Moroccan Journal of Chemistry
ISSN: 2351-812X
http://revues.imist.ma/?journal=morjchem&page=login
Berradi & al. / Mor. J. Chem. 7 N°2 (2019) 230-235

Ultrafiltration of wastewater-models loaded with indigo blue by membranes composed of organic polymers at different percentages: Comparative study

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Received 14 Oct 2018, Revised 14 May 2019, Accepted 26 may 2019

Abstract

The development of membranes intended for the ultrafiltration of wastewater-models loaded with vat dyes, the case of indigo, has taken during this work a major interest. This development was based on the synthesis of films, by phase inversion, permselective based on organic polymers such as polysulfone (PSU), polyvinyl chloride (PVC) and expanded polystyrene (PSE) on the one hand and based on octaglycidylether tetraaniline resin for methylene dianiline (OGTAPMDA), bisphenol A (BA) and titanium dioxide TiO_2, on the other hand. During the first step, we synthesized five types of membranes named M_1, M_2, M_3, M_4 and M_5 according to different percentages of the composition of the collodium (polymer/solvent/non-solvent) obtained namely; M_1 (12 % PSU/88 % DMF), M_2 (9 % PSU/ 1% PVC/ 90% DMF), M_3 (13.5 % PSU/0.5 % BA/1 % PSe/85 % DMF), M_4 (8.5 % PSU/1.5 % OGTAPMDA/90 % DMF) and M_5 (9.5 % PSU/0.5 % TiO_2/90 % DMF). The resulting membranes have already been characterized by researchers from the ECOP team, using microscopic tools such as FTIR and SEM, and macroscopic tools, based on hydrodynamic conditions, namely permeability and selectivity. The results obtained during this study show that the membranes obtained having excellent mechanical and static properties, on the one hand, and the ultrafiltration of the rejection-model, on the other hand, gave significant percentages on the rate scale. Fading and which are obtained as follows; 80.36 % for the M_1 membrane, 89 % for the M_2 membrane, 87.24 % for the M_3 membrane, 64.79 % for the M_4 membrane and 86 % for the M_5 membrane

Keywords: Ultrafiltration; Wastewater-model; Organic membranes; Indigo dye; Percent collodium; Discoloration rate.

1. Introduction

There are many techniques for treating textile wastewater, namely biological, chemical and physical techniques. In the latter techniques, the membrane separations of the dyes by microfiltration (MF), ultrafiltration (UF) and nanofiltration (NF) [1-4] processes have in recent years become very useful. Important to the scientific community because of their ability to retain a significant number of dyes. This is well observed when using the ultrafiltration technique whose interest is due to the performance of the properties of the membranes exploited through certain numbers of variables such as long life, ease of design of the membrane without making a module, a possibility of sterilization and declogging, a good resistance to chemical agents, temperature and solvents, and finally a separation carried out at room temperature, suitable for species, sometimes biodegradable [3]. The majority of commercially available ultrafiltration membranes are currently prepared by the method of phase inversion [3], from various organic polymers in this case, polysulfone, polyfluorovinylidene, polyacrylonitrile, cellulose, polyvinyl chloride, polyimides and polyamides [3-7]. The technique of ultrafiltration membrane synthesis is generally intended for the retention of the dyes contained in the effluents resulting from textile finishing [7]. Although some types of dyes cannot be separated due to their solubility in water, sulfur dyes and vat dyes, especially indigo, can be recovered by membrane filtration because they are insoluble in water and can be easily separated from the effluent [7], hence the interest to consider this work. And this, by synthesizing permeable asymmetric membranes from organic polymers such as polysulfone, expanded polystyrene and polyvinyl chloride based on the technique of phase inversion. The objective of this work is to develop the ultrafiltration membranes and to apply them in the purification of waste water-models containing indigo dye in order to make a comparative study of the separative performance of the different membranes obtained in during this study.

