Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting information

Combining the Converse Humidity/Resistance Response behavior of RGO films for Flexible Logic Devices

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This PDF file includes:

1. Experimental videos.

   Video-1# Real-time negative resistance response; **Video-2#** Real-time positive resistance response; **Video-3#** Real-time humidity insensitive performance; of rGO film to human-index finger/human breath.

   Video-4# Application to gesture recognition.

   Video-5# Application to 3-dimensional-noncontact sensing technique.

2. Further characterization of experimental samples, electrical impedance spectroscopy (EIS), and test instrument/method from **S1**, and **Figure S1** to **S7**.

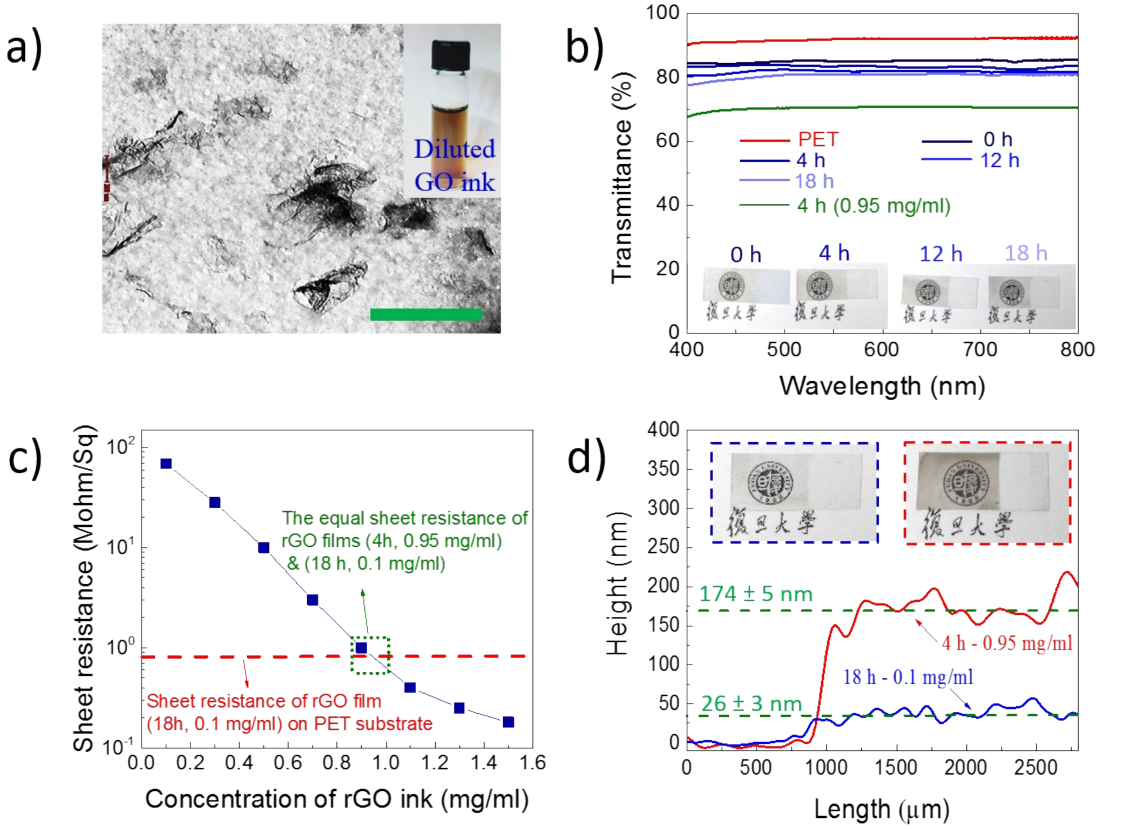
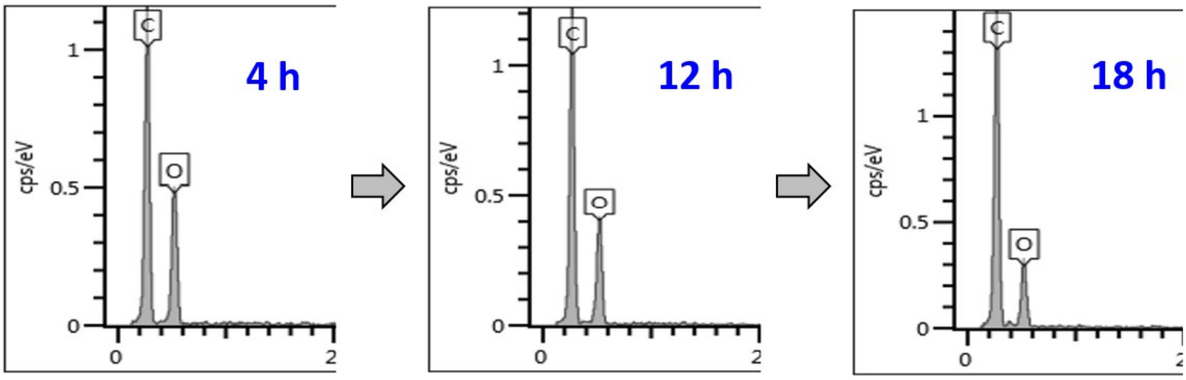


