Separation and Purification Technology 123 (2014) 1–8

Contents lists available at ScienceDirect

Separation and Purification Technology

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

Coagulation performance and flocs characteristics of recycling pre-sonicated condensate sludge for low-turbidity surface water treatment

Zhiwei Zhou [a], Yanling Yang [a,*], Xing Li [a], Weiqiang Wang [a], Yan Wu [a], Changyu Wang [b], Jianliang Luo [b]

[a] Key Laboratory of Beijing for Water Quality Science and Water Environment Recovery Engineering, Beijing University of Technology, Beijing 100124, PR China

[b] Beijing Waterworks Group Co., Ltd., Beijing 100031, PR China

ARTICLE INFO

Article history:
Received 17 September 2013
Received in revised form 26 November 2013
Accepted 1 December 2013
Available online 11 December 2013

Keywords:
Ultrasonic pre-treatment
Drinking water treatment sludge
Recycling
Flocs characteristics
Breakage and re-growth

ABSTRACT

This research innovatively investigated the coagulation performance of recycling pre-sonicated condensate drinking water treatment sludge under different ultrasound (US) conditions (ultrasonic time of 5, 10, 15, 20 and 30 min and US frequencies of 25, 40, 125 and 160 kHz) for low-turbidity surface water treatment. The formation, breakage and re-growth of re-coagulated flocs generated by recycling processes were examined using photometric dispersion analyzer to explore flocs growth properties, flocs strength and re-aggregation potential. The morphological analysis was further conducted to study the structural properties of the broken flocs to elucidate the reversibility/irreversibility of re-growth process. The results indicated that 25 or 40 kHz within 5 min sonication was more favorable for turbidity removal, while 125 or 160 kHz with 10 or 15 min for organics removal. The recoverability of broken flocs was irreversible for the recycling processes regardless of US frequencies. Additionally, the flocs formed preliminarily exposed to 25 or 40 kHz were stronger and more resistant to breakage, and the recoverability was more irreversible. Furthermore, the flocs formed at 25 or 40 kHz were larger and more irregular with more porous and ramose structure. The average size obtained from morphological analysis could also strongly demonstrate the irreversibility of recovery process.

1. Introduction

In water or wastewater treatment, coagulation/flocculation is an essential process for the removal of particulate and/or organic matters. During flocculation, a large quantity of sludge is formed, which generally characterized with elevated concentrations of inorganic or organic substance, as well as precipitation hydroxide that derived from inorganic alum-based or ferric-based coagulants. Sludge return process is an effective method to enhance pollutants removal [1–5]. However, during recycling the breakage of recycled flocs will result in the release of organics and increase the number of fine particles, as well as distribution variability of broken particles, which would deteriorate the coagulated water quality. Therefore, it is necessary to degrade or solubilize the constituents that difficulty to be eliminated by coagulation prior to recycling [6], and improve the destabilized flocs characteristics of recycling sludge for subsequent solid–liquid separation.

Ultrasound (US), one of the most effective pre-treatment and environmentally benign option to disintegrate the sludge flocs size, increase the adsorption sites on pollutants and oxidize/inactivate the refractory toxic substances in sludge, has been under consideration over the last decade [2,7–10]. Sonication can cause a series of compression and rarefaction cycles leading to the generation of cavitation bubbles. Millions of these bubbles implode yielding localized temperatures as high as 5000 °C, pressures of 100 MPa and free radicals e.g., $^{\cdot}OH$, $HO_2^{\cdot}$ and O· [11]. Chu [10] has pointed out that US has no effects on the surface charges of the suspended particles of activated sludge regardless of sonication time under ultrasonic energy of 0.11 and 0.33 W/mL. Lehne et al. [12] found that specific energy of 3000 kJ kg^{-1} TS (total solid) or more could maximally decrease the particles' size down to 5 μm.

Additionally, according to the findings of Guan et al. [2], the positive effects of US on pollutants removal of recycling process were mainly reflected in producing large quantity particles with moderate size and larger specific surface. Thus, recycling pre-sonicated sludge can somewhat affect the coagulation efficacy and resultant flocs characteristics. In particular, when recycling pre-sonicated sludge, the size of particles in raw water and that in pre-sonicated sludge is essentially different and both can interact with fresh addition of coagulation. Furthermore, the particles in raw water can be adsorbed onto the surface of the destabilized pre-sonicated

* Corresponding author. Tel./fax: +86 10 67391726.
E-mail address: yangyanling@bjut.edu.cn (Y. Yang).

1383-5866/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.seppur.2013.12.001

particles. As a consequence, the flocs formation characteristics and pollutants removal efficacy will become complex, as compared to conventional coagulation without sludge.

Generally, the effectiveness of flocculation is measured by the parameters such as residual turbidity or organics removal because they are directly related to the solid–liquid separation and water quality. However, they only provide limited information of flocculation process. The information of flocs size distribution and structure would be much needed [13]. The flocculation dynamic monitoring technology, photometric dispersion analyzer (PDA), was widely adopted to observe the flocs formation, breakage and re-growth properties. In this study, the PDA was used to explore flocs growth properties, flocs strength and re-aggregation potential in recycling processes.

Although the literature on US as pre-treatment of drinking water treatment sludge (DWTS) was rare, in current study US was firstly applied to pre-sonicate DWTS, aiming to enhance subsequent recycling processes. The overall purpose of this research was, therefore, to compare and assess the coagulation performance and re-coagulated flocs characteristics, as compared to control without DWTS. The formation, breakage and re-growth properties of re-coagulated flocs generated by recycling processes were examined. The morphological analysis was further carried out to study the structural property of the broken flocs to explore the reversibility/irreversibility of re-growth flocs generated by recycling processes.

