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**Supplementary information for**

# Ionotronic thermometry

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**This PDF file includes:**

Figures S1 to S10
Supplementary Notes 1 to 5
Description for Movie S1

**Other supplementary materials for this manuscript include the following:**

Movie S1

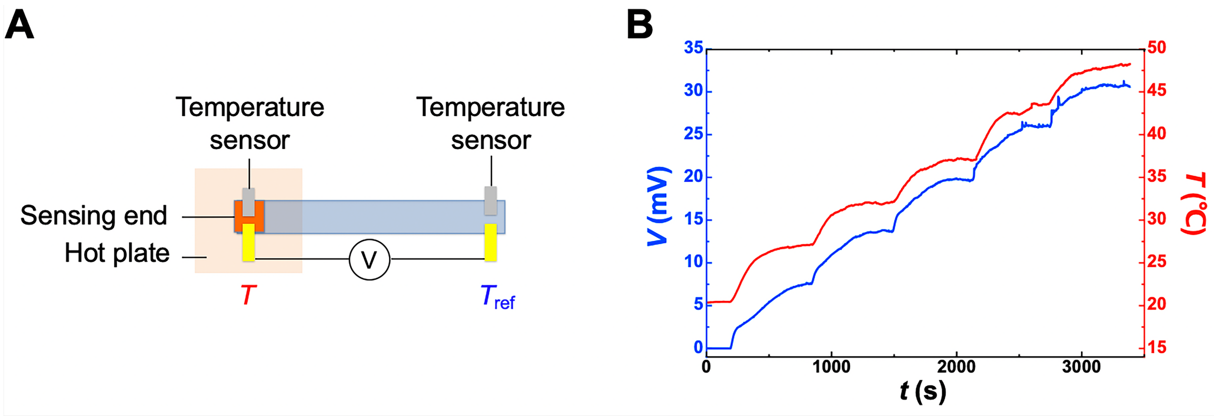

**Fig. S1.** Experimental setup for characterization of the ionotronic thermometer. (*A*) The sensing end is placed on a hot plate, and the reference end is connected to a voltmeter in open air. The temperatures at the sensing end and reference end are separately measured by using commercial temperature sensors. When temperature at the sensing end changes, a change in the open-circuit voltage is recorded. (*B*) Profiles of the voltage recorded by the voltmeter and the temperature of the sensing end recorded by the temperature sensor.

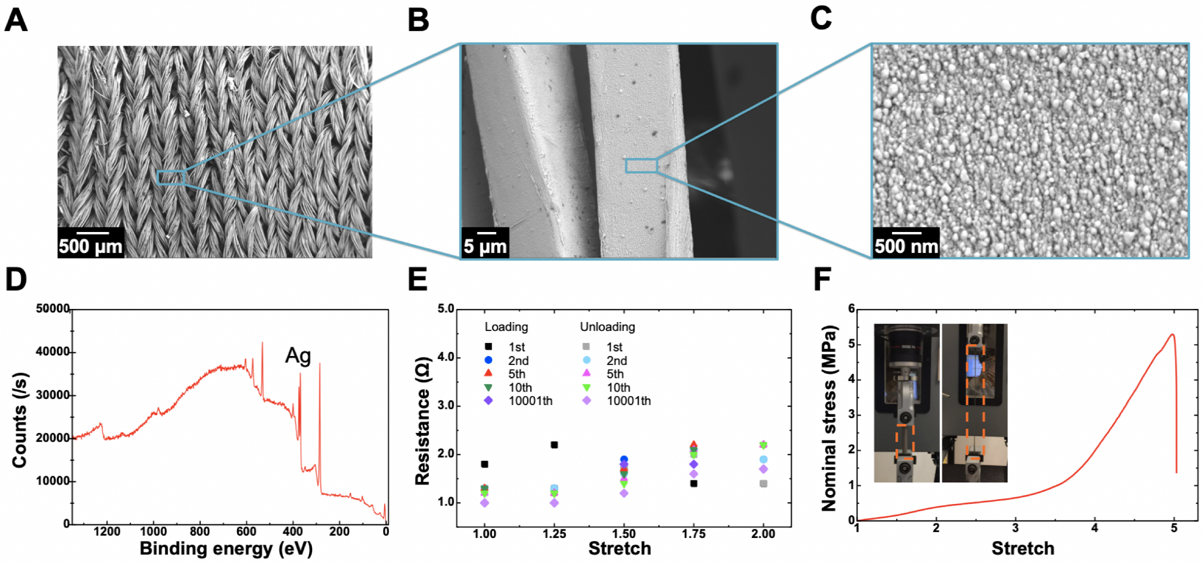

**Fig. S2.** Characterization of the silver-plated fabric. (*A*), (*B*), (*C*) SEM images at three magnifications. (*D*) X-ray photoelectron spectroscopy (XPS) of the fabric. The peak at binding energy of 368 eV indicates the existence of silver. (*E*) Resistance of the fabric is recorded as a function of stretch under cyclic loading. The sample is of length 2 cm, width 1.3 cm, and thickness 0.45 mm. (*F*) The stress-stretch curve of the fabric under uniaxial tension. Inset: photos of the fabric in the unstretched state and the stretched state.

