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Ultrafiltration ceramic membrane performance during the treatment of model solutions containing dye and salt

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ABSTRACT

The present study reports the results of the ultrafiltration with a ceramic membrane of model solutions containing NaCl and a reactive dye, which simulate textile wastewaters. The effect of salt concentration (1, 2.5 and 4 g/L NaCl) and transmembrane pressure (1, 2, and 3 bar) on the evolution over time of both permeate flux and solutes rejection was tested. Membrane fouling was also investigated in terms of analysis of resistances-in-series, determining the contribution of both reversible and irreversible fouling under any condition tested. More severe flux decline and a slight reduction in color rejection were observed under higher pressure. Furthermore, it was demonstrated that the membrane performance was significantly worsened in the presence of salt, according to permeate flux, dye rejection and the values of the different resistances. Higher flux decline and total resistance together with lower color removal were observed when adding NaCl to the solution. These results were attributed to membrane charge and repulsion-attraction phenomena. It was then suggested that electrostatic interactions between the solutes and the membrane materials have an important role in the ultrafiltration performance, as well as influencing the kind of fouling affecting the membrane.

1. Introduction

Textile industry is characterized by the consumption of high quantities of water for processes such as dyeing, washing, rinsing and others [1]. Moreover, the disposal of its effluents creates both an aesthetic and environmental wastewater problem. This is due to the high dye content, often toxic and clearly visible if discharged into public waterways, and to the presence of a great quantity of salts, organic matter and other auxiliaries coming from the different textile processes, which contribute to aquatic toxicity [2,3].

Among the different types of industrial dyes available, mainly reactive azo dyes are applied in textile dyeing processes because of their advantageous characteristics. They are soluble anionic dyes containing azo bonds (—N=N—) and several reactive groups which are able to form a covalent bond with the hydroxyl groups in the fiber [4,5]. However, some disadvantages related to the use of this kind of dyes are their low fiber fixation and the significant amount of electrolyte (mainly NaCl) required during the dyeing process [6]. As a consequence, considerable concentrations of dyes, which have a complex and poorly degradable aromatic structure, are present in the wastewater as well as high amounts of salts, hindering its treatment by conventional systems [7]. Furthermore, textile effluents are subjected to significant variations in their composition due to the changing properties of the textile products and the different processes involved, making its final treatment even more difficult [8].

Due to all these aspects, textile industry is considered one of the most environmentally unfriendly manufacturing industries worldwide. In this way, wastewater reuse would probably offer solutions to the problem of textile effluents disposal. Nowadays, membrane technologies are recognized as the most promising methods for textile wastewater reuse [3,9]. These processes have demonstrated to enable the recovery of valuable products (dyestuffs and chemical auxiliaries), the reduction of both total wastewater volume and contaminant load, together with the minimization of water usage by recycling water in production processes such as rinsing or dyeing [1,10,11].

Ceramic membranes present several advantages over polymeric ones, including high thermal, chemical and mechanical resistance, among others [12]. These characteristics make them particularly appropriate for the treatment of textile effluents, which usually present high temperatures and alkaline conditions [13].

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One negative aspect of the use of membrane processes for the treatment of textile effluents is that during the period of operation, textile water constituents (suspended particles, salts, organic matter, etc.) tend to accumulate within a thin boundary layer adjacent to the membrane surface, in a phenomenon called concentration polarization. This fact increases the osmotic pressure near the membrane–solution interface, which decreases the driving force and thus the permeation of solvent. Furthermore, membrane–solute interactions could lead to the adsorption and deposition of these components onto the membrane surface or into the pores, resulting in membrane fouling [14].

The accumulation of species at the membrane surface adversely affects the process performance since it leads to an increase in the membrane resistance over time. As a consequence, flux decline is observed and separation efficiency could be affected. If the buildup of solids on the membrane is significant enough, it may act as a secondary membrane and change the selective properties of the system [15].

Due to the decline in both flux and separation efficiency caused by rapid fouling while using textile wastewaters directly as influent to nanofiltration (NF) or reverse osmosis (RO) membranes, a proper pretreatment is required. Ultrafiltration (UF) is currently proposed as pretreatment previous to NF or RO processes because of its ability to retain plugging particles present in textile effluents [16].

The nature and extent of membrane fouling is strongly influenced by the hydrodynamic conditions of the process (i.e. transmembrane pressure, flow rate) and by the physicochemical nature of the membrane and the solutes [17]. Besides, membrane–solute and solute–solute interactions can significantly affect the selective properties of the membrane as well as its tendency to fouling [18].

