Biomimetic Nanocomposite Membranes with Ultrahigh Ion Selectivity for Osmotic Power Conversion

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ABSTRACT: Ion transport in nanoconfinement exhibits significant features such as ionic rectification, ionic selectivity, and ionic gating properties, leading to the potential applications in desalination, water treatment, and energy conversion. Two-dimensional nanofluidics provide platforms to utilize this phenomenon for capturing osmotic energy. However, it is challenging to further improve the power output with inadequate charge density. Here we demonstrate a feasible strategy by employing Kevlar nanofiber as space charge donor and cross-linker to fabricate graphene oxide composite membranes. The coupling of space charge and surface charge, enabled by the stabilization of interlayer spacing, plays a key role in realizing high ion selectivity and the derived high-performance osmotic power conversion up to 5.06 W/m². Furthermore, the output voltage of an ensemble of the membranes in series could reach 1.61 V, which can power electronic devices. The system contributes a further step toward the application of energy conversion.

INTRODUCTION

Ion transport across the biological ion channels is a prerequisite for regulating the vital functions in life processes.1−5 Different from that in the bulk, ion transport in such nanoconfinement with the characteristic dimensions comparing to the Debye length presents ionic rectification, ionic selectivity, and ionic gating properties.6−9 A series of significant progresses have been achieved in designing the artificial nanofluidic nanochannel systems,4−6 along with the applications in fields such as DNA sequencing, water desalination, and energy conversion.10−16 Single-pore nanofluidics have provided a simplified research platform to carry out fundamental mechanism studies on biomimetic osmotic energy, which highlight great potential for sustainable energy conversion.11,12 Two-dimensional nanofluidic based systems, reassembled with nanosheets such as graphene and molybdenum disulfide,13,14 provide an easy and cheap strategy for large-scale integration. Their well-defined configuration and angstrom-scale interlayer distance as biological channels, have been demonstrated to guarantee the selective transport of ions.15 Graphene oxide (GO) nanosheet has been discovered to be an ideal material for nanofluidic ion transport,16,17 as well as osmotic energy conversion.18 Ions can transport in the capillaries formed by the adjacent GO nanosheets. And the interlayer spacing (also known as d-spacing), which is close to the diameter of hydrated ions, provides the possibility to achieve ultrafast and ultrahigh selectivity.19,20 However, the oxygen-containing functional groups of GO nanosheets tends to induce the expansion of the d-spacing and even delamination in aqueous solutions, which challenge the success of long-term stable nanofluidics.21 Besides, the limited charged functional groups makes it difficult to pursue a high-efficiency generator. To address this challenge, mixed-dimensional heterostructures have been adopted.22

Herein, we design a GO-based composite membrane as nanofluidic devices for ion transport (Figure 1a). The one-dimensional charged aramid nanofibers (ANFs) are introduced as an interlocking agent to cross-link the two-dimensional GO nanosheets. This nacre-like structure endows the GO/ANF with chemical and physical robustness. The oxygen-containing groups on GO nanosheets and the abundant amide groups on ANFs can form hydrogen bonds, which are essential to prevent membrane swelling and stabilize the interlayer spacing (Figure 1a). The stabilization of d-spacing allows the precisely selective transport of ions through the GO/ANF membranes. The ion transport across the composite membranes shows the typical surface-charge-governed features, offering a precondition for
the osmotic power generation under salinity gradient. Therefore, the ultrahigh ion selectivity, enabled by the space charge donor ANF, is demonstrated to improve the osmotic output power density up to 5.06 W/m² with artificial river water and seawater. In addition, an ensemble of the membranes in series can offer an output voltage of 1.61 V to power the electronic device. This work also provides an effective guidance for 2D membrane designs with promising ion transport behavior and osmotic power conversion performance.

