Chemical Engineering Journal 280 (2015) 634–642

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Effects of epichlorohydrin–dimethylamine on coagulation and membrane performance of ferric chloride in coagulation–ultrafiltration hybrid process

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HIGHLIGHTS

- Coagulation efficiency of FC on HA–Kaolin could be enhanced apparently by DAM–ECH.
- Initial pH would remarkably influence the coagulation performance and floc properties.
- Application of DAM–ECH result in larger flocs with better recovery ability.
- DAM–ECH addition would generate flocs with loose and open structure.
- Membrane performance was significantly improved by DAM–ECH.

ARTICLE INFO

Article history:
Received 12 March 2015
Received in revised form 3 June 2015
Accepted 4 June 2015
Available online 18 June 2015

Keywords:
Membrane fouling
Epichlorohydrin–dimethylamine
Ferric chloride
Floc properties
Coagulation–ultrafiltration

ABSTRACT

Cationic polymer epichlorohydrin–dimethylamine (DAM–ECH) was applied as coagulation aid of ferric chloride (FC) in treatment of humic acid–kaolin (HA–Kaolin) simulated water in this study. Impacts of DAM–ECH on coagulation performance, flocs characteristics and membrane fouling of FC in coagulation–ultrafiltration (C–UF) process were investigated under different pH conditions. Results showed that the removal rates of UV_{254} (ultraviolet adsorption at 254 nm) and DOC (dissolved organic carbon) of FC/DAM–ECH was much better than that of FC at same dosage. The optimum coagulation condition was FC at 10 mg/L with FC/DAM–ECH mass ratio (MR) of 3:1 based on the consideration of treatment cost and performance. Moreover, turbidity, UV_{254} and DOC removal rates increased when pH increased from 4 to 6 and then decreased as pH grew from 7 to 10. Thus, the optimum pH for coagulation was also determined to be 6 accordingly. Addition of DAM–ECH also increased sizes of produced flocs and made them looser in structure comparing with FC–HA flocs. Meanwhile, results obtained from ultrafiltration experiments showed that membrane fouling would be reduced by introducing DAM–ECH in coagulation treatment; the optimum membrane performance was given by FC/DAM–ECH under pH 6. In conclusion, proper dose of FC/DAM–ECH under pH 6 could ensure an ideal HA removal rate and meanwhile increase operating duration of C–UF system by restricting membrane fouling.

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1. Introduction

It is now widely recognized that as a kind of natural organic matter (NOM) which commonly existing in most of drinking water resources, humic substances are jeopardizing human health [1,2]. Thus, removal of humic substances has drawn worldwide attention recent decades in modern water treatment. As an extensively used coagulant in water treatment processes, ferric chloride (FC) can remove humic acid molecules in water bodies by forming strong and large flocs [3–5]. Moreover, many researchers have proved that compounding of iron salt flocculants with organic polymer would be more effective in HA and turbidity removal than single use of iron salts [6]. Thus, a cationic polymer epichlorohydrin–dimethylamine (DAM–ECH) was applied as coagulation aid of ferric chloride (FC) in this study. Meanwhile as a membrane filtration process, ultrafiltration has been broadly applied as postprocessing of water treatment process to further improve the reduction of

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http://dx.doi.org/10.1016/j.cej.2015.06.011
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particle concentration and NOM in portable water resources. This is because that UF processes could not remove most of dissolved organic matter from water bodies because of its large pore sizes [7]. Another main bottle-neck in ultrafiltration utility is the drop of permeate flux caused by membrane fouling, which would hugely increase the power demand to maintain an acceptable flow rate of the UF device [8,9]. It has been proved that pretreatment (such as adsorption, coagulation or peroxidation) can remarkably reduce improve membrane performance by achieving larger permeate flux in combined water treatment systems, in which coagulation has been proved to be more effective than the other pretreatments in reduction of membrane fouling [10]. Breakthroughs have been made by previous researches in all aspect of C–UF system, including influence of coagulants/flocculants by Xu et al. and Zhao et al. [11,12]; optimization of operating conditions in application of C–UF in drinking water treatment and pilot-scale hybrid municipal wastewater reclamation [13,14]. Therefore, coagulation–ultrafiltration hybrid process (C–UF) is becoming more and more popular in high quality water treatment processes for its high removal efficiency of NOM. Generally, C–UF is significantly affected by many elements such as feed water types, coagulation methods and hydraulic conditions (shearing force) [15].

As mentioned above, combination of coagulation and ultrafiltration process is mutually beneficial which can further improve the water quality comparing with single use of each process. Furthermore, combine use of these two processes should be optimize through designed experiment processes. Thus, proper addition method of coagulants was studied to achieve acceptable coagulation and membrane performance in this study.

2. Materials and methods

2.1. Preparation of coagulants

FC solution with concentration of 2 g/L (as Fe^{3+}) was prepared by an analytical reagent $FeCl_3 \cdot 6H_2O$ (CAS No: 10025-77-1). 0.9661 g of $FeCl_3 \cdot 6H_2O$ was completely dissolved in 100 ml of deionized water stirred by a magnetic stirring apparatus until the solution became clarified.

