



Cellulose nanocrystals/polydimethylsiloxane hybrid membranes for air dehydration at elevated temperatures

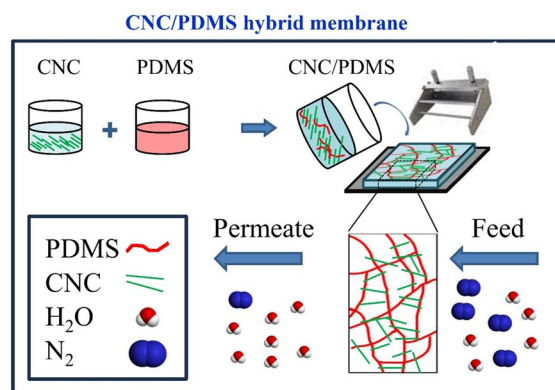
Nasim Alikhani · Ling Li · Jinwu Wang

Received: 10 July 2024 / Accepted: 25 July 2025 / Published online: 20 August 2025
© The Author(s), under exclusive licence to Springer Nature B.V. 2025

Abstract This study presents a cellulose nanocrystals (CNCs)/polydimethylsiloxane (PDMS) composite membrane for enhanced water vapor separation at elevated temperatures. CNCs/PDMS membranes were fabricated via a casting method and characterized for their permeability, selectivity, and thermal stability. Water vapor permeability was measured using a Payne diffusion cell coupled with a Dynamic Vapor Sorption instrument, while nitrogen gas permeability was determined with a gas permeation cell. The results indicate that incorporating 2% CNCs increased water vapor permeability by 24.8%, 30.9%, and 11.2% at 25 °C, 50 °C, and 80 °C, respectively, with a slight improvement in selectivity (up to 3.1%). However, increasing CNC concentration beyond 2% led to slight reductions in permeability, attributed to nanoparticle aggregation. The thermal dimensional stability of the optimized membranes improved, as evidenced by an 8.9% reduction in the coefficient of thermal expansion. These findings suggest that

CNC-reinforced PDMS membranes could be promising candidates for energy-efficient air dehydration applications, though further studies are needed to optimize CNC dispersion and long-term performance under industrial conditions.

Graphical abstract



Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s10570-025-06707-4>.

N. Alikhani · L. Li (✉)
School of Forest Resources, University of Maine, Orono,
ME 04469-5755, USA
e-mail: ling.li@maine.edu

J. Wang
USDA Forest Service, Forest Products Laboratory
Madison, Madison, WI 53726, USA

Keywords CNCs · Membrane · Moisture Separation · PDMS · Permeability · Temperature

Introduction

In recent years there has been increased interest in air dehumidification due to its potential to improve the efficiency of many industries. Examples include

food production, chemical synthesis, pharmaceuticals manufacturing, wood lumber drying and more (Qu et al. 2018; Thiel and Atkinson 2021). The three main air dehumidification technologies are condensing-based systems, desiccant-based systems, and isothermal membrane-based air dehumidification (IMAD). Advantages and limitations of these methods are summarized in Table 1.

In an isothermal membrane-based air dehumidification (IMAD) process, water vapor is separated from the air through a dense membrane without any phase change or temperature change (Zhao et al. 2015). IMAD offers superior energy performance by avoiding the cooling/heating cycles (required in condensing methods) and the desiccant regeneration step (seen in absorption-based systems). IMAD achieves moisture removal efficiency exceeding 50% under certain initial parameters such as temperature and moisture content. Performance studies have shown that IMAD saves up to 42.9% energy compared to traditional systems. Additionally, IMAD reduces dependency on external conditions, lowers operational costs, and offers versatile applications in various industries, including food processing, gas separation, and air drying (Metz et al. 2005; Bui et al. 2015).

Many polymer-based membrane materials may be used to make a dense membrane for air dehumidification in an IMAD process (Bergmair et al. 2012). A membrane functions as a discerning transport barrier separating two contiguous phases. It facilitates the traversal of certain molecules (the permeate) while excluding others (the retentate) through a driving force, such as pressure or concentration gradient (Ger et al. 2007). As the partial pressure difference of the

crucial component between the feed and permeate sides of the membrane increases, the driving force for permeation correspondingly increases (Baker 2012). For air dehydration using polymeric membranes, a dense polymeric membrane is required (Baker 2012). In dense membranes, gas permeation follows the solution-diffusion mechanism, where permeability depends on diffusivity and solubility.

Polymeric membranes

The efficiency of gas separation technology relies heavily on the choice of membrane materials (Metz 2003). Dense polymeric membranes are widely used in gas separation due to their cost-effectiveness and low operating expenses, despite challenges like the “polymer upper bound limit”. This is a trade-off between permeability and selectivity where selectivity increases as permeability decreases (and vice versa) (Rezakazemi et al. 2012).

In the past, commercial polymeric membranes were fabricated using rubbery and glassy polymers such as polyamides, polyetherimides, polypropylene, polyvinyl chloride, and polyvinyl fluoride. However, traditional polymeric membranes have a number of limitations including inadequate resistance to contaminants, low chemical stability, and limited thermal stability (Robeson 2008).

Rubbery polymers, such as polydimethylsiloxane (PDMS), operate above their glass transition temperature and achieve selectivity through solubility differences. Glassy membranes, like polysulfone and polyimide, function below their glass transition temperature, relying on excess free volume for gas

Table 1 Technologies used for air dehumidification (Metz et al. 2005; Bui et al. 2015)

Method	Working principle	Energy efficiency	Limitations
Condensing-based	Cools humid air to its dew point along the saturation curve, condensing moisture	Poor; energy-intensive due to overcooling and reheating	Overcooling is inefficient; dependent on cooling and reheating cycles
Desiccant Absorption-based	Uses solid or liquid desiccants to absorb moisture.	Moderate; energy is required for regeneration and cooling	Limited capacity. Requires additional cooling to control heat generated by absorption
Isothermal Membrane-based (IMAD)	Uses dense polymer membranes to selectively separate water vapor from humid air, driven by a chemical potential gradient	High; eliminates cooling, heating, and regeneration energy needs	Requires advanced materials with high selectivity and permeability; operational optimization needed

separation via diffusivity. While rubbery membranes offer better resilience and simpler synthesis, glassy membranes provide superior diffusivity-based selectivity. However, glassy membranes suffer from high material costs and reduced chemical stability, making both types of membranes valuable depending on the application.

Polydimethylsiloxane (PDMS) stands out as one of the most gas-permeable rubbery polymers. This is due to its flexible siloxane linkages and satisfactory $\text{H}_2\text{O}/\text{N}_2$ selectivity, making it a promising candidate for air dehydration (Metz 2003; Fang et al. 2014; Liang and Chung 2018).

Air dehydration with membranes

In previous studies (Metz et al. 2005; Sijbesma et al. 2008), the water vapor and nitrogen permeability of various polymers were compiled. The data in Fig. 1 represent values of water vapor permeability and $\text{H}_2\text{O}/\text{N}_2$ selectivity. Completely non-polar membranes (such as those based on natural rubber, polystyrene, polypropylene, and polyethylene) tend to have poorer permeabilities. Poly (vinyl alcohol), or PVA exhibits low permeability despite its prevalence in commercial membranes (Bolto et al. 2011). The presence of polarity, such as ether or sulphonate groups,

is considered advantageous in membrane materials. Introducing sulphonate or carboxylate groups into the structure of PVA appears to be a worthwhile modification (Sijbesma et al. 2008).

Many polymer-based membrane materials, such as PDMS, Polyether-block-amide (PEBAX), and Sulfonated poly (ether ether ketone) (SPEEK), can be used to make a dense membrane for air dehumidification (Bergmair et al. 2012). As a result, these membranes are used in certain air conditioning units to enhance the energy-saving capabilities of building systems (Blume et al. 1991; Bergmair et al. 2012; Jia et al. 2022).

Among these materials, PDMS dense membrane is the most common commercially available membrane for air dehumidification (Alikhani et al. 2020). PDMS offers numerous advantages including low cost, chemical stability, nontoxicity, ease of scale-up, good processability, and a high surface area-to-volume ratio (Liang and Chung 2018; Alikhani et al. 2020). In particular, PDMS membranes in hollow-fiber form offer a high surface area-to-volume ratio and scalability. Moreover, PDMS is acknowledged as one of the most permeable rubbery polymers, due to its flexible siloxane linkages, resulting in exceptional water vapor permeability. Also, PDMS exhibits a satisfactory $\text{H}_2\text{O}/\text{N}_2$ selectivity, typically around 140 (Metz 2003; Liang and Chung 2018; Alikhani et al. 2020).

