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Effect of Different Solvents, Pore-Forming Agent and Solubility Parameter Differences on the Properties of PES Ultrafiltration Membrane

Seren ACARER*[1]

Abstract

In the production of polymeric membranes used in water treatment by the non-solvent-induced phase separation (NIPS) method, the materials used in the membrane casting solution and the interaction of these materials greatly affect the properties and performance of the obtained membranes. In this study, polyethersulfone (PES) membranes are produced by the NIPS method using two different solvents, dimethyl sulfoxide (DMSO) and N-methyl-2-pyrrolidone (NMP), and polyvinylpyrrolidone (PVP) as pore-forming agent. Chemical functional groups and morphologies of the produced membranes are investigated by Fourier transform infrared (FTIR) spectroscopy and scanning electron microscopy (SEM), respectively. The viscosity of the membrane casting solutions and the hydrophilicity, porosity, mean pore size, and mechanical properties of the membranes are characterized. The pure water flux (PWF) of the membranes is determined at 1 and 3 bar pressures. The Hansen solubility parameters (HSP) of the materials used in membrane production are calculated and the effect of the interactions of DMSO, NMP, and PVP with PES and/or non-solvent (water) on the membrane properties are investigated.

Keywords: Membrane characterization, phase separation, solvent, PVP, hansen solubility parameters

1. INTRODUCTION

Membrane processes are among the advanced treatment methods commonly used in the treatment of drinking water and wastewater [1, 2]. NIPS is a widely used method in the production of commercially available polymeric membranes used in water treatment [3]. The fact that the NIPS method is simpler than other methods used in the production of polymeric membranes and that the porosity, pore size, and thickness of the membrane can be easily controlled by changing some conditions during production are among the main reasons for the widespread use of the NIPS method [4].

Polyethersulfone (PES) is widely preferred in the production of polymeric membranes due to its high chemical resistance, good chlorine resistance, good thermal and mechanical

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stability, low cost, and good miscibility with solvents [5, 6]. During the production of polymeric membranes, large amounts of conventional organic solvents such as NMP, DMSO, dimethylformamide (DMF), dimethylacetamide (DMAc), and tetrahydrofuran (THF) are used [7]. PES membranes are produced by many researchers using different solvents such as NMP [5, 8-9], DMSO [10], and DMF [8].

In membrane production with NIPS, the polymer-solvent relationship in the membrane casting solution and the solvent-non-solvent relationship when the membrane is immersed in the coagulation bath significantly affect the morphology of the membrane [11]. The thermodynamic stability of the casting solution, polymer-solvent affinity, and solvent-non-solvent affinity cause accelerated or delayed liquid-liquid demixing during phase separation, resulting in membranes with different structures, pore size, and porosity. For instance, Fahrina et al. reported that PES-NMP and PES-DMF membranes exhibit denser membrane surfaces and internal structure compared to PES-DMSO membranes, due to the lower affinity of NMP and DMF for water (non-solvent) than DMSO [12].

Pore-forming agents (such as PVP and PEG) added to the membrane casting solution cause the formation of membranes with different morphologies, properties, and performance due to their effects on the stability of the casting solution, their hydrophilicity, and solution viscosity [13-16]. In a recent study by Tofighy et al., it has been reported that 3% wt. PVP-doped PVDF membranes have higher hydrophilicity, porosity, and pore size compared to undoped PVDF, and the membrane's PWF is improved by incorporating PVP into PVDF membrane [17]. In the study of Kourde-Hanafi et al., it was found that PES-PVP membranes have a denser top layer than pure PES regarding casting solution viscosity, but PES-PVP membrane has higher permeability than pure PES membrane due to the hydrophilic nature of PVP [18].

In this study, PES-DMSO, PES-NMP, and PES-PVP-NMP membranes are produced by the NIPS method to determine the effects of different solvents (DMSO and NMP) and pore-forming agent (PVP) on PES membrane properties and performance. The effects of DMSO, NMP, and PVP on the chemical groups, morphology, hydrophilicity, mechanical properties, and PWF of the membrane are revealed as a result of characterization studies.

