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Discoloration of water loaded with vat dyes by the membrane process of ultrafiltration

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Abstract
During the present study, we conducted a research which is to develop an organic and asymmetric membrane polysulfonePSU UDEL P1700 by the technique of phase inversion, and to apply this film active supported in the discoloration of wastewater loaded with templates of vat dyes; indigo and sulphur black, while using and optimizing the treatment of effluents from textile finishing by ultrafiltration. According to the optimization and characterization of the hydrodynamic conditions of the membrane obtained, we purified directly the colored solutions models of water. The results of the optimization of the membranes showed that the synthesized membranewith a more efficient mechanical and hydraulic properties in percentage by weight of polymer PSU is of the order of 12%on the one hand, and those obtained for the purification of water colored by ultrafiltration have showed that the rate of discoloration is calculated in the range of 80.36% forthe indigo and of the order of 60.78%for the blacksulphur to the other.

Keywords: Asymmetric membrane, Polysulfone, Ultrafiltration, Colored water, Vat dyes, Optimization, Discoloration.

Introduction
In recent years, the exploitation of groundwater and surface water has increased sharply over the world because of the demographic development, agricultural, technological and industrial. This area includes several types of industries, namely the food, pharmaceutical, plastics, paper, metallurgy, aerospace, electroplating, painting, textiles,etc. The steps of spinning, warping, dyeing, weaving, finishing and making are all methods of the textile industry. Such as the steps in the dyeing and finishing make the noble textile and ready to use for making and are called textile finishing processes. During these processes, more additives, dyes, adhesives and other chemicals [1] are used in a mixture with a large volume of water to prepare a dye bath. What happens against a large amount of wastewater loaded with dyes, organic materials, auxiliaries and chemicals that are toxic effects on aquatic life [2-6]. This requires a preliminary treatment of the wastewater before discharge into receiving waters as required by the laws of industrial effluent treatment [7].
To treat this kind of wastewater, there are several treatment techniques, namely the advanced oxidation processes [8-10], adsorption methods on the activated carbon [11-17],the coagulation and flocculation processes [7, 18-23], the membrane processes of microfiltration (MF),ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO) [24,25,49-51] and others.According to some authors, the technique of ultrafiltration is most common in the purification of wastewater loaded with textile dyes including sulphur, vat and azoic [26-28,47,48] a hand and sizing products, rinse bath macromolecules and sizing agents (polyvinyl alcohol) [28-30], etc. Some authors applied the ultrafiltration technique combination with the coagulation and flocculation processes [31-33] and others have applied the reverse osmosis process [34]. Among the most used in the synthesis of the ultrafiltration membranes are polymers of polyurethane (PU) [35], polystyrene (PS)[36, 37], polyvinylchloride (PVC) [38], polysulfone (PSU) [28], etc. The latter polymer is answered in the preparation of permeable membranes because of their good mechanical properties, chemical, their high thermal stability and acid, alkaline resistance during the circulation [28].

The main objective of this study is to synthesize an asymmetric membrane made of an organic polymerwhich is polysulfone PSU UDEL P1700 based on the method of phase inversion [39, 40] after optimizing their percentage by weight colluding in the first time, and second of purifying the wastewater charged with modelstype of vat dyes indigo and sulphur black exploiting the technique of ultrafiltrationusing a membrane synthesized and optimized after their hydrodynamic and microscopic characterization.

2. Materials and methods

2.1. Materials operated

In conducting experiments of the present work, we used the following products: polysulfone (PSU) UDEL P1700 the mass molar of approximately 45 kDa, the chemical structure shown below (**Fig. 1** (a)) [41] and which has been supplied by Union Carbide; dextran of different molar masses (19; 40; 162; 298 kDa) with a mass concentration of 1.5% and was supplied by the company Life Technologies Limited; N, N'-Dimethylformamide (DMF) chemical formula $(CH_3)_2NCHO$, molecular of weight 73.09g/mole, supplied by BASF Company Inc. (Canada), and was used as a solvent PSU; acetic acid; sodium hydrosulfite $(Na_2(SO_4)_2)$; 38°Be causticsoda (NaOH);tenside used as a surfactant and wetting agent; cold and hot distilled water (70°C); indigo dye chemical formula $C_{16}H_{10}N_2O_2$ and a molecular weight of 262.2628 ± 0.0145g/mole; sulphur black of dye, the chemical structure shown in **Fig. 1** (b):

