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Ultrafast Water Transport in Two-Dimensional Channels Enabled by Spherical Polyelectrolyte Brushes with Controllable Flexibility

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Abstract: Fast water transport channels are crucial for water-related membrane separation processes. However, overcoming the trade-off between flux and selectivity is still a major challenge. To address this, we constructed spherical polyelectrolyte brush (SPB) structures with a highly hydrophilic polyelectrolyte brush layer, and introduced them into GO laminates, which increased both the flux and the separation factor. At 70 °C, the flux reached 5.23 kg m⁻² h⁻¹, and the separation factor of butanol/water increased to ~8000, which places it among the most selective separation membranes reported to date. Interestingly, further studies demonstrated that the enhancement of water transport was not only dependent on the hydrophilicity of the polyelectrolyte chains, but also influenced by their flexibility in the solvent. Quartz crystal microbalance with dissipation and molecular dynamics simulations revealed the structure-performance correlations between water molecule migration and the flexibility of the ordered polymer chains in the 2D confined space.

Introduction

The fabrication of high-performance membranes with fast water-transport capacity is a significant challenge to overcome the widespread trade-off between flux and selectivity in waterrelated membrane processes.^[1] Recently, two-dimensional (2D) materials, such as graphene oxide (GO),^[2] MXenes,^[3] 2D metalorganic frameworks (MOFs)^[4] and covalent-organic frameworks (COFs),^[5] have attracted increasing attention for the fabrication of next-generation advanced membranes owing to their atomic thickness, fast transport channels, and micro-sized lateral dimensions. In particular, GO membranes have generated enormous interest as previous theoretical calculations and experimental studies have demonstrated promising water transport properties. However, water transport in the GO interlayer channels is limited by the strong capillary force and the inhomogeneous channel structure of the nanoscale interlayer spacing.^[6]

Therefore, many molecular engineering strategies have been implemented to regulate the water transport behaviors in GO interlayers, including partial reduction,[7] molecule/ion crosslinking,^[8] and the addition of polymers^[9] or nanoparticles,^[10] and the resultant membranes showed excellent performance with respect to the membrane flux or selectivity.[11] However, simultaneously increasing the flux and selectivity remains a challenge. Partial reduction and molecule/ion cross-linking approaches to tune the interlayer spacing are often accompanied by an undesirable decrease in flux due to a reduction in the effective channel size.^[12] Similarly, incorporating disordered polymer additives or rigid nanoparticles often reduces the selectivity or flux of the GO membranes due to uncontrolled swelling or an increase in the transport resistance.^[9b, 13] Recently, Zhang et al.^[10a] intercalated GO membranes with soft particles (polyacrylonitrile gel) to adjust the lamellar structure of GO. The polymer spheres formed by simply twisting polymer chains were able to redisperse in the solvent, which increased the mass transport resistance or degraded the well-assembled membrane structure.

To overcome these limitations of GO membranes, we designed a spherical polyelectrolyte brush (SPB) with a highly hydrophilic polyelectrolyte brush layer (Figure 1a), which was prepared by grafting a polyelectrolyte monomer with a double bond onto polystyrene (PS) spherical nanoparticles (Figure 1b). Owing to the electrostatic repulsion between charged chains, the hydrophilic polyelectrolyte chains are distributed around the PS core in an orderly manner, and the covalent bonds between the polymer chains and PS core can prevent the PS core from solvent attack.^[14] The as-synthesized SPBs consisting of polyacrylic acid (PAA-SPBs) were successfully introduced into the 2D interlayers of GO sheets. As shown in Figure 1c, the spherical soft polymeric structure and the good elasticity of the SPB chains facilitated their well incorporation between the GO sheets, effectively protecting against non-selective defects. In addition, the hydrophilicity of the polyelectrolyte chains significantly facilitates water transport in the GO interlayer channels. The resultant membrane exhibited excellent water transport performance when evaluated for the green biofuel butanol pervaporation dehydration process. Interestingly, by embedding different SPBs into the GO structure, we observed that the water-transport ability was dependent on the hydrophilicity and arrangement of polymer chains, and also highly correlated with their flexibility under real testing conditions (Figure

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1d). In addition, molecular dynamics (MD) simulations also proved that stretched chains with high ordering and flexibility were

conducive to the enhancement of water transport through GO membranes.



