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Arghyadeep Paul and N. R. Aluru

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# Surge-protected mechanical amplifier of ion transport devised using a bipolar nanopore with broken symmetry

Arghyadeep Paul<sup>1</sup> and N. R. Aluru<sup>2</sup>

<sup>1</sup>*Department of Mechanical Science and Engineering, Beckman Institute for Advanced Science and Technology, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801, United States*

<sup>2</sup>*Walker Department of Mechanical Engineering, Oden Institute for Computational Engineering and Sciences, The University of Texas at Austin, Austin, Texas 78712, United States*

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## ABSTRACT

Electrokinetic ion transport under coupled external electric field and pressure drops has recently exhibited giant, but monotonous, mechanical amplifications of ionic current in some nanochannels. While such effects can be exploited to develop novel pressure-sensitive ionic circuits, they can also yield large undesirable current surges if the control parameter, i.e., the pressure drop, overshoots. To overcome this critical issue, we design a nanopore that displays a similar mechanical activation of ionic current, but up to a limited magnitude of pressure, after which current ceases to grow. We integrate a bipolar surface charge distribution with an asymmetric channel shape in a nanopore to alter the effect of an applied pressure drop on the quasi-static distribution of ions in the pore, which directs how ionic conductance varies with pressure, in a way that yields the desired current-pressure response. Via such steps, probed using numerical simulations, we devise a funnel-shaped nanopore with a negatively charged conical mouth and a positively charged elongated cylindrical stem, that acts as the intended ‘surge-protected’ mechanical amplifier of ion transport. It yields a substantial pressure-driven growth in ionic current (by factors of  $\sim 4$ ) up to a limited intensity of pressure drop exerted (along a constant external electric field) towards the stem, beyond which current begins to fall. Besides exhibiting this hitherto undetected electrokinetic behavior, our study implements and paves the way to an innovative ‘function to design’ approach in the field of electrokinetics that can help build improved nanofluidic devices with desired advanced ion transport functionalities.

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Lately, in the field of electrokinetics, the simultaneous exertion of an electric field and pressure drop across nanofluidic channels has displayed a plethora of intriguing ion transport behaviors [1–8]. These revelations demonstrate the potential to mimic, by bearing resemblances to, the intricate ion conduction functionalities of mechanosensitive biological ion channels [9–11] across artificial

nanoscale conduits. Such pressure-sensitive electrokinetic ion transport characteristics, due to their ability to further enhance the regulation of ionic flow at nanoscale, can be exploited to formulate novel nanofluidic devices with improved capabilities in biosensing [12–18], drug delivery [18,19], and ionic sieving [20–24], to be operated with a pressure differential (as an additional flow control parameter) besides the (conventionally applied) voltage.

Under coexisting voltage and pressure drops, while single-digit carbon nanotubes [1] (graphene nanopores [3]) manifest a symmetric and nearly quadratic (linear) activation of ionic current with the applied pressure, angstrom-scale graphene slits [2] exhibit a quadratic voltage-gated growth of the pressure-initiated streaming current. Such studies were then extended to asymmetric nanopores. In non-polarizable conical nanopores [4–6], bidirectional pressure drops nullify the voltage-driven ionic current rectification (ICR) [4]. In contrast, a pressure drop along a conical nanopore embedded in a polarizable dielectric membrane [7] promotes ICR, by anomalously impeding ionic current if applied along the external electric field. Meanwhile, imparting a bipolar surface charge distribution to a symmetrically shaped nanopore, via oppositely charged sections on the pore surface [8], yields a pressure-driven amplification and suppression of ion transport at opposite voltages. However, the elements of asymmetric channel shape and surface charge inhomogeneity are yet to be combined in a nanopore when investigating nanoscale ion transport under coupled voltage and pressure drops, which presents a viable next step in the field to unearth new advanced routes towards a more robust, sensitive, and tunable electromechanical manipulation of ion transport.