2. MATERIAL AND METHOD

2.1. Materials

PES (VERADEL® 3000P) is obtained from Solvay Specialty Polymers. NMP and DMSO of ≥99.5% and ≥99.9% purity, respectively, are purchased from Merck. PVP with a molecular weight of 40,000 g/mol is purchased from Sigma-Aldrich.

2.2. Membrane Production

PES-DMSO, PES-NMP, and PES-PVP-NMP flat sheet membranes are produced by the NIPS method. Solutions containing 16% wt. PES and 84% wt. solvent (DMSO or NMP) are mixed in a heated magnetic stirrer (WiseStir, MSH-20A) for 24 hours at 60 °C. After obtaining a homogeneous solution, the solution is kept in an ultrasonic bath (Weightlab Instruments) at 25 °C for 30 minutes to remove air bubbles in the membrane casting solution. Membranes are cast on a glass plate to form polymeric films using a 200 μm thick casting blade. After waiting for 10 seconds, the casting solution in the form of a film on the glass plate was immersed in a coagulation bath containing non-solvent (distilled water) and left for 15 minutes. Membranes are obtained as a result of the change between solvent-non-solvent. After the obtained membranes are thoroughly washed with distilled water, they are kept in distilled water at 4 °C for 24 hours, and then the membranes are characterized. The production process of membranes produced with different solvents is shown schematically in Figure 1. The PES-PVP-NMP membrane is also produced using the same procedure using 8% wt. PVP and 76% wt. NMP. The composition of all produced membranes is given in Table 1.

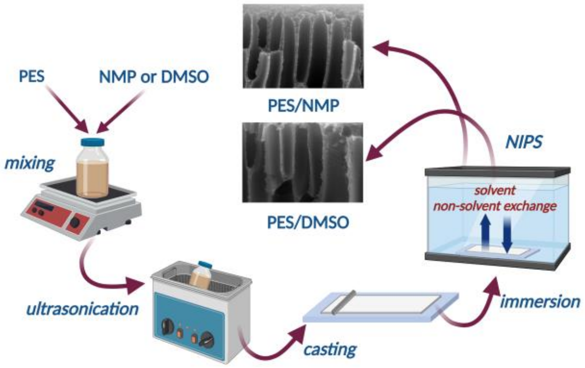

Figure 1 Schematic diagram of the membrane production process

Table 1 Composition of the produced membranes

Membrane Code	***PES (wt.%)***	***DMSO (wt. %)***	***NMP (wt.%)***	***PVP (wt.%)***
PES-DMSO	16	84	-	-
PES-NMP	16	-	84	-
PES-PVP-NMP	16	-	76	8

2.3. Membrane Characterization

2.3.1. Membrane casting solution viscosity

The viscosities of the casting solutions of the membranes are measured by a viscometer (AND, SV-10). Viscosity measurements are carried out at 25 °C.

2.3.2. FTIR

Chemical groups of the membranes are determined using the FTIR spectrometer (Perkin Elmer, Spectrum 100). The FTIR spectra of the membranes are recorded in the wavenumber range of 4000-650 cm^{-1}.

2.3.3. SEM

The surface and cross-sectional morphology of the produced membranes are investigated using SEM (FEG, FEI Quanta 250). To obtain SEM views, firstly, the insulating membrane surfaces are made conductive by coating them with gold. While the surface views of the membranes are examined at 20000x magnification, the cross-sectional views are examined at 10000x and 40000x magnifications.

2.3.4. Contact angle

The surface hydrophilicity of the membranes is determined by the sessile drop technique by contact angle meter (KSV Instruments, CAM 101). Contact angle measurements are carried out at 25 °C. The contact angle results are obtained by determining the angle between the distilled water and the membrane surface by dripping distilled water on the membrane surface. Distilled water is dropped at least three times in different locations of each membrane sample and contact angle results are given as the average of three measurements.