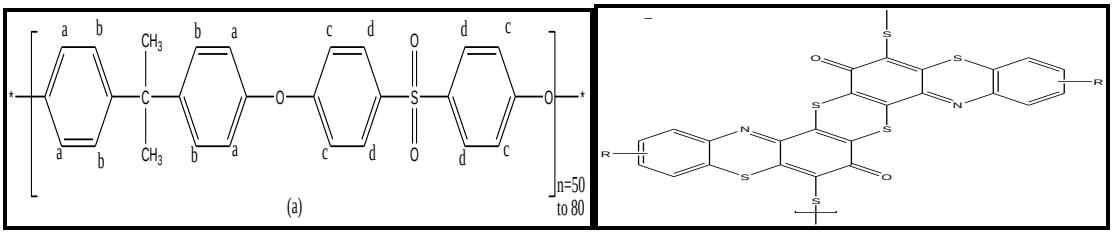

Figure 1: Chemical structures of PSU UDEL P1700 (a) and sulphur black (b).

2.2. Optimization of the synthesis of membranes

In a glass bottle and 250 ml capacity, we have prepared six colluding's compounds percentages by mass mixtures PSU/DMF(%)following: 8/92; 10/90; 12/88; 14/86, 15/85 and 16/84 by dissolving the PSU UDEL P1700 in DMF for 45 min with agitation with a speed of 580 tr/min using of a magnetic stirrer. As each colluding obtained was left in a first few hours to degas and cast on a glass plate and spread with a glass strip giving a precise thickness. The cast glass plate is emerged directly in the precipitation bath containing a non-solvent (water), as water diffuses into the film and causes the solidification of the polymer [41] according to the mechanisms of the method ofphase inversion [18, 24, 42] (**Fig. 2**, (a)). Finally the prepared membranes were cut as discs (**Fig. 2** (b)), the diameter equal to that of the Cell Amicon ultrafiltration (UF Cell Test (**Fig. 3**, b)) with a capacity of 36 cm^3.

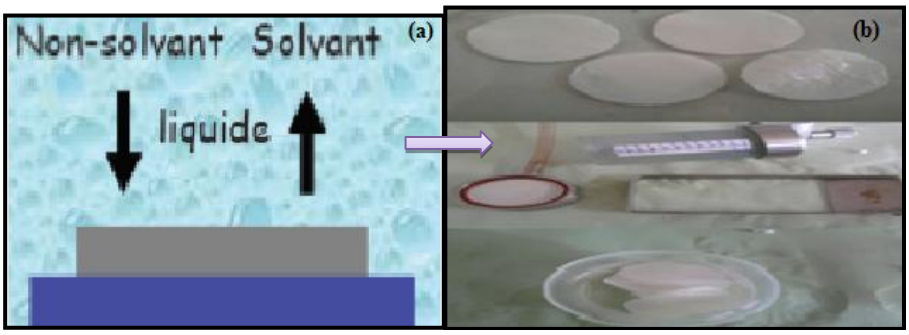

Figure 2: Principle of the phase inversion (a) and samples of the synthesized membranes (b).

2.3. Hydrodynamic characterization of the synthesized membrane

The resulting membranes were characterized hydro-dynamically measuring their permeability's and selectivity's using the ultrafiltration assembly schematically in the following figure:

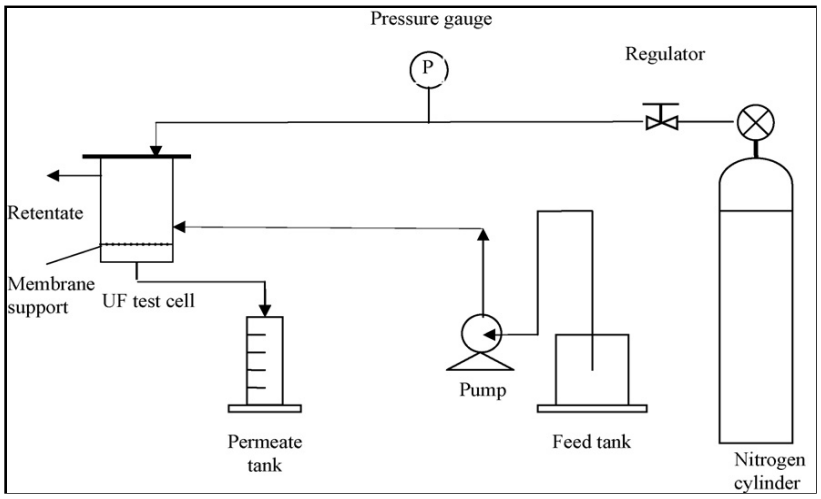

Figure 3: Mounting ultrafiltration (UF) [43].

