Original Article: Investigation of Experimental Variables on Polyether Sulfone (PES) Membrane in Dehydration of Natural Gas

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ABSTRACT

This study investigated the dehydration of natural gas of Tabnak sweet gas field using membrane process, with PES membrane as a new, simple and cheap separation process. The effect of various experimental variables, including pressure, temperature, and nano silica particles in dehydration of natural gas by PES were investigated. PES and its silica nanocomposite membranes were prepared by solution blending in N,N-Dimethylformamide (DMF) and casting-evaporation method. The homogeneity and nano-scale distribution of prepared PES-silica membranes were characterized using Fourier transform infra-red spectroscopy (FTIR) and scanning electron microscope (SEM). Dehydration tests were performed at 2 to 10 bar and 20 to 50 °C, with nitrogen sweep gas. With an increase in the feed pressure, the permeated water was inhanced. On the other hand, decreasing temperature, caused to improve the dehydration on process. Using PES-silica membrane increased water permeation.

Introduction

Since the first industrial membrane gas separation unit was established in 1980, the use of membranes in this field has grown significantly. Membrane processes of gas separation are divided into three groups. The first group includes processes that have been developed and become popular and include the production of nitrogen from air, hydrogen recycling and air drying. These processes account for more than 80% of the market for gas separation processes with membranes. The second group is developing processes that include the separation of carbon dioxide from natural gas, organic vapors from air, nitrogen, as well as recovery light hydrocarbons are gases removed from various refinery and petrochemical units.

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All of these processes are running on a large industrial scale and have been installed in hundreds of different units, and these applications are growing and developing dramatically due to the increased quality of membranes and improved process design.

For example, cellulose acetate membranes are used to separate carbon dioxide from natural gas for 15 years. The production of membranes with higher flux and greater selectivity has caused these processes to compete with the amine absorption process today. The use of silicon rubber membranes to separate vapors in the refining and petrochemical industries is also increasing. The third category of gas separation processes with membranes are processes that must be developed. The natural gas refining process including dehydration, recovery of liquefied hydrocarbons (containing more than three carbons) and separation of hydrogen sulfide have been studied.

Comrade et al. consider membrane technology to be the best alternative to traditional natural gas dehumidification methods. These researchers consider a wide range of polymer membranes to have a high selectivity for separating water from methane [34].

Salem et al. [35] reported that water vapor permeation from zeolite membranes is based on the adsorption-diffusion model, but in polymer membranes is the dissolution-penetration model.

Sander et al. evaluated the simultaneous separation of CO_2 and H_2O by polyethylene oxide from combustion exhaust gases. In this separation, H_2O and CO_2 are practically separated from the mixture of H_2O, CO_2 and N_2 at the same time. Water permeability increases powerfully due to its activity coefficient, while CO_2 permeability decreases slightly with increasing water activity coefficient. Due to the ability of this copolymer to simultaneously separate water and carbon dioxide, it is a suitable membrane for separating combustion gases [37-40].

Test description

First, the effect of temperature in dehydration of natural gas and the amount of water in the retentate phase of the PES membrane in the real temperature range of industry, the temperature of 20-50 ° C, and for better comparison at two constant pressures of 5 and 8 bar, was investigated. Then the effect of pressure in the range of 2 to 10 bar on the amount of water removal from the remaining phase was performed. The amount of dehydration of natural gas in the pressure range of 2 to 10 bar and temperature of 20 to 50 ° C in the presence of nitrogen sweeping gas was performed by using PES membrane and its nanocomposite 2.5 and 10% by weight of silica. With increasing feed pressure, the amount of water permeation from the membrane increased. On the other hand, lowering the temperature increased the rate of dehydration [41-45].

The following principles were considered in performing the tests:

1-The contact between the feed and the sweeping gas was continuous and, in the counter, current.

2-The amount of water in the feed was 910 mg/sm^3.

3-Feed flow was 1000 cm3/min and the ratio of sweeping gas to feed ratio was set to 1.

