



Removal of natural organic matter by titanium tetrachloride: The effect of total hardness and ionic strength



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ABSTRACT

This study is the first attempt to investigate the effect of total hardness and ionic strength on coagulation performance and the floc characteristics of titanium tetrachloride (TiCl₄). Membrane fouling under different total hardness and ionic strength conditions was also evaluated during a coagulation–ultra-filtration (C–UF) hybrid process. Coagulation experiments were performed with two simulated waters, using humic acid (HA, high molecular weight) and fulvic acid (FA, relatively low molecular weight), respectively, as model natural organic matter (NOM). Results show that both particle and organic matter removal can be enhanced by increasing total hardness and ionic strength. Floc characteristics were significantly influenced by total hardness and ionic strength and were improved in terms of floc size, growth rate, strength, recoverability and compactness. The results of the UF tests show that the pre-coagulation with TiCl₄ significantly improves the membrane permeate fluxes. Under different total hardness and ionic strength conditions, the membrane permeate flux varied according to both NOM and floc characteristics. The increase in total hardness and ionic strength improved the membrane permeate flux in the case of HA simulated water treatment.

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1. Introduction

Natural organic matter (NOM) is a diverse mixture of organic compounds with different molecular weights, such as humic acid (HA) with high molecular weight and fulvic acid (FA) with relatively low molecular weight (Kabsch-Korbutowicz, 2005). The NOM causes odor, taste, color, and bacterial re-growth in potable water, and has the potential to form carcinogenic disinfection-byproducts (DBPs) (Hu et al., 2006). Thus, the effective removal of NOM has become increasingly important in modern water treating systems. Coagulation and flocculation have been the most common methods of removing NOM and large particles in water treatment processes (Jarvis et al., 2005).

Aluminum (Al) and iron (Fe) salts are widely used as effective coagulants in removing a broad range of impurities, including colloidal particles and organic substances (Duan and Gregory, 2003). However, the production of large amounts of sludge requiring further disposal is the main drawback of using

conventional Al and Fe coagulants. The sludge post-treatment is considered to be one of the most costly and environmentally problematic challenges of all water treatment works (Kane, 1987; Nassar et al., 2009). Recently, titanium tetrachloride (TiCl₄) has been investigated as an alternative coagulant because its efficiency is comparable to, or higher than, both Al and Fe salts (Zhao et al., 2011a, 2011b). Additionally, the TiCl₄ coagulant produces floc of larger size and higher growth rate than conventional coagulants, resulting in better settleability. The most significant advantage is that the TiCl₄ flocculated sludge can be recycled to produce valuable byproduct by calcination (Zhao et al., 2011a; Shon et al., 2009, 2007; Lee et al., 2009; Okour et al., 2010), namely titanium dioxide (TiO₂), which is the most widely used metal oxide and whose applications include photocatalysts, cosmetics, paints, electronic paper, and solar cells (Hoffmann et al., 1995; Obee and Brown, 1995). Therefore, utilizing TiCl₄ as a coagulant combines comparable or superior coagulation efficiency and sludge disposal.

Floc characteristics are key influences in the solid/liquid separation process and affect coagulation efficiency (Yu et al., 2009). Smaller particles settle more slowly than larger particles of similar density (Boller and Blaser, 1998). Thus, small particles generally have lower removal efficiency by coagulation–flocculation than

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larger particles. The ability of flocs to withstand rupture and recoverability after they are broken has a significant impact at water treatment works (WTW), since the unit processes at WTW have prevalent regions of high shear (McCurdy et al., 2004). Floc strength and recoverability are therefore considered to be important parameters in understanding coagulation behaviour. Floc fractal dimension (D_f) is another particularly important operational parameter influencing the solid/liquid separation process (Gregory, 1998).

Both coagulation performance and floc characteristics depend not only on characteristics of the water source and coagulants, and hydraulic parameters (shear force and break-up period), but also on various water quality parameters, such as solution pH, total hardness and ionic strength (Hu et al., 2006; Yukselen and Gregory, 2004). Although much attention has been paid to the effect of coagulant dose, solution pH and hydraulic conditions on coagulation efficiency and floc characteristics by TiCl_4 (Zhao et al., 2011a, 2011b, 2011c, 2011d), there is no available report on the effect of total hardness and ionic strength on coagulation performance and floc properties by TiCl_4 .

Ultrafiltration (UF) technology has been extensively investigated as an effective water treatment process for the removal of suspended solids, colloidal material ($>0.1 \mu\text{m}$), NOM and DBPs precursors (Jucker and Clark, 1994; Zularisam et al., 2006). However, serious membrane fouling necessitated the employment of coagulation pre-treatment prior to UF to improve NOM removal and reduce membrane fouling (Guigui et al., 2002; Park et al., 2002; Oh, 2005). Nonetheless, there is no available literature concerning the effect of total hardness and ionic strength on membrane fouling using TiCl_4 as a coagulant during the coagulation-ultrafiltration (C–UF) hybrid process.