For optimized the permeability of synthetic membranes, we have completed the Cell Amicon with distilled water and applied to increasing values of the trans-membrane pressure until total or near-total permeation distilled water by measuring the rate of permeation each pressure applied by the following relationship [43]:

$$\boldsymbol{Jw = \frac{Q}{A.\Delta t}} \qquad (1)$$

With Jw: ultrapure water flow (l/m^2.j), Q: volume of water permeates (l), A: effective membrane area (m^2) and Δt: permeation of time (j).

To measure the selectivity of synthetic membranes, we calculated for each membrane rate of retention (% R) of different solutions 1.5% by weight of dextran with molecular weight in kilo-Dalton (kDa): 17.5; 19; 40; 162; 298; 515 in a successive manner by subtracting the measurements of refractive index by an ABBE refract-meter according to the following relationship [44]:

$$\boldsymbol{\%R = \frac{(ni - nH2O) - (np - nH2O)}{(ni - nH2O)} \times 100} \qquad (2)$$

With: n_{H2O}: refractive index of distilled water used; n_i: refractive index of the dextran solution; n_p: refractive index of the ultrafiltered solution of dextran.

2.4. Spectroscopic and microscopic characterization of the membrane optimized

The optimized membrane was characterized by infrared spectroscopy Fourier Transform-Attenuated Total Reflection (FTIR-ATR), spectroscopy of nuclear magnetic resonance (NMR) and scanning electron microscope (SEM). The characterization of this membrane was carried out in the laboratories of the UATRS-CNRST-Rabat-Morocco.

2.5. Preparing the models of wastewater coloured

During this study and to test the bleaching performance of the optimized membrane, we prepared two types of wastewater loaded models, such as the first type is loaded with indigo dye and the second charge with that of the black sulphur.

The first type of waste water was prepared from 1.25g of indigo; 1.5g of sodium hydrosulfite and 2.5g of tenside. These three compounds were placed in a flask containing one litter of distilled water at 30°C with stirring the mixture a few minutes after the addition of caustic soda 38°Beto pH 11.80.

The second kind of wastewater was prepared by adding to a litter of hot distilled water at 70°C; 1.5g black sulphur; 5g tenside; 10ml of acetic acid concentration of 1.5g/l. The resulting mixture was stirred to dissolve and homogenize the solution after the addition of caustic soda 38°Be, such that, the pH of the resulting mixture was measured at 11.20.

The colourful waters models obtained were characterized by UV/Visible (JP. SELECTA SA. Model.2100)covering wavelengths from 190 to1100nm.This is intended to determine the peak wavelength of the water sample stained with a maximum absorbency based on the Beer-Lambert law. Note that in this study, the maximum absorbency A is the optical density (D.O.) was measured by gradually varying the wavelength of the light beam [45]:

$$\boldsymbol{A = \varepsilon . l . c} \qquad (3)$$

With A: the amount absorbed from the solution; Ɛ: the specific extinction coefficient ($cm^{-1}.mol^{-1}.L$); l: the thickness of the tank (cm); C: concentration of the sample solution (mol/l).

To determine the retention rates of various solutions of dextran, we based on the following equation [44]:

$$\%\boldsymbol{R} = \frac{\boldsymbol{Ci - Cf}}{\boldsymbol{Ci}} \times \boldsymbol{100} \qquad (4)$$

With C_i: is the initial concentration of the solution and C_f: is the final concentration of the ultra-filtered solution. The bleaching rate colourful model solution is determined according to the following equation [44]:

$$\%\boldsymbol{D} = \frac{\boldsymbol{DOi - DOf}}{\boldsymbol{DOi}} \times \boldsymbol{100} \qquad (5)$$

With DO_i: initial optical density of the coloured solution and DO_f: the optical density of the ultra-filtered solution.

