Supplementary Materals

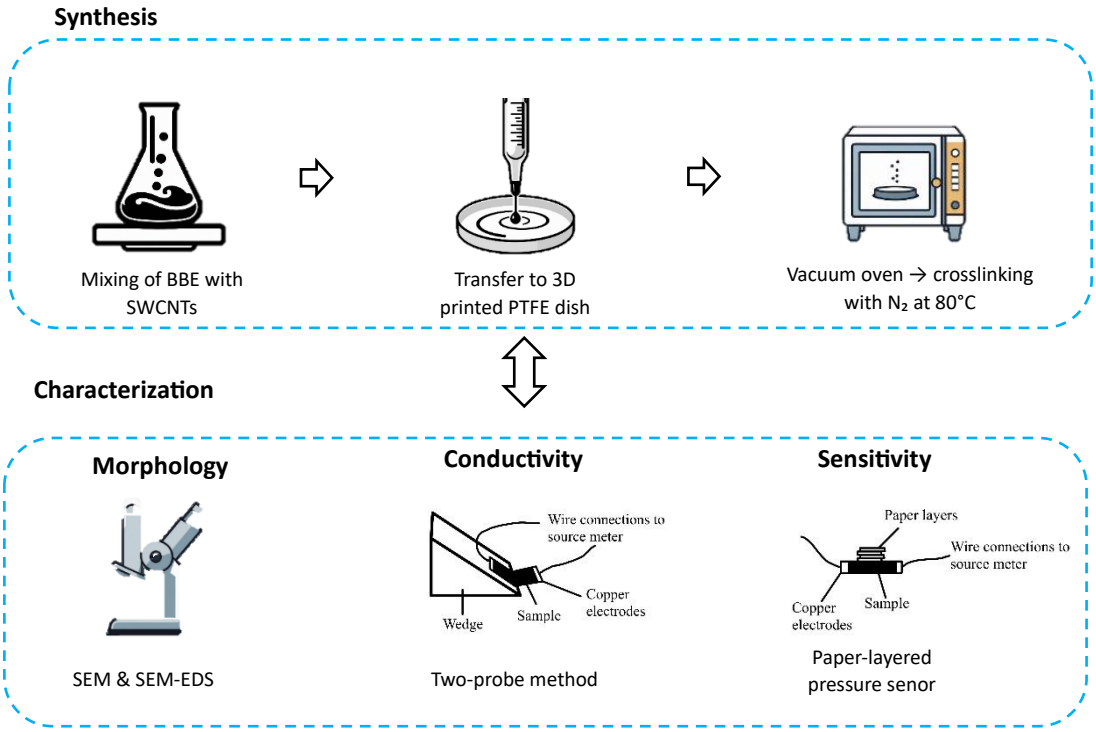


Figure S1. Schematic of the overall process: synthesis involved mixing of the BBE precursor with PDMS:CL ratios of 600:1, 800:1 and 100:1 with SWCNTs at different wt% of 0.04, 0.06 and 0.08%. Next the mixture was transferred into a 3D printed PTFE dish using a serological pipette and placed in a vacuum over overnight. The mixture was then transferred to an 80°C oven with N2 for 12 hours. Once synthesized, the BBE/CNT composite was characterized and the morphology, conductivity, sensitivity, and fatigue were tested. The method used for conductivity test in this schematic diagram is the two-point probe approach.

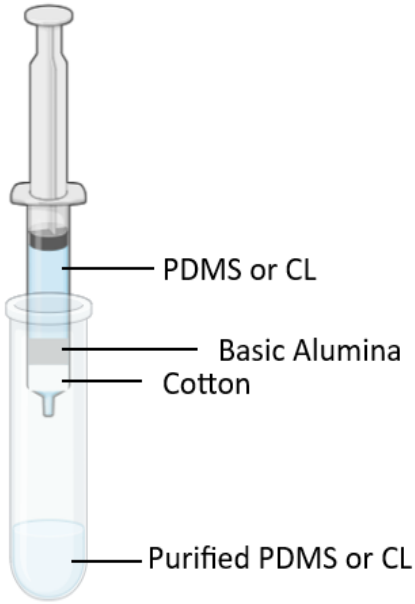


Figure S2. In-house purification column to purify PDMS and crosslinker. A 10mL syringe without a needle was packed with a layer of cotton (~3mm) at the bottom, followed by 3g of basic alumina powder. The syringe was placed on top of a 50mL collection tube and stabilized using aluminum foil. The PDMS and crosslinker were passed through the purification column separately under gentle pressure to remove impurities.

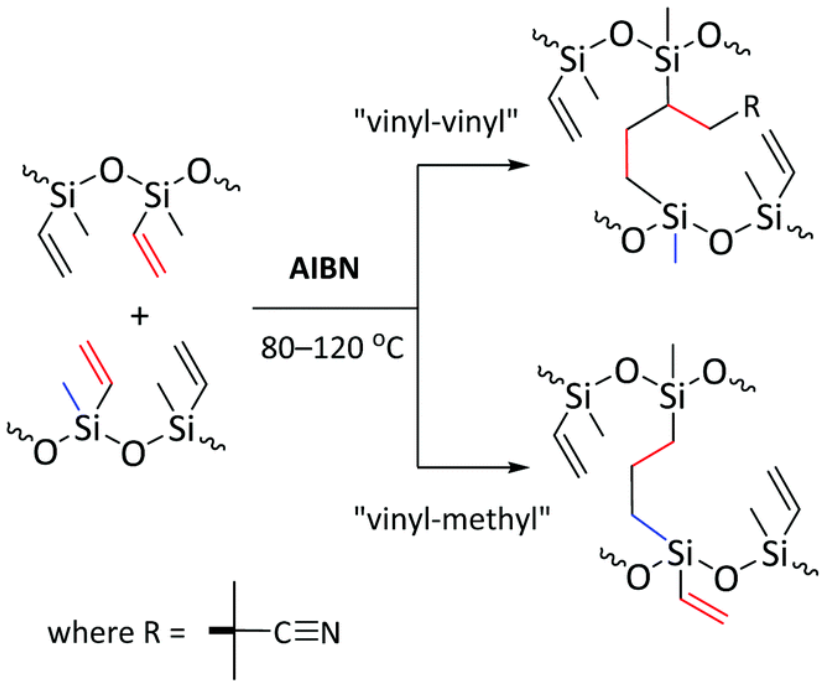


Figure S3. Reaction mechanism of AIBN-initiated crosslinking of PDMS-based bottlebrush elastomers [20].

