (5 min). The permeate weight was recorded by electronic balance connected with a computer and the sampling-time interval is 30 s. To keep the same standard, the discrepancies of PWF of virgin membranes between each other were less than 10%. Then these selected membranes were used in the following fouling experiments. (3) 250 mL SA solution was poured into the cell and pressured through the membrane by compressed air for 1 h under 0.05 MPa at 200 rpm. The experimental temperature was kept at 20 °C. After fouling, the excess feed solution was poured out of the cell. The fouling operations were varied with different feed concentrations (50, 100, 200 and 400 mg/L SA), agitation velocities (200, 300 and 400 rpm) and operation pressures (0.025, 0.05 and 0.075 MPa). Each operation was conducted three times to obtain the average value. Comprehensively considering the cost and the final fouling state, the fouling condition was determined to conduct the following rinsing experiments. (4) The water rinsing of the fouled membrane was performed with 100 mL DI-water at different water rinsing conditions (cleaning time (0–60 min), agitation velocities (200–500 rpm), temperatures (20–50 °C) and operation pressures (0.025 and 0.05 MPa)). After water rinsing, the water was poured out of the cell. The PWF of the cleaned membrane was measured with 300 mL DI-water under 0.05 MPa at 20 °C until the flux remained stable. Each operation was conducted three times to obtain the average value.

2.3. Analytical methods

2.3.1. Surface analysis

Static water contact angle measurements were performed using a Ramé-Hart Model 250 Goniometer (USA). Three liquids (DI water, glycerol and diiodomethane) were selected to perform contact angle measurements for calculating the surface tension parameters [16]. For this test, the virgin PAN membrane samples should be immersed in DI water for 24 h then dried at 35 °C for 24 h [17]. The fouled and cleaned membranes were also dried at 35 °C for 24 h. Each sample was conducted at least five times at different locations to obtain the average value. The measurements of Zeta potential of the membrane surface were conducted by a streaming potential analyzer (Aaton Paar SurPASS, Austria) at 25 °C. Triplicate measurements were conducted for each sample to obtain the average value. The particle size distribution of the SA solution was measured by the Malvern laser particle diameter distribution instrument (MAF-5001, Malvern CO., England). The pore size distribution of the PAN membrane was obtained by modified bubble point method. ATR-FTIR data were recorded on an IRAffinity-1 instrument (Shimadzu CO., Japan). The roughness of the membrane surface was measured with AFM (Pico Scan TM 2500, MI CO., USA).

2.3.2. The determination of the shear stress

The shear stress for the solution at the membrane surface was calculated as reported by Kosvintsev et al. [18].

$$\tau = 0.825\mu\omega\, r\frac{1}{\delta}, \quad r < r_c \tag{1}$$

$$\tau = 0.825\mu\omega\, r_c\left(\frac{r_c}{r}\right)\frac{1}{\delta}, \quad r > r_c \tag{2}$$

$$r_c = \frac{D_i}{2}1.23\left(0.57 + 0.35\frac{D_i}{D_c}\right)\left(\frac{b}{D_c}\right)^{0.036} n_b^{0.116}\frac{\mathrm{Re}}{1000+\mathrm{Re}} \tag{3}$$

$$\mathrm{Re} = \frac{\rho\omega {D_i}^2}{2\pi\mu} \tag{4}$$

$$\delta = \sqrt{\frac{\mu}{2\rho\omega}} \tag{5}$$

where *Re* is the Reynolds number in the stirred dead end system; ρ is fluid density, kg/m^3; ω is the angular velocity, rad/s; D_i is the diameter of the impeller blade, m; μ is the dynamic viscosity of fluid, Pa s; δ is the thickness of momentum boundary layer, m; r_c is the critical radius where the shear stress is maximum, m; D_c is the diameter of stirred cell, m; b is the blade height, m; n_b is the number of stirred blades; τ is the shear stress, Pa; r is the distance to the center point in the stirred cell, m.

2.3.3. The calculation of the diffusion coefficient

Temperature affords an important role in the diffusion of SA particles in the DI water. The diffusion coefficient (D) is calculated according to the Stokes–Einstein equation [19]:

$$D = \frac{k_B T}{6\pi\mu\, r_a} \tag{6}$$

where k_B is the Boltzmann constant (1.38×10^{-23}J/K), T is the operating temperature, K; r_a is the average radius of SA particles, m.

2.3.4. The calculation of surface energy

The interfacial energies for aqueous systems including Lifshitz–van der Waals (LW) and acid–base (AB) interaction energy per unit area between two infinite planar surfaces are, respectively, determined from [20]:

$$\Delta G_{d_0}^{LW} = 2\left(\sqrt{\gamma_w^{LW}} - \sqrt{\gamma_m^{LW}}\right)\left(\sqrt{\gamma_a^{LW}} - \sqrt{\gamma_w^{LW}}\right) \tag{7}$$

$$\begin{aligned}\Delta G_{d_0}^{AB} = {} & 2\sqrt{\gamma_w^+}\left(\sqrt{\gamma_m^-} + \sqrt{\gamma_a^-} - \sqrt{\gamma_w^-}\right) \\ & +2\sqrt{\gamma_w^-}\left(\sqrt{\gamma_m^+} + \sqrt{\gamma_a^+} - \sqrt{\gamma_w^+}\right) \\ & -2\left(\sqrt{\gamma_m^+\gamma_a^-} + \sqrt{\gamma_m^-\gamma_a^+}\right)\end{aligned} \tag{8}$$

where γ^{LW} and γ^{AB} is the LW and AB component of surface tension, respectively, J/m^2; $\gamma+$ and $\gamma-$ are electron-acceptor and electron-donor parameters of the polar component of the surface tension, respectively, J/m^2; the subscript m denotes the membrane, w refers to the DI water and a corresponds to SA.

The free energy of interaction between two identical surfaces immersed in water (ΔG_{sws}) was used to evaluate the surface hydrophobicity/hydrophilicity of a substance.

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