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Fouling characteristics and fouling control of reverse osmosis membranes for desalination of dyeing wastewater with high chemical oxygen demand

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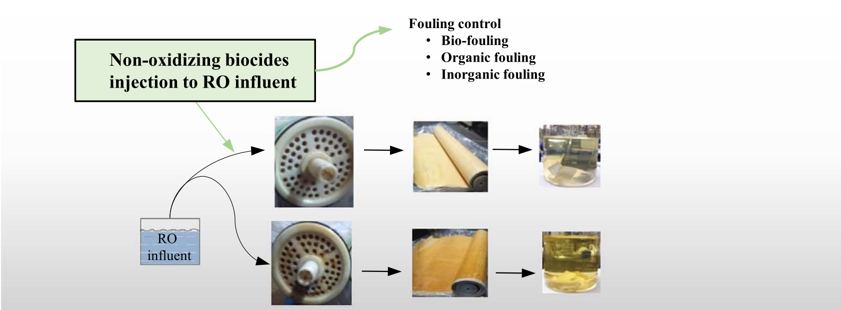
GRAPHICAL ABSTRACT



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ABSTRACT

Fouling of reverse osmosis (RO) membranes is a severe problem for desalination of dyeing wastewater with high chemical oxygen demand (COD). In this paper, fouled RO membranes in a pilot-scale desalination system of biologically treated dyeing wastewater containing high COD (143–228 mg/L) with or without injection of non-oxidizing biocides were analyzed. The two non-oxidizing biocides used are K5030 (a kind of isothiazoline biocide) and FR110 (an inorganic base type of combined chlorine agent), respectively. For the RO membrane from the system without non-oxidizing biocide injection to the influent, a typical biofilm occurred on the RO membrane in a short operation time of 19 days. The injection of non-oxidizing biocides caused a decline of nearly 99.0% in active biomass on the RO membrane. *Burkholderiales* ruled mature biofilm on the RO membrane without injection of non-oxidizing biocide. *Pseudoxanthomonas* exhibited a certain resistance to non-oxidizing biocides and ruled the bacterial community on the RO membrane with injection of non-oxidizing biocides. Fe, P and Al were major inorganic elements on RO membranes, decreasing with injection of non-oxidizing biocides. Hydrophobic neutral (HON) fraction was the most major organic component of dissolved organic matter (DOM) on RO membranes, decreasing notably with injection of non-oxidizing biocides.

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1. Introduction

The textile industry is a huge water consumer and wastewater generator. Dyeing, a combined process of bleaching and coloring, produces the vast majority of textile wastewater. Dyeing wastewater usually has a high chemical oxygen demand (COD), strong color, high hardness and low biodegradability [1]. Due to the poor biodegradability of dyeing wastewater, biological treatments are common with low contaminant removal efficiencies. The contaminant concentrations of secondary effluent are difficult to meet the environmental permissible levels. Conventional advanced treatments, e.g., coagulation-flocculation, advanced oxidation processes (AOPs) and adsorption, failed to be effective in the desalination and complete mineralization of refractory organic matter.

After years of development, reverse osmosis (RO) process has been widely applied in municipal and industrial wastewater reclamation and reuse, no longer confined to seawater desalination only. The striking attributes of RO process are high contaminant rejection, simple operation, modular design and to produce high quality water [2]. In dyeing wastewater desalination, COD concentration of RO influent might be higher in respect to seawater and municipal wastewater desalination. Ciardelli et al. [3] reported that the COD concentration of RO influent after biological-sand filter and ultrafiltration (UF) in a pilot-scale textile plant was 139 mg/L, with high quality produced water for reusing in dyeing processes.

However, membrane fouling has been the biggest limitation factor to the application of RO process. The types of membrane fouling are summarized as colloid matter fouling, biofouling, inorganic/organic foulings. Fouling of RO membrane is affected by pretreatment set-up, membrane properties and operation conditions, but the fouling profile is most likely impacted, to a major extent, by the source water quality [4,5]. Proper disinfection technologies before feed water being charged into RO membranes are necessary for mitigating membrane fouling. Biocides are widely used as a conventional disinfection technology in seawater and municipal wastewater reclamation to control membrane biofouling [6,7]. These are oxidizing and non-oxidizing biocides. Oxidizing biocides are incompatible with polymeric membranes and must be scavenged before being fed in RO membranes, which may lead to post-growth of deactivated microorganisms [7]. Some studies reported chlorination (a kind of oxidizing biocide) did not effectively control seawater RO membrane biofouling [8,9]. Non-oxidizing biocides are compatible with polyamide membranes with greater persistence of bactericidal effect [6]. But the effect of non-oxidizing biocides on controlling fouling of RO membranes for desalination of industrial wastewater with high COD is yet unclear.

