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- theory of membrane transport
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Journal of Membrane Science

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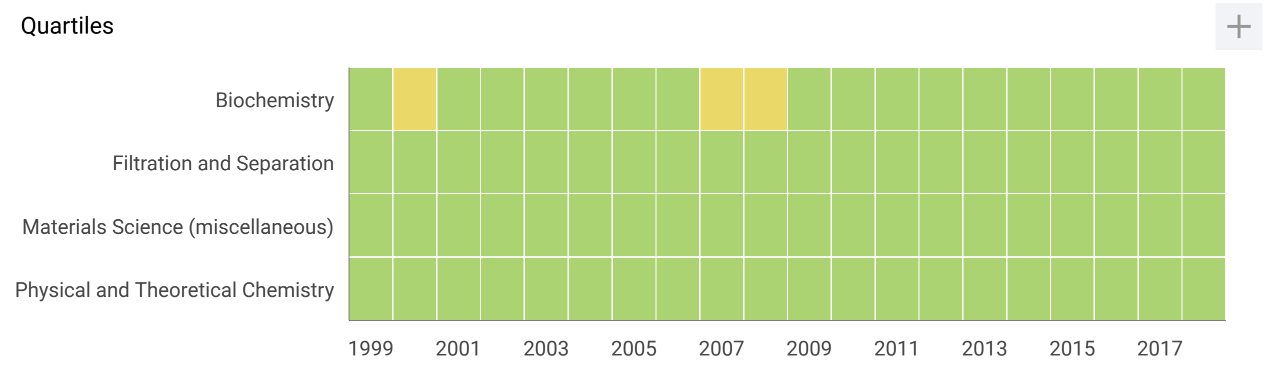
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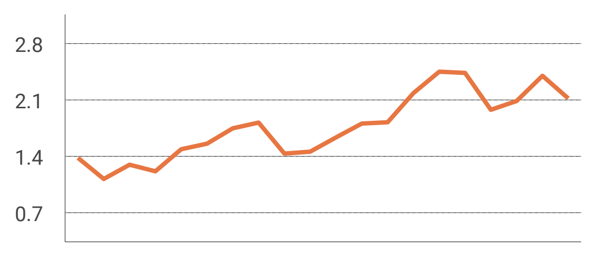

