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Water Purification using Recycled Polymeric Microfiltration Membranes.

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***Abstract*—In our previous work, microfiltration membranes were successfully manufactured by non-solvent induced phase separation (NIPS) method using two concentrations (30 and 35 wt%) of recycled high impact polystyrene (HIPS-R). N, N-dimethyl formamide (DMF) was used as solvent and water as a coagulation bath. These membranes were characterized in terms of chemical composition, surface hydrophilicity, surface cross-section morphology, porosity, and pores size distribution. Accordingly, the membrane's surfaces showed a semi-hydrophilic behavior with contact angles of 81º and 91º for 30 wt% and 35 wt% membranes respectively. In the current study, The prepared membranes were examined for the removal of Humic acid (HA) and Rhodamine B (RhB) dye in a microfiltration process. Filtration experiment showed that pure water flux of 30 wt% membrane was higher than of 35 wt% membrane, also 30 wt% membrane has a higher humic acid and Dye removal efficiency than of 35 wt% membrane. Thus, the results suggest active membranes could be obtained using recycled high impact polystyrene. And then, solve the polymer waste accumulation problem in parallel with help in drinking water crisis solution.**

Keywords—porous membranes, recycled waste, filtration, and hydrophilicity.

1.INTRODUCTION

The demand for pure water is a severe environmental concern [1]. Polymeric membranes are widely used recently for water purification. Wastewater treatment, modifying the membranes to give superior performance, like performing higher fluxes with higher rejection and lower fouling, all of which reduces energy usage, becomes a target [1-2]. In the water purification process, natural organic materials, such as dyes and acids, are removed by ultrafiltration (UF) or microfiltration (MF) [2]. Humic acid (HA) is usually considered as a microbial incubator in water pipes, membrane foulant, and primary source of carcinogenic and mutagenic compounds such as disinfection byproducts [16]. Besides, Rhodamine B (RhB) dye also has a harmful biological impact when drained into water. RhB dye existence in water may cause the death of aquatic species, photosynthetic activities. Complex in nature due to their aromatic structures, which are nonbiodegradable as a result of their optical, thermal and physic-chemical stability [17].

Polymeric membranes as polymer-based materials have distinctive characteristics, like lightweight, easy manufacturing, low cost, and even electrical conductivity as in conductive polymers [3]. Accordingly, their usage covers several fields and applications. Plastics are produced at a considerable rate may reach 230 million tons annually. Besides, plastics left behind a large amount of waste, about 12 % of the solid waste. Efficient treatment of waste polymers is still a severe challenge. Combustion or burying underground are conventional methods to eliminate plastics waste amounts. Unfortunately, these methods have adverse effects on the environment. Consequently, to limit the side effects of polymer waste, plastic recycling is highly considered [4]. Among these polymers that can be recycled, high-impact polystyrene (HIPS) which is formed by the polybutadiene rubber and polystyrene (PS) interaction. HIPS showed significant advantages such as high impact resistance, stability, and ease preparation [1].

Polymers were used widely in separation applications [1, 5-9]. There are different methods for the preparation of PS membranes due to their effective performance for many applications. Zhuang et al. fabricated flat sheet membranes from diverse types of waste PS for the gas separation [6]. It was found that the recycled high-impact polystyrene (HIPS-R) displayed excellent thermal stability and higher gas permeation performance [6]. Garcia et al. studied the application of HIPS-R flat sheet membranes in low-pressure membrane processes [1]. An asymmetric structure with slightly higher porosity, when compared to the commercial HIPS membranes, were successfully obtained [1]. Bussi et al. fabricated PS membranes by phase inversion method with finger-like structure cross-sectional [7]. Ke et al. used the electrospinning method to fabricate nanofibrous PS membranes with a superhydrophobic surface [8]. Microporous PS membranes with enhanced surface hydrophobicity were successfully synthesized in [9]. Furthermore, in our previous study [18], semihydrofilic HIPS-

R membranes were successfully fabricated with different HIPS-R concentrations (20, 25, 30, 35 wt%).

In the current work, HIPS-R membranes with 30 and 35 wt%, were used in the microfiltration (MF) process for water purification. HA and RhB dye were selected to be removed from their aqueous solution. The microfiltration process was investigated in terms of the permeate water flux and the rejection percentage.