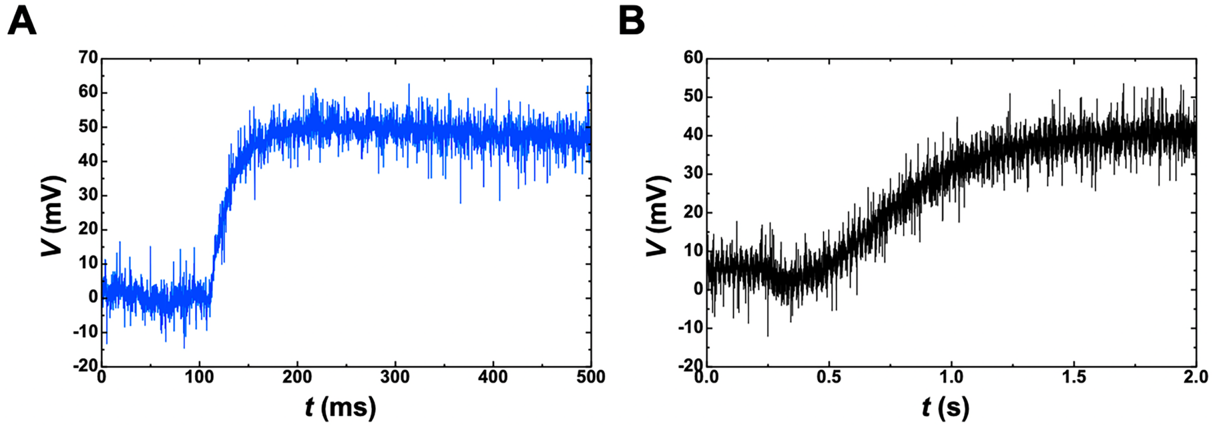

**Fig. S3.** Thermal response of sealed ionotronic thermometer. Voltage recorded as a function of time for a sensor (the design in Fig. 2a) sealed by (*A*) pre-stretched VHB (9473, 3M; ~10 μm thickness) and (*B*) VHB (9473, 3M; 100 μm thickness). In both cases, the ionic conductor is PAAm hydrogel containing 0.03 mol/L NaCl and the two electronic conductors are gold.

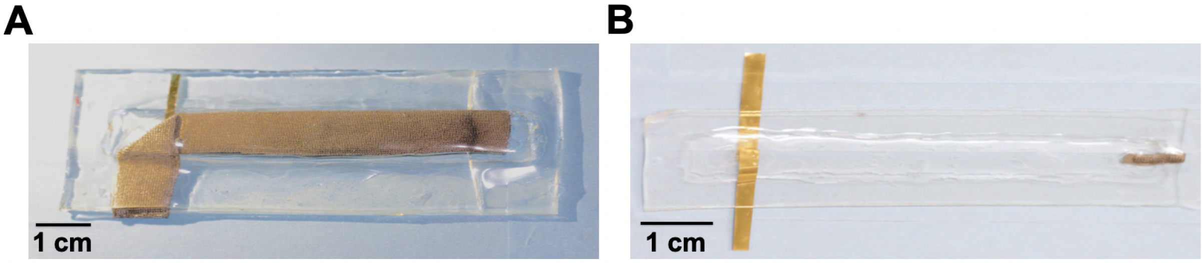

**Fig. S4.** (*A*) Photo of a stretchable ionotronic thermometer (the design in Fig. 3*A*). (*B*) Photo of a stretchable and transparent ionotronic thermometer (the design in Fig. 3*C*).