4-The permeation side pressure was equal to 1 absolute atmosphere

Results and data analysis

Scanning electron microscopy images of PES membranes and PES-silica nanocomposite membranes

According to figure (1), the membrane thickness was reported 47.23 µm.

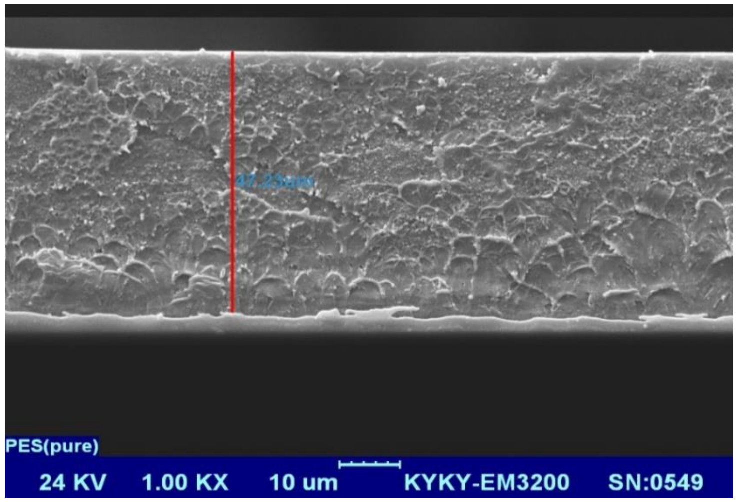

Figure 1. Cross-sectional image of a PES membrane

The results of scanning electron microscopy test show the presence and nanometer distribution of silica particles evenly all over the membrane. The average particle size of silica, is less than 40 nm, and this number is a good indication of the particle size of silica in the nano range.

The size of silica particles, for better observation and investigation of their distribution and nanometer size is presented in the pictures above

Scanning electron microscopy test images show that the cross-sectional roughness increased with inhancing silica content in the membranes. At high percentages of silica, the accumulation of particles in the polymer matrix is increased. In all scanning electron microscope images, the presence and distribution of nanometers of silica particles can be seen, which confirms the optimal dissolution of silica particles in the polymer as well as good compatibility in the two phases [46-48].

Investigation and analysis of Fourier transfer infra-red spectroscopy of PES membrane and its nanocomposites with silica particles

B Infra-red spectroscopy is widely used to identify organic compounds. In order for infra-red radiation to be absorbed by a molecule, the dipole moment of this molecule must undergo a general change as a result of its rotational and vibrational motion. If the frequency of the radiation is equal to a natural vibrational frequency of the molecule, an energy transfer takes place, leading to a change in the vibration amplitude of the molecule, resulting in the absorption of radiation. When co-nuclear species, such as oxygen and nitrogen molecules, vibrate or rotate, no general change in their dipole moment occurs, and therefore these compounds do not absorb infra-red radiation 3].

The infrared spectrum of simple and composite membranes is presented in figures (2) to (3).

