Chemical Engineering Science 114 (2014) 51–57

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Nanofiltration hollow fiber membranes for textile wastewater treatment: Lab-scale and pilot-scale studies

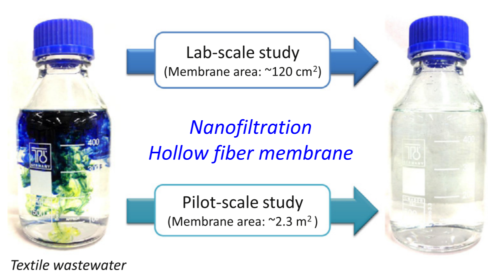
Yee Kang Ong, Fu Yun Li, Shi-Peng Sun **, Bai-Wang Zhao, Can-Zeng Liang, Tai-Shung Chung *

Department of Chemical & Biomolecular Engineering, National University of Singapore, 4 Engineering Drive 4, Singapore 117585, Singapore

HIGHLIGHTS

- The performance of the membrane was evaluated under various operating conditions.
- The membrane was scaled-up and potted in a 2-in. pilot-scale module.
- The pilot-scale module was tested with industrial wastewater.
- The membrane shows robust performance during the lab- and pilot-scale evaluations.
- The membrane offers high potential to recover and reuse the salts.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 16 February 2014
Received in revised form
4 April 2014
Accepted 5 April 2014
Available online 13 April 2014

Keywords:
Nanofiltration
Textile wastewater
Hollow fiber membrane
Pilot-scale
Dyes
Chemical oxidation demand (COD)

ABSTRACT

The textile industry is a water intensive industry that generates a vast amount of wastewater. The wastewater generated from the textile industry is generally loaded with pollutants comprising spent textile dyes, suspended solids, mineral oils, electrolytes, surfactants, etc. Therefore, it must be properly treated before disposal or reuse. A systematic study was conducted to evaluate the performance of newly developed polyamide-imide hollow fiber nanofiltration (NF) membrane in various operating conditions such as feed temperature (i.e., 25, 40, 50, 70 °C), solute concentration (i.e., 100, 500, 1000 ppm) and pH (i.e., 3, 7, 10). The results indicate that the NF membrane has satisfactory rejections (average: > 90%) against various dyes at most testing conditions. In addition, more than 80% of NaCl and 90% of Na_2SO_4 permeate through the membrane. As a result, these salts have the potential to be be recovered and reused for the next dyeing process. The robustness of the membrane was proven by showing satisfactory and stable performance under cycles of chemical cleaning during the lab-scale and pilot-scale evaluations.

1. Introduction

The textile industry is regarded as one of the major industries in the world. The demand of textile related products is expected to increase rapidly along with the exponential growth of world's population in the 21st century. The aforementioned industry is key driver to the economics of many countries and China is regarded to emerge as the largest textile producing country in the world (Heymann, 2011).

Apart from the bright side of the textile industry, it is also being identified as one of the most polluting industry. As a water intensive industry by nature, the quantity of wastewater generated from this industry is reported to be in the range of 2–180 L of wastewater per kg of textile products based on the processes and the type of textile materials (i.e., wool, cotton, synthetic fiber, etc)

* Corresponding author. Tel.: +65 6516 6645; fax: +65 6779 1936.
** Corresponding author. Tel.: +65 6601 2433; fax: +65 6779 1936.
E-mail addresses: chesuns@nus.edu.sg (S.-P. Sun), chencts@nus.edu.sg (T.-S. Chung).

http://dx.doi.org/10.1016/j.ces.2014.04.007
0009-2509/

(World Bank Group, 2007). In addition, the wastewater generated from the textile industry is generally high in both biochemical oxygen demand (BOD) and chemical oxygen demand (COD) because it comprises spent textile dyes, suspended solids, mineral oils, electrolytes, surfactants, etc (World Bank Group, 2007; Lau and Ismail, 2009). Therefore, this severely polluted wastewater must be properly treated before proceeding to the disposal or reuse.

To date, several methods have been developed to treat the wastewater produced from the textile industry and they can be generally classified into biological and physicochemical processes. Biological treatment or activated sludge is reported as an effective and common practice to reduce the COD of wastewater. However, this process was unable to fully eliminate the color of the wastewater which posts severe threat to the environment and the aquatic ecosystem (Kapdan and Kargi, 2002; Dafale et al., 2008). Although physicochemical processes such as coagulation–flocculation (Verma et al., 2012; Golob et al., 2005), adsorption (Demirbas, 2009; Rafatullah et al., 2010) and oxidation processes (including chemical-/photo-/sono-/electro-oxidation) (Madhavan et al., 2010; Nidheesh et al., 2013) show huge potentials in decolorization of textile wastewater, the main hurdle of these techniques lies on their relatively high operating costs and some technical difficulties. Zhang et al. (2004) reported that the ozonation process was expansive and may produce toxic by-products from the wastewater and Can et al. (2006) stated that the advanced oxidation processes were not economic viable. In addition the chemical coagulation process was reported to be ineffective in the removal of dissolved reactive dyestuffs (Vandevivere et al., 1998).