2.3.5. Water content

The produced membranes are left to dry in an oven at 60 °C (NUVE, EN 500) for 24 hours. The dry weights of the membranes are first determined using a precision balance (KERN, 573). Afterward, the membranes are immersed in a container filled with distilled water, and the excess water on them is quickly removed with a blotting paper, and the membranes are weighed again to determine their wet weight. The water content of the membranes is calculated using Equation 1.

$$\text{Water content (\%)} = \frac{W_w - W_d}{W_w}\ x\ 100 \qquad (1)$$

where W_w and W_d are wet and dry weights of membranes (g), respectively.

2.3.6. Porosity and mean pore size

The porosity (ε) and mean pore size (r_m) of the membranes are calculated using Equation 2 and Equation 3 (Guerout-Elford-Ferry Equation), respectively.

$$\varepsilon\,(\%) = \frac{W_w - W_d}{A\,l\,\rho}\,x\,100 \qquad (2)$$

where W_w is the wet weight of the membrane (g), W_d is the dry weight of the membrane (g), A is the membrane area (cm^2), l is the membrane thickness (cm), and ρ is the density of the water (0.998 g/cm^3).

$$r_m = \sqrt{\frac{(2.9-1.75\varepsilon)\ \mathrm{x}\ 8\eta l Q}{\varepsilon\ \mathrm{x}\ A\ \mathrm{x}\ \Delta P}} \qquad (3)$$

where ε is membrane porosity, η is the viscosity of water (8.9 x 10^{-4} Pa.s), l is membrane thickness (m), Q is the volume of permeate water per unit time (m^3/s), A is effective membrane area (m^2), ΔP operating pressure (0.3 MPa).

2.3.7. Mechanical tests

Tensile tests of the membranes are carried out using a universal testing machine (Shimadzu, AG-IS (50kN)). The membranes are left to self-dry for 24 hours at room temperature at 25°C. The membranes are then adhered to the aluminum plates to prevent the slipping of the tips on the tester. The tensile strength and elongation at break of the produced membranes are determined.

2.3.8. PWF

A dead-end filtration system (Tin Mühendislik) is used in PWF tests of the membranes. The effective surface area of the membranes placed in the filtration system is 19.6 cm^2. In the pure water tests, firstly, pure water is passed through the membranes at 5 bar pressure using nitrogen gas (N_2) to obtain a stable flux and to open the pores of the membranes thoroughly. In other words, the membranes are first compressed at 5 bar. Then, distilled water is passed through each membrane at 1 bar and 3 bar, respectively. The permeate collected from the outlet of the filtration system is collected in a beaker on a precision scale and the time-weight graph is transferred to the computer and the PWF of the membranes is calculated in L/m^2.h unit. The PWF of the membranes is calculated using Equation 4.

$$J = \frac{V}{A\,\Delta t} \qquad (4)$$

where J is the membrane flux (L/m^2.h), V is the permeate volume (L), A is the effective membrane area (m^2), and Δt is the time (h).

2.4. Calculation of HSP

The solubility parameter (δ) is useful in estimating the relative attraction force between solvents and solutes numerically [19]. Using the HSP, the total solubility parameter of the polymer or solvent is calculated using Equation 5.

$$\delta_t = \sqrt{{\delta_d}^2 + {\delta_p}^2 + {\delta_h}^2} \qquad (5)$$

where δ_t represents the total solubility parameter and δ_d, δ_p and δ_h are dispersion, polar, and hydrogen bonding parameters respectively.

The difference between the solubility parameter of the polymer and solvent can be calculated using Equation 6.

$$\Delta\delta_{P-S} = \sqrt{(\delta_{d,p}-\delta_{d,s})^2 + (\delta_{p,p}-\delta_{p,s})^2 + (\delta_{h,p}-\delta_{h,s})^2} \qquad (6)$$

The difference between the solvent and non-solvent solubility parameters can be calculated using Equation 7.

$$\Delta\delta_{S-NS} = \sqrt{(\delta_{d,s}-\delta_{d,ns})^2 + (\delta_{p,s}-\delta_{p,ns})^2 + (\delta_{h,s}-\delta_{h,ns})^2} \quad (7)$$

where P, S, and NS represent polymer, solvent, and non-solvent (water), respectively.

