Journal of Industrial and Engineering Chemistry 21 (2015) 363–368

Contents lists available at ScienceDirect

Journal of Industrial and Engineering Chemistry

journal homepage: www.elsevier.com/locate/jiec

Split injection of coagulant to enhance the water permeability of a cake layer in a coagulation–microfiltration process

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ARTICLE INFO

Article history:
Received 11 February 2014
Accepted 24 February 2014
Available online 2 March 2014

Keywords:
Coagulation
Split injection
Microfiltration
Cake layer
Permeability

ABSTRACT

A split injection of an aluminum coagulant at the initial stage of rapid mixing was investigated to improve the water permeability of a cake layer composed of chemical flocs on a microfiltration membrane surface. The split ratio of the first to second injections was changed to find an optimized injection condition. With an increase in the split ratio, the cake compressibility index decreased because compact chemical flocs were produced with reduced dosage of the second injection, but the specific cake resistance increased due to small size of chemical flocs with the insufficient amount of coagulant at the second injection. At a small split ratio, the particle size distribution of the split injection was similar to that of normal single injection, and the compressibility index was small, leading to better water permeability of the cake layer at higher pressure.

1. Introduction

A coagulation–membrane filtration process has been widely studied as an efficient water treatment process [1–6]. It is applicable not only to new construction of water treatment plants but also to modification of existing ones, which is relatively simple because coagulation and flocculation basins may be re-utilized with no or slight modification. In addition, a membrane filtration process can be substituted for a sedimentation as well as a sand-filtration process so as to considerably reduce the footprint of overall processes. Without sedimentation, however, chemical flocs produced in coagulation and flocculation processes may deposit on membrane surface during filtration to form a cake layer especially in dead-end filtration [7–11]. The water permeability of the cake layer affecting overall membrane filterability depends significantly on the properties of chemical flocs [12–17]. Therefore, coagulation conditions affecting chemical floc formation are of great important in a coagulation–membrane filtration process.

The major properties related to membrane filtration are the size and structure of chemical flocs, which rely on diverse coagulation conditions such as coagulant dosage as well as the type of coagulants, concentration and type of constituents in raw water, solution pH and temperature, mixing time and intensity, and so on [18–34]. More dosage of coagulant or higher ionic strength by adding more salt generally induces more charge neutralization or double-layer compression to probably make chemical flocs larger [35]. Wang et al. [36] observed that average floc size increased as a function of the dosage of aluminum chlorohydrate. Xu et al. [37] reported that, in coagulation experiments with humic acids and two types of coagulants containing polymeric aluminum species, floc size increased with increasing dosage. The dosage or salt concentration also affects floc structure. Waite et al. [38] showed that hematite (α-Fe_2O_3) aggregates became compact when KCl concentration was reduced. Slow aggregation rate due to low concentration of coagulant or salt led to the compact structure of flocs [38,39].

This effect can be applied to coagulation–membrane filtration processes in different manner. A certain dosage of coagulant is selected generally by preliminary experiments in which water quality is investigated to find optimized dosage. With the selected dosage, it is difficult to change floc size and structure practically because pH, temperature, mixing time and intensity are usually fixed in real water treatment plants even though they are variously changeable in lab-scale experiments. When the dosage is split up into two or more, however, the concentration ratio of coagulant to constituents in raw water can be adjusted to control floc properties. Liu et al. [40] demonstrated that two-stage coagulant injection into coagulation (slow mixing) and flocculation (rapid mixing) stages

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http://dx.doi.org/10.1016/j.jiec.2014.02.046
1226-086X/

with 3:1 of coagulant injection ratio was successful to achieve the improvement of membrane filtration performance. By the injection methods, floc size was enhanced and floc structure was more open to be favorable to reducing rise of trans-membrane pressure.

In this study, a split injection of coagulant only at the initial stage of rapid mixing was investigated to improve the water permeability of a cake layer made up of chemical flocs on a microfiltration membrane surface. Specific cake resistance, which is inversely proportional to the water permeability, was obtained from filtration data to identify at which split ratio of the first to second injections the split-injection method could enhance membrane filterability compared to normal single-injection one. Cake compressibility index was calculated to assess the effect of pressure on cake permeability. Particle size distributions were measured to examine how the cake properties depended on the characteristics of chemical flocs.

2. Experimental

2.1. Materials and analysis

Water samples were taken from Lake Maeji in the Republic of Korea. The raw water quality is listed in Table 1. Aluminum sulfate hydrate (Alum) (54%wt. as $Al_2(SO_4)_3$, Duksan pure chemicals, Republic of Korea) was used as coagulant to be injected into the water samples. The microfiltration membrane used for filtration experiments was cellulose nitrate 0.45-μm membrane filter (Whatman®, GE healthcare UK Ltd., UK). Every membrane filter was pressurized for pre-compaction by the filtration of deionized water at 50 kPa using a filtration cell unit (Amicon® 8050, Millipore, USA) with nitrogen gas before measuring water permeability followed by each filtration experiment (Fig. 1).