Table S1. Formulations for each PDMS, crosslinker and AIBN mixtures with different CNT loadings. Each composition was based on 10mL of PDMS, with crosslinker volumes adjusted according to the PDMS:CL ratio and 2 mol% AIBN initiators relative to PDMS.

PDMS:CL ratio	CNT loading wt.%	PDMS (mL)	Crosslinker (uL)	AIBN mol.%
600:1	0.4	10	163.2	2
600:1	0.6	10	163.2	2
600:1	0.8	10	163.2	2
800:1	0.4	10	122.5	2
800:1	0.6	10	122.5	2
800:1	0.8	10	122.5	2
1000:1	0.4	10	97.96	2
1000:1	0.6	10	97.96	2
1000:1	0.8	10	97.96	2

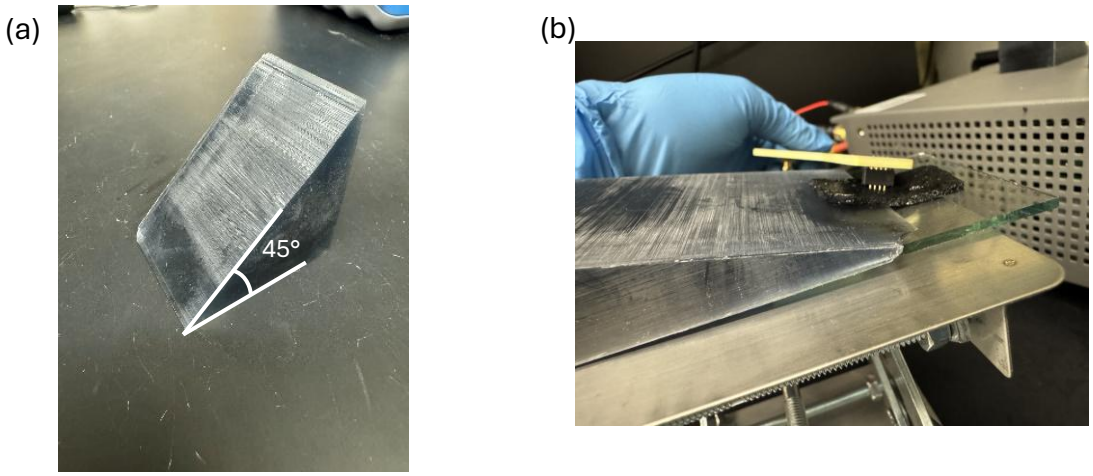


Figure S4. (a) Example of the 3D printed wedge (45°) used for angle dependent conductivity testing. (b) Example of the 3D printed wedge (30°) with half the sample was placed on the wedge and the other half placed on the surface with the four-point probe placed at the bending angle.

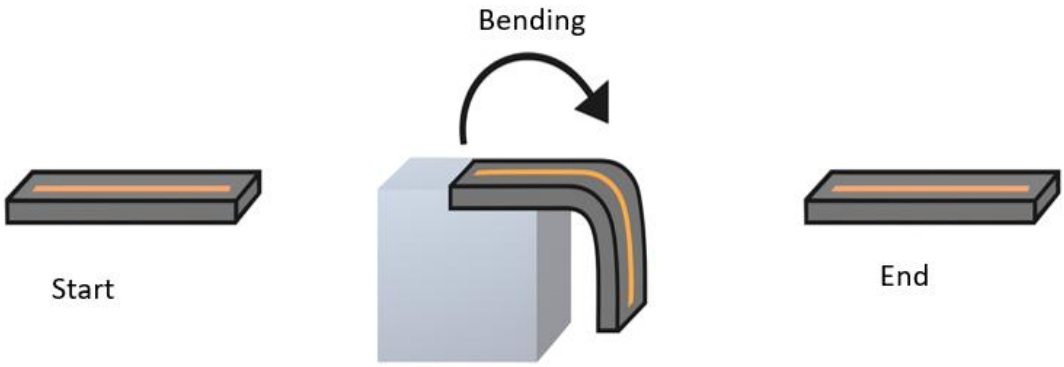


Figure S5. Fatigue testing method. Samples were manually bent over a 90° angle and then returned to flat for repeated cycle counts of 100, 200 and 300. Each bend-release cycle was performed at a rate of ~ 1 cycle every 2 seconds. Bending was applied at random positions on each sample surface to stimulate real-world conditions. The resistance was measured after each set of cycles using the two-point probe method.

