

Ionic conductivity and excitation frequency effects in iontronic pressure sensing

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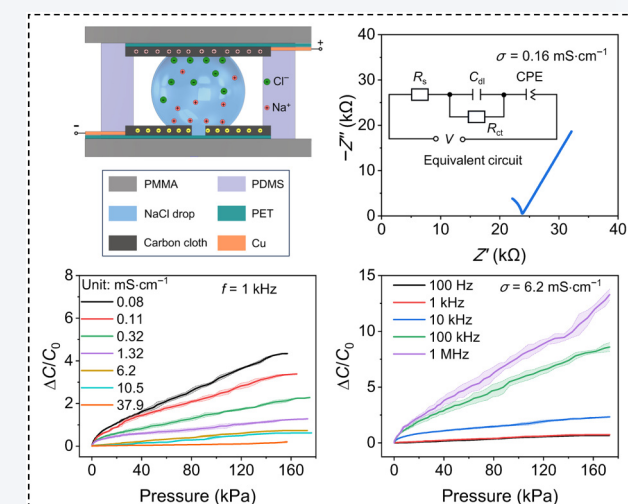
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ABSTRACT: Iontronic pressure sensors, which combine high sensitivity, superior signal stability, and resistance to electromagnetic interference, are regarded as promising candidates for next-generation pressure sensing systems. However, the roles of ionic conductivity and excitation frequency in determining sensing behavior remains unknown. Here, we employ an ionic-droplet-based research model to elucidate these effects through electrochemical impedance spectroscopy (EIS). Based on the equivalent circuit, we derived equations that link apparent capacitance change to ionic conductivity and excitation frequency. The equations reveal that droplet compression leads to larger apparent capacitance variation at lower ionic conductivity and higher excitation frequency, consistent with our experimental observations. Accordingly, tuning these parameters alone enhances the sensitivity of the droplet-based pressure sensor by up to 8772% without any structural modification. Under the optimized conditions, the sensor delivers highly repeatable pressure responses. Integrated into a robotic arm, it enables real-time pressure detection and discrimination of materials with different softness levels. By substituting ionic-droplets with hydrogels, the sensing model is successfully extended to solid-state ionic materials. A hydrogel sensor array of 4×4 with a pixel size of 2 mm is fabricated, enabling spatial pressure mapping. These findings provide new design principles for elastic ionic materials and underscore the critical role of measurement parameters in determining the sensing behavior of ionic sensing systems.

KEYWORDS: iontronic electronics, pressure sensor, flexible electronics, excitation frequency, ionic conductivity

1 Introduction

Iontronic pressure sensing has emerged as a promising paradigm for next-generation flexible electronics since its first report in 2011 [1]. This approach leverages the ultrahigh interfacial capacitance of the electric double layer (EDL) to achieve superior sensitivity, excellent immunity to electromagnetic interference, and high signal stability [2–4]. Owing to ubiquitous presence of ionic charge



carriers in natural media, iontronic sensors also enable facile integration with biological tissues [5]. Moreover, iontronic sensing systems can detect both static forces and dynamic forces, greatly expanding their application potential in tactile electronics [3, 6–11].

Over the past decade, tremendous progress has been made in iontronic pressure sensors through innovations in materials [12–16], structures [17–20], and interfacial engineering [21–24]. Material advances include the development of conductive polymers [25], particle-doped composites [26], ionic nanofiber membranes [27], and ionogels with improved dielectric and mechanical properties [21, 28–32]. Structural innovations, such as micro-pyramids [17, 19], micro-hemispheres [22, 29], micro-pillars [18], and gradient architectures [33, 34], enhance the deformability of the

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dielectric layer under compression, leading to significant improvements in pressure sensitivity, hysteresis, and signal stability. These efforts have yielded record sensitivities (e.g., 9280 kPa⁻¹ using ionic-liquid-infused foams [35]) and highly linear responses over broad pressure ranges [18, 36], pushing iontronic sensors toward practical systems.

Despite these achievements, the intrinsic electrical parameters that fundamentally govern iontronic sensing remain unexplored. In particular, the effects of ionic conductivity (σ) and excitation frequency (ω), two parameters central to ion transport and EDL formation [37–39], have received little systematic attention. In most reported studies, the electrical readout of such sensors is recorded using an inductance–capacitance–resistance (LCR) meter (Table S1 in the Electronic Supplementary Material (ESM)), which extracts the apparent capacitance of the device through impedance measurements. Although electrochemical impedance spectroscopy (EIS) has been used to probe ionic interfaces [40–43], quantitative understanding of how these parameters determine capacitance responses is still lacking. Addressing this gap is essential for building a unified physical framework that links ion transport dynamics to pressure sensitivity, beyond structural or material-level improvements.

Here, we construct an ionic-droplet-based research model to uncover how ionic conductivity and excitation frequency govern iontronic sensing. This platform enables precise modulation of ionic conductivity while eliminating structural variations. By combining experimental impedance analysis with an equivalent circuit framework, we derive explicit expressions linking the apparent capacitance to ionic conductivity and excitation frequency. The analysis reveals that reducing ionic conductivity and increasing excitation frequency markedly amplify compression-induced changes in apparent capacitance, in quantitative agreement with experimental results. The stress distribution, droplet deformation, and ion concentration distribution of the droplet-based pressure sensor under different pressures were simulated using COMSOL finite element analysis (FEA) software [44]. Under optimized conditions, the sensor exhibits an enhancement of 8772% (ratio of maximum to minimum) in sensitivity without any structural modification and high sensing stability. When integrated into a robotic arm, it enables real-time monitoring of contact pressure and discrimination of material softness [45]. By replacing ionic-droplets with hydrogels, we successfully extended the sensing model to iontronic pressure sensors based on solid-state ionic materials such as hydrogel and were able to verify conclusions consistent with the droplet-based pressure sensor. We also fabricated an array device of 4 × 4 based on hydrogel pressure sensors, with a sensing pixel size of 2 mm, and demonstrated its application in spatial pressure mapping. This work identifies ionic conductivity and excitation frequency as key governing parameters for iontronic sensing and establishes a quantitative framework for their modulation. The findings not only deepen the fundamental understanding of iontronic mechanisms but also offer a practical route toward high-sensitivity tactile sensors for next-generation human-machine interfaces.

2 Results and discussion

2.1 Research model structure and sensing mechanism

Figure 1(a) depicts the structural schematic of the research model

and the illustration of controlling the electrode displacement using an electronic universal tester. Outer polymethyl methacrylate (PMMA) plates were used to maintain parallel alignment of the electrodes, preventing complex deformation that would complicate model analysis. Polyethylene terephthalate (PET) served as the flexible substrate for current collecting electrode (Cu). Hydrophobic carbon cloth was used as porous electrode to enable reversible change of contact area with the NaCl droplet. 10 μ L of NaCl droplet was trapped inside the research model cavity and anchored by the 1 mm-diameter hydrophilic pore of the bottom carbon cloth. To decouple mechanical effects, we deliberately removed the elastic supporting frame and used mechanical testing system to precisely control the electrode displacement between two carbon electrodes.

Figure 1(b) illustrates the iontronic sensing mechanism of the device. Distinct displacements (with the reference zero defined at the onset of the abrupt electrical signal transition) corresponded to different operational states. Under an applied voltage, ions within the NaCl droplet migrate to form EDL at the electrode–droplet interfaces. When electrode displacement is reduced, the droplet is compressed, leading to deformation of the droplet and changes in the interfacial contact area, which in turn modulate the device's capacitance and impedance.

