Supporting Information

Flexible and Transparent Strain Sensors Based on Super-aligned Carbon Nanotube

Films

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I. Supplemental Figures



Figure S1 Stress-strain curves of the PDMS film and the SACNT/PDMS film.



Figure S2 Number of SACNT bundles along the strain direction of the SACNT film at different applied strains.



Figure S3 Normalized resistance changes of the SACNT/PDMS strain sensor in response to 100%, 200%, 300%, and 400% step strains.



Figure S4 Normalized resistance changes of the SACNT/PDMS strain sensor during cycling under 1 Hz, 0.2 Hz, and 0.1 Hz frequencies at 400% strain.



Figure S5 Normalized resistance changes of the continuous cycles from the 2nd to 5000th stretching-releasing cycle at 400% strain.



Figure S6 Stress-strain curves of the SACNT/PDMS film during the 1st, 10th, 500th, and 5000th stretching-releasing cycles at 0-400% strains.



Figure S7 Normalized resistance changes of the SACNT/PDMS strain sensor attached on single finger at different bent angles.



Figure S8 A lab-designed tensile module to stretch the SACNT/PDMS films within the SEM chamber for *in situ* morphology observation.

II. Experimental Details

Preparation of the SACNT film and the PDMS substrate:

SACNT arrays with a diameter of 20–30 nm and a height of 300 µm were synthesized on silicon wafers by chemical vapor deposition with acetylene as the precursor and iron as the catalyst. Details of the synthesis procedure of the SACNT arrays can be found in previous reports^[34]. Continuous SACNT films were directly drawn from the SACNT arrays by an end-to-end joining mechanism. The thickness of the SACNT film was about 200 nm. The alignment of CNTs in the film was parallel to the drawing direction. The PDMS substrate with a thickness of 0.6 mm was prepared by mixing the base and the curing agent (Sylgard 184, Dow Corning) at a weight ratio of 20:1. The mixture was cured at room temperature for 24 h. Then the PDMS substrate was cut into a size of 50 mm × 50 mm.

Preparation of the SACNT/PDMS composite:

The SACNT film was directly coated onto a PDMS substrate with the alignment of the SACNTs perpendicular to the strain axis. Alcohol was dropped onto the SACNT film to shrink the film. This process resulted in the development of a strong van der Waals contact between the SACNT film and the PDMS substrate. After the evaporation of the alcohol, the SACNT/PDMS composite was cut into sizes of 50 mm \times 5 mm, with silver paste applied at both ends.

In situ SEM observation:

The morphologies of the SACNT/PDMS films were examined by scanning electron microscopy (SEM, Sirion 200, FEI). A lab-designed tensile module (Figure S8) was integrated within the SEM chamber to stretch the SACNT/PDMS samples. The gauge length was 5 mm and the strain rate was 10% s⁻¹.

Characterization:

The resistance of the SACNT/PDMS strain sensor applied at a strain up to 400% was characterized. The dimension of the sample was 10 mm × 5 mm × 0.6 mm. Two Cu foils were attached at both ends of the sample using a conductive silver paste for resistance measurements. Tensile strain was applied using an Instron 5848 microtester and the resistance of the SACNT/PDMS sample was monitored by a Keithley 2400 Source Meter. The durability of the SACNT/PDMS conductor was characterized by performing cyclic tensile tests with 400% strain for 5,000 cycles and the resistance of the sample was measured at each stretching cycle. The morphologies of the SACNT/PDMS film were further examined by SEM before and after the cyclic tensile testing. The optical transmittance of the SACNT/PDMS sample was measured using a PerkinElmer-Lambda 950 (UV-VIS-NIR) spectrophotometer.