Figure 1. a) Illustration of the SPB structure. b) Schematic diagram of PAA-SPB synthesis, including the two key reaction steps: photoinitiator attachment to the PS core and photoemulsion polymerization. The as-prepared PAA-SPB particles show the typical Tyndall effect when illuminated with a laser beam. c) Schematic diagram of molecular transport in the PAA-SPB/GO membrane. d) Schematic diagram of the influence of the SPB state on water transport. e) Hydrodynamic size and size distribution of the PS cores, PAA-SPBs, PSS-SPBs, PAEMH-SPBs, and PDMC-SPBs measured by dynamic light scattering. Inset: Transmission electron microscopy (TEM) image of the PS core. Scale bar: 50 nm. f) Illustrations of the molecular structures of four SPBs and their corresponding TEM images. Scale bar: 50 nm.

Results and Discussion

SPB synthesis and membrane preparation

PAA-SPBs with negative charges were prepared via the photoemulsion polymerization method (Figure 1b).^[15] A solution containing PS cores of uniform size (Figure 1e) was synthesized via an emulsion polymerization method (Figure S1a), followed by the covalent attachment of 2-[p-(2-hydroxy-2methylpropiopenone)]-ethylene glycol methacrylate (HMEM, Figure S1b-c) as a photoinitiator. Then, PAA chains were grafted

onto the surface of the PS core under UV irradiation. The assynthesized PAA-SPB consisted of a solid PS core and a flurry brush layer (Figure 1a). The solid PS core was clearly observed by the transmission electron microscopy (TEM, Figure 1f). The corresponding Fourier transform infrared spectroscopy (FTIR, Figure S2a), and 1H nuclear magnetic resonance (1HNMR, Figure S2b) further confirmed that the PAA-SPBs possessed good chain structural characterizations.^[15] The PAA-SPBs were steadily dispersed in an aqueous solution, which showed a stretching hydrodynamic size of about 120 nm (Figure 1e) and a significant Tyndall effect (Figure 1b). Furthermore, the particle size of the PAA-SPBs (Figure S3) showed good stability in aqueous solutions over a wide temperature range (25–70 °C) owing to the synergistic effect between the polyelectrolyte chains

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and the PS core, whereas the pure polyelectrolyte disaggregated and randomly distributed in the aqueous phase. Water vapor adsorption tests showed that the PAA-SPBs have good hydrophilicity (due to the high affinity of the carboxyl groups to water molecules), with an adsorption capacity of 66.7 mg/g (Figure S4), whereas the PS cores had a value of 3.96 mg/g owing to the hydrophobic structure. Moreover, the pressure developed as a result of the structure being stretched (as the ionized chains with the same negative charge repel each other) can facilitate water intake.^[16]

The PAA-SPBs were mixed with GO nanosheets dispersed in water (Figure S5) to form a homogeneous solution (Figure S6a), followed by the assembly of the PAA-SPB/GO membranes on porous polyethersulfone (PES) substrates using a pressureassisted filtration method (Figure S6b). The PAA-SPB/GO membranes can also be easily fabricated on other substrates, such as cellulose acetate, AAO, or polyacrylonitrile (PAN) (Figure S6c-e). The membrane thickness was precisely controlled from 52 to 539 nm by changing the volume of the filtration solution used (Figure S7-8). Well-stacked PAA-SPB/GO membranes with some wrinkles were obtained, similar to the pristine GO membranes (Figure 2a-d). However, excessive addition of SPBs would cause partially aggregation of SPBs (Figure S9a-c), and even formed a filter cake layer (Figure S9d).