Turbidity of water samples was measured with a turbidimeter (2100N, Hach company, USA). UV_{254}, which represents UV absorbance of filtered water samples at 254 nm, indicating relative amount of dissolved organics in water, was measured with a spectrophotometer (8453 UV-Vis, Agilent Technologies, USA). Dissolved organic carbon (DOC) was measured with a total organic carbon analyzer (Sievers® 5310C, GE analytical instrument, USA) after filtering raw water samples. The filters used before measuring UV_{254}, DOC, and turbidity of filtered water were 0.45-μm syringe filters (Advantec®, Toyo Roshi Kaisha Ltd., Japan).

Particle size distributions of raw and coagulated water were investigated using a particle size analyzer (S3500, Microtrac, USA) for the range as large as a thousand μm and a zeta-potential and particle-size analyzer (ELS-Z, Otsuka Electronics, Japan) for small size less than 4 μm.

2.2. Coagulation experiments

800 mL of each water sample was agitated during coagulation and flocculation with a jar-tester (Mtops® SF-4, Misung S&I, Republic of Korea). Rapid and slow mixing steps were conducted at 150 rpm for 3 min of coagulation and 20 rpm for 15 min of flocculation, respectively. A coagulant stock solution with the concentration of 8000 ppm as Al_2O_3 was prepared in advance with the alum and deionized water to be injected into water samples during coagulation by the rapid mixing. The temperature of water samples was controlled in the range of 19.8–20.1 °C by using a water bath. Water pH at the initial stage of coagulation was adjusted to be 7.49 ± 0.05 with 1-N sodium hydroxide solution for both single and split injections. The injection amount of the hydroxide solution as a function of coagulant dosage was estimated at preliminary experiments by dropping the hydroxide solution into the same water sample mixed with the coagulant to make water pH to be ca. 7.5. For single injection, whole of the coagulant and corresponding hydroxide solution were injected into a water sample at one injection whereas, for the split injection, the first injection of a portion of the coagulant and hydroxide solutions was followed by the second injection of the remainder after 10-s time interval. The coagulant split ratios of the first to second injections were 1:1, 1:3, and 1:7 in this study.

Table 1
Characteristics of water samples used in this study.

Parameters	
pH (at 20 °C)	7.29 ± 0.09
Turbidity[a] (NTU)	3.00 ± 0.43
UV_{254}[b] (cm^{-1})	0.037 ± 0.001
DOC[c] (ppm C)	1.80 ± 0.03
Suspended solid (mg/L)	11 ± 2

[a] Turbidity without filtration using 0.45-μm filter.
[b] UV absorbance at 254 nm.
[c] Dissolved organic carbon.

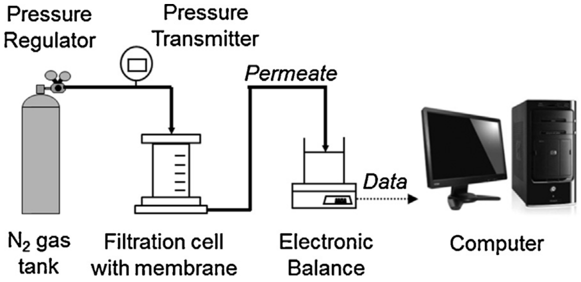

Fig. 1. Schematic diagram of microfiltration setup.

2.3. Filtration experiments

2.3.1. Formation of cake layer on microfiltration membrane

Each coagulated water sample after the rapid and slow mixing was left to sit for 30 min of gravity settling. After supernatant was removed, the remaining water including chemical flocs concentrated by the settling was transferred to the filtration cell unit. It was filtered at 5 kPa by using the cell unit with a pre-compacted 0.45-μm membrane filter to form a cake layer on the membrane filter (Fig. 1).

2.3.2. Measurement of filterability of the cake layer

Supernatant was filled with the filtration unit over each cake layer and filtered at 10, 15, 20 and 25 kPa in steps to measure the flow rate of permeate. The mass of permeate was measured with an electronic balance (BSA-2202S, Sartorius, Germany), and the measured mass data were sent and logged to a computer every 5 s. The mass was converted to the volume of permeate using water density, and then water flux, J, was calculated:

$$J = \frac{\Delta V}{A_m \cdot \Delta t} \tag{1}$$

where ΔV is the volume of permeate, A_m is effective membrane surface area, which was $1.2 \times 10^{-3}\ m^2$ in this study, and Δt is time interval. The hydraulic resistance of cake layer, R_c, was estimated using the following equations:

$$J = \frac{\Delta P}{\mu(R'_m + R_c)} \tag{2}$$

where ΔP is trans-membrane pressure, μ is the viscosity of permeate water, and R'_m is the hydraulic resistance of fouled membrane [41]. R'_m was estimated with separate filtration experiments using 0.45-μm filtered supernatant which contained