Electronic Supplementary Material (ESI) for Materials Horizons.

Supporting Information

An integrated electronic skin with multiple-dimensional sensibility from layered biphasic liquid metal-polymers

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Materials and Methods

Materials: Liquid metals (LM) (Ga, melting point: 29.8 °C; Sn, melting point: 231.9 °C) were purchased from Shenyang Jiabei Trading Company; PDMS (Sylgard 186, Dow Corning) was obtained from the Shanghai Educational Consumables Center; sodium chloride (NaCl) was purchased from China National Pharmaceutical Group Chemical Reagent Company; Ecoflex (00-35, Smooth-On) was acquired from Beijing Tiantong Huayi Company.

Methods

Preparation of biphasic liquid metal: Biphasic liquid metal $Ga_{0.3}Sn_{0.7}$ (at.) was a GaSn alloy formed by thoroughly stirring and mixing Ga and Sn (molar ratio 3:7) at 300 °C. During stirring, N_2 was continuously input to reduce the formation of metal oxides.

Preparation of conductive films: Firstly, 5.2 g $Ga_{0.3}Sn_{0.7}$ (at%) was mixed thoroughly with 0.56 g Ecoflex A, and then 0.56 g Ecoflex B was added. Grind and blend the mixture evenly. Pour the mixture into a mold, spread it evenly, and cure at room temperature for 5 minutes.

Preparation of PDMS foam: 26 g NaCl (80 vol%) was thoroughly mixed with 3.3 g PDMS (Sylgard 184 with a curing agent ratio of 10:1) and pressed into a mold. The mixture was then vacuum-dried at 100 °C for 2 hours. Subsequently, it was immersed in deionized water and soaked at 85 °C for 24 hours, followed by rinsing with deionized water. The final product was dried at 80 °C for 2 hours to remove any remaining moisture.

Preparation of integrand electronic skin: As illustrated in **Figure S1**, before the conductive film was completely cured, PDMS foam was embedded between two layers of the film to create a "sandwich" structure. The entire assembly was cured at 60 °C for 30 minutes. Additionally, sensors with a dielectric layer made of PDMS (without pores) were prepared as control samples and referred to as p-IES.

Characterization

Morphology characterization: Observation of the Morphological Features of the Electrode and Dielectric Layers of SSMFS under Tensile and Compressive Stimuli using Scanning Electron Microscope (FEI 3D) and Optical Microscope (XTL-16B).

Measurement of sensing performance: At ambient temperature (25 oC), compression and tensile forces were simulated using a tensile testing machine (MTS E42). Real-time monitoring of the capacitance and resistance signals of SSMFS was conducted using a digital multimeter (Keysight 34461A).

Evaluation of mechanical performance: Compressive and tensile tests were conducted on a tensile testing machine (MTS E42) at a speed of 0.2 mm·s-1 under ambient conditions. All reported mechanical performance values are averages based on at least three independent measurements for each sample.

-Supplementary Discussion I: The preparation process of IES.

-Supplementary Discussion II: Electrical performance and morphology illustrations for IES.

-Supplementary Discussion III: The wearable applications of IES for simulating natural skin.

Supplementary Discussion I: The preparation process of IES.