Table 2 shows the solubility parameters of PES, DMSO, NMP, and water.

Table 2 Solubility parameters of PES, PVP, NMP, DMSO, and water

	δ_d (MPa)$^{0.5}$	**δ_p (MPa)$^{0.5}$**	**δ_h (MPa)$^{0.5}$**	**δ_t (MPa)$^{0.5}$**
PES	19.6	10.8	9.2	24.19
PVP	21.4	11.6	21.6	32.54
NMP	18	12.3	7.2	22.95
DMSO	18.4	16.4	10.2	26.67
Water	15.5	16	42.3	47.80

3. RESULTS

3.1. Membrane Casting Solution Viscosity

The viscosity of the membrane casting solution plays an important role in the morphology of the membranes produced by NIPS. The viscosity results of membrane casting solutions at 25 °C are shown in Figure 2. The viscosity of the PES-DMSO casting solution (6.77 Pa.s) is found to be approximately 5 times higher than the viscosity of the PES-NMP solution (1.37 Pa.s). This result can be attributed to the fact that DMSO is more viscous than NMP [20]. With the inclusion of 8% wt. PVP in the casting solution of the PES-NMP membrane, the viscosity increased 6.7 times to 9.2 Pa.s. The higher polymer content of the PES-PVP-NMP solution (16% wt. PES and 8% wt. PVP) and lower solvent content (76% wt.) of the PES-NMP solution are increased viscosity. In this study, the increase in the membrane casting solution with the addition of PVP is consistent with the studies in the literature on the increase in viscosity of polymeric solutions with the addition of PVP [21-23] and with the increase of PVP concentration [21, 24] in the solution.

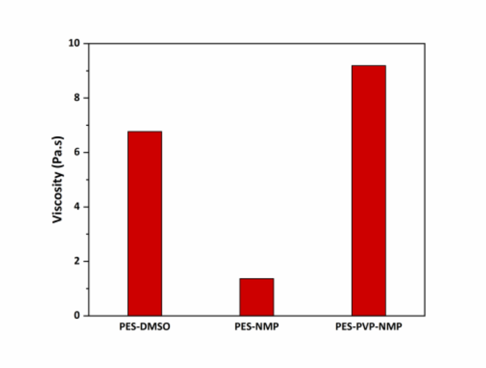

Figure 2 Viscosity of membrane casting solutions

3.2. Confirmation of the Chemical Structure of Membranes

To confirm the chemical structure of PES and PES-PVP membranes, the chemical groups of the membranes are determined by FTIR analysis. FTIR spectra of PES-DMSO, PES-NMP, and PES-PVP-NMP membranes are shown in Figure 3. Similar peaks are obtained in PES-DMSO and PES-NMP membranes. In the NIPS method, an exchange occurs between the solvent in the casting solution and the non-solvent (water) in the coagulation bath, and the solvent moves away from the membrane structure. Therefore, similar bands are obtained as a result of the chemical characterization of PES-DMSO and PES-NMP membranes with FTIR, regardless of the solvent type used in the membrane production process. The C=C stretching vibration originating from the aromatic benzene rings of PES corresponded to the bands at 1485 and 1577 cm^{-1}. The C-O-C stretching vibration of aromatic ether is confirmed by the band at 1237 cm^{-1}. The peaks occurring at 1147 cm^{-1} and 1103 cm^{-1} are caused by the O=S=O vibration [25, 26]. In the FTIR spectrum of the PES-PVP-NMP membrane, unlike the PES-NMP and PES-DMSO membrane, a new peak occurred at 1675 cm^{-1} corresponding to the stretching vibration of the carbonyl (C=O) group of PVP.