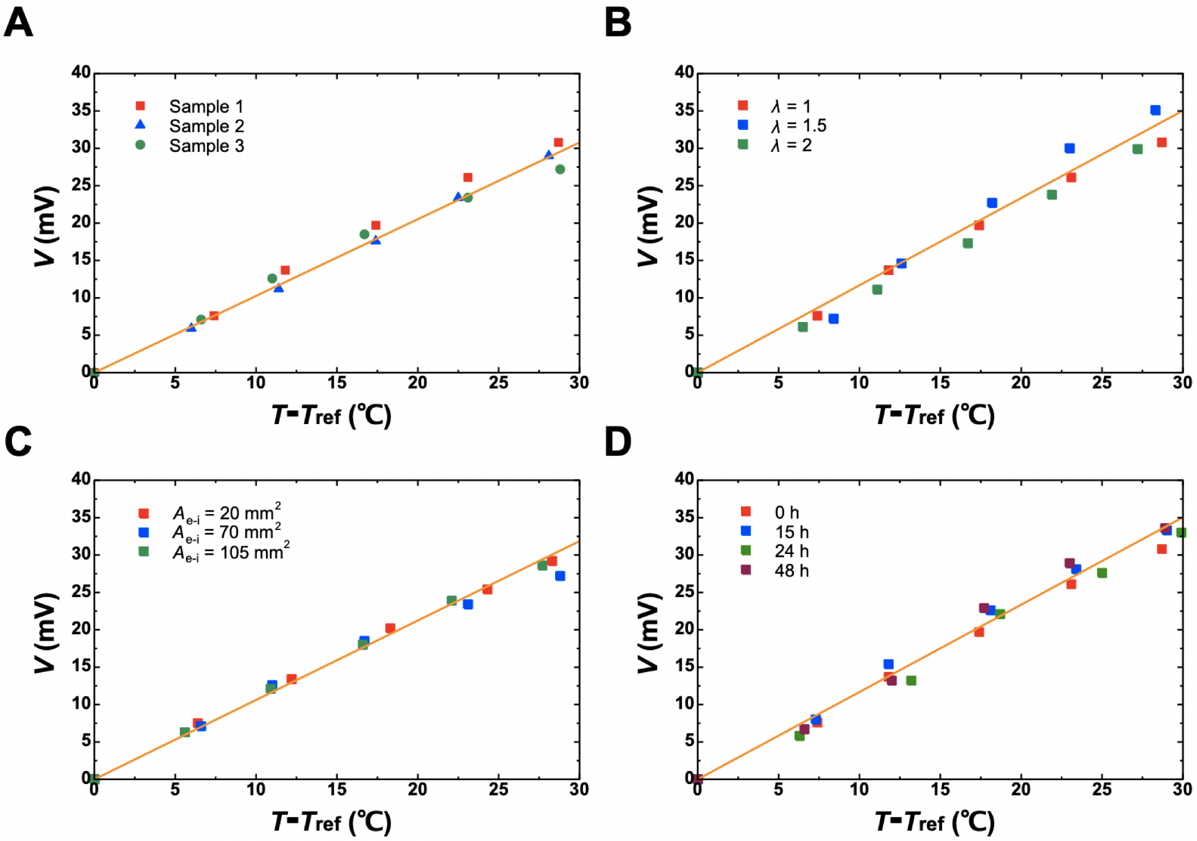

**Fig. S5.** Voltage-temperature relation of a stretchable and transparent ionotronic thermometer (the design in Fig. 3*C*). The open-circuit voltage $V$ is measured at the reference end using a voltmeter. The temperatures at the sensing end and the reference end, $T$ and $T_{\mathrm{ref}}$, are measured using two commercial temperature sensors. Each solid line is a linear fit to the data. (*A*) Voltage-temperature relation. (*B*) A stretch of the e-i junction at the sensing end, $\lambda$, negligibly affects the voltage-temperature relation. (*C*) The area of the e-i junction at the sensing end, $A_{\mathrm{e-i}}$, does not affect the sensitivity. The area of the e-i junction at the reference end is fixed at $28\ \mathrm{mm}^2$. (*D*) The voltage-temperature relation is stable over time.

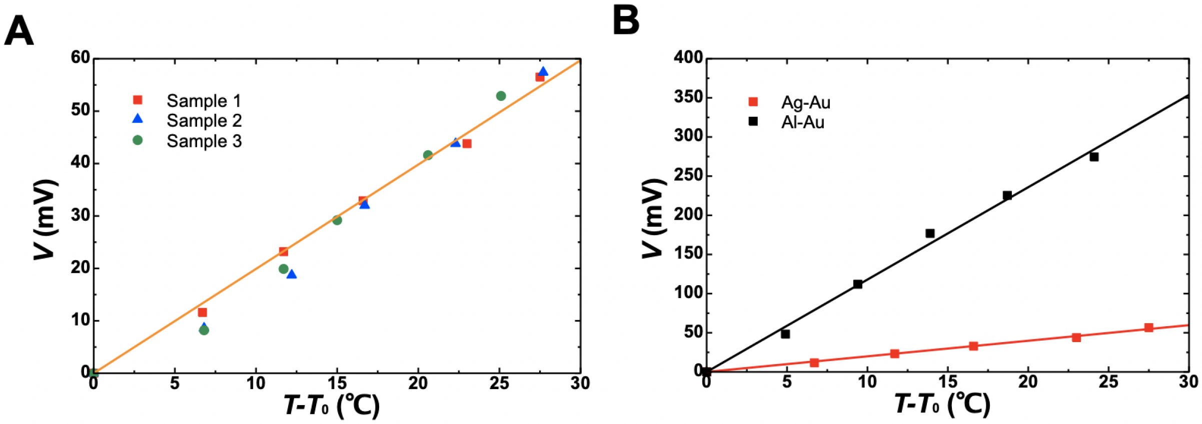

**Fig. S6.** Voltage-temperature relation of an e-i-e junction (the design in Fig. 3*E*). The open-circuit voltage $V$ is recorded using a voltmeter. The temperature is recorded using a commercial thermometer. $T_0$ is set to be the room temperature. Each solid line is a linear fit to the data. (*A*) Voltage-temperature relation. Silver-plated fabric and gold-coated PET are used as the two electronic conductors to sandwich the hydrogel. (*B*) Sensitivity depends on the type of the electronic conductors.

