Supplementary Information

Humidity-resistive, elastic, transparent ion gel and its wearable strain-sensing device

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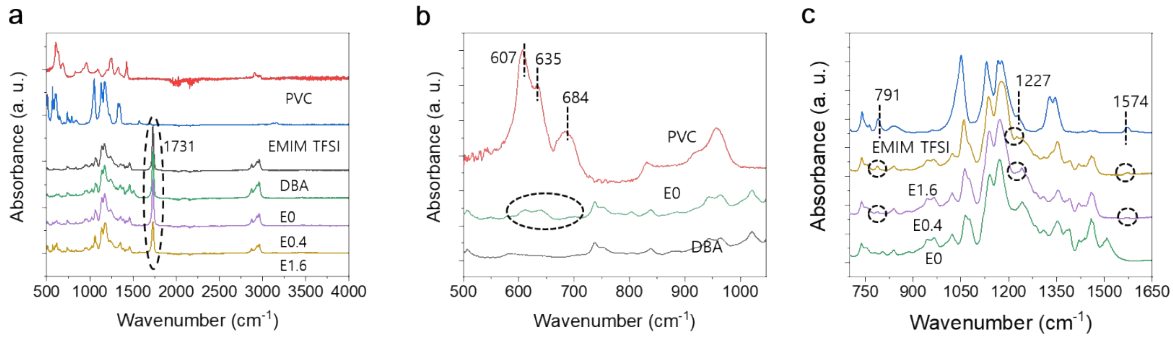

Fig. S1 FT-IR analysis of ion gels (a) The FT-IR results of PVC, DBA, and [EMIM$^+$][TFSI$^-$], which are the component of the gel, were obtained and compared with the results of ion gel (E0, E0.4, E1.6). Ester stretching bands (1731 cm^{-1}) for the DBA were observed at E0, E0.4 and E1.6 gels. This peak can be only observed in the DBA among three substances (PVC, DBA, and [EMIM+][TFSI-]) which are constituents of the ion gel. Therefore we could confirm DBA in an ion gel. The results of E0, E0.4 and E1.6 are almost similar to that of the DBA, because the DBA has the largest portion in an ion gel and the weight ratio of PVC and [EMIM$^+$][TFSI$^-$] in the gel was much lower than that of the DBA. (b) The C-Cl band peak (607, 635 and 684 cm^{-1}) can be observed at E0, so the existence of PVC in the gel is confirmed. However, it is hard to identify these peaks for E0.4 and E1.6, because the peaks of [EMIM+][TFSI-] are overlapped in the range of 600–700 cm^{-1}. (c) The peaks of C-S stretching (791 cm^{-1}), –CF_3 symmetric stretching (1227 cm^{-1}), and CH_2N/CH_3N symmetric and asymmetric stretching (1574 cm^{-1}) peaks, which can be seen only in [EMIM$^+$][TFSI$^-$], were the largest at E1.6. For E0.4, the peak size was reduced and the peak was not observed for E0. The corresponding results indicate that the DBA plasticizer and the [EMIM$^+$][TFSI$^-$] additive were physically distributed in the PVC network without forming specific intermolecular interactions with the PVC chains. As a result, the [EMIM$^+$][TFSI$^-$] ions in the gel are moving without forming chemical bonds with PVC or DBA.[1]

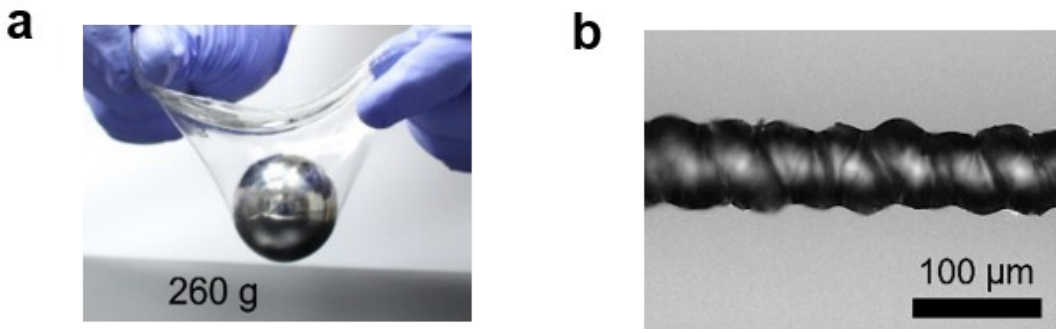

Fig. S2 (a) The 260-g steel ball is put on an ion gel film. (b) The gel is twisted to form a micro-sized yarn.