2. EXPERIMENTAL

A. Materials

HIPS-R cups obtained from the local Egyptian market were used as the membrane's polymer base material. The cups were washed and dried before use, cut into small square pieces (5 mm average side length), ultrasonicated in distilled water, and dried overnight at 75 ºC. N, N-dimethyl formamide (DMF) was employed as the organic solvent. Both DMF, HA, and Dye were purchased from Sigma-Aldrich. Distilled water was utilized for washing and solution preparation processes.

B. HIPS-R membranes preparation

The membranes preparation procedure is as mentioned in our previous study [18]. Typically, 30, and 35 wt% of HIPS-R were added to 10 mL DMF solvent with stirring at elevated temperature to prepare the polymer homogeneous solutions. Then the solutions were then cast on glass slides with a 200 µm doctor blade at room temperature. Distilled water at room temperature was utilized as a coagulation bath. Finally the membranes were washed and dried overnight at 70 ºC.

C. Membrane characterization

The surface and cross-sectional morphology of the prepared membranes were observed using a Field-emission scanning electron microscopy (FESEM, FEI Quanta 200 scanning electron microscope, FEI Company BV, Netherlands). Also, the pores' size and its distribution on the membranes' surface were analyzed using image analysis free software (Image-J). CA measurements (OCA 15EC Contact angle model, Data Physics Instrument Gmbh) were employed to measure the water contact angle of the prepared membranes. The average porosity of the surfaces of the membrane was calculated by the wet-dry weighting method. Accordingly, a small part of each membrane was soaked in water for 48 h until water filled the pores. Then, the excess water on the sample surface was removed. The surface porosity for each membrane was calculated using Eq. 1:

$$\% \text{ porosity} = \frac{\frac{W_W - W_D}{\rho_w}}{\frac{W_W - W_D}{\rho_w} + \frac{W_D}{\rho_p}} * 100 \quad (1)$$

where, Ww and W_D are the weight of the wet and dry samples (g), respectively. ρ_w is the water density at room temperature (g/cm^3), and ρ_p is the density of the polymer (g/cm^3).

D. Filtration experiments

Separation performance of the resulting membranes was studied in terms of permeate flux (Jp, L/m2 h) and rejection index (R, %) by filtration experiments of 50 mg/L HA solutions and 15 mg/L Dye (RhB) solutions. HA concentration was selected according to previous studies about membrane characterization [1]. These experiments were performed at 25 ± 1 oC and 10 bars. HA and RhB rejections were measured by obtaining their concentrations in each stream using a UV–Visible spectrophotometer (Hewlett-Packard 8453) at a wavelength of 254 and 543 nm for HA and RhB, respectively. Figure 1 represents a schematic diagram for the microfiltration unit used. Therefore, rejection index was calculated as follows:

$$R\ (\%) = \frac{C_f - C_p}{C_f} . 100 \quad (2)$$

where, C_f and C_p are the solute concentration (mg/L) in the feed and permeate streams respectively.

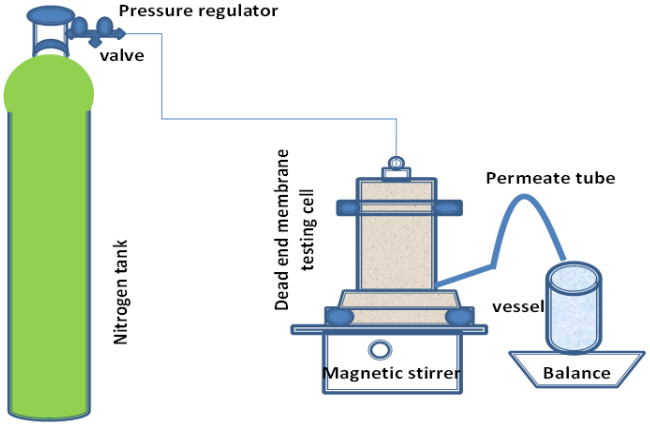

Fig.1. Schematic diagram of bench membrane testing cell.

3. RESULTS AND DISCUSSION

A. Membranes properties

As presented in our previous study [18], membranes with a sponge-like structure with noticeable longitudinal macrovoids were noticed, as shown in Fig. 2. These macrovoids are formed due to the high exchange rate between DMF (solvent) and distilled water (non-solvent) due to the high miscibility [1]. The walls around these voids have a sponge-like structure, especially for the (35wt %) polystyrene membrane. Furthermore, it was clearly illustrated that the surface membrane porosity reduced with the rise of polystyrene concentration.

