

# **Robust, ultrathin, and highly sensitive reduced graphene oxide/silk fibroin wearable sensors responded to temperature and humidity for physiological detection**

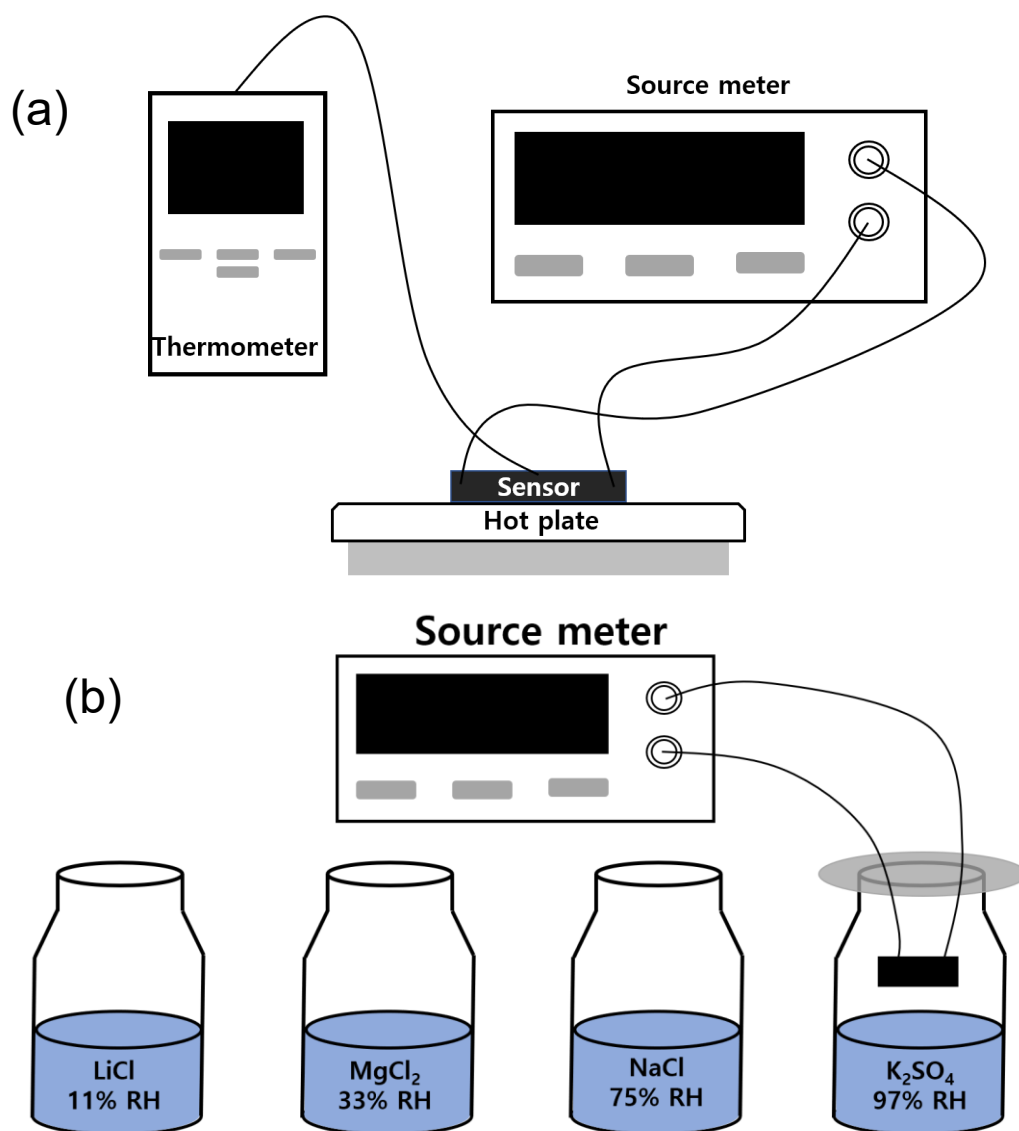
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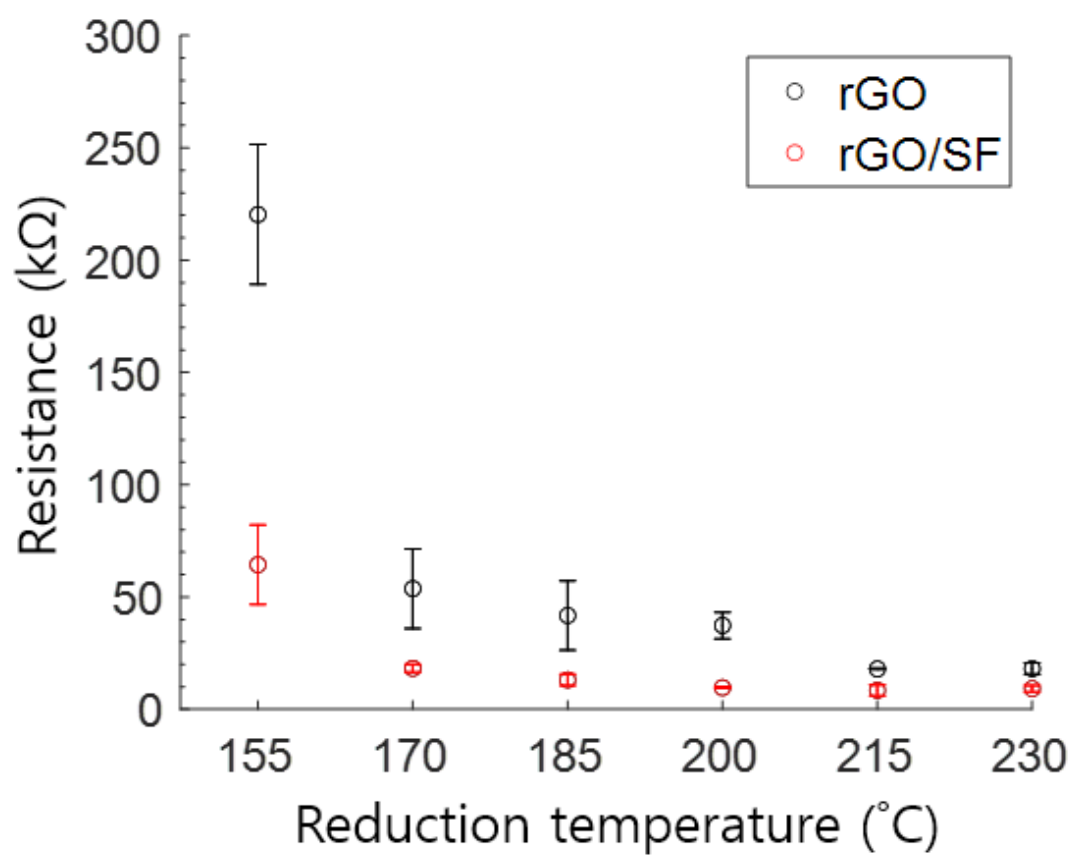
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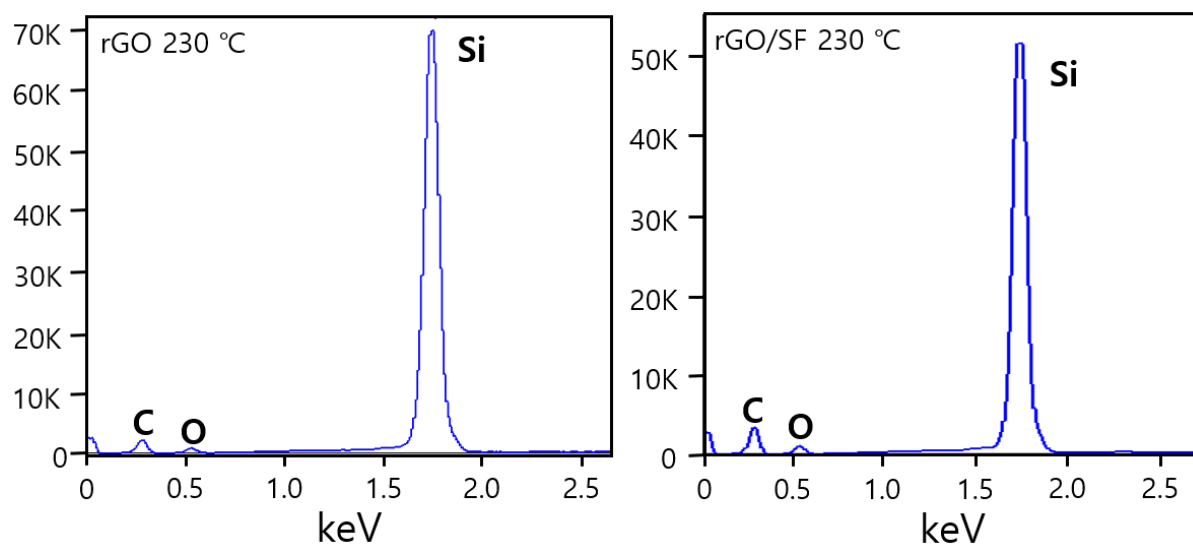
†Hyeonho Cho and Chanui Lee contributed equally to this work.