Figure 1(c) presents the Nyquist plot of the research model. In this plot, the high-frequency region exhibits a characteristic semicircular arc, while the low-frequency region displays a linear response. The corresponding equivalent circuit model is shown in Fig. 1(d), where R_s represents the droplet resistance, C_{dl} denotes the electrochemical double-layer capacitance at the interface between the hydrophobic carbon cloth electrode and the water droplet, and R_{ct} corresponds to the charge-transfer resistance at the electrochemical interfaces, originating from ion adsorption and desorption on the carbon cloth surface. A constant phase element (CPE) was incorporated to account for the non-ideal behavior of the EDL.

2.2 Impedance response characteristics of the research model

To systematically evaluate the influence of droplet σ on the impedance response, Nyquist and Bode plots were measured for droplets with low ($\sigma = 0.08$ – 1.32 mS·cm⁻¹) and high ($\sigma = 6.2$ – 511.0 mS·cm⁻¹) ionic conductivities at a fixed displacement of 1.0 mm. The corresponding measurement schematic is shown in Fig. 2(a). For low ionic conductivity droplets (Fig. 2(b)), the Nyquist plots exhibited the characteristic high-frequency semicircular arcs and low-frequency linear tails. As ionic conductivity increased, the curves shifted leftward, the semicircular arcs became progressively shorter, and the linear tails extended. Figure 2(c) presents the frequency-dependent variation of total impedance ($|Z|$), which decreased monotonically with increasing frequency. Higher ionic conductivity accelerated the rate of $|Z|$ reduction in the low-frequency region. Figure 2(d) depicts the evolution of the phase angle (θ): The absolute value of the phase angle ($|\theta|$) initially decreased, reached a minimum between 10^4 and 10^5 Hz, and subsequently increased with frequency. Increasing σ elevated $|\theta|$ in the low-frequency region while reducing it in the high-frequency region.

For high ionic conductivity droplets (Fig. 2(e)), the Nyquist plots were dominated by a linear response that shifted leftward with increasing σ . This behavior occurred because the transition frequency between the semicircular and linear regions exceeded the

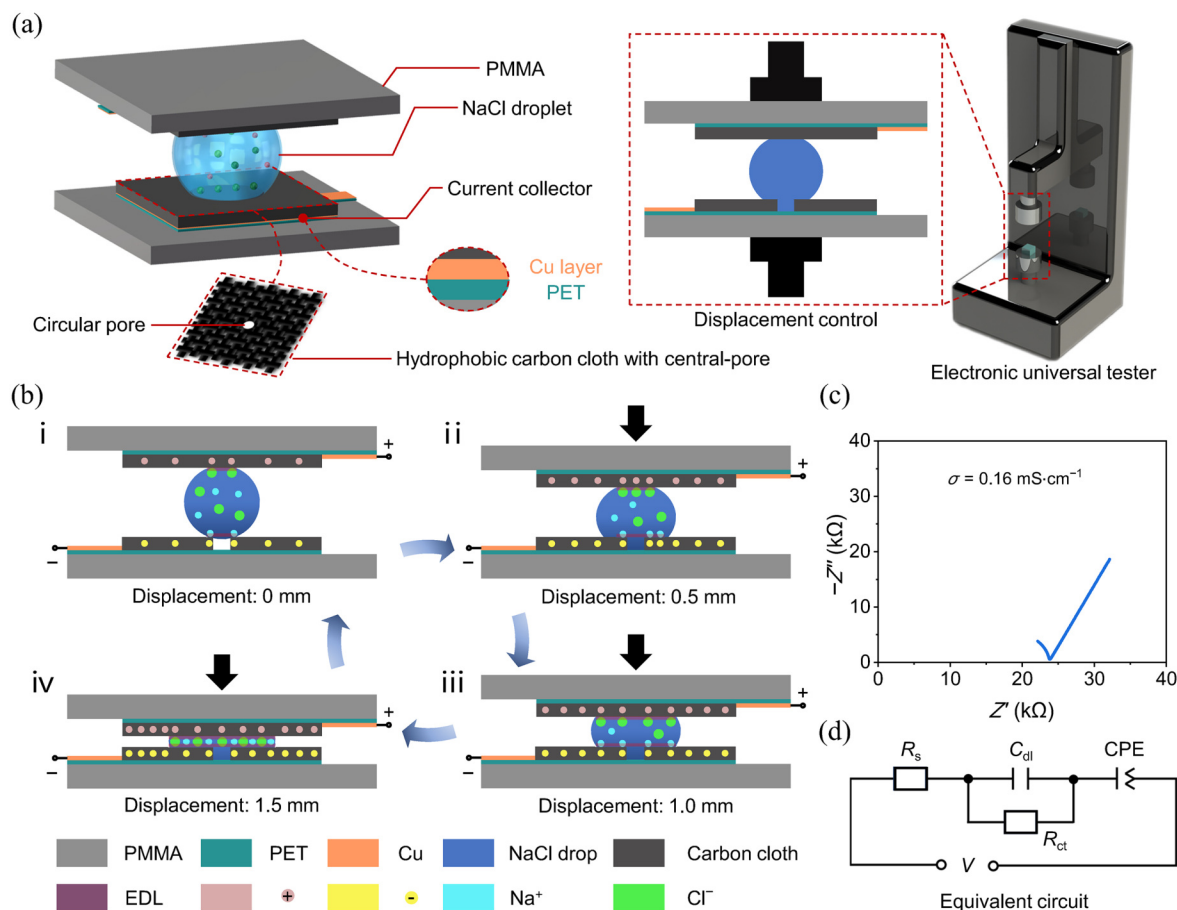


Figure 1 Structure, sensing mechanism, and equivalent circuit model of the research model. (a) Schematic diagram of the research model structure and the setup for controlling displacement using an electronic universal tester. (b) Schematic of the sensing mechanism: (i) initial state and ((ii)–(iv)) evolution under displacements of 0.5, 1.0, and 1.5 mm, respectively. (c) Nyquist plot and (d) corresponding equivalent circuit model.

upper measurement limit of the instrument (1 MHz) at high σ . As shown in Fig. 2(f), $|Z|$ consistently decreased with increasing ionic conductivity. Figure 2(g) illustrates that $|\theta|$ increased with higher σ , while the turning point, defined as the frequency corresponding to the most pronounced slope change, shifted toward higher frequencies. Similar trends were observed at other displacements (0.5 and 1.5 mm; Figs. S1 and S2 in the ESM). These results confirm that ionic conductivity significantly influences the impedance response of the research model, thereby directly affecting their sensing performance. Additionally, ω plays a critical role in modulating system impedance, warranting further mechanistic investigation. Unless otherwise specified, subsequent analyses are based on the displacement of 1.0 mm.

To validate the incorporation of the CPE in the equivalent circuit model, measured capacitance values were compared with calculated results for models with and without (w/ and w/o) the CPE (Figs. S3–S5 in the ESM). The coefficients of determination (R^2) for each case are shown in Fig. 2(h) and Fig. S6 in the ESM. Models incorporating the CPE consistently exhibited $R^2 > 0.99$, while those without the CPE yielded negative R^2 values, confirming the necessity of the CPE for accurate modeling. The corresponding derivations are provided in Fig. S8 in the ESM. Moreover, the quality of the EIS fitting was high (Fig. 2(i)), with most Chi-squared values located within the well-fitted region (green zone), demonstrating the reliability of the extracted parameters (orange zone indicates fair fits).

Furthermore, to assess the nonlinear polarization effects observed in the electrochemical impedance spectroscopy measurements of the droplet-based device at high frequencies or under large deformations, the raw experimental data were systematically compared with the corresponding fitted results for droplet devices with different ionic conductivities (Fig. S7 in the ESM). Excellent agreement between the experimental and fitted data is achieved in the high-frequency regime for both low and high ionic conductivity cases. These results indicate that the nonlinear polarization effect is intrinsically embedded in the experimental response and is effectively captured by the proposed equivalent circuit model, thereby validating its applicability under practical operating conditions.