**RESULTS AND DISCUSSION**

The GO nanosheets obtained by Hummers method possess abundant functional groups (Supporting Information Figure S1a), such as -COOH, -C=O-C, and -OH, on both the edge and the surface. The guest material is ANF. During the ANF exploitation process, the strong base could weaken or eliminate the interchain interactions of polymer chains (Figure S1b), demonstrating the deprotonation of amide groups. The dispersions of GO/DMSO and ANF/DMSO were mixed, and then water was added, which contributes to the reprotonation of ANFs to form hydrogen bonds between the oxygen-containing groups on GO and the abundant amide groups on ANFs. The GO/ANF membranes are obtained by using the vacuum-assisted filtration with the compact and lamellar structure (Figure 1b, Figure S2), and the assemblies exhibit a dense interlocked hybrid morphology facilitated by the hydrogen bonds (Figure 1c). The ANFs serve as cross-linkers to improve the interfacial concatenation between GO nanosheets, also to enlarge the interlayer space (Figure 1d). The nacre-like brick-and-mortar structure in which the hard GO nanosheets correspond to “brick” and soft ANFs correspond to “mortar”, contributes excellent chemical and physical robustness to the GO/ANF membranes. The membranes exhibit great stability in acidic and basic saline solutions for 1 month (Figure S3). Besides, ANFs in graphene-based membranes play a key role in the high-performance adhesive that withstands the enormous stresses at the interfaces. Such structure promotes the mechanical strength of the composite membrane with a tensile strength of 92.89 MPa and a toughness of 6.069 MJ m⁻³ (Figure 1e, Figure S4, and Table S1). These results demonstrate that the GO/ANF membranes as nanofluids are chemically and physically robust enough for ion transport.

**Figure 1.** Nanofluidic ion transport system fabricating with graphene oxide/aramid nanofibers (GO/ANF) composite membrane. (a) Schematic illustration of the ion transport across the composite membrane. The cross-linked structure can prevent the plane swelling and thus achieve a stable interlayer spacing. (b) Freestanding GO/ANF composite membrane obtained by vacuum-assisted filtration. (c) SEM image of the cross-sectional GO/ANF membrane with the lamellar structure. Scale bar, 1 μm. (d) XRD patterns for showing the changes in interlayer spacing of different membranes. (e) Comparison of stress–strain curves of the pristine GO and GO/ANF membranes.
Typical nanofluidic devices are fabricated using the GO/ANF composite membranes, and the ion transport behaviors of the membranes are determined by ionic current−voltage (I−V) measurements (Figure 2a and Figure S5). The I−V responses of the GO, ANF, and the composite membranes in 0.1 M KCl electrolytes all present linear ohmic behavior (Figure 2a). Since the ANFs can enlarge the interlayer spacing of the composite membrane by reducing the ion transport energy barrier, the ion transport through the composite membrane is promoted.32 The ion conductance shows a linear relationship with the temperature, in accordance with the Arrhenius behavior (Figure 2b). According to the Arrhenius equation

\[ G = G_0 e^{-E_a/R T} \]

where \( G \), \( G_0 \), and \( E_a \) are the conductance, the constant, and the activation energy, respectively. \( R \) and \( T \) are the gas constant and the temperature. The corresponding results indicate that the calculated energy barrier of K⁺ ion transport in the composite membrane is 6.62 kJ mol⁻¹, suggesting the improved ion transport.33 In addition, Figure 2c shows the temperature indeed enhances the ion activity and facilitates transmembrane transport behaviors.

To get insight into the ion transport behaviors of the GO/ANF membranes, different pH values and types of salt solutions are applied (Figure 2d,e). The ion conductance across the composite membranes can be tuned by changing the pH values of electrolytes (Figure 2d). Along with the increase of pH values, the dissociation of the carboxyl and hydroxyl groups generates much more charges on the surfaces of nanochannels (Figure 2d, Figure S6), reflecting in an increased ion conductance. Besides, ion conductance of the membrane decreases with increased hydration radius, following the size effect (Figure 2e). Since the composite membranes have an interlayer distance of 0.90 nm, calculated from the single diffraction peak (Figure 1d), while the hydrated diameters (\( D_H \)) of the cations range from 6.6 to 9.1 Å. Nevertheless, complete exclusion of large ions is not found, indicating that the hydration shell of the cations under confinement are compressed.35