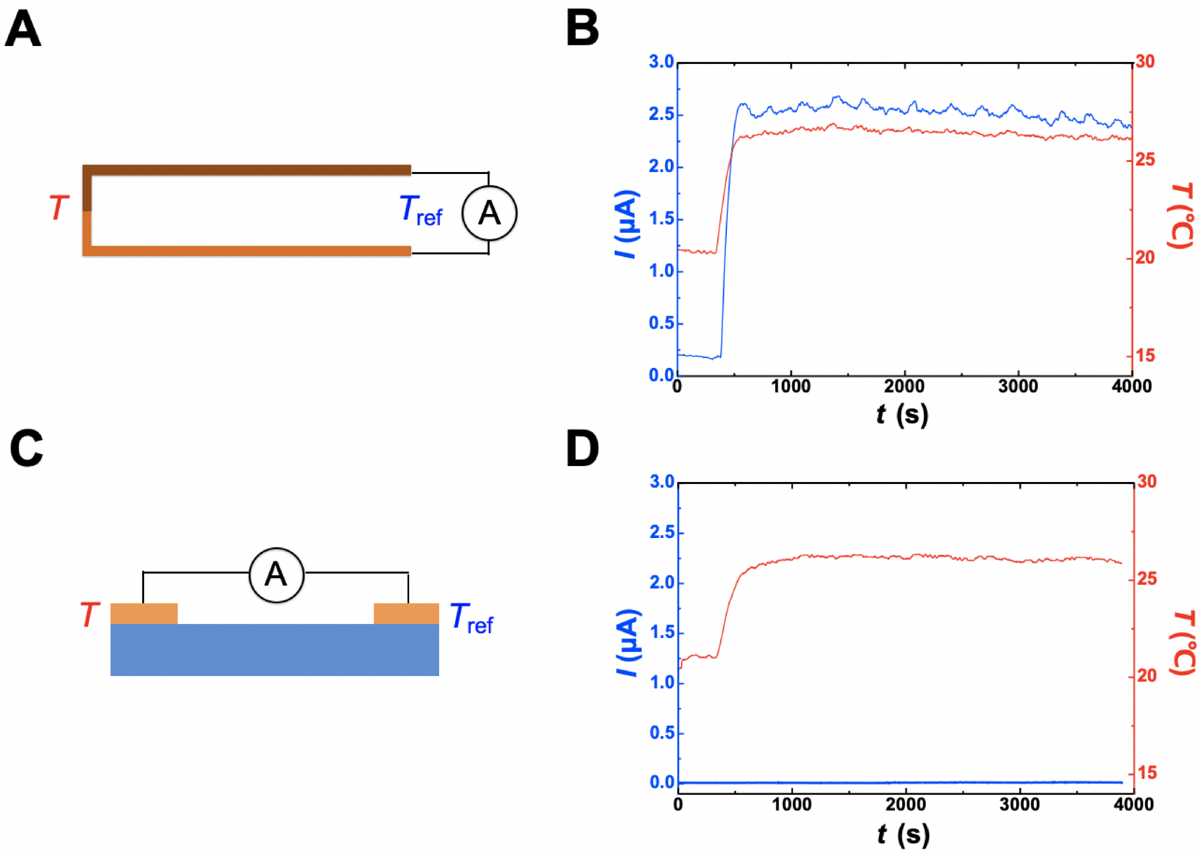

**Fig. S7.** Short-circuit current of a thermocouple and an ionotronic thermometer. (*A*) A K type thermocouple is made of two alloys, chromel and alumel, and is connected to an ammeter. (*B*) When the temperature at the sensing end increases and the temperature at the reference end is unchanged, a current is generated. When the temperature difference is maintained, the current is steady. (*C*) An ionotronic thermometer is made of PAAm hydrogel containing 0.03 mol/L NaCl as the ionic conductor, and silver-plated fabric and gold-coated PET as the electronic conductors at the sensing and reference ends, respectively. (*D*) When the temperature at the sensing end increases and the temperature at the reference end is unchanged, negligible current is generated. In both (*B*) and (*D*), the temperature is independently measured using a commercial resistive thermometer.