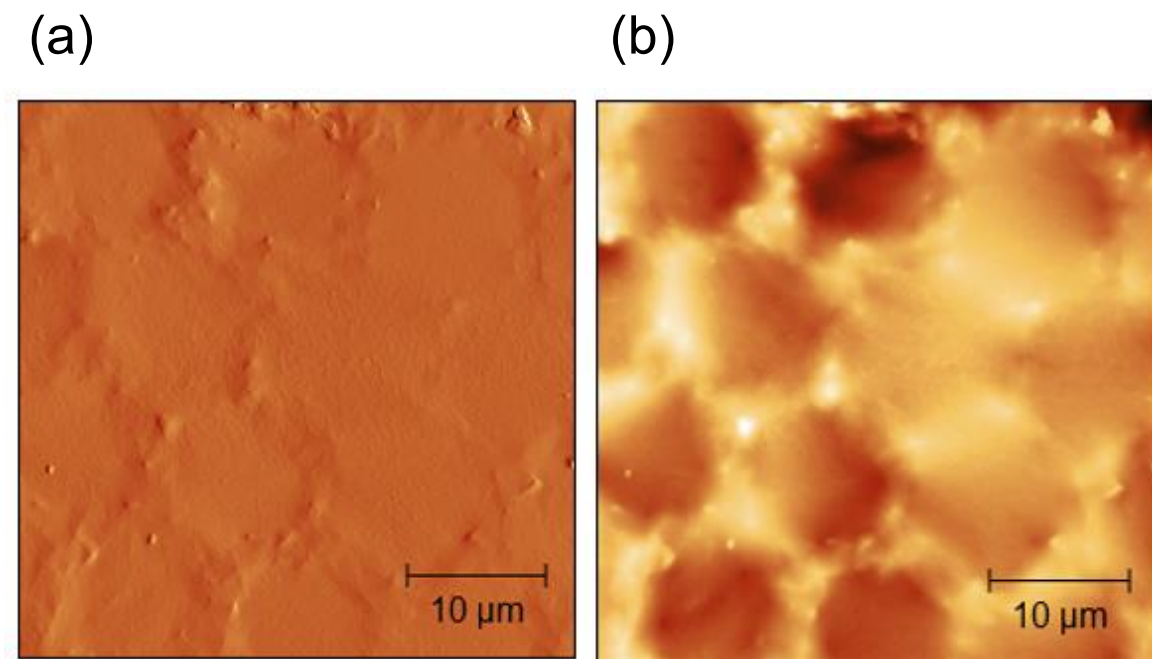
**Figure S1.** Schematic of the measurement methods used for the responsivity of a sensor: (a) varying the temperature and (b) varying the humidity.