2.3 Analysis of the influence of frequency and ionic conductivity on device capacitance variation

As established, ω and σ critically influenced impedance behavior. To quantify their effects on $|Z|$ and to further investigate their regulatory effects on device apparent capacitance variation, the partial derivatives $\partial|Z|/\partial\omega$ and $\partial|Z|/\partial\sigma$ were derived from the equivalent circuit model (Fig. 1(d); full derivation in Fig. S8 in the ESM). These expressions provide a rigorous mathematical framework for describing the regulatory influence of ω and σ . Figures 3(a)–3(c) compare theoretical and experimental values of $\partial|Z|/\partial\omega$ (for low and high σ) and $\partial|Z|/\partial\sigma$, with coefficients of determination calculated. Theoretical values were obtained directly

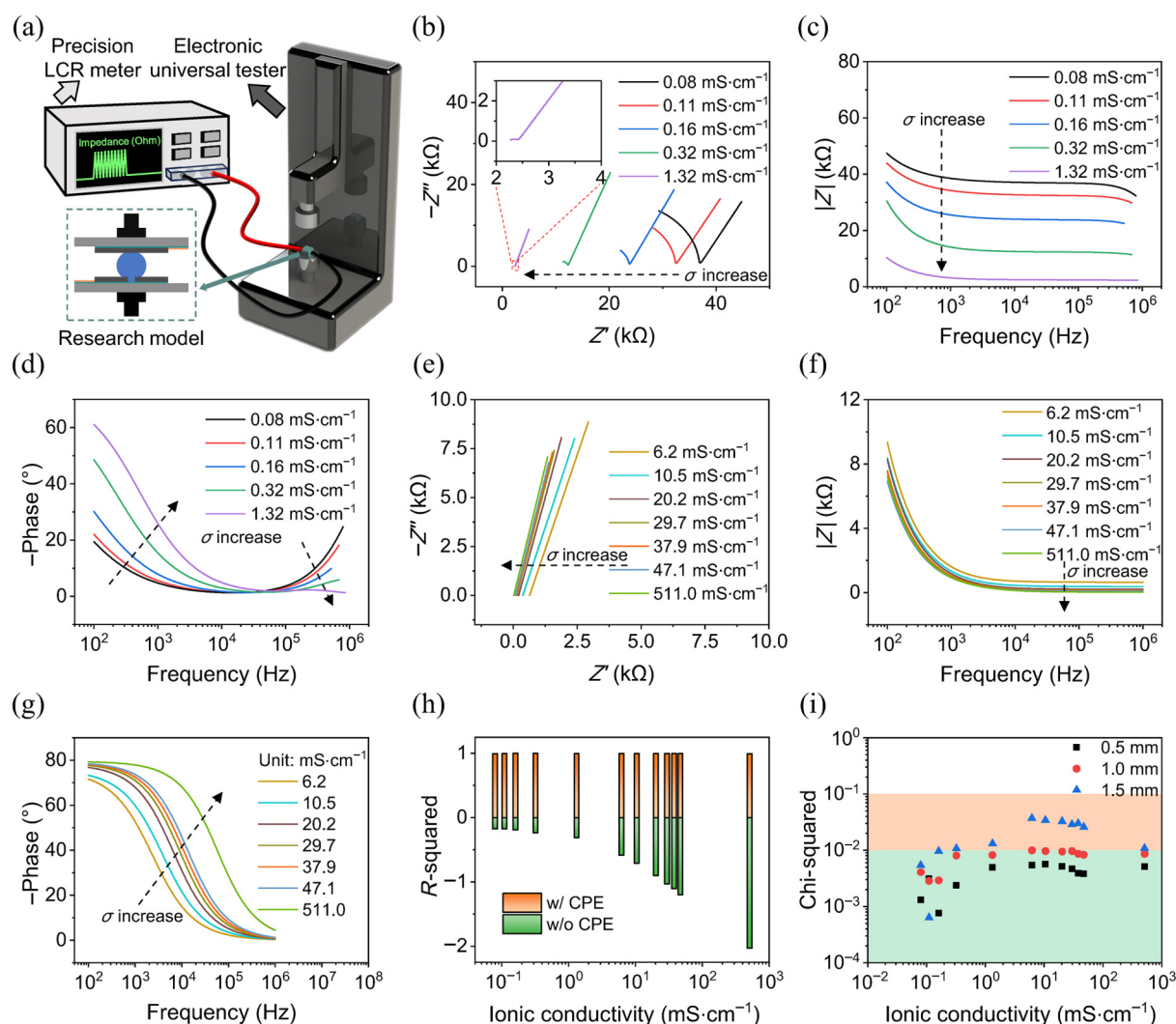


Figure 2 Electrochemical impedance spectroscopy analysis of the research model. (a) Schematic diagram of EIS measurement setup. (b) Nyquist plots for low ionic conductivity droplets. ((c) and (d)) Bode plots for low ionic conductivity droplets. (e) Nyquist plots for high ionic conductivity droplets. ((f) and (g)) Bode plots for high ionic conductivity droplets. (h) Comparison of R^2 values for models with and without the CPE. (i) EIS fitting results.

from the derived equations, whereas experimental values were obtained by numerical differentiation of measured $|Z|$. High R^2 values confirm excellent agreement between theoretical and experimental results. The concordance of the partial derivatives ($\partial|Z|/\partial\omega$ and $\partial|Z|/\partial\sigma$) across other displacements and excitation frequencies further validates the predictive accuracy of the derived equations (Figs. S9–S12 in the ESM). Both partial derivatives are negative ($\partial|Z|/\partial\omega < 0$ and $\partial|Z|/\partial\sigma < 0$), confirming that total impedance decreases with increasing frequency or ionic conductivity, consistent with the trends in Fig. 2. Importantly, ionic conductivity affects $|Z|$ far more strongly than frequency: The absolute values of $\partial|Z|/\partial\sigma$ are orders of magnitude larger than those of $\partial|Z|/\partial\omega$ (comparing the scales in Figs. 3(a)–3(c)). Moreover, the sensitivity of $|Z|$ to either parameter increases as the parameter decreases; that is, both $\partial|Z|/\partial\omega$ and $\partial|Z|/\partial\sigma$ attain larger magnitudes in the low-frequency and low-conductivity regions, as clearly illustrated in Figs. 3(a)–3(c).

The above results firmly establish the validity of the equivalent circuit model (Fig. 1(d)), providing a reliable foundation for using this mode, together with additional measurements, to investigate how ionic conductivity and excitation frequency regulate the

apparent capacitance variation ($\Delta C/C_0$) of the device. In the equivalent circuit, the CPE accounts for EDL non-ideality, which mainly originates from electrode roughness. Because the electrode material is identical across devices, the influence of the CPE can be neglected in the comparative analysis, thus the equivalent circuit was simplified accordingly (Fig. 3(d)).