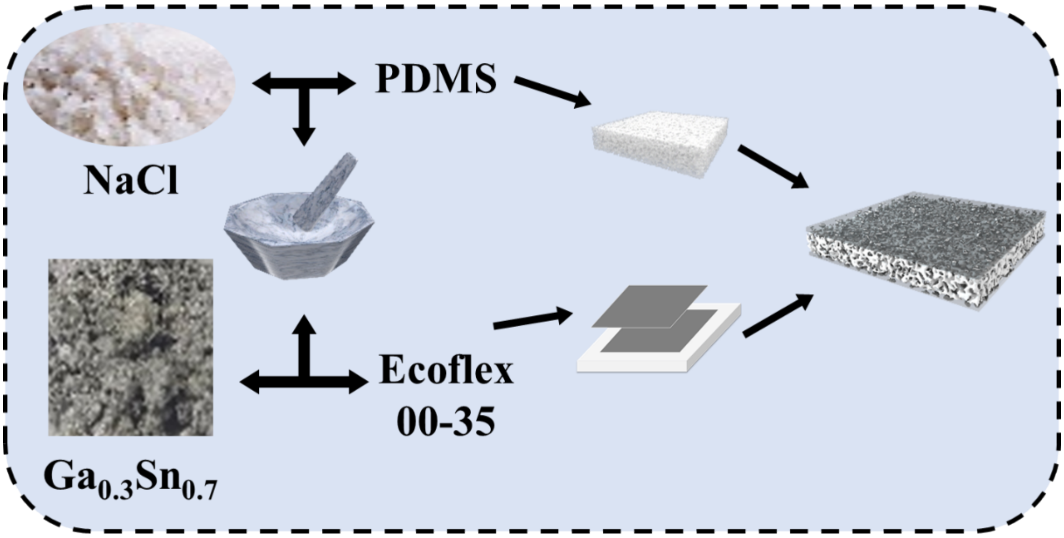

Figure S1. Schematic of the preparation process for flexible integrated electronic skin. Porous insulating foam was prepared by incorporating sodium chloride (NaCl) through grinding in polydimethylsiloxane (PDMS). Additionally, a biphasic liquid metal ($Ga_{0.3}Sn_{0.7}$, at%) and silicone rubber elastomer (Ecoflex), after thorough grinding and mixing, were cured into conductive films in a rectangular polytetrafluoroethylene mold. The flexible integrated electronic skin, IES, was conveniently prepared by sandwiching a layer of insulating foam between two layers of conductive film.

Supplementary Discussion II: Electrical performance and morphology illustrations for IES.

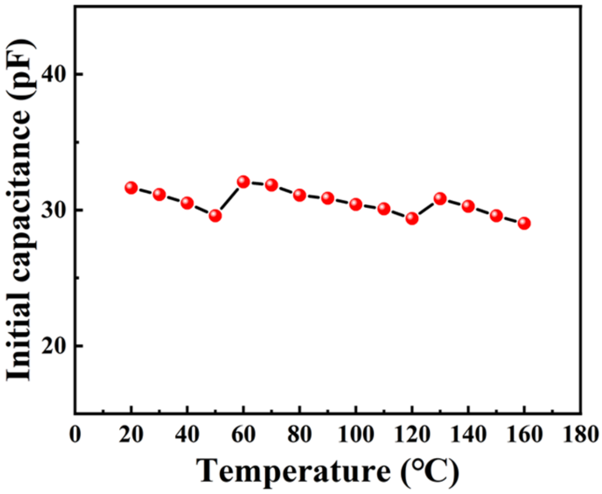

Figure S2. The initial capacitances of the IES were measured at intervals of 10 °C in an environmental range from 20 °C to 160 °C. The capacitances obtained above all fall within the range of 29 to 31 pF. Therefore, the initial capacitances of the IES in this study (30 pF) were not influenced by external environmental temperatures, demonstrating excellent environmental stability.