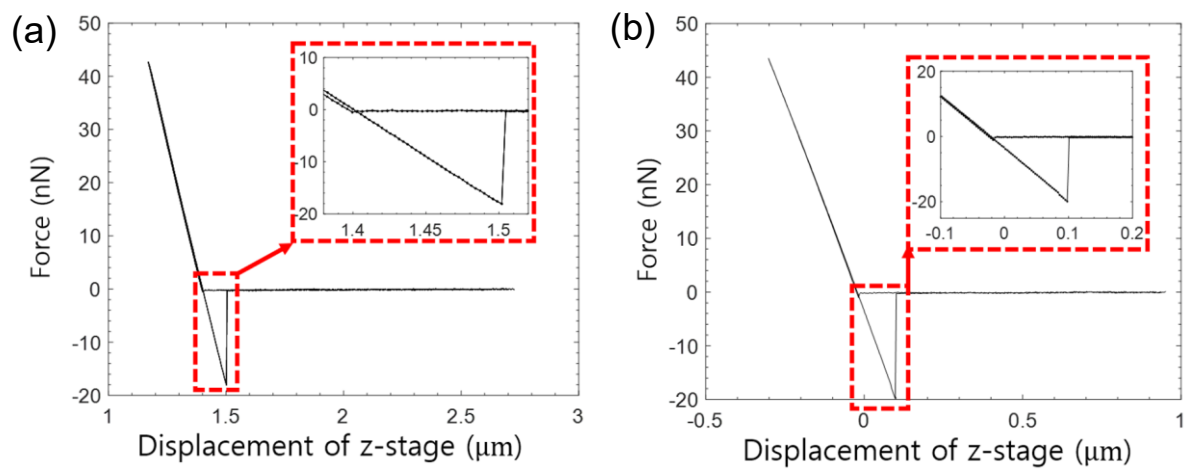
**Figure S2.** Resistance of rGO and rGO/SF sensors fabricated with different reduction temperatures.



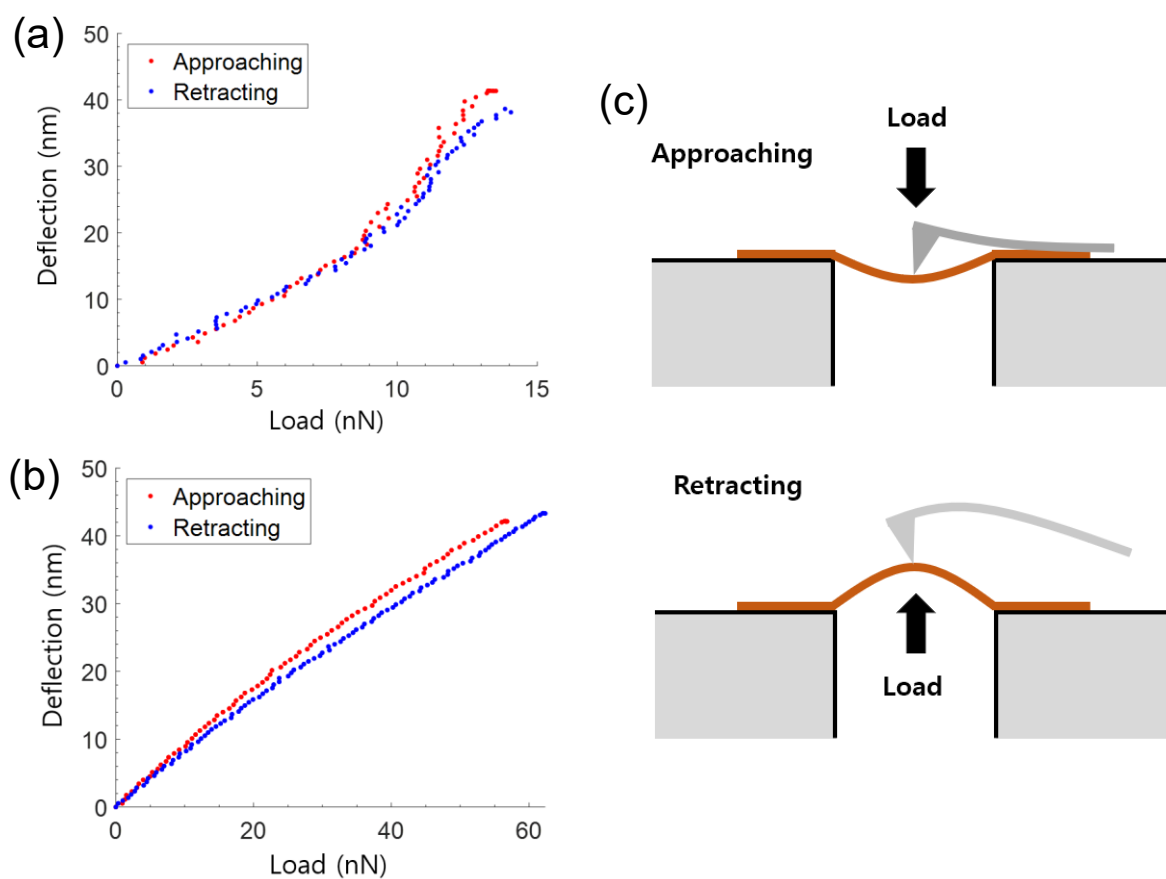
**Figure S3.** Representative EDS plots of rGO and rGO/SF films fabricated with a reduction temperature of 230 °C.



**Figure S4.** AFM images: (a) a force image and (b) a morphological image of an rGO/SF film scanned in the contact mode. The scale bars are 10 μm in length.