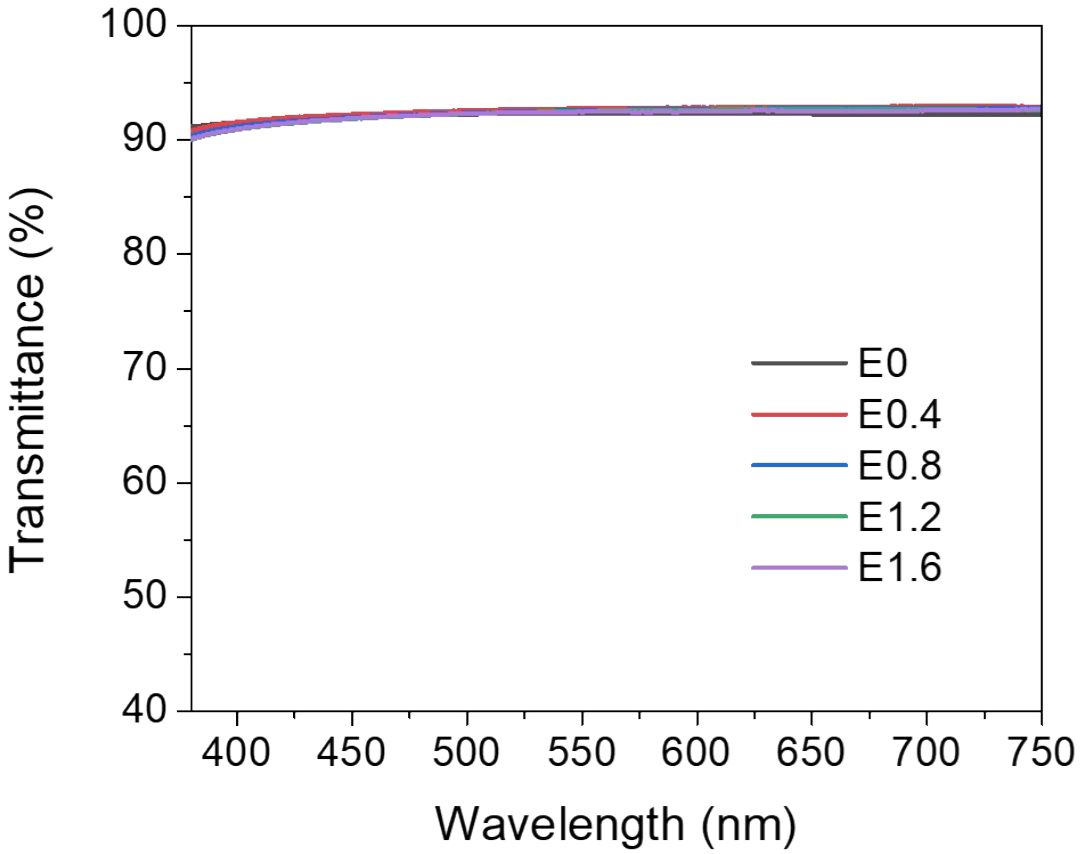

Fig. S3 Ultraviolet spectroscopy analysis of ion gels**.** The transmittance of an ion gel was measured by ultraviolet spectroscopy. The results show excellent transparency of an ion gel. For all ion gels, a transmittance between 90 ~ 93% in the visible light range of 380 to 750 nm was measured.

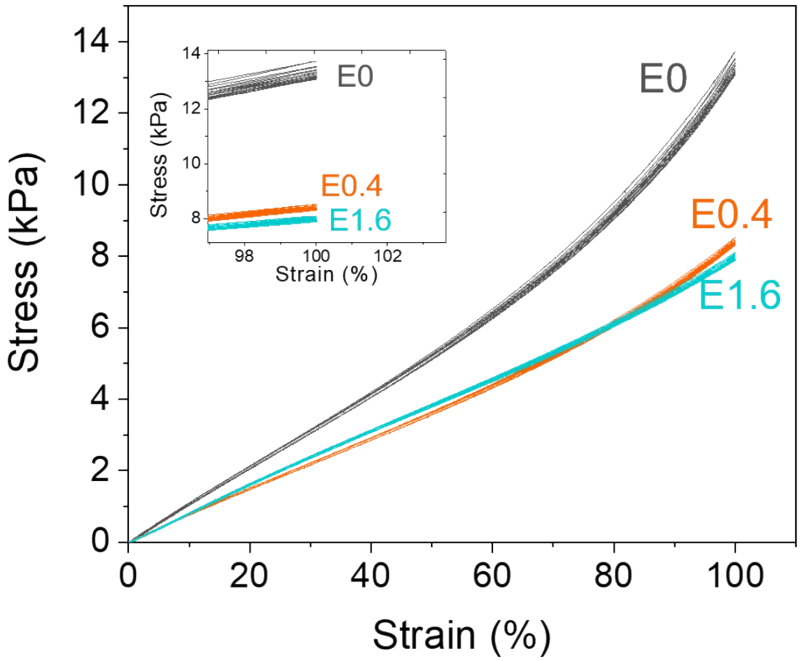

Fig. S4 Mechanical hysteresis of ion gels. Stress-strain curve is hardly changed when repeated tests on the E0 sample without ionic liquid. In the first cycle, the strain at 100% strain was 13.72 kPa, which decreased by 4.74% to 13.07 kPa in the ninth cycle. The E 0.4 and E1.6 curves show nearly the same stress-strain curves and are below the E0 stress-strain curves and show little mechanical hysteresis as in E0. E0.4 stress decreased 2.48% from 8.51 kPa to 8.30 kPa at 100% strain and 2.83% from 8.1 kPa to 7.87 kPa for E1.6. These results show that the addition of ionic liquid slightly reduces the stiffness but still maintains the mechanical stability under repeated loads.