Membrane separation processes have demonstrated robust performances in water treatment, reclamation and desalination applications (Shannon et al., 2008; Greenlee et al., 2009; Lee et al., 2011; Shen et al., 2012). As an intermediate process between the ultrafiltration (UF) and reverse osmosis (RO), the nanofiltration (NF) process requires less operating pressure than RO and offers a better rejection than UF. It has been widely applied in various applications ranging from water treatment, pharmaceutical, oil and food industries (Wang and Chung, 2006; Caus et al., 2009; Lau and Ismail, 2009; Sereewatthanawut et al., 2011; Rundquist et al., 2012; Sun et al., 2012; Xing et al., 2014). The separation mechanisms of this process include size exclusion and Donnan exclusion that can be favorable in removing dyes and water softening applications in industrial effluents (Peng and Escobar, 2003; Verliefde et al., 2008; Zhong et al., 2012).

The commercially available NF membranes are predominantly made of thin-film composite (TFC) flat membranes installed in a spiral-wound module which consists of a thin polyamide selective layer deposited on top of the support (Majamaa et al., 2011). Qin et al. (2007) evaluated the performance of several NF membranes obtained from Osmonics (DK4040C), TriSep (4040-TS40-TSA) and Saehan (NE-70) to treat the wastewater from dyeing facilities, the NE-70 membrane showed a reasonable flux and 99% dye removal efficiency among the tested membranes. Kurt et al. (2012) compared the performance of commercial NF (NF-270, Dow Filmtec) and reverse osmosis (XLE, Dow Filmtec) membranes in decolorization as well as reduction of COD and salts of wastewater from the weaving industry. The concentration polarization, osmotic pressure of the feed solution and inorganic scaling were identified to be the crucial factors in affecting the fouling of the aforementioned membranes.

On the other hand, the membrane in a hollow fiber configuration was reported to offer several advantages over flat membrane as it eliminates the need of spacers, possesses a higher surface area per unit of membrane module volume (which translates into a higher productivity), provides self-supporting structure and permeate channel (Li et al., 2011; Ong and Chung, 2012, 2013). These advantages prompted researchers to explore the feasibility of NF hollow fiber membranes in various applications (Wang and Chung, 2006; Darvishmanesh et al., 2011; Sun et al., 2011, 2012; Maurya et al., 2012; Shao et al., 2013; Zheng et al., 2013). Sun et al. (2012) developed a double-repulsive NF hollow fiber membrane which comprises a thin positively charged selective layer on top of a negatively charged substrate. The membrane possesses over 99% rejections to both positively and negatively charged dyes. Shao et al. (2013) fabricated a TFC NF hollow fiber membrane using the combination of *m*-phenylenediamine (MPDA) and piperazine (PIP) as the amine monomers during the interfacial polymerization process to alter the sub-nano structure of the selective layer. Their membrane demonstrated good performance in rejecting Safarnin O and Aniline blue. Zheng et al. (2013) utilized a TFC NF hollow fiber membrane in simultaneous decolorization and COD reduction of biologically treated textile effluent via submerged NF technique. They could achieve a 99% color removal and a COD reduction rate of more than 90%. The rejection of electrolytes or salts is greatly dependent on the surface charge of the membrane. Shao et al. (2013) observed that the negatively charged membranes were found to show a higher rejection towards divalent anion (SO_4^{2-}) over the monovalent anion (Cl^-) as well as monovalent cation (Na^+) over divalent cation (Mg^{2+}), vice versa for the positively charged membranes as reported by Sun et al. (2012).

Since limited studies have been devoted to evaluate the performance of pilot-scale NF hollow fiber modules, this study aimed to systematically investigate a newly developed NF membrane in treating textile wastewater. The performance of the NF membrane was examined under various operating conditions such as dye types, dye concentrations, feed temperatures, pH values as well as cleaning cycles. Subsequently, the membrane was potted in a 2-in.-diameter half-meter-length pilot-scale module with an effective membrane area of $\sim 2.3\ m^2$ and tested on-site with industrial generated wastewater. The outcome of this study may provide valuable insights in the evaluation and design of novel pilot-scale NF hollow fiber membranes for various industrial applications.

2. Experimental

2.1. Materials

The polyamide-imide based NF hollow fiber membrane with a pure water permeability of $6 \pm 1\ L/m^2\ h\ bar$ was fabricated based on the procedure reported in Sun et al. (2011) with minor modifications. The NF hollow fiber membrane consists of an integrally skinned substrate with a polyethyleneimine (PEI) functionalized outer surface. Organic textile dyes consisting of reactive blue 19, reactive black 5 and reactive yellow 81 (Sigma-Aldrich Corporation Singapore) were used as the model solutes to evaluate the membrane performance. Table 1 shows the respective chemical structures and molecular weights of these solutes. All the dyes in the feed solution were expected to remain in the anionic state throughout the experimental conditions due to the low pK_a value of the sulfonic acid group (pK_a: ~ 1) (Muruganandham et al., 2006; Pirillo et al., 2009). Deionized (DI) water was used to prepare the model solutions during the experiments. Hydrochloric acid (HCl) and sodium hydroxide (NaOH) were utilized to adjust the pH values of the feed solutions.

2.2. Nanofiltration experiments

2.2.1. Lab-scale

The lab-scale NF experiments were carried out in a self-fabricated NF setup. The effective membrane area of each hollow