The ion transport across the GO/ANF membranes also exhibits the surface-charge-governed features (Figure 2f), a prerequisite for the osmotic power generation under salinity gradient.34−38 When the concentration of the electrolyte is high, the ion transport agrees well with that in bulk solution. Once the concentration is below 1 M, the nanofluidic ionic conductance gradually deviates from the bulk value, suggesting that ion transport is governed by the surface charge. It can be

Figure 2. Nanofluidic ion transport of the GO/ANF membranes. (a) Current−voltage curves of the ANF, GO/ANF, and GO membranes recorded in 0.1 M KCl electrolyte. (b) Arrhenius plot of conductance through the composite membrane and the corresponding ion conductance curves versus temperature. (c) Conductance as a function of temperature across the composite membrane. (d) Curves of the ion conductance and surface charge densities at different pH values. The ion conductance of the GO/ANF membrane is in positive proportion to the surface charge density. (e) Ion conductance of various salt electrolytes (0.1 M) solutions. Metal cations are presented on the top x axis, and their hydrated diameter (\( D_H \)) values can be found on the bottom x axis. (f) Nanofluidic ion conductance and Debye length versus electrolyte concentration. When the concentration of the salt is below 1 M, the size of the nanochannel is less than the Debye length (orange dashed line), which means that the electronic double layers close to the channel walls are overlapped. The nanofluidic ion conductance (green dashed line) gradually deviates from the bulk value (pink line), indicating the surface-charge-governed ion transport.
ascribed to the size of the nanochannel being less than the Debye length of the low-concentration region, which means that the electrical double layers near the channel surface are overlapped. In this way, a unipolar confinement is formed with counterions. When the counterions pass along the channel, the co-ions are excluded, and thus selectively ion transport can be achieved.

To elucidate the physical mechanism of ion selectivity, current–voltage measurements, known as the drift–diffusion experiment, are conducted to measure ion flows actuated by the concentration gradient and the applied voltage (Figure 3a).40 The composite membranes were mounted between electrochemical testing reservoirs. 0.1 and 0.01 M salt solutions filled the feed and drain reservoirs, respectively. As the applied voltage ranges from −0.2 to 0.2 V, the electrophoretic component of the overall ionic flows contributed by the voltage difference, named as drift current. Due to the salt concentration gradient across the composite membrane, the diffusive current can be measured at zero applied voltage. The contribution of redox reaction (\(E_{\text{redox}}\)) deriving from unequal chloride concentration at electrodes and the pure electroosmotic contribution (\(E_{\text{diff}}\)) arising from the chemical potential generating the open-circuit voltage (\(V_{\text{OC}}\)). The cation/anion mobility ratio, \(\mu^+ / \mu^-\), can be evaluated by employing the Henderson equation,41

\[
\frac{\mu^+}{\mu^-} = \frac{z_+ \ln(\Delta) - z_+ FE_{\text{diff}}/RT}{z_+ \ln(\Delta) - z_- FE_{\text{diff}}/RT}
\]

where \(z\) is the valences (\(z_+,\) cations; \(z_-,\) anions); \(\Delta\) is the ratio of concentration and \(F\) represents the Faraday constant, and \(T = 298\) K. The surface charge contribution is negligible because the conductivity is estimated at high-concentration regime (\(C = 100\) mM). Therefore, the ion conductivity can be deduced as follows:

\[
\sigma \approx F(C_+ \mu^+ + C_- \mu^-)
\]