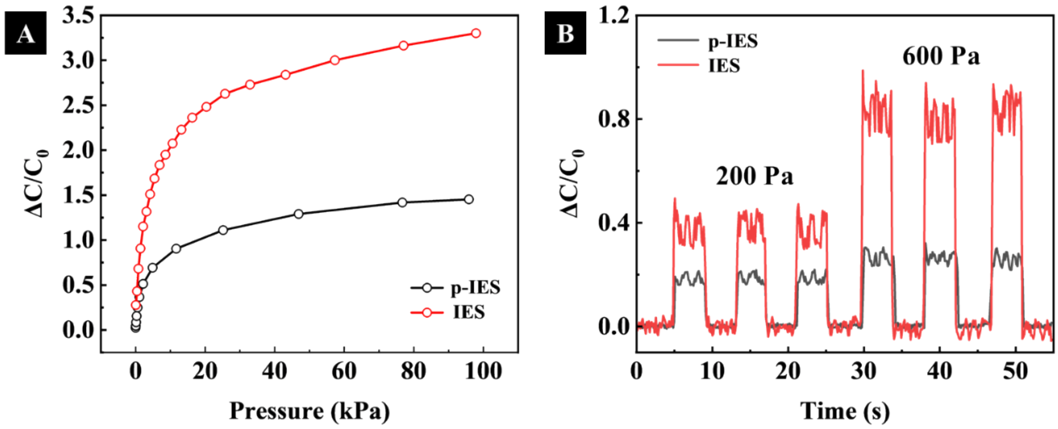

Figure S3. The capacitance transmission capabilities between p-IES and IES were compared. The dielectric layer of p-IES lacked pores, with the dielectric material being solely PDMS, whereas the dielectric layer of IES consisted of porous PDMS foam, with the dielectric material being a combination of air and PDMS. **(A)** Under dynamic pressure ranging from 0 to 100 kPa, the maximum capacitive response value ($\Delta C/C_0$) of p-IES was significantly lower than that of IES, measuring 1.2 and 3.2, respectively. **(B)** Similar to **Figure 3C**, cyclic pressure loading-unloading tests were conducted on p-IES and IES at 200 Pa and 600 Pa. The results indicated that at the same pressure level, the $\Delta C/C_0$ of p-IES was only half of that of IES. Due to the differences in capacitive response performance mentioned above, the use of porous PDMS foam as the dielectric layer for IES was the most reasonable choice.

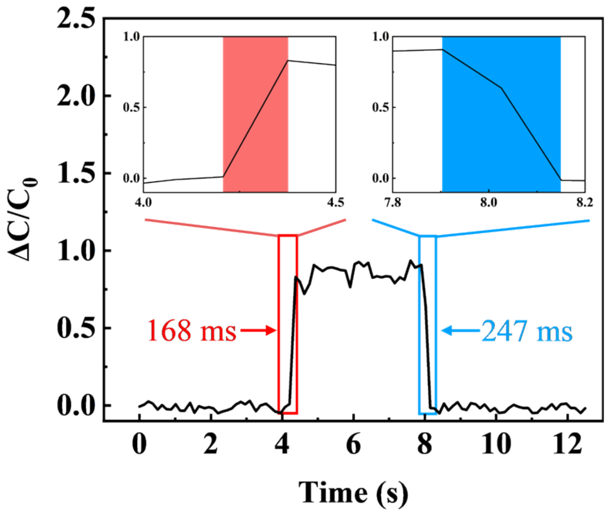

Figure S4. The capacitive response time of IES under 600 Pa. When subjected to a 600 Pa pressure load, the IES responded to the increase in pressure with an increase in capacitance after 168 ms. Upon pressure unloading, the IES required 247 ms to restore the capacitive value to its initial state. The red and blue portions in the illustration respectively depicted the capacitance variations of the IES during pressure loading and unloading.

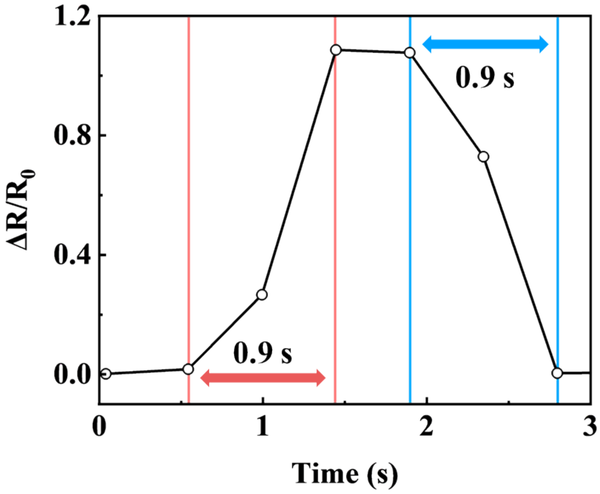

Figure S5. The resistive response time of IES under 200% strain. After applying a 200% strain to the IES and rapidly unloading the additional tension, the resistance signal of the IES required 0.9 seconds to respond to the change in tension.