To observe the distribution of SPBs in the GO interlayer channels, the SPB/GO membranes were mechanically exfoliated (Figure 2e and Figure S10a). Figure 2f-g and Figure S10b-d show the SPB distributions in a series of randomly exfoliated interlayers. The SPBs were well intercalated into the multilayered GO nanosheets and did not aggregate together, which was further demonstrated by TEM (Figure 2h-i). The SPBs adhered well onto the GO nanosheets (Figure 2h) and the layered stacked structure of GO was clearly visible Figure 2i. The incorporation of hydrophilic SPBs creates more water transport channels in the membrane. In addition, the isolated distribution of the SPBs is beneficial for maintaining the integrity of the membrane structure and the finely ordered arrangement of polymer chains. Notably, the isolated SPB particles did not change the main stacking structure of the GO membrane, which was further confirmed by X-ray diffraction (XRD) measurements. As shown in Figure 2j, both the SPB/GO and pure GO membranes had similar XRD patterns, with characteristic peaks at ~10° in the dry state and at ~9.5° in the wet state. In addition, PAA-SPB/GO membranes with different doping ratio also showed similar XRD patterns (Figure S11). These results indicate that the SPBs had a negligible influence on the interlayer d-spacing of the GO membrane, in accordance with the above hypothesis.

Synchrotron small-angle X-ray scattering (SAXS) was used to further investigate the distribution state between PAA-SPBs and GO nanosheets. Generally, the introduction of GO nanosheets was supposed to increase the scattering intensity. However, as shown in Figure 2k, when a small amount of GO nanosheets was added to the PAA-SPB solution, an apparent decrease in the scattering intensity was observed, indicating that the GO nanosheets were distributed around the SPBs (insets of Figure 2k) and partially shielded the electrostatic repulsion force between SPBs themselves, rather than intertwining with the polyelectrolyte chains of PAA-SPBs. Figure S12 shows that the scattering intensity increases with increasing SPB concentration, demonstrating the uniform dispersion of SPBs. The uniform dispersion is closely related to the strong electrostatic repulsion force between SPBs. In addition, as the SPBs are integrated in the GO laminates, the SPBs simultaneously experience various forces (Figure S13), including an external pressure, interaction forces between GO nanosheets and PAA-SPBs, and swelling force of the SPB chains, further making them well incorporation with the GO nanosheets. As a result, well-stacked SPB/GO membranes were obtained, which contributed to the membrane separation process. Meanwhile, Shao et al.^[17] revealed that largesized molecules inhibit the overlap of GO nanosheets, resulting in lower degree of alignment and shorter transmembrane pathway. We expected the introduction of PAA-SPBs would also create more efficient water transport channels in membrane.

Water transport properties

The selective water transport properties of the SPB/GO membranes were evaluated for the dehydration of butanol-water mixtures, which require membranes with fast and selective water transport. Figure 3a shows the effect of the amount of PAA-SPBs in the GO membrane on the flux and separation factor during the dehydration tests. The highest flux was achieved using a membrane with 4.62 wt.% PAA-SPBs, while the maximum separation factor was observed at 1.54 wt.%. The separation factor increased significantly with increasing PAA-SPB content in the range of 0-1.54 wt.%, while the flux increased linearly. However, with the further addition of PAA-SPBs (1.54-4.62 wt.%), the flux increased more rapidly, accompanied by a rapid reduction in the separation factor, due to the formation of non-selective defects. Furthermore, excessive doping (>4.62 wt.%) introduced higher transport resistance (Figure S9), which resulted in an unsatisfying membrane separation performance. Hence, a PAA-SPB content of 1.54 wt.% was considered optimal and was used for all further analyses of the SPB/GO membranes.