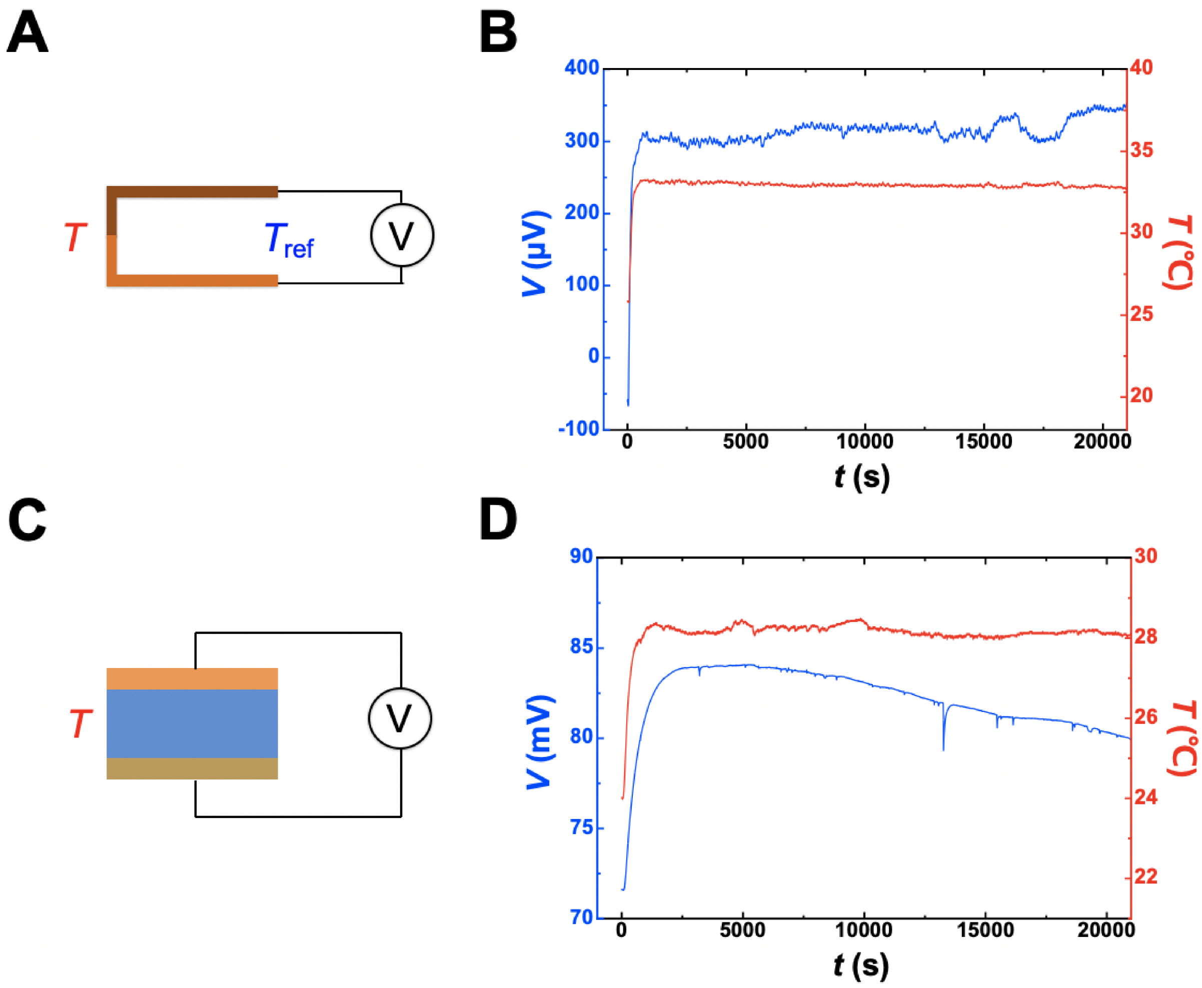

**Fig. S8.** Open-circuit voltage of a thermocouple and an ionotronic thermometer. (*A*) A K type thermocouple is made of two alloys, chromel and alumel, and is connected to a voltmeter. (*B*) The thermocouple maintains a stable voltage when the two ends are kept at a fixed temperature difference. (*C*) An e-i-e junction is made of PAAm hydrogel containing 0.03 mol/L NaCl as the ionic conductor, and silver-plated fabric and gold-coated PET as the two electronic conductors. (*D*) The voltage starts to drop after the junction is connected to a voltmeter for a long time. The sensitivity is on the order of 1 mV/K for the e-i-e junction, and $10^{-2}$ mV/K for the thermocouple. In both (*B*) and (*D*), the temperature is independently measured using a commercial resistive thermometer.