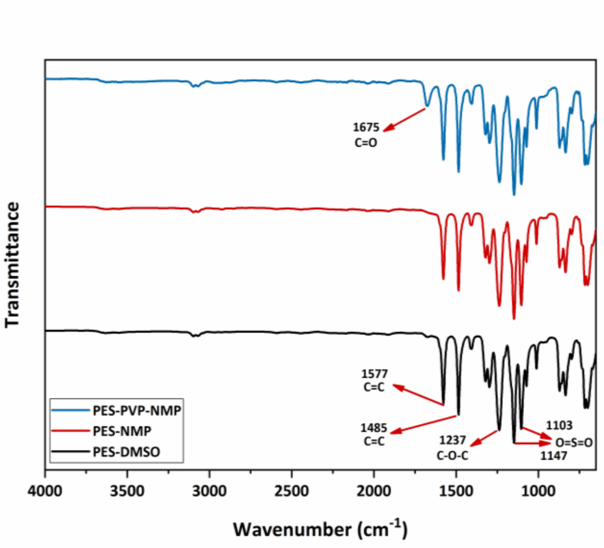

Figure 3 FTIR spectra of membranes

3.3. Membrane Morphology

3.3.1. Effect of solvents on membrane morphology

SEM surface and cross-section views of the membranes are given in Figure 4. All membranes exhibited an asymmetrical structure consisting of a top layer responsible for selectivity and a porous sublayer. When DMSO was used as a solvent in the production of PES membrane, the surface porosity of the membrane increased significantly compared to the use of NMP. It is seen that both PES-DMSO and PES-NMP membranes exhibit finger-like pore structures. However, fewer but larger finger-like pores extending from the surface of the membrane to the sublayer occurred in PES-DMSO compared to PES-NMP. Another remarkable point is that the regions surrounding finger-like pores in the PES-NMP membrane are thicker and more pronounced sponge-like, while the thickness of these regions is less in the PES-DMSO membrane. In addition, a thin top layer responsible for selectivity is formed in the PES-DMSO membrane structure, while a thicker and sponge-like top layer is formed in the PES-NMP membrane. The membrane structure is sponge-like and the thick top layer is the result of the delay of liquid-liquid demixing during phase separation [27]. SEM views revealed that the type of solvent used in membrane production affects membrane surface porosity, top layer thickness, top layer structure, and pore size.

The changes in membrane morphology depending on the solvent type can be explained by the solubility parameter (δ). Two factors, equilibrium thermodynamics and kinetics of the solution play an important role in the morphology of membranes produced with NIPS. High interaction between polymer and solvent, i.e. lower $\delta_{P\text{-}S}$, indicates high thermodynamic stability [28, 29]. On the other hand, the lower the $\delta_{S\text{-}NS}$ value, the greater the interaction between the solvent and the non-solvent (water). Table 3 shows the polymer-solvent interaction (P-S) and solvent-non-solvent (water) interaction (S-NS) results calculated from the dispersion, polar, and hydrogen bonding parameters of PES, NMP, DMSO, and water. The fact that the interaction between PES-NMP (2.97 MPa$^{0.5}$) is stronger than the interaction between PES-DMSO (5.81 MPa$^{0.5}$) indicates that NMP is a better solvent for PES and its solution stability is higher. On the other hand, the fact that the DMSO-water interaction (32.23 MPa$^{0.5}$) is stronger than the NMP-water (35.38 MPa$^{0.5}$) interaction indicates that the affinity of DMSO to water is higher than that of NMP to water. The low affinity of DMSO for PES and high for water compared to NMP causes the acceleration of liquid-liquid demixing by increasing the interaction between non-solvent and solvent during the NIPS. If DMSO is preferred as the solvent in membrane production, the formation of a membrane with a less sponge-like structure and a thinner top layer is compatible with the affinity results calculated from Hansen solubility parameters.