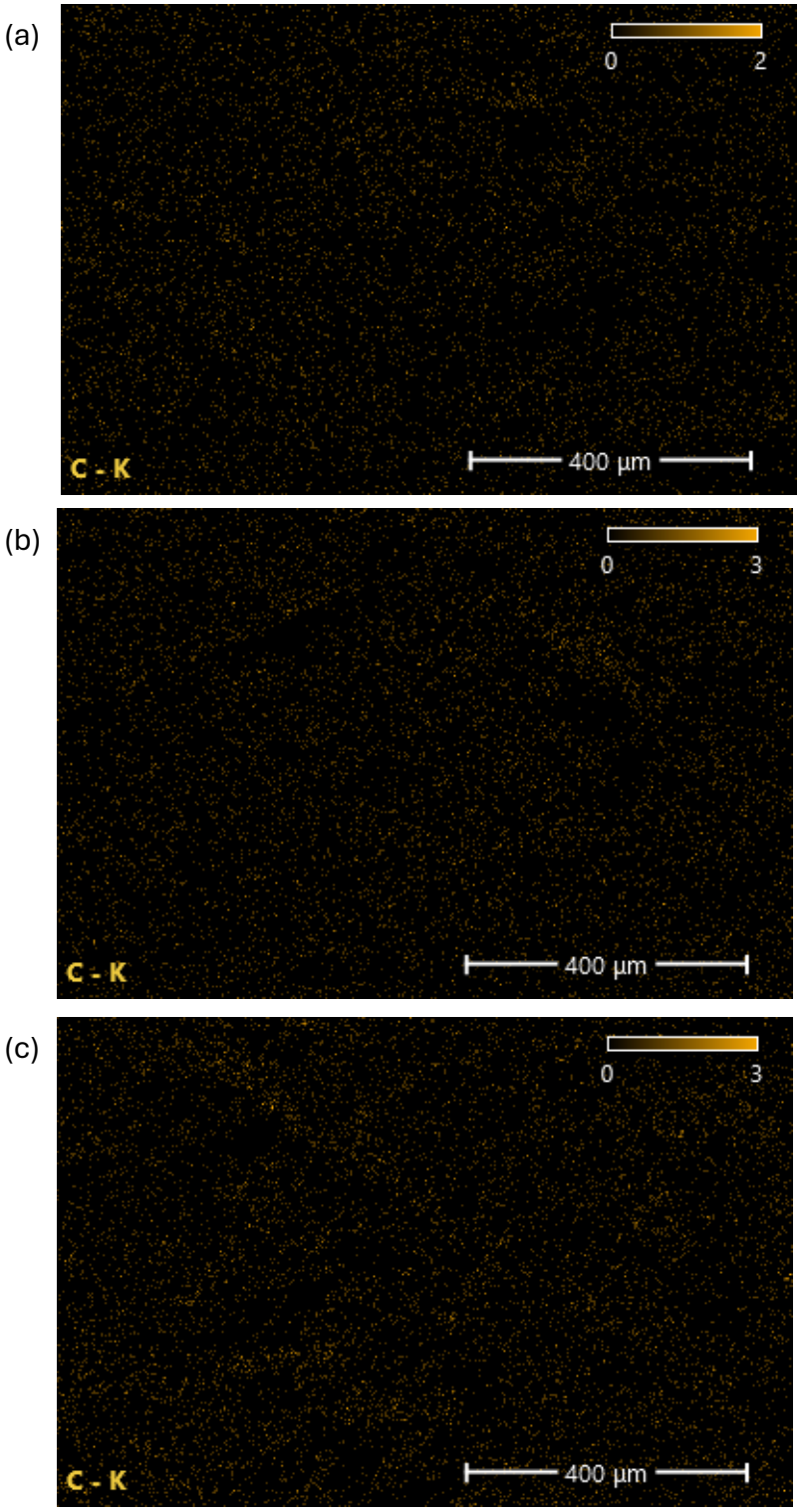


Figure S6. Elemental carbon (C-K) count maps of BBE/CNT composites with CNT loadings of (a) 0.4 wt. %, (b) 0.6 wt.%, (c) 0.8 wt.% at PDMS:crosslinker ratio of 600:1. The carbon signal is uniformly distributed across all samples, confirming homogenous CNT dispersion.

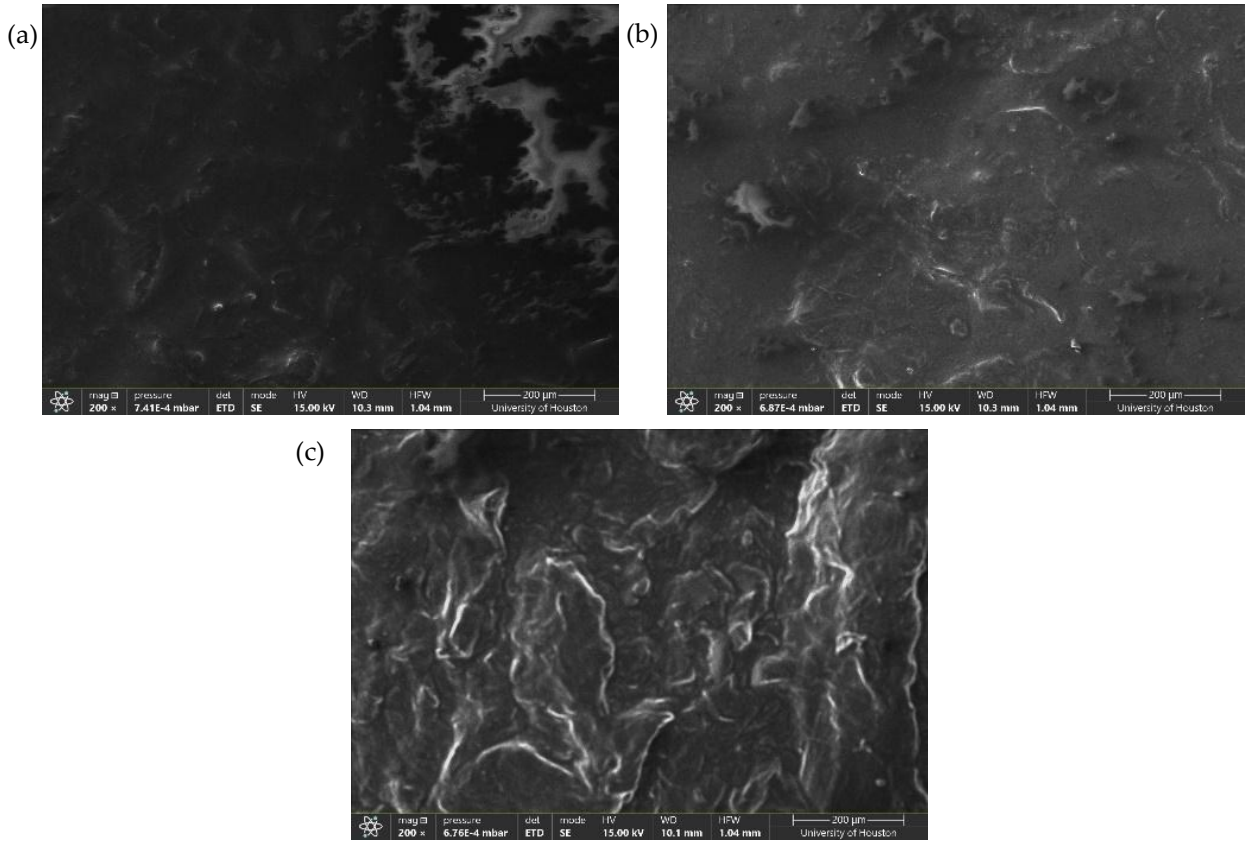


Figure S7. SEM images of the 600:1 BBE/CNT composite after 1000 manual bending cycles, showing a continuous surface morphology and preserved CNT network, compared to Figure 1 in the main text; (a) 0.4 wt.% (b) 0.6 wt.% (c) 0.8 wt.%.

