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**JOURNAL**


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Recovery of Flux Decline in Cross Flow Nanofiltration of Two-Component Dye and Salt Mixture by Low Frequency Ultrasonic Irradiation

Jal M. Patel, Kaushik Nath

Gradual reduction of steady state permeate flux (throughput) over a period of time often mars the performance of nanofiltration process. In the present study, nanofiltration of two-component dye and NaCl mixture containing Reactive Black 5 and C.I Reactive yellow 160 was carried out in a flat sheet membrane module with hydrophilized polyamide membrane (molecular weight cut off 400) over a range of operating conditions. Results indicate that the flux decline with time could be significantly mitigated under low frequency (34 ± 3 kHz) ultrasonic irradiation when compared with the conventional unassisted nanofiltration. Various filtration resistances were determined using a simple resistance-in-series model, which revealed that concentration polarization resistance could be effectively decreased by ultrasonic irradiation. Effect of several parameters such as mode of irradiation (viz. continuous and intermittent), trans-membrane pressure and temperature were studied and discussed with respect to existing theories. The power consumption of the high pressure plunger pump at different feed pressures and the ultrasonic power transferred to the water bath at three different temperatures were estimated. More than 96% of color, and 65% TDS were removed from the treated water accompanied by a marked reduction of COD. The process allowed the production of permeate stream with great reutilization possibilities.

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being classical, has not only found its wide application in cleaning of fouled polymeric membranes to ensure its reusability, but has also been applied for flux improvement of pressure driven membrane processes at times in combination with some other techniques [10,11]. Feng et al. [12] studied ultrasonic cleaning to remove fouling from a commercially important polyamide based reverse osmosis membrane during cross-flow filtration of CaSO₄, Fe³⁺ and carboxyl cellulose solutions. In each case, the permeate flux of the membrane increased significantly, with virtually no decrease in rejection in the presence of ultrasonication [12]. Muthukumaran et al. [13] reported the use of low frequency ultrasound to facilitate cross flow ultrafiltration of dairy whey solution. The study revealed that the use of turbulence promoters (spacers) in combination with ultrasound could lead to a doubling in the permeate flux. Tarleton and Wakeman [14] demonstrated that both electric and ultrasonic fields could reduce membrane fouling and in turn enhance flux. It is suspected that the internal and elastic forces created by the acoustic waves cause changes to the interfacial phenomena of the solid phase leading to reduction of viscosity of the liquid phase in a suspension to allow for improved dewatering [14]. In another study ultrasound effects on permeate flux and rejection of solute were tested by using two types of polyacrylonitrile (PAN) ultrafiltration (UF) membranes reinforced with non-woven cloth. It was found that the ultrasound irradiation significantly increased the permeate flux of different molecular weight (MW) dextran solutions [15].

Although, ultrasonic effect has been frequently applied in the ultrafiltration and microfiltration processes for the enhancement of permeate flux as well as cleaning of fouled membranes, this almost remains to be an underexploited topic in nanofiltration application for aqueous waste treatment. To the best of the knowledge of authors, the use of acoustic irradiation to mitigate the flux decay in cross-flow nanofiltration has so far not been reported in the literature. The present work aims to understand the effects of low frequency ultrasonic irradiation on flux behavior and contribute various resistances during nanofiltration of a feed containing a mixture of two different reactive dyes in varied proportion and Filtration resistances were determined under various operating conditions using the resistance-in-series model. Effect of several parameters such as irradiation mode, transmembrane pressure temperature were studied and discussed with respect to existing theories. The experimental data presented show that the ultrasound irradiation could both modify and improve performance of nanofiltration over a range of operating conditions.