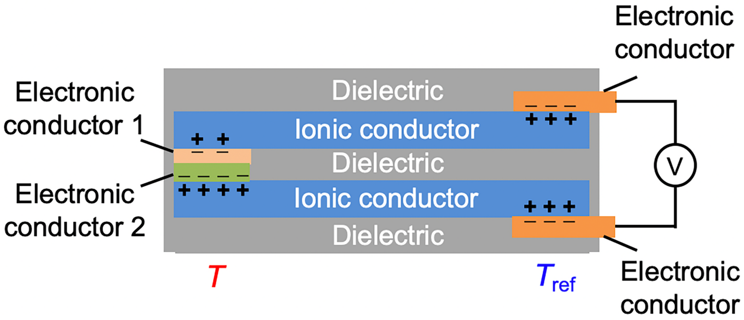

**Fig. S9.** An alternative design of stretchable and transparent ionotronic thermometry. Two dissimilar electronic conductors are placed in contact, and are connected to two stripes of ionic conductor. The two ionic conductors can have either the same ionic concentrations or different ionic concentrations.

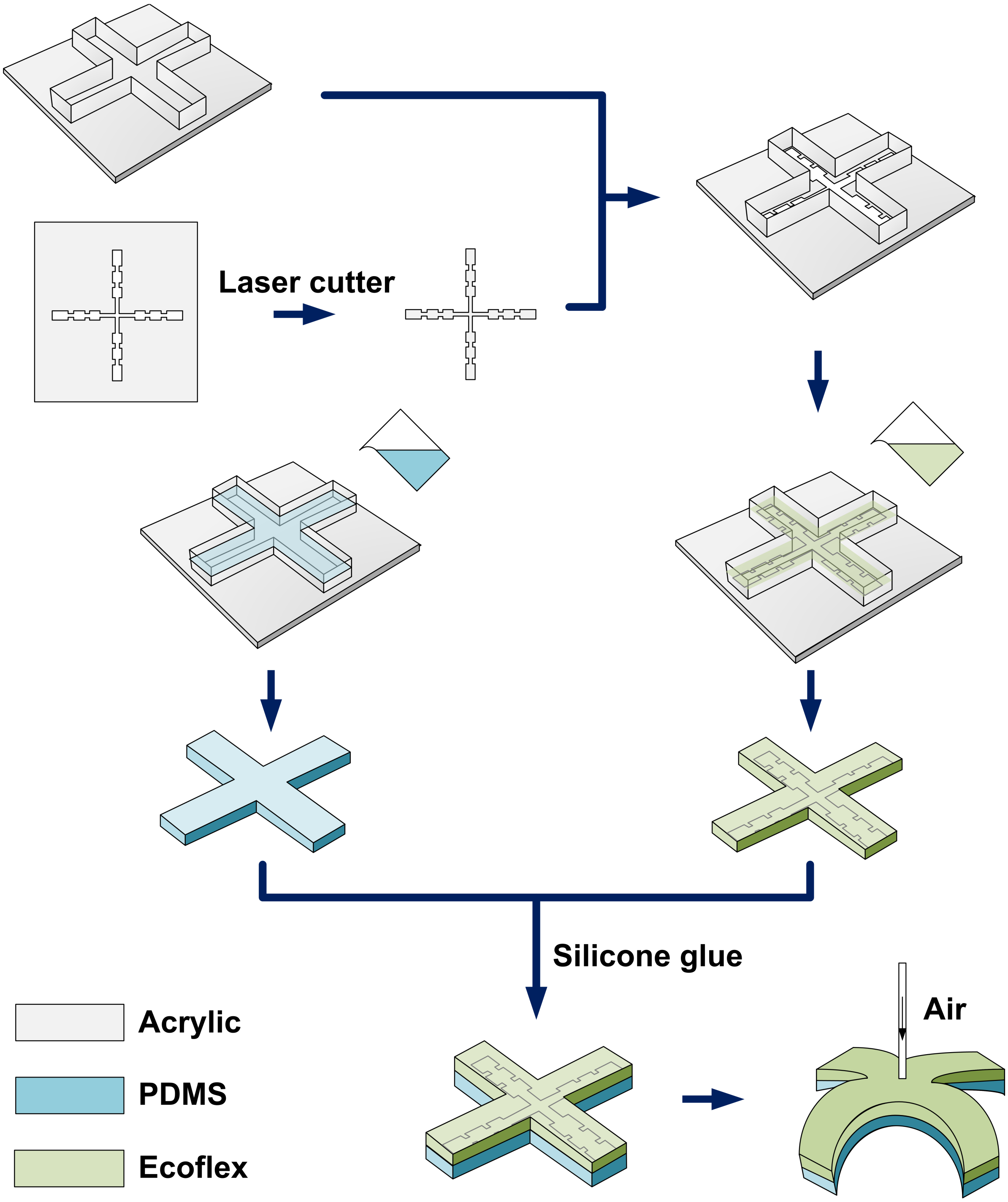

**Fig. S10.** The procedure to fabricate a pneumatic soft gripper.

## Supplementary Note 1 | Derivations of voltage-temperature relation.

The relation between the electric displacement $D$ and the electric charge density $\rho$ is $\frac{\mathrm{d}D}{\mathrm{d}x} = \rho$. The relation between the electric displacement $D$ and the electric field $E$ is $D = \varepsilon_\mathrm{e} E$, where $\varepsilon_\mathrm{e}$ is the permittivity of the electrolyte. The relation between the electric field $E$ and a voltage $V$ for an electric charge at position $x$ is $E = -\frac{\mathrm{d}V}{\mathrm{d}x}$. Combining the above three equations gives the Poisson's equation $-\frac{\mathrm{d}^2V}{\mathrm{d}x^2} = \frac{\rho}{\varepsilon_\mathrm{e}}$.