Under the same displacement, the variations of C_{dl} and R_{ct} primarily reflect changes in the contact area between the droplet and the carbon electrodes, while the variation of R_s arises from both droplet deformation and contact area changes. Thus, the rates of change of C_{dl} , R_{ct} , and R_s are governed by contact area and droplet geometry, and are largely independent of ionic conductivity and excitation frequency. Under these conditions, the double-layer capacitance per unit contact area can be considered constant. Consequently, the variation of C_{dl} mainly arises from the change in the droplet-electrode contact area as the electrodes approach each other. At the identical displacements, the change rates of these three parameters can be considered consistent, since ionic conductivity has little influence on the droplet deformation. To further examine this assumption, we fitted impedance spectra obtained at two fixed electrode displacements (1.0 and 1.5 mm). Using the initial values

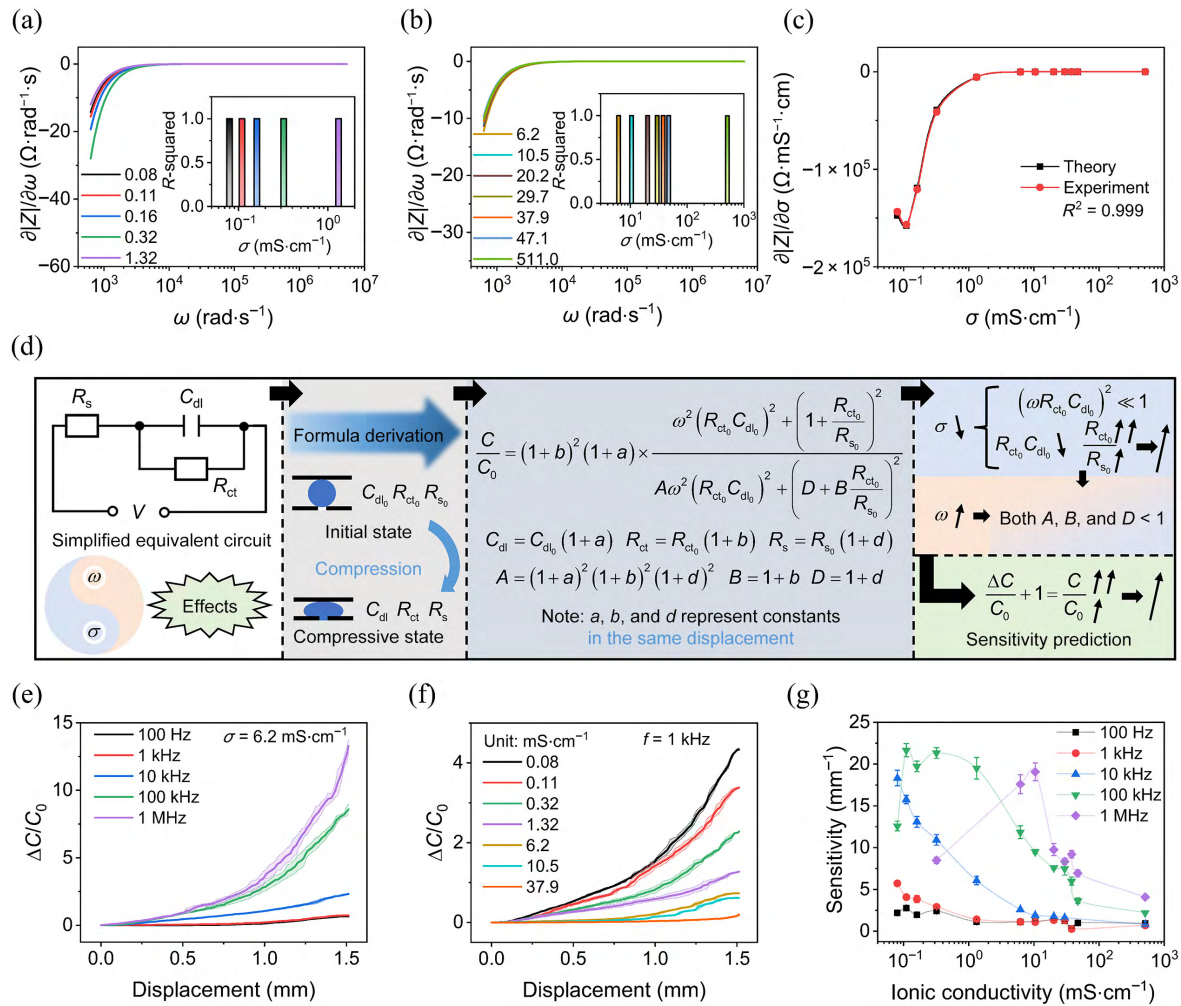


Figure 3 Influence of frequency and ionic conductivity on device capacitance variation. ((a) and (b)) Comparison of theoretical and experimental values of $\partial Z/\partial \omega$ for devices with (a) low and (b) high ionic conductivity droplets (inset: R^2 values). (c) Comparison of theoretical and experimental values of $\partial Z/\partial \sigma$ at a fixed frequency of 1 kHz for droplets with different ionic conductivities. (d) Theoretical analysis of excitation frequency and ionic conductivity effects on device capacitance variation based on simplified equivalent circuit. ((e) and (f)) Capacitance variation versus displacement curves for devices (e) at different frequencies and (f) with different ionic conductivities. (g) Device displacement sensitivity variation with ionic conductivity and frequency at the displacement range of 1.0–1.5 mm.

R_{s_0} , C_{dl_0} , and R_{ct_0} at 0.5 mm displacement as references, the extracted ratios R_s/R_{s_0} , C_{dl}/C_{dl_0} , and R_{ct}/R_{ct_0} were found to be generally comparable across different ionic conductivities (Table S2 in the ESM). The small deviations observed are attributed mainly to fitting uncertainties and experimental variations, such as slight differences in the initial droplet-electrode contact condition and surface inhomogeneity of the carbon cloth. Therefore, we assigned them with constant values a , b , and d , respectively.

Based on the simplified circuit, we derived the equation for apparent capacitance variation linking to frequency and ionic conductivity as Eq. (S25) in Fig. S8 in the ESM. At the same frequency, since the order of magnitude about $R_{ct}C_{dl}$ is 10^{-7} and the maximum frequency is 10^6 Hz (Fig. S13 in the ESM), the value of $(\omega R_{ct}C_{dl})^2$ is much less than 1, allowing omission of the numerator's $(\omega R_{ct}C_{dl})^2$ term in the equation. As ionic conductivity decreases, $R_{ct}C_{dl}$ decreases, while R_{ct}/R_s increases (Fig. S13 in the ESM). Since the values of A , B , and D are all less than 1, the numerator increases more rapidly than the denominator, thus the apparent capacitance variation increases with decreasing ionic conductivity. Similarly, at the same ionic conductivity, the variable of this formula is only frequency. Since A is less than 1, the numerator increases faster

than the denominator as frequency increases, thus the device apparent capacitance variation increases with frequency. In other words, since the initial capacitance decreases with increasing excitation frequency, the normalized sensitivity ($\Delta C/C_0$) of the device will increase with higher frequency if the capacitance variation is similar. This behavior is attributed to measurement-defined effects at high frequencies. As indicated by the analytical expression in Fig. 3(d), under constant ionic conductivity, the capacitance variation of device increases monotonically with frequency.

Figure 3(e) shows capacitance variation versus displacement curves for devices at different frequencies ($\sigma = 6.2$ mS·cm⁻¹). Figure 3(f) displays the results under different ionic conductivities at a fixed frequency (1 kHz). The influence of these two parameters on apparent capacitance variation exhibited consistent across different ionic conductivities and frequencies (Figs. S14 and S15 in the ESM). At the displacement range of 1.0–1.5 mm, the device displacement sensitivity varied with ionic conductivity and frequency, as depicted in Fig. 3(g), which was defined as the slope of the $\Delta C/C_0$ versus displacement curve. Experimental results demonstrated that sensitivity generally increased with higher

frequency and lower ionic conductivity, aligning with the theoretical predictions in Fig. 3(d). However, at frequencies of 100 kHz and 1 MHz, the displacement sensitivity slightly decreased in the low-conductivity regime. This reduction is speculatively attributed to diminished capacitive reactance and consequently smaller capacitance variations under high frequency conditions. Similar trends in sensitivity were observed at the displacement range of 0–1.0 mm (Fig. S16 in the ESM).