As mixing a heterogeneous surface charge distribution with an irregular channel shape can lead to endless possible nanopore designs, formulating the proposed analysis can get overcomplicated, unless it is aimed to extract a specific (useful) ion transport characteristic in such a nanopore that is thus designed accordingly. One such challenge is to build a ‘surge-protected’ mechanical amplifier of ion transport. A current surge in ionic circuits due to an overshooting control parameter, e.g., a voltage or pressure drop, leads to excessive Joule heating [25]. The resulting temperature jump in the device-channel can yield unwanted surface reactions [26–28], vapor bubble formation [25], and affect the solution pH [29], altering the channel’s size, surface chemistry, surface charge [30], and thus its ion transport properties. Such current surges can be averted in voltage-driven ion transport via, e.g., a negative differential ionic resistance above a threshold voltage, detected in several past studies [31–38]. Meanwhile, applying a simultaneous pressure drop, next to a voltage, has revealed large mechanical amplifications of ionic current across single-digit carbon nanotubes [1], graphene

nanopores [3], and bipolar nanopores [8]. While such phenomena offer the potential to add an extra layer of mechanical tunability and control over voltage-regulated ionic flow in nanofluidic devices, the monotonous nature of the pressure-induced growth of ionic current they display can yield giant current surges if the pressure drop overshoots beyond control. However, analogous mechanisms to shield ionic circuits from a current upsurge for such mechanical activation of ion transport remain unexplored. In this study, we consequently integrate an inhomogeneous surface charge distribution with an asymmetric channel geometry in a nanopore, as a potential strategy, to design a mechanical amplifier of (voltage-initiated) ion transport with an added safeguard against current surges in case of overshooting stimuli. We devise the nanopore to yield a significant amplification of ionic current with a pressure drop applied along a specific direction of the pore-axis (under a constant voltage), but only up to a certain intensity of applied pressure, beyond which the current displays either an ideal stagnation or a more practical gradual suppression instead of a continued growth. This desired non-monotonous current-pressure (I-P) response is sketched in fig. 1(a).

A pressure drop modulates the voltage-initiated ionic flux in an electrolyte-filled nanopore by distorting the quasi-static distribution of ions across the pore via hydrodynamic flow, which alters the ionic conductance [7,8]. We use this fundamental principle here to attain our goal.

For the design operation, we use a suitable model of a nanopore, embedded in a membrane of dielectric constant  $\epsilon_m$ , depicted in fig. 1(b). It is axisymmetric about the centerline DE and its centre coincides with the origin of the cylindrical coordinates ( $r, z$ ). Consequently, the half-section space within the segment CDEF is adequate to numerically simulate ion transport across the pore. The membrane separates two electrolyte reservoirs containing a KCl solution of bulk salt concentration  $c_0$ , density  $\rho = 1000 \text{ kg/m}^3$ , viscosity  $\eta = 1 \text{ mPa-s}$ , and dielectric constant  $\epsilon_f = 80$ , with  $D(K^+) = 1.95 \times 10^{-9} \text{ m}^2/\text{s}$  and  $D(Cl^-) = 2.03 \times 10^{-9} \text{ m}^2/\text{s}$  as the diffusion coefficients of the electrolyte ions. We assume a constant system temperature  $T = 298 \text{ K}$ . The surfaces AI and AB (HI and HG) carry a negative (positive) inherent surface charge density of  $\sigma_1$  ( $\sigma_2$ ), prior to the dielectric polarization of the membrane under an external electric field [7,8,39,40] which induces additional charges on the surfaces. The consideration of this widely analyzed bipolar surface charge distribution [8,41–44] is justified later. The pore width is dictated by the tip-radius  $R_t$ . The angle of inclination of the pore surface AI (HI) to the radial ‘ $r$ ’ direction is  $\theta_1$  ( $\theta_2$ ). Taking  $\theta_1 = \theta_2 = 90^\circ$ ,  $\theta_1 < \theta_2$  with  $\theta_2 = 90^\circ$ ,  $\theta_1 = \theta_2 < 90^\circ$ , and  $\theta_1 + \theta_2 = 180^\circ$  with  $\theta_1 \neq \theta_2$  yields a cylindrical, funnel-shaped, hourglass-shaped, and conical nanopore, respectively, indicating the geometric adaptability of our model. The

depth of AI (HI) in the axial ‘z’ direction is  $h_1$  ( $h_2$ ). A voltage  $\Delta V$  and pressure drop  $\Delta P$  are exerted in the negative-z direction, along which ionic current is considered positive.