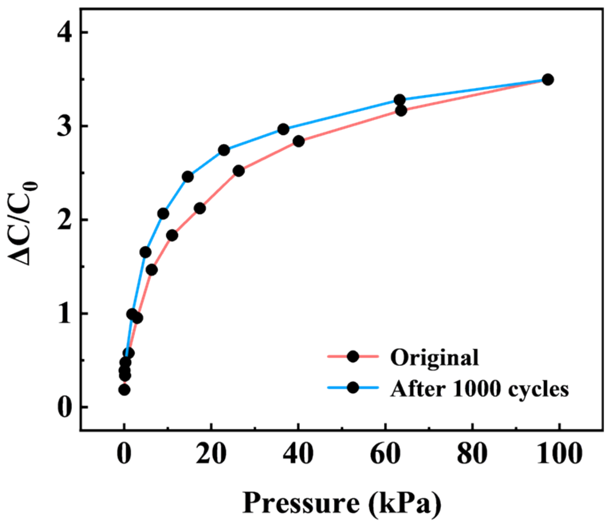

Figure S6. Under pressure from 0 to 100 kPa, the initial capacitive response curve of the IES (red) and the capacitive response curve of the IES (blue) after 1000 times cycles of pressure loading and unloading at 10 kPa. Obviously, the maximum capacitive response value for both two curves was 3.4. However, under the same pressure, the capacitance response value of the blue curve was greater than that of the red curve. Compared to the initial state (red curve), the 1000 times compressive cycles made the insulating dielectric layer (PDMS foam) in the IES more compressible. Based on equation (1), under the same pressure, the distance (*d*) between the conductive layers in the IES corresponding to the blue curve was larger than that of the red curve, leading to a larger capacitive response value.

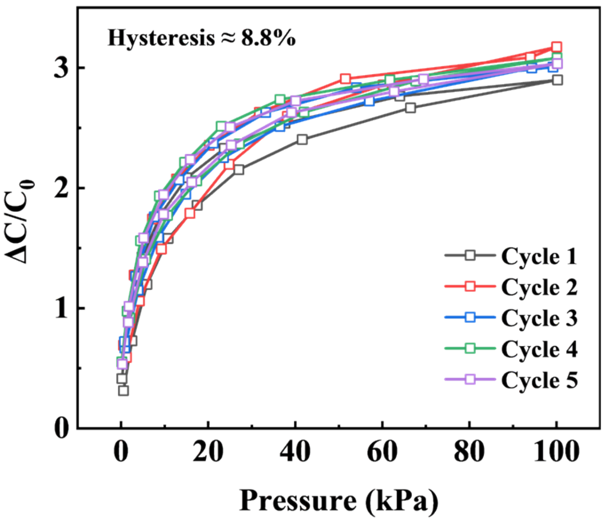

Figure S7. Under 0 to 100 kPa, the capacitive response hysteresis rate was calculated by the response curves from 5 times compressive loading-unloading cycles. The hysteresis rate, defined as the ratio of the maximum difference between loading and unloading curves to the full-scale range, was approximately 8.8% for IES.[1] The lower hysteresis rate indicated that IES still maintained good response stability after pressure loading and unloading at 100 kPa.

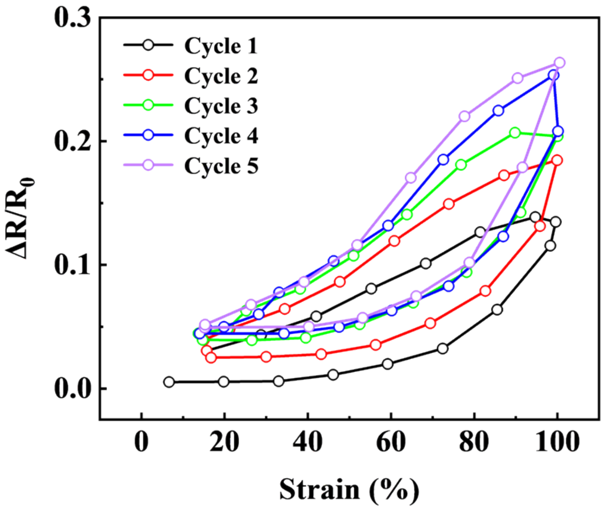

Figure S8. In the loading-unloading cycle, tension was applied to IES 5 times under 0 to 100% strain. As the number of cycles increased, the maximum $\Delta R/R_0$ value of the hysteresis loop increased from 0.1 to 0.25. Due to the biphasic state of the $Ga_{0.3}Sn_{0.7}$ (at%) in the conductive films, which possessed a certain degree of mobility, it could continuously reorganize in response to changing external tension.

