

## **Supporting Information**

### **Measuring adhesion of the Interfaces of Carbon Nanotube Bundles and Electrospun Polymer Fibers**

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More details of the nano-cheese-cutter are shown in Figure S1. Using a high resolution micro-manipulator (AutoMate Scientific) combined with a stereomicroscope (Olympus, BX51), two identical glass microspheres with a diameter of 30–50  $\mu\text{m}$  (Potters Industries) were attached to the free end of an AFM cantilever using epoxy (AeroMarine TM 400) with a designated separation of 50–120  $\mu\text{m}$  (Fig. 1(a)). The epoxy was then cured after 2 h at 90°C. Small amount of epoxy was put on top of the particles before a sample fiber was attached. Despite our strenuous efforts, the sample fiber was not perfectly leveled inevitably. Note that when the cantilever was lowered, it subtended a small but nonzero angle to the leveled fiber on mica because of the AFM configuration. Similar method was used to suspend a freestanding fiber sample on the mica surface.

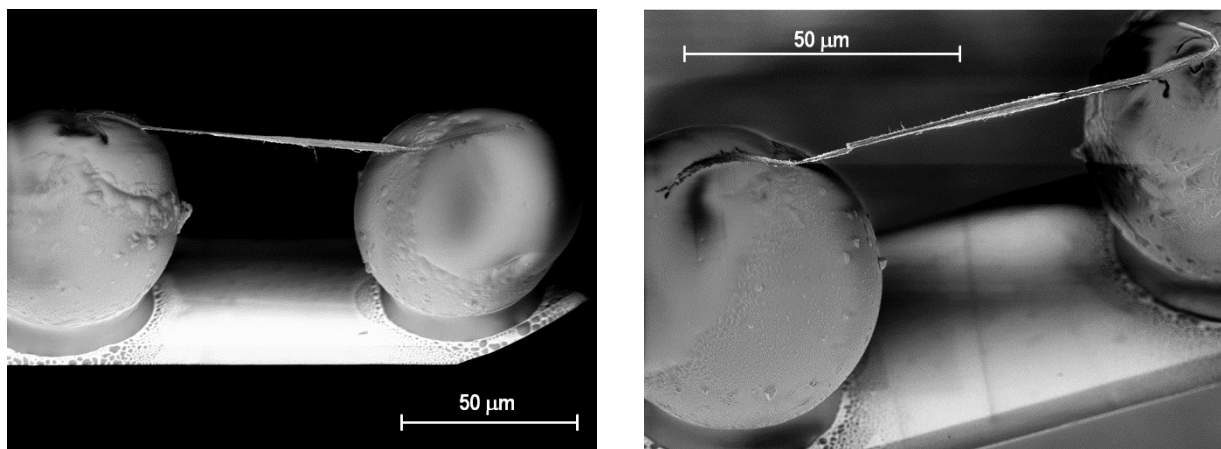


Figure S1. A SWCNT bundle attached to two microspheres at the end of a planar AFM cantilever.

In the presence of an external compression applied to the AFM cantilever, the cheese-cutter deformed into an inverted V-shape, while the lower fiber turned into V-shape geometry as shown in Figure S3. Since the fibers were relatively thick, central deformation at the contact was relatively small, resulting in rod bending instead of string stretching. The force displacement was measured to be linear as expected.

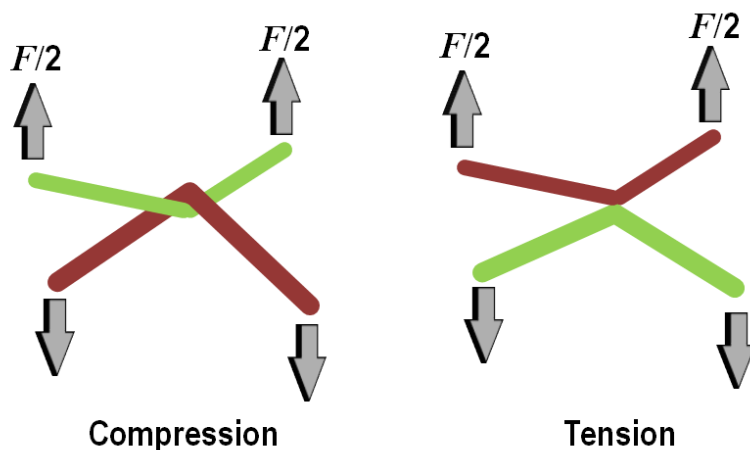


Figure S2. Deformation of the fiber upon loading (compression) and unloading (tensile). At pull-off, the inter-fiber contact area is small.

After several loading-unloading cycles, surface damage accumulated leading to loosened CNTs sticking out of the sample fiber as shown in Figure S3. At large loading displacement, the Espun fiber remained intact, but the CNT bundle broke. Figure S4 show different magnification of the broken fiber. In all our samples, the CNT bundles were always first to break.