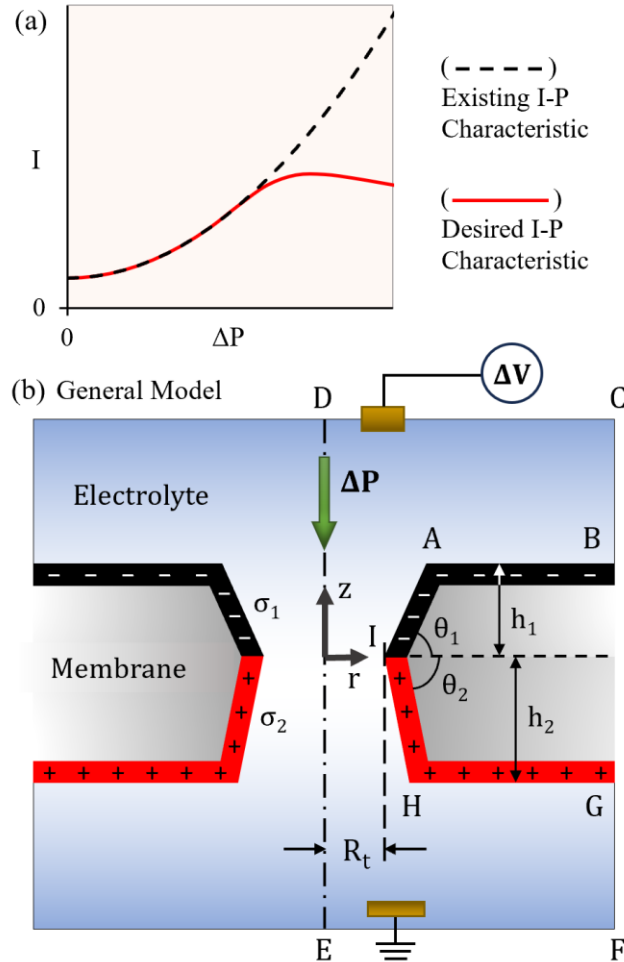


**Figure 1:** (a) The existing and the desired current-pressure (I-P) characteristics are sketched by the dashed (black) and solid (red) lines, respectively. (b) System schematic.

We use a continuum model based on the Poisson, Nernst-Planck, and Navier-Stokes equations to numerically simulate ionic flow in the pore via the finite-element solver COMSOL Multiphysics. The governing equations with their boundary conditions across the two-dimensional axisymmetric computational space within the segment CDEF are detailed in the supplemental material (SM) [45]. The fluid (membrane) domain, enclosed within the segment ABCDEFGHI (BAIHG), is discretized into triangular elements. The mesh is refined sufficiently near the pore tip, denoted by the point ‘I’ in fig. 1(b), to capture the high gradients of ionic concentration and electric potential across the electrical double layers (EDL) [46] of ions formed over the charged nanopore surfaces. We ensure that the numerically measured ionic current ( $I$ ), described in eq. (S11) of SM [45], is precise, grid-

independent, and unaffected by the (adequately enlarged) computational space dimensions. We also validate our computational model in fig. S1 of SM [45], using it to replicate the pressure-modulated ionic conductance across nanochannels from past experiments [1,3,4] and numerical studies [7,8].

We first select a tactical set of the parameters  $\sigma_1$ ,  $\sigma_2$ ,  $R_t$ ,  $h_1$ ,  $\theta_1$ ,  $h_2$ , and  $\theta_2$ . Subsequently, they are altered selectively to modify the deformation of the quasi-static distribution of EDL ions across the pore by an applied pressure drop, which determines the variation of ionic conductance with  $\Delta P$ , in a way necessary to make the current initially grow with  $\Delta P$  up to a restricted intensity of pressure applied in a specific direction and then reduce. As we try to attain this non-monotonic I-P function for a unidirectional  $\Delta P$ , e.g., say in the positive- $\Delta P$  regime as exemplified in fig. 1(a), we are not directly interested in the resultant I-P response in the opposite (i.e., negative) polarity of  $\Delta P$ .