Figure S1. Characterization of rGO ink and rGO based films. a) TEM image of rGO nanoplates; Inset is the typical rGO ink with the concentration of 0.1 mg/ml. b) Optical transmittance of the prepared rGO-based transparent conductive films on a PET substrate with the thermal-reduction time of 0 h, 4 h, 12 h, 18 h, respectively; Note that pure PET was used as a reference, and rGO (4h, 0.95 mg/ml) was also compared; Insets are the typical rGO films with different thermal-reduction times. c) Relationship between sheet resistance and concentration of rGO ink to balance the relative resistance for the fabrication of the following flexible logic devices. Note the balance point is that the resistance of rGO film (4 h, 0.95 mg/ml) is equal to that of (18 h, 0.1 mg/ml), and d) demonstrate the typical profiles of both; Inset in d) is the corresponding digital image of both rGO films. Note that each sample was tested three times and a typical result is presented.



Element	Wt%
CK	61.22
OK	38.78

Element	Wt%
CK	69.34
OK	30.66

Element	Wt%
CK	78.96
OK	21.04

Figure S2 EDS analysis (element ratio of carbon and oxygen) of rGO films with different thermal-reduction times, 4 h, 12 h, 18 h, respectively, at 150 °C.

S1. EIS Data Analysis and Equivalent Circuit Modeling

The data obtained from the electrical impedance spectroscopy (EIS) represents the complex impedance (Z) along with its phase angle (θ) measured at different frequencies. The variation of Z and θ or these impedance parameters such as real and imaginary part of the impedance are plotted over frequency to analyze the sample under test. The Nyquist curve which is the plot of the imaginary part (X) of the complex impedance over its real part (R) represents the key information about the equivalent circuit of the material. The martials with capacitive or inductive components parallel to some resistance produces a Nyquist plot which is a part of a semicircular arc representing the values of the circuit components within it. In this direction Nyquist curve of a particular sample are essentially required to be analyzed to model its equivalent circuit for extracting the information of the material composition or structure in terms of its equivalent circuit components.

Curve fitting techniques can be utilized to fit the Nyquist plot with a circular arc and deduce the equivalent circuit model. In the present study a Matlab based curve fitting algorithm has been developed to analyze the Nyquist plot obtained from the EIS data collected from the sample tested using the impedance analyzer Agilent 4980A. The Algorithm loads the impedance data matrix formed with real and imaginary part of the sample impedance data collected at 20000 different frequency points within a frequency band of 20 Hz to 2 MHz.

If a material contains the composition which equivalently behaves like a circuit model containing a series resistance (r) along with the parallel branch ($R_p||C_p$) as shown in Figure S1-1a, the Nyquist plot exerted by the material will be a semi-circular arc which cuts the real axis at a distance of r. The semi-circular impedance spectra will be representing the parallel combination of R_p and C_p. When the material contains a constant phase element (CPE), the Nyquist plots are found as the semi-circular arcs with their centers some distance below the x-axis as shown in Figure S1-1b. As shown in the Figure S1-1b, the Nyquist plot presents a depressed semi-circular arc which indicates an equivalent circuit containing the CPE in parallel with R (R||CPE) and a series resistance (r) is connected with that parallel branch (R||CPE).

