I. Possible factors affecting pressure-sensitive ion transport

To reveal the possible causes of this phenomenon, here we explored several potential issues and discussed the possibilities for leading to the experimental observations.

i. Polymer residual contamination on surface of graphene

Due to the hydrophobic nature of the graphene membrane, it is inevitable that there would be some polymer residuals on the surface of graphene. Through our TEM imaging of 156 devices, we have found that the graphene membrane is easily contaminated by hydrocarbon which may come from the PMMA residual most likely. Atomic force microscope (AFM) imaging results showed that one of the graphene sample contained a 0.8 nm thick residual PMMA film as shown in Figure S23. We have explored three methods of graphene transfer to obtain a relatively clean graphene film (Figure S1). Figure S2 showed the images of monolayer graphene on a 60 nm supporting pore with three transfer methods. We found that there were polymer residuals on the surface of graphene by the PMMA transfer method (Figure S2 A-C) and tape transfer method (Figure S2 G-I). The relatively clean graphene nanopores were obtained by PVA assisted transfer method, as shown in the TEM results (Figure S2 D-F).

ii. Wetting issues or nanobubbles formed in graphene nanopores

The incomplete wetting of graphene nanopores can increase the probability of forming nanobubbles in graphene nanopores, which may be a potential factor
influencing the pressure-sensitive ion transport. We increased the volume fraction of ethanol in KCl solution to tune the wettability of the solution in nanopores. Figure S19A showed the conductance of salt solution decreased with the increase of ethanol in the solution due to the change of conductivity. We did not observe considerable sensitivity changes with the change of the hydrophilicity of the solution (Figure S19C).

The current power spectral density (PSD) has been reported as a feature to identify the presence of nanobubbles in the nanopore. The noise may increase dramatically by 5 orders of magnitude due to the existence of nanobubbles(59). The PSD results of our data showed that the low frequency current noise did not change considerably under different applied pressures (Figure S20), which suggested that there were no such issues for the current case.

iii. Deformation of monolayer graphene membranes

Graphene is known to be one of the strongest materials and has been mechanically tested to various deformations(36). We designed AFM indentation experiments to examine the mechanics of suspended graphene membranes. An AFM (Molecular Vista VistaScope) equipped with air temperature controller for minimizing drift was used for AFM imaging and indentation experiment(36). In the indentation experiment, the tip was first lifted to 20 nm above the sample and then moved 50 nm downwards to record an indentation force curve (Figure S21). This process includes three loading stages as shown in Figure S23C. AFM probe spring constants were
calibrated using reported calibration method (60). The results revealed the Young’s modulus of graphene from 10 to 145 N/m (Figure S21C), agreeing with reported results obtained for CVD graphene membranes (36).

