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Evaluation of starch-based flocculants for the flocculation of dissolved organic matter from textile dyeing secondary wastewater



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HIGHLIGHTS

G R A P H I C A L A B S T R A C T

- Two starch-based flocculants with different chain architectures have been employed.
- Flocculation of dissolved organic matters in dyeing secondary effluent was studied.
- Humic acid and bovine serum albumin (BSA) were used as synthetic organic matters.
- Flocculant with cationic branch chains owns better flocculation performance.
- BSA flocculation at pH 7 appears an unusual phenomenon.

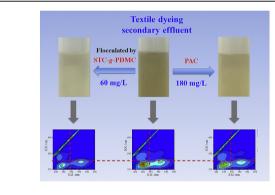
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ABSTRACT

China is a major textile manufacturer in the world; as a result, large quantities of dyeing effluents are generated every year in the country. In this study, the performances of two cationic starch-based flocculants with different chain architectures, i.e., starch-graft-poly[(2-methacryloyloxyethyl) trimethyl ammonium chloride] (STC-g-PDMC) and starch-3-chloro-2-hydroxypropyl trimethyl ammonium chloride (STC-CTA), in flocculating dissolved organic matter (DOM) in dyeing secondary effluents were investigated and compared with that of polyaluminum chloride (PAC). In the exploration of the flocculation mechanisms, humic acid (HA) and bovine serum albumin (BSA) were selected as main representatives of DOM in textile dyeing secondary effluents, which were humic/fulvic acid-like and proteinlike extracellular matters according to the studied wastewater's characteristics based on its threedimensional excitation-emission matrix spectrum. According to experimental results of the flocculation of both the real and synthetic wastewaters, STC-g-PDMC with cationic branches had remarkable advantages over STC-CTA and PAC because of the more efficient charge neutralization and bridging flocculation effects of STC-g-PDMC. Another interesting finding in this study was the reaggregation phenomenon after restabilization at an overdose during the flocculation of BSA effluents by STC-g-PDMC at a very narrow pH range under a nearly neutral condition. This phenomenon might be ascribed to the formation of STC-g-PDMC/BSA complexes induced by some local charge interactions between starchbased flocculant and the amino acid fragments of protein due to charge patch effects.

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1. Introduction

As a huge water consumer and wastewater producer in China, the textile dyeing industry has always faced the formidable challenges of wastewater treatment and water reuse in view of the current water resource shortage (Lau and Ismail, 2009; Soares et al., 2006) as well as stricter regulation (Zahrim et al., 2011). Textile dveing effluents are distinguished by high levels of chrominance. chemical oxygen demand, and salinity (Yang et al., 2015). Biological treatment is essential to the purification of dyeing wastewater (Marcucci et al., 2001). However, the residual contaminants after biological treatment are still difficult to handle. Among the various types of contaminants in secondary treated wastewater, dissolved organic matter (DOM) is one of the most disconcerting threats to human health and environmental safety. DOM generally includes natural organic matter (NOM), soluble microbial products (SMPs), and trace harmful chemicals (Shon et al., 2006). NOM, which is the main precursor of many disinfection by-products, significantly degrades water environment quality (Kim and Yu, 2005; Matilainen et al., 2002). The complexity and indeterminacy of the constituents of DOM account for the challenge in studying its identification and removal efficiency (Imai et al., 2002; Zhu et al., 2014).

In view of the increasingly stringent discharge and reuse standards, a variety of methods have been employed to treat dyeing secondary effluents, including activated carbon adsorption (Bes-Piá et al., 2003), advanced oxidation (Bes-Piá et al., 2004), ultrafiltration/microfiltration (Żyłła et al., 2006), reverse osmosis (Marcucci et al., 2001), and magnetic ion-exchange resin treatment (Yang et al., 2015). In fact, a single method can never achieve complete treatment of the diverse objects from wastewater (Lotito et al., 2014), and the extensive application of these methods is hindered by their respective limitations. Therefore, flocculation is generally applied as a pretreatment technology for tertiary treatment (Kornaros and Lyberatos, 2006). The integration of flocculation allows for a comprehensive treatment, which can evidently alleviate the load of contaminant removal on the subsequent processes and reduce the operation cost. For example, flocculation has significant effects on membrane fouling when ultrafiltration/microfiltration (Żyłła et al., 2006), nanofiltration (Zahrim et al., 2011) or reverse osmosis (Marcucci et al., 2001) is applied subsequently.