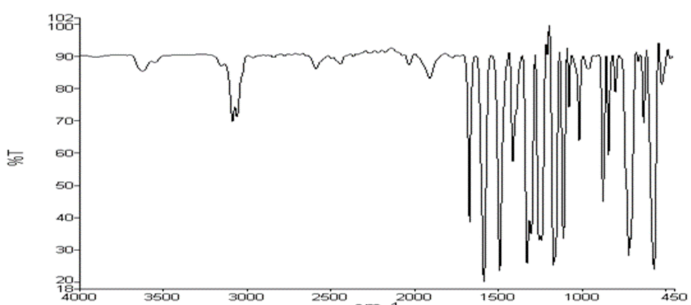

Figure 2. Infra-red spectrum of PES membrane

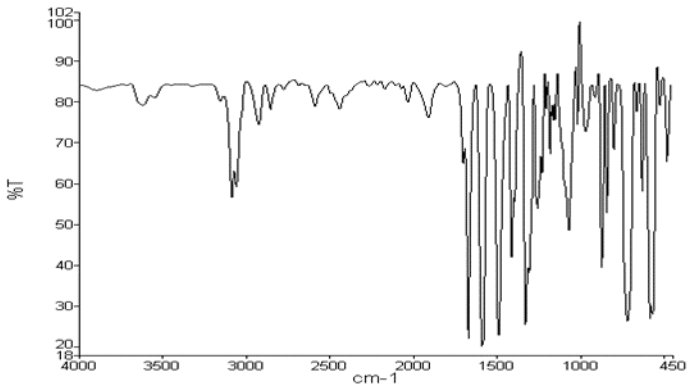

Figure 3. Infra-red spectrum of 10% silica -PES nanocomposite

As it can be seen, the presence of strong absorption peaks in the wavelength range of 1170 cm indicates asymmetric tensile bonds of Si-O-Si silica particle formation. The presence of a broad peak in the wavelength range of 3300 cm-1 and 1150 cm^{-1} indicates the presence of O-H polar groups in silica particles [7].

By examining the infra-red spectrum of silica particles, the symmetric tensile peak of Si-O-Si in the range of 794 cm^{-1} and the tensile peak of Si-OH in the range of 958 cm^{-1} can be seen.

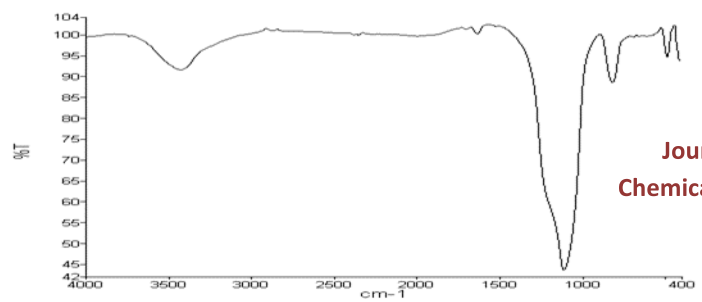

Figure 4. Infra-red spectrum of silica nanoparticles

The effect of temperature on the amount of water removal from natural gas

Changes in the amount of water in the retentate phase at constant pressures of 5 and 8 bar and at normal industrial temperatures in the range of 20 to 50 ° C in the presence of sweeping gas are shown in figure (5).

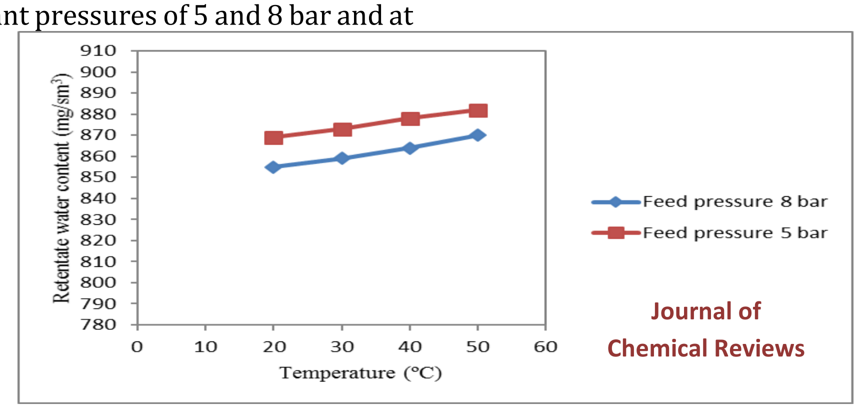

Figure 5. Changes in the amount of water in the retentate phase at pressures 5 and 8 bar relative to temperature changes

The graph of changes in the amount of water in the retentate phase is ascending in terms of temperature changes at constant pressures of 5 and 8 bar, which shows that the permeation of water vapor from the PES membrane has decreased with increasing temperature. This issue can be explained qualitatively based on the dissolution-diffusion model, permeation is defined as the solubility and penetration of component into the membrane. The overall temperature dependence of the permeability of water vapor, which is defined as the activation energy of the permeation, depends on the activation energy of the diffusion and the heat of absorption [5].