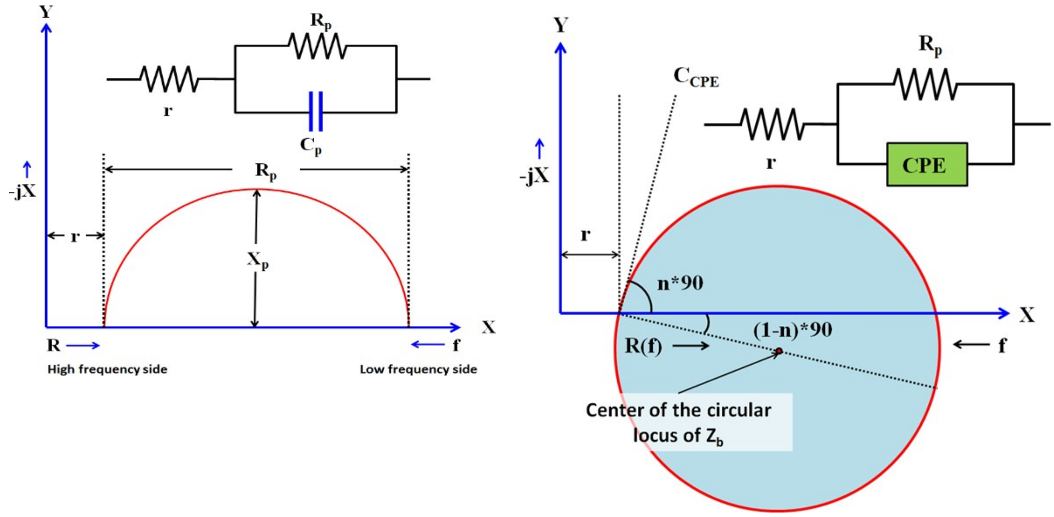


Figure S1-1: EIS data analysis and the equivalent circuit modelling (a) Nyquist plots for a circuit combination containing two sub-circuit blocks with a series resistance of r and the parallel combination of R_p and a capacitor C_p ($R_p||C_p$), (b) Nyquist plots of circuit combinations containing two sub-circuit blocks: one with series resistance, R_s, and another containing a CPE in parallel with resistance R_p (R_p || CPE).

The impedance of the CPE (Z_{CPE}) shown in Figure S1-1b is represented as:

$$Z_{CPE} = Q\left(\frac{1}{j.\omega}\right)^{n} = \frac{1}{Y_0}(j.2\pi f)^{n},$$

where Q is the magnitude of Z_{CPE} and Y_0=1/Q, j=$\sqrt{-1}$ and 0<n<1.
As the phase angle (ϕ) of Z_{CPE} is frequency independent, the phase remains constant throughout the frequency range of EIS and hence it is called the constant phase element.

The phase angle (ϕ) of Z_{CPE} is given by:

$$\phi = -(90 * n)$$

For an material containing a CPE within it, if n is less than 1, the CPE resembles a real capacitor producing a phase angle ϕ less than 90°. As the value of n approaches toward 1, CPE gradually becomes more capacitive and Y_0 becomes a pure capacitor (C_{CPE}) at n = 1.

If a material contains CPE and a resistive element, R_p, parallel to each other, the R-X plot (Nyquist plot) of the material will be depressed by an angle of (1-n)*90° (Figure S1-1b).

For example, for a material having an equivalent circuit of a series parallel combination as shown below (Figure S1-2), the Matlab base algorithm calculates the f_{Max}, X_{Max}, r, R_p, X_p, C_p, n and Z_p.