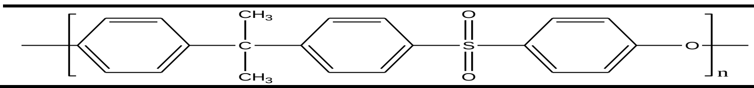
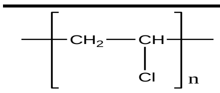
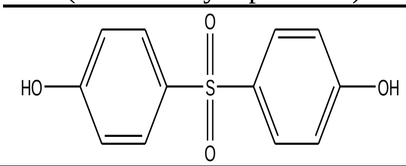
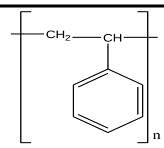
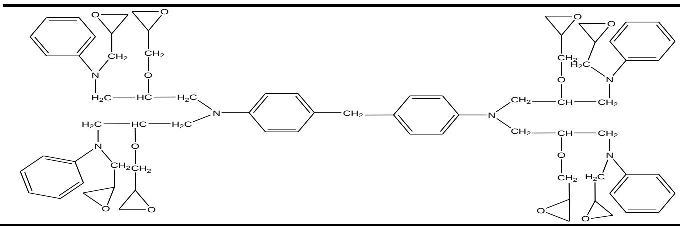
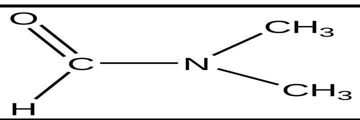
2. Material and methods

2.1. Preparation of membrane films

The polymers used to form the membranes, denoted M_1, M_2, M_3, M_4 and M_5, are of the polysulfone, expanded polystyrene and polyvinyl chloride type, and the adjuvant used are of the octaglycidylether tetraaniline paramethylenedianiline resin type and the titanium dioxide feed whose chemical structures are grouped in **Table 1** below.

These polymers have properties of rigidity, stability much higher than other thermoplastics, and especially at high temperatures [3, 4, 9-12]. The membrane samples are obtained, by the team of researchers within the team of Organic Chemistry & Polymers of the Laboratory LARPGP-FSK-ITU-Morocco, by the technique of phase inversion, by solubilizing different percentages by mass polymers either solely or in admixture in a solvent which is dimethylformamide (**Table 1**), according to the prototype indicated in **Figure 1**

Table 1: Percentages of composition of synthetic membranes and chemical structures of polymers.

Composition as a percentage of membranes and Chemical structures of polymers and adjuvant		
M_1 (12% PSU/88% DMF)	PSU 	
M_2 (9% PSU/1% PVC/90% DMF)	PVC 	
M_3 (13.5% PSU/0.5% BA/1% PSe/85% DMF)	BA (4.4- sulfoxyldiphenol S) 	PSe 
M_4 (8.5% PSU/1.5% OGTAPMDA/90% DMF)	OGTAPMDA 	
M_5 (9.5% PSU/0.5% TiO_2/90% DMF)	DMF 	

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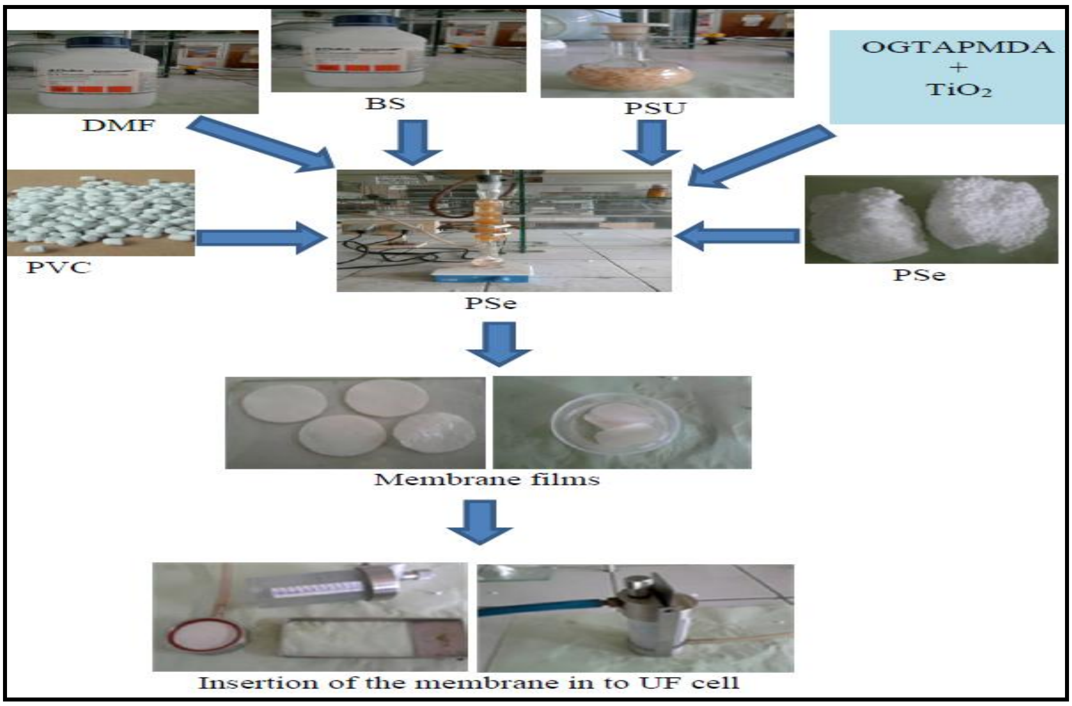

Figure 1: Prototype of UF membrane film synthesis.