2. Materials and methods

2.1. Raw water and DWTS

The raw water and DWTS was simultaneously collected from a drinking water treatment plant in northern China during the period of this study. The raw DWTS was the water treatment residual generated by ferric salt coagulants. The average total solids (TS), volatile suspended solids (VS), total chemical oxygen demand (TCOD) concentration of raw DWTS was 4.56 g L^{-1}, 3.37 g L^{-1}, 132.07 g L^{-1} respectively and the soluble chemical oxygen demand (SCOD) in supernatant of raw DWST was 27.59 g L^{-1} and its pH was 7.60, as indicated in Table 1. All tests started within 2 h after sampling to prevent sludge changes.

The turbidity and levels of organic carbon of the tested raw water was low, which were the same characteristics as low-turbidity and micro-polluted water. The main physicochemical characteristics of the raw water were summarized in Table 1.

2.2. Recycling trials

The procedure of the whole experiment was as follows: the raw DWTS was firstly exposed to the US apparatus under different US conditions, and then the sonicated DWTS was sampled at a given sonication time and put aside for settling 120 min. The supernatant of each sonicated DWTS was discharged meanwhile maintaining the condensate factor of the thickened sludge was 0.6–0.8. Finally, the pre-sonicated condensate sludge was added into the beaker of the jar tester at the beginning of the flocculation stage of 120 rpm (seen the coagulation procedure in Section 2.2.2).

2.2.1. Sonication of DWTS

For each sonication experiment, each 15 L of raw DWTS sample was sonicated with the sound waves emitting from a sonochemical reactor with configuration of 250 mm (length) × 250 mm (width) × 300 mm (height), equipped with a transducer and an outlet. Sonication of DWTS was conducted at US frequencies of 25, 40, 125 and 160 kHz using an ultrasonic batch system under corresponding ultrasonic energy density as low as 0.03 W/mL respectively. The actual ultrasonic power dissipated into the suspension was measured using calorimetry [14]. Such low energy densities were used since in large scale applications higher intensities may not always be practical due to the consideration of energy consumption. Each sample was pre-sonicated for 5, 10, 15, 20 and 30 min. At each sampling point, about 250 mL pre-sonicated sludge was withdrawn. Each sonication experiment was repeated twice and average values were reported. All US reaction was conducted under air atmosphere without controlling the rise of temperature considering the practical application.

2.2.2. Determination of coagulant dosage and recycling ratio of DWTS

A series of jar test using ZR4-6 (Zhongrun, China) was primarily carried out to determine the optimum dosage of $FeCl_3$ and recycling ratio of unsonicated DWTS. Control test without DWTS was conducted at different $FeCl_3$ dosages ranging from 2.17 to 15.20 mg/L (calculated as Fe) and constant initial pH about 8.00. The jar test for control test or recycling trial of control was initially started with a rapid mixing at 350 rpm($G = 449\ s^{-1}$); after 1 min a certain dose of $FeCl_3$ was added with or without DWTS (recycling ratio 2–10%) was introduced, with 1 min of rapid mixing at 120 rpm($G = 106\ s^{-1}$) again; the sample was subsequently exposed to a slowing mixing at 50 rpm ($G = 33\ s^{-1}$)for 15 min followed by a 20 min settling period, after which the supernatant samples were collected for the measurements of turbidity, UV_{254} absorbance and DOC.

2.3. Flocs formation, breakage and recovery

Experiments on the kinetics of formation, breakage and re-growth of re-coagulated flocs were performed using the "turbidity fluctuation" technique, as used in the Photometric Dispersion Analyzer. The procedures were as follows: a rapid mixing at 350 rpm ($G = 449\ s^{-1}$) for 1 min, then with 1 min of rapid mixing at 120 rpm ($G = 106\ s^{-1}$) followed by a slow mixing at 50 rpm ($G = 33\ s^{-1}$) for 3 min. Flocs were then exposed to an increased shear of 350 rpm ($G = 449\ s^{-1}$) for 1 min followed by a restoration of the 50 rpm ($G = 33\ s^{-1}$) slow mixing phase for 11 min re-growth. A slow mixing at 50 rpm ($G = 33\ s^{-1}$) lasting 3 min in this study was enough to reach a steady balance between breakage and re-aggregation and obtain a plateau Flocculation Index (FI), as shown in Fig. 1S (seen in Supplementary data).

The average transmitted light intensity (dc value) through the flowing sample and the root-mean-square value (rms) of the fluctuating component are monitored. The ratio (rms/dc) provides a sensitive measure of particle aggregation and it is often called FI. The ratio value is strongly correlated with the respective floc size and always increases as flocs grow larger. In this work, after the FI value reached an initial steady value, the FI value was recorded by a PC data acquisition system at 2s intervals.

In the floc formation process, three parameters were adopted to analyze the data collected by PDA: a flocs growth rate of the growth region, a time-weighted average steady-state ratio value

Table 1
Main physicochemical characteristics of raw water and DWTS.

Analytes (Units)	Raw water Average	Analytes (Units)	Raw DWTS Average
Temperature (°C)	15	Temperature (°C)	15
pH	8.09	pH	7.60
Turbidity (NTU)	2.09	Moisture content (%)	99.54
UV_{254} (cm^{-1})	0.022	TS (g L^{-1})	4.56
DOC (mg/L)	1.875	VS (g L^{-1})	3.37
SUVA (L/mg m)	1.173	TCOD (g L^{-1})	132.07
Zeta potential (mV)	−16.8	SCOD in supernatant (g L^{-1})	27.59