Let $\nu^+$ and $\nu^-$ be the valences of positive and negative ions. Let $n_0^+$ and $n_0^-$ be the numbers of positive ions and negative ions per unit volume in the electrolyte far away from any interfaces. Charge neutrality requires that $n_0^+\nu^+ + n_0^-\nu^- = 0$. Take the voltage far away from the interface to be zero, $V(\infty) = 0$. We can represent the charge density by $\rho = n^+\nu^+e - n^-\nu^-e$, where $n^+$ and $n^-$ are the numbers of positive and negative ions per unit volume, and $e$ is the elementary charge. The numbers of ions per unit volume are assumed to follow Boltzmann distribution, i.e., $n^+ = n_0^+\exp\left(-\frac{\nu^+eV}{kT}\right)$ and $n^- = n_0^-\exp\left(\frac{\nu^-eV}{kT}\right)$. Combining the Poisson's equation with the Boltzmann distribution, we can have

$$-\frac{\mathrm{d}^2V}{\mathrm{d}x^2} = \frac{n_0^+\nu^+e}{\varepsilon_\mathrm{e}}\left[\exp\left(-\frac{\nu^+eV}{kT}\right) - \exp\left(\frac{\nu^-eV}{kT}\right)\right]. \tag{S1}$$

When $\left|\frac{\nu^{+,-}eV}{kT}\right| \ll 1$, we can linearize the above differential equation:

$$\frac{\mathrm{d}^2V}{\mathrm{d}x^2} = \frac{V}{L^2}, \tag{S2}$$

where $L = \sqrt{\frac{\varepsilon_\mathrm{e}kT}{n_0^+\nu^+(\nu^+ + \nu^-)e^2}}$ is the Debye length. General solution is $V(x) = A\exp\left(\frac{x}{L}\right) + B\exp\left(-\frac{x}{L}\right)$, where $A$ and $B$ are the constants to be determined. $V(\infty) = 0$ results in $A = 0$. Then, $V(x) = B\exp\left(-\frac{x}{L}\right)$. The charge-voltage relation gives

$$V(0) = \frac{D(0)}{\varepsilon_{\mathrm{e}}} L. \tag{S3}$$

We assume that no space charge exists in the dielectric, so that the electric displacement in the dielectric is a constant, $D_{\mathrm{d}} = \text{constant}$. In the electrode, the density of mobile charge is high, so that the electric displacement vanishes, $D = 0$. Applying Gauss's law to a closed surface including the dielectric/electrolyte interface, we obtain that

$$D(0) - D_{\mathrm{d}} = \sigma_{\mathrm{i}}. \tag{S4}$$

Applying Gauss's law to a closed surface including the dielectric/electrode interface, we obtain that

$$\sigma_{\mathrm{e}} = D_{\mathrm{d}}. \tag{S5}$$

The voltage drop in the dielectric is

$$V(0) - V_{\mathrm{e}} = \frac{\sigma_{\mathrm{e}}}{\varepsilon_{\mathrm{d}}} d. \tag{S6}$$

Combining equations (S3)-(S6) gives

$$V_{\mathrm{e}} = \frac{\sigma_{\mathrm{e}} + \sigma_{\mathrm{i}}}{\varepsilon_{\mathrm{e}}} L - \frac{\sigma_{\mathrm{e}}}{\varepsilon_{\mathrm{d}}} d. \tag{S7}$$

## Supplementary Note 2 | Estimation of the minimum sensing area for ionotronic thermometry.

The sensing area $A_{\mathrm{e-i}}$ affects the output current by $I_{\mathrm{out}} = \frac{c_{\mathrm{e-i}} A_{\mathrm{e-i}} V}{t}$, where $c_{\mathrm{e-i}}$ is the capacitance of the e-i junction per unit area, $V = \frac{\mathrm{d}V}{\mathrm{d}T}(T - T_{\mathrm{ref}})$ is the voltage generated by the temperature change $T - T_{\mathrm{ref}}$, and $t$ is the thermal response time. The output current must be higher than the resolution of the measurement apparatus, $I_{\mathrm{resolution}}$, so that the minimum sensing area is $A_{\mathrm{e-i\,min}} = \frac{I_{\mathrm{resolution}} t}{c_{\mathrm{e-i}} V}$. In particular, $I_{\mathrm{resolution}} \sim 1$ fA (Keithley 6482 Picoammeter),

$t \sim 10$ ms, $c_{\mathrm{e-i}} \sim 0.1\ \mathrm{F/m^2}$, and $\frac{\mathrm{d}V}{\mathrm{d}T} \sim 1\ \mathrm{mV/°C}$. When $T - T_{\mathrm{ref}} \sim 1$ °C, the minimum sensing area is $A_{\mathrm{e-i\,min}} \sim 0.1\ \mathrm{\mu m^2}$.