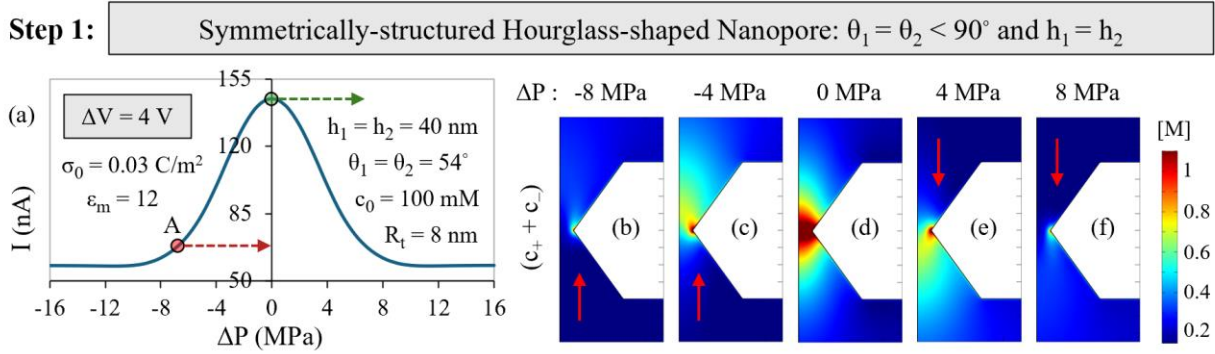


**Figure 2:** (a) The current-pressure (I-P) curve of a bipolar symmetrically shaped hourglass-like nanopore is plotted at  $\Delta V = 4$  V,  $\sigma_0 = 0.03$  C/m<sup>2</sup>,  $\epsilon_m = 12$ ,  $R_t = 8$  nm,  $\theta_1 = \theta_2 = 54^\circ$ ,  $h_1 = h_2 = 40$  nm, and  $c_0 = 100$  mM. (b)-(f) 2D contours of total ionic concentration ( $c_+ + c_-$ ) across the channel are plotted with varying pressure drop  $\Delta P$ .  $c_+$  ( $c_-$ ) is the concentration of  $K^+$  ( $Cl^-$ ) ions.

**Step 1:** We consider a bipolar symmetrically shaped hourglass-like pore ( $\theta_1 = \theta_2 < 90^\circ$  and  $h_1 = h_2$ ) with  $|\sigma_1| = |\sigma_2| = \sigma_0$ , from our past work [8], under a positive  $\Delta V$ . At  $\Delta P = 0$ , the negatively (positively) charged surface AI (HI) attracts  $K^+$  ( $Cl^-$ ) ions which, under a downward electric field, migrate downwards (upwards), elevating the density of both EDL ions throughout the channel. This ionic enrichment in the pore at  $\Delta P = 0$  boosts the conductance and  $I(\Delta P = 0)$ . A positive or negative  $\Delta P$  pushes the quasi-static position of the ions accumulated in the pore to its exterior [see figs. 2(b)-2(f)] where the electric field intensity is negligible [8]. Thus,  $\Delta P$  lowers the conductance to suppress  $I$  in fig. 2(a). This I-P response is a crucial starting point, as being able to morph its shape by shifting its maxima (denoted by the green pointer in fig. 2(a)) to a specific direction, e.g., the positive  $\Delta P$ -regime, achieves our target (partially) by making the current grow from  $\Delta P = 0$  to a certain intensity

of positive  $\Delta P$  and then dip with higher magnitudes of  $\Delta P$ , as formulated in fig. 1(a). It also justifies the prior consideration of the bipolar surface charge profile. However, this may not be adequate to develop an effective mechanical amplifier of ion transport due to a low factor of current activation, from  $\Delta P = 0$  to the  $\Delta P$  value where the new maxima occurs. To resolve the issue, we aim to induce a low conductance state at  $\Delta P = 0$  by effectively shifting such a condition occurring at a negative  $\Delta P$ , say at the point 'A' marked by the red pointer in fig. 2(a), towards  $\Delta P = 0$ .