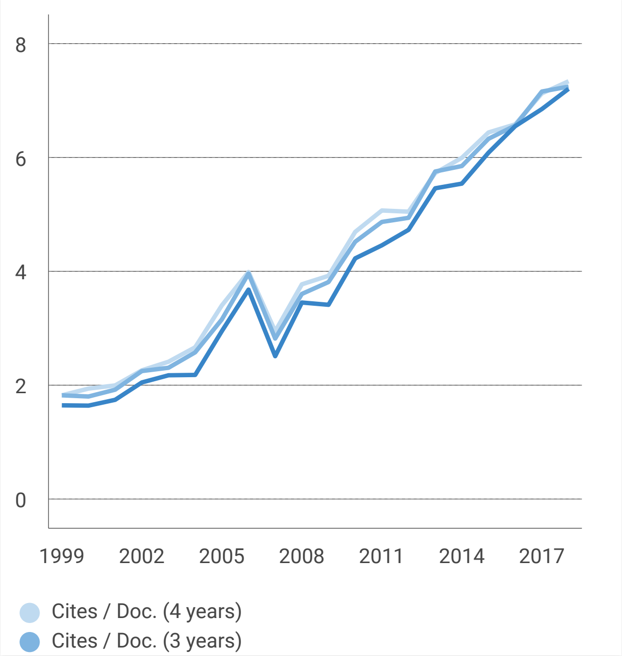

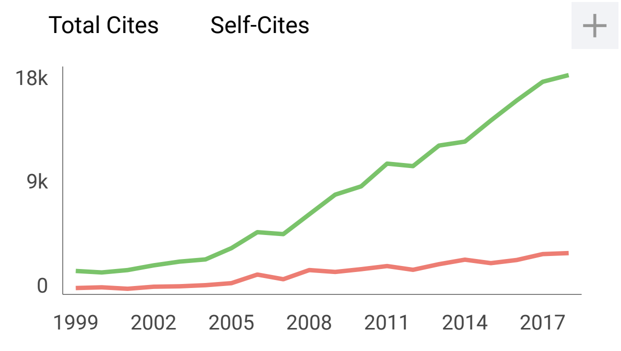

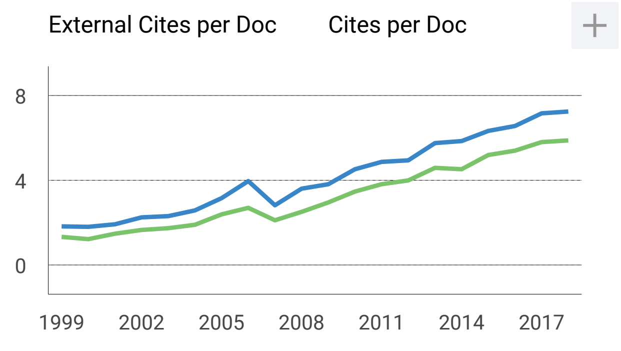

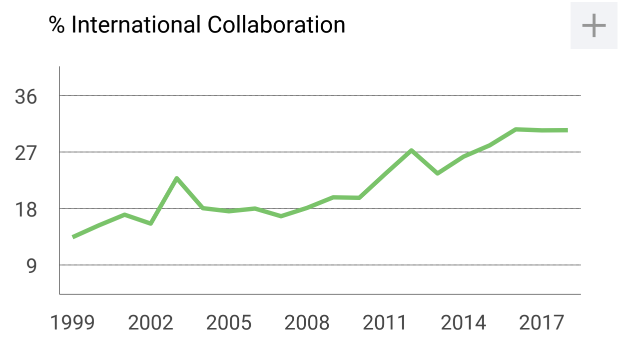

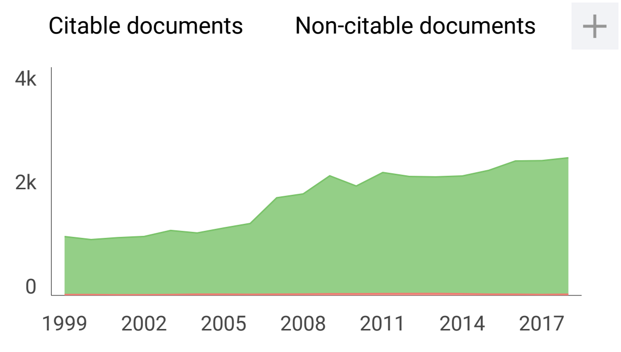

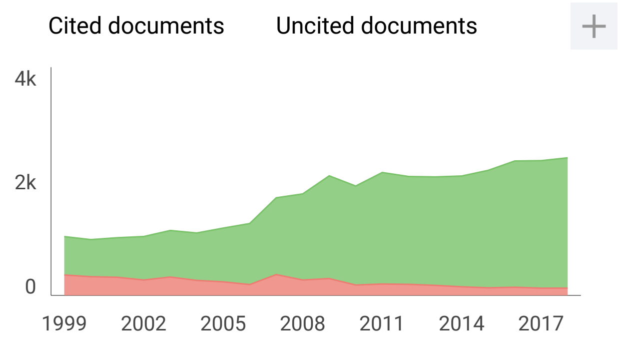

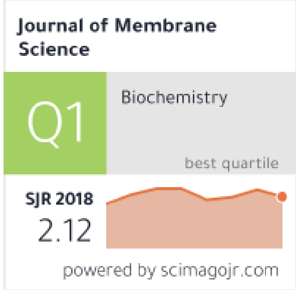

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Interlayer-free hybrid carbon-silica membranes for processing brackish to brine salt solutions by pervaporation

Hong Yang, Muthia Elma, David K. Wang, Julius Motuzas, João. C. Diniz da Costa

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Interlayer-free hybrid carbon-silica membranes for processing brackish to brine salt solutions by pervaporation

Hong Yang[1], Muthia Elma[2,3], David K. Wang[2], Julius Motuzas[2], João C. Diniz da Costa[2*]

[1]College of Environmental Science and Spatial Informatics, China University of Mining and Technology, Xuzhou, Jiangsu 221116, P.R. China.
[2]The University of Queensland, FIM^{2}Lab-Functional and Interfacial Materials and Membranes Laboratory, School of Chemical Engineering, Brisbane, Qld 4072, Australia.
[3]Chemical Engineering Department, Lambung Mangkurat Uiversity, Jl. A. Yani KM 36, Banjarbaru, South Kalimantan 70714, Indonesia.

[*]Corresponding author. Phone: +62-7-33656960; fax: +62-7-33654199. E-mail address: j.dacosta@uq.edu.au

Abstract

Mesoporous, interlayer-free, hybrid carbon-silica matrices and membranes based on tetraethoxysilane (TEOS), organosilica of triethoxyvinylsilane (TEVS) and pluronic triblock copolymer poly(ethylene oxide)-poly(propylene oxide)-poly(ethylene oxide) (P123) were successfully prepared using an acid-base catalysed sol-gel method for desalination applications. These membranes were carbonized to form the hybrid carbon-silica structures under inert conditions in vacuum and nitrogen. The effects of calcination conditions on the structure-property relationship of the carbon-silica xerogels were elucidated, and the membrane performances were systematically studied using brackish (1 wt%) to brine (15 wt %) feed concentrations of sodium chloride solution and feed temperatures (25 – 60 °C) under pervaporation process. Vacuum calcined (CS-Vc) membrane produced a slightly more mesoporous matrix and higher carbon yield than the nitrogen calcined (CS-N_2) membrane, and hence, led to comparatively superior desalination performance. CS-Vc membranes produced high water fluxes of 26.5 (1 wt%, 60 °C) and 9.2 (15 wt%, 60 °C) L m^{-2} h^{-1} with salt rejections of 99.5% and 98.6%, respectively. This study demonstrates that the combined strategy of hybrid organosilica with polymeric template and vacuum calcination offered the carbonized silica mesostructure membranes with excellent separation of water from the hydrated salt ions, and importantly, high water fluxes particularly for processing brine salt solutions.

Keywords: Interlayer-free; desalination; carbon-silica membrane; carbonizing templates; brine concentration.

1. Introduction

Access to potable water is one of the major global problems facing our current society. Coupled with population explosion and industrial development, the demand for fresh water is concerning, especially in developing countries [1]. To address this critical concern of water scarcity, desalination technologies have attracted the attention of researchers with promising potential outcomes [2]. Reverse osmosis (RO) has undergone major development and improvements for the last 3 decades, and currently it is the dominant desalination technology. RO requires high pressure to overcome the seawater osmotic pressure (~ 25 bar), thus operating at pressures as high as 80 bar [3]. The commercially available polymeric RO membranes for desalting seawater are now delivering water fluxes in the range from 14.7 to 28 L m^{-2} h^{-1} [4], though RO membranes are prone to biofouling and regenerative instability [5]. Comparative studies of ceramic and polymeric membranes have shown that ceramics foul less than polymers regardless of the chemistry of the membrane [6]. Therefore, recent progress in inorganic membrane desalination research has shown improvement in performance.

The pore size of porous inorganic membranes for seawater desalination is very small, generally in region of 5Å, thus making them very difficult to operate in a RO configuration due to extremely high pressures needed to drive water molecules through these ultra-micropores. Hence, inorganic membranes have been reported to operate mainly in pervaporation (PV) set up, which is a subset of membrane distillation (MD). PV does not require the feed to be pressurized, a major advantage for RO processes. However, MD and PV require energy to condense the water vapor that permeates through the membrane. PV is driven by a vapor pressure process, thus requiring that the feed vapor pressure to be higher than the permeate vapor pressure. As the vapor pressure is a function of the temperature, water vapor diffuses through the membranes by simply maintaining the feed temperature higher than the permeate temperature. There are three most common operation configurations for MD or PV set ups for desalination based on (i) vacuum, (ii) air gap, and (iii) sweep flow [7].

Initial PV inorganic membranes based on zeolites [8, 9] and microporous derived silica [10] delivered high salt rejection though water fluxes were very low, offering below 1 L m^{-2} h^{-1} for processing 3.5 wt% NaCl solutions. These initial results from a decade ago showed that water fluxes of PV inorganic membranes were very low and uncompetitive against RO membranes, and even against MD polymeric membranes as shown in Table 1. A note of caution must be observed when comparing different membranes tested in different set ups and under different conditions. Nevertheless, Table 1 provides a yard stick to compare the performance of different membranes and the evolution of membrane performance for similar MD/PV organic and inorganic membranes. Recent results show that PV inorganic membranes undergone major improvements, as water fluxes are now reaching values as high as below 25 L m^{-2} h^{-1} (NaCl 3.5 wt%). These results clearly show that PV inorganic membranes closed the performance gap as compared with PV organic membranes, with performances now in the same range as commercial RO membranes. It is interesting to observe that the number of inorganic materials used for membrane preparation is expanding ranging from crystalline (i.e. zeolites), to amorphous (i.e. silica), doped silica (i.e. metal oxides and carbonized templates), hybrid silica (i.e. organic link), titania and mixed matrix (i.e. carbon alumina).