**Supplementary Note 3 | Analysis of the i-e-i junction.**

For the design in Fig. 3*C*, the voltage can be expressed as

$$V = \frac{\sigma_{\mathrm{i1}} + \sigma_{\mathrm{e1}}}{\varepsilon_{\mathrm{e}}} \sqrt{\frac{\varepsilon_{\mathrm{e}} k (T - T_{\mathrm{ref}})}{2 c_1 N_{\mathrm{A}} v^2 e^2}} - \frac{\sigma_{\mathrm{i2}} + \sigma_{\mathrm{e2}}}{\varepsilon_{\mathrm{e}}} \sqrt{\frac{\varepsilon_{\mathrm{e}} k (T - T_{\mathrm{ref}})}{2 c_2 N_{\mathrm{A}} v^2 e^2}}, \tag{S8}$$

where $\sigma_{\mathrm{e1}}$ and $\sigma_{\mathrm{e2}}$ are the accumulated electronic charges per unit area at the two e-i junctions, $\sigma_{\mathrm{i1}}$ and $\sigma_{\mathrm{i2}}$ are the accumulated ionic charges per unit area at the two e-i junctions, and $c_1$ and $c_2$ are the concentrations of ions in the two ionic conductors. When $c_2 \gg c_1$, the second term in the above expression is negligible and equation (S8) can be simplified as $V = \frac{\sigma_{\mathrm{i1}} + \sigma_{\mathrm{e1}}}{\varepsilon_{\mathrm{e}}} \sqrt{\frac{\varepsilon_{\mathrm{e}} k (T - T_{\mathrm{ref}})}{2 c_1 N_{\mathrm{A}} v^2 e^2}}$. As a result, the design in Fig. 3*C* and the design in Fig. 2*A* have similar sensitivities, which is consistent with the experimental measurement (Fig. 2*B* and Fig. S5*A*).

**Supplementary Note 4 | Analysis of the e-i-e junction.**

For the design in Fig. 3*E*, the voltage *V* can be expressed as

$$V = \frac{(\sigma_{\mathrm{i1}} + \sigma_{\mathrm{e1}}) \pm (\sigma_{\mathrm{i2}} + \sigma_{\mathrm{e2}})}{\varepsilon_{\mathrm{e}}} \sqrt{\frac{\varepsilon_{\mathrm{e}} k (T - T_0)}{2 c N_{\mathrm{A}} v^2 e^2}}. \tag{S9}$$

Here $T_0$ is a pre-determined temperature for reference and is set to be room temperature in the experiment for convenience. As indicated by equation (S9), changes in the voltages of the two e-i junctions caused by temperature changes can be either additive or subtractive, depending on the type of the two electronic conductors. Dissimilar electronic conductors can anchor ionic charges of either same or opposite signs.

When aluminium is used as one of the electronic conductors, the sensitivity is increased to as high as ~$10\ \mathrm{mV/°C}$ (Fig. S6*B*). This may be because the hydroxyl groups on the native aluminium oxide layer result in increase in the ionic charges on the hydrogel/dielectric interface, $\sigma_{\mathrm{i}}$.

**Supplementary Note 5 | Electrical response of the ionic cable.**

Resistance of hydrogel can be calculated by $R_{\mathrm{hydrogel}} = \frac{\rho l}{S} \sim \frac{1 \cdot 10^{-1}}{10^{-5}} = 10^4\ \Omega$, where $\rho$ is the resistivity, $l$ is the length, and $S$ is the cross-sectional area. Capacitance of VHB can be calculated by $C_{\mathrm{VHB}} = \frac{\varepsilon_{\mathrm{VHB}} A_{\mathrm{VHB}}}{h_{\mathrm{VHB}}} \sim \frac{10^{-11} \cdot 10^{-4}}{10^{-3}} = 10^{-12}$ F, where $\varepsilon_{\mathrm{VHB}}$ is the permittivity, $A_{\mathrm{VHB}}$ is the surface area, and $h_{\mathrm{VHB}}$ is the thickness. As a result, the electrical response time of the ionic cable is given by $t_{\mathrm{RC}} = R_{\mathrm{hydrogel}} C_{\mathrm{VHB}} \sim 10^{-8}$ s, which is much shorter than the thermal response time.

## Description of Other Supplementary Materials

File Name: Movie S1

Description: Ionotronic thermometer integrated to a soft gripper for temperature monitoring for curved surfaces.