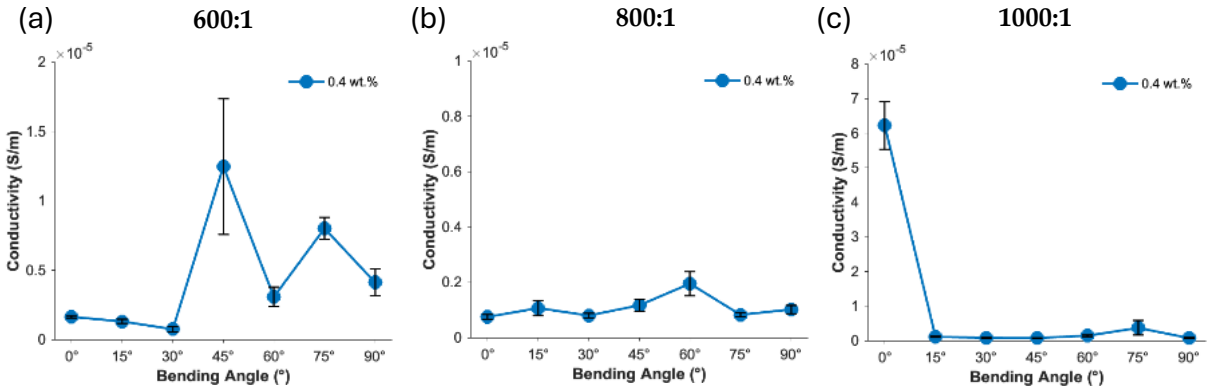


Figure S8. Conductivity of 0.4 wt.% CNT/BBE composites under bending angles (0-90°) at PDMS:crosslinker ratios of (a) 600:1, (b) 800:1, and (c) 1000:1. These rescaled plots highlight the small-magnitude fluctuations that are not visible in the main text due to the low absolute conductivity of the samples.

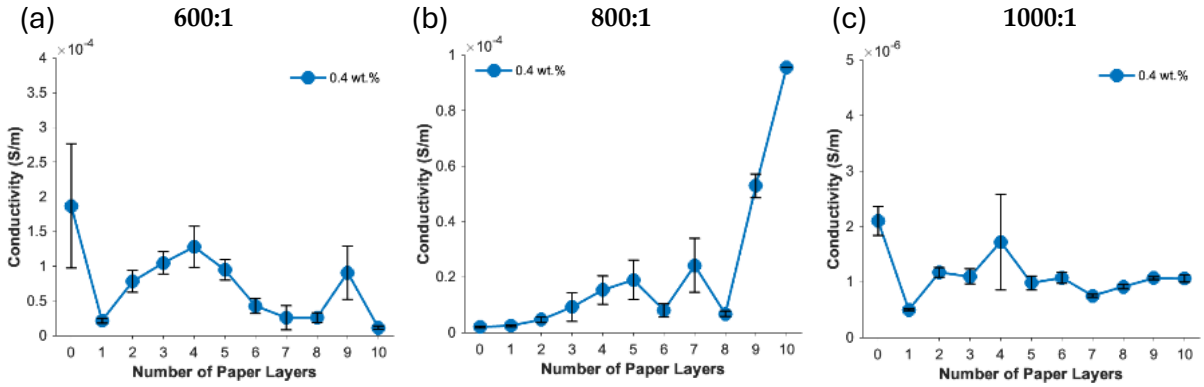


Figure S9. Conductivity response of 0.4 wt.% CNT/BBE composites as a function of paper layers with PDMS:crosslinker ratios of (a) 600:1, (b) 800:1, and (c) 1000:1.

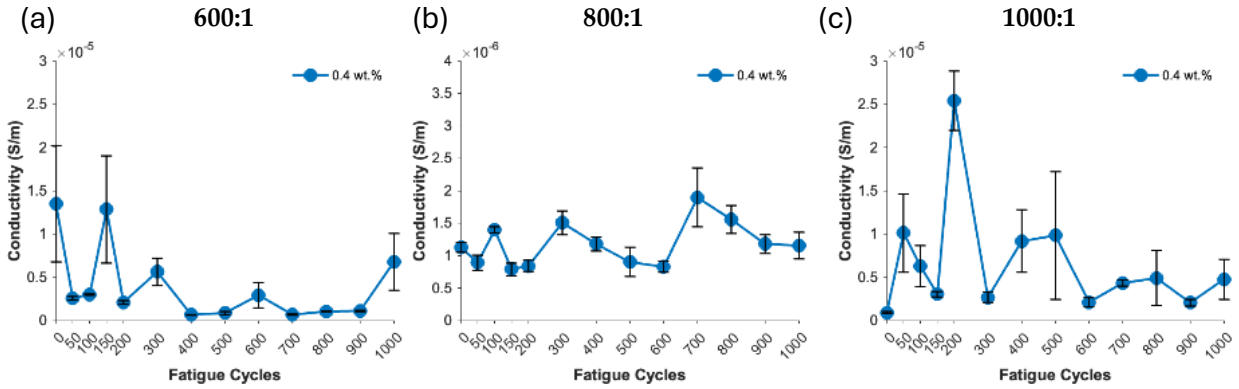


Figure S10. Conductivity response of 0.4 wt.% CNT/BBE composites measured over 1000 bending cycles at PDMS:crosslinker ratios of (a) 600:1, (b) 800:1, and (c) 1000:1

Additional Information on Normalization of Conductivity Data for Comparison

To compare electromechanical performance across compositions, all conductivity measurements were normalized to the baseline (unstrained) conductivity for each sample made with a specific formulation of CNT loading and PDMS:crosslinker molar ratio. For each formulation, the conductivity measured at each bending angle, pressure level, or fatigue cycle was divided by its initial value at 0° bending, 0 paper layers, or cycle 0. In other words, the normalization baseline changes across different formulations.

This normalization highlights the relative changes in conductivity within each formulation. Because the baseline conductivity varies among compositions, the normalized data are intended for trend comparison rather than quantitative comparison across samples. This approach enables clear visualization of each sample's response to mechanical deformation or loading conditions.

Conductivity was measured using both a two-point method and a four-point probe approach: the two-point data are presented first for comparison with previous studies as summarized in Table S2 below, followed by the four-point probe results, which provide higher measurement accuracy.