Table 1 – MD/PV organic and inorganic membranes for desalting water.

Membrane	Feed temp. (^{o}C)	Salt conc. (wt%)	Water flux (L m^{-2} h^{-1})	Salt Rej. (%)	Ref.
Polypropylene	53	3.5	5.2		[11]
PVDF - polyvinylidene fluoride	53	3.5	12.0	99.9	[12]
PTFE - polytetra fluoroethylene	60	3.5	30.0	99.9	[13]
Zeolite MFI-S1	75	3.5	4.1	99	[14]
Zeolite NaA	113	3.5	4.2	99.9	[15]
Titania	75	3.5	7.0	99.1	[16]
Nickel oxide silica	60	0.3	7.0	93.0	[17]
Carbonised P123 silica	60	3.5	8.0	99.9	[18]

Mesoporous hybrid silica	60	3.5	11.0	99.9	[19]
Mesoporous Cobalt Oxide Silica	60	3.5	25.8	96.0	[20]
Carbon alumina mixed matrix	75	3.5	25.0	93.0	[21]

Initial work reported on silica membranes included the formation of microporous structures by carbonizing ionic surfactants embedded in silica matrices, where water fluxes improved from 1.6 to 2.2 L m^{-2} h^{-1} (3.5 wt% and 25 °C) as the carbon chain increased from 6 (C6) to 16 (C16) [10]. These water fluxes were very low, and many of the recent performance improvements of silica based membranes were realized by the formation of mesoporous structures. This has been achieved by using triblock copolymer Pluronic F127 [22] or F68 templates or [23], or co-assembling Pluronic F68 template with organo-bridged silica precursors such as 1, 2-bis (triethoxysilyl) ethane (BTESE) [19]. Mesoporous structures allow for an increase in the total pore volume through the membrane matrix, thus reducing resistance to the diffusion of water vapors and increasing water fluxes.

A very recent approach reported in the literature is the preparation of interlayer-free membranes, a departure from the conventional method of preparing silica derived membranes which required very smooth surfaces (i.e. like interlayers) on macroporous substrates to avoid silica film cracking or pin-holing, thus rendering silica membranes ineffective for separation processes. This recent achievement of interlayer-free silica derived membranes has been attributed to change of the silica precursors and silica sol-gel synthesis. Examples of interlayer-free mesoporous membranes tested for desalination include cobalt oxide silica calcined in air [20], and carbonized non-ionic pluronic triblock copolymer (P123) silica calcined in inert vacuum pressure [18]. The advantage of interlayer-free membranes is the reduction of the overall membrane thickness, likewise reducing the resistance to water vapour diffusion.

In order to further explore the interlayer-free and mesoporous silica derived membranes, in this work we report for the first time a preparation method containing the carbonization of ligand and non-ligand templates. To this end, we used triethoxyvinylsilane (TEVS) which contains a vinyl group as a ligand pendant of silica. TEVS has been previously used to prepare microporous silica membrane on interlayered porous substrates [24]. Further, the ligand methyl pendant groups of silica have been previously demonstrated to deliver high quality microporous silica membranes [25, 26]. To achieve mesoporosity, we incorporated TEVS with another silica precursor using tetraethyl orthosilicate (TEOS), and added a non-ligand triblock copolymer P123 template. Then, we used a base-catalyzed sol-gel synthesis which allowed deposition of sols directly onto porous substrates without the need of interlayers [27].

In this work, our method differs from those listed in Table 1, where non-ligand surfactants were embedded into the silica matrix followed by carbonization. We demonstrate that high quality carbon derived membranes can be prepared by using both ligand and non-ligand templates together with the co-polymerisation reaction of two different silica precursors where TEOS has no templates whilst TEVS has a ligand template based on a vinyl group. The resultant xerogels and silica derived membranes were calcined under vacuum or N_2 atmosphere conditions to carbonise the carbon template groups (vinyl ligand group and the P123), and characterized via FTIR, TGA, SEM and N_2 adsorption-desorption. The performance of the interlayer-free carbonised mesoporous silica membranes was evaluated systematically using a series of testing conditions, including feed salt (NaCl) concentrations from 1 to 15 wt% at varying temperatures from 25 to 60 °C via a pervaporative process.

2. Experimental

2.1 Material and membrane synthesis

Hybrid silica sol were synthesized by acid-base sol-gel method using TEOS (99.0%, GC, Sigma-Aldrich), P123 ($(EO)_{20}(PO)_{70}(EO)_{20}$, Sigma Aldrich), TEVS (97.0%, Sigma-Aldrich), ethanol (EtOH), dilute nitric acid (0.0008 M HNO_3, Merck), ammonia (NH_3, 25%, Merck) and deionized water. The preparation of hybrid silica sol was performed in a sequential two-step sol-gel process as described in a recent work. Firstly, TEVS and TEOS were added drop-wise into ethanol, stirred at 0 °C for 5 min followed by the addition of HNO_3. Then the solution was stirred and refluxed for 1 h at 50 °C. In the second step, base-catalysed polycondensation was carried out by adding ethanol-diluted NH_3 and stirred for another 2 h. P123 was then added and further stirred at room temperature for 45 min to obtain the resultant sol. The final sol pH was measured to be approximately 6 ± 0.1, which is considered to be basic because it is above the isoelectric point of the silica (1-3) [28-31]. The final molar ratios of the TEVS: TEOS: EtOH: HNO_3: H_2O: NH_3

mixture sols was 0.1: 0.9: 38: 0.0008: 5: 0.003 whilst the concentration of P123 was kept at 50 wt% based on the SiO_2 theoretical yield.