In the flocculation process, the flocculant always plays a key role (Lee et al., 2014; Matilainen et al., 2010; Yang et al., 2016). Natural polymer-based flocculants, a sort of organic polymers, are distinct from traditionally commercial coagulants/flocculants, such as polyaluminum chloride (PAC), in many aspects because of their several significant advantages, such as biodegradability and high efficiency (Guibal et al., 2006; Huang et al., 2016; Yang et al., 2016). Among these natural polymer-based flocculants, starch-based flocculants have received a considerable attention owing to their abundant supply and low cost; many kinds of starch-based flocculants have been designed and reported (Lekniute et al., 2013; Lin et al., 2012; Wang et al., 2013; Wei et al., 2008; Wu et al., 2016). However, the precise control of the materials' molecular structure on the basis of the structure-activity relationship is highly important to improve their final application performance (Flory, 1953; Sperling, 2006). In our previous study (Wu et al., 2016), several different starch-based flocculants with various chain architectures and charge properties were employed to flocculate humic acid (HA). Starch-graft-poly[(2methacryloyloxyethyl) trimethyl ammonium chloride] (STC-g-PDMC) was proved to have the highest flocculation efficiency among the studied starch-based flocculants and traditional PAC because of its distinct chain architecture with cationic branches, resulting in its strong charge attractions and bridging effects. However, the flocculation performance of the studied starch-based flocculants, including the effects of the chain architecture, still requires further examination using real wastewater.

In the current study, the textile dyeing secondary effluents were chosen as the target wastewater for treatment, since textile dyeing effluents remain to be a thorny scientific research challenge because of their high levels of chrominance, chemical oxygen demand, and salinity (Yang et al., 2015). The textile dyeing secondary effluents were obtained after a primary sediment and anaerobic anoxic-oxic (A²O) process, which were collected from a textile dyeing plant located in Yixing, China. Two cationic starch-based flocculants, STC-g-PDMC and starch-3-chloro-2-hydroxypropyl trimethyl ammonium chloride (STC-CTA), were then applied to flocculate the collected wastewater to demonstrate their potential practical applications. In comparison to previously used inorganic coagulants and the synthesized organic polymeric flocculants, those natural polymer-based flocculants are more environmental friendly due to their biodegradability. Moreover, PAC, which is a widely used commercial coagulant, was also employed for further comparison. Besides, in consideration of the significantly complicated composition of real wastewater, the differences among the different components of the target pollutants treated by various flocculants should be investigated. Such an investigation can further provide a valuable reference for the design and selection of suitable flocculants to obtain better synergetic and complementary flocculation effects in practical applications. Therefore, flocculation of specific pollutants in wastewater separately for the mitigation of their interferences is necessary for a clear and in-depth understanding of the flocculation mechanisms. In this study, HA and bovine serum albumin (BSA) were selected as the simulated pollutants. The selection was in accordance with the characteristic composition of the studied textile dyeing secondary wastewater based on its three-dimensional excitation-emission matrix (3D EEM) spectrum and findings in the relevant literature (Dizge and Tansel, 2011). The flocculation behaviors of the aforementioned two starch-based flocculants and PAC for removing HA and BSA, aside from the textile dyeing secondary effluents, have been evaluated in detail.

2. Materials and methods

2.1. Materials

The two starch-based flocculants, i.e., STC-g-PDMC and STC-CTA, were prepared meticulously and characterized in detail according to the procedure described in our previous study (Wu et al., 2016). The contents of positive quaternary ammonium salts in STC-g-PDMC and STC-CTA were approximately 2.21 and 1.16 mmol/g, respectively. HA (sodium salt, Aladdin Industrial Co., Ltd.), BSA (molecular biology grade, Aladdin Industrial Co., Ltd.), and PAC ([Al₂(OH)_nCl_{6-n}]m, n = 3.6-5, m < 10, Al₂O₃ content > 28%) were used directly without further purification in this study. Other chemicals were all obtained from Nanjing Chemical Reagent Co., Ltd.)

2.2. Flocculation experiments

2.2.1. Real and synthetic wastewaters

The textile dyeing secondary effluents were obtained from a textile dyeing plant in Yixing, China, and used as wastewaters for treatment. The textile dyeing wastewater, mainly containing reactive dyes, was initially treated with a primary sedimentation unit and subsequently by $A^{2}O$ process. The textile dyeing secondary effluents were prefiltered with a 0.45 µm cellulose nitrate filter to remove particulate matter. The filtrate was preserved at 4 °C and used in experiments as raw water. The typical characteristics of the filtrate are shown in Table 1.

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