3. Results and discussion

3.1. Hydrodynamic characterization of membranes

3.1.1. Effect of PSU concentration on the permeability of membranes synthesized

The figure 4 shows the influence of the weight percentages of the PSU UDEL P1700 on the permeation rate according to the volume ultra-filtered for each membrane at a fixed pressure in 20 cm Hg:

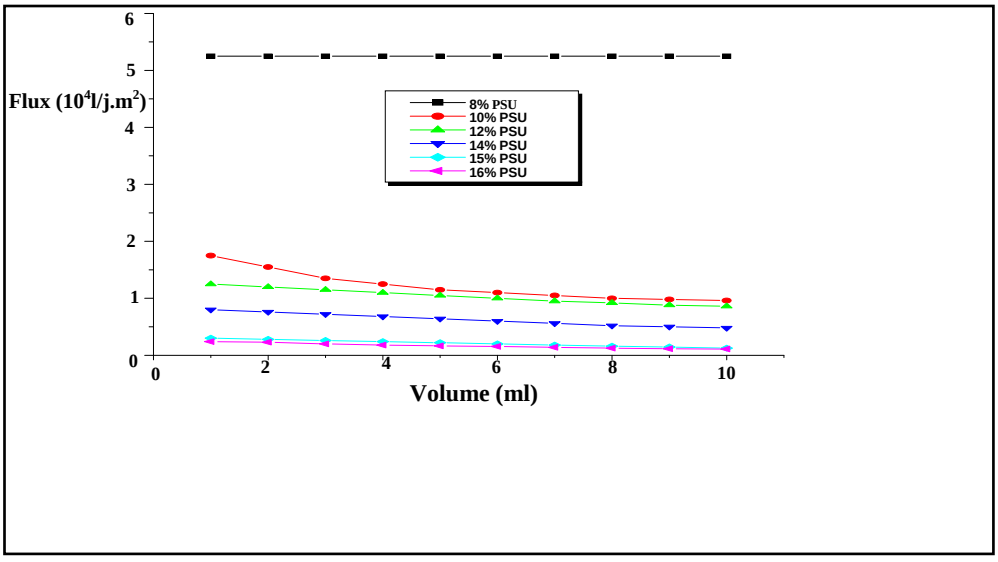

Figure 4: Variations of the flow permeability of each membrane as a function of the volume (ml).

From the curves in **Fig.4**, except that it shows the membrane in which the percentage 8% of PSU, the permeation of flux gradually decreased to its stabilization, this indicates that each membrane was compacted under the effect of applied pressure after 3 to 4 hours [43]. Moreover the increase of the concentration of polymer in the PSU colluding reduces permeability of distilled water. This is due to the fact that the walls of the membrane pores become closer, more dense and more uniform, leading to the reduction of the size of these pores and therefore the flow during compaction [43, 46], which approves the percentage by weight of polymer

PSU is an important influence in the colluding composition required for the synthesis of a permeable and asymmetric membrane.

3.1.2. Effect of PSU concentration on the selectivity of the synthesized membranes

The figure below shows for each membrane, the change in retention rates (%) by the increase in molecular weight dextran at a constant pressure of 20 cm Hg:

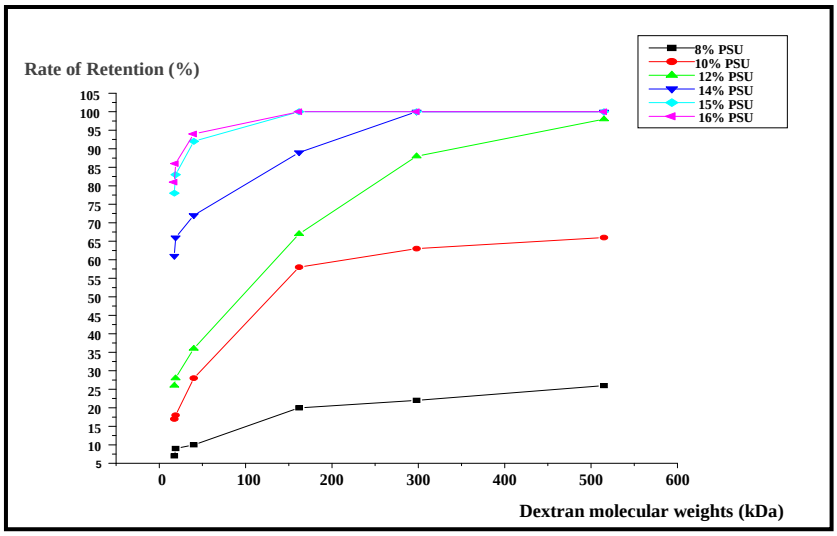

Figure 5: Rate of retention (%) of all membrane as a function of the molecular weights of dextran.