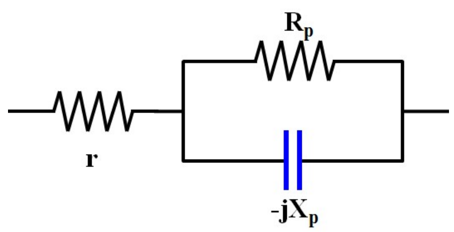


Figure S1-2: An equivalent circuit model containing two sub-circuit blocks with a series resistance of r and the parallel combination of R_p and a capacitor C_p ($R_p||C_p$)

For the circle parameters calculated by the algorithm the f_{Max}, X_{Max}, r, R_p, X_p and n are found. X_p is the capacitive reactance produced by the capacitor C_p as given below:

$$X_p = \frac{1}{2\pi f C_p} \qquad \text{(S1)}$$

The impedance of the parallel branch ($Z_p = R_p||C_p$ or $R_p||(-jX_p)$) is represented and calculated as:

$$Z_p = R_p \,||\, (-jX_p)$$

$$|Z_p| = \sqrt{\left(\frac{R_p (X_p)^2}{(R_p)^2 + (X_p)^2}\right)^2 + \left(\frac{X_p (R_p)^2}{(R_p)^2 + (X_p)^2}\right)^2} \qquad \text{(S2)}$$

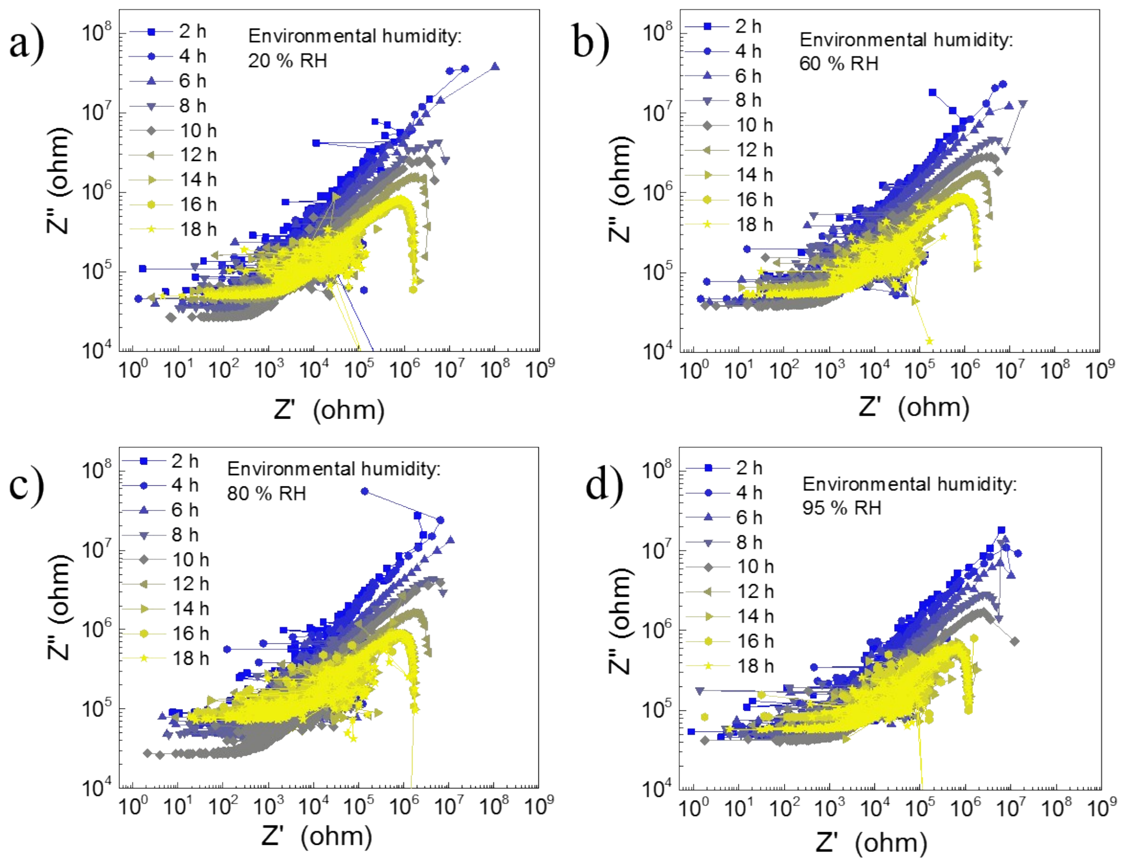


Figure S3. Electrical impedance spectroscopies of rGO films with different thermal-reduction times under different environmental humilities, a) 20 % RH; b) 60 % RH; c) 80 % RH; d）95 % RH; respectively.