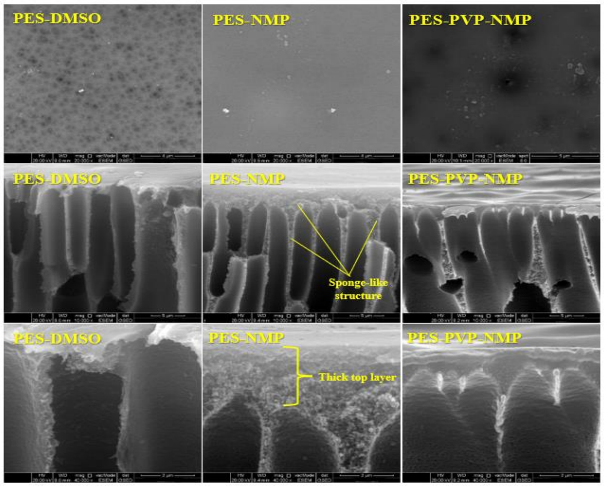

Figure 4 SEM surface and cross-section views of the membranes

Table 3 Hansen affinity parameter between PES-DMSO, PES-NMP, DMSO-water, and NMP-water

	PES-Solvent Interaction (P-S) (MPa)$^{0.5}$	**Solvent-Non-Solvent Interaction (S-NS) (MPa)$^{0.5}$**
NMP	2.97	35.38
DMSO	5.81	32.23

3.3.2. Effect of PVP on PES membrane morphology

As seen in Figure 4, the surface porosity of PES-NMP membranes increased with the addition of PVP. The increase in porosity can be attributed to the fact that the presence of a third component (PVP) other than polymer and solvent in the membrane casting solution causes an increase in the thermodynamic instability of the solution [30]. The fact that the δ_t value of PVP is closer to the δ_t value of water compared to the δ_t value of PES indicates that PVP has a higher affinity for water (Table 2). Due to the high affinity of PVP for water and its solubility in water, large pores were formed on the membrane surface and the top layer of the membrane was thinned during the PVP washing from the near-surface parts of the membrane and passing from the casting solution to the water. On the other hand, the higher viscosity of the PES-PVP-NMP membrane resulted in a more sponge-like and tighter structure of the membrane interior than the PES-NMP membrane. This result revealed that in addition to the kinetic factor, viscosity is also an important factor in membrane morphology.

3.4. Surface Hydrophilicity

Membrane surface hydrophilicity is characterized by the contact angle, and hydrophobic membrane surfaces are expressed with high contact angle while hydrophilic membrane surfaces are expressed with a low contact angle [31]. The contact angle and water content results of the membranes are shown in Figure 5. The fact that the contact angle of PES-DMSO (61.1°) is 28% lower than the contact angle of PES/NMP (78.4°), indicates that PES-DMSO has higher hydrophilicity. The significantly higher surface porosity of the PES-DMSO membrane (Figure 4) allowed increased diffusion of water and resulted in higher surface hydrophilicity. Similarly, PES-PVP-NMP membrane exhibited a more porous structure compared to PES-NMP and the hydrophilic carbonyl group (C=O) of PVP decreased the contact angle by 19.4% and increased the hydrophilicity of the membrane surface [32]. Since membranes with higher surface hydrophilicity exhibit higher antifouling properties, the order of membrane surfaces to exhibit higher fouling resistance under the same conditions are PES-DMSO > PES-PVP-NMP > PES-NMP.

Another parameter related to membrane hydrophilicity is the water content of the membranes. The water content of all membranes is above 70%. The water content of PES-DMSO is 5.5% higher compared to PES-NMP. The high surface porosity and the thinner top layer of PES-DMSO allowed water to pass into the membrane structure more easily and absorb more water in finger-like pores. The contact angle and water content results of the membranes produced with different solvents revealed that DMSO is a more suitable solvent than NMP in the production of PES membrane to increase the hydrophilicity. On the other hand, the water content of PES-PVP-NMP was found to be 3% higher than that of PES-NMP, although the internal pores of PES-PVP-NMP were less than the internal pores of PES-NMP. This may be related to the fact that the large pores on the surface of the PVP-doped membrane and its thin top layer allow the water to pass through more and faster than PES-NMP, which has a denser surface and thicker top layer.