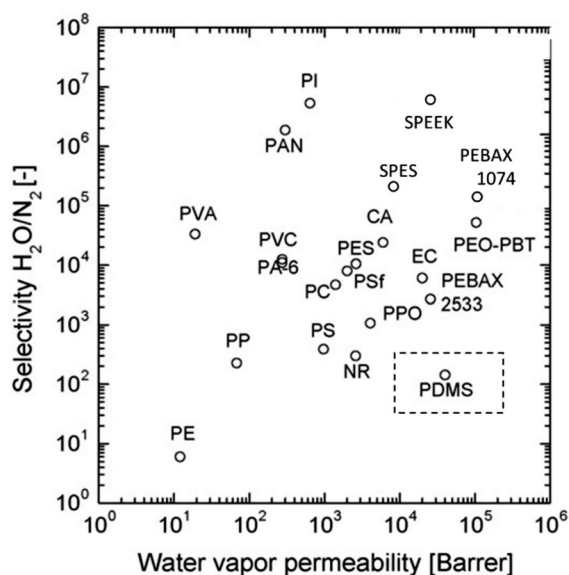


Fig. 1 Water vapor permeability and selectivities of different polymeric membranes. Adapted from (Sijbesma et al. 2008)

Modification of polymeric dense membrane using nanoparticles

Most polymeric membranes, including PDMS, are recommended for use with temperatures less than 50 °C because at higher temperatures there is a reduced gas separation efficiency. However, in most drying processes, the air temperature reaches up to about 82 °C (Simpson 1991). Similarly, in other industries, the gas streams often exhibit high temperatures that exceed the tolerance of the membranes used. Therefore, in the study of flue gas dehydration using PEBAX®1074 and sulfonated SPEEK membrane materials, field tests were performed after the flue gas was cooled below 50 °C (Sijbesma et al. 2008). In another study, feed gas streams were cooled solely to accommodate a membrane gas separation process and subsequently heated back up (Rezaka-zemi et al. 2018). However, cooling and reheating gases introduces additional costs and energy

consumption in the process (Rezazazemi et al. 2018; Jahan et al. 2018).

The goal of this study is to improve these aspects of membranes: (1) Their thermal and mechanical properties, and (2) their vapor/gas separation performance. One promising approach is to develop mixed matrix membranes that combine the favorable characteristics of both polymeric and nanomaterials (Moaddeb and Koros 1997; Sohn et al. 2000; Jahan et al. 2018). Various nanoparticles are employed to enhance the performance of polymeric membranes. Examples include ZnO (Ammar et al. 2016), TiO₂ (Vaimakis-Tsogkas et al. 2019), SiO₂ (Beltran et al. 2011), titanate nanotubes (TNTs) (Li et al. 2010), Zeolite (Mao et al. 2019; Şahin et al. 2020), and others. However, it is important to note that some of these nanoparticles may have non-sustainable petroleum-based origins.

Cellulose nanocrystals (CNCs) are a class of nanomaterials with dimensions of 100–300 nm length and 5–70 nm diameter derived from sustainable and renewable lignocellulosic biomass, mitigating some of the environmental problems related to other inorganic nanoparticles. They have many attractive features such as large specific surface area, high tensile strength and stiffness, abundance of surface hydroxy groups, and extremely low coefficient of thermal expansion (CTE) (Nechyporchuk et al. 2016; Cruz-Tato et al. 2017; Saba and Jawaid 2018; Gomri et al. 2022). As a result, CNCs have been used to substitute for inorganic nanoparticles (such as TiO₂, Al₂O₃) in film-type composites as reinforcement fillers to improve the strength, thermal expansion stability, and optical property of the composites (Azizi et al. 2017).

In the solution-diffusion mechanism that governs gas diffusion in the dense polymeric membranes, the gas molecules dissolve into a membrane, diffuse across the membrane's thickness, and then desorb from the other side of the membrane (Alikhani et al. 2020). The current study aimed to develop a high-performance hybrid composite membrane for air dehumidification at elevated temperatures for wood drying applications. CNCs were chosen as an additive to PDMS because of two hypotheses: (1) the surface hydroxy groups of CNCs could increase the moisture adsorption sites in the PDMS membrane, improve the solubility of water vapor on the membrane surface and create a large water vapor concentration difference for fast diffusion; and (2) CNC's low thermal

expansion, could restrain the PDMS thermal expansion at elevated temperatures if CNCs disperse in PDMS uniformly.

To achieve this goal, four objectives of this study were to (1) synthesize CNCs/PDMS membranes using CNC suspension with four concentrations (0%, 2%, 4%, and 6%); (2) measure the water vapor permeability, nitrogen gas permeability, selectivity, and coefficient of thermal expansion of the membrane samples at three different temperatures (25 °C, 50 °C, and 80 °C); (3) characterize the morphology of the membrane samples, and (4) determine the optimal CNC concentration for making a high-performance CNCs/PDMS composite membrane.

Materials and methods

Materials

PDMS prepolymer and curing agent kits (Dow SYLGARDTM 184 Silicone Elastomer kit) were purchased from Dow Inc. (MI, USA). A CNC suspension at 11.8% solids was purchased from the USDA Forest Products Laboratory, distributed by the Process Development Center of the University of Maine (ME, USA). CNCs were subjected to comprehensive characterization in a previous study (Reid et al. 2017). The results of the study revealed that the CNC particles had an average length of 134 ± 52 nm and a width of 7 ± 2 nm. The aspect ratio of the particles was determined to be 19. Additionally, the study reported a sulfate half-ester ($-\text{OSO}_3^-$) content of 335 mmol kg^{-1} for the CNCs.

Methods

Membrane preparation

The PDMS prepolymer and curing agent at the ratio of 10:1 by weight (i.e., 10 g of PDMS prepolymer and 1 g of curing agent) was mixed mechanically for 5 min at room temperature. The solution was degassed for 45 s with a planetary centrifugal bubble free mixer (THINKY ARE-310, Tokyo, Japan). Then the solution was cast on a Teflon plate using a casting knife. The wet film had a nominal thickness of 150 µm. The wet film was cured in an oven at

50 °C for 24 h. The pure PDMS film was used as a control group (i.e., 0% CNCs).

For making the hybrid CNCs/PDMS membranes, 10 g of the PDMS prepolymer and 1 g of the curing agent were mixed mechanically for 5 min at room temperature. An appropriate amount of CNC suspension was added to the solutions to achieve the CNC solid concentrations of 2%, 4%, and 6% in total solids. The mixture was then mechanically mixed for 1 h, degassed for 45 s, and cast on a Teflon plate using a casting knife to obtain a wet membrane with a nominal thickness of 130 and 150 μm for Pure PDMS and CNCs/PDMS films, respectively (Planes et al. 2016; Xia et al. 2018). To remove any remaining air bubbles, the membranes were first kept in a vacuum desiccator with a vacuum pressure of 88 kPa for 1 h at room temperature. Afterward, they were transferred to an oven and cured at 50 °C for 24 h (Planes et al. 2016). A schematic of the membrane fabrication process is presented in Fig. 2.

The thickness of dry film samples was measured using a digital micrometer (Mitutoyo, IL, USA) with an accuracy of 0.001 mm. Multiple measurements were performed to determine the average thickness of the film samples.

Scanning electron microscopic (SEM) analysis

The morphology of the CNCs/PDMS membrane samples was observed using a scanning electron microscope (SEM) (NVision 40, Zeiss, Germany). The sample was placed on a specimen mount using carbon tape and then was coated with conductive silver. The regions of interest of the sample were sputter coated with gold (23 nm) using a Cressington 108 auto sputter coater (Ted Pella Inc., Redding,

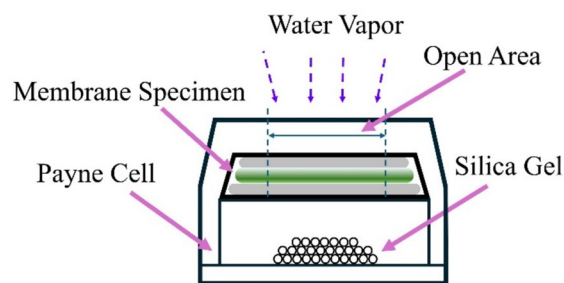


Fig. 3 Schematic of the WVTR test setup on a dynamic vapor sorption instrument (dry cup) indicating a test sample is sealed on the top of the Payne cell

CA, USA). The images were taken at an accelerating voltage of 3 kV.

Atomic force microscopy (AFM) analysis

The PDMS and CNCs/PDMS samples were cut into 5 mm $\times$ 5 mm areas for observation on an AFM (MFP-3D, Oxford Instruments Oxon, UK). Images were collected with an AC200TS-R3 probes with 9 ± 6 (nN/nm) spring constants. Alternating current mode produced images with a set point of (0.4 V) and constant free air amplitude of (1 V).

ATR-FTIR analysis

The ATR-FTIR analysis was performed using a PerkinElmer Spectrum Two™ FTIR spectrometer (Shelton, CT, USA) to evaluate the nature of the interaction between the CNCs and PDMS in the membranes. Each sample was measured at a resolution of 4 cm^{-1} . The data were obtained under an accumulation of 16 scans in the range of 450–4000 cm^{-1} .