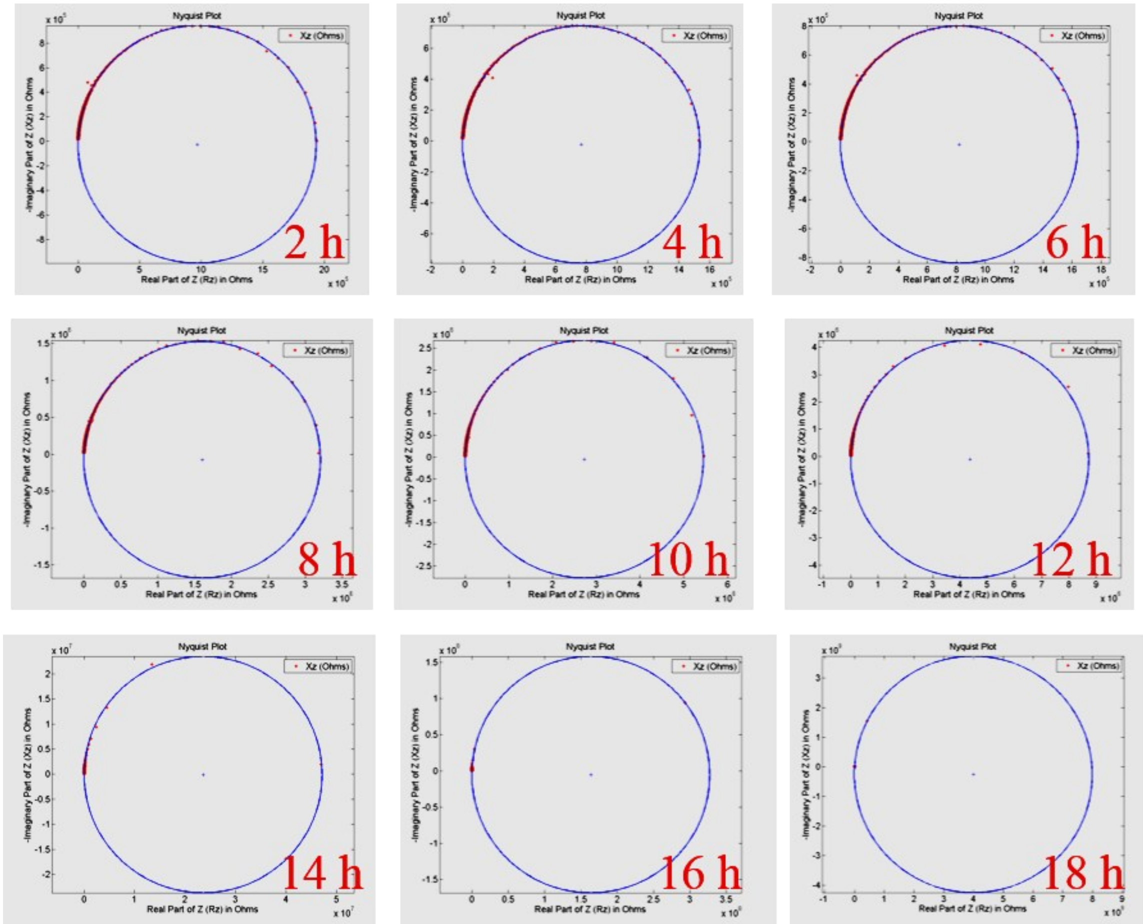


Figure S4. The fitted electrical impedance spectroscopies of rGO films with different thermal-reduction times at room humidity (41.3 RH %) from 2 h to 18 h, respectively. This image presents the typical fitted results via a MATLAB-based impedance curve fitting program, indicating the efficiency of this strategy. Note that the red-dot line is the test results, and the blue circle is the fitted results.