$$EP = ED + \Delta HS$$

EP = Permeation Activation Energy
ED = penetration activation energy
ΔHS = heat absorption

As the temperature increases, the diffusion also increases and the penetration activation energy is always positive. Under normal conditions, the heat of absorption for a gas is negative, and thus the gas permeation through a polymer membrane increases with increasing temperature. However, for the emission of condensable gases such as water vapor, the heat of absorption becomes more important and may overcome the heat of activation of penetration and reduce the permeation of water vapor by increasing the temperature. With increasing temperature, the amount of water in the remaining phase has increased and this has meant a decrease in water permeation [4].

At a certain concentration of water in a gaseous mixture, with increasing temperature, the vapor pressure of water saturation increases and therefore its activity decreases thermodynamically. Therefore, the negative effect of temperature on water vapor permeation increases significantly with increasing temperature. As we know, with increasing temperature, the emission of gases such as methane, which are normally incondensable under conditions, increases. But condensable gases such as water vapor, as the experimental results of this study show, their permeation decrease with increasing temperature, therefore, it can be concluded that natural gas dehydration is more beneficial at lower operating temperatures [7].

Respectively, the diagram of the changes in the amount of water in the retentate phase in the nanocomposite membrane 2.5 and 10% by weight of PES-silica at a pressure of 5 and 8 bar are shown in relative to the temperature changes [9].

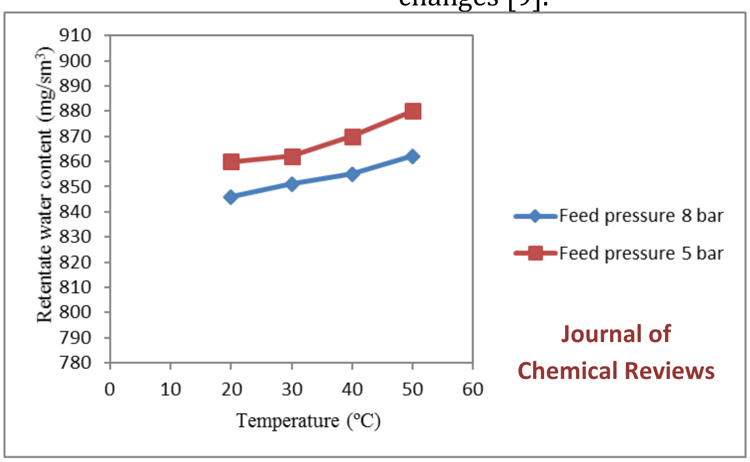

Figure 6. Changes in the amount of water in the retentate phase in the 2.5% PES-silica at pressures of 5 and 8 bar relative to temperature changes

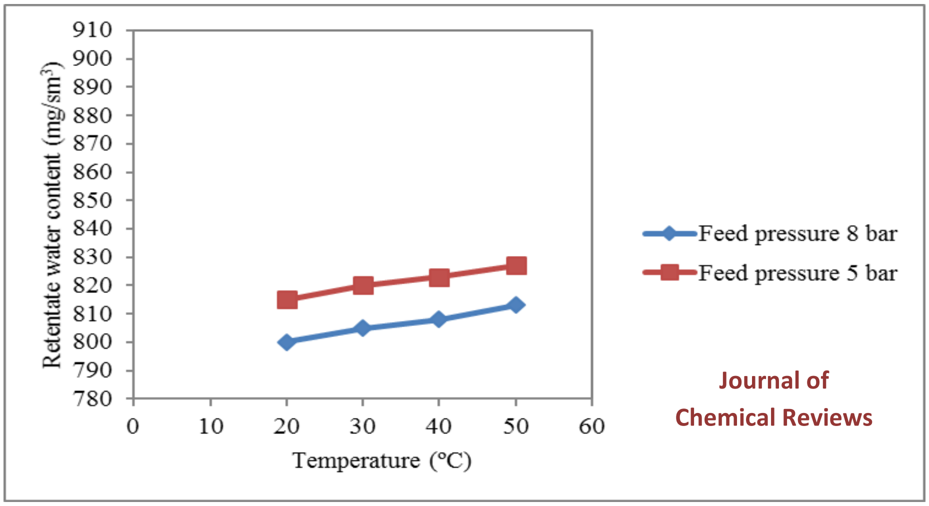

Figure 7. Changes in the amount of water in the retentate phase in the 10% PES-silica at pressures of 5 and 8 bar relative to temperature changes