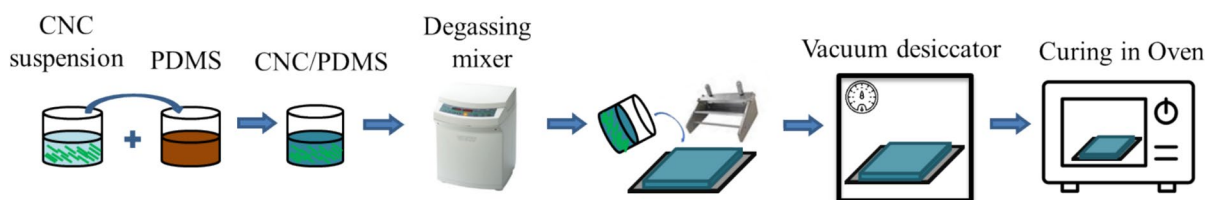


Fig. 2 Schematic of membrane preparation process

Measurement of water vapor permeability of membranes and diffusion coefficients

The water vapor transmission rate (WVTR) was measured using a dynamic vapor sorption (DVS) instrument (Model: DVS Advance; Surface Measurement Systems, London, UK) as shown in Fig. 3. The sample was placed to seal a Payne cell. Silicone gel beads were placed in the cell to absorb water vapor that entered the cell to maintain a 0% RH in the headspace. The surrounding environment of the Payne cell was controlled by DVS to desired testing conditions. For each sample type, nine (9) replicates cut from three same membranes were tested. The thickness of each sample was measured using a digital micrometer. Each sample was tested at 25 °C, 50 °C, and 80 °C and a constant RH of 60%. The increase in mass of the Payne cell assembly with time was recorded simultaneously. The tests followed the ASTM E96 Standard test method for water vapor transmission of materials (ASTM 2016). The H_2O permeability (P_{H_2O}) was calculated using Eq. (1):

$$P_{H_2O} = \frac{WVTR \times T_{mem}}{\Delta_{Vaporpressure}} = \frac{(\Delta m / \Delta t) \times T_m}{A \times \Delta p \times 18} \left(\frac{mol}{m \cdot s \cdot Pa} \right) \quad (1)$$

where, $\Delta m / \Delta t$ is the slope of a linear section of the mass versus time, g/s; T_m is the thickness of the sample membrane, m; A is the open area of the Payne cell, $1.18 \times 10^{-4} \text{ m}^2$; Δp is the partial pressure difference of water vapor, $\Delta p = \frac{\Delta RH}{100} \times \text{Saturated water pressure}$ at 25 °C, 50 °C, and 80 °C, Pa (Saturated water vapor pressure at 25 °C = 3171 Pa, 50 °C = 12,351 Pa, 80 °C = 47,415 Pa); 18 g/mol is the molecular weight of H_2O .

For measuring the diffusion coefficient, rectangular specimens (15 mm × 20 mm) were cut from the films and pre-dried in an oven at 60 °C for 24 h prior to testing. The relative humidity (RH) was incrementally increased from 0 to 60% in 10% steps at a temperature of 25 °C. The membrane samples were considered to have reached equilibrium when the mass variation was less than 0.0009% per minute. The water vapor diffusion coefficient was determined using DVS software based on Eq. (2):

$$\frac{M_t}{M_\infty} = \frac{4}{d} \sqrt{\frac{Dt}{\pi}} \quad (2)$$

where M_t represents the amount adsorbed at time t , M_∞ is the amount adsorbed at thermodynamic equilibrium, D denotes the diffusion coefficient, and d is the film thickness. This equation is applicable for $M_t / M_\infty < 0.4$.

Measurement of nitrogen gas permeability of membranes

The pure nitrogen gas transmission rate (NGTR) was measured using a single gas transmission cell (Custom Scientific Instruments Inc, PA, USA) as shown in Fig. 4, following the ASTM D1423-82(2015) Standard test method for determining gas permeability characteristics of plastic film and sheeting (ASTM 2015). Samples were cut into circles with a 0.12 m diameter. Thickness was recorded. One sample was mounted to separate the cell into the upper and lower chambers. Compressed nitrogen gas flowed into the lower chamber, diffused through the sample, and entered the upper chamber. The volume expansion of the upper chamber was measured by recording the movement of a liquid color slug in the capillary tube. Nine (9) replicates for each sample type were tested. The transmission cell was placed in a water bath to maintain the desired temperatures of 25 °C, 50 °C, and 80 °C. The pressure difference between the lower chamber and upper chamber was measured by a pressure gauge. The N_2 gas permeability was calculated using Eq. (3):

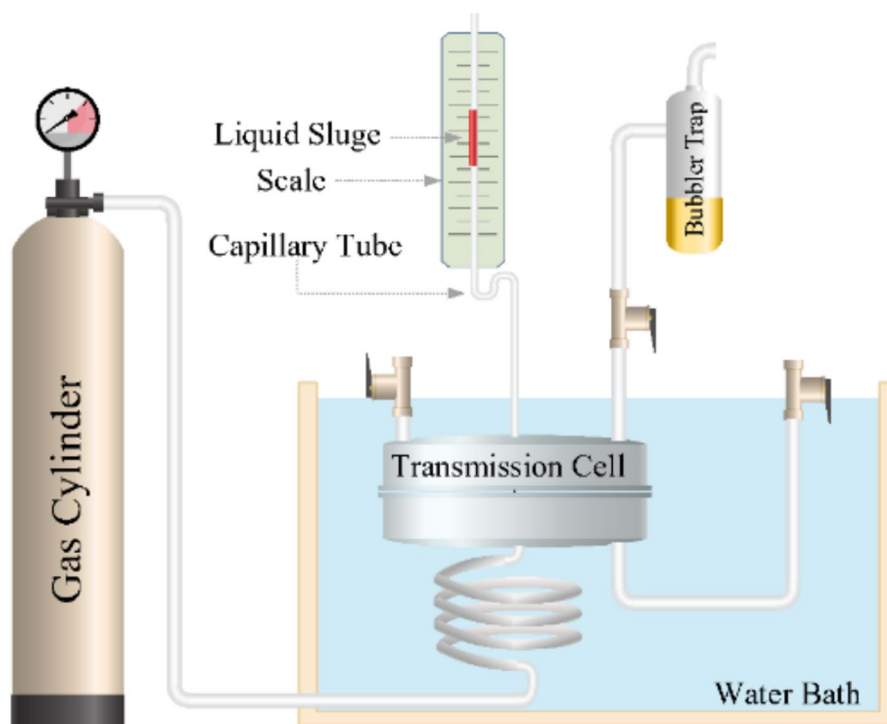
$$P_{N_2} = \frac{NGTR \times T_m}{\Delta p} = \frac{P_0 \times S \times \pi D^2 \times T_m}{\Delta p \times 4ART} \left(\frac{mol}{m \cdot s \cdot Pa} \right) \quad (3)$$

where P_0 is the ambient pressure, 101,325 Pa, S is the rate of rising of the capillary slug (m/s); D is the capillary tube's inner diameter, $0.4957 \times 10^{-3} \text{ m}$; A is the open area of the cell, $0.6655 \times 10^{-2} \text{ m}^2$; T_m is the thickness of the sample membrane, m; Δp is the pressure difference between the lower and upper chamber, i.e., gauge pressure, Pa; T is the temperature of gas, K; R is the gas constant ($8.3143 \text{ m}^3 \cdot \text{Pa} / (\text{mol} \cdot \text{K})$).

Calculation of selectivity of water vapor and nitrogen

The selectivity (α) was obtained from the ratio of H_2O to N_2 permeability, Eq. (4).

Fig. 4 Schematic of a single gas transmission cell. Adapted from (Alikhani et al. 2025)



$$\alpha = \frac{P_{H_2O}}{P_{N_2}} \quad (4)$$

Measurement of coefficient of thermal expansion (CTE)

The in-plane coefficient of thermal expansion (CTE) of the PDMS and CNCs/PDMS membrane samples were measured using a thermomechanical instrument (Q400 TMA, TA Instrument). Samples were cut into 5 mm × 5 mm areas and one sample at a time was sandwiched between two crystal wafers to ensure force dispersion. An expansion probe rested on the surface of the samples with a 0.02 N preload. As the temperature was raised from 25 to 100 °C at a heating rate of 5 °C/min, the change in thickness of the sample was recorded, allowing the calculation of CTE from the slope of the resulting expansion temperature plots. CTE is calculated using Eq. (5) (Saba and Jawaid 2018).

$$CTE = \frac{\Delta L}{L_0 \times \Delta t} \quad (5)$$

where L_0 is the initial length of the sample (μm); Δt is the temperature change (°C), and ΔL is the thermal expansion (or contraction) of the sample after the temperature change.

For each concentration, 3 samples were prepared and tested, where performed in triplicate. Therefore, the average of Nine (9) replicates for each sample type were measured and reported.

Statistical analysis

A two-way analysis of variance (ANOVA) was performed for two independent variables (CNC concentration and temperature) to understand their main effects and their interaction effects on the water vapor and nitrogen gas permeability, and selectivity of the membrane samples. One-way ANOVA was conducted for the effect of the independent variable (CNC concentration) on CTE. All statistical analyses were run in OriginPro software version 2022, with a level of significance of 0.05 (OriginLab Corporation 2022).