2.2. Preparation of model sewage

The wastewater samples used to conduct the experimental part of this study were prepared by solubilizing 1 g of the of the powdered indigo dye, the chemical structure indicated in **Figure 2**, in one liter of hot water (70°C), in the presence of acetic acid, tenside and sodium hydrosulfite. To solubilizing this dye, the mixture obtained was stirred several times with the magnetic stirrer, knowing that the measured pH of the obtained colored water sample is alkaline (pH = 11.87).

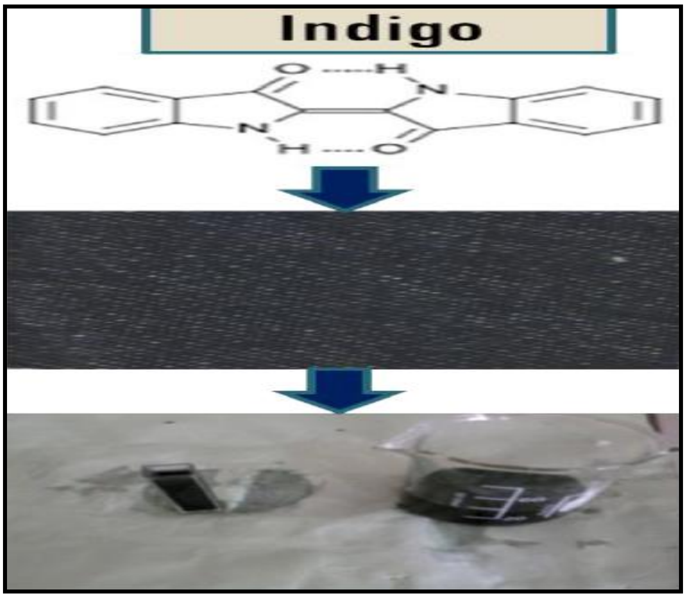

Figure 2: Chemical structure and wastewater model of indigo dye.

2.3. Method of analysis and ultrafiltration of model effluents

The spectroscopic analysis of the samples of the wastewater obtained was carried out using a spectrometer of JP.SELECTA.sa type, model 2100. By which, we determined the maximum wavelength λ_{max} of these samples, following the Beer Lambert law ($A = \varepsilon_{\lambda max}.l.C.$) [5], as shown in the diagram of **Figure 3**.

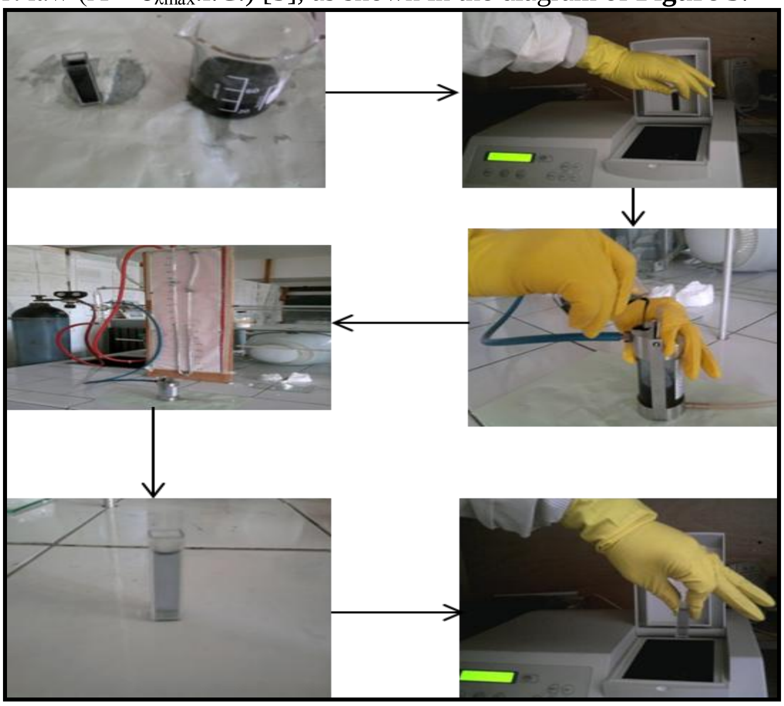

Figure 3: Experimental prototype of ultrafiltration of model effluents.