The two-dimensional graphene membrane is crumpled because of the inherent flexural phonons and static wrinkling from fabrication process. Interferometric profilometry measurements suggest the intrinsic crumpling effect soften the Young’s modulus of graphene to 10 N/m before reaching the values for flat graphene (61). We scanned the topography of graphene membrane in tapping mode using cantilever with $k = 40$ N/m (MikroMasch NSC15/AlBS). This crumpling effect was confirmed in our case by AFM imaging of static wrinkles (Figure S23), indicating that the crumpling allowed for graphene deformation possibilities. To know the role of membrane deformation for ionic conduction, we then developed computational models using MD to account for contributions from membrane deformations. However, the MD simulations showed that the deformation was not the main reason for the highly pressure-sensitive ion transport (Figure S24). First, the deformation of graphene membrane (diameter of supporting pore is 60 nm) occurs at two orders of higher force than the pressure force used in the conductance experiments (Figure S24D). Second, even if we assume the deformation, the simulation results show no enhancement of conductance (Figure S24E and F). We thus eliminated this possibility.
Figure S1. Methods used for graphene transfer. (A) PMMA based transfer method. (B) PVA assisted transfer method. (C) Tape assisted transfer method.
Figure S2. Typical TEM imaging of monolayer graphene samples prepared using different transfer methods. (A) Monolayer graphene with PMMA residuals on a 60 nm supporting pore before drilling, transferred by PMMA method. (B) A graphene nanopore with PMMA residuals on a 60 nm supporting pore after drilling. (D) Monolayer graphene without PMMA residual on a 60 nm supporting pore before drilling, transferred by PVA assisted method. (E) A graphene nanopore without PMMA residual on a 60 nm supporting pore after drilling. (G) Monolayer graphene with organic residuals on a 60 nm supporting pore before drilling, transferred by tape method. (H) A graphene nanopore with organic residuals on a 60 nm supporting pore after drilling. (C, F, I) Diffraction patterns of monolayer graphene from the three transfer methods.
Figure S3. Electron diffraction pattern of monolayer graphene membrane under different tilt angles in TEM. (A, C) Diffraction patterns of graphene monolayer with tilt angles of 0 and 15°. (B) Diffraction intensity of graphene monolayer without tilt angle along the turquoise dotted line in A. Outer and inner diffraction spots are indicated with 1 and 2, respectively. (D) Diffraction intensity of graphene monolayer with tilt angle of 15° along the turquoise dotted line in C. Outer and inner diffraction spots are indicated with 3 and 4, respectively. As we can see, the intensity of inner dots (2, 4) was higher than that of outer dots (1, 3) before and after tilting 15°. This result was consistent with the literature(62). With the change of tilt angles, the diffraction intensities have a weak monotonic variation in monolayer graphene, which is a unique feature and can be used to identify the monolayer graphene in TEM.
Figure S4. TEM images of monolayer graphene nanopores with different pore sizes. (A-F) Graphene nanopores with size from 2.6 nm to 16.4 nm.
Figure S5. Ionic current time trace of graphene nanopore Device #8 showing strong pressure-sensitive conductance response. (A) Ionic current time trace (orange) under a step-like pressure (blue) at 100 mV bias. (B) Ionic current time trace under a step-like pressure at -100 mV bias. The pressure is gradually applied to 2.5 bar with a step of 0.25 bar for 5 s intervals. The diameter of the graphene nanopore is 1.7 nm. The measurements correspond to 1 M KCl solution.
Figure S6. Ionic current time trace graphene nanopore Device #9 showing weak pressure-sensitive conductance response. (A) Ionic current time trace (green) under a step-like pressure (blue) at 100 mV bias. (B) Ionic current time trace under a step-like pressure at -100 mV bias. The pressure is gradually applied to 2 bar with a step of 0.25 bar for 5 s intervals. The diameter of the graphene nanopore is 1.8 nm. The measurements correspond to 1 M KCl solution.
Figure S7. Graphene nanopores Device #10 under different voltages and a pressure difference of 2 bar. Ionic current time trace under a stepwise voltage in 1 M KCl solution at a fixed pressure (2 bar). The voltage is gradually applied in the range of 0-100 mV with a step of 50 mV.
Figure S8. Ionic current time trace of graphene nanopore Device #11. Ionic current time trace under 100 mV voltage at a fixed pressure (2 bar) in 1 M KCl solution. The pressure (2 bar) is applied uninterruptedly to the device for more than one hour. The diameter of Device #11 is 3.8 nm.
Figure S9. Current–voltage curves for the same device (Device #7-a). Current–voltage curves are tested before (A) and (B) after the pressure measurements in 1 M KCl solution.
Figure S10. Diverse pressure-sensitive conductance responses of graphene nanopores with similar pore sizes. (A) Ionic conductance as a function of applied pressure in 1 M KCl solution (blue) and 0.1 M KCl solution (orange) for two graphene nanopores devices with similar pore sizes. (B) Pressure-sensitive conductance change ratios as a function of applied pressure in 1 M KCl solution for two graphene nanopores devices with similar pore sizes. The diameter of Device #12 and Device #13 are 6.7 nm and 7.0 nm, respectively.
Figure S11. Pressure-sensitive conductance for KCl solutions with different concentrations. (A) Ionic conductance as a function of applied pressure in different concentrations of KCl solution for Device #4 in Figure 3B. (B) Ionic conductance as a function of applied pressure in different concentrations of KCl solution for Device #5 in Figure 3E. The diameter of Device #4 and Device #5 are 7.5 nm and 7.2 nm, respectively.
Figure S12. Pressure-sensitive conductance of graphene nanopore Device #14 for different salt solutions. (A) Ionic conductance as a function of applied pressure in different salt solutions. (B) Histograms of pressure-sensitivities for different salt solutions at different applied pressures. The diameter of Device #14 is 2.2 nm.
Figure S13. Streaming current of graphene nanopore Device #14 for different salt solutions. Streaming current as a function of applied pressure in different salt solutions at 0 mV. The diameter of Device #14 is 2.2 nm.
Figure S14. Pressure-sensitive conductance for 1 M KCl solution under different pH. (A) Ionic conductance as a function of applied pressure in different pH values of 1 M KCl solution for Device #3 in Figure 3A. (B) Ionic conductance as a function of applied pressure in different pH values of 1 M KCl solution for Device #5 in Figure 3D. The diameter of Device #3 and Device #5 are 7.1 nm and 7.2 nm, respectively.
Figure S15. Streaming current for 1 M KCl solution under different pH. (A-C) Streaming current as a function of applied pressure in different pH values of 1 M KCl solution for Device #3 in Figure 3A. (A: 0 mV, B: 50 mV, C: 100 mV). (D-E) Streaming current as a function of applied pressure in different pH values of 1 M KCl solution for Device #5 in Figure 3D (D: 0 mV, E: 50 mV, F: 100 mV). The diameter of Device #3 and Device #5 are 7.1 nm and 7.2 nm, respectively.
Figure S16. Pressure-sensitive conductance change ratios $G_{sr}/G_0$ under pH control (pH=3.0, 5.5 and 8.9) for a 2.4 nm graphene nanopore Device #15. The concentration of KCl solution is 0.1 M.