Results

Appearance and morphology of CNCs/PDMS membrane samples

Four different membrane samples are shown in Fig. 5. The Pure PDMS has the highest transparency, and the transparency of the film decreases with increasing CNC concentration. The SEM images (Fig. 6) show the top surface morphology of four membrane

samples. The dispersion of CNCs in PDMS depends on the concentration of CNC added. CNC particles are relatively small and distributed randomly in PDMS when 2% of CNCs was added. In contrast, when the concentration of CNC was increased to 4% and 6%, the CNCs aggregation became more significant. In addition, these SEM images show the PDMS and CNCs/PDMS membranes are dense membranes, and no pores or defects are observed at this level of magnification. AFM images (Fig. 7) were used to

Fig. 5 Appearance of the membrane samples

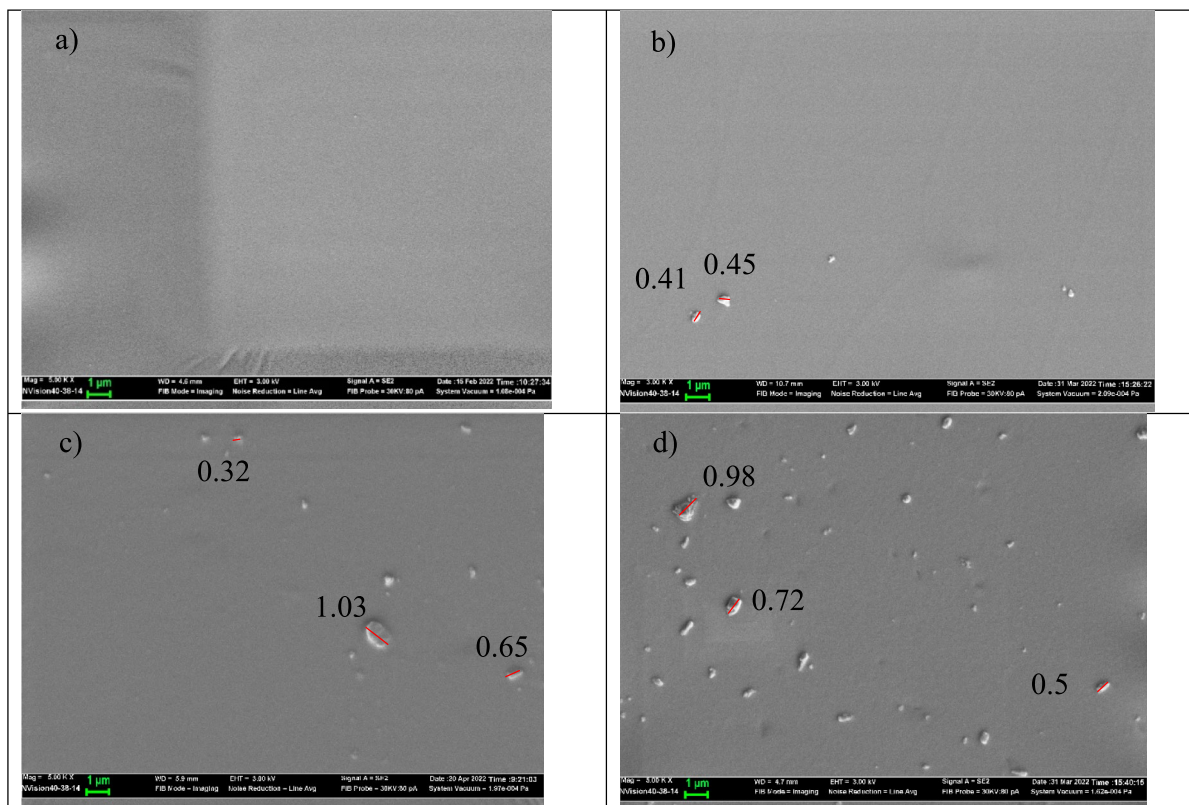
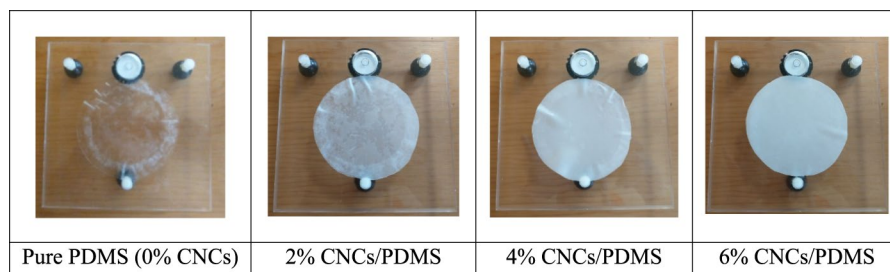


Fig. 6 SEM images of hybrid membrane materials of CNCs/PDMS: **a** 0%, **b** 2%, **c** 4%, and **d** 6%. CNCs appear as various-sized particles

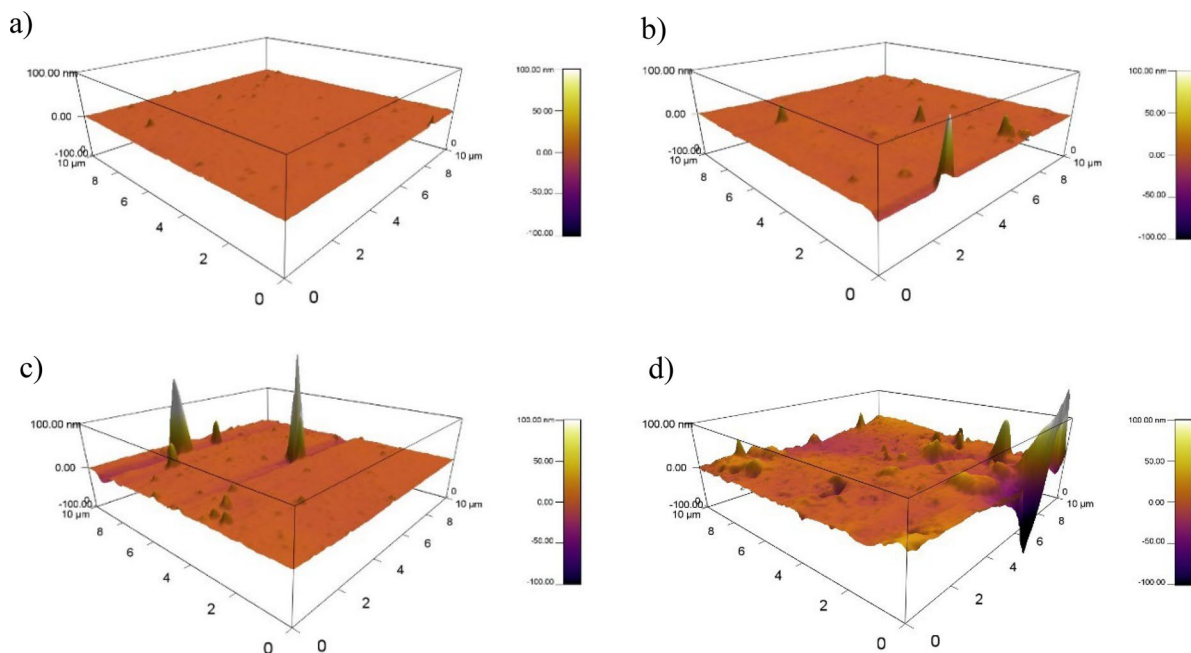


Fig. 7 3D images of AFM results of hybrid membrane materials **a** Pure PDMS, **b** 2% CNCs/PDMS, **c** 4% CNCs/PDMS, and **d** 6% CNCs/PDMS

estimate the 3D shape and dimensions of CNC nanoparticles agglomerations. Most of the CNC particles have a height in the range of 10 nm to 100 nm. The maximum height of agglomerated CNC particles could reach over 100 nm. More high peaks are observed with the increase in CNC concentration.

The CNCs/PDMS membranes in this study were synthesized using varying CNC concentrations of 0%, 2%, 4%, and 6%. Higher CNC concentrations (8% and 10%) were also explored; However, the hydrophilicity of CNCs and the hydrophobicity of PDMS led to agglomeration. As a result, 6% was determined to be the highest concentration at which a uniform and continuous membrane sheet could be successfully synthesized.

ATR—FTIR analysis

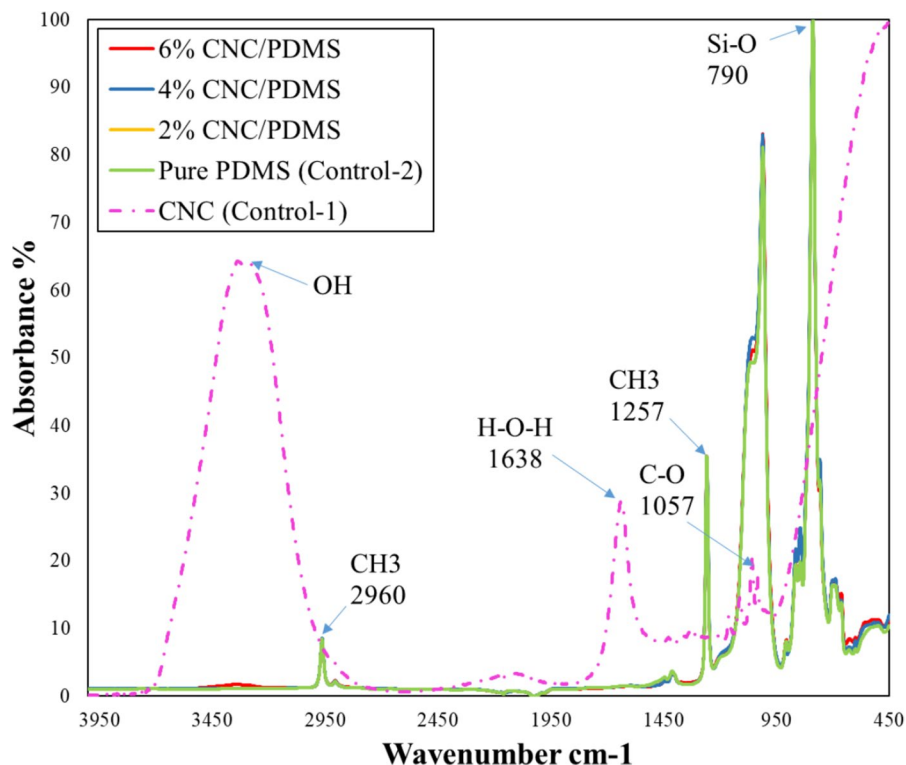
Figure 8 shows the FTIR spectra of an air-dried CNC film (Control-1), pure PDMS sample (Control-2), and CNCs/PDMS samples with CNC concentrations of 2%, 4%, and 6%. For Control-1 CNC film, a strong O–H stretching vibration at 3332 cm^{-1} and bending band of H–O–H bond at 1638 cm^{-1} are observed (Jayaramudu et al. 2018; Inácio and Souza 2020).