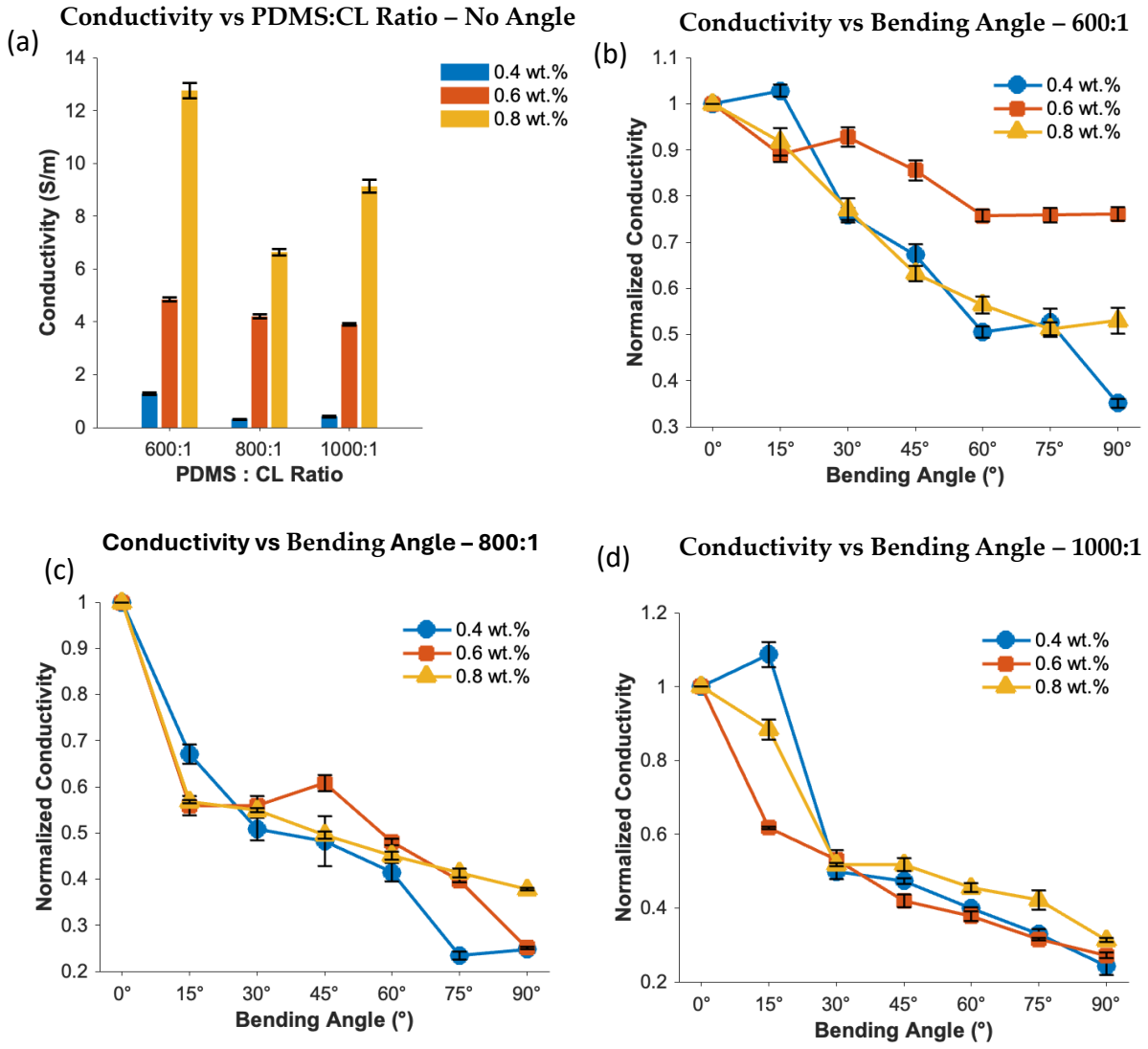


Figure S11. (a) Conductivity vs PDMS:crosslinker (CL) ratio (600:1, 800:1, 1000:1) at 0.4, 0.6, and 0.8 CNT wt%. (b-d) Normalized conductivity to 0° of 600:1 (b), 800:1 (c), and 1000:1 (d) samples with 0.4, 0.6, and 0.8 CNTs wt% at bending angles of 15°, 30°, 45°, 60°, 75°, 90° using the two-point probe method.

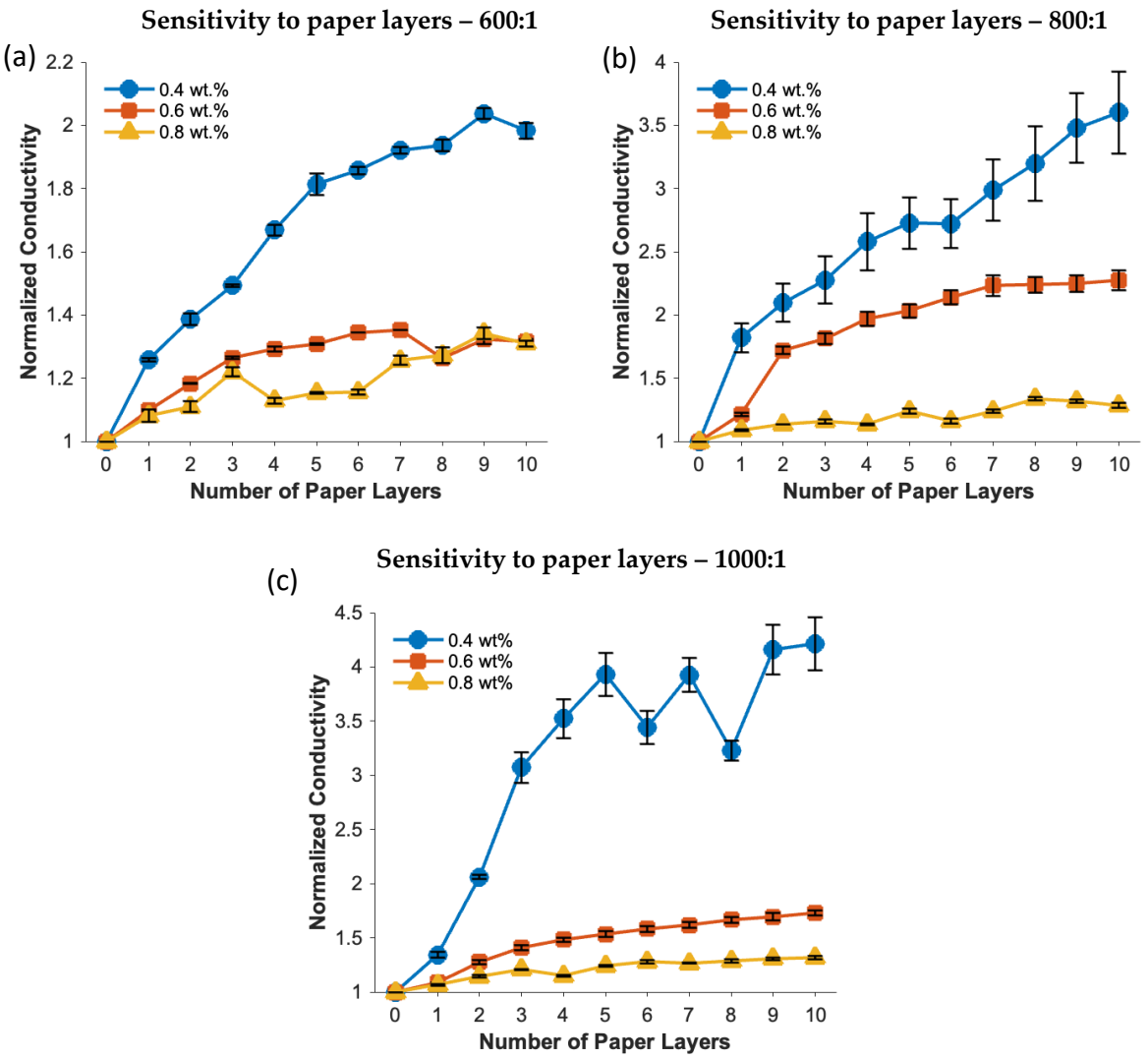


Figure S12. (a-c) Normalized conductivity or the sensitivity of 0-10 paper layers for each BBE:CL ratio (600:1, 800:1, 1000:1) and CNT wt% (0.4, 0.6, 0.8 wt%) using the two-point probe method