From this figure, it shows that increasing the concentration of the polymer in the PSU colluding causes an increase in the selectivity of the aqueous solutions of dextran of various molecular weights at a constant pressure. Indeed, in agreement with Fick's law [52], the curves in **Fig. 5** show as increasing amounts of PSU UDEL P1700 (%): 8; 10; 12; 14; 15; 16, allow obtaining selectivity also increasing. This explains the increase in polymer concentration, in turn, increases the thickness of the membrane and consequently the narrowing of the pore size of the membrane.From these results, it shows that equalled cut-off threshold at 298 kDa of dextran, membranes having the composition by weight percentages of the PSU (%) 12, 14, 15, 16 and selectivity's data increasing based on the results of permeability obtained in **Fig. 4**, the flow rate is decreasing. This pushed us to the percentage 12% PSU is the optimum value required for the synthesis of the membrane, on the one hand, and that the latter has a high porosity by reports to other diaphragms (14%, 15%, 16 %). Porosity gives the membrane the trans-membrane low energy properties, resistance to clogging of the pores and thus increasing their service life, that no longer exist to the other membranes, on the other hand.Hence the optimized membrane which was taken to carry out this study is that of 12% percentage by weight of polysulfone.

3.1.3. Mechanical characterization of the membrane optimized

The mechanical characterization of the optimized membrane was achieved by measuring the flux of the permeability of water at different pressure values: 5; 10; 15; 20 cm Hg. The curves in **Fig. 6** show the effect of pressure on the permeability of the flow.

From this figure, it shows that the flux of permeation decreases depending on the volume filtered, followed by a steady state at all pressure values. This decrease may be attributed to compaction of the membrane under applied pressure [24].

The **Fig. 7** below shows the evolution of distilled water stream about the pressure applied to quantify the permeability of the membrane optimized.

From this figure, we see that the profile obtained is a straight right, such as the proportional relationship between the flow and the pressure applied is verified. This allows us to say that the experimental results partially satisfy the law of Darcy [24, 53-55].

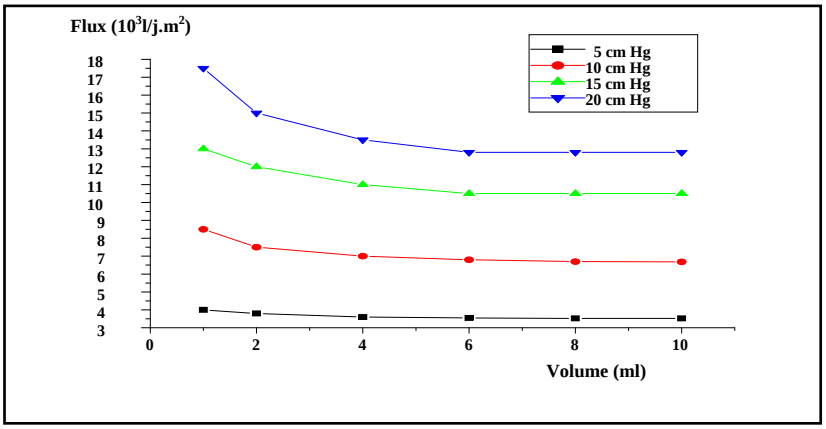

Figure 6: Variation of distilled water flows in the volume to be filtered in different pressure values.

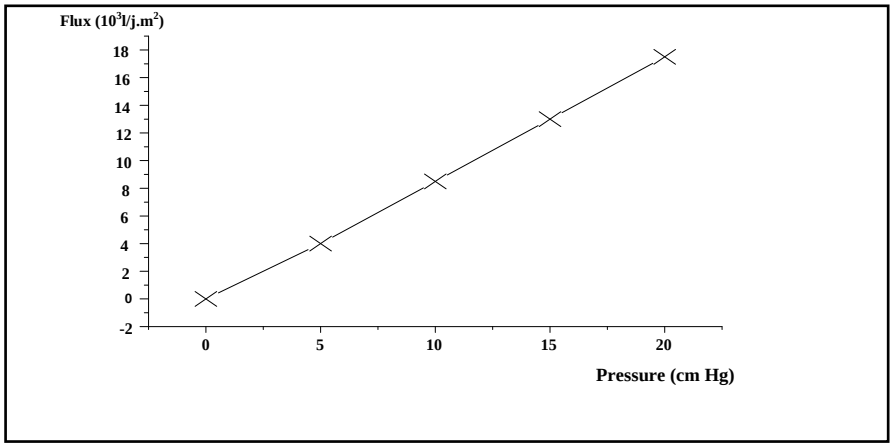

Figure 7: Evolution of the flow of distilled water depending on the pressure.