For Control-2 PDMS membrane, the featured peaks of PDMS include 2960 cm^{-1} for CH_3 group vibration, 1257 cm^{-1} for the symmetric deformation of the CH_3 group, 887 cm^{-1} for Si–C vibration, and 790 cm^{-1} for Si–O vibration (Cui et al. 2018). When low concentrations of CNCs were blended in PDMS, the spectra of CNCs/PDMS samples show that peaks at 3332 cm^{-1} and 1638 cm^{-1} almost disappeared but the peak at 1057 cm^{-1} representing the stretching vibrations of C–O in CNCs became slightly distinctive (Jayaramudu et al. 2018). The results reveal that the addition of CNCs from 2% to 6% did not change the general spectra pattern of CNCs/PDMS samples. Therefore, the performance of CNCs/PDMS membrane is mainly governed by the PDMS.

Permeability of water vapor and diffusion coefficient

The values of water vapor permeability (Fig. 9) increased with the CNC concentration from 0% (pure PDMS) to 2% and then decreased. The mean $\pm$ SD values of water vapor permeability at 25°C for Pure PDMS, 2% CNCs/PDMS, 4% CNCs/PDMS, and 6% CNCs/PDMS were $30,683.6 \pm 615.9$, $38,295.6 \pm 1070.9$, $35,883.6 \pm 1,248.8$, and

Fig. 8 FTIR spectra of all membrane samples and control groups



$35,846.1 \pm 1338.3$ Barrer, respectively. The addition of 2% CNC resulted in an increase of 24.8%, 30.9%, and 11.2% at 25 °C, 50 °C, and 80 °C, respectively. Higher concentrations of CNC (4% and 6%) show a slightly decreasing trend for water vapor permeability, while the permeability values remained higher

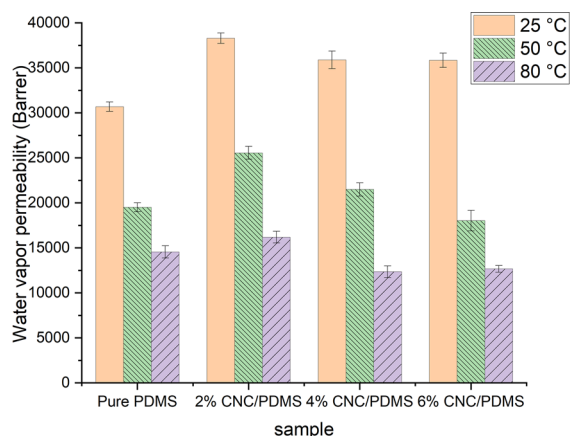


Fig. 9 Water vapor permeability at elevated temperatures and humidity differential of 60%

than those for pure PDMS membranes. This trend is the same in all three temperatures except for 80 °C. When the temperature increased from 25 °C to 80 °C, the permeability of water vapor decreased dramatically. The rationale behind this phenomenon is elaborated in the discussion. Figure 9 illustrates that increasing temperature reduces the water vapor permeability of PDMS; however, the results indicate that the addition of CNC nanofillers helps mitigate these negative effects.

Two-way ANOVA results reveal that the effects of CNC concentration, temperature and their interaction on water permeability were statistically significant (Table S.1 Supplementary document). Furthermore, the comparison between different CNC concentrations shows that the 2% CNCs/PDMS is significantly different from other samples, but there is not any significant difference between pure PDMS (0% CNCs), 4% CNCs/PDMS, and 6% CNCs/PDMS samples (Table S.2). Therefore, 2% CNCs is the optimal concentration in terms of water vapor permeability.

In addition, the interaction between CNC concentration and temperature (Table S.1) is statistically significant ($p < 0.05$), indicating that the effect of CNC

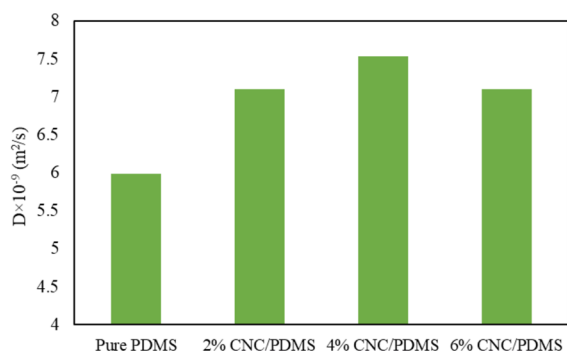


Fig. 10 Diffusion coefficients of the PDMS and CNCs/PDMS membranes, 25 °C and RH 60%

concentration on water vapor permeability varies depending on the temperature.

The diffusion coefficients for PDMS and CNCs/PDMS membranes at 25 °C and RH=60%, are presented in Fig. 10. The diffusion coefficient for PDMS is $5.99 \times 10^{-9} \text{ m}^2/\text{s}$, which aligns well with previous studies (Favre et al. 1994; Watson and Baron 1996). The results indicate an increasing trend in diffusion coefficient with the addition of 2% and 4% CNCs, suggesting improved diffusion properties. However, at 6% CNCs, a decreasing trend is observed, which may be attributed to CNC particles agglomeration.

Permeability of nitrogen gas

Nitrogen gas permeability (Fig. 11) increased with CNC concentration from 0% (pure PDMS) to 2% and then slightly decreased. The mean $\pm$ SD values of nitrogen gas permeability at 25 °C for Pure PDMS, 2% CNCs/PDMS, 4% CNCs/PDMS, and 6% CNCs/PDMS were 263.7 ± 4.6 , 324.9 ± 18.7 , 310.0 ± 8.4 , and 310.3 ± 7.3 Barrer, respectively.

The nitrogen gas permeability (Fig. 11) increased by 23.2%, 27.8%, and 16.0% at three temperatures when adding 2% CNCs to the PDMS membrane. Similarly, higher concentrations of CNCs (4% and 6%) show a slightly decreasing trend for nitrogen permeability compared with 2% CNCs. The nitrogen permeability increased as the temperature was elevated from 25 to 80 °C.

Two-way ANOVA results show that the effect of CNC concentration on nitrogen permeability was not statistically significant; however, the effect of temperature on permeability was statistically significant

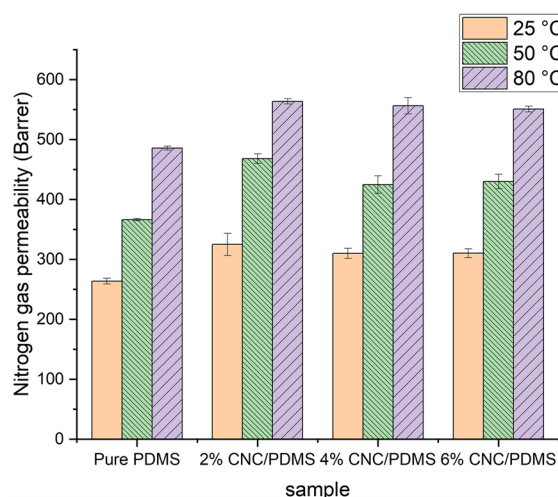


Fig. 11 Nitrogen permeability at elevated temperatures

(Table S.3). The interaction between temperature and concentration was not statistically significant either. Furthermore, the comparison between different CNC concentrations reveals that there was not a statistically significant difference between 2%, 4%, and 6% CNCs/PDMS membranes but all of the CNCs/PDMS membranes were significantly different from the Pure PDMS (Table S.4). Therefore, 2% CNCs is the optimal concentration in terms of nitrogen gas permeability. In addition, the interaction between CNC concentration and temperature (Table S.3) was found to be statistically insignificant ($p=0.46$), indicating that the effect of CNC concentration on nitrogen gas permeability does not vary depending on the temperature.

Selectivity of water vapor and nitrogen

The selectivity of water vapor over nitrogen (Fig. 12) at 25 °C of pure PDMS, 2% CNCs/PDMS, 4% CNCs/PDMS, and 6% CNCs/PDMS were 116.3, 117.9, 115.7, and 115.5, respectively. A slight increase of about 3.1% was observed at 2% CNCs addition, followed by a slight decrease when increasing the concentration of CNCs at three temperatures. Two-way ANOVA results (Table S.5) show that the effect of CNC concentration on selectivity was not statistically significant but the effect of temperature on selectivity was statistically significant. The interaction between temperature and concentration was not statistically significant for selectivity. In addition, the

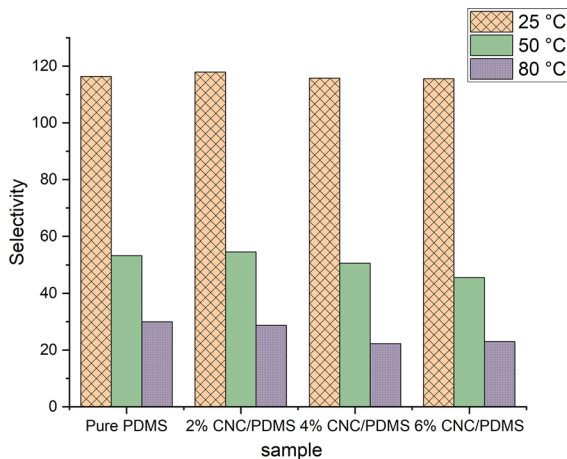


Fig. 12 Selectivity of H₂O/N₂ at elevated temperatures

interaction between CNC concentration and temperature (Table S.5) was found to be statistically insignificant ($p=0.72$), indicating that the effect of CNC concentration on selectivity does not vary depending on the temperature.