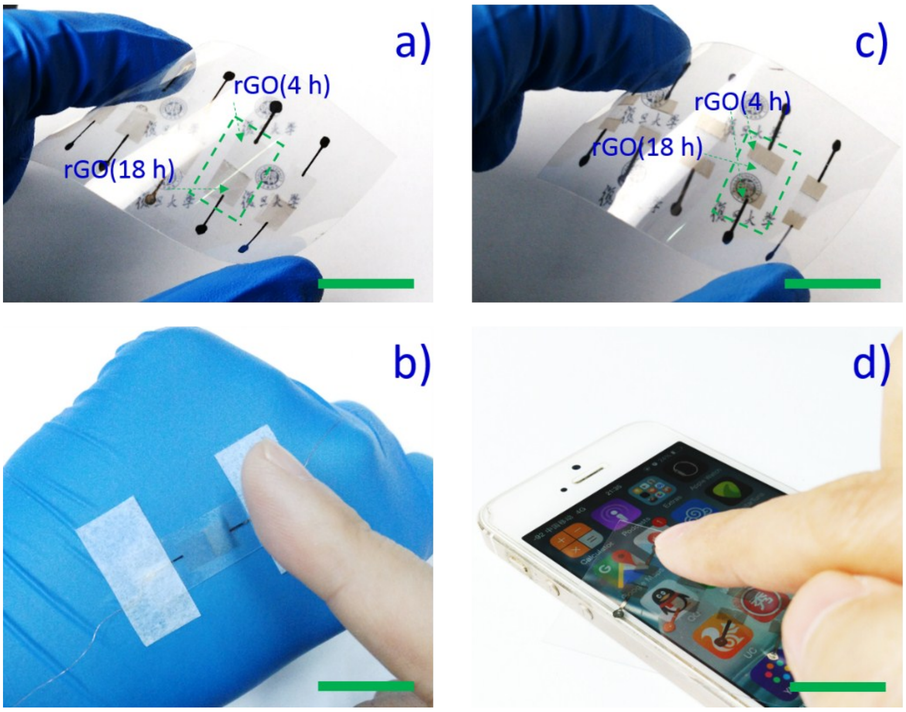


Figure S5. Digital images of the fabricated flexible logic devices. a) and b) rGO-based Janus pattern for gesture recognition; c) and d) rGO-based sandwiched pattern for 3D noncontact sensing. All scale bar is 1 cm.

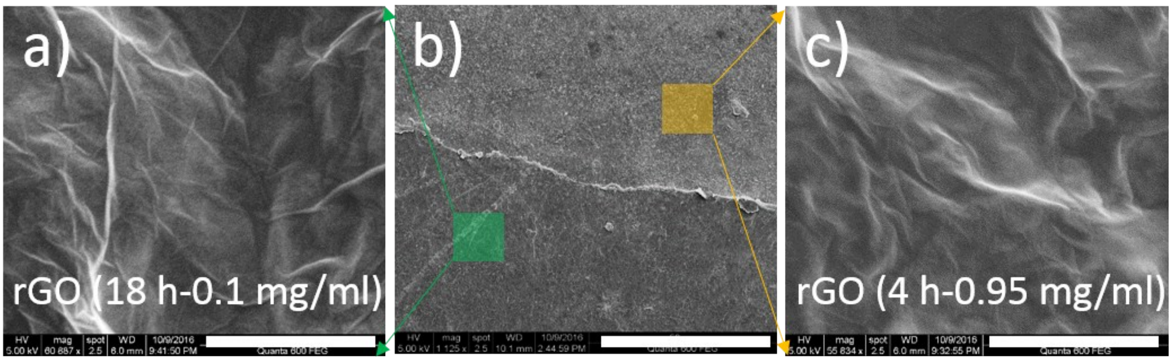


Figure S6. SEM images of the flxible logic devices. b) the connection between rGO film (4 h, 0.95 mg/ml) and rGO film (18 h, 0.1 mg/ml)) with a) and c) different magnifications for Janus or sandwich conductive patterns. The scale bar in a) and c) is 1 μm, and that in b) is 50 μm. Both films, and their contact line in b) can be seen clearly.