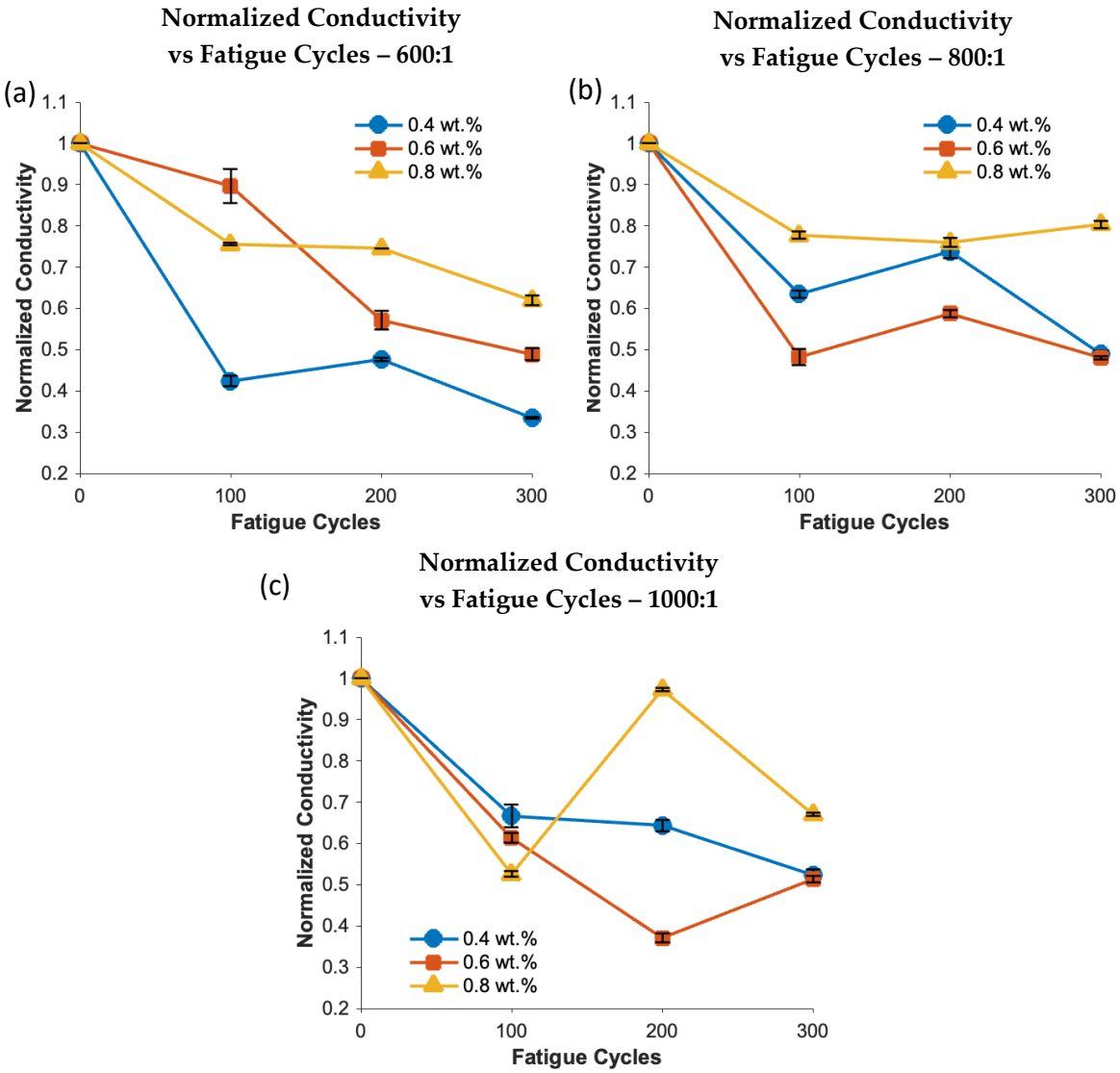


Figure S13. (a-c) Normalized conductivity of BBEs/CNTs composites over multiple fatigue cycles. Samples were tested at intervals of 100, 200, 300 cycles under repeated mechanical deformation using the two-point probe method. Results are shown for various PDMS:crosslinker ratios and CNT content.