Coefficient of thermal expansion (CTE) of membranes

The dimensional stability of CNCs/PDMS membranes influences how well the membrane operates at elevated temperatures. It was evaluated in terms of the coefficients of thermal expansion (CTE) of pure PDMS and CNCs/PDMS membranes (Fig. 13). The mean $\pm$ SD of CTE for pure PDMS, from room temperature up to 100 °C, was $301.1 \pm 12.6 \mu\text{m/m}^\circ\text{C}$. This measured value is in good agreement with the published values for CTE of PDMS which is 309 ppm/°C (Müller et al. 2019). After adding 2% CNCs, the mean $\pm$ SD of CTE was reduced to $274.4 \pm 8.5 \mu\text{m/m}^\circ\text{C}$, which is about 12% lower than that of pure PDMS. This decrease in CTE was expected because CNC has a very low CTE of $9 \mu\text{m/m}^\circ\text{C}$ (Diaz et al. 2013). It helps to restrain the expansion of PDMS at elevated temperatures when CNCs are dispersed in PDMS (Saba and Jawaid 2018). However, the mean $\pm$ SD of CTE of 4% CNCs/PDMS and 6% CNCs/PDMS was 269.5 ± 13.2 and 264.3 ± 8.6 , respectively. Increasing the CNC concentration to 4% and 6% did not further decrease the CTE greatly. One-way ANOVA results (Table S.6) show that the decrease in the CTE between pure PDMS

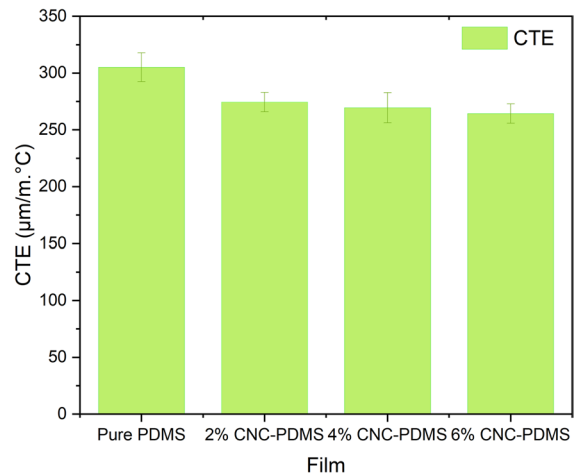


Fig. 13 The CTE values for different samples

sample and 2% CNCs/PDMS sample was statistically significant. Also, the comparison between different CNC concentrations (Table S.7) shows that there is not a significant difference between 2%, 4% and 6% CNCs/PDMS samples and all of these samples are significantly different from Pure PDMS. Once again, 2% CNCs is the optimal concentration in terms of CTE.

Furthermore, in our previous study (Alikhani 2023), the mechanical properties of pure PDMS were compared with those of CNCs/PDMS membranes using Dynamic Mechanical Analysis. The results revealed that incorporating only 2% unmodified CNCs did not significantly enhance the storage modulus of PDMS. This finding aligns with previous studies. Similarly, in a study by Planes et al., the mechanical properties of PDMS films remained largely unchanged upon the addition of 0.5 to 4 w.t.% unmodified CNCs, further supporting the observed trend (Planes et al. 2016).

Discussion

CNC Nanofillers in PDMS: effectiveness and comparison with other nanofillers

In this study, a bio-based material (CNC) was used as a nano filler in the PDMS matrix to improve the PDMS membrane performance for air dehydration applications. To the best of our knowledge, this is the

first report of this usage of CNC. CNC's renewable and sustainable features make it a desirable alternative to non-renewable nanofillers like zeolites (ZIF), silicon dioxide (SiO_2), and titanate nanotubes (TNTs) reported in the previous studies (Li et al. 2010; Beltran et al. 2011; Mao et al. 2019; Şahin et al. 2020). In most of the other studies of mixed matrix membranes, the nano fillers used were not bio-based and not renewable. The CNC used in this study is renewable and can be produced from sustainable sources, two features that make it superior to the other nano particles listed above.

Our study revealed that the water vapor permeability and nitrogen permeability of PDMS membrane at room temperature were 3,683 Barrer and 263 Barrer, respectively, consistent with previous studies (Lee-mann et al. 1996; Koester et al. 2015). In the current study, after adding the optimal concentration of 2% CNCs, the water vapor and nitrogen gas permeability at room temperature were increased by 24.8% and 23.5%, respectively. This improvement may be attributed to an increase in the membrane's free volume resulting from the incorporation of CNCs, a nonporous nanoparticle, as well as potential agglomeration of the CNCs within the PDMS matrix. A similar increase was observed when nanofiller was added to PDMS (Li et al. 2010; Beltran et al. 2011; Şahin et al. 2020). The enlargement of the fractional free volume (FFV) created more diffusion paths for small penetrants and consequently increased the gas permeability of the nanocomposite membranes.

In an earlier study, the addition of approximately 30 wt% of ZIF-L to PDMS led to a significant 8.0% increase in water vapor permeability at 40 °C (Mao et al. 2019). Similarly, Şahin et al. conducted another study where they added 20 wt% of ZIF-71 to PDMS and observed a significant enhancement of around 34% in the permeability of nitrogen gas at 35 °C (Şahin et al. 2020). CNCs have several benefits over ZIF nanoparticles. For instance, they require a significantly lower weight ratio to achieve a similar improvement in water vapor and nitrogen gas permeability.

Beltran et al. reported an increase in the gas permeability of PDMS after adding modified SiO_2 due to an increase in the free volume of the membrane (Beltran et al. 2011). Such an increase in free volume led to an increase in the diffusion coefficient. These findings suggested that PDMS, a rubbery polymer,

may exhibit an increase in free volume upon the incorporation of nonporous fillers of SiO_2 (Beltran et al. 2011). Li et al. found that adding TNTs into the PDMS matrix resulted in a significant enlargement of the FFV (Li et al. 2010).

In our study (Fig. 10), the incorporation of CNCs into the PDMS matrix resulted in an increase in the membrane's diffusion coefficient. This enhancement in diffusivity can be attributed to an increase in free volume, which subsequently contributes to the observed improvement in the permeability of CNCs/PDMS membranes.

In another study (Lovineh et al. 2014), PEBA-nano silica membranes were fabricated using the sol-gel method and compared to neat PEBA membranes. The single-gas CO_2 permeability of the membranes was measured, revealing that the addition of 10 wt% nano-silica filler to the polymeric matrix led to a decrease in CO_2 permeability.

Asghari et al. prepared chitosan/silica mixed matrix membranes for dehydration of ethanol using both experimental and simulation. The permeation of water vapor through the membranes was studied after the addition of silica and as a function of temperature change, ranging from 30 °C to 70 °C. The results showed that the addition of silica nanoparticles to chitosan increased the water vapor permeation flux, attributed to the enhancement of hydrophilic hydroxy groups within the polymeric matrix. In addition, as the feed temperature was raised, the polymer chain mobility increased and led to an increase in permeation flux (Asghari et al. 2017).

In a study by Zhang et al. a mixed matrix membrane was developed by incorporating a Zirconium metal-organic framework (Zr-MOF) into polyetherimide. This gas separation membrane was used for water vapor separation from CH_4 . The addition of this nanofiller introduced a high density of amino groups, which enhanced the membrane's water vapor/ CH_4 selectivity due to strong bonding interactions between the membrane and water molecules (Zhang et al. 2024).

Fernández-Barquín et al. created mixed matrix membranes incorporating small pore zeolites into PTMSP polymer for O_2/N_2 separation. The performance of the membranes was evaluated as the temperature changes from 298 to 333 K. The results show that for pure PTMSP, both O_2 permeability and

selectivity decreases with increasing the temperature (Fernández-Barquín et al. 2016).

In another study, the thermal stability of PVDF membranes were improved by montmorillonite (MMT) incorporation. The membranes operated from 27 to 80 °C for gas separation. The CO₂ absorption flux of the pure PVDF membrane showed a decreasing trend with increasing temperature. However, for the mixed matrix membrane, the reduction of performance with temperature was less pronounced compared to the membrane without filler. A detailed comparison between this study and other mixed matrix membranes is provided in Table 2 (Rezaei-DashtArzhandi et al. 2016).

Effects of temperature on permeability of water vapor and nitrogen gas

Gas transport through dense polymeric membranes is governed by the solution-diffusion mechanism. Therefore, the effects of temperature on both solution and diffusion processes should be considered.