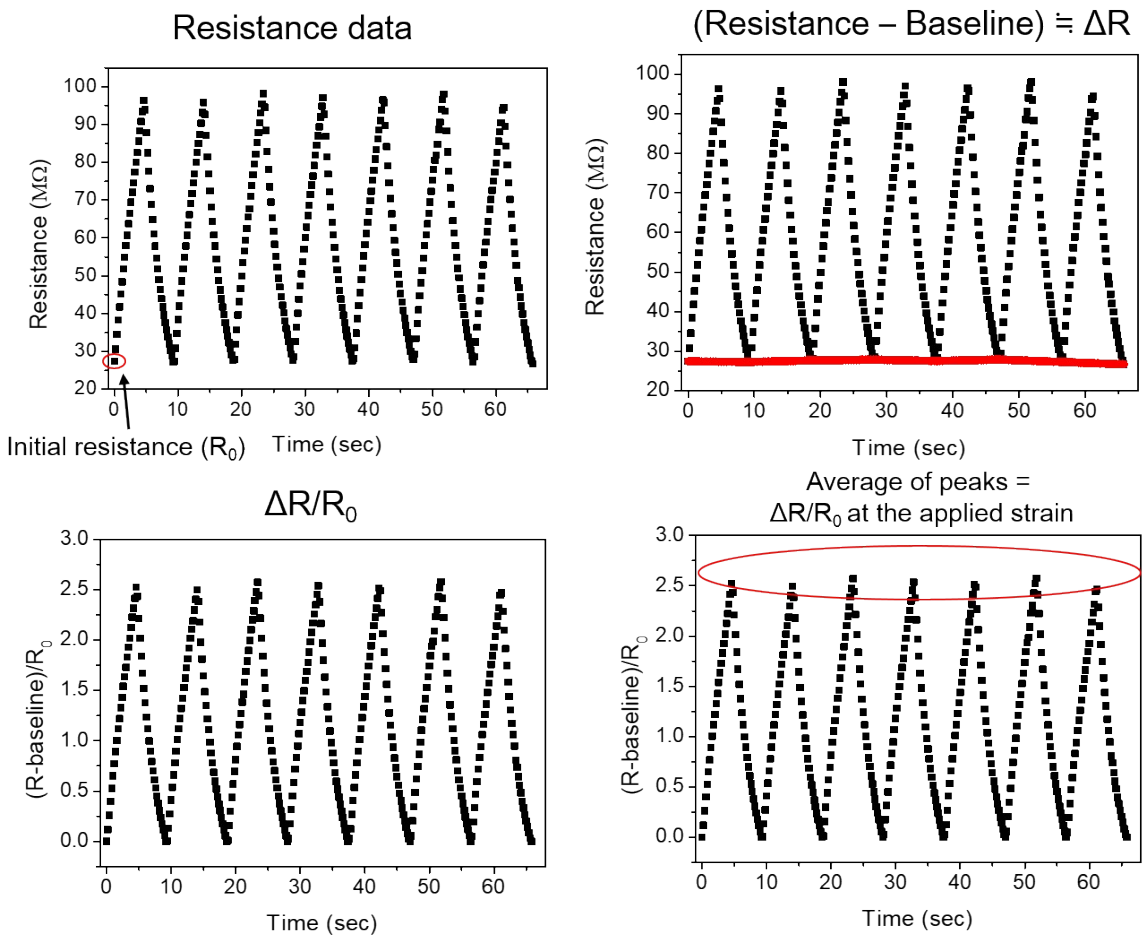

Fig. S5 Calculating method of $\Delta R/R_0$ for the gel with the applied strain.

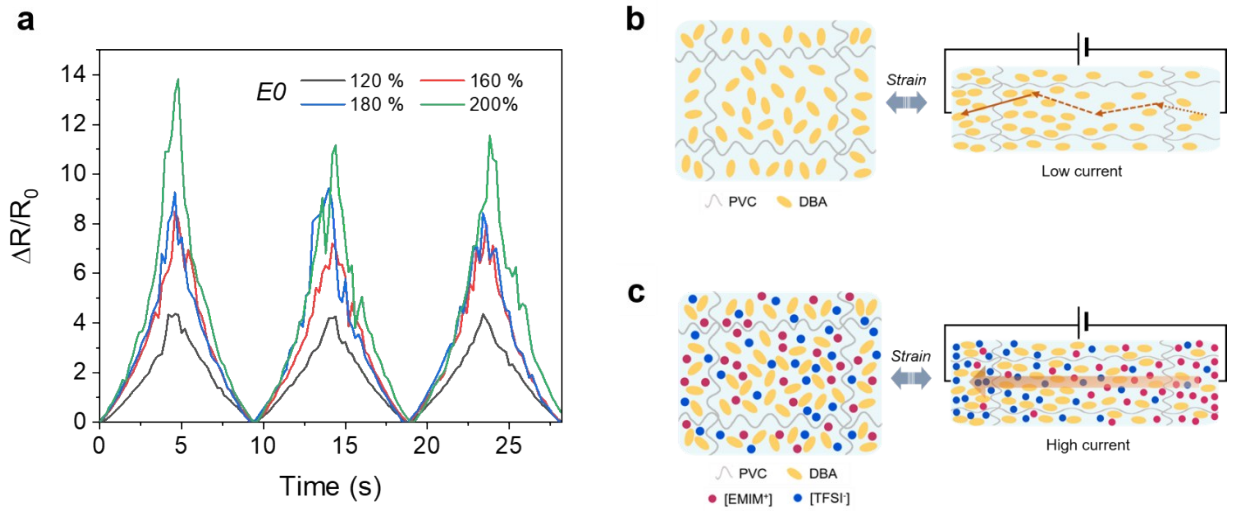

Fig. S6 The resistance change of E0 and schematics of operation mechanism of strain sensing. (a) the resistance of E0 measured while applying a constant voltage of 0.5V with 120~200% strain. (b) When the voltage (less than 10V) applied to PVC/DBA gel without ion liquid (E0), DBA would migrate toward the anode, but the amount of migration would be very small due to relatively low voltage. At the same time, little current flows through the gel because PVC and DBA are dielectric materials. When the gel is stretched, the current flow through the gel becomes unstable due to the increased resistance (c) If the same voltage applied to PVC/DBA gel with ion liquid and apply strain to the gel, only a little amount of DBA migrate toward the anode (less than the gel without the ion liquid).[2] The positive ions ($[EMIM^+]$) are attracted to the cathode and the negative ions ($[TFSI^-]$) are attracted to the anode. There are still some ions in the middle of the gel.[3] These ions help the charge transport through the gel when it is stretched. Therefore, the addition of $[EMIM^+][TFSI^-]$ to PVC/DBA makes the gel a stable strain sensor.