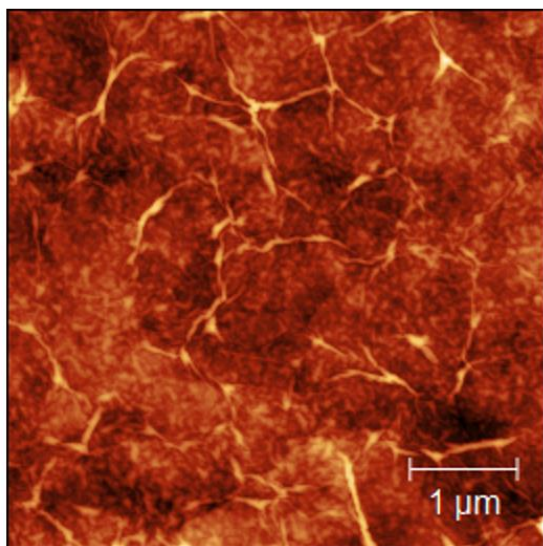


**Figure S5.** Force-displacement (FD) curves of the z-stage for (a) rGO and (b) rGO/SF films.

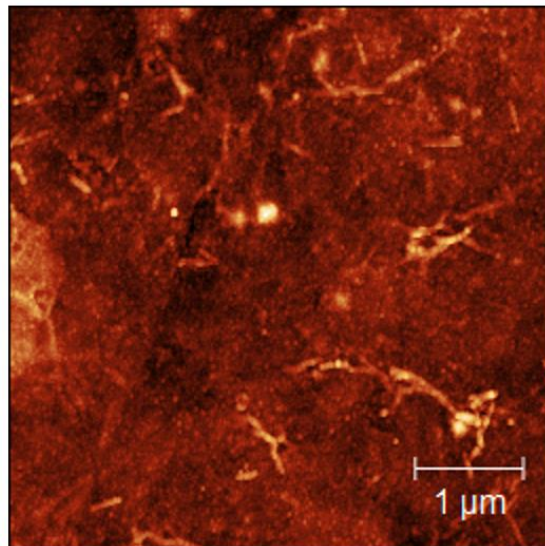


**Figure S6.** Load-deflection plots for (a) rGO and (b) rGO/SF films, constructed for (c) the measurement of the approaching and retracting directions.

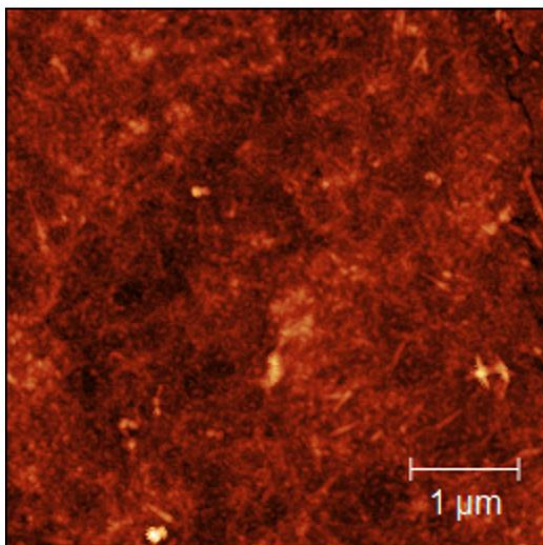
(a)

Roughness ( $R_q$ ):  $3.25 \pm 0.51$  nm

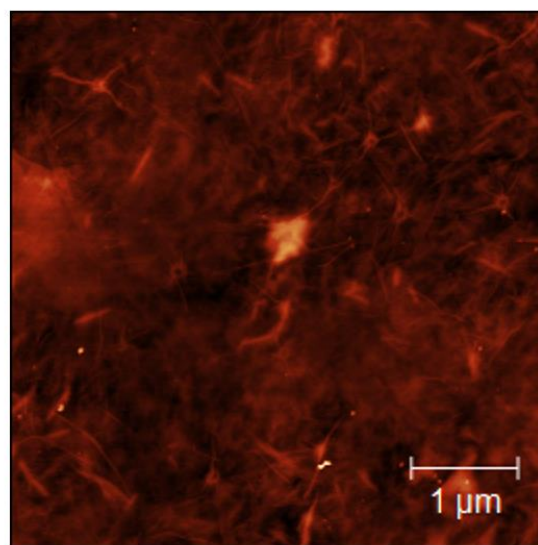
(b)

Roughness ( $R_q$ ):  $3.06 \pm 0.69$  nm

(c)

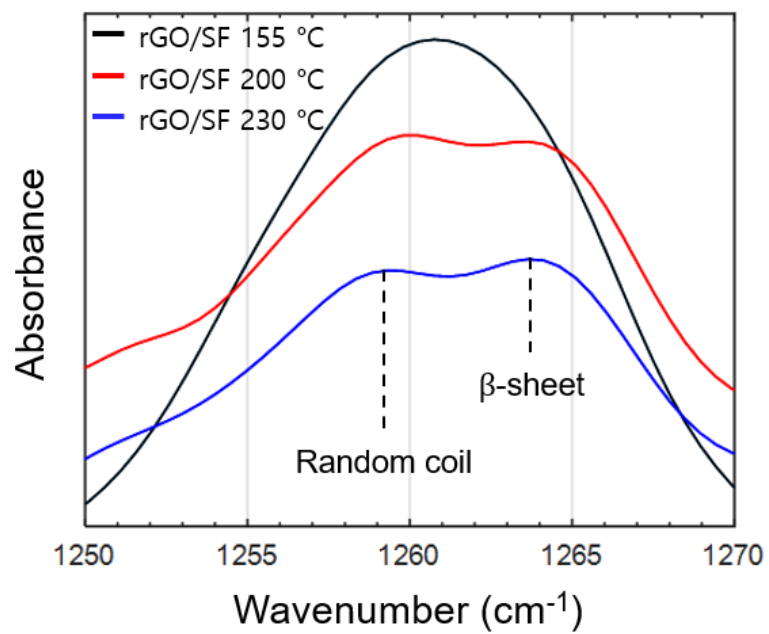
Roughness ( $R_q$ ):  $4.68 \pm 1.02$  nm

(d)