The influence of temperature on solubility has been well addressed in terms of the van't Hoff relationship. Gas solubility correlates with its condensability. For gases that are less condensable (e.g., N₂ and O₂) and often have lower critical temperatures (Table 3), gas solubility increases with an increase in temperature. However, for condensable gases and vapors (e.g., water vapor) solubility decreases with increasing temperature (Rowe et al. 2010; Rezakazemi et al. 2018). The diffusion of gas molecules in a dense membrane is a thermally activated process and increases as the temperature increases, which is well described by the

Table 3 Kinetic diameter and critical temperature of molecules (Metz 2003; Liang and Chung 2018)

Molecule	Kinetic diameter (pm)	Critical temperature (K)
Water vapor	250	647
Nitrogen	364	126
Oxygen	346	154

Table 2 Nano filler/polymer mixed matrix membranes

Mixed matrix membrane (MMM)	Mixture to be separated	MMM properties compared to pure membrane	Conditions	Concentration of nano filler	Reference
CNCs/PDMS	Water vapor/N ₂	24.8% increased water vapor permeability 23.5% increase in nitrogen gas permeability	T = 25 °C	2 wt%	The current study
Modified Silica/PDMS	Nitrogen gas	22.6% Reduction in nitrogen gas permeability	T = 35 °C	10 wt%	Beltran et al. (2011)
ZIF-71/PDMS	Nitrogen gas	33.8% increase in nitrogen gas permeability	T = 35 °C	20 wt%	Şahin et al. (2020)
ZIF-L/PDMS	Water vapor	8% increase in water vapor permeability	T = 40 °C	30 wt%	Mao et al. (2019)
Silica/PEBA	CO ₂	14.9% reduction in CO ₂ permeability	P = 8 bar T = 25 °C	10 wt%	Lovineh et al. (2014)
Silica /Chitosan	Water vapor/ ethanol	29.4% increase in water flux as temperature increases from 30 °C to 70 °C	Temperature: 30–70 °C	10 wt%	Asghari et al. (2017)
Zr-MOF/PEI	Water vapor/ CH ₄	14.28% decrease in water vapor permeability	–	20 wt%	Zhang et al. (2024)
Small pore zeolites / PTMSP	O ₂ /N ₂	31.2% decrease in O ₂ permeability 355.3% increase in selectivity	T = 60 °C	20 wt%	Fernández-Barquín et al. (2016)

Arrhenius equation (Rowe et al. 2010). However, for water vapor permeability, sorption is the dominant factor (Spatafora Salazar et al. 2019), meaning that the magnitude of solubility change is greater than the magnitude of diffusivity change (because of high condensability which correlates with the high critical temperature as shown in Table 2), thereby resulting in a decrease in permeability. This explains why water vapor permeability decreases, as illustrated in Fig. 9, when the temperature increases from 25 to 80 °C.

However, for N₂ and O₂ molecules which are larger in kinetic diameter and less condensable, when the temperature increases, there is an increase in both diffusivity and solubility. This leads to an increase in permeability (Rezazakemi et al. 2018). This phenomenon is evident in the increased nitrogen gas permeability when the temperature rises from 25 to 80 °C, as shown in Fig. 11.

Influence of CNC agglomeration on membrane appearance and properties

When preparing the CNCs/PDMS hybrid membranes, CNC agglomeration occurred due to the poor compatibility of hydrophilic CNCs and hydrophobic PDMS. As the CNC concentration increased, more agglomerated CNC particles were formed, the membrane transparency decreased ascribed to the mismatch of refractive indexes between CNC and PDMS. Combined SEM and AFM images, the CNC particles are present on the membrane surface with needle-like shapes.

The permeability analysis and CTE results show that 2% CNC was the optimal concentration. Increasing the CNC concentration past 2% did not further enhance the membrane performance. The underlying reason is also explained by the poor compatibility. The compatibility of CNCs and PDMS might be improved by modifying the CNCs, such as with the silylation of CNCs (Planes et al. 2016; Xia et al. 2018).

Conclusions

In this study, CNCs/PDMS composite membranes with CNC concentrations of 0%, 2%, 4%, and 6% were successfully fabricated and characterized for their permeability, selectivity, and thermal stability.

The addition of 2% CNCs resulted in the most favorable performance, enhancing water vapor permeability by up to 30.9% while maintaining structural integrity. However, higher CNC concentrations led to particle agglomeration, limiting further improvements. The optimized membranes demonstrated improved thermal stability, as reflected by a lower CTE (8.9% reduction), suggesting their potential for air dehydration applications at elevated temperatures. Despite these promising results, challenges remain in achieving uniform dispersion of CNC and assessing the membrane's long-term durability under industrial conditions. Future work should explore surface modifications of CNCs to enhance compatibility with PDMS and further improve membrane performance.

Acknowledgements This project was supported by the USDA National Institute of Food and Agriculture, McIntire-Stennis (Project number ME0-42205) through the Maine Agricultural & Forest Experiment Station; the U.S. Department of Agriculture's Agricultural Research Service (USDA ARS Agreement No. 58-0204-6-003 & No. 58-0204-9-166); the US Forest Service and US Endowment for Forestry and Communities-P3Nano Advancing Commercialization of Cellulose Nanomaterials (Agreement 21-00166). Any opinions, findings, conclusion or recommendations expressed in this publication are those of the authors and do not necessarily reflect the view of the Department of Agriculture. In addition, the authors would like to thank Dr. Elliot Sanders for assistance with the AFM testing, Dr. Chung Shum for his valuable help in preparing and writing this manuscript, and iLab UMaine for assistance in preparing the SEM images.

Author contributions Nasim Alikhani: Methodology, Experiments, Visualization, Writing original draft. Ling Li: resources, writing—review and editing, and funding acquisition. Jinwu Wang: resources, writing—review and editing, and funding acquisition.

Funding This project was supported by the USDA National Institute of Food and Agriculture, McIntire-Stennis (Project number ME0-42205) through the Maine Agricultural & Forest Experiment Station; the U.S. Department of Agriculture's Agricultural Research Service (USDA ARS Agreement No. 58-0204-6-003 & No. 58-0204-9-166); the US Forest Service and US Endowment for Forestry and Communities-P3Nano Advancing Commercialization of Cellulose Nanomaterials (Agreement 21-00166).

Data availability No datasets were generated or analysed during the current study.

Declarations

Conflict of interest The authors declare no competing interests.

Ethics approval and consent to participate Not applicable. This study did not involve human or animal subjects.

Consent for publication All the images and tables presented in this work are original results. Content adapted from other works has been cited carefully.

References

- Alikhani N, Li L, Wang J et al (2020) Exploration of membrane-based dehumidification system to improve the energy efficiency of kiln drying processes: Part I factors that affect moisture removal efficiency. *Wood Fiber Sci.* <https://doi.org/10.22382/wfs-2020-029>
- Alikhani N, Li L, Wang J (2025) Silylated CNC/PDMS composite membranes with improved selectivity for H₂O/air separation at elevated temperatures. *Ind Eng Chem Res.* <https://doi.org/10.1021/acs.iecr.4c03658>
- Alikhani N (2023) Cellulose nanocrystals/polydimethylsiloxane composite membranes for air dehydration with the aim of energy-use reduction in wood kiln drying
- Ammar S, Ramesh K, Vengadaesvaran B et al (2016) Amelioration of anticorrosion and hydrophobic properties of epoxy/PDMS composite coatings containing nano ZnO particles. *Prog Org Coat* 92:54–65. <https://doi.org/10.1016/J.PORGCOAT.2015.12.007>
- Asghari M, Sheikh M, Afsari M, Dehghani M (2017) Molecular simulation and experimental investigation of temperature effect on chitosan-nanosilica supported mixed matrix membranes for dehydration of ethanol via pervaporation. *J Mol Liq* 246:7–16
- Azizi N, Mohammadi T, Research RB (2017) Comparison of permeability performance of PEBAX-1074/TiO₂, PEBAX-1074/SiO₂ and PEBAX-1074/Al₂O₃ nanocomposite membranes for CO₂/CH₄. *Chem Eng Res*
- Baker RW (2012) *Membrane technology and applications*. Wiley, Hoboken
- Beltran A, Nisola G, Cho E, Lee E (2011) Organosilane modified silica/polydimethylsiloxane mixed matrix membranes for enhanced propylene/nitrogen separation. *Appl Surf Sci.* <https://doi.org/10.1016/j.apsusc.2011.08.061>
- Bergmair D, Metz SJ, De Lange HC, Van Steenhoven AA (2012) Modeling of a water vapor selective membrane unit to increase the energy efficiency of humidity harvesting. *J Phys Conf Ser Inst Phys* p 012161
- Blume I, Schwering P, M.H.V M (1991) Vapour sorption and permeation properties of poly (dimethylsiloxane) films. *J Memb Sci*
- Bolto B, Hoang M, Xie Z (2011) A review of water recovery by vapour permeation through membranes. <https://doi.org/10.1016/j.watres.2011.10.052>
- Bui TD, Chen F, Nida A et al (2015) Experimental and modeling analysis of membrane-based air dehumidification. *Sep Purif Technol* 144:114–122
- Cruz-Tato P, Ortiz-Quiles EO, Vega-Figueroa K et al (2017) Metalized nanocellulose composites as a feasible material for membrane supports: design and applications for water treatment. *Environ Sci Technol* 51:4585–4595. <https://doi.org/10.1021/ACS.EST.6B05955>
- Cui X, Zhu G, Pan Y et al (2018) Polydimethylsiloxane-titania nanocomposite coating: fabrication and corrosion resistance. *Polymer.* <https://doi.org/10.1016/j.polymer.2018.01.063>
- Diaz JA, Wu X, Martini A et al (2013) Thermal expansion of self-organized and shear-oriented cellulose nanocrystal films. *Biomacromol.* <https://doi.org/10.1021/bm400794e>
- Fang M, Wu C, Yang Z, et al (2014) ZIF-8/PDMS mixed matrix membranes for propane/nitrogen mixture separation: experimental result and permeation model validation. <https://doi.org/10.1016/j.memsci.2014.09.040>
- Favre E, Schaetzel P, Nguyen QT et al (1994) Sorption, diffusion and vapor permeation of various penetrants through dense poly(dimethylsiloxane) membranes: a transport analysis. *J Memb Sci* 92:169–184. [https://doi.org/10.1016/0376-7388\(94\)00060-3](https://doi.org/10.1016/0376-7388(94)00060-3)
- Fernández-Barquín A, Casado-Coterillo C, Valencia S, Iribien A (2016) Mixed matrix membranes for O₂/N₂ separation: the influence of temperature. *Membranes (Basel)* 6:28
- Ger D, En S, Kalıpcılar H, Yilmaz L (2007) Development of polycarbonate based zeolite 4A filled mixed matrix gas separation membranes. *J Memb Sci* 303:194–203. <https://doi.org/10.1016/j.memsci.2007.07.010>
- Gomri C, Cretin M, Semsarilar M (2022) Recent progress on chemical modification of cellulose nanocrystal (CNC) and its application in nanocomposite films and membranes-a comprehensive review. *Carbohydr Polym* 294:119790. <https://doi.org/10.1016/J.CARPOL.2022.119790>
- Inácio E, Souza D, Applications MD (2020) Thermal and crystallization behavior of PLA/PLLA-grafting cellulose nanocrystal. *Mater Sci Appl*
- Jahan Z, Niazi MBK, Hägg MB, Gregersenvind W (2018) Decoupling the effect of membrane thickness and CNC concentration in PVA based nanocomposite membranes for CO₂/CH₄ separation. *Sep Purif Technol* 204:220–225. <https://doi.org/10.1016/J.SEPPUR.2018.04.076>
- Jayaramudu T, Ko H, Kim H et al (2018) Electroactive hydrogels made with polyvinyl alcohol/cellulose nanocrystals. *Materials.* <https://doi.org/10.3390/ma11091615>
- Jia L, Xu X, Zhang H, Xu J (2022) Sulfonation of polyetheretherketone and its effects on permeation behavior to nitrogen and water vapor. *J Appl Polym Sci*
- Koester S, Roghman F, Wessling M (2015) Water vapor permeance: the interplay of feed and permeate activity. *J Memb Sci* 485:69–78
- Leemann M, Eigenberger G, Strathmann H (1996) Vapour permeation for the recovery of organic solvents from waste air streams: separation capacities and process optimization. *J Memb Sci* 113:313–322. [https://doi.org/10.1016/0376-7388\(95\)00130-1](https://doi.org/10.1016/0376-7388(95)00130-1)
- Li B, Xu D, Zhang X et al (2010) Rubbery polymer-inorganic nanocomposite membranes: free volume characteristics on separation property. *Ind Eng Chem Res* 49:12444–12451. <https://doi.org/10.1021/IE101142B>
- Liang CZ, Chung TS (2018) Robust thin film composite PDMS/PAN hollow fiber membranes for water vapor removal from humid air and gases. *Sep Purif Technol*

