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Coagulation–bubbling–ultrafiltration: Effect of floc properties on the performance of the hybrid process

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HIGHLIGHTS

- A novel hybrid process of coagulation–bubbling–ultrafiltration was proposed.
- Relationship of air bubble size and gas flow rate was studied.
- Bubbles and floc properties were measured to explain membrane performance.
- Concentration polarization resistance was reduced with air bubbles injected.
- Floc separation by air bubbles holds great promise in reducing membrane fouling potential.

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ABSTRACT

A novel hybrid process of coagulation–bubbling–ultrafiltration was proposed to study membrane fouling phenomena by surface water. Relationship of bubbles, flocs and the hollow fibers was explored. When applying less than 20 mL/min gas flow rate, membrane fouling was accelerated with air bubbles introduced. When gas flow rate increased further to 40 mL/min and 60 mL/min, TMP showed a two-stage development trend, which was a fast development in the first few hours followed with a relatively slow development after about 4 h. Unified membrane fouling index (UMFI) increased from 0.00216 (without bubbles) to 0.00274 m^2/L (40 mL/min gas flow rate) and 0.00219 m^2/L (60 mL/min gas flow rate). As gas flow rate increased, bubble size became bigger, and its distribution range became wider, resulting in higher shear rate in the ultrafiltration column, which led to severe floc breakage. Flocs of small size and compact structure accelerated membrane fouling, resulting in highest UMFI value under 40 mL/min gas flow rate. However, under 60 mL/min gas flow rate, with largest bubbles and highest shear rate examined in this study, concentration polarization was effectively limited. As a result, TMP development slowed down when pore blockage reached equilibrium.

1. Introduction

Membrane process of ultrafiltration (UF) applied in water treatment received much attention in recent years due to its small footprint, high-quality finished water and relatively low cost [1,2]. Hollow fiber is particularly popular because of its high packing density and the convenience of modularized installation [3]. But like other membrane separation process, hollow fiber ultrafiltration is faced up with the problem of membrane fouling which usually results from particles accumulating on the membrane surface and colloids plugging in membrane pores [4]. Several methods including pretreatment before membrane filtration and hydrodynamic disturbance during filtration were developed to solve this problem [5,6].

Huang et al. reviewed the major pretreatment approaches applied before low-pressure membrane filtration [5]. They concluded that compared with adsorption, preoxidation and prefiltration, coagulation has been the most successful pretreatment for fouling reduction. Coagulant type, source water quality and coagulation condition can influence membrane filtration significantly [7]. When coagulant was added in two stages, internal pore fouling could be remarkably limited, Liu et al. explained this as larger and more irregular flocs formed by the addition strategy [8]. Yu et al. observed a lower TMP increase with flocs formed in breakage and regrowth process [9]. They found that breakage followed by regrowth led to lower fractal dimension of flocs and decreased amount of small micro-flocs. Barbot et al. also emphasized the importance of large flocs with open branches to promote permeate flux [10]. Thus, it seems that floc size distribution and structure characteristic had a significant influence on membrane performance.

Among many hydrodynamic disturbance methods such as vibration, pulsating flow and vortex generation, gas sparging has been proved to

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be an effective, simple and low cost technique in membrane filtration [11]. Researches of gas sparging have been applied in different kinds of membrane modules like flat sheet membrane [12], tubular membrane [11] and hollow fiber membrane [13,14]. Membrane performance was related with bubble characteristics, gas flow rate and sparging frequency in these studies. When bubbling was applied inside the membrane modules, larger bubbles or bubble slugs are preferred due to their ability to occupy most of the channel thus promoting mass transfer. For submerged modules, using particle image velocimetry, Yeo et al. calculated bubble induced shear stress around hollow fibers and revealed that one of the key parameters in promoting membrane performance was the fluctuation in shear stress [15]. In their research, small bubbles with high frequency showed some beneficial against large ones. Tian et al. investigated influence of bubble size on the fouling of immersed hollow fiber membrane and concluded that the smallest bubble with diameter of 3.5 mm was most effective [1].

However, hybrid process of coagulation–ultrafiltration and bubbling–ultrafiltration mentioned above were both conducted independently. Scarcely literature integrated the three processes of coagulation, gas sparging and ultrafiltration to explore the interaction of bubbles, particles/flocs and membrane. When air bubbles are introduced in the process of filtration followed by coagulation, bubble induced hydrodynamic disturbance can show some impact on membrane performance. Moreover, air bubbles may adsorb some small flocs and bring them to water surface by floatation or bubble induced shear stress may lead to floc breakage and size/strength redistribution. It is of paramount necessity to reveal how bubbles influence floc property and membrane fouling potential in such a hybrid process of coagulation–bubbling–ultrafiltration.