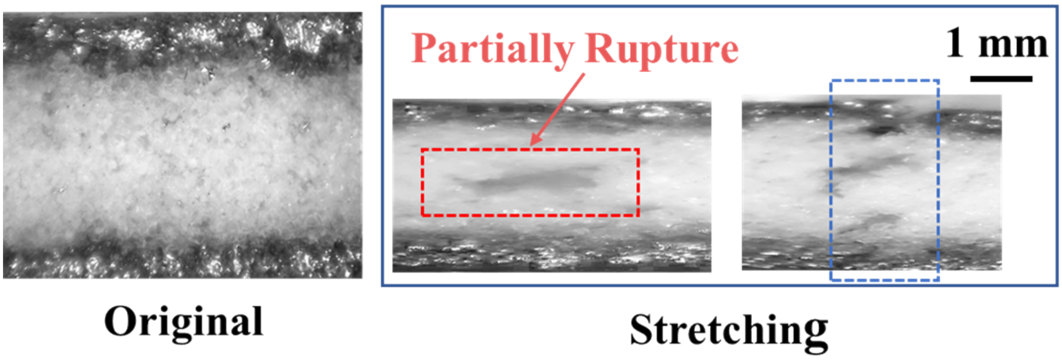

Figure S9. Microscopic images of IES in the original and stretched states, scale bar was 1 mm. During the stretching process, localized tearing first occurred in the PDMS foam dielectric layer of IES (white, middle layer). The electrode layers (gray-black, upper and lower layers) relied on their excellent flexibility and experienced damage after the external tension reached its limit.