where \(C_+\) and \(C_-\) represent the concentrations of cations and anions, respectively. Several chloride solutions are chosen, with hydrated diameter increasing from 6.6 to 9.1 Å. The negatively charged \(\text{Cl}^-\) ions are suppressed compared to the positively charged ions across the composite membrane (Figure 3b). For an example, the mobility ratio of \(\text{K}^+/\text{Cl}^-\) is 10.93 while these ions have similar \(D_H\), deriving from that the negatively charged \(\text{Cl}^-\) ions are electrostatic repulsed by the membrane surface charge. Besides, the phenomena of size exclusion, it is observed that the mobility increases with decreasing \(D_H\), indicating that the cation mobility increases with decreasing \(D_H\) (Figure 3c). Thus, the electrostatic repulsion and activated size exclusion contributes to the ion selectively transport. The GO/ANF composite membranes also exhibit ultrahigh ionic selectivity, which is quantified by the calculated transference number (\(t_i\)) with the value up to 0.97 (Figure 3d). The electric double layer in the nanochannels completely overlaps due to the coupling of surface charge and space charge, which promotes the ionic selectivity of the membrane, hence favoring high-performance osmotic power conversion.
The GO/ANF composite membranes definitely exhibit the surface-charge-governed features, enabled by the ultrahigh ion selectivity, paving the way for osmotic power harvesting (Figure S7). The cations can pass through the channels, whereas the anions are electrostatically repulsed, generating net current across the membranes.\textsuperscript{36} A typical $I$-$V$ response under salt concentration gradient is shown in Figure 4a. The redox reaction occurs at the electrodes due to the unequal chloride concentration, and such deviation is subtracted by a blank test to obtain the pure electro-osmotic contribution. Thus, the diffusion potential ($E_{\text{diff}}$) and short current ($I_{\text{SC}}$) can be deduced from the intercept of the voltage axis and current axis, respectively.\textsuperscript{42} As the concentration gradient gradually increases from 10 to $3 \times 10^{5}$ (Figure 4b and Table S2), the related $E_{\text{diff}}$ and $I_{\text{SC}}$ increase with the salt concentration increase and the corresponding efficiency of osmotic energy conversion decreases to 9.7% (Figure S8). To evaluate the output power performance, a variable resistor ($R_{L}$) is used as the electronic load in the external circuits. The output power density, calculated by the equation $P = I^{2}R_{L}$, has a maximal value when the resistance of the membrane systems is comparable to the load resistance. By mimicking the seawater and river water condition, the current density shows a decrease when increasing the load resistance, and the peak output power density of up to 5.06 W/m$^2$ can be obtained, which surpasses most reported performances (Figure 4c and Figures S9 and S10). As for a hypersaline environment, the output power density could reach a peak value of 14.17 W/m$^2$, offering a promising avenue for converting the osmotic energy existing in the areas of salt lakes meeting the rivers. Furthermore, in order to output high voltages for powering the electronic devices, a tandem of the GO/ANF membranes stacks is fabricated.\textsuperscript{43,44} The output voltages increase linearly with cell numbers (Figure 4d). An ensemble of the composite membranes in series can offer a voltage up to 1.61 V, which can power the electronic devices (inset of Figure 4d).

**CONCLUSION**

In summary, we design GO-based composite membranes as nanofluidic devices for ion transport. The guest material is charged aramid nanofibers (ANFs), which serves as cross-linker to the GO nanosheets, promoting chemical and physical robustness of the membranes. The ion transport behaviors across the GO/ANF membranes exhibit the surface-charge-governed features, a prerequisite for the osmotic power generation under salinity gradient. The coupling of surface charge and space charge, enabled by the stabilization of $d$-spacing, is of key importance to realize high ion selectivity as well as derived high-performance osmotic power conversion up to 5.06 W/m$^2$. An ensemble of devices in series can achieve an output voltage up to 1.61 V, which can be used to power the electronic devices. The current work demonstrates the great promise of graphene-based membranes used in osmotic energy-harvesting generators for large-scale applications.
MATERIALS AND METHODS

Safety Statement. No unexpected or unusually high safety hazards were encountered.

Chemicals. Graphene oxide (thickness, 0.8–1.2 nm; diameter, 0.5–5 μm) was purchased from J&K Beijing Co., Ltd. Kevlar yarns were purchased from Dupont. Other chemicals were all analytical grade and purchased from Sigma-Aldrich, USA. Deionized water was prepared by Milli-Q system. All reagents were directly used without further purification.

Characterizations. SEM images were obtained by a field emission scanning electron microscopy instrument (Hitachi S-4800, Hitachi, Japan) with an acceleration voltage of 5 kV. The morphology of the ANF and GO were tested by transmission electron microscopy (JEOL, JEM-2100, Japan). The XRD patterns were conducted by a Bruker D8 Advance powder diffractometer with Cu Kα radiation (λ = 1.5418 Å) at 40 kV. The mechanical strength of the composite membrane was tested by a force meter M5-2 (ESM301, USA) in tensile mode with a tensile speed of 10 mm min⁻¹. Surface ζ potential was tested by the SurPASSSTM 3 Zeta Potential Analyzer.