- 202:345–356. <https://doi.org/10.1016/J.SEPPUR.2018.03.005>
- Lovineh SG, Asghari M, Khanbabaei G (2014) CO₂ permeation through poly (amide-6-b-ethylene oxide)-nanosilica membranes. *Appl Surf Sci* 318:176–179
- Mao H, Zhen HG, Ahmad A et al (2019) Highly selective and robust PDMS mixed matrix membranes by embedding two-dimensional ZIF-L for alcohol permselective pervaporation. *J Memb Sci* 582:307–321. <https://doi.org/10.1016/J.MEMSCI.2019.04.022>
- Metz SJ, Van De Ven WJC, Potreck J et al (2005) Transport of water vapor and inert gas mixtures through highly selective and highly permeable polymer membranes. *J Memb Sci* 251:29–41. <https://doi.org/10.1016/J.MEMSCI.2004.08.036>
- Metz SJ (2003) Water vapor and gas transport through polymeric membranes
- Moaddab M, Koros WJ (1997) Gas transport properties of thin polymeric membranes in the presence of silicon dioxide particles. *J Memb Sci* 125:143–163
- Müller A, Wapler MC, Wallrabe U (2019) A quick and accurate method to determine the Poisson's ratio and the coefficient of thermal expansion of PDMS. *Soft Matter* 15:779–784. <https://doi.org/10.1039/C8SM002105H>
- Nechyporchuk O, Belgacem MN, Bras J (2016) Production of cellulose nanofibrils: a review of recent advances. *Ind Crops Prod* 93:2–25. <https://doi.org/10.1016/J.INDCROP.2016.02.016>
- OriginLab Corporation (2022) Origin(Pro)
- Planes M, Brand J, Lewandowski S (2016) Improvement of the thermal and optical performances of protective polydimethylsiloxane space coatings with cellulose nanocrystal additives. *ACS Appl Mater Interfaces*. <https://doi.org/10.1021/acsami.6b09043>
- Qu M, Abdelaziz O, Gao Z, Yin H (2018) Isothermal membrane-based air dehumidification: a comprehensive review. *Renew Sustain Energy Rev* 82:4060–4069. <https://doi.org/10.1016/J.RSER.2017.10.067>
- Reid MS, Villalobos M, Cranston ED (2017) Benchmarking cellulose nanocrystals: from the laboratory to industrial production. *Langmuir* 33:1583–1598. <https://doi.org/10.1021/ACS.LANGMUIR.6B03765>
- Rezaei-DashtArzhandi M, Ismail AF, Ghanbari M et al (2016) An investigation of temperature effects on the properties and CO₂ absorption performance of porous PVDF/montmorillonite mixed matrix membranes. *J Nat Gas Sci Eng* 31:515–524
- Rezakazemi M, Shahidi K, Mohammadi T (2012) Hydrogen separation and purification using crosslinkable PDMS/zeolite A nanoparticles mixed matrix membranes. *Int J Hydrogen Energy*. <https://doi.org/10.1016/j.ijhydene.2012.06.104>
- Rezakazemi M, Sadrzadeh M, Matsuura T (2018) Thermally stable polymers for advanced high-performance gas separation membranes. *Prog Energy Combust Sci*. <https://doi.org/10.1016/j.peccs.2017.11.002>
- Robeson LM (2008) The upper bound revisited. *J Memb Sci* 320:390–400. <https://doi.org/10.1016/J.MEMSCI.2008.04.030>
- Rowe BW, Robeson LM, Freeman BD, Paul DR (2010) Influence of temperature on the upper bound: theoretical considerations and comparison with experimental results. *J Memb Sci* 360:58–69. <https://doi.org/10.1016/J.MEMSCI.2010.04.047>
- Saba N, Jawaid M (2018) A review on thermomechanical properties of polymers and fibers reinforced polymer composites. *J Ind Eng Chem* 67:1–11. <https://doi.org/10.1016/J.JIEC.2018.06.018>
- Şahin F, Topuz B, Kalıpçılar H (2020) ZIF filled PDMS mixed matrix membranes for separation of solvent vapors from nitrogen. *J Membr Sci*. <https://doi.org/10.1016/j.memsci.2019.117792>
- Sijbesma H, Nymeyer K, van Marwijk R et al (2008) Flue gas dehydration using polymer membranes. *J Membr Sci* 313:263–276
- Simpson WT (1991) Dry kiln operator's manual. U.S. Dept. of Agriculture, Forest Service, Forest Products Laboratory
- Sohn W-I, Ryu D-H, Oh S-J, Koo J-K (2000) A study on the development of composite membranes for the separation of organic vapors. *J Memb Sci* 175:163–170
- Spatafora Salazar AS, Sáenz Cavazos PA, Mújica Paz H, Valdez Frago A (2019) External factors and nanoparticles effect on water vapor permeability of pectin-based films. *J Food Eng* 245:73–79. <https://doi.org/10.1016/J.JFOODENG.2018.09.002>
- Thiel GP, Atkinson C (2021) The potential for energy savings in U.S. houses by using isothermal dehumidification. *Sci Technol Built Environ* 27:776–787. <https://doi.org/10.1080/23744731.2021.1879564>
- Vaimakis-Tsogkas DT, Bekas DG, Giannakopoulou T et al (2019) Effect of TiO₂ addition/coating on the performance of polydimethylsiloxane-based silicone elastomers for outdoor applications. *Mater Chem Phys* 223:366–373. <https://doi.org/10.1016/J.MATCHEMPHYS.2018.11.011>
- Watson JM, Baron MG (1996) The behaviour of water in poly(dimethylsiloxane). *J Memb Sci* 110:47–57. [https://doi.org/10.1016/0376-7388\(95\)00229-4](https://doi.org/10.1016/0376-7388(95)00229-4)
- Xia T, Yuwen H, Lin N (2018) Self-bonding sandwiched membranes from PDMS and cellulose nanocrystals by engineering strategy of layer-by-layer curing. *Compos Sci Technol*. <https://doi.org/10.1016/j.compscitech.2018.03.038>
- Zhang X, Zeng S, Long J et al (2024) Natural gas dehydration membranes prepared by incorporating environmentally friendly metal-organic framework MIP-202 into polyetherimide. *J Appl Polym Sci* 141:e55253
- Zhao B, Peng N, Liang C et al (2015) Hollow fiber membrane dehumidification device for air conditioning system. *Membranes (Basel)* 5:722–738. <https://doi.org/10.3390/membranes5040722>

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.