To further exclude contributions from ionic/EDL-related effects, we examined the pressure response of the research model by supporting a deionized water droplet with a polydimethylsiloxane (PDMS) frame, which transduces applied pressure into electrode displacement. A similar frequency-dependent sensitivity enhancement was observed (Fig. S17 in the ESM), supporting the conclusion that this effect is not governed by EDL modulation. In addition, we evaluated the pressure response of devices without droplets, which effectively behave as parallel-plate capacitors. In this case, the capacitance signal was markedly weaker due to the absence of EDL formation (Fig. S18 in the ESM). While ions are present, even at low concentrations, ionic conductivity and excitation frequency jointly influence the impedance distribution of the system, leading to a significantly larger change in $\Delta C/C_0$. In this sense, ionic conductivity does not create the frequency-dependent behavior itself but amplifies its effect, resulting in a synergistic enhancement of $\Delta C/C_0$.

Taken together, these results indicate that while ionic/EDL-related effects primarily establish a large baseline capacitance (from the pF to nF level), the enhanced pressure sensitivity observed at higher excitation frequencies arises predominantly from measurement-defined impedance effects, rather than intrinsic frequency-dependent EDL modulation. While lower ionic conductivity and higher excitation frequency can increase $\Delta C/C_0$, they simultaneously reduce the baseline capacitance. As a result, the measurable capacitance signal shifts from the nF range to the pF range, which imposes stricter requirements on measurement precision and reduces the practical readout range. In contrast, systems with moderate ionic conductivity provide a larger baseline capacitance and more robust signal readout.

2.4 Sensor performance characterization

To experimentally validate the effects of frequency and ionic conductivity on sensor performance, the apparent capacitance variation versus pressure curves were measured for devices with droplets of different ionic conductivities under varying frequencies. These curves provided insight into the influence of both parameters on device pressure sensitivity, which was defined as the slope of the $\Delta C/C_0$ -pressure curve. Figure 4(a) depicts the structural schematic and optical image of the sensor. PDMS functioned as both the elastic frame and the seal. Detailed fabrication procedures and optical images are provided in Figs. S19 and S20 in the ESM. The cycling stability of devices with and without a hydrophilic pore was also compared. Devices lacking the hydrophilic pore exhibited pronounced baseline signal fluctuations, whereas those incorporating the hydrophilic pore maintained excellent baseline stability, underscoring the critical role of droplet confinement (Fig. S21 in the ESM).

The Young's modulus (E) of the PDMS frame impacted its deformation under applied pressure, thereby affecting device sensitivity. To investigate this effect, three PDMS frames with different Young's modulus were fabricated by varying the curing

agent ratios in the same temperature of 25 °C (Fig. 4(b)). The results demonstrate that device sensitivity can be effectively tuned by modulating the Young's modulus of the PDMS frame (Fig. S22 in the ESM).

To visualize the coupled mechanical-electrochemical behavior of the droplet-based device under pressure, finite-element simulations were performed. As shown in Fig. S23 in the ESM, the applied stress is primarily concentrated within the PMMA plate, copper electrodes, and PDMS frame, while the droplet deformation gradually increases with increasing pressure. In addition, a pronounced ion concentration gradient develops at the droplet/carbon cloth interface, and the pressure-enhanced deformation significantly promotes ion migration toward the carbon cloth electrode.

Figures 4(c) and 4(d) present the $\Delta C/C_0$ -pressure curves for devices sharing the same PDMS frame but measured at different frequencies ($\sigma = 6.2 \text{ mS}\cdot\text{cm}^{-1}$) and for droplets with varying ionic conductivities at a fixed excitation frequency (1 kHz), respectively. These results clearly demonstrated that pressure sensitivity increases with decreasing ionic conductivity and increasing excitation frequency. Additional results under different frequencies and ionic conductivities are shown in Figs. S24 and S25 in the ESM. Additionally, the initial capacitance of the device under various conditions is shown in Fig. S26 in the ESM, indicating a decrease in capacitance with increasing frequency. Figure 4(e) illustrates the dependence of pressure sensitivity on ionic conductivity and frequency in the pressure range of 5–180 kPa. Overall, sensitivity increases monotonically with lower ionic conductivity and higher frequency, but exhibits a slight decline in the low-conductivity regime at 100 kHz and 1 MHz, consistent with experimental results in Fig. 3. By optimizing ionic conductivity and excitation frequency, the pressure sensitivity is enhanced by 8772% without any structural modification. Similar trends were observed in the low-pressure regime (0–5 kPa; Fig. S27 in the ESM).

Beyond sensitivity, sensing stability is equally critical. To assess whether the optimized ionic conductivity and excitation frequency compromise sensing stability, we performed dynamic force-sensing tests using a droplet with $\sigma = 1.32 \text{ mS}\cdot\text{cm}^{-1}$ at 100 kHz. Figures 4(f) and 4(g) show the apparent capacitance variation under different compressive speeds (fixed strain: 50%) and under different compressive strains (fixed speed: 30 mm·min⁻¹), demonstrating a robust and consistent dynamic response. Moreover, cyclic testing revealed no observable signal drift after 1480 cycles (Fig. S28 in the ESM), confirming excellent long-term stability. These results indicate that the optimized parameters significantly enhance sensitivity without introducing any adverse effects on sensing stability. Additionally, we also tested the response/recovery time, detection limit, and pressure resolution of this droplet-based pressure sensor (Fig. S28 in the ESM).

In Table S3 in the ESM, we compared the key performance metrics of several recently reported iontronic pressure sensors. This comparison reveals that the sensitivity of our current sensor is relatively lower, which can be effectively tuned by adjusting the Young's modulus of the PDMS frame and roughness of the droplet/electrode interface. Importantly, this study primarily focuses on elucidating the influence of ionic conductivity and excitation frequency on iontronic pressure sensing, an aspect that has not been extensively explored in the existing literature. By rationally modulating these two parameters, the sensitivity of iontronic pressure sensors can be further improved and is expected to surpass the levels reported previously.