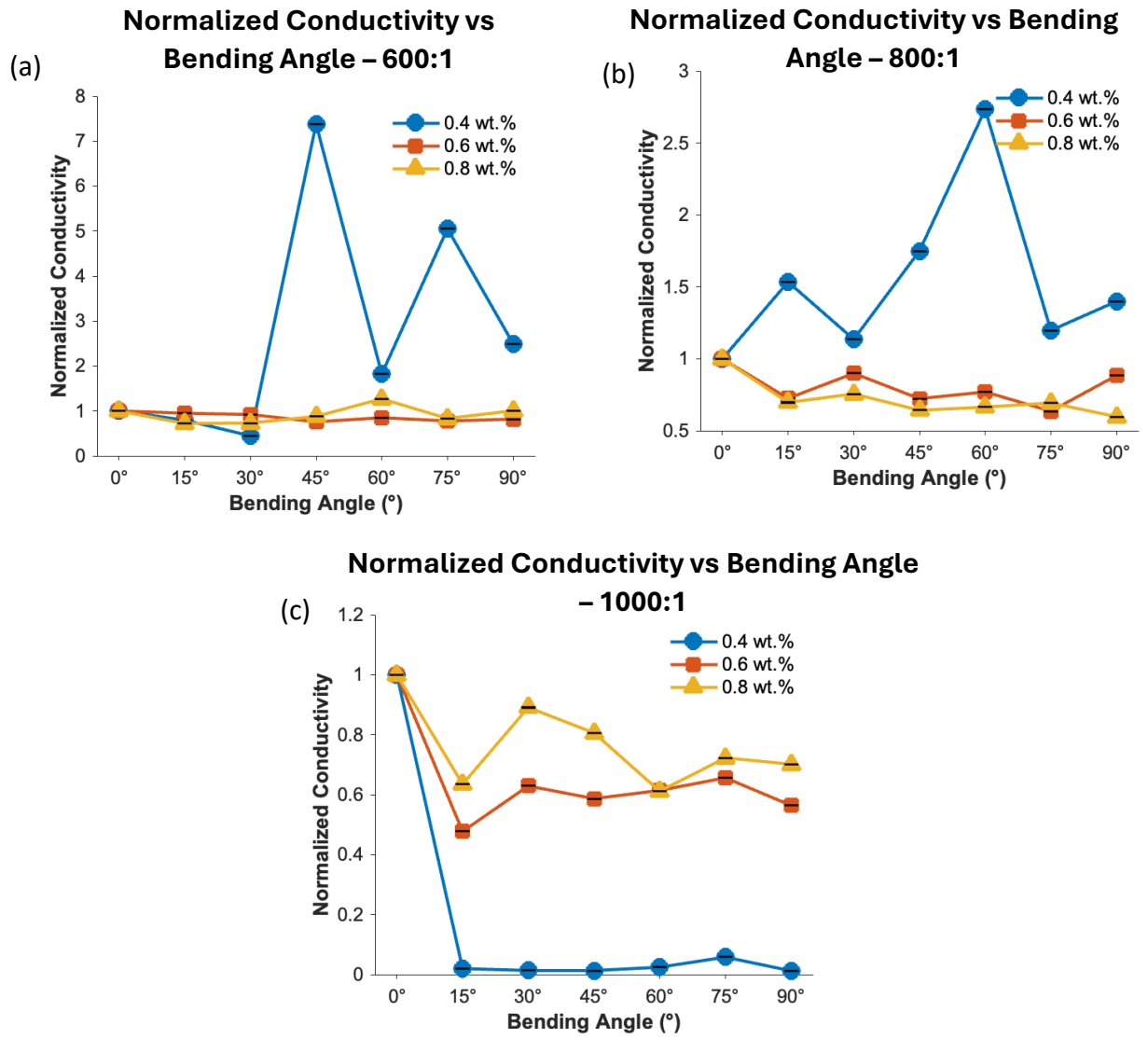


Figure S14. (a-c) Normalized conductivity of CNT/BBE composites measured by the four-point probe under bending using the four-point probe at PDMS:CL ratios of 600:1, 800:1, and 1000:1, respectively with CNT loadings of 0.4, 0.6 and 0.8 wt.%

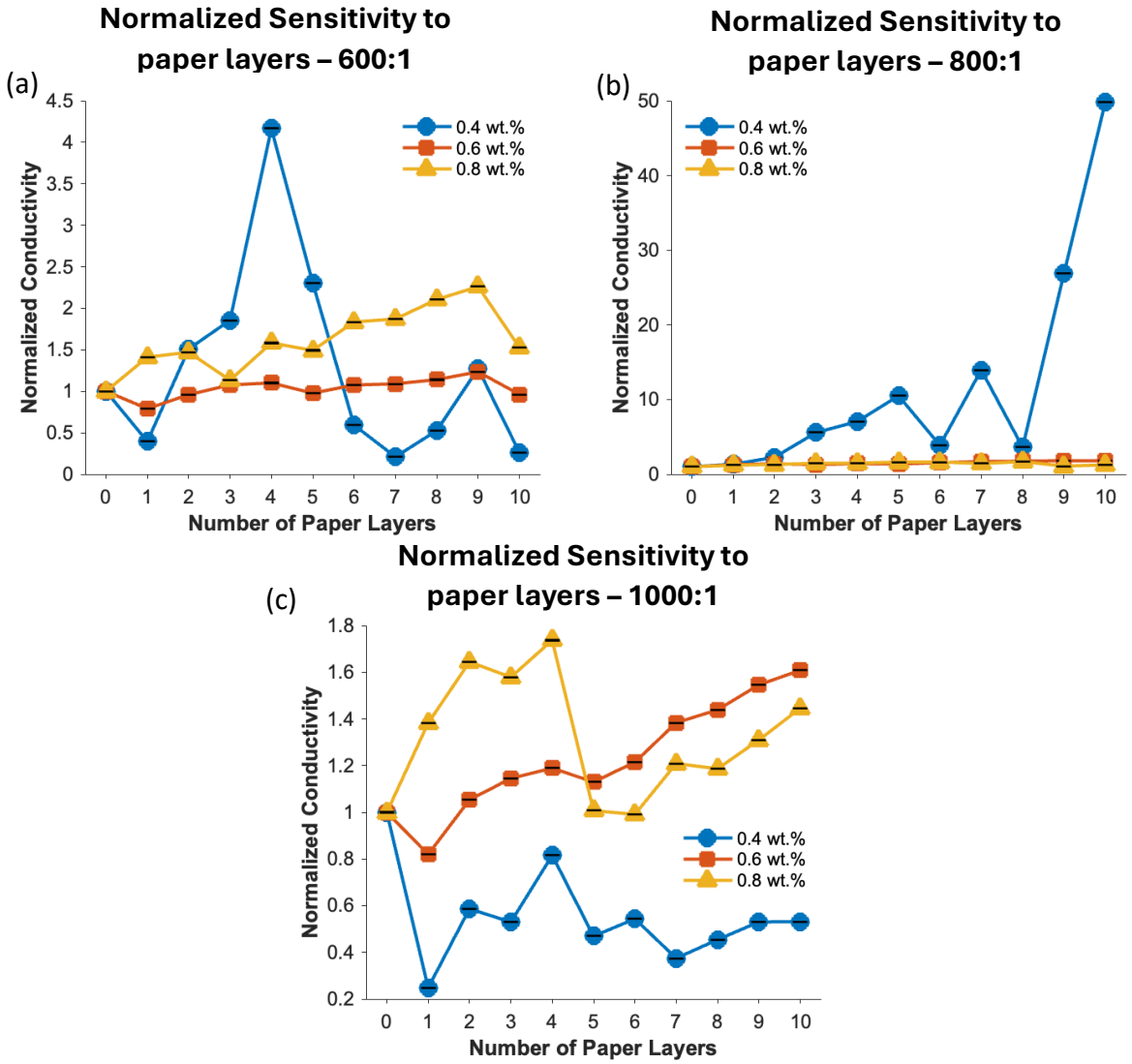


Figure S15. (a-c) Normalized conductivity response of CNT/BBE composites under incremental pressure (0-10 paper layers) using the four-point probe for PDMS:CL ratios of 600:1, 800:1, and 1000:1 respectively with CNT loadings of 0.4, 0.6 and 0.8 wt.%