The main objective of this study was to detect the fouling characteristics and evaluate the effect of non-oxidizing biocides on controlling fouling of RO membranes for desalination of dyeing wastewater containing high COD. An autopsy was performed for the fouled RO membranes in a pilot-scale dyeing wastewater desalination plant with and without injection of non-oxidizing biocides to the influent. A comparative analysis of surface morphology, active biomass, bacterial communities, inorganic/organic components of the fouled RO membranes was carried out.

2. Materials and methods

2.1. Pilot-scale RO processes

A pilot-scale UF-RO system was conducted at a dyeing wastewater treatment plant, located in Shaoxing, Zhejiang Province, China. The effluent of a biological-sand filter of the dyeing wastewater treatment plant was fed to the UF unit, and UF permeate flowed to two sets of RO units with uniform operation conditions (Fig. S1). Table S1 shows the characteristics of the UF and RO membranes used in the pilot system. Each RO unit had one membrane element with the feed water flow rate of 3.4 m^3/d. The recovery of each RO unit was 50%. RO units were operated at a constant permeate flux of 1.7 m^3/d.

Four mg/L sodium hypochlorite (NaOCl) was sequentially added to UF influent to inhibit bacterial growth. Nine mg/L sodium hydrogen sulfite ($NaHSO_3$) was injected as reductant to RO influent for preventing RO membranes oxidation. Ten mg/L T-N770 (Kurita Water Industries Co., Ltd., Japan), a liquid reagent containing organic phosphonic acid, was continuously injected as antiscalant to RO influent to inhibit inorganic scaling of RO membranes. Five mg/L K5030 and 100 mg/L FR110 (Kurita Water Industries Co., Ltd., Japan) were continuously injected as non-oxidizing biocides to one of the RO units successively by different dosing spots of pipeline. The dosing spot of FR110 was closer to RO membrane module than that of K5030. The active ingredient of K5030 is a kind of isothiazoline biocide, which can inhibit slime growth on RO membranes. The active ingredient of FR110 is an inorganic base type of combined chlorine agent. FR110 has function of high sterilizing power and low oxidation potential in the form of combined chlorine in target water. FR110 inhibits slime and prevents fouling of a membrane by a peel off mechanism while RO unit is in operation. The other RO unit worked without non-oxidizing biocide injection to the influent.

2.2. Water samples

RO influent, RO concentrates and RO permeates from the UF-RO pilot system were collected and loaded into amber glass bottles. All water samples, kept in ice, were delivered to the laboratory. Biological analysis of water samples were tested immediately. Water samples were filtered through a 0.45 μm membrane (Tianjin JinTeng Experiment Equipment Co., Ltd.) to minimize the effect of suspended particles, and then stored at 4 °C until analyzed.

2.3. Collection of fouled RO membranes and deposits on RO membranes

2.3.1. Collection of fouled RO membranes

Two sets of RO membrane modules were collected from the pilot system after 19 days of operation and taken to the laboratory. RO membrane samples were stored at 4 °C and autopsied within 24 h.

2.3.2. Collection and extraction of deposits on RO membranes

The deposits at the inlet, middle and outlet parts of fouled RO membranes were collected by physical scrapping. The inlet part of RO membrane was nearest to RO influent and the outlet part of RO membrane was nearest to RO concentrate.

A 10 cm × 10 cm of fouled RO membrane was extracted in 500 mL of 0.01 M NaOH solution (pH = 12) to obtain dissolved organic matter (DOM) of deposits. After gentle agitation for 2 h, pH of the solution was then adjusted to 7.0 ± 0.2. The solution was filtered immediately through the 0.45 μm membrane and stored at 4 °C for further analysis.

2.4. Fouling characterization

2.4.1. Colloidal fouling prediction of water samples

Silt density index (SDI_{15}) is commonly used for colloidal fouling prediction. The operating pressure of SDI_{15} test was 0.21 MPa supplied by a TM21R air compressor (Japan), which was a dead-end filtration with 0.45 μm Millipore membrane, and the filtration time was recorded according to the method by Cha et al. [10].

2.4.2. Loss on ignition (LOI) test

Deposit sample was dried in 110 °C to constant weight, and then combusted at 550 °C for 4 h. The weight of sample after ignition at 550 °C represented inorganic matter content and the weight of mass loss on ignition at 550 °C represented organic matter content.