Figure. 3 showed the pores diameter distribution and water contact angles of the produced membranes. The pore diameter distribution of 30 wt% membrane ranged from 0.1 to 1µm (see Figs. 3a and 3b), and the pore diameter distribution of 35 wt% membrane ranged from 0.1 to 0.5 µm. These results illustrated that the mean pore size decreased with the rise of HIPS-R concentration. Also, the membranes' surfaces showed a semi-hydrophilic behavior with contact angles of 83 º and 91º for 30 wt% and 35 wt% membranes, respectively.The CA increased with the rise of the HIPS-R concentration.

Regarding the surface porosity It was recorded that the membrane with the PS concentration of 30 wt% displayed more significant surface porosity (62.56 %) than that of 35 wt% membrane (61.6%). The porosity declined with the HIPS-R concentrations rise. The large size of the pores and the

formation of a large number of macrovoids in the case of 30 wt% membrane may be the direct reason for the accomplished high porosity. With the increase of PS concentration, the pores size and numbers of macrovoids reduced which in turn lessened the overall porosity of the membrane.

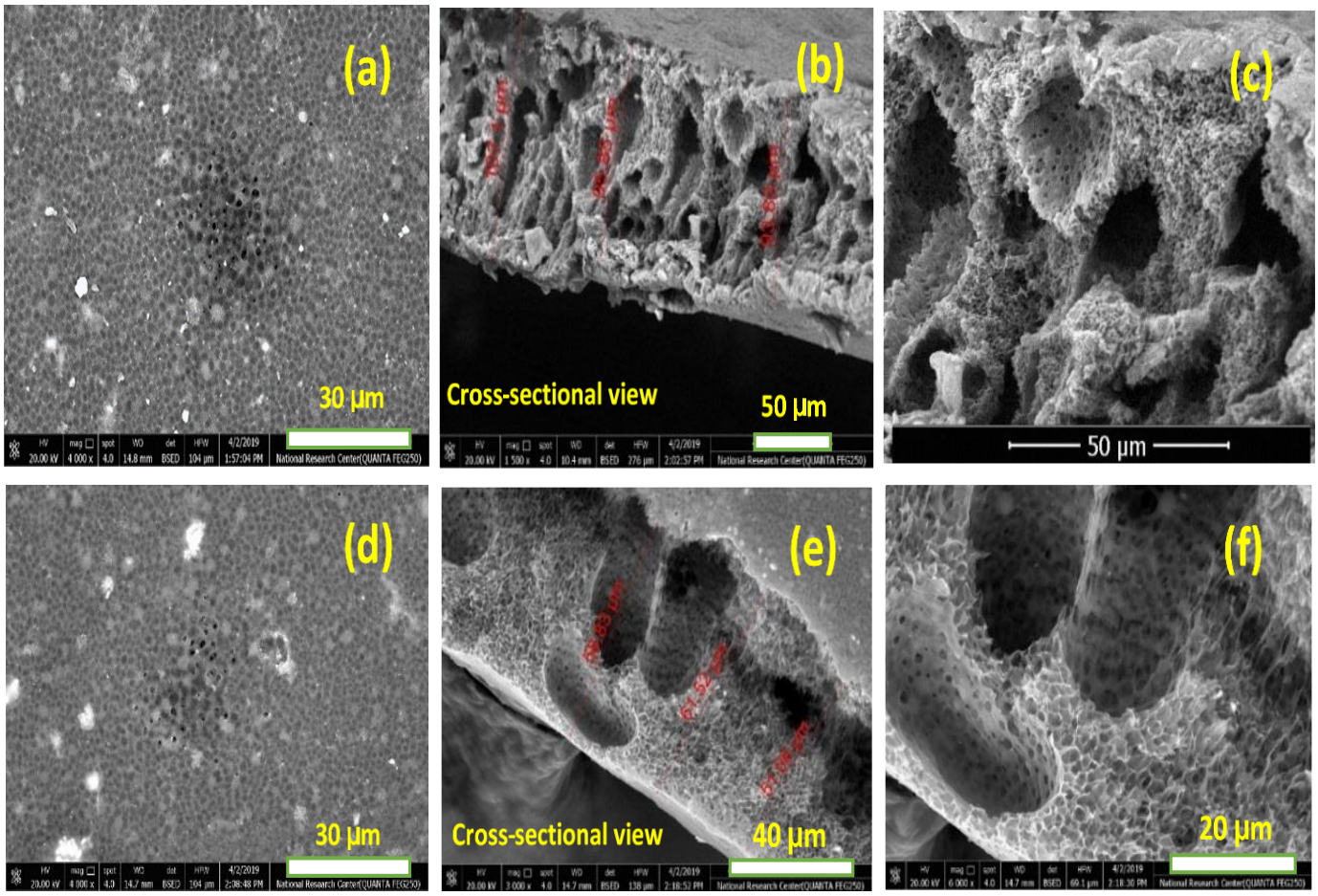

Fig. 2. SEM images of 30 wt% membrane (a) surface, (b) and (c) cross-sections, 35 wt% membrane (d) surface, (e) and (f) cross-sections.