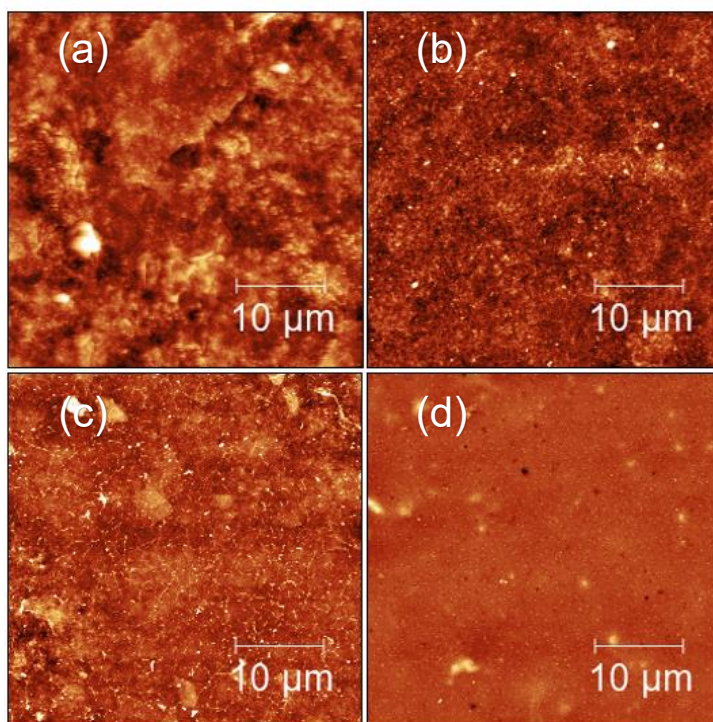
Roughness ( $R_q$ ):  $2.80 \pm 0.93$  nm

**Figure S7.** Morphological AFM images of rGO films fabricated with reduction temperatures of (a) 155 and (b) 230 °C. Morphological AFM images of rGO/SF films fabricated with reduction temperatures of (c) 155 and (d) 230 °C.