DAM–ECH copolymer was polymerized by analytical reagent (A.R.) epichlorohydrin and dimethylamine (A.R.) with ethanediamine (A.R.) as cross-linker. The polycondensation reaction that producing DAM–ECH copolymer was conducted in a 250 mL of four-neck round bottom flask. In addition, several facilities were required during the synthesis process: a mechanical stirrer, a thermometer, a dropping funnel and corresponding glass spigots. Certain dosage of epichlorohydrin was poured into the four-neck round bottom flask and meanwhile the environmental temperature within the flask was kept at 10 °C by a thermostated water bath device. Then dimethylamine was constantly dripped into the flask using a dropping funnel, and the mixture was under steady stirring throughout the whole dripping process. At last, certain dose of ethanediamine was introduced into flask under continuous stirring as the cross-linker [5].

2.2. Simulated water sample

Humic acid–kaolin (HA–Kaolin) water was used as raw water in this study. The HA–Kaolin stock solution was prepared as follow: 1.0 g of HA powder (Aladdin, Shanghai, China) and 0.40 g of NaOH (Tianjin Damao Co., Tianjin, China) were weighted and dissolved in deionized water under continuously stirring of a magnetic stirring apparatus till completely dissolved; then the mixture was transferred into a 1000 mL volumetric flask and diluted to volume (1 L).

Kaolin stock solution was applied in this study to adjust turbidity of HA simulated water, which was prepared as follow: 5.0 g of kaolin (Tianjin Kermel Chemical Reagent Co., Ltd, China) was weighted and mixed with 1.0 L of deionized water, the suspension liquid were stirred by a magnetic stirring apparatus for at least 30 min; then the upper 500 mL liquid was withdrawn for later use.

HA–Kaolin simulated water of this study was prepared by diluting 10 mL of the HA stock solution into 1.0 L of tap water. Initial turbidity of the raw water was adjusted to 15.0 ± 0.5 NTU using kaolin stock solution. Characteristics of raw water were as follow: UV_{254} (ultraviolet absorbance at 254 nm) = 0.320 ± 0.010.

$$\text{DOC(dissolved organic matter(DOM) concentration)} = 3.55 \pm 0.03\ \text{mg/L}$$

$$\text{Raw water pH} = 8.32 \pm 0.02$$

Many researchers have found that UV_{254} can be used as indicator of humic substances and aromatic organic matter contain C=C and C=O in water bodies, that higher humic concentration in water always show high UV_{254} adsorption [13,16]. Furthermore, the relationship between UV_{254} and DOC have been studied by many researchers, results showed that 70% to 80% of DOC removal was aquatic humic fraction with high UV_{254} absorption value, only 10% of DOC was hydrophobic organic with low UV_{254} adsorption. In other words, UV_{254} removal should be positive correlated with DOC removal [16].

The initial pH of the raw water sample was adjusted by 0.1 M HCl and 0.1 M NaOH solution when necessary during the tests.

2.3. Jar test

In this study, jar tests were conducted to determine the optimum coagulation method for HA–Kaolin water, which were carried out by a program-controlled flocculator with six 1.5 L cylindrical plexiglass beakers and six flat paddle impellers (ZR4-6, Zhongrun Water Industry Technology Development Co. Ltd., China). During jar tests, 6 L of prepared HA–Kaolin solution was filled into six breakers averagely (1 L of raw water in each breaker). And the operation program was preseted as follow: first, synthetic water sample in each breakers was stirred rapidly under 200 rpm (rpm) for 0.5 min; then certain dose of coagulant was added into each breakers, followed by another stirring period of 200 rpm for 0.5 min; following the third period, the mixture in each breakers would be stirred under 200 rpm for 1.5 min for dispersion of coagulant; in the fourth stage, floc would grow under slow stirring speed of 40 rpm for 10 min; during the final stage, the shearing force would be removed for flocs to settle for 20 min. After the sedimentation period, 200 ml of supernatant water was withdrawn from each jar to use as water samples in the following tests (Turbidity, DOC, UF, etc.). Turbidity of water sample would be measured by a 2100P turbidimeter (Hach, USA) and zeta potential was measured using a Zetasizer 3000Hsa (Malvern Instruments, UK) at room temperature (around 25 °C). Each water sample was filtered through a 0.45 μm fiber membrane before DOC analysis using a Shimadzu TOC-VCPH analyzer and UV_{254} measurement by a UV-754 UV/VIS spectrophotometer (Precision Scientific Instrument Co. Ltd., Shanghai, China).

2.4. On-line monitoring of floc formation, breakage and re-growth

The evolution of flocs during coagulation process was monitored by a laser diffraction instrument (Malvern Mastersizer 2000, Malvern, UK). Water samples were monitored through the

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