The as-synthesized sol was first mixed with ethanol in 1:0.9 to reduce the sol viscosity prior to dip-coating substrates. Thin films were coated directly on the macroporous α-Al_2O_3 tubular substrates ($\phi \approx 100$ nm) (Ceramic Oxide Fabricates, Australia) with a dwell time of 2 min and a dipping and withdrawal rate of 10 and 5 cm min^{-1}, respectively. After the deposition of each layer, the coated tube was dried briefly in an oven and then calcined in N_2 or vacuum at 450 °C for 4 h with 1 °C min^{-1} ramping and cooling rates in a tubular furnace. The cycle of dip-coating, drying and calcinations was repeated three times to obtain high quality hybrid silica membranes. The hybrid carbon silica membranes are denoted as CS-N_2 (N_2 calcination) and CS-Vc (Vacuum calcination). The equivalent bulk xerogel samples were also synthesized for material characterization. The bulk xerogels were dried at 60 °C for 24 h to obtain the dried gel which was grounded into powder and then calcined emulating the same heating treatment as the membranes.

The membrane morphology was characterized by field emission scanning electron microscopy (FESEM JOEL 7001). The xerogel was characterized by using a Shimadzu IRAffinity-1 Fourier-transform infrared spectrometer with a Pike MIRacle attenuated total reflectance accessory (ATR-FTIR) at wavelength range 400-4000 /cm, for a total of 30 scans. Nitrogen adsorption analysis was performed at 77 K and 1 bar using Micromeritic TriStar 3000 instrument to determine the structural parameters of the carbonized xerogels. Samples were degassed under vacuum for $>$ 6h at 200 °C prior to measurement. Thermogravimetric analysis (TGA) was performed using a differential scanning calorimeter / thermogravimetric analyser (Mettler-Toledo, TGA/DSC 1) from 30 °C to 800 °C using 5 °C min^{-1} in air and/or nitrogen atmosphere.

2.2 Desalination Tests

Desalination tests were carried out using a thermally based pervaporation process as displayed in Fig. 1. Briefly, a dead-end system was employed, where one end of the membrane tube was blocked whilst the other end was connected to a vacuum pump (1.5 kPa) via a cold trap to collect the permeate water. The membrane tube was immersed in the feed tank containing sodium chloride (NaCl, Sigma Aldrich) solutions with concentrations varying from 0 to 15 wt% at a controlled temperature of 25, 40 and 60 °C. The feed solution was recirculated via a peristaltic pump and constantly stirred in a biker to minimize salt concentration polarization on the membrane surface. The water vapor in the permeate stream was collected in the liquid nitrogen cold trap at pre-determined intervals.

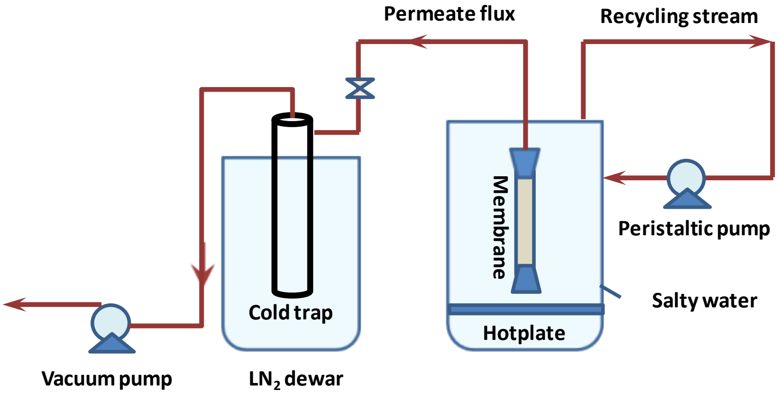

Fig. 1. Schematic of the pervaporation set-up for membrane desalination.

The water flux (F) was determined based on the Eq. 1:

$$F = \frac{m}{A\Delta t} \tag{1}$$

where m is the mass of the permeate (L) retained in the cold trap; A is the surface-active area (m^2) and Δt is the time measurement (h). The salt rejection, R (%) was obtained by Eq. (2),

$$R = \frac{\left(C_f - C_p\right)}{C_f} \times 100\% \tag{2}$$

where C_f and C_p are the feed and permeate concentrations of salt (wt%), respectively. The salt concentrations were determined by using a conductivity meter (labCHEM CP) based on a pre-determined standard curve of salt concentrations vs conductivity. To ensure a steady water flux and rejection, at least 3 permeate collections were taken at steady state conditions.

3. Results and discussion

The FTIR spectra of the xerogels calcined at 450 °C in Fig. 2 show very similar vibrational bands in the region of 1400 − 700 cm^{-1} for both CS-N_2 and CS-Vc samples irrespective of the calcination atmosphere. Bands near 800, 1060, 1220 cm^{-1} correspond to the stretching modes of siloxane bridges (Si-O-Si) whilst a shoulder 960 cm^{-1} was assigned to the silanol groups (Si-OH) [32-34]. These vibration bands are generally found in calcined silica xerogels and represent the final stages of the sol-gel synthesis leading to hydrolyses reaction due to formation of silanol groups and condensation reactions as evidenced by the formation of siloxane bridges.

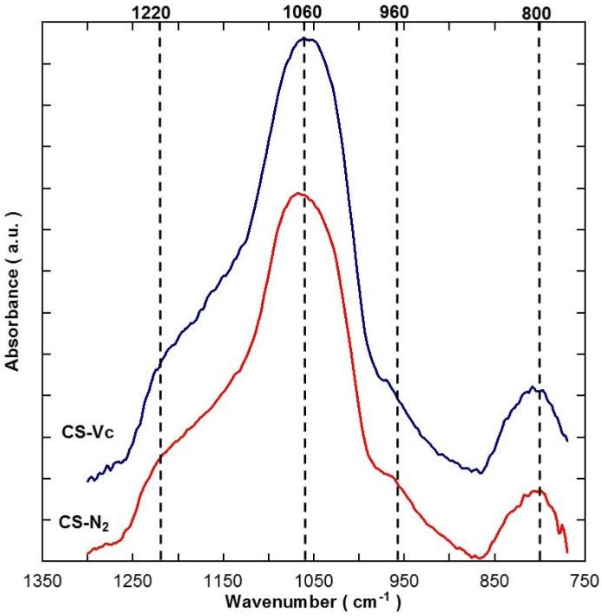

Fig. 2. FTIR spectra of CS-N_2 and CS-Vc xerogels calcined at 450 °C.