In this paper, a novel hybrid process containing coagulation, air bubbling and ultrafiltration to deal with lake water was developed. The interaction of bubbles, particles/flocs and membrane was integrally studied.

2. Materials and methods

2.1. Source water and coagulant

Source water used in this study was obtained from Mingyuan Lake located in Jiang'an campus of Sichuan University. Lake water quality was continuously monitored during the experiment. Water quality was shown in Table 1. Aluminum sulfate hydrate ($Al_2(SO_4)_3 \cdot 18H_2O$; Sigma Aldrich, analytical grade) was used as coagulant. Stock solution was prepared at a concentration of 0.01 M in a measuring flask and saved in dark for use.

2.2. Jar test

Jar test was performed in a floculator (ZR-6, Zhongrun Co., Ltd., China) to determine optimal alum dosage based on UV_{254} and turbidity removal. Firstly, a rapid mixing with rotation speed of 300 rpm was maintained for 1 min, and then rotation speed was reduced to 60 rpm allowing floc growth for 12 min.

Table 1
Characteristic of Mingyuan lake water.

Parameter	Value
pH	7.95–8.63
Turbidity (NTU)	1.08–2.27
Temperature (°C)	21.5–24.6
TOC (mg/L)	2.16–2.92
UV_{254} (cm^{-1})	0.030–0.052
SUVA (mg/L·m)	1.30–1.82

2.3. Floc property measurement

Use of fractal dimension to describe structure characteristic of flocs was presented in numerous studies [16–18]. Detailed concept and measurement of fractal dimension were interpreted in Bushell and coworkers' review [19]. In this study, fractal dimension for cross-sectional area of flocs was determined by box-counting method using the following equation [16,19]:

$$D_f = \lim_{L\to 0} - \frac{\log(N)}{\log(L)} \tag{1}$$

where N is the number of pixel needed to cover floc image and L is pixel size.

After in-line coagulation, outlet of flocculation pipe was put on a flat microscope slide to collect flocs. Coagulated water flowed slowly onto the slide and less than 1 mL sample was collected each time to ensure no floc breakage occurs. Flocs in the ultrafiltration column were collected in the same way at the end of ultrafiltration circle. Image of flocs was captured by a microscope (CX41, Olympus Co., Ltd., Japan) with CCD camera and further processed by a software package Image Pro Plus (Version 6.0, Media Cybernetics Inc., US). Specifically, flocs were separated from their background by brightness and contrast adjustment, sharp edge detection and random noise filtering, which was the same as Chakrabroti et al. did in their research [17,18]. Tests were performed in advance to reach the best similarity between flocs in source image and the processed one. After preprocessing, the longest side value of flocs, which is the parameter adopted to feature floc size, was determined by Image Pro Plus software directly. Box-counting fractal dimension of flocs was calculated using Eq. (1) on a professional graphic processing platform MATLAB 7.0 (Mathworks Inc., USA).

2.4. Air bubbling

Air flow was generated by an air pump and controlled through a glass rotameter. Air bubbles were introduced by a porous stainless steel disk (Nanjing Institute of Metallic Membrane, China) with diameter of 30 mm. According to the manufacturer, the disk has a pore size of 0.1–5.0 μm and porosity of 25–50%. The image of bubbles in ultrafiltration column was captured by a digital camera. Bubble size was determined using Image Pro Plus by applying diameter of the column as scale. More than ten pictures were captured and at least 60 bubbles were calculated to perform bubble size distribution analysis.

2.5. Experiment set-up

Fig. 1 shows the apparatus of the coagulation–bubbling–ultrafiltration set-up. A constant-level tank was used to maintain hydrodynamic condition. Magnetic stirring apparatus was used to provide hydrodynamic gradient in coagulation tanks. In rapid mixing tank, rotation speed was set at 300 rpm for 1 min, then reduced to 60 rpm for 6 min in both of the two flocculation tanks, which was identical with jar test. After in-line coagulation, feed water entered membrane module directly and was sucked by a peristaltic pump (BT100-2J, Longer Pump, China). The ultrafiltration module consists of inner tube and outer tube. Hollow fibers with mean pores of 0.02 μm (Litree Purifying Technology Co., Ltd., China) were installed between the two tubes. Effective length of the fibers is 220 mm, with inner and outer diameter of 1.0 mm and 1.8 mm, respectively. 16 fibers were installed corresponding to a total area of 0.02 m^2. The volume of the annular cylinder is 300 mL. Permeate flux was set at a constant value of 20 $L/m^2 \cdot h$. Thus, the hydraulic retention time (HRT) of feed water in the membrane module is 43 min. TMP data was monitored by a pressure gauge.