Examination of diagrams of changes in the amount of water in the retentate phase in terms of temperature changes at constant pressures 5 and 8 bar in nanocomposite membranes 2.5 and 10 % of PES-silica is ascending, which shows the permeation of water vapor in these membranes shrinks with increasing temperature [13].

By Jennifer Ron Hongdu et al. the effects of temperature on dehydration of natural gas in three feed samples with molar fractions of 0.005, 0.015 and 0.025 water by poly composite membrane (dimethyl amino ethyl methacrylate) - polyacrylonitrile in the temperature range of 30 to 55 °C have been studied by experimental tests, the results of which show that with increasing temperature, water permeation from the membrane decreases, which is in consistent with the results of this study [16].

The effect of pressure on the amount of water removal from natural gas

The effect of pressure changes in the amount of water in the retentate phase of PES membrane at 30 and 50 ° C.

Due to the increase in driving force, the difference in the partial pressure of water on both the feed and permeation sides, the permeability of water vapor increases and as a result the amount of water in the remaining phase decreases. The increase in permeability at higher pressures is due to the increased interaction between the water molecules and PES agents. In other words, with increasing pressure, water solubility is expected to increase, condensability will increase and permeability increase too. As seen in Figure 4-18, at feed pressure 2 bar and at 30 °C, 20 mg of feed moisture is reduced, while at constant conditions, the amount of water is reduced only by increasing the feed pressure to 10 bar from the retentate phase is equal to 60 mg, which shows a 3-fold increase [19].

In Figures (8) and (9), respectively, the diagram of changes in the amount of water in the retentate phase in the nanocomposite membrane 2.5 and 10% by weight of PES-silica at 30 And 50 ° C are shown with respect to pressure changes.

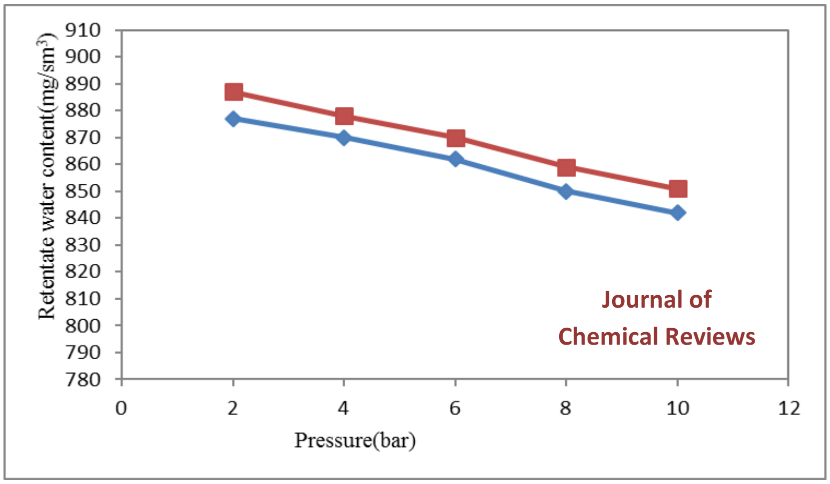

Figure 8. Changes in the amount of water in the retentate phase in the nanocomposite membrane by 2.5 % by weight of PES-silica at 30 °C and 50 °C relative to pressure changes

