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Dissolved organic matter removal using magnetic anion exchange resin treatment on biological effluent of textile dyeing wastewater

Jun Fan, Haibo Li, Chendong Shuang*, Wentao Li, Aimin Li*

State Key Laboratory of Pollution Control and Resource Reuse, Collaborative Innovation Center for Advanced Water Pollution Control Technology and Equipment, School of the Environment, Nanjing University, Nanjing 210023, China

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ABSTRACT

This study investigated the removal of dissolved organic matter (DOM) from real dyeing bio-treatment effluents (DBEs) with the use of a novel magnetic anion exchange resin (NDMP). DOMs in two typical DBEs were fractionized using DAX-8/XAD-4 resin and ultrafiltration membranes. The hydrophilic fractions and the low molecular weight (MW) (<3 kDa) DOM fractions constituted a major portion (>50%) of DOMs for the two effluents. The hydrophilic and low MW fractions of both effluents were the greatest contributors of specific UV₂₅₄ absorbance (SUVA₂₅₄), and the SUVA₂₅₄ of DOM fractions decreased with hydrophobicity and MW. Two DBEs exhibited acute and chronic biotoxicities. Both acute and chronic toxicities of DOM fractions increased linearly with the increase of SUVA₂₅₄ value. Kinetics of dissolved organic carbon (DOC) removal via NDMP treatment was performed by comparing it with that of particle active carbon (PAC). Results indicated that the removal of DOC from DBEs via NDMP was 60%, whereas DOC removals by PAC were lower than 15%. Acidic organics could be significantly removed with the use of NDMP. DOM with large MW in DBE could be removed significantly by using the same means. Removal efficiency of NDMP for DOM decreased with the decrease of MW. Compared with PAC, NDMP could significantly reduce the acute and chronic bio-toxicities of DBEs. NaCl/NaOH mixture regenerants, with selected concentrations of 10% NaCl (m/m)/1% NaOH (m/m), could improve desorption efficiency.

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Introduction

Textile dyeing is one of the most chemically-intensive industries that generate a large amount of wastewater during dyeing processes. It is characterized by high chemical oxygen demand, color, turbidity, pH, temperature, and toxic chemicals (Gao et al., 2007). Direct discharge of textile dyeing wastewater causes great environmental hazard to receiving water bodies through a series of direct and indirect effects, such as light attenuation, depletion of dissolved oxygen, eutrophication, and toxicity to aquatic life (Jo et al., 2008). Aside from esthetic problems, residual persistent organic pollutants affect the quality of potable water. Sporadic and excessive

exposure to colored effluents can cause health disorders in humans (Foo and Hameed, 2010). Hence, textile dyeing wastewater treatment has received wide attention in the past decades.

Given its high heterogeneity, dyeing wastewater can be treated by using only few advanced technologies, such as coagulation (Wang et al., 2011), oxidation (Feng et al., 2010; Mezzanotte et al., 2013), membrane separation (Zheng et al., 2003; Ben Amar et al., 2009), adsorption (Santhi et al., 2010), and combination process (Zainal et al., 2007; Konsowa et al., 2010). Coagulation and adsorption processes are generally used for removing hydrophobic dyes and auxiliaries in textile dyeing wastewater. Traditional flocculant and active carbon are non-recyclable. Advanced oxidation processes, including

* Corresponding authors. E-mails: shuangchendong@nju.edu.cn (Chendong Shuang), liaimin@nju.edu.cn (Aimin Li).

chemical agents (Fenton agents and ozone) and photochemical or electrochemical methods, have been successfully applied in organics degradation. Both ultrafiltration and nanofiltration membranes efficiently perform color removal. However, they have a limited lifetime because of clogging or membrane fouling. These methods are efficient, but are commercially unattractive because of their high costs.

Biological treatment is currently the most economical and popular alternative process (Dogan et al., 2010). Azo dye removal through biological process is based on anaerobic treatment for reductive cleavage of azo linkages in combination with aerobic treatment for further degradation of residual chemicals. Textile wastewater contains different types of dyes and auxiliaries, some of which are toxic to microbes and are non-biodegradable (Sarayu and Sandhya, 2012). Substances in industrial bio-treatment effluents even at low concentrations can still exhibit acute and chronic toxicities (Gurung et al., 2011). Therefore, finding an alternative advanced treatment combined with current biological processes, considering efficiency and economy, is highly needed to reduce dissolved organic matter (DOM) and toxicity of dye wastewater.

DOM in biologically treated dyeing wastewater mainly consists of soluble microbial products, residual dissolved dye, and auxiliaries. Dyes used by the textile industry are made up of chromophores and auxochromes. The chromophores are azo, nitro, and quinoid groups, while the most important auxochromes are carboxyl ($-\text{COOH}$), hydroxyl ($-\text{OH}$), and sulfonate ($-\text{SO}_3\text{H}$), which are typical anion groups. Anion surfactants are also important auxiliaries in dyeing wastewater (Verma et al., 2012). Major components of soluble microbial products SMPs, such as humic substances, are typically negatively charged. Thus, anion resin may be suitable for biologically treated dye wastewater.