2. Materials and methods

2.1. Chemicals

The feed solution for the present study was a mixture of two reactive dyes namely, Reactive black 5 (molecular weight 991.82) and Reactive yellow 160 (molecular weight 818.13) which are largely used in dyeing of cotton knits wear in a winch dye machine at various dye houses locally. The characteristics and the chemical structures of the dyes are presented in Table 1. Concentrations of both dyes were varied in different proportion however fixing the (NaCl) concentration at 500 mg/L. All the chemicals used in experiment were of AR grade supplied by Merck, India and were used without further purification.

Table 1

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>Reactive Black 5</th>
<th>Reactive Yellow 160</th>
</tr>
</thead>
<tbody>
<tr>
<td>Color Index (CI) name</td>
<td>CI. Reactive Black 5</td>
<td>CI. Reactive Yellow 160</td>
</tr>
<tr>
<td>Molecular weight</td>
<td>991.82 Da</td>
<td>818.13 Da</td>
</tr>
<tr>
<td>Melting point</td>
<td>&gt;260 °C</td>
<td>&gt;240 °C</td>
</tr>
<tr>
<td>Application class</td>
<td>Cotton</td>
<td>Cotton</td>
</tr>
<tr>
<td>Chemical Class</td>
<td>Azo</td>
<td>Azo</td>
</tr>
<tr>
<td>Absorption max (water)</td>
<td>590—595 nm</td>
<td></td>
</tr>
<tr>
<td>Chemical formula</td>
<td>C₉₀H₁₄₂Na₂N₂O₂S₀₂S₆</td>
<td>C₉₀H₁₄₂Cl₂N₄Na₂O₄S₃</td>
</tr>
<tr>
<td>Molecular structure</td>
<td><img src="image1" alt="Molecular structure of Reactive Black 5" /></td>
<td><img src="image2" alt="Molecular structure of Reactive Yellow 160" /></td>
</tr>
</tbody>
</table>

UV spectra

![UV spectra of Reactive Black 5](image3)  ![UV spectra of Reactive Yellow 160](image4)

*: Experimental value.
Separation of ternary sodium chloride/ Reactive Black-5 aqueous solutions using two different modules in a nanofiltration pilot plant

T. M. Patel & K. Nath
Separation of ternary sodium chloride/Reactive Black-5 aqueous solutions using two different modules in a nanofiltration pilot plant

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Abstract Nanofiltration of ternary mixtures of sodium chloride and aqueous solutions of Reactive Black-5 was studied in two different modules, namely, flat sheet and spiral wound over a wide range of operating conditions. Hydrophilized polyamide membrane with molecular weight cutoff of 150 was used for the experiments. Combined effect of dye and salt concentration, trans-membrane pressure drop, initial pH of feed solution on the permeate flux, and observed retention were investigated. Extent of color removal, chemical oxygen demand (COD), total dissolved solid (TDS), and conductivity were determined to assess performance of the membrane. The experimental results showed that both the permeate flux and observed retention decreased with increase in dye as well as salt concentration in the feed. Permeate fluxes were lower at higher pH values. Substantial removal of color was achieved in the nanofiltration experiments with a marked reduction in COD and TDS. The process allowed the production of permeate stream with great reutilization possibilities.

Keywords Membrane • Salt rejection • Permeate • Chemical oxygen demand • pH • Transmembrane pressure

Introduction

Considering both concentration and composition, dye-house effluents generated from the textile industry are among the most polluting of the industrial sectors and have been considered a significant environmental concern for several decades. Reactive dyes are soluble anionic dyes that contain one or more reactive groups capable of forming a covalent bond with the hydroxyl groups in the fiber and are not suitable for recycling (Erswell et al. 1988). These dyes have low fixation rates with the highest loss in the effluent compared with other dye types. Besides having strong color, effluents from dye houses contain high concentration of auxiliary chemicals—predominantly inorganic salts, used to regulate the rate of dye fixing on the textile. During the conventional chemical synthesis of dye by precipitation-filtration-drying method the dye is precipitated from an aqueous solution by salt, whereby the final product contains up to 30 % residual salt impurities (He et al. 2009; Yu et al. 2010). Given the great variety of fibers, dyes, and process additives in use, the conventional wastewater treatment of the textile effluent has limited application since the containing pollutants have very high complexity and diversity combined with very low biodegradability (Forgacs et al. 2004).