The structural properties of the carbonized xerogels were studied by N_2 adsorption-desorption technique. Fig. 3a shows that the isotherms of both samples are type IV, a characteristic of a mesoporous material. The isotherms show a strong uptake of nitrogen adsorption for relative pressures $p/p_0 < 0.2$, a region dominated by microporous structures. The increase in nitrogen uptake clearly indicates the formation of mesoporous structures, which is further evidenced by the hysteresis starting at p/p_0 ~0.5. Fig. 3b shows the pore size distributions of the both samples based on the BJH method for the desorption branch of the nitrogen isotherms in Fig. 3a. Both samples share similar features of a bimodal pore size distribution in the mesoporous region. The first cluster of peaks appears at 3.9 nm, and it is slightly broader for the CS-N_2 sample. The second distinct peak is broad peaking at 6.5 nm for the CS-N_2 sample and at 7.5 nm for the CS-Vc sample. The CS-N_2 sample slightly increased its pore size after 14 nm, whilst that of the CS-Vc sample decreased. The inset of Fig. 3b shows the pore size distribution based on the BJH method for adsorption branch which was measured at lower relative partial pressures. The inset shows that the micropores ($d_p < 2$ nm) for the CS-Vc sample increased whilst the CS-N_2 decreased.

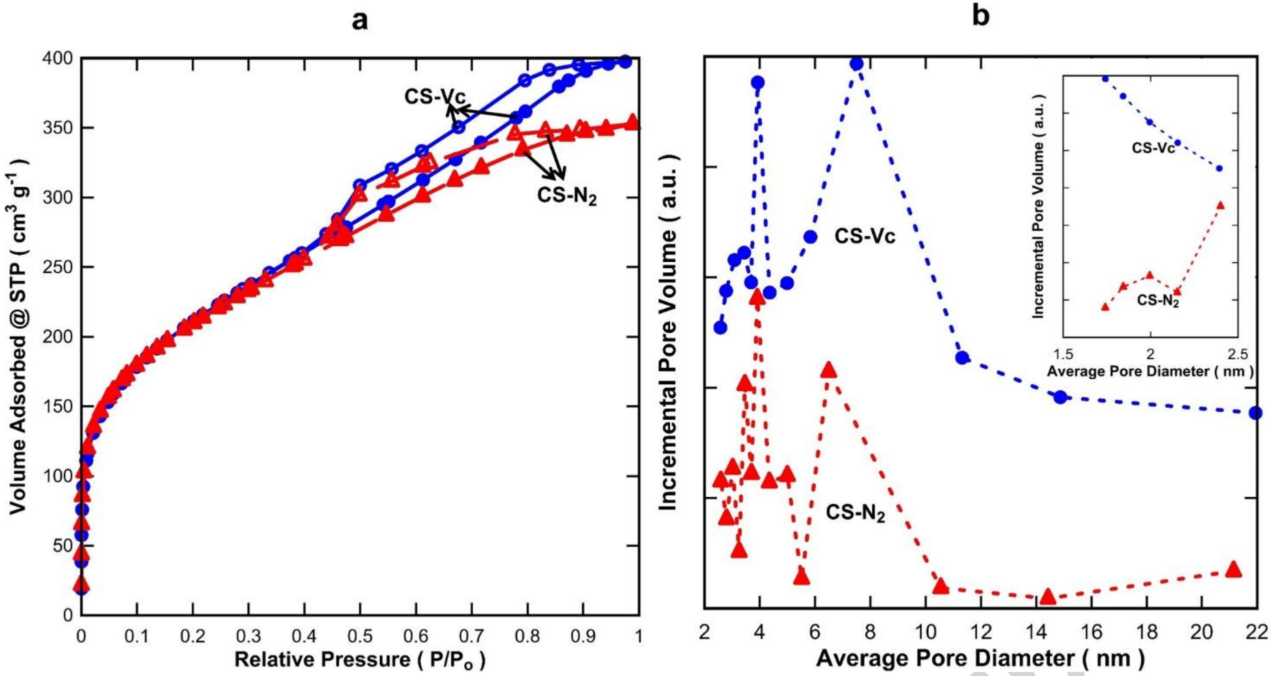

Fig. 3. CS-Vc and CS-N_2 xerogels calcined at 450 °C (a) N_2 isotherms closed symbols (adsorption) and open symbols (desorption), and (b) pore size distributions and the inset image shows the average pore diameter in the region of 1.5 and 2.5 nm.

The small structural variations of both samples related to BET surface areas, total pore volumes and average pore diameters are summarized in Table 2. It is interesting to observe the large surface areas of 761 m^2 g^{-1} achieved by the synthesis and carbonization method in this work. These surface areas are similar to those reported by Fahrenholtz et al. [35] for silica membranes prepared with methyl pendant ligand groups, though ~25% lower than carbonized P123 TEOS xerogels reported by Elma and co-workers [18]. However, the total pore volumes in this work are at least one order of magnitude higher than those reported by Wei et al. [24] for TEVS-TEOS xerogels. Therefore, the carbonization of both vinyl pendant ligand groups together with the non-ligand P123 opposed structural densification contrary to previous reported work for TEVS-TEOS derived xerogels.

Table 2. Surface properties of carbon-silica xerogels calcined in N_2 and vacuum at 450 °C

Sample Code	S_{BET} (m^2/g)	Pore volume (cm^3/g)
CS-N_2	754	0.546
CS-Vc	761	0.615

Although the membranes prepared in this work were carbonized up to 450 °C, TGA analyses were carried out up to 800 °C to understand the components made up of the carbon-silica matrix. It is envisaged that calcination of P123-incorporated TEVS:TEOS sol-gel material using nitrogen or vacuum will lead to the carbonization of the organic components from P123 block copolymer and the TEVS vinyl moiety. Starting with the carbonized xerogel samples tested in the TGA under N_2 atmosphere (Fig. 4a) and in air (Fig. 4b), there are three distinct regions of mass loss. In the first region, the mass decreases rapidly between 30 – 100 °C due to the elimination of the physisorbed water in both samples. This is common as silica is hydrophilic and readily adsorbs water molecules via hydrogen bonding with the OH groups of the silanol species [36, 37], which remain in the carbon-silica matrix as observed in the FTIR spectrum in Fig. 2. However, carbon should impart some degree of hydrophobicity, as water adsorption on carbon surface has very low surface coverage via cluster aggregation [38] onto the carbon defects or functional groups [39]. Nevertheless, the carbon-silica xerogels in this work similar to that of carbonized surfactant silica xerogels reported by Duke and wo-workers [40], and this effect is mainly attributed to the hydrophilic silica rather than the hydrophobic carbon.