3.2. Spectroscopic and microscopic characterization of the optimized membrane
3.2.1. FTIR spectroscopic characterization of the membraneoptimized
FTIR spectroscopic analysis of the membrane exploited in this work is shown in the following **Fig. 8**:

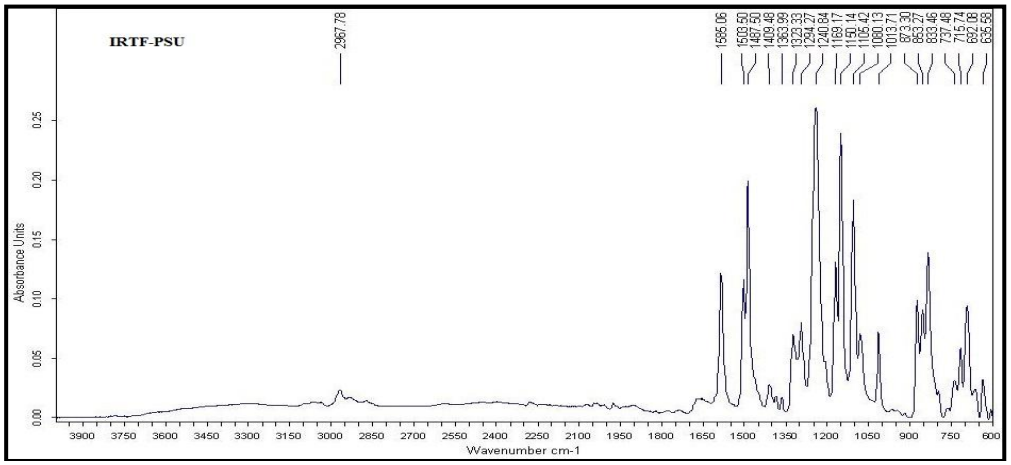

Figure 8: FTIR-ATR spectrum of the membrane optimized (PSU).

According to the FTIR-ATR spectrum of this figure and in the spectra of work [41, 56] of the polysulfone-based membrane, it shows that the adsorption bands are relating to groups of the polymer PSU are grouped in the following table:

Table 1: Absorption bands the corresponding groups of polysulfone membrane.

Absorption bands (cm^{-1})	Corresponding groups
2967.78	C-H(CH_3)
1585.06	$\nu_H$of the arena
1503-1487	Semi-circle ν_H of the arena
1409.48	Asymmetric bond vibration C-CH_3
1363.99	Symmetrical bond vibration C-CH_3
1323.33-1294.27	Asymmetric bond vibration SO_2
1284	Bond vibration ofAryl-O-Aryl
1169.17-1150.14	Symmetrical bond vibration SO_2
1105.42-1113.71	Deformation vibration in para ν_{CH} plan of arena
835	Bending vibration out ν_{CH} plan para to the arena
693	vibration of ν_{CH} the arena
1630 -1710	vibration of group amide (DMF)

From this table, it shows that all the absorption bands belonging to those of the spectrum of the base matrix polysulfone, except that there is another band ofintensity between 1630 and 1710cm^{-1} match the vibration of the amide group which is assigned to operate DMF solvent [56].

3.2.2. (^1H) NMR spectroscopic characterization of the optimized membrane

The spectroscopic characterization of the polysulfone membrane in solution by NMR showed that this polysulfone of the following signals in (^1H) NMR (250 MHz, CD_2Cl_2) δ in ppm: 7.12 (d, J = 8.8, Hd); 7.20 (d, J = 8.5, Hb); 7.00 (d, J = 8.8, Hc); 6.94 (d, J = 8.6, Ha); 1.68 (S, -$C(CH_3)_2$) [41]. The intensities of the peaks obtained were the same for the core matrix that is polysulfone PSU UDEL P1700 [41, 56].