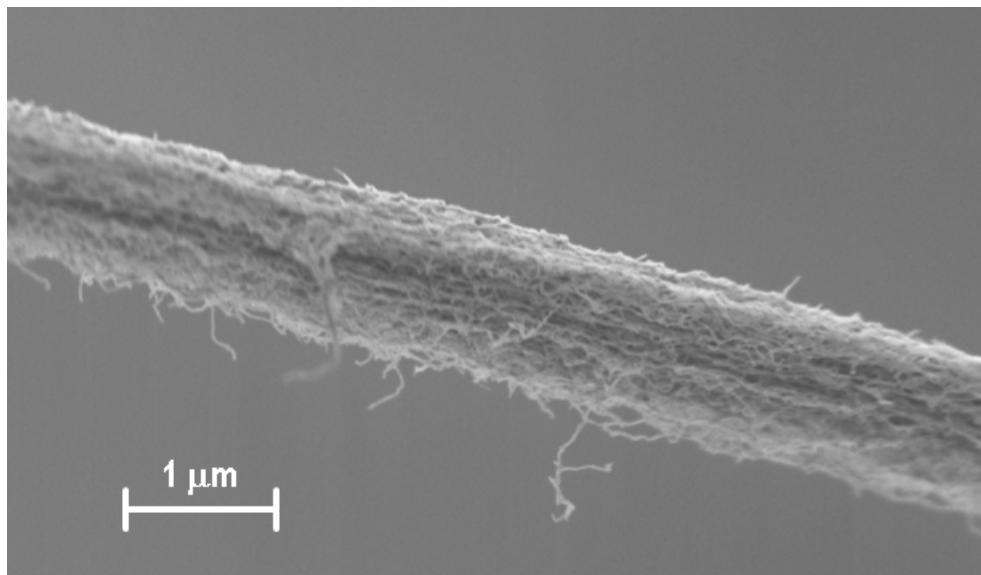


Figure S3. Roughened surface of a CNT bundle after several loading-unloading cycles with an Espun fiber.

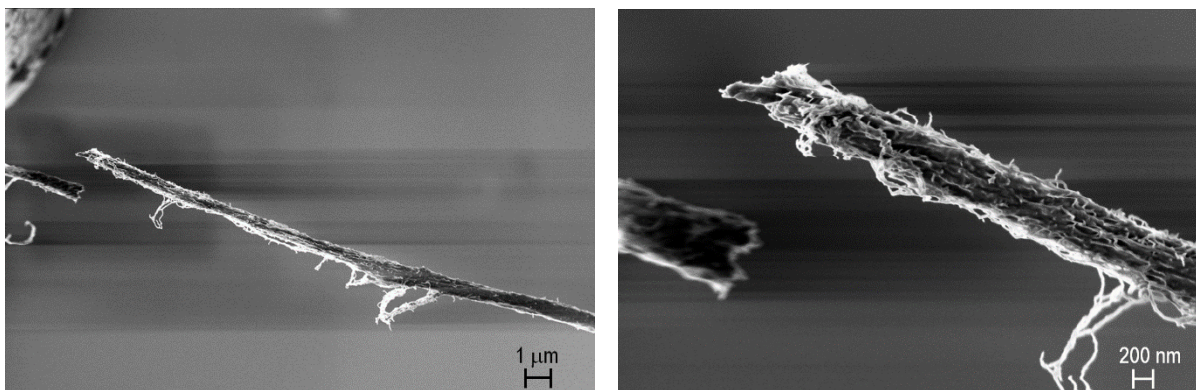


Figure S4. Broken CNT fiber at the contact at different magnifications.

In CNT-CNT measurement (Figure S5), the kink only occurred during unloading in some samples. If it happened, the kink was observed at roughly the same location in the same sample pair. It was likely the result of surface slippage, and did not lead to significant loading-unloading hysteresis. The “pull-off” load varied from measurement to measurement, but fell in the same range. Figure S5 shows several loading-unloading cycles on the same sample pair.

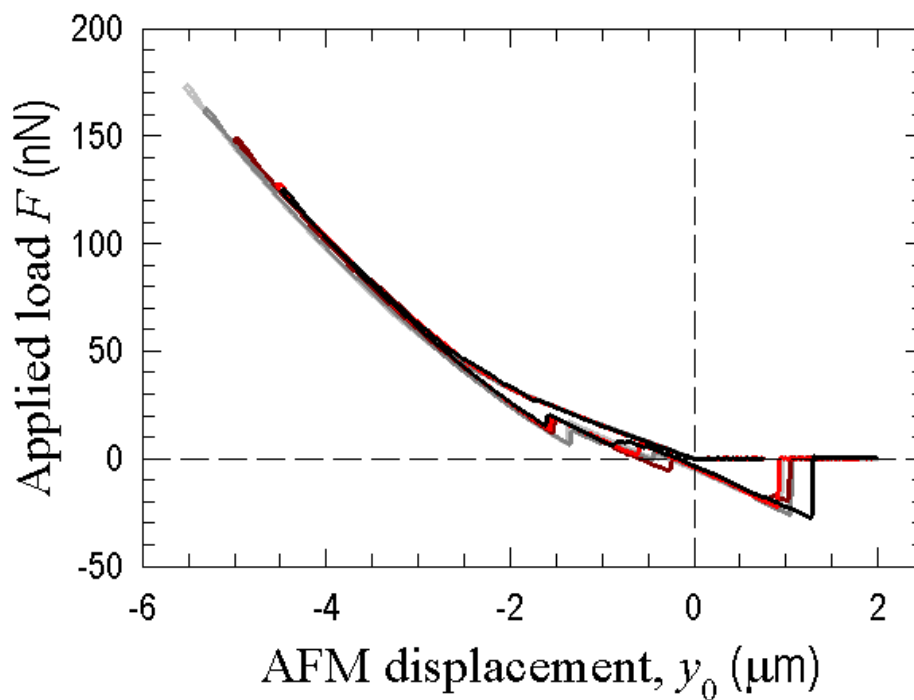


Figure S5. Roughened surface of a CNT bundle after several loading-unloading cycles with an Espun fiber.