It is interesting to observe that the samples calcined under vacuum (CS-Vc) adsorbed less water (3.5 wt%) as compared with the sample calcined in an inert atmosphere (CS-N_2) which shows a higher amount of 9.5 wt% in both Fig. 4a and 4b. These results can be explained by the fact that the vacuum calcination maintained a higher mass of organic carbon in the carbon-silica matrix than the inert N_2 atmosphere calcination, given that both samples contain the same molar ratio of the organic components (P123 and vinyl groups). This can be clearly seen in the second mass loss stage in Fig. 4a, which shows a mass loss difference of ~0.6% between the CS-Vc and CS-N_2 for the temperature range of 100 – 450 °C. In the third stage (>450 °C), the CS-Vc samples showed a higher mass loss as compared to a smaller mass variation in the CS-N_2 samples, thus clearly indicating that the residual organic groups arising from the pendant ligand vinyl groups or the non-ligand P123 templates remained in the matrix up to 450 °C. This can be further observed in Fig. 4b, when the carbon-silica samples were tested in an oxidized atmosphere using air, as the CS-Vc mass loss in the second stage started at lower temperatures of 380 °C. This is due to the earlier onset of thermal and oxidative degradation associated with these functional groups in air compared to N_2 atmosphere

[41]. The loss of these functional groups thus explain the reasons for the CS-Vc and CS-N_2 samples having different affinities to water.

In order to further understand the mass loss behaviour of the P123-TEVS:TEOS derived xerogels, TGA analysis was carried out in air for samples prepared without the TEVS. Fig. 4c shows the mass loss of carbonized P123-TEOS xerogel, and again the trend related to the vacuum calcination retained a higher amount of carbon mass in the second stage is maintained. However, a faster mass loss is observed for the xerogel calcined in vacuum from 100 °C onwards, whereas the mass loss profile of nitrogen treated xerogel appears to be similar to those of the corresponding xerogels in Fig 4a and 4b (black lines). Therefore, this shows that the effect of calcination environments is significant on the thermal behaviours of the P123-TEOS samples, despite both vacuum and nitrogen treatments are considered inert. In the absence of the vinyl groups (Fig 4c), vacuum calcined P123-TEOS sample led to a steady loss of the volatile organic species between 100 and 600 °C, which suggests that degradation of the P123 into smaller fragments continuously occurs over this temperature. On the other hand, when the vinyl groups are present, there is a delay in the degradation process which starts at approximately 450 °C for the CS-Vc sample in vacuum (Fig. 4a) and 300 °C in air (Fig 4b). It is possible that the hydrophobic vinyl groups of the TEVS may associate with the hydrophobic PPO blocks of the P123, which led to a delayed onset of degradation as a result of a stabilization effect. Such hydrophobic-hydrophobic interaction between TEVS and P123 is often described for the preparation of templated mesoporous ordered silica in the literature [42-44].

In contrast, all the nitrogen calcined samples have very similar mass loss profile suggests that the organic components in the gel matrix had been carbonized to some extent giving rise to the properties of a carbonaceous material. Hence, the removal of the organic fragment between 100 and 400 °C accounts for only 1% (Fig 4c). Interesting, the masses of both samples crossed over at ~400 °C contrary to the samples containing TEVS in Figs. 4a and 4b. This result suggests that the vinyl pendant ligand group was less affected by the carbonization under the vacuum conditions.

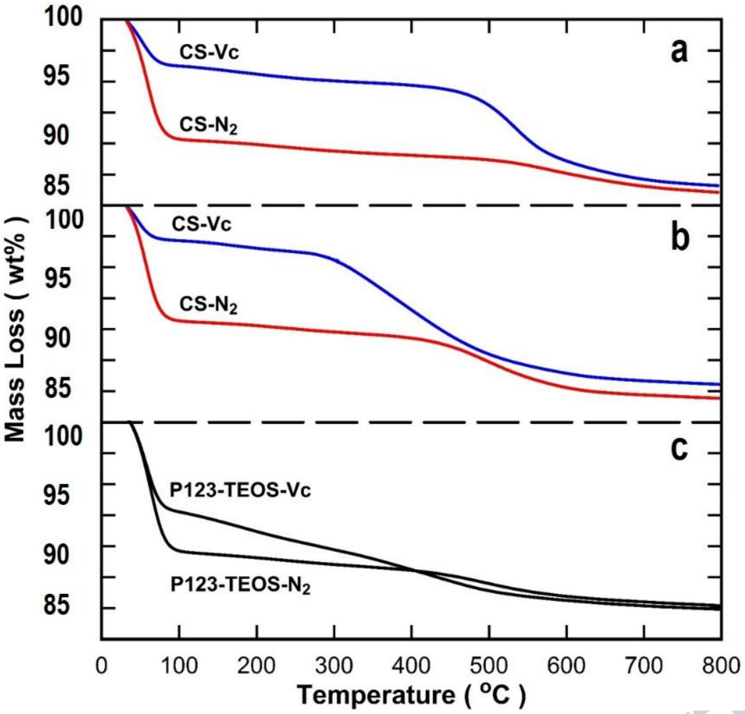

Fig. 4. TG mass loss curves of CS-Vc and CS-N_2 samples as a function of temperature exposed to (a) N_2 flow, (b) air and (c) carbon-silica samples (P123-TEOS) in air.

Fig. 5 shows the SEM images of the prepared membranes. It is observed that there is no clear boundary between the α-Al_2O_3 tubular supports and the carbon silica films in both membranes. The top layers are characterized by rough surfaces, where alumina particles are clearly observed, though full coverage was achieved as pores, pin holes and micro-crack defects were not observed. In fact, the carbon silica film penetrated into the pores of the alumina substrate, similar to these recent reports [20, 27] on the preparation of interlayer-free silica membranes. This is attributed to the contact between a dry porous surface (alumina substrate) and liquid (sol), which induces wetting forces similar to capillary forces, and modulated by surface liquid tension [45] until equilibrium is reached where no further sol infiltration takes place [46]. Upon dip-coating, the sol fills up the macropores of the substrate between the voids of the α-Al_2O_3 particles which forms a rigid structure upon drying and carbonization at 400 °C. The top surface images for the carbon silica membranes prepared under vacuum and in N_2 atmosphere show similar good surface coverages.