Interestingly, as the testing temperature was increased from 40 to 70 °C, the PAA-SPB/GO membrane showed an unusual synchronous enhancement of the flux and separation factor (Figure S14a). In contrast, such a phenomenon was not observed for the pristine GO membrane (Figure S14b), which showed the typical trade-off between flux and selectivity. The sorptiondiffusion mechanism is widely used to explain the separation process in 2D membranes.^[18] Generally, activation energy (E_a) obtained from ln J vs. 1/T plot is used to reveal temperature dependence of water transport properties in the pervaporation membrane, including both the diffusion and sorption processes. An increase in temperature increases the molecule diffusion rate and weakens the sorption ability of the membrane, resulting in a balance between the membrane flux and selectivity. Herein, both the permeation flux and selectivity of the composite membrane were increased significantly with increasing operation temperature, indicating that the water transport capability in the PAA-SPB/GO membrane is highly enhanced compared with that of the pristine GO membrane. Figure S15 shows that E_a of the water molecules transport through the PAA-SPB/GO membrane is higher than that of the pure GO membrane, further indicating that the permeation of water becomes more sensitive to operating temperature after incorporating the PAA-SPBs. This enhanced temperature dependence significantly overcomes the membrane trade-off effect between permeability and selectivity. It is worth noting that the separation factor of the PAA-SPB/GO membrane reached approximately 8000 with a desirable permeation flux of

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 $5.23 \text{ kg m}^{-2} \text{ h}^{-1}$ at 70 °C, which considerably exceeded that of most state-of-the-art membranes used in butanol/water separation to date (Figure 3b and Table S1). In addition, the PAA-SPB/GO membrane showed a stable flux and separation factor over time

(Figure S16), confirming the structural stability of the membrane. Overall, the excellent separation performance of the PAA-SPB/GO membrane highlights the great potential of SPB additives for the improvement of water transport in 2D channel.



Figure 2. Surface SEM images of a) GO and b) PAA-SPB/GO membranes. Cross-sectional SEM images of c) GO and d) PAA-SPB/GO membranes. e) Schematic diagram of the mechanical exfoliation process. Part A and Part B are the tape side and membrane side, respectively. The entire exfoliation process is random. SEM image of the distribution of PAA-SPBs in membrane for f) Part A and g) Part B after exfoliation. TEM images of the h) distribution of PAA-SPB in GO layers and i) GO layers. j) XRD spectra of GO and PAA-SPB/GO membranes in dry state and 90 wt.% butanol/water environment. k) SAXS scattering curves of PAA-SPBs, GO, and PAA-SPB/GO in solution. Inset: schematic diagram of the interaction between PAA-SPBs and GO before and after introducing GO. All composite membranes had the same doping amount (1.54 wt.%).

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Figure 3. a) Pervaporation performance of PAA-SPB/GO membranes with PAA-SPB contents of 0.00–15.40 wt.% measured at 30 °C with 10 wt.% water in butanol as the feed. b) Comparison of pervaporation performance of our PAA-SPB/GO membrane (70 °C) for butanol dehydration with polymeric membranes, inorganic membranes, GO membranes, and a MOF membrane. The dashed purple line provides guidance for the eye and indicates membranes. The corresponding references and details of the specific membranes are listed in Table S1. c) Comparison of the butanol dehydration performance of PAA-SPB/GO, PAA/GO, PS/GO, and GO membranes at 70 °C (90 wt.% butanol/water mixture). d) Pervaporation performance of PAA-SPB/GO, PAS-SPB/GO, PAEMH-SPB/GO, and PDMC-SPB/GO membranes at 70 °C (90 wt.% butanol/water mixture). All composite membranes in b-d have the same doping amount (1.54 wt.%) with a membrane thickness of about 160 nm.