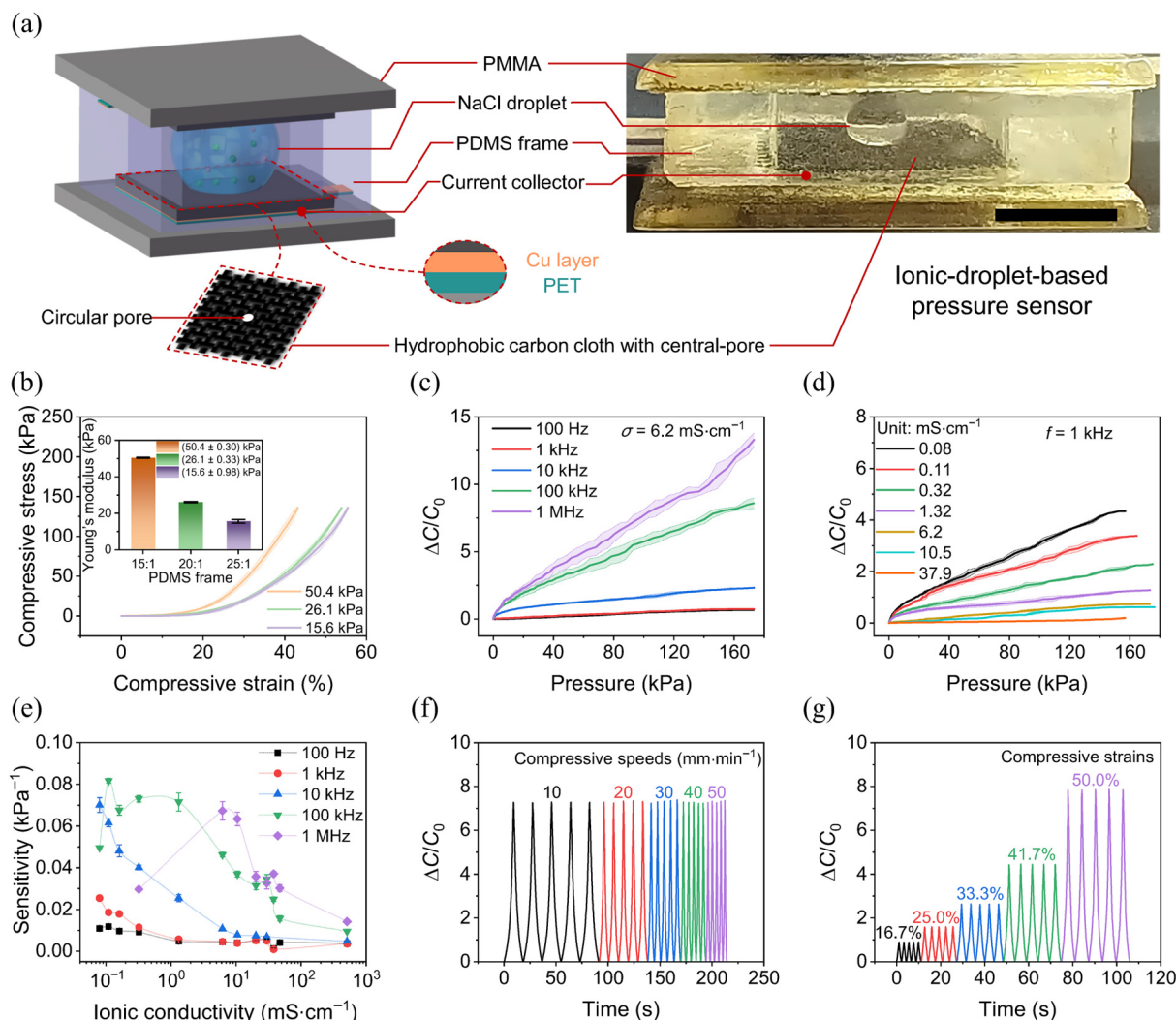


Figure 4 Performance characterization of the sensor. (a) Schematic representation of the sensor architecture and corresponding optical image. Scale bar: 5 mm. (b) Compressive stress-strain curves of different PDMS frames (inset: corresponding Young's modulus indicated by base-to-curing agent weight ratios). ((c) and (d)) Capacitance variation versus pressure curves for devices (c) at different frequencies and (d) with droplets of varying ionic conductivities. (e) Device pressure sensitivity variation with ionic conductivity and frequency within the pressure range of 5–180 kPa. (f) Capacitance variation under a fixed compressive strain (50%) with varying compressive speeds. (g) Capacitance variation at a fixed compressive speed (30 mm·min⁻¹) with varying compressive strains.

2.5 Sensor application demonstrations

To demonstrate the practical potential of the optimized iontronic pressure sensor, it was integrated into the end of a robotic arm for material differentiation (Fig. 5(a) and Fig. S29 in the ESM). As shown in Fig. 5(b), when compressing PDMS cubes under identical displacements, the sensor generated stronger signal variations for samples with higher Young's modulus, clearly demonstrating its ability to distinguish material softness (Fig. S30 and Movie S1 in the ESM).

The sensor was further integrated into a robotic gripper (Fig. 5(c), and Figs. S31 and S32 in the ESM). When deployed in a robotic manipulation platform for object grasping, the sensor provided real-time tactile feedback that enabled discrimination of common objects based on their hardness. As depicted in Fig. 5(c), the signal variation amplitude decreased progressively with decreasing objects stiffness—metal and glass produced the highest responses, followed by plastic, rubber, and sponge, which produced the weakest signal. These results confirm the sensor's broad applicability in object differentiation and tactile perception,

highlighting its potential for integration into intelligent robotic systems (Fig. S33 and Movie S2 in the ESM).

2.6 Hydrogel device and its application demonstration

To overcome the intrinsic limitations of droplet-based pressure sensors, including leakage, evaporation, and positional instability under deformation, we extended the sensing framework to hydrogels as solid-state ionic conductor. The ionic conductivity of the hydrogels was precisely tuned by NaCl incorporation, and surface microstructures were introduced using sandpaper templating to enhance the sensitivity. Compression-induced capacitance variation was systematically evaluated for hydrogels with different ionic conductivities. Consistent with the droplet-based model, the pressure sensitivity increases with decreasing ionic conductivity and increasing excitation frequency (Fig. S34 in the ESM), validating the generality of sensitivity enhancement through modulating of these two parameters.

Hydrogel-based pressure sensors were subsequently fabricated (Fig. 6(a)). Capacitance-pressure characteristics recorded over a

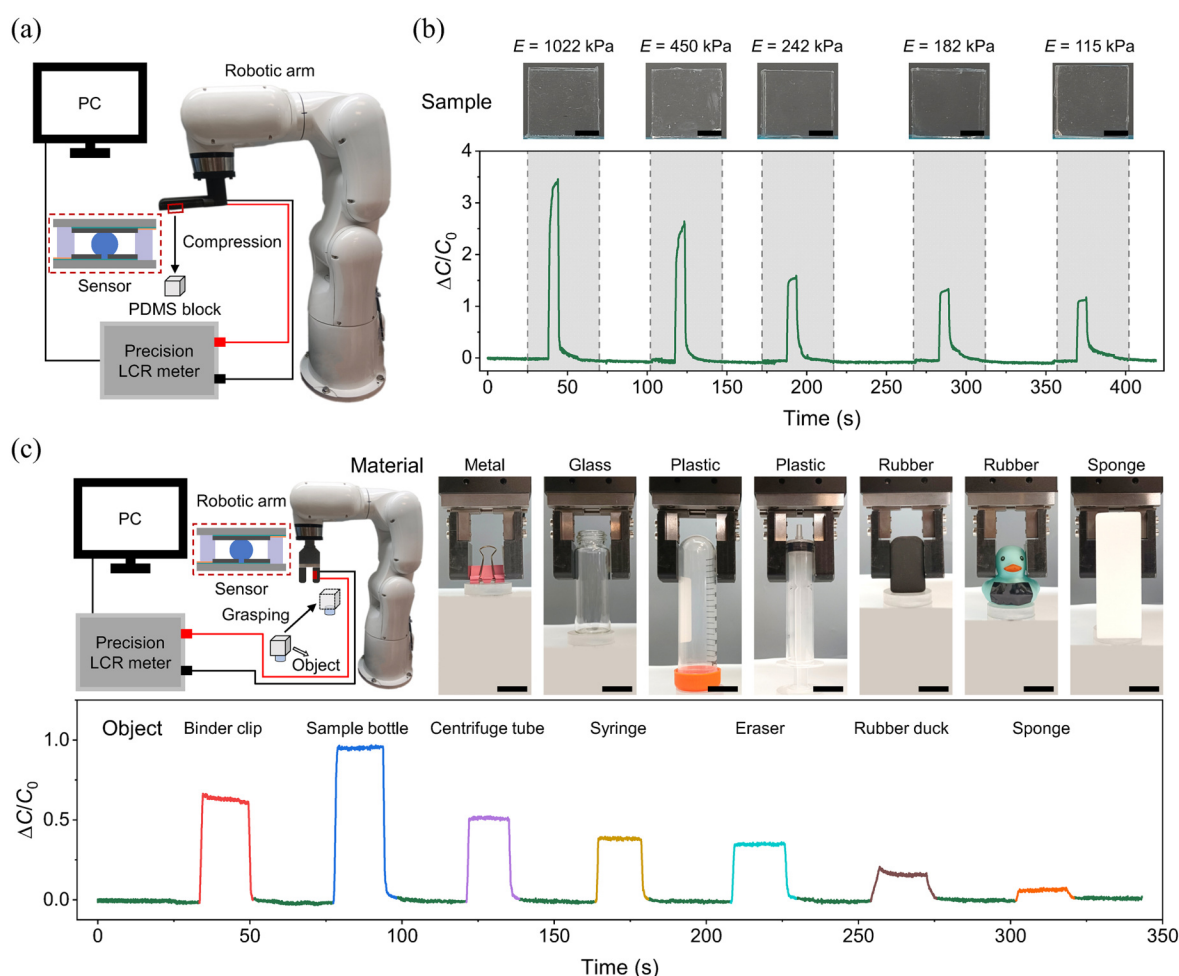


Figure 5 Application demonstrations of the sensor. (a) Schematic diagram of the robotic arm manipulation platform equipped with the sensor for pressing process. (b) PDMS blocks with different E and corresponding device signal variations during pressing. Scale bars: 5 mm. (c) Schematic diagram of the robotic arm manipulation platform equipped with the sensor for grasping, daily objects and corresponding device signal variations during the process. Scale bars: 20 mm.