Figure S17. Pressure-sensitive conductance in 1 M KCl solution for pores with different sizes. (A) Ionic conductance as a function of applied pressure for devices with different pore sizes in Figure 3C. (B) Ionic conductance as a function of applied pressure for devices with different pore sizes in Figure 3F. The measurements are in 1 M KCl solution.
Figure S18. Surface charge measurements for graphene nanopores (Device #16 and Device #17) with different pore sizes. (A) Current–voltage response of graphene nanopores with different pore sizes (red, 5.9 nm; blue, 7.4 nm) in 1 M KCl solution. (B) Conductance as a function of KCl bulk conductivity $\kappa_b$. The dashed lines denote the results by fitting to extract the surface conductance $\kappa_s$. The values are 0.934 nS and 1.50 nS for the two pores, respectively. Surface charge density values are estimated by the reported theories (63, 64). (C) The surface charge as a function of salt concentration for the two pores. We calculate the surface charge at each measurement point by using the values of $\kappa_s$. Surface charge density values are estimated by the reported theories (63, 64). (D) Conductance-concentration curves in log scale. The dashed lines represent the theory suggested by Noh and Aluru(58) with $\alpha=0.12$ and 0.13 for the nanopores with 5.9 nm and 7.4 nm diameters respectively. Inset in D: zoomed conductance values at the lower concentrations.
Figure S19. Pressure-sensitive conductance of graphene nanopore Device #18 in 1 M KCl solution with different amounts of ethanol. (A) Current–voltage curve in 1 M KCl solutions with different amounts of ethanol. (B) Ionic conductance as a function of applied pressure in 1 M KCl solutions with different amounts of ethanol. (C) Histograms for KCl solutions with different amounts of ethanol exhibiting pressure-sensitive conductance change ratios at different applied pressures. The diameter of Device #18 is 6.6 nm.
Figure S20. Power spectral density of the current traces under different applied pressures for Device #19. (A, C) Ionic current time trace under stepwise pressure at a voltage bias of 100 mV and -100 mV, respectively. (B, D) Ionic current power spectral density as a function of frequency under different applied pressures at a voltage bias of 100 mV and -100 mV, respectively. The dashed lines show the fitting of low frequency noise under different applied pressures (blue: 0 bar; red: 1 bar; black: 2 bar). The measurements correspond to 0.1 M KCl solution. The diameter of Device #19 is 7.3 nm. The recording frequency is 100 kHz.
Figure S21. Indentation of monolayer graphene membranes by AFM for supporting pores with different diameters. (A) AFM image of monolayer graphene membrane suspended over a supporting pore. (B) Height profile along the blue dotted line in A. The step height at the edge of the membrane is approximately 8 nm. (C) Fitted indentation curves of supporting pores with different diameters and histogram of young’s modulus (inset).
Figure S22. Deformation force curve of monolayer graphene membranes. (A) Amplitude modulation AFM image of a 12 × 12 μm SiNx window, 20 nm in thickness. Monolayer graphene was suspended on the window, partially covered by PMMA. Pore 1 is of 300 nm in diameter and free of PMMA and pore 2 is of similar diameter as pore 1 and with PMMA covering the graphene membrane. (B, C) Zoomed AFM images of pore 1 and 2, respectively. Supporting pores are denoted by dashed circles. (D) Height profile along the blue dotted line in B. The step height is about 0.8 nm, indicating the thickness of PMMA layer is about 0.8 nm. (E) Indentation curves of pore 1 and pore 2 suggest that extra PMMA layer enhances the stiffness of graphene layer.
Figure S23. The intrinsic crumpling of monolayer graphene membrane. (A, C) AFM topography of graphene membrane suspended on a 450 nm diameter supporting pore and 80 nm diameter supporting pore, respectively. (B, D) Reconstructed surface image of the intrinsic crumpling in the graphene membrane suspended on corresponding supporting pores. The results are consistent with previous work (61). (E) Illustrations of three stages in the indentation curve. Before AFM tip reaches the surface of graphene membrane, static wrinkle can be distinguished from the topography; when the tip goes down and touches the membrane, the wrinkles are first flattened, where small Young’ s modulus is measured; with the tip going downwards another several nanometers, large Young’ s modulus is measured.
Figure S24. Effect of mechanical deformation on the ion transport in graphene nanopore. (A) top view of the system (B) initial side view (C) side view after a pressure is applied. (D) Maximum deformation versus applied pressure on the membrane. The dashed line is the bulge theory\(65\), \(\delta_{\text{max}} = \frac{3}{16EL} \Delta p R^4 (1-\nu)^{\frac{1}{2}}\), where \(E\), \(L\), \(\nu\) are the elastic modulus, the thickness of membrane, and the Poisson’s ratio, respectively. For the curve, we use \(E=1\) TPa, \(L=0.34\) nm, \(\nu=0.16\) \(66\). The deformation due to intrinsic crumpling (0.45 nm) is subtracted from MD data. (E) Current over time with and without mechanical deformation in \(D=2.6\) nm nanopore. (F) Conductance in deformed graphene normalized by conductance in flat graphene over various degrees of deformation.
Figure S25. Pressure-sensitive ion current demonstrated by MD simulation. (A) Current-voltage response with and without an applied pressure. (B) Current-pressure response with and without surface charge in the graphene membrane at 3 V. (C) Current-pressure response with and without an applied voltage. For all data, nanopore with accessible diameter of 2.6 nm is considered in the simulation.
Figure S26. PNP-NS calculations for pressure-sensitive ion transport. (A) Geometry considered in PNP-NS calculation and boundary conditions. The accessible diameter ($2R$) is 2.32 nm. Colors represent net charge distributions: blue, red, and green indicate positive net charge, negative net charge, and zero net charge. (B) Net charge distribution along the cutting plane for different applied voltages. (C) Total net charge on one side of the membrane surface ($\varepsilon_m = 3$ and no pore) and the parallel-plate capacitor model.
Figure S27. Schematics of nanopore membrane with electric permittivity subjected to pressure-driven and electrically driven flow. The blue shaded area represents net positive layer and the red shaded area represents net negative layer due to the capacitive mechanism.
Figure S28. Pressure-sensitive ion current from PNP-NS calculation (symbols) and theory (dashed lines). Current-voltage curve in (A) Linear regime for various applied pressures ($\epsilon_m = 3$). (B) Non-linear regime for various dielectric permittivity of membrane ($\Delta \rho = 0$). (C) Current-pressure response for an uncharged nanopore. (D) Current-pressure response for a charged nanopore. For all data, nanopore with accessible diameter of 2.32 nm is considered.
Figure S29. Estimated permeation coefficient and slip length. (A) Permeation coefficient over pore radius. Permeation coefficients are estimated from equation (11) and parameters in Table S2. Orange line indicates the permeation coefficient from the Hagen-Poiseuille (HP) equation with Dagan’s hydrodynamic pressure drop length(67):