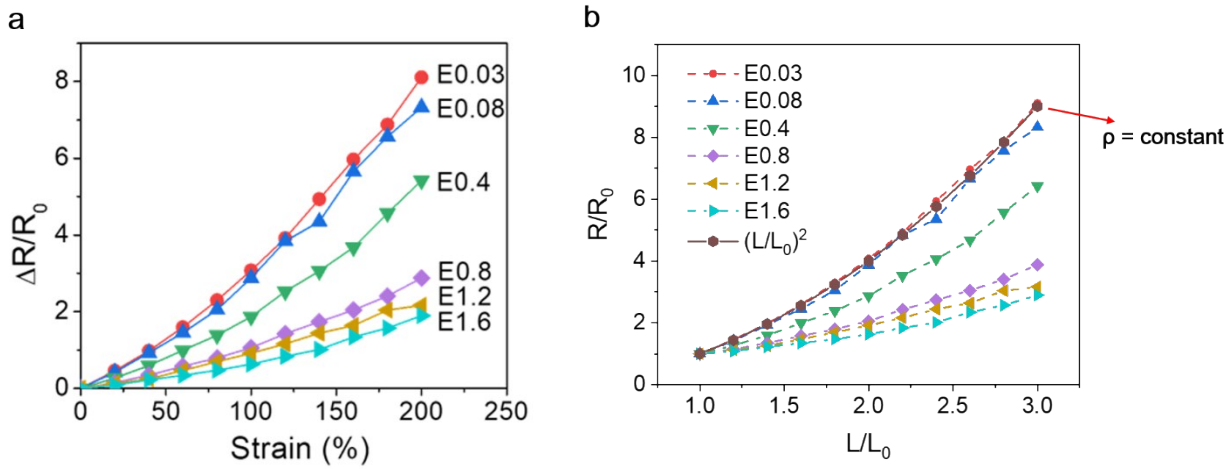

Fig. S7 Comparison strain vs. sensitivity with experimental and theory. (a) Strain vs. $\Delta R/R_0$ graph of ion gels with various ion liquid content. (b) L/L_0 vs R/R_0 graph of ion gels with various ion liquid content. If the resistivity (ρ) is constant regardless of the strain, the resistance changes only depending on the shape of the gel. The resistance change rate R/R_0 is equal to $(L/L_0)^2$. Samples in this range are E0.03 and E0.08. From E0.4, R/R_0 is below the $(L/L_0)^2$ curve. This means $R/R_0 < (L/L_0)^2$. Let's define the initial resistance (R_0) as $R_0 = \rho_0 L_0/A_0$, and the resistance of an ion gel after stretch (R) as $R = \rho L /A$. If the length of an ion gel increases from L_0 to L, the area of gel would approximately decrease from A_0 to $A_0/(L/L_0)$ because the volume of an ion gel is constant. Then the resistance of an ion gel after stretch is $R = \rho L /A = \rho_0(L_0/A_0)\times(L/L_0)^2$, so from $R/R_0 < (L/L_0)^2$, it must be $\rho < \rho_0$.