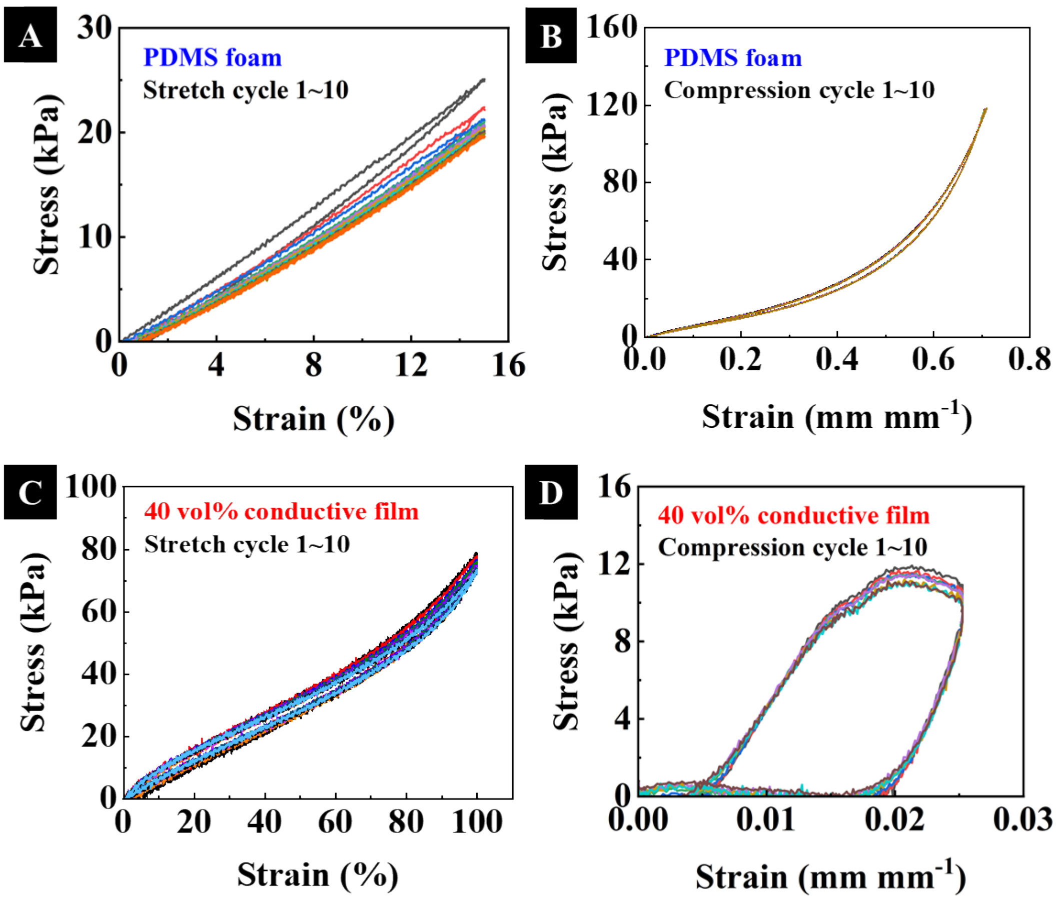

Figure S10. The fundamental mechanical properties of the dielectric layer (PDMS foam) and the electrode layer (biphasic liquid metal conductive film with a metal volume fraction of 40%) constitute the IES. **(A)** The cyclic tensile test of PDMS foam under 25% strain. **(B)** The cyclic compressive test of PDMS foam at 120 kPa. **(C)** The cyclic tensile test of the conductive film under 100% strain. **(D)** The cyclic compressive test of conductive film at 12 kPa. Both the electrode layer and the dielectric layer exhibited excellent mechanical recovery performance.

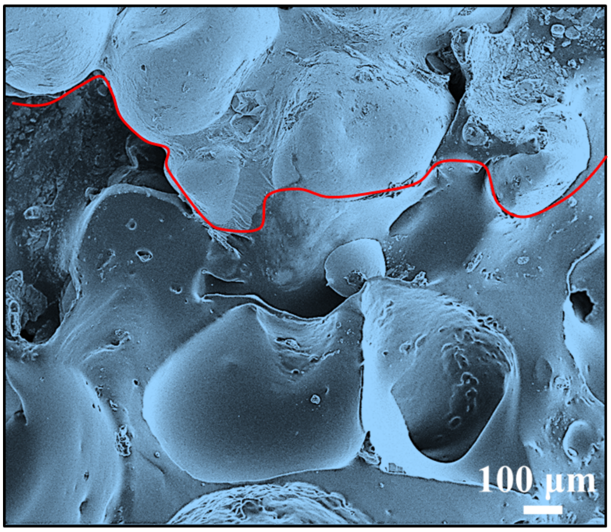

Figure S11. SEM image of the interface between the conductive film (mixture of Ecoflex and biphasic liquid metal $Ga_{0.3}Sn_{0.7}$, at%) and the PDMS foam, scale bar was 100 µm. The red curve was used to distinguish between the electrode layer and the dielectric layer. The adhesive force between the two interfaces arises from the physical bonding between the cured Ecoflex and the PDMS layer.

Supplementary Discussion III: The wearable applications of IES for simulating natural skin.

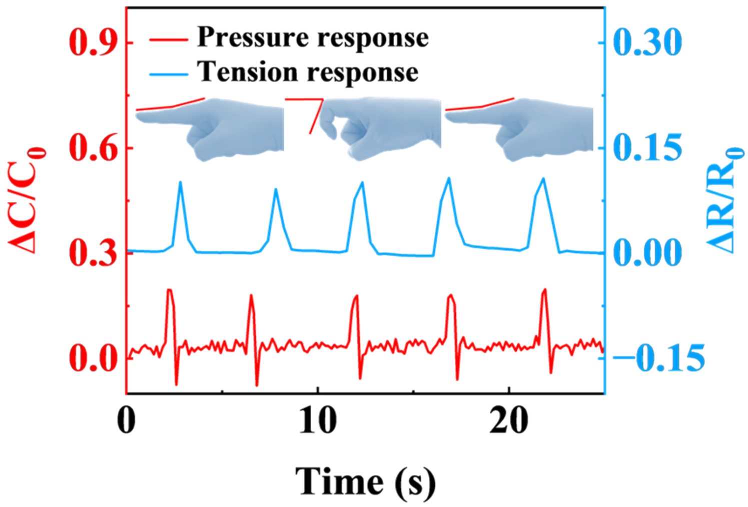

Figure S12. IES exhibited dual-response peaks in capacitance and resistance in response to the bending motion of a finger. A piece of IES measuring 3 cm (length) × 1 cm (width) × 5 mm (thickness) was affixed at the joint of the index finger. The IES responses were recorded in real-time while the index finger performed 5 times horizontal bending motions within 25 seconds which was shown in the insets. The experiments indicated that IES could perfectly respond to external force variations through the dual signals of capacitance and resistance.