Electrical Tests. The I–V measurements and osmotic energy conversion tests were performed with a Keithley 6487 semiconductor picoammeter (Keithley Instruments, Cleveland, OH, USA). And the testing membrane area was about 3 × 10⁴ μm², the same as in previous reports. ⁴⁵

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acscentsci.1c00633.

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REFERENCES

(1) Gouaux, E.; MacKinnon, R. Principles of Selective Ion Transport in Channels and Pumps. Science 2005, 310, 1461–1465.
(2) Xiao, K.; Jiang, L.; Antonietti, M. Ion transport in nanofluidic devices for energy harvesting. Joule 2019, 3, 2364–2380.
(3) Zhang, Z.; Wen, L.; Jiang, L. Bioinspired Smart Asymmetric Nanochannel Membranes. Chem. Soc. Rev. 2018, 47, 322–356.
(4) Hou, X.; Hu, Y.; Grinthal, A.; Khan, M.; Aizenberg, J. Liquid-based gating mechanism with tunable multiphase selectivity and antifouling behaviour. Nature 2015, 519, 70–3.
(5) Lu, J.; Zhang, H.; Hou, J.; Li, X.; Hu, X.; Hu, Y.; Easton, C. D.; Li, Q.; Sun, C.; Thornton, A. W.; et al. Efficient metal ion sieving in rectifying subnanochannels enabled by metal-organic frameworks. Nat. Mater. 2020, 19, 767–774.
(6) Acar, E. T.; Buchsbaum, S. F.; Combs, C.; Fornasiere, F.; Siwy, Z. S. Biomimetic Potassium-selective Nanopores. Sci. Adv. 2019, 5, eaav2568.
(7) Kim, S. J.; Ko, S. H.; Kang, K. H.; Han, J. Direct Seawater Desalination by Ion Concentration Polarization. Nat. Nanotechnol. 2010, 5, 297–301.
(8) Huang, S.; Romero-Ruiz, M.; Castell, O. K.; Bayley, H.; Wallace, M. I. High-throughput Optical Sensing of Nucleic Acids in a Nanopore Array. Nat. Nanotechnol. 2015, 10, 986–991.
(9) Schroeder, T. B. H.; Guha, A.; Lamoureux, A.; VanRenterghem, G.; Sept, D.; Shtein, M.; Yang, J.; Mayer, M. An electric-eel-inspired soft power source from stacked hydrogels. Nature 2017, 552, 214–218.

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(10) Shehzad, M. A.; Wang, Y.; Yasmin, A.; Ge, X.; He, Y.; Liang, X.; Zhu, Y.; Hu, M.; Xiao, X.; Ge, L.; et al. Biomimetic Nanocones that Enable High Ion Permeability. Angew. Chem., Int. Ed. 2019, 58, 12646–12654.

(11) Siria, A.; Poncharal, P.; Biance, A. L.; Fulrund, R.; Blase, X.; Purcell, S. T.; Bocquet, L. Giant Osmotic Energy Conversion Measured in a Single Transmembrane Boron Nitride Nanotube. Nature 2013, 494, 455–458.

(12) Feng, J.; Graf, M.; Liu, K.; Ovchinnikov, D.; Dumcenco, D.; Heirian, M.; Nandigana, V.; Aluru, N. R.; Kis, A.; Radenovic, A. Single-layer MoS2 Nanopores as Nanopower Generators. Nature 2016, 536, 197–200.

(13) Cheng, C.; Jiang, G.; Simon, G. P.; Liu, J. Z.; Li, D. Low-voltage Electrostatic Modulation of Ion Diffusion through Layered Graphene-based Nanoporous Membranes. Nat. Nanotechnol. 2018, 13, 685–690.

(14) Sapkota, B.; Liang, W.; VahidMohammadi, A.; Karnik, R.; Noy, A.; Wannimpla, M. High Permeability Sub-nanometre Sieve Composite MoS2 Membranes. Nat. Commun. 2020, 11, 2747.