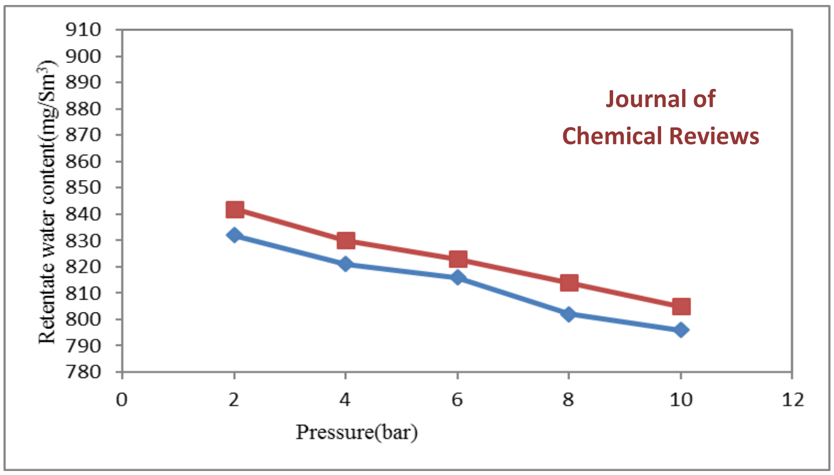

Figure 9. Changes in the amount of water in the retentate phase in the 10% by weight PES-silica membrane at 30 °C and 50 °C with respect to pressure changes

Examination of diagrams of changes in the amount of water in the retentate phase at 30 °C and 50 °C relative to pressure changes in nanocomposite membranes of 2.5 and 10% by weight of PES-silica is decreasing, which shows the permeation of water vapor in these membranes has increased. The increase in permeability at higher pressures in PES-silica is due to the increased interaction between water molecules and polyether sulfone agents.

The effect of adding silica nanoparticles to PES membrane in dehydration process of natural gas

In figure (10) comparison of the amount of water in the retentate phase in PES-silica membranes at a pressure of 8 bar and different temperatures by changing the weight percentage of silica nanoparticles is shown. In figure (11) changes in the amount of water in the retentate phase in PES-silica membranes at temperature of 30 °C and different pressures by changing the weight percentage of silica nanoparticles are presented [24].

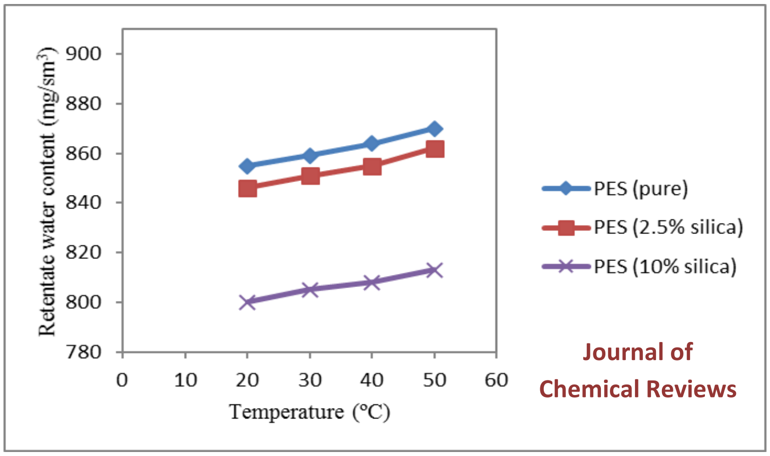

Figure 10. changes in the amount of water in the retentate phase in PES-silica membranes at a pressure of 8 bar and different temperatures by changing the weight percentage of silica nanoparticles