\[ P_{o}^{\text{HP-Dagan}} = \frac{\pi R^4}{8\eta} \frac{1}{L + \frac{3\pi}{8} R} \], where \( \eta \) is viscosity in unit Pa·s. Red line is corrected HP (CHP) suggested by Heiranian and Aluru(68) \( P_{o}^{\text{CHP}} = \frac{\pi \left( R^4 + 4R^3 \eta' \lambda' \right)}{8\eta'} \frac{1}{L + \frac{3\pi}{8} R} \), where \( \eta' \) is corrected viscosity and \( \lambda' \) is corrected friction coefficient. (B) Slip lengths (\( \eta/\lambda \)) extracted from CHP over pore radius.
Figure S30. Comparison of streaming currents for devices with different pressure-sensitive conductance responses under zero bias. (A) Streaming current as a function of applied pressure for a 1.7 nm nanopore (response: 177.9% at 2 bar) and a 1.8 nm nanopore (response: 3.7% at 2 bar). (B) Streaming current as a function of applied pressure for a 2.2 nm nanopore (response: 141.3% at 2 bar) and a 2.4 nm nanopore (response: 2.2% at 2 bar). The detailed response under different applied pressures is shown in Figure 3C and Figure 3F. The measurements correspond to 1 M KCl solution. Our model suggests that slip length and dielectric modulation may be reasons for the pressure-sensitive conductance in graphene nanopores. Capacitive charging due to the intrinsic dielectric constant of graphene should not occur at 0 mV. The electroosmotic mobility $\mu_{eo}$ for device with strong response is much higher than that of device with weak response at 0 mV as shown in A and B, which suggests that the slippage contribution is the major cause of the phenomenon.
Figure S31. Comparisons of pressure-sensitive conductance for devices with different substrates. (A) Profiles of SiNₓ substrate and SiO₂ substrate for suspending the monolayer graphene nanopores. (B) Pressure-sensitive conductance change ratios for graphene nanopores with SiNₓ substrates (10 nanopore devices, Device #20-29) and SiO₂ (10 nanopore devices, Device #30*-39*) substrates. The SiO₂ substrate was prepared by sputtering a 60 nm thick SiO₂ membrane on the SiNₓ substrate. We applied FIB drilling to prepare a number of supporting pores with a diameter of 150 nm (See Methods for details).
Figure S32. Comparisons of pressure-sensitive conductance for graphene devices with different thickness. Sensitivity for monolayer (Device #20-29) and three-layers (Device #40*-48*) graphene nanopores on SiN$_x$ substrates at 2 bar in 1 M KCl solution.
Figure S33. Comparisons of pressure-sensitive ion transport effect for graphene nanopores and MoS$_2$ nanopores. (A) Current–voltage curves measured as a function of applied pressure in a 2.1 nm MoS$_2$ nanopore Device #49* in 1 M KCl solution. (B) Pressure-sensitive conductance change ratios for monolayer graphene (Device #20-29) and monolayer MoS$_2$ nanopores (Device #50*-59*) on SiN$_x$ substrates at 2 bar in 1 M KCl solution. (C) Streaming current comparisons for monolayer graphene nanopores (Device #60-63) and monolayer MoS$_2$ nanopores (Device #50*-53*) on SiN$_x$ substrates under 0 mV at 1 bar and 2 bar in 1 M KCl solution.
### III. SM Table