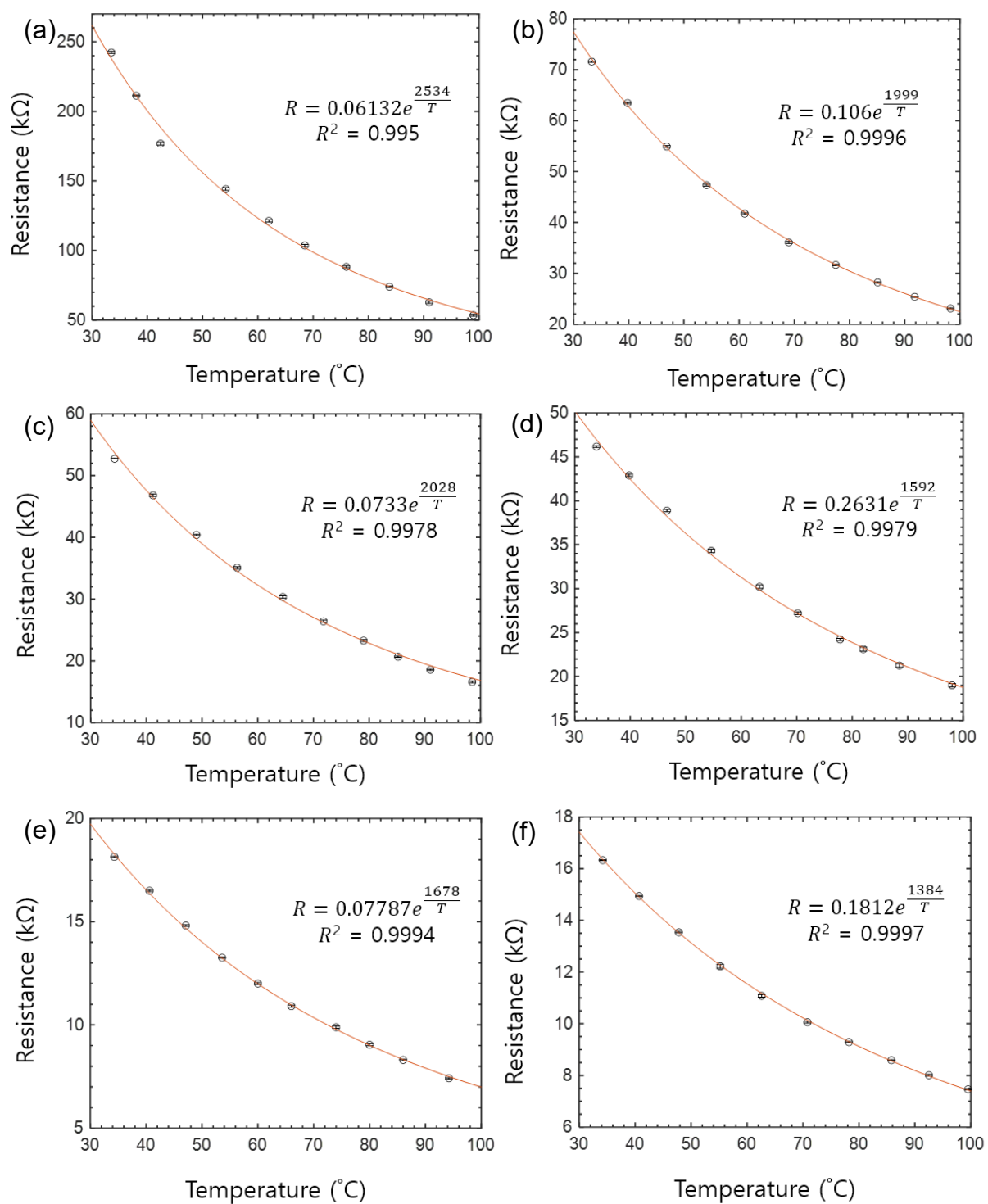




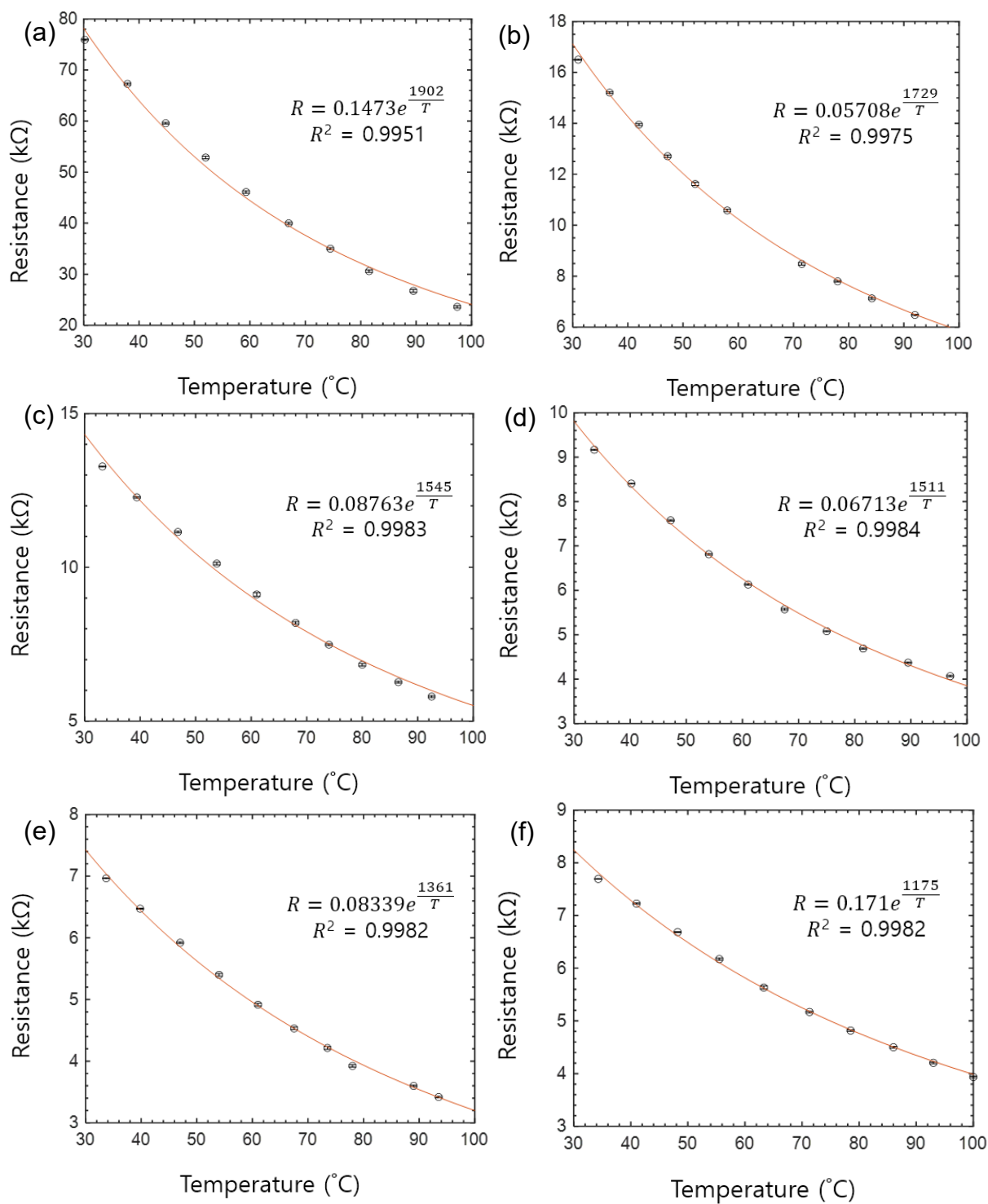
**Figure S8.** FTIR spectra of rGO/SF films fabricated with reduction temperatures of 155 (black), 200 (red), and 230 °C (blue).



**Figure S9.** AFM images of (a) an rGO/SF film without an LbL structure (z-scale: 400 nm), (b) an rGO/SF film with an LbL structure (z-scale: 70 nm), (c) an rGO film (z-scale: 30 nm), and (d) an SF film (z-scale: 70 nm).



**Figure S10.** Resistance-temperature curves of rGO-sensor-based temperature sensors fabricated with reduction temperatures of (a) 155, (b) 170, (c) 185, (d) 200, (e) 215, and (f) 230 °C.



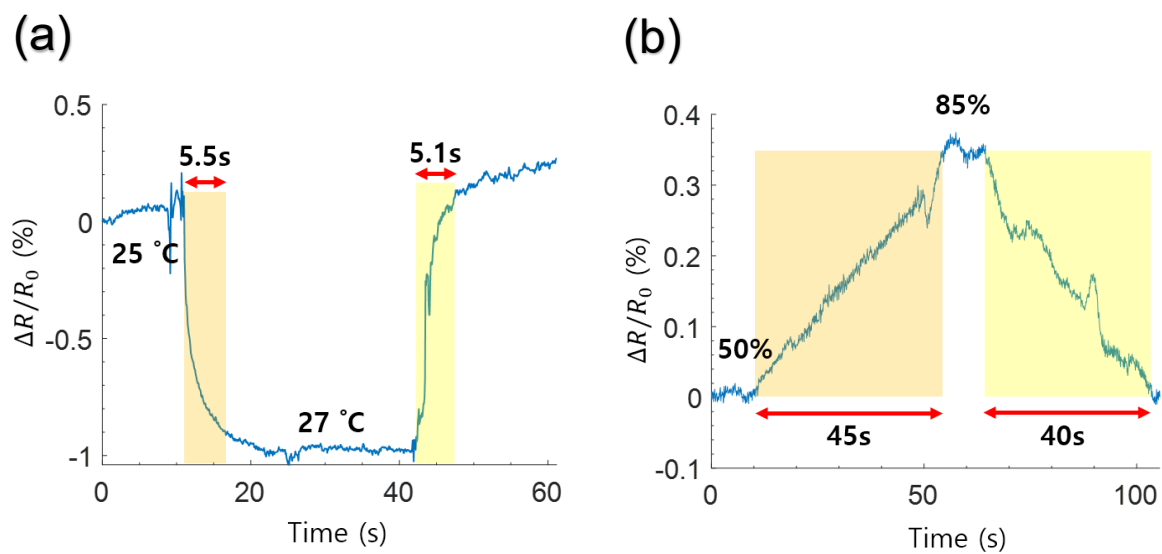
**Figure S11.** Resistance-temperature curves of rGO/SF-sensor-based temperature sensors fabricated with reduction temperatures of (a) 155, (b) 170, (c) 185, (d) 200, (e) 215, and (f) 230 °C.