Meanwhile, the I-P response at  $\Delta V < 0$ , manifesting a pressure-induced current amplification, cannot be similarly transformed into the desired I-P characteristic of fig. 1(a), as illustrated in fig. S2 of SM [45]. Therefore, we consider a positive  $\Delta V$  throughout the remaining analysis.

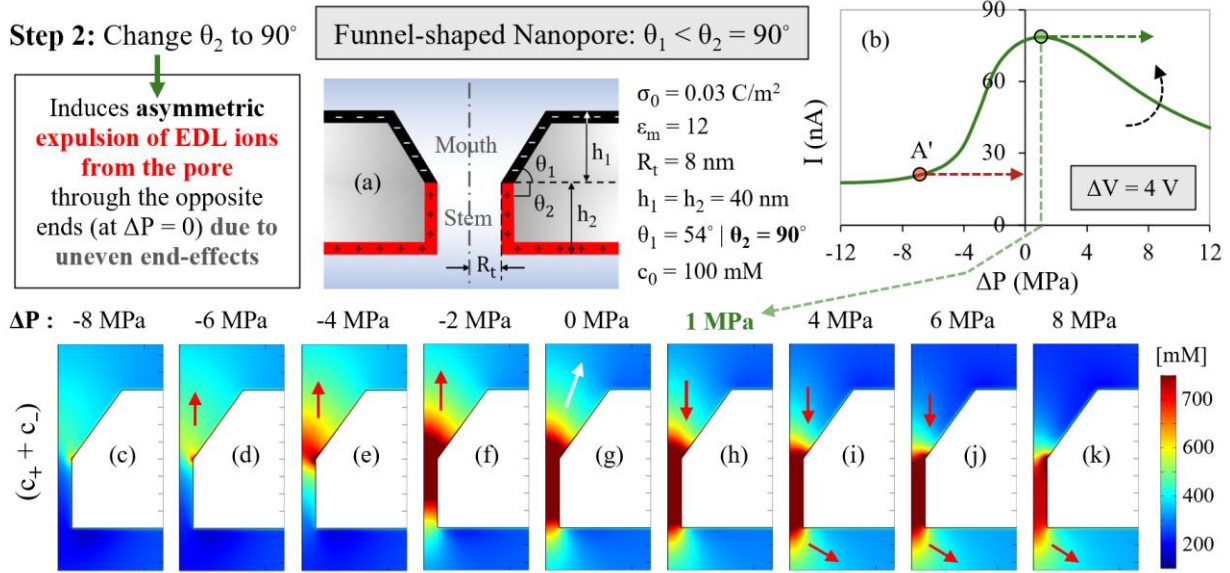


**Figure 3:** (a) Schematic of a bipolar funnel-shaped nanopore, obtained by modifying  $\theta_2$  to  $90^\circ$ . (b) The current-pressure (I-P) response of the nanopore is plotted with the same parameters used in fig. 2, except for  $\theta_2 = 90^\circ$ . (c)-(k) 2D contours of total ionic concentration  $(c_+ + c_-)$  across the pore are plotted at varying pressure drops  $\Delta P$ .

**Step 2:** Shifting the maxima of the I-P plot in fig. 2(a) to  $\Delta P > 0$  while diminishing  $I(\Delta P = 0)$  essentially needs the I-P curve to be transposed rightwards. For that, we raise  $\theta_2$  to  $90^\circ$ , yielding a funnel-shaped pore in fig. 3(a). Now, the ionic accumulation at  $\Delta P = 0$  resides mainly in the lower cylindrical pore section, or the ‘stem’ [see fig. 3(g)]. A weaker lateral channel confinement in the upper conical section, or the ‘mouth’, than that in the stem, as  $\theta_1 < \theta_2$ , expels most of the expected ionic accumulation across the mouth into the top reservoir (as depicted by the white arrow in fig. 3(g)), an end-effect prevalent at channel-reservoir interfaces [47]. While this EDL expulsion occurs