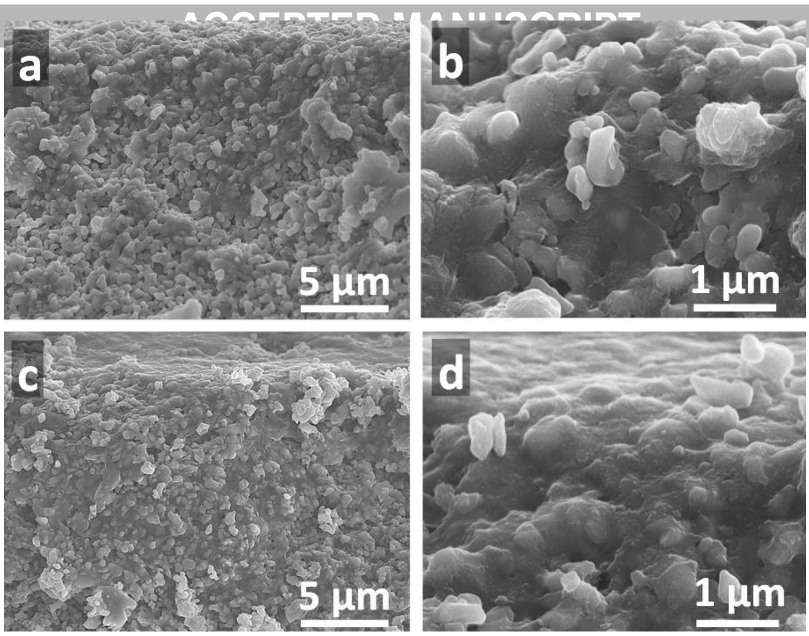

Fig. 5. SEM images of the (a, c) cross-section and (b, d) top surface of (a,b) CS-N_2 and (c, d) CS-Vc membranes.

Figure 6 displays water fluxes and salt rejection values of both membranes as a function of feed temperatures and salt concentrations. There are three important trends related to the performance of the membranes. The first trend is associated with water fluxes decreasing as a function of the feed salt concentrations. This effect is attributed to the affinity of the silica surface to retain hydrated ions as reported by de Lint et al. [47]. As hydrated salt ions such as Cl^--H_2O (6.64 Å) and Na^+-H2O (7.16 Å) [48, 49] are much larger than the kinetic diameter of water (2.6 Å), and as salt rejection is high and in excess of 98%, the retention of larger hydrated ions may cause pore blockage of the carbon-silica films, particularly the pores sizes below 7.16 Å, thus reducing the ability of the smaller water molecules to diffuse through the membrane. However, the reduction of water fluxes of 49% (CS-Vc) and 29% (CS-N_2) with increasing feed salt concentration from sea water (3.5 wt%) to brine (15 wt%) at room temperature is not as severe as a 77% flux reduction reported for pure silica membranes [50]. These results further support the beneficial effect of carbon structures in PV desalination, where the effect of hydrated ion retention on the membrane surface is greatly reduced for carbonized P123 silica [18] or negligible for carbon [21] membranes.

The second trend is related to the water fluxes consistently increased as a function of temperature for both membranes. For instance, the water fluxes measured at 25, 40 and 60 °C at 3.5 wt% feed salt concentration for the CS-Vc membrane increased in a sequential manner from 9.5 to 11.3 and 19.8 L m^{-2} h^{-1}. As PV is driven by vapour pressure difference, raising the feed temperature leads to higher vapour pressure in the feed side of the membrane, thus increasing water fluxes. It is also noteworthy that the CS-Vc membrane produced a very high water flux of 26.5 L m^{-2} h^{-1} with excellent salt rejection of 99.5% for processing brackish water (1 wt% NaCl) at 60 °C. Similarly, the same membrane delivered 9.2 L m^{-2} h^{-1} for processing brine waters (15 wt%) at 60 °C, and reaching 98.4% of salt rejection. Processing brine wastes containing 15 wt% NaCl requires overcoming osmotic pressures of 139 bar [51] at 25 °C, plus extra hydraulic pressure to drive water molecules through the RO membranes. These pressure requirements are beyond the reach of engineering RO systems, whilst the PV carbon-silica membranes in this work demonstrate they were able to handle these high salt concentrations very well. Further, the membranes in this work are generating high water fluxes in the top range of PV polymeric and inorganic membranes as compared to performance results listed in Table 1.

A third and interesting trend is that the CS-Vc membranes generally delivered high water fluxes than the CS-N_2 membranes for pure water permeation (outside the experimental error ± 8%) and slightly higher for processing saline waters. The salt rejections of the membranes were also very similar, though slightly higher for the CS-Vc membranes. These results strongly suggest that the vacuum calcination method delivered superior structural formation of the carbon-silica membranes than the N_2 atmosphere calcination method. The superior water fluxes of the CS-Vc membrane are attributed to the increase total pore volume of the CS-Vc xerogels as observed in Fig. 3 and Table 2. In other words, the CS-Vc membranes undergone lower matrix densification than the CS-N_2 membranes, which reduces the resistance to water diffusion, thus explaining the improved water fluxes.

Another interesting point associated with the vacuum calcination method is that the CS-Vc materials were more hydrophobic due to the higher amount of carbon retained in the carbon-silica matrix as ascertained by TGA analysis (Fig. 4). Hydrophobic microporous polymeric materials are generally preferred in MD membranes to avoid pore wetting [52-54]. In the case of hydrophobic materials, there is less water adsorption occurring on the surface of the CS-Vc membrane, and in this case water vapour diffusion through micro and mesopores proceeded at faster rate than the slower surface diffusion. Therefore, the combined effect of higher pore volumes and higher degree of hydrophobicity as compared to the membranes calcined in N_2 atmosphere explains the superior performance of the CS-Vc membranes.

The carbon silica membranes derived from P123/TEVS/TEOS in this work delivered water fluxes 248% higher than the P123/TEOS membranes at 3.5 wt% and 60 °C reported by Elma et al. [18]. As both membranes were interlayer-free and prepared on the same type of alumina substrates, this significant improvement is associated with the role played by the TEVS precusor. Of particular attention, the potential hydrophobic to hydrophobic interactions of the vinyl pendant ligand group in TEVS to the PPO blocks in P123 copolymer conferred the final carbon silica matrix with superior structural domains. In addition, the P123/TEVS/TEOS proved to have a superior integration into the inter-particle void of the porous alumina substrate where previous P123/TEOS derived membranes failed when carbonized in N_2 inert atmosphere [18].