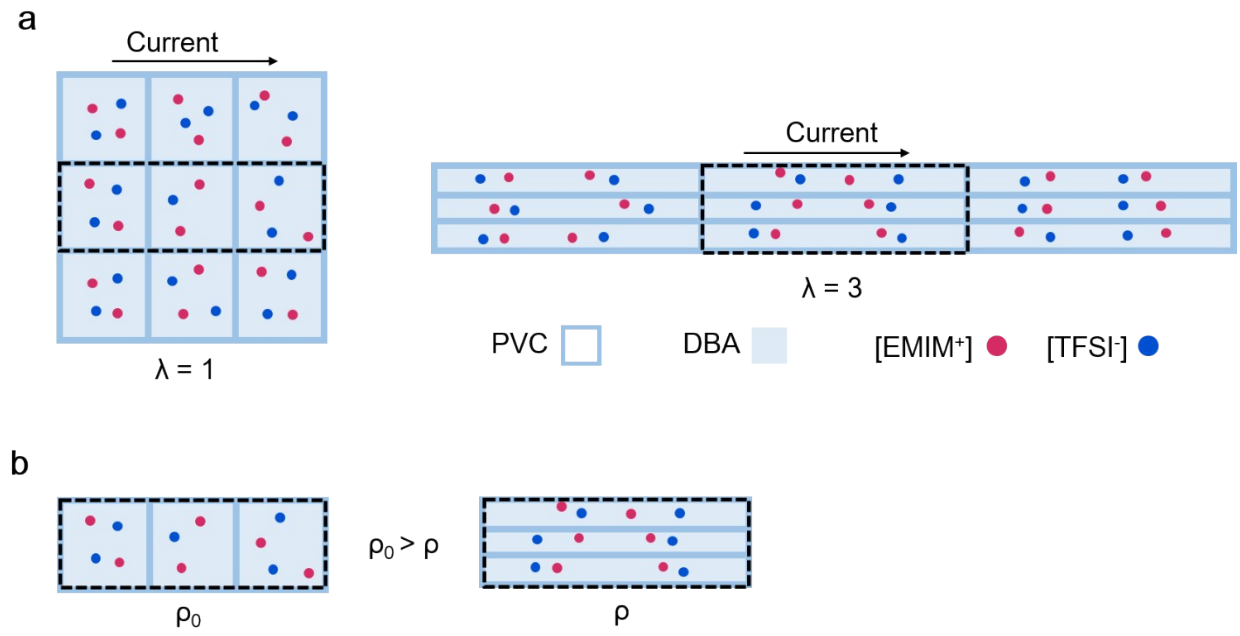

Fig. S8 Resistivity reduction mechanism of an ion gel. (a) When an ion gel is stretched, the resistivity of an ion gel decreases gradually, and this tendency increases with gels with higher ion content. If there are some interactions between PVC chain and ions which would impede the charge transfer, the decrease of resistivity could be explained. We guess these interaction is the ion-dipole interaction of C-Cl bond in the PVC chain and ions of [$EMIM^+$][$TFSI^-$].[4,5] As the length of an ion gel is tripled, the density of the PVC chain in the current path direction is reduced by one third. The ion-dipole interaction decreases in the direction of current due to reduced density of PVC chain in the current direction. (b) This leads to the decrease of the resistivity of an ion gel. The decreased resistivity compensates the increase of resistance of the stretched ion gel. For an ion gel with higher ion content, the decrease of resistivity is large, due to the ion-dipole interaction.

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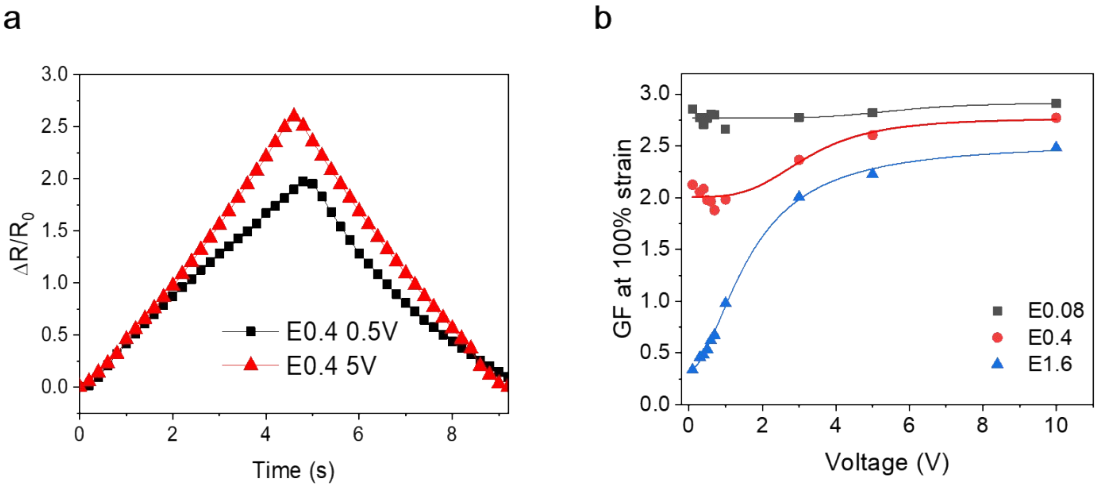

Fig. S9 (a) Resistance change of E0.4 gel with applied voltage 0.5 and 5V. (b) Applied voltage vs GF at 100% strain of ion gels (E0.08, E0.4 and E1.6)