Water transport mechanism

To further investigate the interesting behavior of the SPBs, we directly introduced a polyelectrolyte polymer (polyacrylic acid) with the same weight ratio into GO membranes, henceforth referred to as PAA/GO membranes. As expected, the PAA/GO membrane showed a similar result to that of the pure GO membrane. As shown in Figure 3c, Figure S14 and Figure S17a, the introduction of the polyelectrolyte fails to significantly increase the separation factor, and did not provide a noteworthy improvement in the flux for the PAA/GO membrane. This result is consistent with our assumptions. Although polyelectrolytes with high hydrophilicity can enhance the water sorption ability, their disordered self-volume would occupy the spacing between GO layers and thus influence the membrane performance from two aspects: unlimited swelling and enhanced transport resistance. We also prepared GO-based composite membranes doped with only PS spherical nanoparticles, referred to as PS/GO membranes, whose performance did not show significant improvement (Figure. 3c, Figure. S14 and Figure S17b). This result indicates that the constraint of the PS core for polyelectrolyte chains plays a key role in water transport behavior; that is, the grafting of the polyelectrolyte chains can indeed enhance the separation performance of the GO membrane. Furthermore, we introduced a PAA-SPB layer on the surface of as-prepared GO membranes, referred to as PAA-SPB@GO membranes, which did not show enhancement of the flux and separation factor (Figure S18). Combined with the sorptiondiffusion theory, PAA-SPBs mainly promote the diffusion of water in the 2D confined spacing rather than the sorption of water on the membrane surface, thereby improving the separation performance.

Further characterization was performed to better understand how PAA-SPBs facilitate water transport in 2D confined spaces and the water transport mechanism. In general, water transport behavior is closely related to membrane hydrophilicity. A homemade self-evaporation weight-loss setup (Figure S19a) was utilized to investigate the pure water permeation behavior of the membranes as a function of time. As expected, the PAA-SPB/GO membrane showed a faster weight loss than the pure GO membrane (Figure S19b), indicating a higher pure water flux with stronger hydrophilicity.

To further investigate the key role of the hydrophilicity of the additive polyelectrolyte, we synthesized three additional SPBs with different hydrophilicities by grafting poly (p-styrene sulfonate) (PSS), poly (2-aminoethyl methacrylate hydrochloride) (PAEMH),

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and poly (methacryloxyethyl trimethyl ammonium chloride) (PDMC) onto the PS core (Figure 1e and f, Figure S20). The corresponding SPBs are labeled PSS-SPB, PAEMH-SPB, and PDMC-SPB, following the order of hydrophilicity of PSS-SPB > PDMC-SPB > PAA-SPB > PAEMH-SPB (Figure 4a). Furthermore, the dipole moments of the SPB monomers, which are linearly related to the polarity of SPB, were calculated using density function theory. The calculated results showed that dipole moments have the same sequence as the hydrophilicity of the four SPBs (see the inset data in Figure 4a and Table S2). The pure water permeation behaviors of the corresponding composite membranes also followed the above order of the four SPBs (Figure S21). In theory, the stronger hydrophilicity of SPBs could increase the affinity for water molecules, resulting in a higher separation performance for the associated composite membrane. However, unexpectedly, their separation performance in the dehydration of aqueous butanol by the pervaporation (PV) separation process did not show this order. As shown in Figure. 3d and Figure S22 a-c, the PAA-SPB/GO membrane had the best separation performance for both the flux and separation factor, implying that the intrinsic hydrophilicity of SPBs is not the only factor influencing water transport. This unexpected discovery motivated us to explore other potential factors that influence the membrane transport process.

The PAA-SPBs were well dispersed in both water and butanol solutions (Figure S23a), suggesting that the PAA-SPB chains could maintain the stretched state during the PV separation process. However, when the PSS-SPBs, PAEMH-SPBs, and PDMC-SPBs were dispersed in a butanol environment, all of these SPBs underwent excessive aggregation (Figure S23ab) as organic solvents can shield the electrostatic repulsion force in adjacent charged chains, resulting in shrinkage of the chains, but have little influence on PAA-SPBs. This shrinkage restrains polyelectrolyte chain flexibility, which inhibits the transport of water molecules, and thus greatly reduces the water carrying capacity of the SPBs (Figure 1d and Figure S24a). In contrast, when pure water was fed, the water diffusion rate did not show this unusual phenomenon (Figure S21) and had the same order as the hydrophilicity of the SPBs owing to the absence of butanol molecules (Figure S24b).