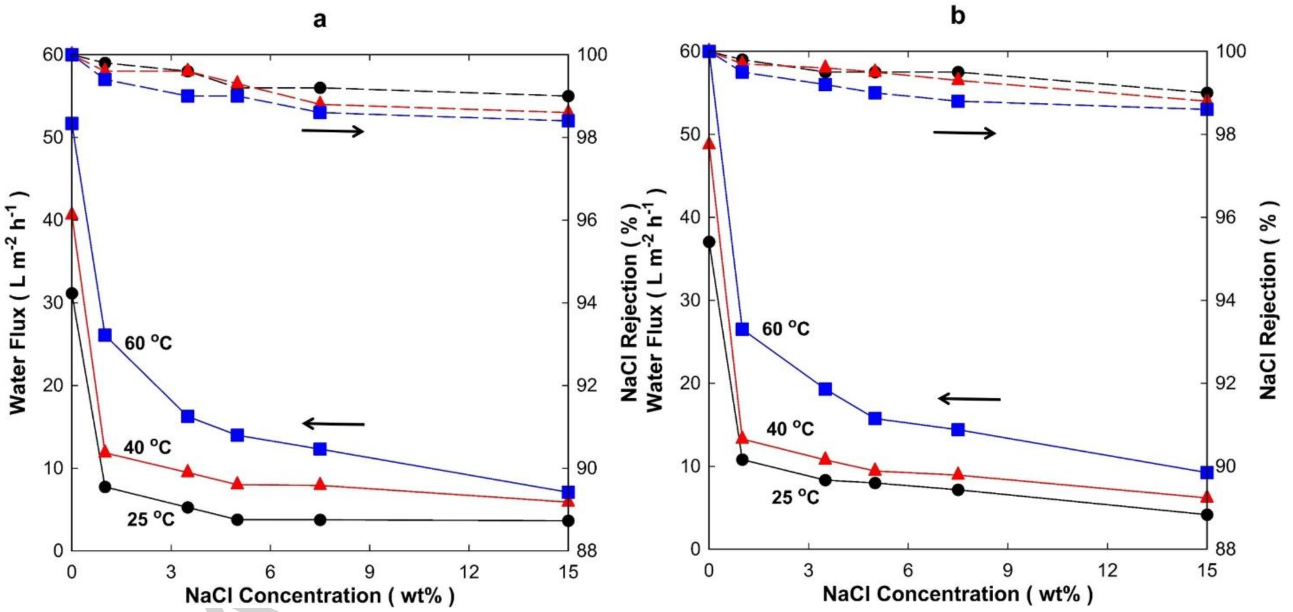

Fig. 6. Desalination performance of (a) CS-N_2 and (b) CS-Vc membranes as a function of feed salt concentrations at feed temperatures of 25 °C (●),40 °C (▲) and 60 °C (■). Water flux (± 8%) (full line ▬▬) and NaCl rejection (± 1%) (broken line -----).

Finally, the performance of the CS membranes in this work to process brines are noteworthy. There has been a number of recent publications using MD polymeric membranes for processing RO brines or coal seam gas water brines. PTFE membranes reached 10 L m^{-2} h^{-1} (at 35 °C and 5 wt% brine) [55] and 5 L m^{-2} h^{-1} (at 85 °C and 12 wt%) [56] while PVDF membranes delivered 9.5-10.8 L m^{-2}

h^{-1} (65 °C and ~7.5 wt%) [57]. These values are below the performance of the CS-Vc membrane which processed even highly concentrated brines of 15wt% and produced 9.2 L m^{-2} h^{-1} at 60 °C. The CS membranes also performed well for processing seawater concentrations of 3.5wt% at 60 °C and delivering fluxes of up to 19.3 L m^{-2} h^{-1}. This water flux is 75% and 119% higher than the carbonized template [18] or hybrid silica [19] membranes in Table 1, respectively. However, mesoporous cobalt oxide silica [20], carbon alumina mixed matrix [21] PV polymeric membranes [13] showed higher fluxes from 25 to 30 L m^{-2} h^{-1}.

4. Conclusions

In this work, interlayer-free hybrid carbon-silica membranes were prepared by templating and carbonizing pluronic triblock copolymer (P123) and vinyl pendant ligand in TEVS in a sol-gel synthesis also containing TEOS as a second silica precursor. Both vacuum and N_2 calcined samples showed mesoporous properties with high pore volume, but vacuum calcined sample (CS-Vc) produced more carbon structures within the final matrix so that a membrane with better desalination performance was prepared. CS-Vc membranes produced water fluxes of 26.5 (1 wt%, 60 °C) and 9.2 (15 wt%, 60 °C) L m^{-2} h^{-1} with salt rejections of 99.5% and 98.6%, respectively, much higher than the previously reported state-of-the-art for processing brines. Therefore, interlayer-free mesoporous carbon-silica membranes prepared with hybrid TEVS and TEOS with P123 template is a promising potential route for preparing high performance inorganic membranes for pervaporative desalination with capabilities to process a range of salt concentration from brackish to brine waters.

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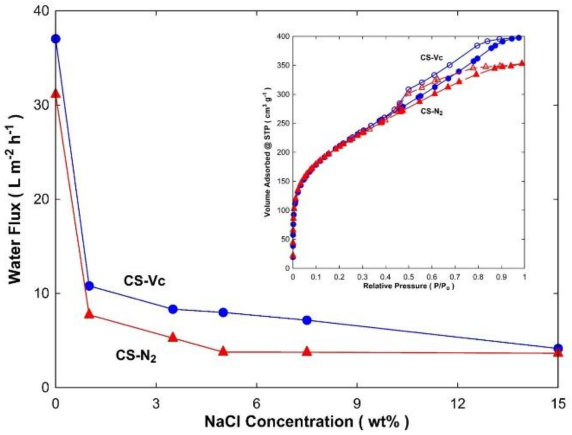
Highlights

- Interlayer-free carbon-silica matrices carbonized under vacuum and N_2 atmosphere
- Pendant ligand vinyl group and triblock copolymer embedded in the silica matrix
- Higher mesoporosity and carbon yield via vacuum as compared to N_2 carbonization
- High water flux of 26.5 $Lm^{-2}h^{-1}$ for brackish feed with 99.5% salt rejection
- Also high water flux of 9.2 $Lm^{-2}h^{-1}$ for brine (15 wt%) feed with 98.6% salt rejection

Graphical abstract



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