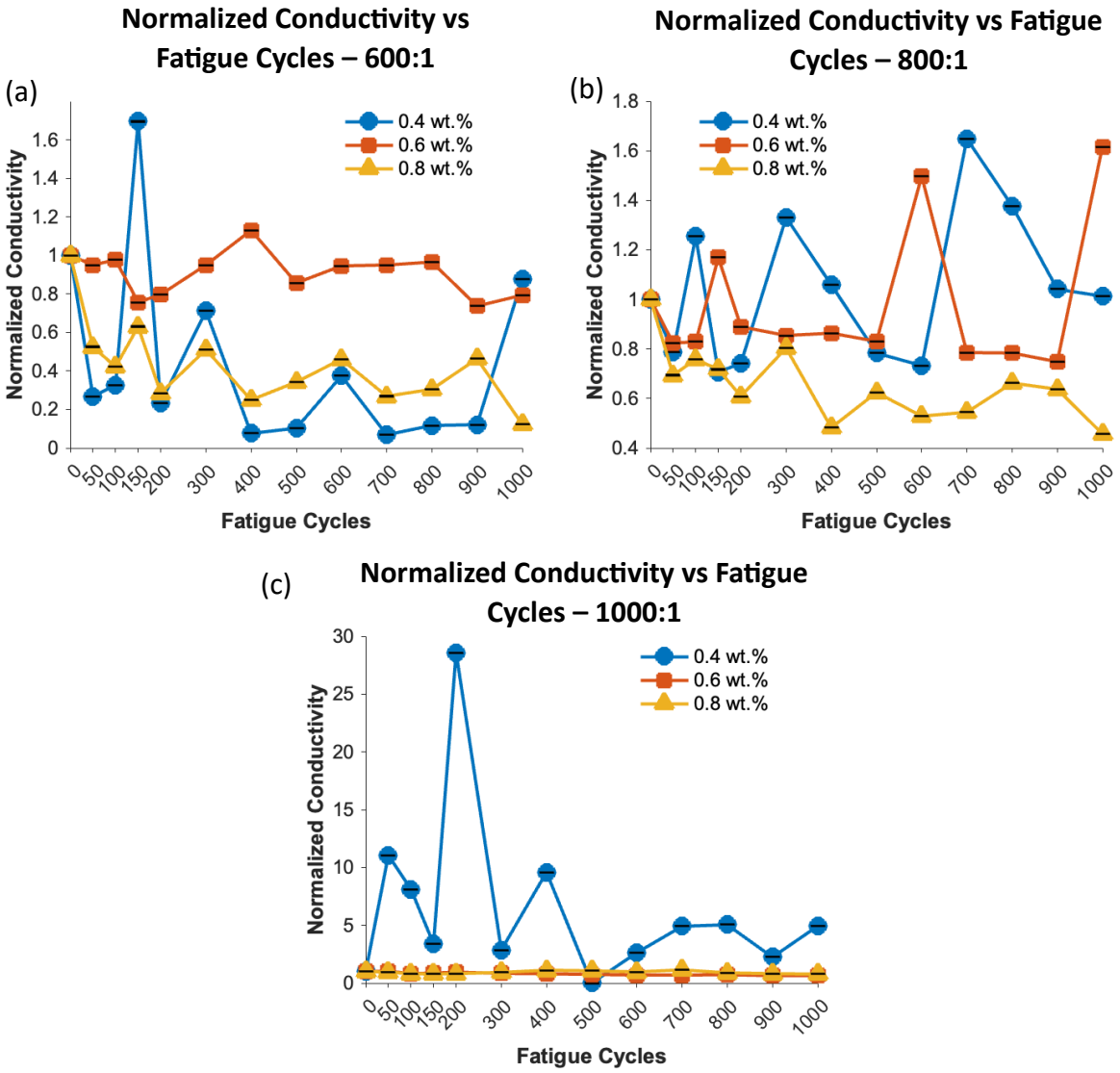


Figure S16. (a-c) Normalized conductivity of CNT/BBE composites over 0-1000 fatigue cycles using the four-point probe. Data are shown for PDMS:CL ratios of 600:1, 800:1 and 1000:1 respectively and CNT loadings of 0.4, 0.6 and 0.8 wt.%

Table S2. Comparison of electrical and mechanical performance of this work with previously reported CNT/PDMS-based and bottlebrush elastomer conductors.

Different studies	Conductivity at 0% strain (S/m)	Conductivity under strain	Strain survivability	Gauge factor	Retention after N cycles
This work (measurements of one sample with the balanced conductivity and signal stability are provided for brevity)	Four-point: 0.013 S/m (600:1, 0.8 wt.%) Two-point: 12.75 S/m (600:1, 0.8 wt.%)	Four-point: 0.013 S/m (600:1, 0.8 wt.%) Two-point: 6.74 S/m (600:1, 0.8 wt.%)	Bending to 90°	Not measured	~90% after 1000 cycles - 800:1, 0.8 wt.%
P. Xu et al., [11]	2.68 -13.78 S/m (0.2 - 0.6 wt.%) (Two-point method)	Retains conductivity	2,500 cycles at 100% strain	Decreases as SWCNT loading rises	Retained after 2,500 cycles
J. Shao et al., [34]	0.038 S/m (Two-point method)	Constant up to 160% strain – rise before fracture	Elongated up to 200%	Not reported	Drops slightly after 1000 cycles but stabilizes up to 5000 cycles
J. Zeng et al., [35]	30 S/m (Two-point method)	Decrease in conductivity as fiber is stretched	Ultra-wide sensing range – 471%	368	200 cycles at 100% strain

Our conductivity measurements demonstrate good agreement with previously reported values for PDMS- and bottlebrush-based elastomer composites [11, 34, 35]. Specifically, the two-point probe measurements for the 600:1, 0.8 wt.% composite (~12.75 S/m) fall within the range reported in the literature (2.68–13.78 S/m) using the similar two-point method, which confirms the validity of our approach for comparison with prior studies. As expected, the four-point probe measurement for the same sample (0.013 S/m) is lower due to elimination of contact resistance, but it provides more accurate absolute conductivity values. Fatigue testing also shows trends consistent with literature reports, which performed 200–5000 bending cycles; our composites maintained similar relative performance despite applying larger bending angles. These results collectively indicate that our composites achieve comparable electromechanical stability and conductivity to previously reported materials while demonstrating robust performance under more extreme mechanical deformation.