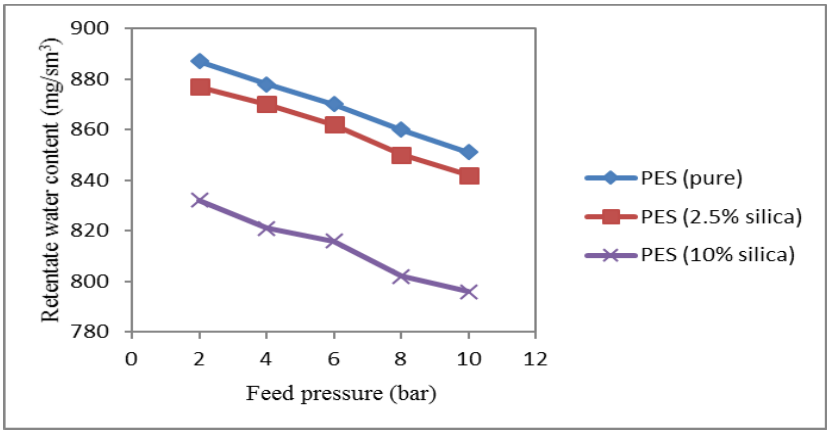

Figure 11. Changes in the amount of water in the retentate phase in PES-silica membranes at a temperature of 30 °C and different pressures by changing the weight percentage of silica nanoparticles

According to figures (10) and (11), it is clear that with increasing the percentage of silica nanoparticles in the polyether sulfone membrane, the amount of water in the retentate phase decreases and this indicates an increase in the permeability of water molecules from PES-silica membrane with increasing percentage of silica. For example, in figure (11) at a temperature of 30 °C and a pressure of 8 bar, the pure PES membrane reduced 50 mg of feed moisture while the 10% PES-silica reduced 110 mg in the same conditions, and this means a more than 2-fold increase in the dehydration rate of the feed with 10% PES-silica nanocomposite compared to the pure PES membrane [29].

In 2008, Sadeghi et al. investigated the gas penetration properties of ethylene vinyl acetate and silica nanocomposite membranes. They investigated the gas permeability of nanocomposite membranes with contents of 5, 6, 10% by weight of silica for N_2, O_2, CO_2 and CH_4 gases at pressures of 4, 6 and 8 bar. The results showed that the increase of silica nanoparticles will significantly increase the permeability of all gases and will increase the selectivity of CO2 to N_2 and CH_4.

Iron et al. made a poly sulfone matrix membrane mixed with silica nanoparticle filler. The addition of silica nanoparticles significantly increased the permeability, and this increase in permeability was directly related to the amount of silica content. In addition, they found that as silica nanoparticles increased, the permeability of large gases would increase, resulting in an increase in voids. Increasing the empty space drastically increases the diffusion and permeability and thus reduces the selectivity [9].

Conclusion

1-The results of infrared spectra (FT-IR) in nanocomposite membranes revealed that the Si-O-Si peak is tensile and close to 1170 cm^{-1} and the Si-OH peak is in the range of 3100-3700 cm^{-1}. These spectra are related to silica nanoparticles added to PES.

2-The results of the SEM analysis demonstrated the presence of nanometers of silica particles evenly across the polymer volume.

3-The results of increasing the removal of water in the retentate phase by increasing the weight percentage of silica nanoparticles in PES-silica membranes are due to the reduction of polymer compaction and shortening of the gas penetration path.

4-The increase in water vapor permeability at higher pressures is due to the increased interaction between water molecules and PES agents. In other words, water solubility and permeability are expected to increase.

5-The process of removing water from natural gas by PES membrane and its nanocomposites was decreasing with increasing temperature at 5 and 8 bar pressures. The effect of temperature on permeability and solubility is the opposite. As the temperature increases, the permeability increases too, but the solubility decreases. Therefore, the effect of temperature on membrane permeability is a combination of these effects. In most cases, the effect of temperature on the permeability is dominant and with increasing temperature, the permeability increases. However, in the case of water vapor, the effect of temperature on solubility was dominant and with increasing temperature, its permeability decreased.

6-One reason for increasing water permeability by increasing the percentage of nanoparticles as a result of poor compatibility of silica and PES surface is that polymer chains cannot be firmly bonded with silica nanoparticles and this causes narrow cracks or seams around the nanoparticles. As a result, the gas penetration path is shortened and the penetration coefficient and permeability increase.

Conflict of interest

The authors declare no conflict of interest.

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