broad frequency range further confirm that lower ionic conductivity and higher excitation frequency synergistically amplify pressure sensitivity (Figs. 6(b) and 6(c), and Fig. S35 in the ESM). These results establish a unified sensing model applicable to both liquid and solid-state ionic systems. An optimized hydrogel composition ($6.9 \text{ mS}\cdot\text{cm}^{-1}$) operated at 10 kHz was therefore selected for subsequent characterization, including response/recovery time, detection limit, and pressure resolution (Fig. S36 in the ESM). Owing to the high mechanical stability of the hydrogel, the device maintains stable sensing performance under repeated compression, stretching, and bending cycles (Fig. S37 in the ESM).

To demonstrate scalability and spatial sensing capability, a hydrogel sensor array of 4×4 with a pixel size of 2 mm was fabricated (Fig. 6(d)). Fully elastic materials and stretchable serpentine electrodes impart excellent deformability, allowing stable operation under stretching and bending (Figs. 6(e) and 6(f)). The array enables spatial pressure mapping, highlighting its potential for distributed tactile sensing application (Fig. 6(g)).

3 Conclusions

In summary, we established an ionic-droplet-based research model platform and systematically elucidated how ionic conductivity and excitation frequency regulate iontronic sensing behavior. By

integrating EIS measurements with an equivalent-circuit analysis, we derived analytical expressions linking apparent capacitance variation to these two parameters. The excellent agreement between theory and experiment validates the model and reveals a clear mechanism: Pressure sensitivity is enhanced at lower ionic conductivity and higher excitation frequency.

Beyond fundamental insights, tuning these parameters alone, without any structural modification, led to an enhancement of 8772% in sensitivity. Under optimized conditions, the sensor delivered stable and repeatable responses and enabled real-time discrimination of materials with different stiffness when integrated into robotic systems, underscoring its potential for tactile perception, material classification, and human-machine interaction in next-generation soft and humanoid robotics. By replacing ionic-droplets with hydrogels, the model sensor was successfully extended to solid-state ionic systems, with conclusions consistent with those obtained from droplet-based devices. Furthermore, an array of 4×4 of hydrogel pressure sensors featuring a pixel size of 2 mm was fabricated and demonstrated for spatial pressure mapping application.

This model framework is broadly applicable to iontronic sensing platforms, offering a generalizable route for performance optimization through controlled modulation of ionic conductivity and measurement frequency. Unlike previous efforts that primarily

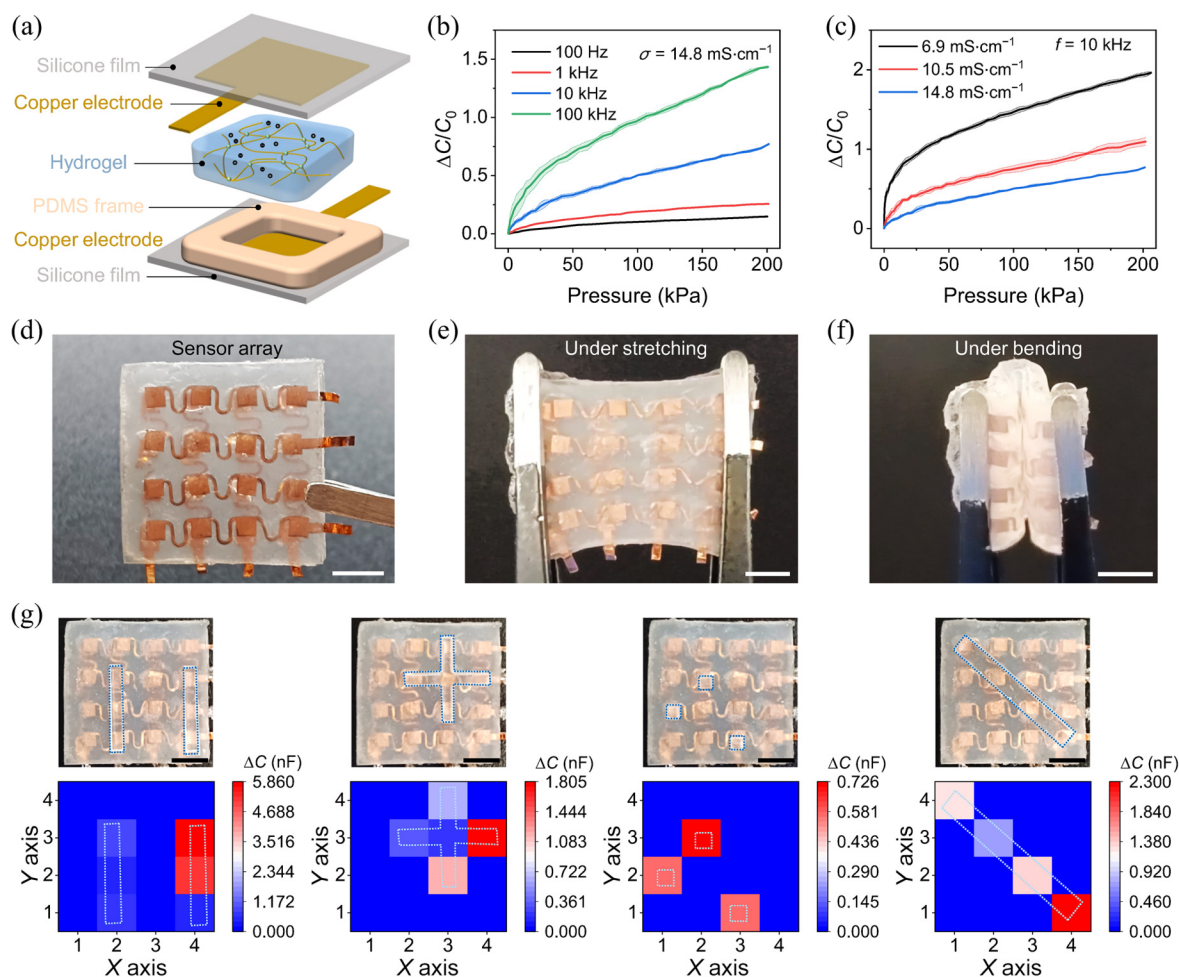


Figure 6 Hydrogel device and its application demonstration. (a) Schematic diagram of the hydrogel pressure sensor structure. ((b) and (c)) Capacitance variation versus pressure curves for devices (b) at different frequencies and (c) with hydrogels of varying ionic conductivities. (d) Optical image of a 4×4 array based on hydrogel pressure sensors. ((e) and (f)) Demonstration of the array device under (e) stretching and (f) bending. (g) Spatial pressure mapping demonstration of the array device. For a more pronounced effect, a weight of 185 g (not shown in the image) was placed on acrylic plates of different shapes. Scale bar: 5 mm.

advanced sensing performance through new materials or structural designs, our findings reveal that tuning these two parameters alone can markedly enhance sensitivity even in sensors with identical materials and architectures and the conclusions are primarily applicable to impedance-based capacitance readout schemes. Importantly, tuning ionic conductivity and excitation frequency does not alter the mechanical framework of the device. The resulting sensitivity enhancement therefore originates from impedance redistribution during electrical measurement. Optimizing these electrical parameters can thus effectively increase the measured sensitivity of iontronic pressure sensors. We believe that this work opens a complementary pathway for the development of high-performance iontronic sensing technologies, expanding the design space beyond conventional material and structural innovation.