(15) Esfandiar, A.; Radha, B.; Wang, F. C.; Yang, Q.; Hu, S.; Garaj, S.; Nair, R. R.; Geim, A. K.; Gopinadhan, K. Size Effect in Ion Transport through Angstrom-scale Slits. Science 2017, 358, 511–513.

(16) Raidongia, K.; Huang, J. Nanofluidic Ion Transport through Reconstructed Layered Materials. J. Am. Chem. Soc. 2012, 134, 16528–16531.

(17) Xin, W.; Lin, C.; Fu, L.; Kong, X.-Y.; Yang, L.; Qian, Y.; Zhu, C.; Zhang, Q.; Jiang, L.; Wen, L. Nacre-like mechanically robust heterojunction for lithium-ion extraction. Matter 2021, 4, 737–754.

(18) Wang, J.; Huang, L.; Wu, J.; Xiong, L.; Hu, Z.; Yu, H.; Li, T.; Zhou, J. Microwave Combustion for Rapidly Synthesizing Pore-size-controllable Porous Graphene. Adv. Funct. Mater. 2018, 28, 1800382.

(19) Wang, J.; Zhang, Z.; Zhu, J.; Tian, M.; Zheng, S.; Wang, F.; Wang, X.; Wang, L. Ion sieving by a two-dimensional TiC6T6 alginic lamellar membrane with stable interlayer spacing. Nat. Commun. 2020, 11, 3540.

(20) Hong, S.; Constans, C.; Surmani Martins, M. V.; Seow, Y. C.; Guevara Carrió, J. A.; Garaj, S. Scalable Graphene-Based Membranes for Ionic Sieving with Ultrafast Charge Selectivity. Nano Lett. 2017, 17, 728–732.

(21) Yeh, C.-N.; Raidongia, K.; Shao, J.; Yang, Q.-H.; Huang, J. On the origin of the stability of graphene oxide membranes in water. Nat. Chem. 2015, 7, 166–170.

(22) Chen, C.; Liu, D.; He, L.; Qiu, W.; Wang, J.; Razal, J. M.; Kotov, N. A.; Lei, W. Bio-inspired nanocomposite membranes for osmotic energy harvesting. Joule 2020, 4, 247–261.

(23) Chen, C.; Liu, D.; Yang, G.; Wang, J.; Wang, L.; Lei, W. Bioinspired Ultrastable Nanocomposite Membranes for Salinity Gradient Energy Harvesting from Organic Solutions. Adv. Energy Mater. 2013, 10, 1904098.

(24) Chen, C.; Liu, D.; Qiu, W.; Yang, G.; Wang, X.; Lei, W. Robust Membrane for Osmotic Energy Harvesting from Organic Solutions. ACS Appl. Mater. Interfaces 2020, 12, 52771–52778.

(25) Flouda, P.; Shah, S. A.; Lagoudas, D. C.; Green, M. J.; Lootens, J. L. Highly Multifunctional Dopamine-Functionalized Reduced Graphene Oxide Supercapacitors. Mater. Today 2019, 1, 1532–1546.

(26) Xiong, R.; Kim, H. S.; Zhang, L.; Korolovych, V. F.; Zhang, S.; Yingling, Y. G.; Tsukruk, V. V. Wrapping Nanocellulose Nets around Graphene Oxide Sheets. Angew. Chem., Int. Ed. 2018, 57, 8508–8513.

(27) Wu, Y.; Wang, F.; Li, X.; He, J.; Huang, Y. Fabrication of a graphene oxide/nanocellulose armed fiber composite membrane with improved hydrophilicity and mechanical strength via a fast-drying method using absolute ethanol as proton donor. J. Mater. Sci. 2018, 53, 16383–16392.

(28) Xin, W.; Xiao, H.; Kong, X. Y.; Chen, J.; Yang, L.; Niu, B.; Qian, Y.; Teng, Y.; Jiang, L.; Wen, L. Biomimetic Nacre-Like Silk-Crosslinked Membranes for Osmotic Energy Harvesting. ACS Nano 2020, 14, 9701–9710.