3.2.3. SEM microscopic characterization of the optimized membrane

Microscopic characterization of morphological features of the optimized membrane affected by the SEM is given as follows:

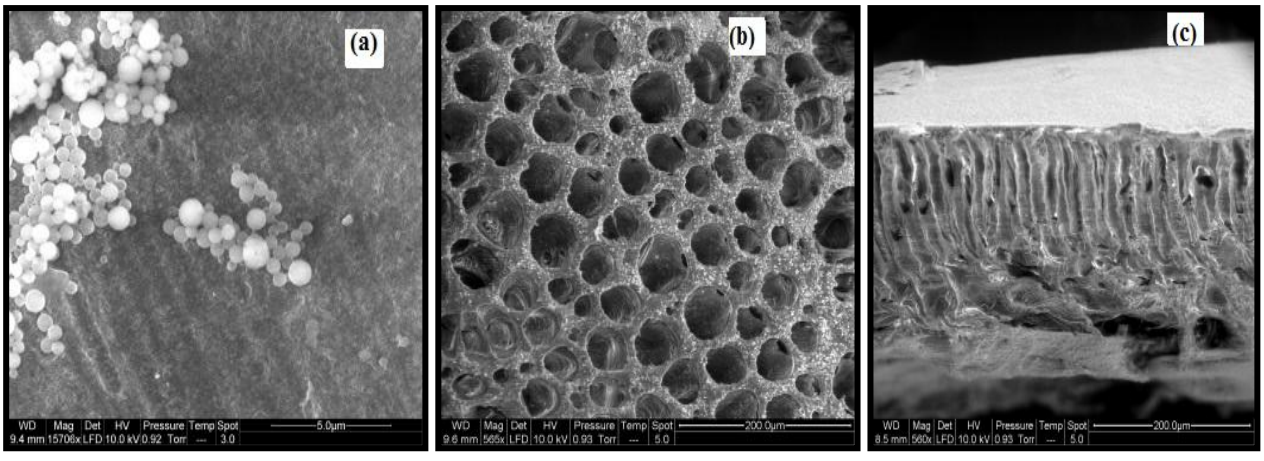

Figure 9: SEM photographs of the optimized membrane (a, b, c).

The observation images optimized polysulfone membrane by the scanning electron microscope, enabled us to see that the top surface of the membrane is a thin dense layer (**Fig. 9** (a)) [24]. The lower surface is porous size of 5 to 60 µm (**Fig. 9** (b)) constituting the membrane support has approximately the same density and much higher porosity than those observed in diameters by setting face [24]. On the other hand, the study of the same

membrane profile showed that the latter has an asymmetrical structure. Indeed, this structure consists of two layers a very fine and dense skin is supported by a layer of high porosity and thickness of 180 µm (**Fig. 9** (c)).

3.3. Characterization of colored waste water models

The results of the characterization of the colored waste water models that have been usedin this work are summarized in the following table:

Table 2: Characteristics of colored solutions models.

Colored solutions models	pH	λ_{max} (nm)	D.Oi.
Indigo	11.80	665	0.256
Black sulphur	11.20	597	0.375

3.4. Results of ultrafiltration of the colored wastewater models

The table below gives the values of the discoloration rate (%RD) of solutions purified by ultrafiltration technique operator optimized membrane.

Table 3: Discolorations rate of water loaded with dyes.

Colored solutions models	pH	λ_{max} (nm)	D.Oi.	D.Of.	% RD
Indigo	11.80	665	0.256	0.050	80.36
Black sulphur	11.20	597	0.375	0.147	60.78

From this table, it shows that the discoloration rate is about 80.36% for the colored solution loaded with indigo and approximately 60.78% for the one charged with the black sulphur. The difference between the values obtained, returns to the degree of the dissolution of the dyes operated in water on the one hand, and second the clogging of the pores of the membrane [47].

Conclusion

The synthetic membrane polysulfone PSU UDEL P1700 by the method of phase inversion is permeable asymmetric in structure and mechanical, hydrodynamic properties are better. The optimization of experimental conditions plays a very important role in the bleaching of colored wastewaters models with vat dyes (indigo and sulfur black) by the ultrafiltration membrane method. Except that the membrane was clogged exploited when disposing of this type of dyestuffs and we will fix this clogging problem in the next study.

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