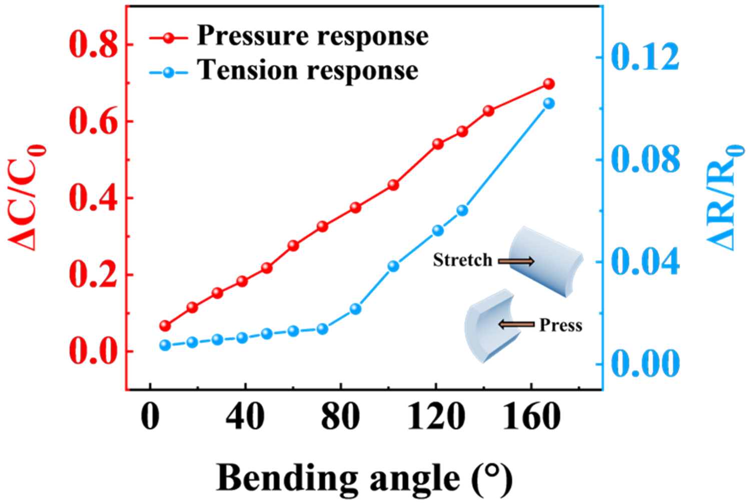

Figure S13. Applying bending stress to the IES at intervals of 10° (bending angle), while recording the response values of $\Delta C/C_0$ and $\Delta R/R_0$ of the IES. The illustration demonstrated that the bending stress applied to the IES could be decomposed into compression on the inner side and tension on the outer side. With the bending angle increasing from 0° to 160°, the $\Delta C/C_0$ and $\Delta R/R_0$ increased from 0 to 0.7 and 0 to 0.1, respectively. Therefore, as an e-skin possessing dual-response capabilities of capacitance and resistance, the IES was able to instantaneously and accurately perceive external bending action.

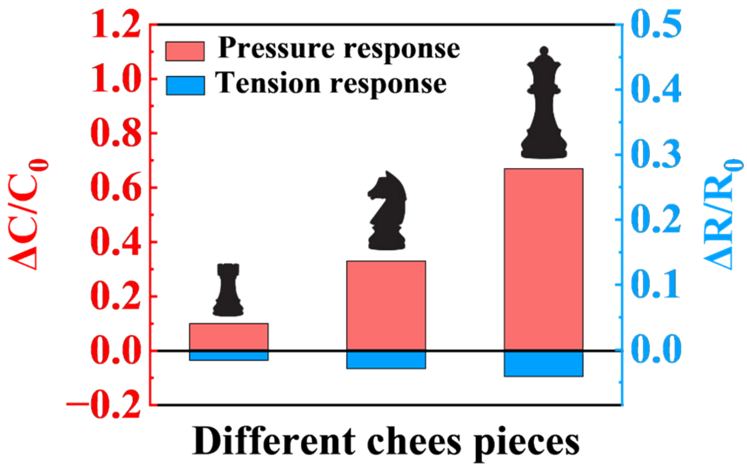

Figure S14. By statically placing chess pieces with varying masses and different base diameters, IES is capable of discriminating and distinguishing them through capacitive and resistive signals. Chess pieces of different masses (10 g, 20 g, and 30 g) and base diameters (17 mm, 21 mm, and 27 mm) were statically placed on IES, and $\Delta C/C_0$ (red column heights) and $\Delta R/R_0$ (blue column heights) were recorded. For example, the $\Delta C/C_0$ of the "rook" chess piece (mass 10 g, base diameter 17 mm) was 0.1, and the $\Delta R/R_0$ was -0.02. The $\Delta C/C_0$ of the "queen" chess piece (mass 30 g, base diameter 27 mm) was 0.7, and the $\Delta R/R_0$ was -0.1. Due to the differences in the mass and base diameter of the chess pieces, variations in the pressure applied by the chess pieces on IES (vertical) and the range of pressure application (horizontal) differed. This allows IES to discriminate between different chess pieces based on $\Delta C/C_0$ and $\Delta R/R_0$.