The overall objectives of this paper are to: i) investigate the effect of total hardness and ionic strength on coagulation performance by TiCl_4 for both HA and FA removal; ii) assess the influence of total hardness and ionic strength on floc properties, including floc growth rate, size, strength, recoverability and structure; iii) examine membrane fouling during the C–UF process under different total hardness and ionic strength conditions. To our best knowledge, this is the first attempt to elucidate the effect of total hardness and ionic strength on the coagulation behaviour of TiCl_4 . Since TiCl_4 has only recently been investigated as a coagulant, this study helps to acquire a better understanding of the correlation of the coagulation performance of TiCl_4 with the characteristics of raw water.

2. Experimental

2.1. Coagulant and test water

TiCl_4 stock solution (20%, density = 1.150 g/ml) was obtained from Photo & Environment Technology Co. Ltd. (South Korea).

HA and FA simulated water samples were used in this study. The HA stock solution (1.0 g/L) was prepared as follows: 1.0 g of HA (supplied by Ju-Feng Chemical Technology Co., Ltd, Shanghai, China), together with 0.04 g sodium hydroxide (NaOH), was dissolved in deionized water under continuous stir for 30 min, and the solution was then diluted to 1.0 L. NaOH was used to promote the dissolve of HA. The stock solution of FA (1.0 g/L) was prepared by dissolving 1.0 g of FA (biochemical reagent, purchased from Yinong Biochemical Technology Co. Ltd., Shanghai, China) directly in deionized water.

Coagulation experiments were performed with two simulated water samples: i) containing 10 mg/L of HA prepared in deionized water and tap water (Yang et al., 2010), in which the turbidity, UV_{254} absorbance, dissolved organic carbon (DOC), zeta potential

and pH of the suspension was 3.59 ± 0.02 NTU, $0.432 \pm 0.005 \text{ cm}^{-1}$, $4.096 \pm 0.144 \text{ mg/L}$, $-14.5 \pm 0.5 \text{ mV}$ and 8.2–8.4, respectively; ii) containing 10 mg/L of FA prepared in tap water (Wang et al., 2012), in which the turbidity, UV_{278} absorbance, DOC, zeta potential and pH of the suspension was 0.45 ± 0.02 NTU, $0.089 \pm 0.002 \text{ cm}^{-1}$, $4.905 \pm 0.145 \text{ mg/L}$, $-15.0 \pm 1.0 \text{ mV}$ and 8.3–8.4, respectively.

2.2. Jar-test

Standard jar tests were conducted using a programmable jar-tester (ZR4-6, Zhongrun Water Industry Technology Development Co. Ltd., China). The water sample (1000 mL) was mixed rapidly for 30 s at 200 rpm before coagulant was added. After addition of the coagulant, rapid mixing (200 rpm) was performed for 1 min followed by slow mixing at 40 rpm for a duration of 15 min, which was then followed by 15 min of quiescent settling. Water samples were collected from 2 cm below the water surface for measurements. The water samples were pre-filtered using a $0.45 \mu\text{m}$ fibre membrane syringe filter before testing UV_{254} or UV_{278} (absorbance at 254 nm or 278 nm using a UV-754 UV/VIS spectrophotometer), while turbidity was directly measured without filtration using a 2100P turbidimeter (Hach, USA).

Coagulation–flocculation experiments under different total hardness conditions were conducted after the total hardness of the test water was adjusted to different levels by CaCl_2 solids. The ionic strength of the test water was adjusted from 0.05 mol/L to 0.4 mol/L by NaCl solid before the coagulation test.

2.3. Floc characterization

A laser diffraction instrument (Mastersizer 2000, Malvern, UK) was used to measure dynamic floc size as the coagulation and flocculation process proceeded. The schematic diagram of the on-line monitoring system for dynamic floc size can be found in Zhao et al. (2011b). The median equivalent diameter, d_{50} , was selected as the representative floc size, although the same trends were observed for d_{10} and d_{90} floc sizes. The floc growth rate was calculated by the slope of the rapid growth region (Xiao et al., 2010):

$$\text{Growth rate} = \frac{\Delta \text{size}}{\Delta \text{time}} \quad (1)$$

Following the floc growth phase, the aggregated flocs were exposed to a shear force at 200 rpm for 5 min, followed by a slow mixing at 40 rpm for 15 min to allow floc regrowth, with the aim of studying the floc breakage and recovery properties. Floc strength factor (S_f) and recovery factor (R_f) are used to compare floc breakage and recoverability (Jarvis et al., 2005; Zhao et al., 2011b, 2012; Cao et al., 2011; Zhao et al., 2012):

$$\text{Strength factor} (S_f) = \frac{d_2}{d_1} \times 100 \quad (2)$$

$$\text{Recovery factor} (R_f) = \frac{d_3 - d_2}{d_1 - d_2} \times 100 \quad (3)$$

where, d_1 is the average floc size of the plateau before breakage, d_2 is the floc size after the floc break-up period, and d_3 is the floc size after regrowth to the new plateau. The higher the S_f value, the less sensitive the flocs are to breakage. Likewise, the floc with a larger R_f shows better recoverability after being subjected to high shear force.

Previous researches have reported the determination of aggregate mass fractal dimension (D_f) using Mastersizer 2000 (Jarvis

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