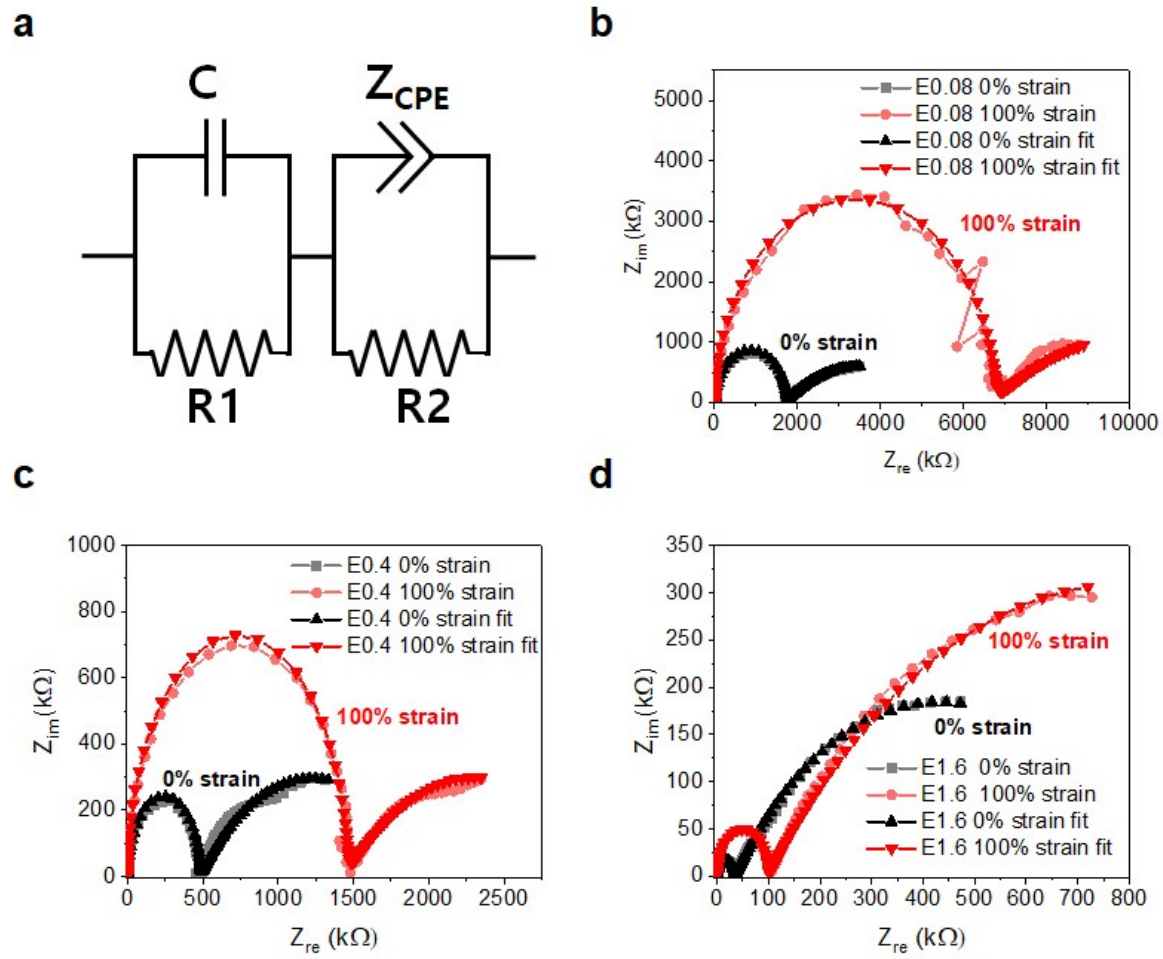

Fig. S10 (a) Equivalent circuit of the ion gel. (b), (c), and (d) Experimental results of impedance spectroscopy and fit results from the equivalent circuit for the initial state and stretched state of the gels (E0.08, E0.4, and E1.6)

	R1 (Ω)	C (F)	R2 (Ω)	Q (F)	n
E0.08 0% strain	1.7e+6	8.5e-11	3.7e+6	1.55e-6	0.4
E0.08 100% strain	6.7e+6	8.5e-11	7.2e+6	1.48e-6.	0.38
E0.4 0% strain	4.8e+5	8.5e-11	1.5e+6	4e-6	0.48
E0.4 100% strain	1.45e+6	8.5e-11	1.7e+6	3.3e-6	0.43
E1.6 0% strain	3.4e+4	8.5e-11	8.0e+5	1.1e-5	0.55
E1.6 100% strain	1.0e+5	8.5e-11	1.4e+6	8.2e-6	0.53

Table S1 Properties of elements in the equivalent circuit (Fig. S10a) for curve fitting.

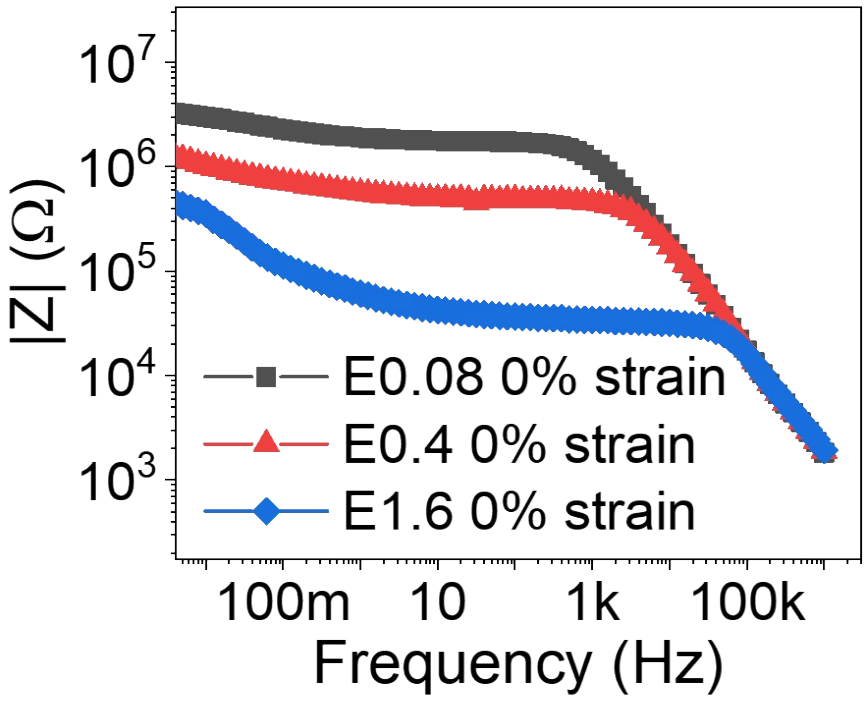

Fig. S11 Bode plot of impedance spectroscopy for E0.08, E0.4, and E1.6.

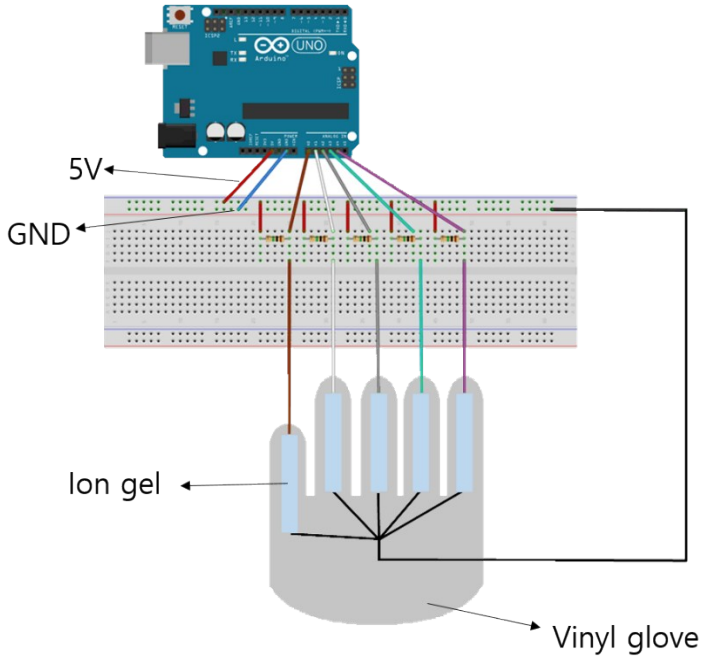

Fig. S12 The schematic diagram of the connection between the Arduino and the gel attached to a vinyl glove.