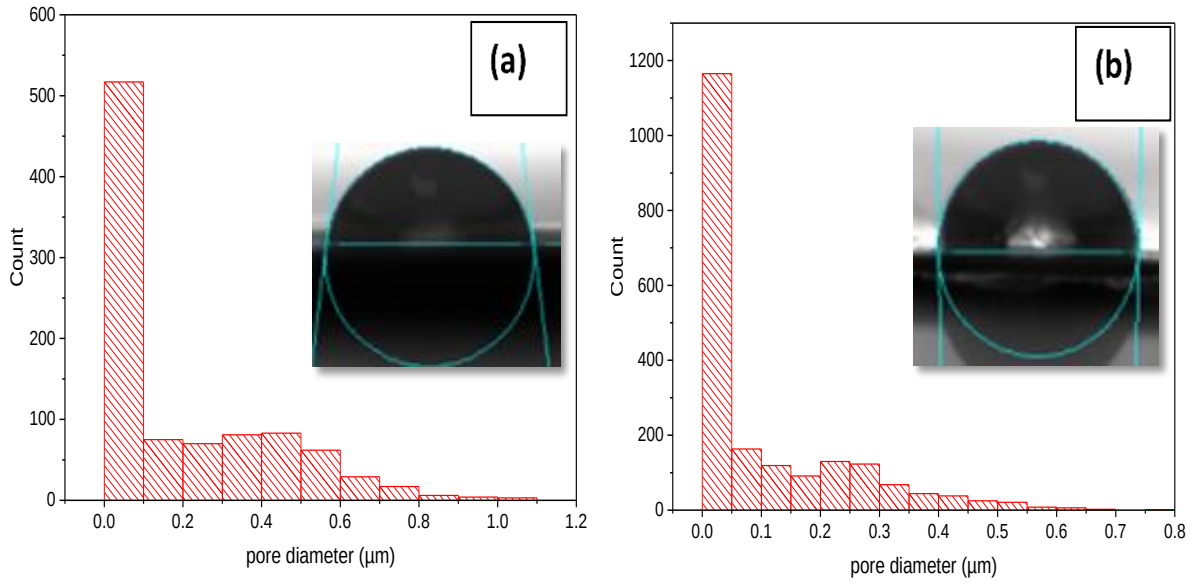

Fig. 3. Pore size distribution and water contact angle of (a) 30 wt%, (b) 35 wt% membranes.

B. Filtration experiments

Permeation flux and solute rejections are the main parameters to evaluate membrane performance [1]. Permeation flux values of HA, RhB, and water for both membranes are shown in Figs 4 and 5, The water permeability value of 30 wt% membrane was higher than that obtained for 35 wt% membrane, mainly due to the high porosity of 30 wt% membrane. The effect of the material and the porosity of membranes on their water flux was studied on many pieces of research [13]. The surface hydrophilicity of the membrane has a strong effect on the water flux thus high surface hydrophilicity leads to high water permeability. The 30 wt% membrane showed higher water permeability than 35 wt% membrane because it has higher surface hydrophilicity.

Finally, Fig. 9. shows the solute rejection of each membrane for HA and Dye solutions. The HA and Dye rejection index of 30 wt% membrane was higher than that obtained by 35 wt%. Previous studies reported the large effect of porosity on water permeability [14]. The higher porosity and lower water contact angle could cause an improvement in hydrophilicity [15-16].