Commercially available anion exchange resins have been employed in dye wastewater (Raghu and Ahmed Basha, 2007). However, traditional ion exchange processes are usually performed in fixed beds, which are expensive and exhibit flux restriction (Shuang et al., 2012a). A commercial magnetic ion exchange resin (MIEX®) was developed by Orica Limited (Australia) in the last decade. This resin has the following characteristics: small size (150 to 180 μm), polyacrylic matrix, and quaternary amine functional group. The magnetic property of this resin enables it to self-agglomerate easily for separation. MIEX® is widely used for removing DOMs in natural water and bio-treatment effluent (Drikas et al., 2011; Aryal and Sathasivan, 2011; Ho et al., 2012; Kingsbury and Singer, 2013). Shuang et al. (2012a, 2012b, 2013) recently developed a novel magnetic anion exchange resin (NDMP) that has higher adsorption capacity, faster kinetics, and regeneration advantage than MIEX®, and also demonstrated that NDMP has higher adsorption capacity for active dyes and is resistant to humic acid fouling. Thus NDMP is more suitable than MIEX® for treating textile dyeing bio-treatment effluent (DBE) efficiently and economically. However, only few studies investigated DOM removal and toxicity in real biologically treated textile wastewater by using NDMP.

This article aims to investigate the removal efficiency of NDMP for DOM in real DBE. To achieve this objective, (1) the characteristics of DBE were analyzed using resin chromatography, membrane filtration, and fluorescence excitation–emission–matrix (EEM); (2) the adsorption/desorption capacities of NDMP for DOM in DBEs were investigated; and (3) the removal efficiency of NDMP treatment for biotoxicity in DBEs was assessed.

1. Materials and methods

1.1. Textile DBEs and sample collections

Two typical DBEs (Lao San LS and Dong Nan DN) were obtained from two textile wastewater treatment plants (WWTPs) located

in Changzhou, China. Reactive anionic dyes were the main toxic organics in influent. Treatment processes of the two WWTPs were divided into three steps, namely, primary sediment, anaerobic bio-treatment, and aerobic bio-treatment. Raw effluents (250 L) were collected and filtered immediately through 0.45 μm membrane filters. Then, the filtered water samples were stored at 4°C in the dark.

1.2. DOM fractionation

Different affinities of Supelite DAX-8 (Sigma-Aldrich Co. LLC USA) and Amberlite XAD-4 (Rohm and Haas Corporation, USA) adsorbent resins were used to divide DOM in DBEs into the following five fractions: hydrophobic acids (HPOA), transphilic acids (TPIA), hydrophobic neutrals (HPON), transphilic neutrals (TPIN), and hydrophilic matters (HPI). Before placing them into the resins, samples were acidified to a pH level of 2.0 with 1 mol/L H_3PO_4 . Then, they were passed through the resins in glass chromatography columns, in which hydrophobic DOM and transphilic DOM were adsorbed onto the DAX-8 and XAD-4 resins, respectively. HPI were not adsorbed onto either the DAX-8 or XAD-4 resin. After adsorption, DAX-8 and XAD-4 resins were rinsed with 0.1 mol/L NaOH and distilled water to obtain HPOA and TPIA. These resins were then cleaned with 75% acetone and 25% distilled water to obtain HPON and TPIN. Acetone-free HPON and TPIN were separated by using vacuum rotary evaporation.

Samples were fractionated using Merck Millipore Ultracel PL-1, PL-3, and PL-10 ultrafiltration membranes Merck Group, Germany) with molecular cut-offs of 1, 3, and 10 kDa, respectively. Ultrafiltration was performed using a stirred 400 mL ultrafiltration device. Nitrogen pressure was maintained at 0.5 MPa. Starting with the PL-10 membrane, the process continued through the PL-3 and PL-1 membranes. With the ultrafiltration method, four DOM fractions were collected with molecular weights (MWs) of >10 kDa, 3 to 10 kDa, 1 to 3 kDa, and <1 kDa. Adsorption and ultrafiltration methods were based on the modified schemes described by Hua and Reckhow (2007). A freeze dryer was used to obtain lyophilized isolates for further analysis.

1.3. Toxicity assessment

Acute biotoxicity was assessed by testing its inhibitory effect (15 min) on the metabolism of *Photobacterium phosphoreum* T3 spp. (Institute of Soil Science, Chinese Academy of Sciences, China), which is indicated by its luminescence inhibition rate (LIR, %). LIR can be expressed as Eq. (1):

$$\text{LIR} = (1 - L_1/L_2) \times 100\% \quad (1)$$

where, L_1 is the luminous intensity of luminescent bacteria cultivated in the samples, and L_2 is the luminous intensity of luminescent bacteria cultivated in the blank samples (Meriç et al., 2005).

LIR values were converted to acute toxicity units (TUa) as shown by Eq. (2).

$$\text{TUa} = 100/(100 - \text{LIR}). \quad (2)$$

Daphnia magna was used as the testing species for chronic biotoxicity tests, which are based on chronic toxicity by using

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