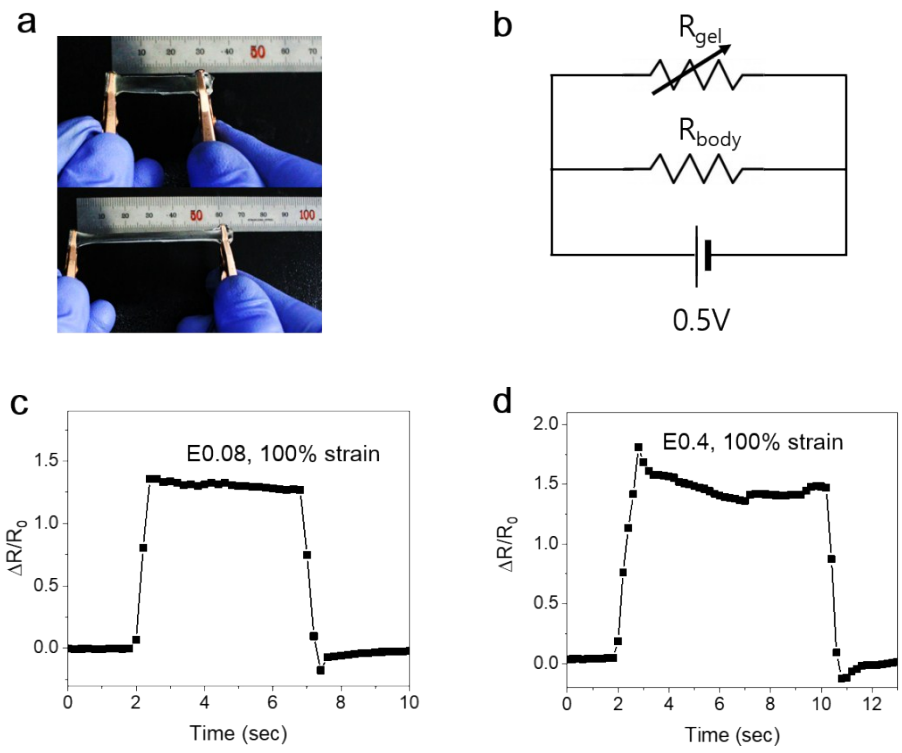

Fig. S13 Resistance change of PVC gels when strain is applied and electrodes are held by hands. (a) Photo of the experiment. Holding both electrodes at the end of an ion gel with hands wearing the experimental gloves, the ion gel was stretched to a 100% strain. In this state, the current can flow through the body, thus it can be described as a parallel connection with the ion gel and the body. An equivalent circuit diagram of the experiment is shown in (b). (c) The $\Delta R/R_0$ of E0.08 at a 100% strain is 1.5 or less, which is lower than the those (2.5-3) measured with the strain tester. The resistance of the ion gel is relatively large, and current also flows by hand, which reduces the resistance change due to the parallel connection.. d) In the case of E0.4 (higher ion concentration), there was a little difference in the ΔR/R0 between this experiment (1.5-1.7) and the experiment of the strain tester (1.5-2.0)

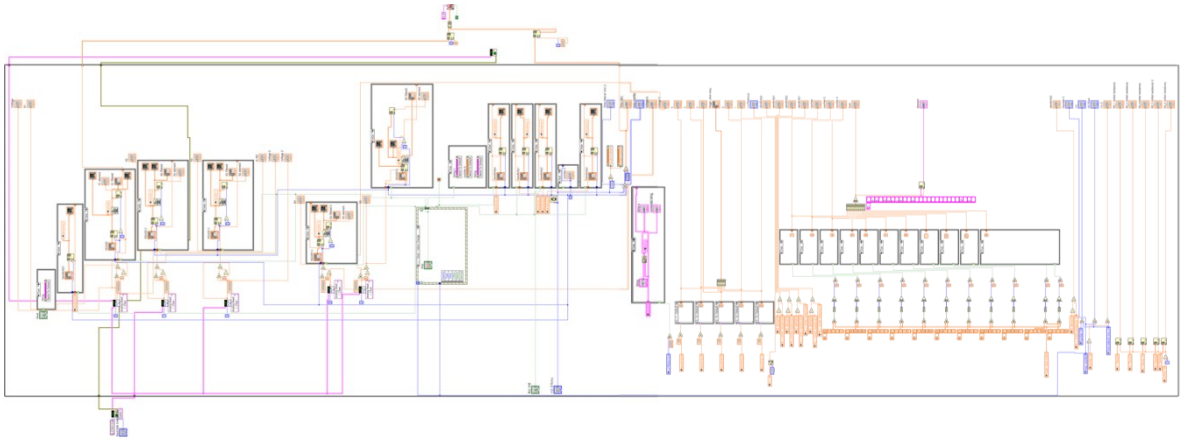

Fig. S14 LabVIEW block diagram for conducting sign language.