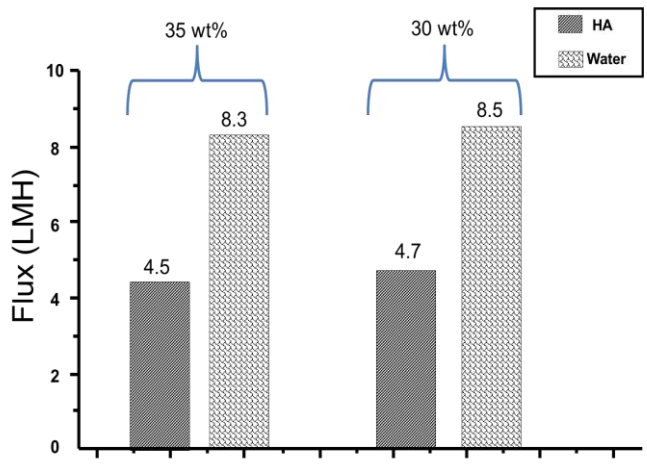

Fig. 4. The flux of the HA and water using both prepared membranes.

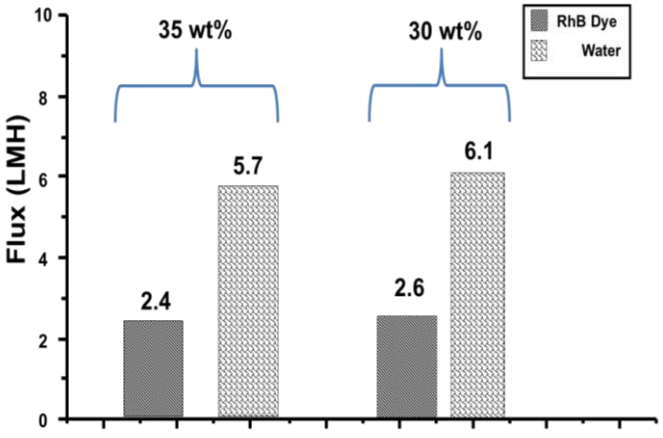

Fig. 5. The flux of the RhB dye and water using both prepared membranes.

The increasing in all these parameters (porosity, hydrophilicity, and water permeability) for 30 wt% membrane could be the reason for the improvement of their HA and Dye rejection index, especially the membrane hydrophilicity which inhibits interactions between the membrane surface and the organic solutes[14].

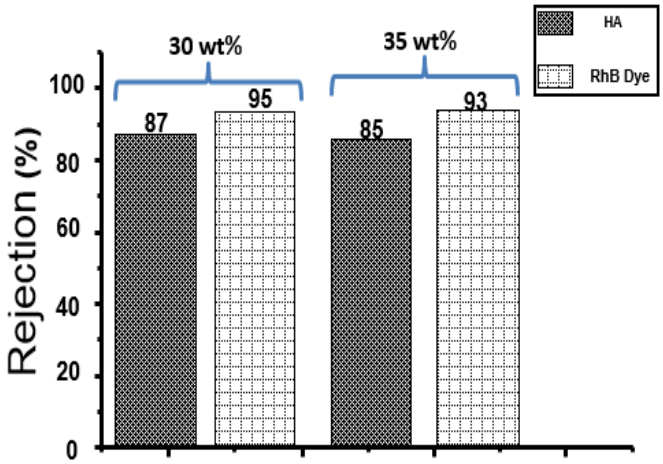

Fig. 6. Rejection of HA and RhB dye using both prepared membranes.

4. Conclusion

Tow hydrophilic flat sheet membranes were successfully fabricated from recycled high impact polystyrene HIPS-R (30 and 35 wt%) in DMF solution. These membranes showed a porous cross-sectional structure with macrovoids. The porosity decreased with the rise of HIPS-R concentrations. The diffusivity of the DMF/HIPS-R solution with the coagulant, water, effect on the formation of the macrovoids and the sponge structure was approved. Also, the porosity gradually diminished from 62.56 to 61.6 % when the concentration raised from 30 to 35 wt%, respectively. Contact angle measurements showed that the membranes had a semi-hydrophilic surface. 30 wt% membrane has the largest water permeability (8.5 L/m^2 h) and rejection index () due to its high porosity and hydrophilicity. Finally, these results suggested that polystyrene wastes can be used to prepare effective flat sheet membranes.

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