In order to quantify the electrokinetic interactions involved in membrane fouling and solute rejection, the zeta potentials of the membranes are usually calculated from the streaming potential data [19,20]. The zeta potential is an indicator of the membrane surface charge; its evaluation is of great importance since it provides information which is useful for the understanding of the membrane filtration performance.

Considering the variety of factors affecting the phenomenon of fouling, there is a need for a detailed study of the different effects controlling the process. Numerous studies have been performed in order to evaluate the behavior of ultrafiltration [21–23] and nanofiltration [24–27] membranes treating effluents containing dyes and salts. From those studies, it is remarkable the significant influence of the solute–solute and solute–membrane interactions on the final performance of the filtration process. However, most of the studies on ultrafiltration membranes involve the use of tight ultrafiltration membranes. Thus, scarce literature may be found dealing with the treatment of these dye–salt binary systems by loose ultrafiltration ceramic membranes and the influence of the interactions between the solution constituents on the final performance. To that end, in the present work the influence on the filtration process of different dye–salt model solutions, containing a reactive dye and NaCl, was evaluated by means of a loose UF ceramic membrane.

The effect of various salt concentrations and transmembrane pressures on the membrane performance was analyzed. The evolution of permeate flux and solutes retention was determined for each solution in order to compare the membrane behavior. Furthermore, the various filtration resistances were estimated using the resistance-in-series model in order to evaluate the membrane fouling according to operating conditions and feed characteristics. For a better understanding of the membrane performance, streaming potential measurements were also carried out in order to determine the membrane charge.

2. Materials and methods

2.1. Membrane filtration unit

Fig. 1 presents a schematic drawing of the pilot plant used for the study. It was a cross-flow membrane filtration system whose main components were two tanks (containing the feed and the cleaning solution respectively), a variable speed pump (3CP1140, Cat pumps, US) to circulate the feed solution throughout the system and a stainless steel housing where the membrane module was placed.

Two manometers at each side of the membrane module were used to measure the transmembrane pressure, regulated by means of a needle valve placed at the retentate stream. Two filters of 100 and 25 μm were situated upstream the pump and the membrane module respectively in order to prevent them to be damaged by the pass of suspended particles through the system. A temperature regulating system permitted the experiments to be performed at constant temperature with an accuracy of ±1 °C. An electronic balance (KB120 2N, Kern®, Germany), connected to a computer with a data acquisition software (Balance Connection 4.0, Kern®, Germany), made it possible to determine the permeate flux gravimetrically, recording data at one minute interval.

The set-up operated at constant concentration mode, via permeate and retentate recirculation between the membrane module and the feeding tank.

2.2. UF membrane

In this work, an INSIDE CéRAM™ tubular ultrafiltration membrane purchased from Tami Industries (Nyons, France) was used. The membrane was 250 mm long, had an external diameter of 10 mm and consisted of seven channels with a hydraulic diameter of 2 mm each. Table 1 collects the main membrane characteristics according to manufacturer.

Membrane permeability was determined experimentally using deionized water before each filtration experiment. Flux values of deionized water, labeled as J_w, were measured at different operating pressures (ΔP), constant temperature of 25 °C and cross-flow velocity of 3 m/s. Then, they were plotted against pressure. The Darcy's law was used to fit the pure water permeability data [17]:

$$J_w = L_p \cdot \Delta P = \frac{\Delta P}{\mu \cdot R_m} \quad (1)$$

where μ is the water viscosity. The slope of the straight line J_w versus pressure gives the pure water membrane permeability coefficient (L_p). The value of the intrinsic membrane resistance (R_m) was then calculated from the permeability coefficient according to Eq. (1). The average value of membrane permeability as measured was 142.86 L m^{-2} h^{-1} bar^{-1}, corresponding to an intrinsic membrane resistance of 2.83×10^{12} m^{-1}.

2.3. Feed solutions

The membrane performance during the filtration of an azo reactive dye solution was studied with feed containing 100 mg/L of dye Reactive Black 5 (RB5), purchased from Sigma–Aldrich (Germany). This dye was selected since it is one of the most commonly used in the textile industry throughout the world [28]. The RB5 has two sulfonate groups and two sulfatoethylsulfon groups with negative charges within an aqueous solution [29]. It has a molecular weight of 991.82 g/mol and its molecular structure is presented in Fig. 2.

The membrane was also tested with feeds consisting of mixtures of both dye and sodium chloride salt at different concentrations of NaCl (1, 2.5 and 4 g/L) and constant dye concentration