**Table S1.** Specifications and comparison of some wearable temperature sensors and their performance

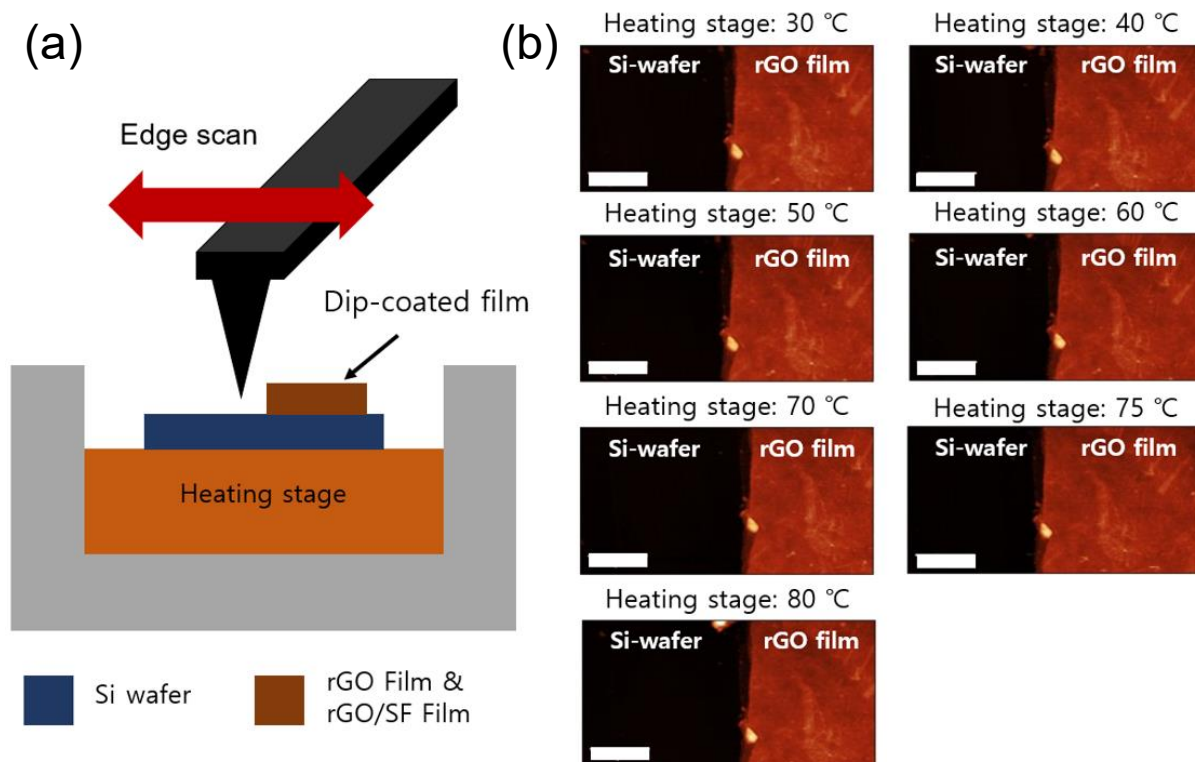
Sensing material	Sensitivity [%/K] at 300 K	Temp. range [°C]	Method	Response (recovery) time(s)	Reference
rGO	0.63	30–80	Wet-spinning	7 (20)	[1]
Crosslinked PEDOT:PSS	0.77	25–50	Printing	1.5 (6)	[2]
rGO/CB	0.6	20–60	Spray-coating	100	[3]
rGO	1.30	25–45	Spray-coating	0.33	[4]
rGO/PU	1.20	30–80	Wet-spinning	7 (70)	[5]
GO/PEDOT:PSS	1.09	25–100	Printing	18 (32)	[6]
rGO-based materials	2.77	30–100	Dip-coating	5.5 (5.1)	This work