at both ends, it is more intense at the much wider interface between the mouth and the top reservoir. A positive (downward)  $\Delta P$  propels the spilled ions back into the pore, especially into its stem [see figs. 3(h)-3(j)], which is also the zone of high electric field intensity, as seen in fig. S3 of SM [45]. This slightly elevates the conductance and thus the current in fig. 3(b). However,  $\Delta P$  exceeding 1 MPa steers these ions outside the pore into the bottom reservoir [see figs. 3(i)-3(k)] to deplete the current. Therefore, as strategized, the I-P maxima (depicted by the green pointer in fig. 3(b)) now occurs at a positive  $\Delta P$  ( $= 1$  MPa). But as suspected, the current activation factor, from  $\Delta P = 0$  to 1 MPa, is extremely low. To fix this, we can lower  $I(\Delta P = 0)$ . Notice that the current weakens with a negative (upward)  $\Delta P$  as the channel ions are forced out through the pore mouth [see figs. 3(c)-3(f)]. We attempt to induce a similar condition, say the state at point ‘A’ marked by the red pointer in fig. 3(b), at  $\Delta P = 0$ . We additionally aim to extend the range of  $\Delta P$  across which current grows, for a greater tunability of current amplification, and to lower the rate at which current falls with  $\Delta P$  beyond the maxima so that the level of the amplified current can be maintained to a practical extent rather than letting it plummet drastically for an overshooting  $\Delta P$ .

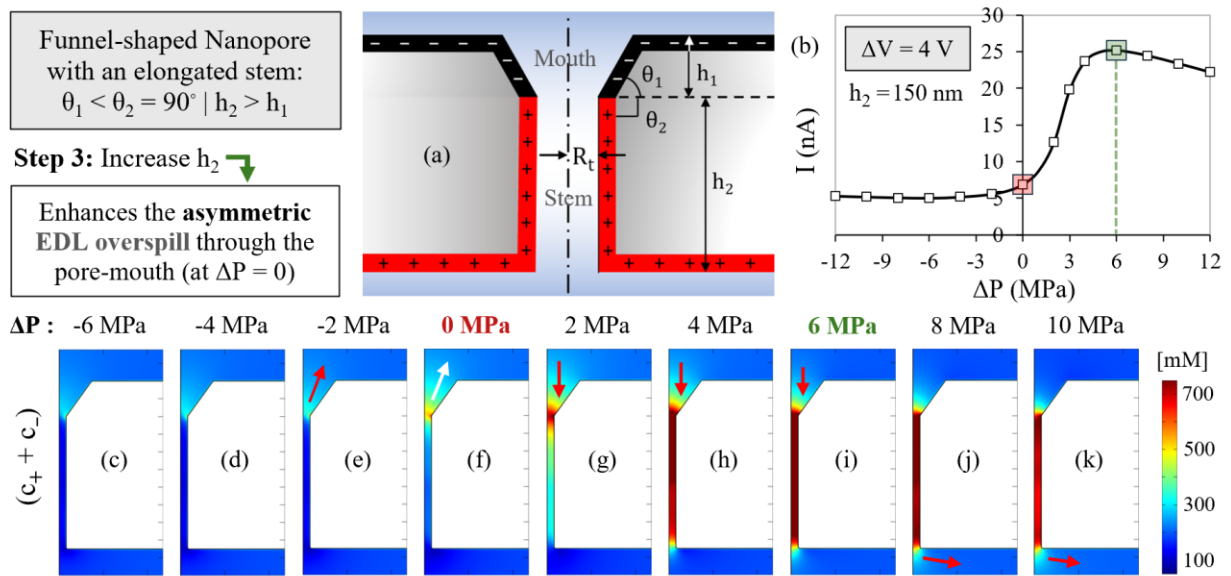


**Figure 4:** (a) Schematic of the bipolar funnel-shaped pore with an elongated stem. (b) The current-pressure (I-P) response of the nanopore is plotted using the same parameters of fig. 3, except for  $h_2 = 150$  nm. (c)-(k) 2D contours of total ionic concentration ( $c_+ + c_-$ ) across the channel are plotted with varying pressure drop  $\Delta P$ .