This allowed us to measure their optical densities before and after their decolorization by ultrafiltration, and consequently, we determined their fading rate according to the following formula [5]:

$$TD\% = \frac{DOi - DOf}{DOi} \times 100$$

With DO_i is the optical density of the colored water before ultrafiltration and DO_f is the optical density of permeate after ultrafiltration. Note that the fading rate is influenced by several parameters (pH, T°, etc.) [13], this is why we worked at a temperature of 25°C and at basic pH by adding a NaOH solution with a mass concentration of 3 g/l.

3. Results and discussion

3.1. Discoloration of effluents by ultrafiltration membranes

The analysis of the maximum adsorption of the model effluents used in this study was obtained at a maximum wavelength λ_{max} = 665 nm and the results of their discoloration by the different exploited membranes (M_1, M_2, M_3, M_4 and M_5) are well represented in **Figure 4** below.

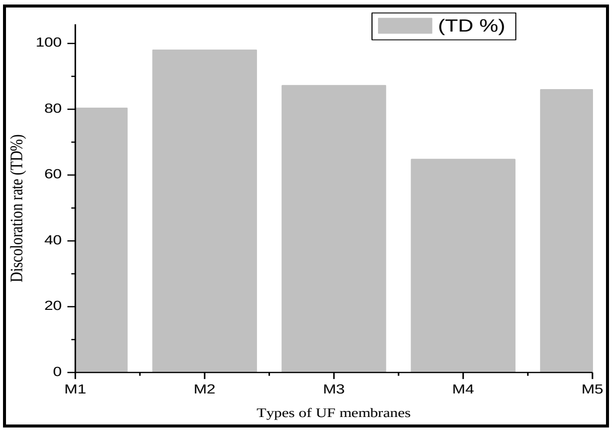

Figure 4: Discoloration rate of the model effluents by the different membranes.

According to the analysis of the results indicated in this figure, it turns out that membrane ultrafiltration of model effluents exploited by asymmetric membranes synthesized by polysulfone polymers, polyvinyl chloride and expanded polystyrene, and octaglycidylether resin adjuvant tetraaniline paramethylenedianiline, bisphenol A and titanium dioxide TiO_2 as filler, gave us significant values at the end of the fading rate and which are recorded respectively as follows; 80.36 % for the M_1 membrane (12% PSU/88% DMF) [5], 89% for the M_2 membrane (9% PSU/1% PVC/90% DMF) [8], 87.24% for the M_3 membrane (13.5% PSU/0.5% BA/1% PSe/85% DMF) [9], 64.79% for the M_4 membrane (8.5 %PSU/1.5 % OGTAPMDA/90 % DMF) [10] and 86 % for the M_5 membrane (9.5 % PSU/0.5 % TiO_2/90 % DMF) [11]. From a comparative point of view, it shows us the best yield, 89%, was obtained in the case of operating membranes based on the optimal mixture 9% PSU/1% PVC/90% DMF, followed by compound membranes, 87.24%, 13.5% PSU/0.5%BA/ 1% PSe/85% DMF. This can be explained by the permselectivity of the membranes obtained and more particularly by the geometric radius of the particles of the dye used relative to the porosity of the membranes.

3.2. Comparative study of other techniques for discoloring effluents loaded with indigo

The effluents from the denim finishing industry are loaded with vat dyes namely sulfur black and indigo. The decolorization of effluents loaded with this last dye (indigo), according to previous studies, has been made by several techniques such as their cobalt-catalyzed degradation and the nanoparticles of zirconium oxide modified by carbon

nanotubes (Co- ZrO_2-MWCNT) under simulated visible light achieved a degradation efficiency of about 98 % [14]. Cyanobacteria of the Phormidium sp. and Anabaena sp. are able to decolorize the indigo dye, after 14 days of incubation at 25 °C, respectively by the percentages 91.22 % and 71.92 % [15], so much so that the discoloration by yeast Diutina rugosa is of the order of 99.97 % at pH 2, temperature 30 °C and 2.0 g of cellular biomass within 5 days [16]. According to *Buscio et al.* PVDF-based ultrafiltration membranes eliminated the blue color by up to 99 % [7].

4. Conclusion

In this work, the discoloration of model wastewater loaded with vat dyes, indigo, by the ultrafiltration technique using five types of asymmetric membranes synthesized different percentages of polymers and additives namely resin and fillers has reached very high fading rates. This encouraged us to make, for a future study, a discoloration to the real samples of the effluents of the textile finishing industry, and also to make a comparative study on the scale of the mechanical and physical properties of these membranes, on the one hand and to apply them in combination processes namely coagulation-ultrafiltration, on the other hand.

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