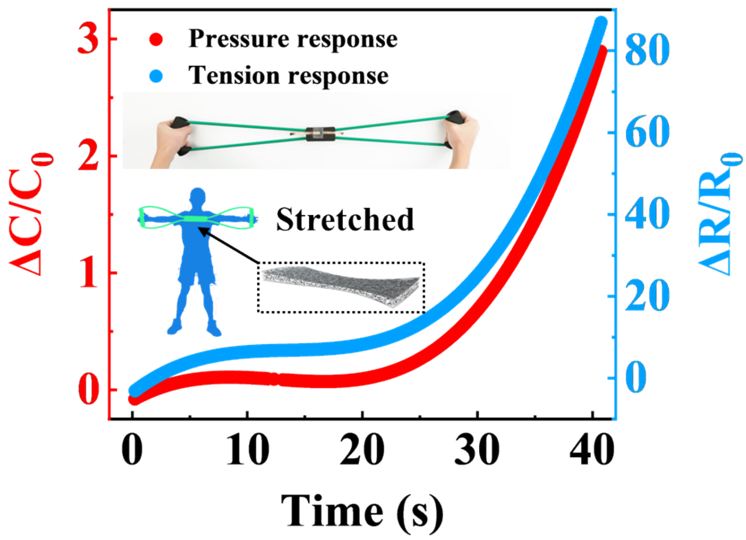

Figure S15. After attaching IES to the exercise equipment, IES is capable of monitoring the motion status of the device in real-time under high strain (over 200% strain) by changes in $\Delta C/C_0$ and $\Delta R/R_0$. The illustration depicts the experimental setup and the morphological changes of the IES during the stretching exercise. Within 40 seconds, the IES, along with the exercise equipment, is gradually stretched to 207% strain. $\Delta C/C_0$ and $\Delta R/R_0$ increase from 0 to 2.8 and 80, respectively. Obviously, similar to **Figure 2F**, both capacitance and resistance can continuously and simultaneously generate responses as tension increases. Therefore, within the stretching limit, the IES can stably generate dual responses to achieve real-time perception of dynamic tension.

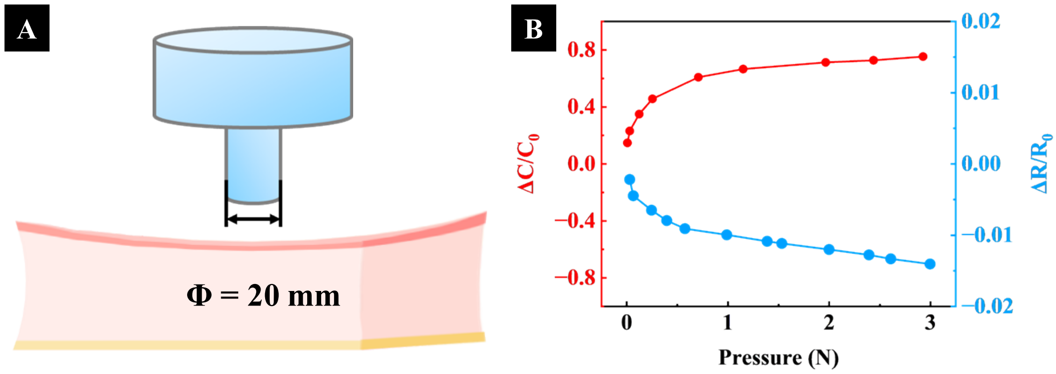

Figure S16. IES can establish a correlation with dynamic pressure through capacitance and resistance signals. **(A)** When pressing natural skin with a mold (diameter 20 mm), the pressure ranges could be perceived between the mold and the skin. **(B)** Based on the scenario depicted in Figure S16A, the pressure was applied to IES using the molds, capturing the pressure intensity range (0 to 3 N). Clearly, the capacitive response values increased to 0.8 and the resistive respone value decreased to -0.015. The pressure applied to IES was capable of displacing air and causing a number of semi-solid metal particles to be compressed and brought closer together. Therefore, the biaxial response capabilities of IES enhanced the integrated sensor's ability to identify individual mechanical stimuli.

Reference

1. L. Pan, A. Chortos, G. Yu, Y. Wang, S. Isaacson, R. Allen, Y. Shi, R. Dauskardt and Z. Bao, *Nat. Commun.*, 2014, **5**, 3002.