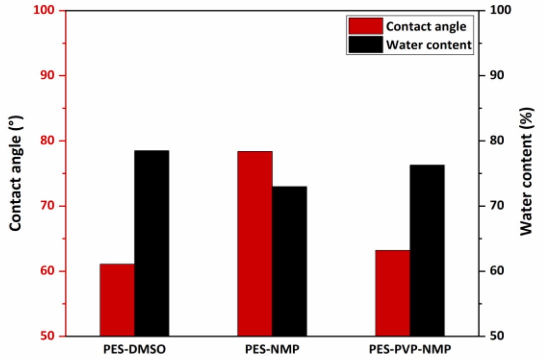

Figure 5 Contact angle and water content of membranes

3.5. Porosity and Mean Pore Size

The calculated porosity and mean pore size values of the membranes are given in Table 4. The highest porosity is obtained in the PES-DMSO membrane, and the lowest porosity is obtained in the PES-PVP-NMP membrane. The calculated porosity results are consistent with the SEM cross-sectional views. The mean pore sizes of the membranes varied in the range of 18.2-27.8 nm, and these mean pore sizes indicate that the produced membranes are ultrafiltration (UF) membranes [33].

Table 4 Porosity and mean pore size of the membranes

	Porosity (%)	**Mean pore size (nm)**
PES-DMSO	66.4	19.4
PES-NMP	60.1	18.2
PES-PVP-NMP	54.3	27.8

3.6. Mechanical Properties

To determine the mechanical properties of the membranes, tensile strength and elongation at break are investigated and the results are shown in Figure 6. The tensile strengths of PES-NMP and PES-DMSO membranes are found to be 15.1 MPa and 13.9 MPa, respectively. Since the high surface porosity of PES-DMSO causes a decrease in the membrane surface area, it has a negative effect on the mechanical strength of the membrane, causing it to exhibit lower tensile strength [34, 35]. Also, the elongation at break of PES-NMP membrane is higher than that of PES-DMSO. Also, the elongation at break of PES-NMP membrane is higher than that of PES-DMSO.

When the effect of PVP on the mechanical properties of the PES-NMP membrane is investigated, it was observed that the tensile strength and elongation at break decreased. The large diameter surface pores, high mean pore size, and thin top layer of PES-PVP-NMP resulted in a decrease in the maximum stress that the membrane can withstand before rupture. In addition, the elongation at break of the membrane is reduced by the incorporation of PVP into the PES matrix. In other words, PVP reduced the flexibility and ductility of the membrane [36].