**Step 3:** All the above issues are addressed simultaneously by elongating the stem of the funnel-shaped pore to an axial length ( $h_2$ ) nearly 4 times that of the mouth ( $h_1$ ), as shown in fig. 4(a). Now, the ions expected to accumulate predominantly in the pore at  $\Delta P = 0$  are  $\text{Cl}^-$  ions, i.e., counterions

to the much longer positively charged stem. A positive  $\Delta V$  pushes these ions up and into the mouth. The  $\text{Cl}^-$  ions are thus easily spilled (along with the  $\text{K}^+$  ions expected to gather across the negatively charged pore mouth) into the upper reservoir, as the mouth now lacks lateral confinement due to  $\theta_1 < \theta_2$  and  $h_1 < h_2$ , yielding massive end-effects [47]. As  $\Delta V$  pushes the ‘net excess’  $\text{Cl}^-$  ions up, it generates an upward electroosmotic (fluid) flow [see fig. S4 of SM [45]], which further enhances the displacement of the  $\text{K}^+$  ions into the upper reservoir. Hence, the anticipated ionic accumulation at  $\Delta P = 0$  is almost entirely displaced into the upper reservoir [see fig. 4(f)], which accomplishes our aim as this is a condition that occurred at negative  $\Delta P$  in step 2. Thus,  $I(\Delta P = 0)$  is substantially reduced, as observed in fig. 4(b). A positive  $\Delta P$  shifts those ions (displaced into the upper reservoir) back into the pore via its mouth and into the stem [see figs. 4(g)-4(i)], which is also the region of high electric field intensity, as shown in fig. S5 of SM [45], thus amplifying the current in fig. 4(b). However, as the downward  $\Delta P$  increases further, the ions are pushed outside the pore via its stem [see figs. 4(i)-4(k)], depleting  $I$ . It yields a maxima in the  $I$ - $P$  curve, and at a higher  $\Delta P$  ( $= 6$  MPa), as larger pressures are required to propel a much greater quantity of ions into the channel (than in step 2), enhancing the tunability of the mechanical activation of ion transport. The diminished  $I(\Delta P = 0)$ , moreover, magnifies the current activation factor to  $\sim 4$ . The reduction rate of  $I$  with  $\Delta P$  past the maxima is also greatly weakened, as raising the pore length decelerates the fluid flow at a given  $\Delta P$ , making it more difficult for the hydrodynamic flow to dislodge the ions from within the pore. Ultimately, the  $I$ - $P$  curve in fig. 4(b) mimics the desired  $I$ - $P$  characteristic in fig. 1(a).

Effectively, we begin with an hourglass-structured bipolar nanopore that enables a mechanical suppression of ionic flux at  $\Delta V > 0$  (analyzed in our previous study [8]) and alter  $\theta_2$  and  $h_2$  to attain a funnel-shaped bipolar nanopore acting as a surge-protected mechanical amplifier of ion transport. The quasi-static position of the ‘ $\Delta V$ -induced’ ionic accumulation within the hourglass-shaped pore at  $\Delta V > 0$  and  $\Delta P = 0$  is expelled into the upper reservoir by modifying  $\theta_2$  ( $h_2$ ) to  $90^\circ$  ( $\sim 4h_1$ ). This enables a downward  $\Delta P$  to first hydrodynamically propel the quasi-static positions of the dislodged ions back into the channel where the electric field intensity is substantially greater (to enhance the ionic conductance) and then expel them out into the bottom reservoir (to diminish the conductance), yielding the desired non-monotonous  $I$ - $P$  response. For a clearer visualization of the developments in step 2 (3), we display its intermediate stages at gradual stepped increments of  $\theta_2$  ( $h_2$ ) in fig. S6 (fig. S7) of SM [45]. Increasing  $\Delta V$  augments the extent of ionic accumulation at  $\Delta P = 0$  within the (initial) hourglass-shaped nanopore, since it originates primarily due to  $\Delta V$ . This enhances the ionic