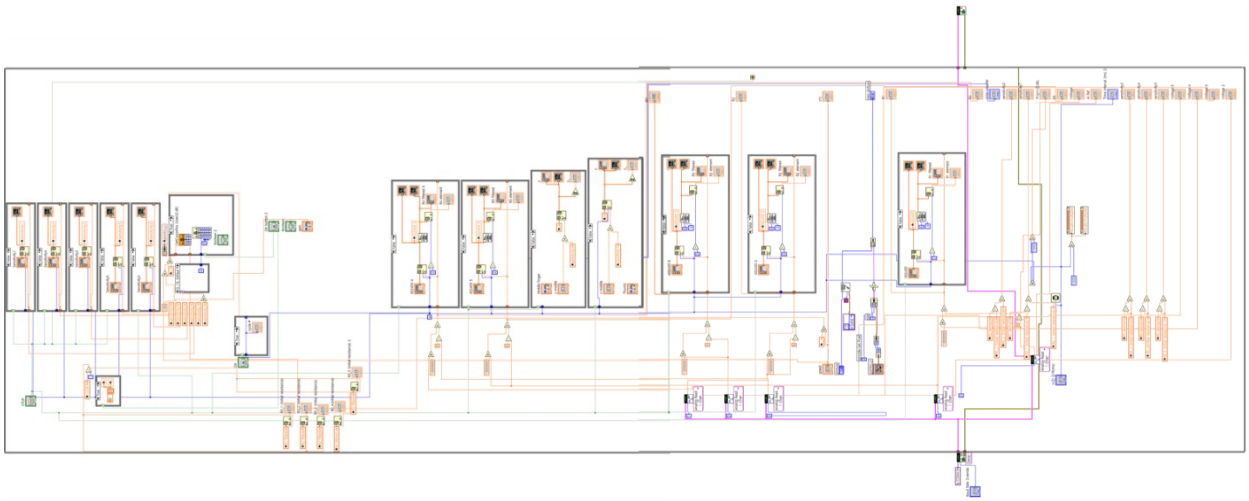

Fig. S15 LabVIEW block diagram for conducting mouse control.

The resistance change mechanism of an ion gel when it is stretched (Fig. S6b, Fig. S8)

To deeply investigate the relationship between the resistance change and the ion content of the stretched ion gel, we plotted a L/L_0 vs. R/R_0 graph (Fig. S6b). L_0 is initial length of an ion gel, and L is the length of an ion gel after stretching. Conventionally the resistance of a material (R) is defined as

$$R = \rho\frac{L}{A} \qquad (1)$$

(ρ is the resistivity, L it the length, and A referes to the area of cross section)

If the material stretched from L_0 to L, then cross section area A_0 decreases to $(L_0/L)\times A_0$ by assuming that the volume of materials does not change.[6] Then the resistance of the stretched material changes to

$$R = \frac{\rho(L)}{(\frac{L_0}{L}A_0)} = \rho\frac{L}{A} \times (\frac{L}{L_0})^2 = R_0(\frac{L}{L_0})^2$$

Therefore R/R_0 is equal to $(L/L_0)^2$. This happens when the resistivity is constant regardless of the strain. The ion gels which coincide with this equation are E0.03 and E0.08 (Fig. S5b). For ion gels with higher ion content (E0.4, E0.8, E1.2 and E1.6), R/R_0 have lower value, which is below $(L_0/L)^2$. If we define ρ_0 as the resistivity before stretching and ρ as the resistivity after stretching, then $\rho_0 > \rho$ by equations (1) and (2). This means that the resistivity (ρ) decreases when an ion gel is stretched. A decrease of the resistivity suppresses the resistance change of the stretched gel. Also, the gel with a higher ion content shows a lower resistance change. Based on this analysis, we assume the mechanism of the resistance change of an ion gel when the strain is applied. The PVC chain has C-Cl bonds and these bonds have a dipole moment, and there may be an ion-dipole interaction between the PVC and $[EMIM^+][TFSI^-]$ ion liquid.[4,5] We presume that this interaction hinders charge transfer through an ion gel.[7] If an ion gel is stretched as thrice its length, the networks of the PVC chain also elongates thrice for the

stretched direction (Fig. S8a). This causes a decrease of the PVC chain density in the stretched direction (also current direction), and it becomes easier for the charges to flow through the stretched gel in this direction. This also results in a decrease in the resistivity of an ion gel when it is stretched (Fig. S8b), and a decrease in the resistivity causes the the resistance change to reduce conrrespondingly. As the ion content in the gel increases, the ion-dipole interaction also increases, so there are more reductions in the resistance change.

The calculation method for mouse controlling

The click motion is determined by the differentiation of $\Delta R/R_0$ (t). When this value exceeds 0.2, a click motion of the mouse can be executed. $d(\Delta R/R_0(t))/dt$ means the folding speed of a finger, because $\Delta R/R_0$ (t) is related to the folding degree of a finger. To move the mouse pointer horizontally, the thumb should be folded or extended to stretch or contract the ion gel on the thumb. The program recognizes the integration of $(0.25 - \Delta R/R_0(t))$ by real time and the horizontal displacement of the pointer is determined by $\int(0.25 - \Delta R/R_0(t))dt$. Therefore $0.25 - \Delta R/R_0$ (t) determines the speed of the pointer. Here, 0.25 refers to the middle value between 0 to 0.5 (0.5 is a maximum value of $\Delta R/R_0$ (t)). When $\Delta R/R_0$ (t) exceeds 0.25, $0.25 - \Delta R/R_0$ (t) becomes a negative value, and the pointer goes left, while the pointer moves right when $\Delta R/R_0$ (t) drops below 0.25, $0.25 - \Delta R/R_0$ (t) to become a positive value. When $\Delta R/R_0$ (t) is equal to 0.25, $0.25 - \Delta R/R_0$ (t) becomes zero, and the pointer stops. Likewise we can control the vertical movement of the pointer. By composing $\int(0.25 - \Delta R/R_0(t))dt$ of the ion gel on the thumb and the middle finger as x and y displacement, we can draw the trace of the pointer.

Supplementary Videos

1. Video S1 : Stretching and contraction of the ion gel in water
2. Video S2 : Signaling and monitoring sign language using an ion glove
3. Video S3 : Controlling mouse pointer using an ion glove

Supplementary Information References

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