#### Table S1. Radii of hydrated ions. The data are from two references (69, 70).

| Ion       | K⁺   | Li⁺   | Na⁺   | Mg²⁺  | Bmim⁺ | Cl⁻  |
|-----------|------|-------|-------|-------|-------|------|
| Hydrated radii | 0.33 nm | 0.38 nm | 0.36 nm | 0.43 nm |       | 0.33 nm |

*Length: 1.10 nm Width: 0.58 nm

#### Table S2. Parameters used in the fitting of experimental data.

| Parameters | Expression/Value | Comments |
|------------|------------------|----------|
| \(\mu^+\) | \(\mu_{\text{pore}}^+ = \left( \frac{0.004104 \text{ V} \cdot \text{s} \cdot \text{m}^{-1}}{R} + \frac{1}{\mu_{\text{bulk}}^+} \right)\) | Suk and Aluru (71) |
| \(\mu^-\) | \(\mu_{\text{pore}}^- = \left( \frac{0.006215 \text{ V} \cdot \text{s} \cdot \text{m}^{-1}}{R} + \frac{1}{\mu_{\text{bulk}}^-} \right)\) | Suk and Aluru (71) |
| \(\mu_{\text{bulk}}^+\) | \(5.6388 \times 10^{-8} \text{ m}^2\cdot\text{s}^{-1}\cdot\text{V}^{-1}\) | Bulk mobility at 1M |
| \(\mu_{\text{bulk}}^-\) | \(5.8608 \times 10^{-8} \text{ m}^2\cdot\text{s}^{-1}\cdot\text{V}^{-1}\) | Bulk mobility at 1M |
| \(\varepsilon_m\) | 3 | From Santos et al. (56) |
| \(\alpha\) | \(\frac{\pi}{2}\) | Maxwell-Hall access resistance (57) |
| \(\beta_{\text{dep}}\) | 0.1 | From MD simulation at 1M |
| \(\beta_{\text{ap}}\) | 6.4 | From MD simulation at 1M |
| \(p_Q\) | Fitting parameter | By fitting \(G_{e,Ap}\) |
| \(\tilde{\psi}_p\) | -0.3 mV ~ -0.05 mV | Fitting classical streaming current (\(\Delta \phi = 0\)) |
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