To gain further evidence of the effect of structural change on the flexibility and the water carrying capacity, a quartz crystal microbalance with dissipation (QCM-D) system was used to detect the microscopic variations in the SPBs. Fluctuations in the frequency shift (Δf) and energy dissipation shift (ΔD) represent the adsorption ability and flexibility degree of polyelectrolyte chains, respectively.^[19] In the initial state, all the SPB samples

were kept in a butanol environment, as shown in Figure 4b. The four polyelectrolyte chains are restrained to a certain extent, resulting in intertwining. In contrast, the PAA chains can stretch to a larger extent, owing to their better dispersibility in butanol (see Figure S23). With the introduction of a trace amount of water, the ΔD of the PAA-SPBs increased sharply and the corresponding Δf significantly declined (Figure 4c and d), whereas the other SPBs showed a relatively small change in both D and f. Water adsorption on PAA-SPBs facilitates their chain ionization, which makes the chains more ordered owing to mutual repulsion between negatively charged chains (Figure 4bi and 4bii). Usually, Δf depends on the mass adsorbed on the SPBs, and thus can serve as an indicator of the binding affinity of water molecules with the polyelectrolyte, while ΔD is closely related to the shrinkage and densification of the polyelectrolyte. Hence, the larger Δf and ΔD values resulting from water adsorption indicate that PAA-SPBs have stronger affinity for water molecule (Figure 4c) and more flexible chain structures (Figure 4d) than PSS-SPBs. PAEMH-SPBs, and PDMC-SPBs in the given solvent. The QCM-D results support the above inference that the shield effect derived from the organic solvent could induce shrinkage of the polymer brush, thereby limiting the flexibility and water-capture ability of SPBs, which is a non-negligible factor in the PV process. Furthermore, the strong interaction between the oxygencontaining groups of GO and the amines of PAEMH and PDMC further constrained the flexibility and water-capture ability of PAEMH-SPBs and PDMC-SPBs.^[20] Therefore, the separation performance of the corresponding PSS-SPB/GO, PAEMH-SPB/GO, and PDMC-SPB/GO membranes (Figure 3d) did not follow the sequence of the hydrophilicity due to their poor water transport capability. The orderly stretching state of PAA-SPBs in butanol was responsible for the superior separation performance of the PAA-SPB/GO membrane.

In addition, PAA-SPBs are sensitive to the variation of solution pH because PAA is a weak polyelectrolyte. With a decrease in pH, the diameter of PAA-SPB decreases, indicating the shrinkage of polyelectrolyte chain of the PAA-SPBs (Figure S25). This property of PAA-SPBs can be further used to evaluate the effect of the shrinkage of polyelectrolyte chains on the membrane separation performance. We measured the separation performance of the PAA-SPB/GO membrane under different pH conditions, which showed an obvious decrease of separation factor with a slight decrease of pH (Figure S26), similar to the results varying with the different SPBs (Figure 3d). This drop is attributed to the shrinkage of PAA-SPBs and declines their water carrying capacity.

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Figure 4. a) Weight of water adsorbed by the PAA-SPB, PSS-SPB, PAEMH-SPB, and PDMC-SPB membranes using the QCM-D. Inset: dipole moments of the monomers of the four SPBs calculated by DFT simulation. b) Schematic diagrams of the changes in the state of the chains of PAA-SPB and the other three SPBs. (D_0 , D_1) and (D_0' , D_2) in (i) and (ii) are the dissipation values of the SPBs in the respective environments. The given equations were used to calculate the amount of water adsorption and D values from the QCM-D data. c) Frequency shift (Δf) of the four SPBs placed on the QCM-D sensor upon dropping a trace amount of water after stabilization in a butanol environment. d) Energy dissipation shift (ΔD) of the four SPBs simultaneously measured with Δf by QCM-D. All *f* and *D* data were from the fifth overtone (n = 5). The sharp shifts or oscillations at ~325 s for all curves were attributed to the switching of the solution (water and butanol) with different density and viscosity. e) Schematic of the modeled structure of the GO composite membrane system, where (i) and (ii) correspond to GO membranes with stretched or agglomerated brush chains, respectively. f) The number of permeated water molecules of the simulated water transport process for the different GO composite membranes over 100 ns.