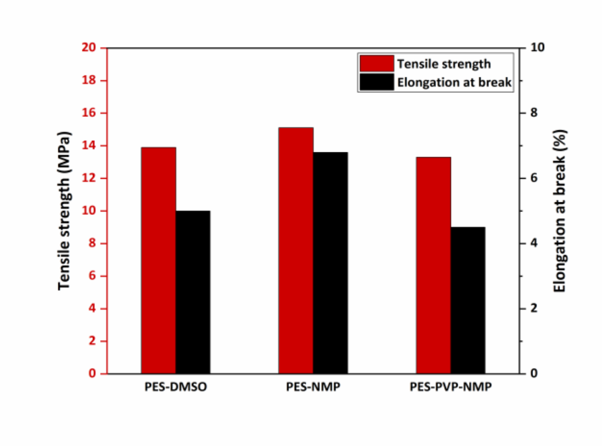

Figure 6 Mechanical properties of membranes

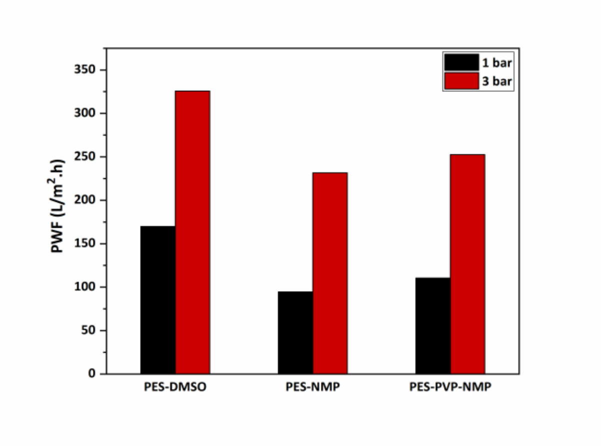

Figure 7 PWF performance of membranes

3.7. PWF Performance of Membranes

The PWF performance of the membranes at 1 bar and 3 bar pressure is presented in Figure 7. With the increase in pressure, the PWF performance of the membranes also increased. PWF of PES-DMSO is found to be 44.2% and 33% higher than PES-NMP at 1 bar and 3 bar, respectively. In addition, the results of the contact angle and water content of the membranes are compatible with the PWF performance results. The relatively high surface hydrophilicity of PES-DMSO increased the interaction between water and the membrane surface. In addition, the high porosity, larger pore sizes, and thinner skin layer of the PES-DMSO membrane reduced the hydraulic resistance of the membrane and increased the passage of water through the membrane.

Although the inclusion of PVP in PES-NMP increased the surface porosity and hydrophilicity, increasing the PWF, the PWF of PES-PVP-NMP is found to be lower than that of PES-DMSO. Since the addition of 8% wt. of PVP to the casting solution increased the viscosity, the formation of finger-like voids was suppressed, and this may have prevented much higher flux values from being reached in the PVP-doped membrane.

4. CONCLUSION

Determination of the most suitable membrane casting solution composition in terms of desired membrane properties and performance contributes to the production of water treatment membranes more economically and with the use of chemicals that are more harmless to the environment. In this study, PES-DMSO, PES-NMP, and PES-PVP-NMP membranes are produced by NIPS method and the characterization of the produced membranes was performed. In addition, the effect of different solvents (DMSO and NMP) on the membrane, especially their morphology, is explained by Hansen solubility parameters. As a result of the study, the following results were obtained:

1) According to Hansen solubility parameters calculations, the relatively stronger and weaker affinity of NMP to PES and water (non-solvent), respectively, caused a delay in demixing during the formation of the PES-NMP membrane. As a result of the demixing delay, the PES-NMP membrane with less porosity and a thicker internal structure is obtained. On the contrary, relatively weaker, and stronger interactions of DMSO with PES and water, respectively, resulted in a more porous and less dense membrane.

2) PES-DMSO membrane produced with DMSO, which is a more “green” solvent compared to NMP, has a more porous surface, thinner top layer, higher hydrophilicity, higher porosity and mean pore size compared to PES-NMP. PWF of PES-DMSO is found 44.2% higher than PES-NMP at 1 bar. However, PES-DMSO exhibited lower tensile strength and elongation at break compared to PES-NMP.

3) With the inclusion of PVP, a pore-forming agent, in the PES-NMP membrane, the pore size, hydrophilicity, and PWF of the membrane increased, but the tensile strength and elongation at break decreased. Since the inclusion of 8% wt. PVP in the membrane casting solution provides a significant increase in the viscosity of the casting solution, a denser membrane interior structure was obtained.

This study revealed that the use of different solvents in membranes produced by the NIPS and the inclusion of a pore-forming agent in the membrane casting solution led to different membrane properties and performance, provided that the polymer (PES) ratio in the membrane casting solution is the same.

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Authors' Contribution
The first author S.A. contributed 100%.

The Declaration of Conflict of Interest/ Common Interest
No conflict of interest or common interest has been declared by the authors.

The Declaration of Ethics Committee Approval
This study does not require ethics committee permission or any special permission.

The Declaration of Research and Publication Ethics
The authors of the paper declare that they comply with the scientific, ethical and quotation rules of SAUJS in all processes of the paper and that they do not make any falsification on the data collected. In addition, they declare that Sakarya University Journal of Science and its editorial board have no responsibility for any ethical violations that may be encountered, and that this study has not been evaluated in any academic publication environment other than Sakarya University Journal of Science.

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