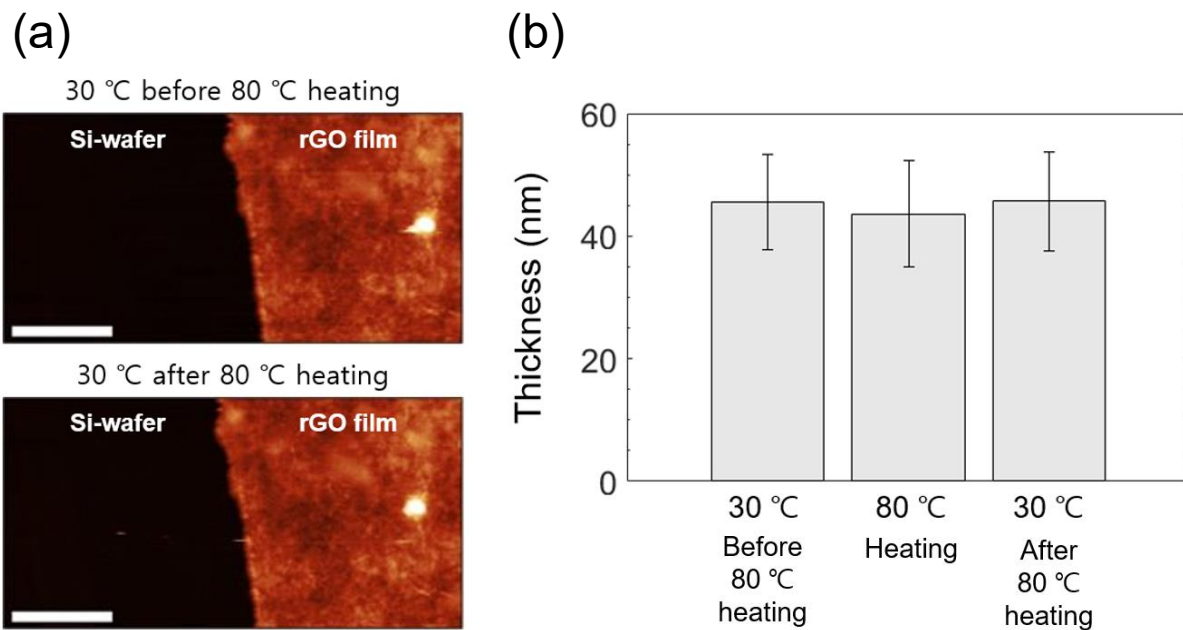
**Table S2.** Specifications and comparison of some wearable humidity sensors and their performance

Sensing material	Sensor type	Sensitivity	Humid. range [%RH]	Response (recovery) time(s)	Reference
rGO/CB	Resistance	161.16%	16–95	300 (100)	[3]
PDDA/RGO	Resistance	8.69%–37.43%	11–97	108–147 (94–133)	[7]
Pt-nRGO	Resistance	4.51%	6.1–66.4	-	[8]
GO	Capacitance	3215.25 pF/%RH	10–90	15.8	[9]
LIG/GO	Capacitance	9150 pF/%RH	11–97	49 (2)	[10]
rGO/ND	Capacitance	13086 pF/%RH	11–97	8 (1.5)	[11]
rGO-based materials	Resistance	48.28%	11–97	45 (40)	This work



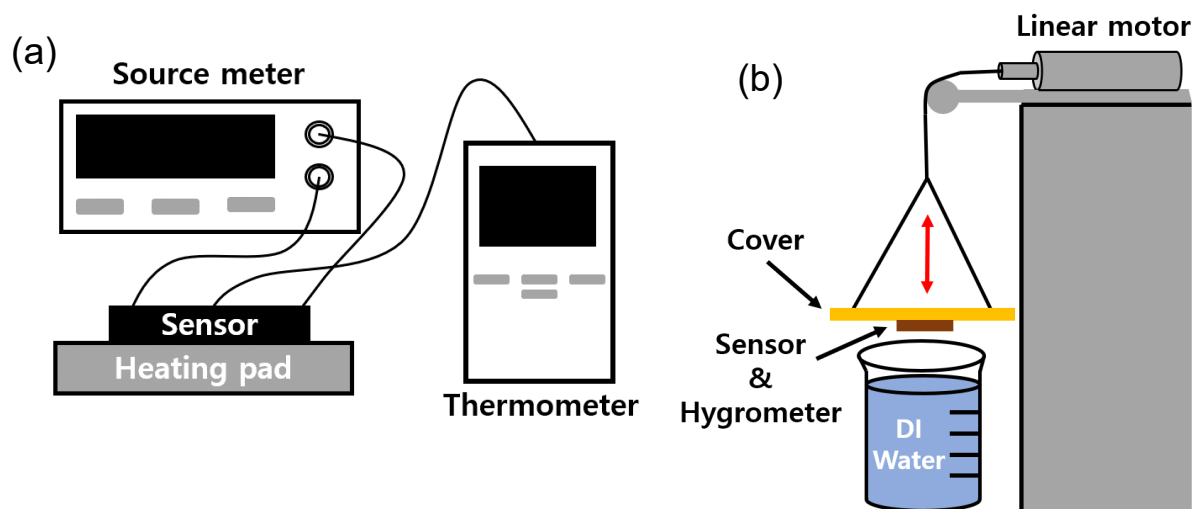
**Figure S12.** Response and recovery curves of the rGO/SF sensor for a reduction temperature of 170 °C: (a) between the temperatures of 25 °C to 27 °C and (b) between relative humidity values of 50% to 85%.





**Figure S14.** (a) AFM images of an rGO film at 30 °C before and after heating the film to 80 °C. (b) Thickness of the rGO film at 30 °C before heating, at 80 °C, and at 30 °C after heating and cooling. All scale bars are 3  $\mu\text{m}$ .

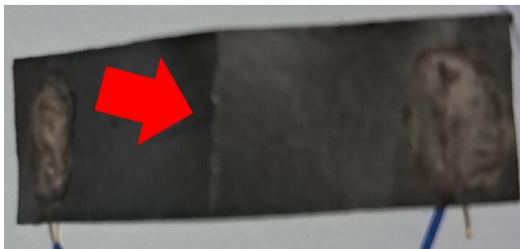




**Figure S15.** Schematic of the measurement methods used for the cyclic responsivity of sensors: (a) varying the temperature and (b) varying the humidity.

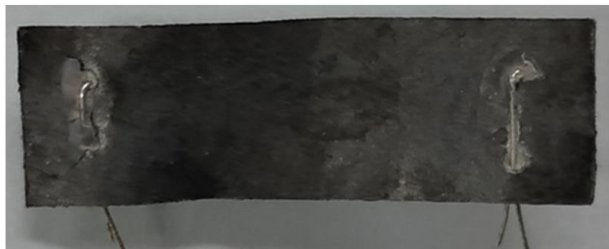
(a)

rGO 200 °C: Delamination



(b)

rGO/SF 200 °C: No delamination



**Figure S16.** (a) Surface of rGO 200 °C and (b) rGO/SF 200 °C after the repeated bending test. The surface of rGO 200 °C was delaminated in the region indicated by the red arrow.

## References

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