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To further elucidate the benefits of stretching the polyelectrolyte chains, MD simulations were performed to study the water transport through the SPB/GO membrane with stretched chains or agglomerated chains. As schematically shown in Figure 4e, when the chains stretch, there are three types of interlayer spacings: (1) GO interlayer spacing (d_1) ; (2) GO interlayer spacing produced by polyelectrolyte chains (d_2) ; (3) GO interlayer spacing produced by the PS core and chains (d_3). In contrast, the GO membrane with agglomerated brush chains has only two types of interlayer spacings (d_1 and d_2). By comparing the water transport behavior in the GO lamina between stretched SPB and agglomerated SPB, faster water permeation occurs in the GO membrane with stretched chains. It was found that stretched chains enlarged the water transport channels, and their flexible hydrophilic chains were beneficial for fast water permeation (Figure 4f).

Overall, SPBs are highly beneficial for improving the water transport ability in 2D confined spaces. Their stretched arrangement is closely related to the solvent response of the polymer chains. The PAA-SPBs showed better flexibility in PV measurements, which effectively promoted the transport of water molecules. In contrast, the other three SPBs showed major shrinkage of the chains, which greatly reduced their flexibility and water transport capacity, although some of them display stronger hydrophilicity than PAA-SPBs.

Conclusion

Rationally incorporating functional PAA-SPBs with hydrophilic flexible PAA polyelectrolyte chains into GO 2D confined spaces led to the successful fabrication fast water transport channels in membranes and at the same time hamper the transport of butanol molecules, offering the simultaneous enhancement of flux and selectivity that exceeds most state-ofthe-art membranes reported to date. Notably, the key roles of the hydrophilicity and flexibility of the polyelectrolyte chains during the butanol pervaporation dehydration process were confirmed using the QCM-D technique, comparison experiments, and molecular dynamics simulations. It is especially noteworthy that the structural state of the polyelectrolyte chains in the solvent, which has been neglected to date, is strongly related to water transport ability. This work may result in more attention being paid to the intrinsic properties of incorporated materials, including flexibility, polarity, and hydrophilicity, among others, which can be used to create better membranes with highly efficient, fast water transport channels. Additionally, the interesting structures of the SPBs may lead to the modification of other materials, such as MOF crystals and TiO2 nanoparticles, and may also arise more possibility for the design of other 2D membranes (e.g., MXenes, 2D MOFs, 2D COFs, etc.). Finally, the design and construction of 2D confined channels with excellent water transport capacities that use highly hydrophilic, highly flexible, and controllable polyelectrolyte chains may lead to outstanding 2D membranes that have great potential for use in biofuel-production, water-remediation, and wastewatertreatment applications, as well as many other industrial water processes.

((Note: Please place comprehensive details in the Supporting Information.))

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Keywords: water transport • ordered polymer chains • chain flexibility • two-dimensional confined spacing • graphene oxide

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Entry for the Table of Contents

Ultrafast water transport in two-dimensional channels enabled by spherical polyelectrolyte brushes with controllable flexibility

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Fast and high-efficiency water transport behaviour in 2D GO confined spacing assisted by spherical polyelectrolyte brushes can be easily realized. Further studies revealed that water transport capacity was not only dependent on the hydrophilicity of the polyelectrolyte chains but also influenced by their flexibility in the solvent.