4 Materials and methods

4.1 Fabrication of the sensor

The fabrication procedure is illustrated in Fig. S19 in the ESM. The sensor was assembled in a layer-by-layer configuration. A PET substrate served as the base layer, onto which cut-to-size copper

tape was adhered. Subsequently, anisotropic conductive film (ACF, 3M 9703) and hydrophobic carbon cloth (W1S1011, CeTech Co., Ltd.) were sequentially laminated. The upper section was completed by attaching the PET stack to a PMMA plate using double-sided adhesive. The lower section was fabricated using a similar approach, except that the carbon cloth included a central hydrophilic pore and was integrated with a PDMS (Dow Corning 184) frame. A 10 μL NaCl droplet was dispensed into the hydrophilic pore using a micropipette before aligning and encapsulating the upper and lower sections. Electrical connections were established by soldering wires to the upper and lower copper electrodes.

4.1.1 Regulation of droplet ionic conductivity

Sodium chloride (Tianjin Damao Chemical Reagent Factory) was dissolved in deionized water at varying concentrations to prepare droplets with different ionic conductivities. The ionic conductivity of each solution was measured using a conductivity meter (DDS-11A, Shanghai INESA Scientific Instrument Co., Ltd.). Each value represented the average of five repeated measurements.

4.1.2 PDMS frame and block preparation

Acrylic molds were fabricated to define the desired frame and block geometry firstly. The PDMS base and curing agents (agents A and

B) were mixed at specified mass ratios, followed by vacuum degassing to remove trapped air bubbles. The resulting mixture was then poured into the molds, degassed again, and cured at 50 °C for 8 h. After curing, the PDMS frames and blocks were carefully demolded for subsequent assembly and experiment.

4.1.3 Hydrogel-based sensor and array preparation

The hydrogel was prepared via a photopolymerization process. First, 18 wt.% acrylamide (99.0%, Shanghai Macklin Biochemical Co., Ltd.) was dissolved in deionized water, followed by sequential addition of 0.054 wt.% crosslinker N,N'-methylenebis(acrylamide) (MBA, 99.0%, Shanghai Aladdin Biochemical Technology Co., Ltd.), 0.05 wt.% photoinitiator lithium phenyl(2,4,6-trimethylbenzoyl)phosphine (LAP, 98.0%, Shanghai Aladdin Biochemical Technology Co., Ltd.), and NaCl solution at varying concentrations. The mixture was stirred to obtain a homogeneous precursor solution. Subsequently, using sandpaper as a template, the solution was exposed to ultraviolet (UV) light at 365 nm for 2 min to produce hydrogels with different ionic conductivities. The microstructure of the hydrogel was imaged using a trinocular stereo microscope (SOPTOP SZM704, Ningbo Sunyu Instrument Co., Ltd.). Stretchable electrodes were fabricated with a fiber laser marker (FOL-50M, Dongguan Hunst Laser Technology Co., Ltd.). Hydrogel pressure sensors and an array device of 4×4 were obtained through layer-by-layer assembly of silicone film, copper electrodes, hydrogels, and a PDMS frame.

4.2 Device performance characterization

4.2.1 Electrochemical impedance spectroscopy testing

Alternating current (AC) impedance measurements were performed using a precision LCR meter (TH2840B, Changzhou Tonghui Electronics Co., Ltd.) over a frequency range of 100 Hz–1 MHz.

4.2.2 Pressure sensing performance testing

Device characterization was performed using an electronic universal tester (EM2.502, Shenzhen Tesmart Instrument and Equipment Co., Ltd.) to apply and record mechanical compression. Capacitance measurements were acquired using the precision LCR meter at an AC voltage of 1 V root mean square (RMS).

4.2.3 Robotic arm manipulation platform

The finger and gripper models were fabricated using a light-curing three-dimensional (3D) printer (HALOT-SKY, Shenzhen CREALITY Co., Ltd.) and mounted onto a robotic arm (SJ603-A, RobotAnno (Shenzhen) Co., Ltd.) via screws. Capacitance signals were collected using the same precision LCR meter under a droplet ionic conductivity of $1.32 \text{ mS}\cdot\text{cm}^{-1}$, a frequency of 100 kHz, and an AC voltage of 1 V RMS.

4.3 Data processing

4.3.1 Electrochemical impedance spectroscopy fitting

Electrochemical impedance spectroscopy fitting: The resulting data were fitted and analyzed with ZView2 software to extract the relevant equivalent circuit parameters.

4.3.2 Formulas derivation

Detailed derivations are provided in Fig. S8 in the ESM. The

physical significance of all parameters, as described in the main text, was determined from electrochemical impedance spectroscopy measurements and subsequent data fitting.

4.4 FEA simulation

The FEA simulations were performed using COMSOL Multiphysics software. Specific simulation parameters were as follows: In the solid mechanics physics field, a linear elastic material model was used. The Young's modulus of PMMA was set to 3.3 GPa with a Poisson's ratio of 0.35; the Young's modulus of the copper electrode was set to 110 GPa with a Poisson's ratio of 0.3; the Young's modulus of the carbon cloth was set to 300 MPa with a Poisson's ratio of 0.2; and the Young's modulus of PDMS was set to 2.5 MPa with a Poisson's ratio of 0.49. The boundary between the droplet and the carbon cloth was defined as a contact condition, with the penalty function selected as the contact method and a penalty factor of 10. The bottom surface of the lower PMMA layer was assigned a fixed constraint, while a boundary load of pressure type was applied to the top surface of the upper PMMA layer.

In the dilute species transport physics field, the charge numbers of Na^+ and Cl^- were set to +1 and −1, with corresponding diffusion coefficients of 1.33×10^{-9} and $2.03 \times 10^{-9} \text{ m}^2\cdot\text{s}^{-1}$, respectively, and an initial concentration of $1 \text{ mol}\cdot\text{m}^{-3}$ for both ions. In the electrostatics physics field, charge conservation was enforced, and a voltage of 0 V was applied to the top electrode and 1 V to the bottom electrode.

The entire study employed a transient solver with a simulation time of 1 μs . By coupling the solid mechanics, dilute species transport, and electrostatics physics fields, the stress distribution, droplet deformation, and ion concentration distribution of the droplet-based device were successfully simulated under different pressure conditions.

Electronic Supplementary Material: Supplementary material (device fabrication process, performance characterization, application demonstration, electrochemical impedance spectroscopy analysis, equation derivation, supplementary tables and movies, etc) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908691>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Y. M. W.: Writing – original draft, writing – review & editing,

visualization, investigation, formal analysis, data curation. J. S. X.: Writing – review & editing, visualization, investigation. M. W. G.: Writing – review & editing. Y. Y.: Writing – review & editing. X. Y. M.: Writing – review & editing. L. Z.: Writing – review & editing. J. S. C.: Writing – review & editing. Y. J. L.: Writing – review & editing. J. H. G.: Writing – review & editing. J. G.: Conceived project idea, writing – revising & improving, conceptualization, methodology, supervision, funding acquisition. All the authors have approved the final manuscript.

Use of AI statement

None.

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