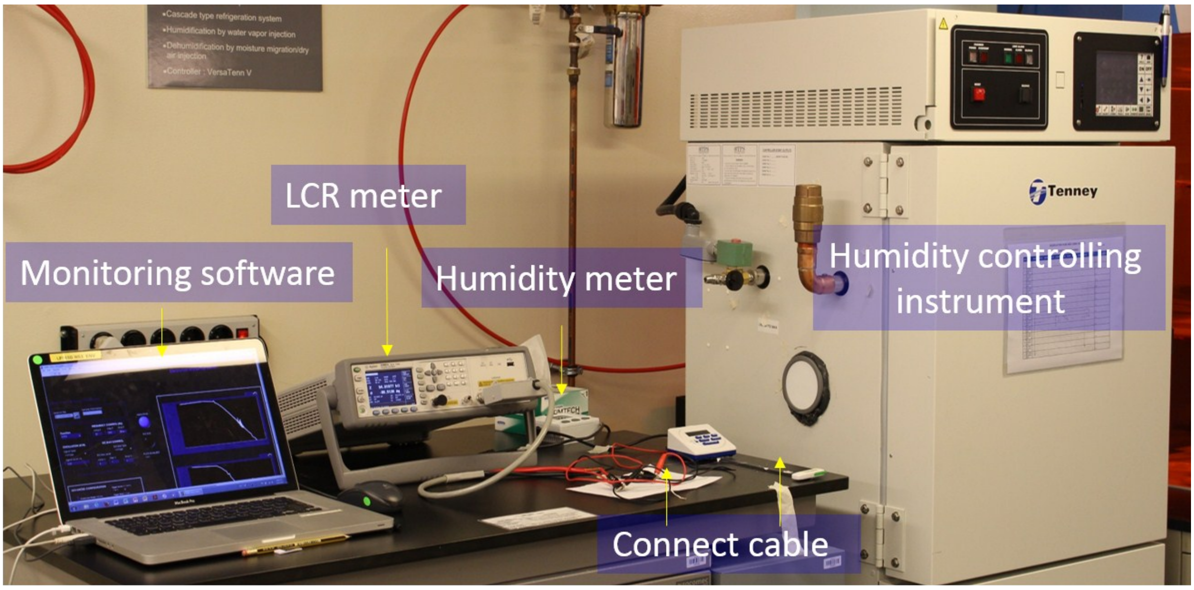


Figure S7. Digital image for the measurement of electrical impedance spectroscopy (EIS), which is composed of a monitoring software, LCR meter, humidity meter, and a climatic chamber.

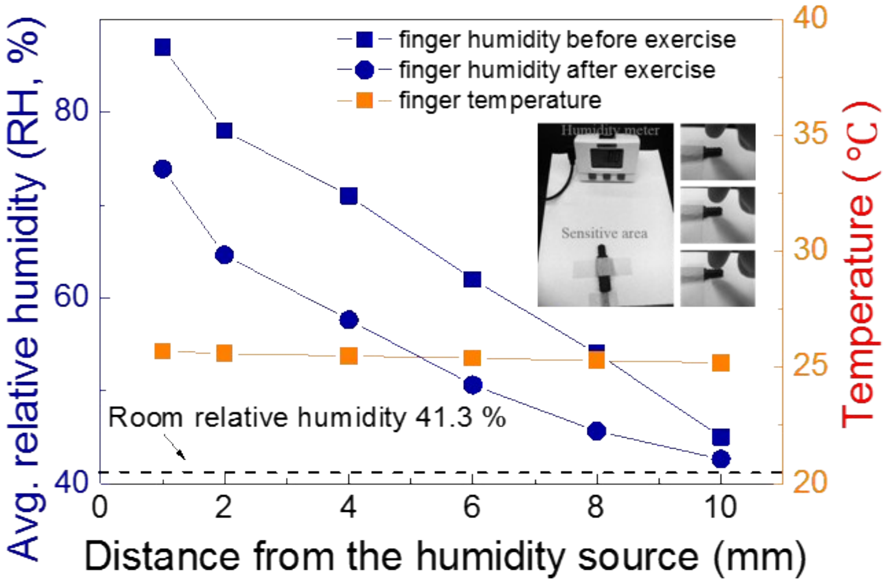


Figure S8. Relative humidity before and after exercise (measured by an RH reference sensor TM325 Dickson) and temperature detected as a function of the distance to the finger. Note that changes in temperature were minimal compared to changes in humidity, and thus any effect of temperature was neglected in this study.