In order to overcome the disadvantages encountered in the conventional treatment processes for dye wastewater, constant research efforts are put up to look for alternative treatment methods. Membrane-based separation processes, which were the focus of attention of the separation technologists for the past three decades, have the potential to offer an improved separation and several cost advantages over these more traditional techniques. Various works have reported the use of nanofiltration (NF) membranes for dye separations either for wastewater treatment (Nataraj et al. 2009; Mo et al. 2008; Yu et al. 2010; Uzal et al. 2010) or process applications (Levenstein et al. 1996). NF is becoming widely accepted in the dye-house wastewater treatment. Their separation mechanisms involve both steric (sieving) and electrical (Donnan) effects. This combination allows NF
Comparative performance of flat sheet and spiral wound modules in the nanofiltration of reactive dye solution

Tejal M. Patel • Harsh Chheda • Abhisek Baheti • Punit Patel • Kaushik Nath

Abstract

Background and purpose. Besides the opportunities for reuse, stringent regulations and growing public awareness demand an enhanced quality of effluent from dye industries. Treatment of an aqueous solution of dye (reactive red 198) was carried out in a nanofiltration unit using both flat sheet and spiral wound modules to obtain a comparative performance evaluation in terms of permeate flux and quality.

Methods. Hydrophilized polyamide membrane with molecular weight cutoff of 150 was used for the experiments. Effects of trans-membrane pressure (TMP), feed concentration and addition of salt on permeate flux were investigated. Percent reduction of color, chemical oxygen demand (COD), total dissolved solid (TDS), and conductivity were determined to assess performance of the membrane.

Results. The maximum flux decline was 16.1% of its initial value at 490 kPa TMP with 50 ppm feed concentration in spiral wound module, whereas the same in flat sheet under same conditions was 7.2%. The effect of TMP showed a quasi-linear increase in flux with increasing pressure. Increased permeate concentration led to the reduction in observed retention of dye in the membrane. The average reduction in color, COD, and TDS were 96.88%, 97.38%, and 89.24%, respectively. The decline in permeate flux was more in case of spiral wound module compared to flat sheet. However, spiral wound module performed better in terms of color removal, COD reduction, and TDS removal.

Conclusion. Substantial removal of color was achieved in the nanofiltration experiments with a marked reduction in COD and TDS. The process allowed the production of permeate stream with great reutilization possibilities.

Keywords. Nanofiltration • Reactive dyes • Flux • Flat sheet • Spiral wound

1 Introduction

Dye house waste water is the major source of pollutants in process industries like textile, paper, and pulp, paints, ink, etc. Occurrence of dyestuffs and other inorganic chemical compounds on crops irrigated with waste water treatment plant effluent has adverse health effects. Ingestion of the dye-containing waste water may cause skin irritation, ulceration of the skin, and mucous membranes (Golka et al. 2004; Levine 1991). Their presence in water bodies may decrease the absorption of light by water, plants, and phytoplankton thereby reducing photosynthesis and the oxygenation of water (Kouba and Zhuang 1994). The effluent from the dyeing and finishing processes is characterized by strong color, high pH, high temperature, high chemical oxygen demand (COD), and low biodegradability. Therefore, effective treatment of dye contaminated waste streams assumes paramount importance in view of stringent environmental regulations and growing public awareness (Rott and Minke 1999). The conventional methods for removal of dyes from waste water include chemical coagulation and flocculation (Joo et al. 2007), flotation (Kabil and Ghazy 1994), chemical oxidation (Liu et al. 2010; Banerjee et al. 2007), and adsorption (Dotto and Pinto 2011). However, reactive dyes cannot be easily removed by conventional coagulation, flocculation, and flotation processes as these are recalcitrant.

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