expulsion from the (formulated) funnel-shaped nanopore to the upper reservoir at  $\Delta P = 0$ . Thus, a subsequently applied downward  $\Delta P$  can propel a greater quantity of ions back into the pore. Hence, a larger  $\Delta V$  amplifies both the current activation factor and the magnitude of  $\Delta P$  required to reach the maximum current, as depicted in fig. S8 of SM [45]. But, at  $\Delta V = 0$ , fig. S8(d) shows a negative (upward) current at positive (downward)  $\Delta P$  that varies linearly with  $\Delta P$ . It stems from the pressure-driven hydrodynamic advection of the net excess  $\text{Cl}^-$  ions, which is independent of  $\Delta V$  [7]. Hence, its contribution to the current at  $\Delta V = 4$  V and a certain  $\Delta P$  is equal to the (purely pressure-induced) current at  $\Delta V = 0$  and the same  $\Delta P$ , which is 2-3 orders of magnitude smaller in intensity than the current at  $\Delta V = 4$  V [see fig. S8(d)]. Therefore, the pressure-driven hydrodynamic streaming of net excess counterions does not influence the behavior of the I-P response in fig. 4(b).

In fig. S9 (fig. S10) of SM [45], we inspect the effects of varying  $\theta_1$  ( $h_1$ ) on the I-P curve while retaining the other parameters in fig. 4, elaborated thoroughly in sec. IX of SM [45]. Interestingly, increasing  $\theta_1$  and  $h_1$  (till  $0.5h_2$  to retain the effects of step 3) both diminish the  $\Delta P$  magnitude at which maximum current is recorded, elevate the reduction rate of current with  $\Delta P$  past the maxima, and cause the current activation factor (CAF) to initially rise and then diminish which suggests that CAF can be optimized via  $\theta_1$  and  $h_1$ . However, the highest CAF obtained at  $\theta_1$  ( $h_1$ )  $\approx 63^\circ$  (30 nm) in fig. S9 (fig. S10) is only  $\sim 2\%$  (4%) greater than that yielded in fig. 4(b).

Although our analysis accounts for dielectric polarization via eq. (S4) in SM [45], the additional charges it induces on the pore (and outer membrane) surfaces and thus its influence on the I-P curve in fig. 4(b) are insignificant [see fig. S11 of SM [45]], as the membrane bearing the funnel-shaped nanopore in fig. 4(a) is not axially thin enough to develop substantial polarization-induced surface charges [7,8,39,40]. Furthermore, the I-P response demonstrates no noticeable changes if the outer membrane surface AB (HG) is inherently uncharged, unless  $h_1$  ( $h_2$ ) is diminished significantly [see fig. S12 of SM [45]], indicating the much greater influence of the surface charges on AI (HI).

Hence, we observe that a bipolar funnel-shaped nanopore with an elongated positively charged cylindrical stem and a negatively charged conical mouth manifests a ‘surge-protected’ mechanical amplifier of ion transport. A mechanical stimulus in the form of a pressure drop exerted, alongside a constant parallel electric field, towards the stem yields a controlled amplification of ionic current up to a restricted magnitude of applied pressure, which effectively marks the ‘operational’ pressure range of the amplifier. If the pressure drop overshoots this limit, current starts to reduce gradually,

inhibiting a monotonous current surge. We simultaneously focus on boosting the current activation factor and tunability of the amplifier.

In essence, unlike the past conventional studies in the field which prioritized a fixed nanopore design to analyze its ion transport features, we began with the aim to realize a useful ion transport characteristic and formulated a nanopore that exhibits it by strategically integrating an asymmetric channel shape and surface charge inhomogeneity into it. Therefore, besides manifesting a hitherto undetected electrokinetic behavior, our study exhibits an innovative ‘function to design’ approach in the field of electrokinetics that can help develop novel nanofluidic devices with more effective and accurate routes of regulating ionic and molecular flow under electrical and mechanical stimuli.

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