Supporting Information

Alignment of Carbon Nanotubes in Carbon Nanotube Fibers Through Nanoparticles: A Route for Controlling Mechanical and Electrical Properties

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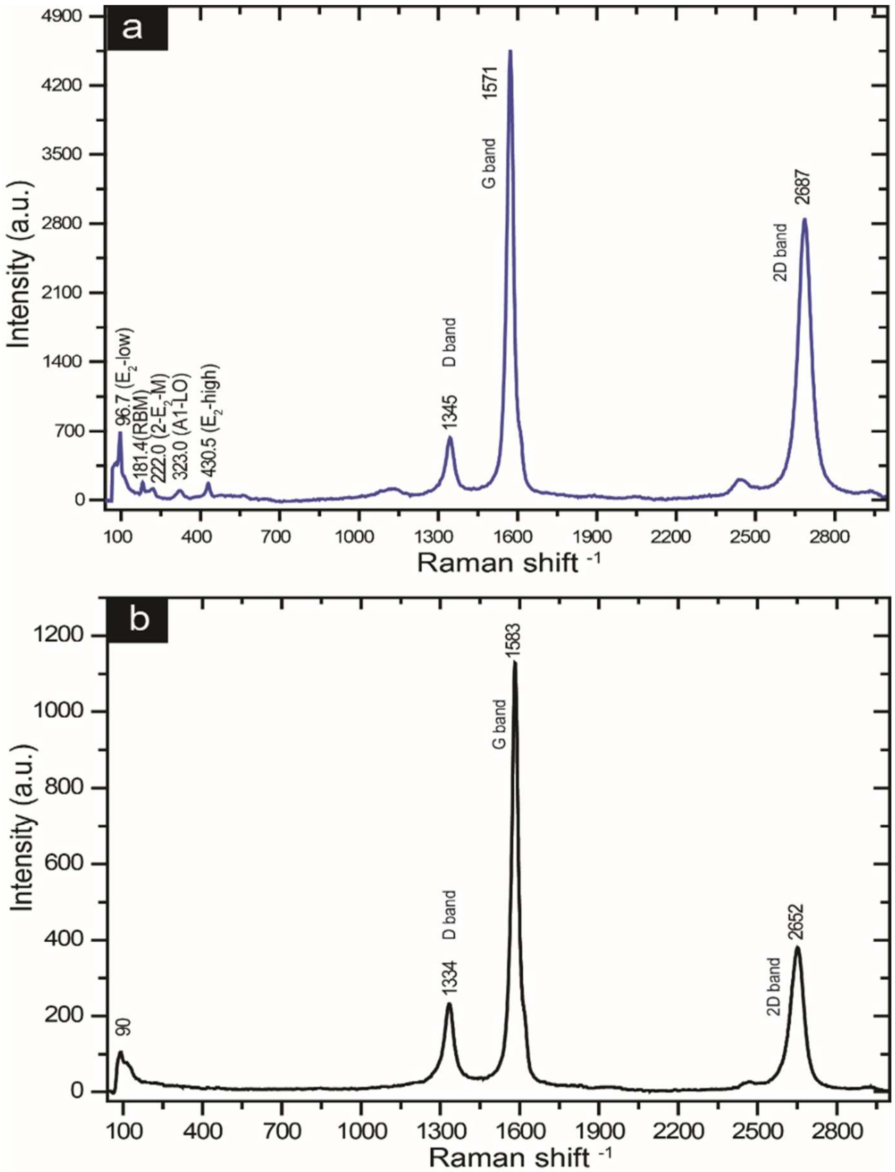

Figure S1. Raman spectra of (a) aligned CNT fiber and (b) bare CNT fiber.

Raman Spectra

Raman spectra (Figure S1) of aligned and bare CNT fibers were taken to monitor ZnO nanoparticles, CNTs, and their interactions in the aligned CNT fiber. The presence of D-bands in the bare CNT fiber (Figure S1a, b) indicates the structural defects in the CNT fiber, recognized as oxygen-containing functional groups inside the aligned and bare CNT fibers. New peaks were also detected at 96.7, 222.0, 323.0, and 430.5 cm^{-1} (Figure S1a) for the case of ZnO nanoparticles in aligned CNT fibers. On the other hand, the bare CNT fiber (Figure S1b) did not display any peaks in this region. Thus, these new peaks indicate that the ZnO nanoparticles are available inside the aligned CNT fiber. The peaks for ZnO nanoparticles at 96.7, 323.0, and 430.5 cm^{-1} are associated with the E_2 (low)[1], A1-LO (longitudinal optical),[1, 2] and E_2 (high)[1] phonon peaks of ZnO. Of note, the radial breathing mode (RBM) peak was observed at 181.4 cm^{-1}, indicating that the radial expansion and contraction of carbon atoms of CNTs inside the aligned CNT fiber arises from this vibration. Consequently, the radial vibration of Zn atoms in –O-Zn-C or C-Zn-C systems in the aligned CNT occurred in presence of a 514 nm laser for radial polarization. Figure S1(a, b) of the aligned CNT and bare CNT fiber shows the D band and 2D band, which are shifted 11 and 35 cm^{-1} respectively, to the higher frequency direction in aligned CNT fiber. On the other hand, G band, which are shifted to the lower frequency position in aligned CNT fiber. The peak shift specifies that a strong interaction between ZnO and the CNT fiber is available. The incidence of 1D, G, 2D band, E_2 (high), and a high intensity RBM peak in the aligned CNT fiber suggest chemical bonding between the CNT and ZnO nanoparticles in the aligned fiber. Since bare CNT fibers (Figure S1b) show a D band, some oxygen-containing functional groups (–OH, –COOH-, C=O) must be present which create defects in the graphene lattice of the CNTs. Additionally, the aligned CNT fiber also showed a D band and of higher intensity. This suggests a new chemical reaction due to the increased functionalization.

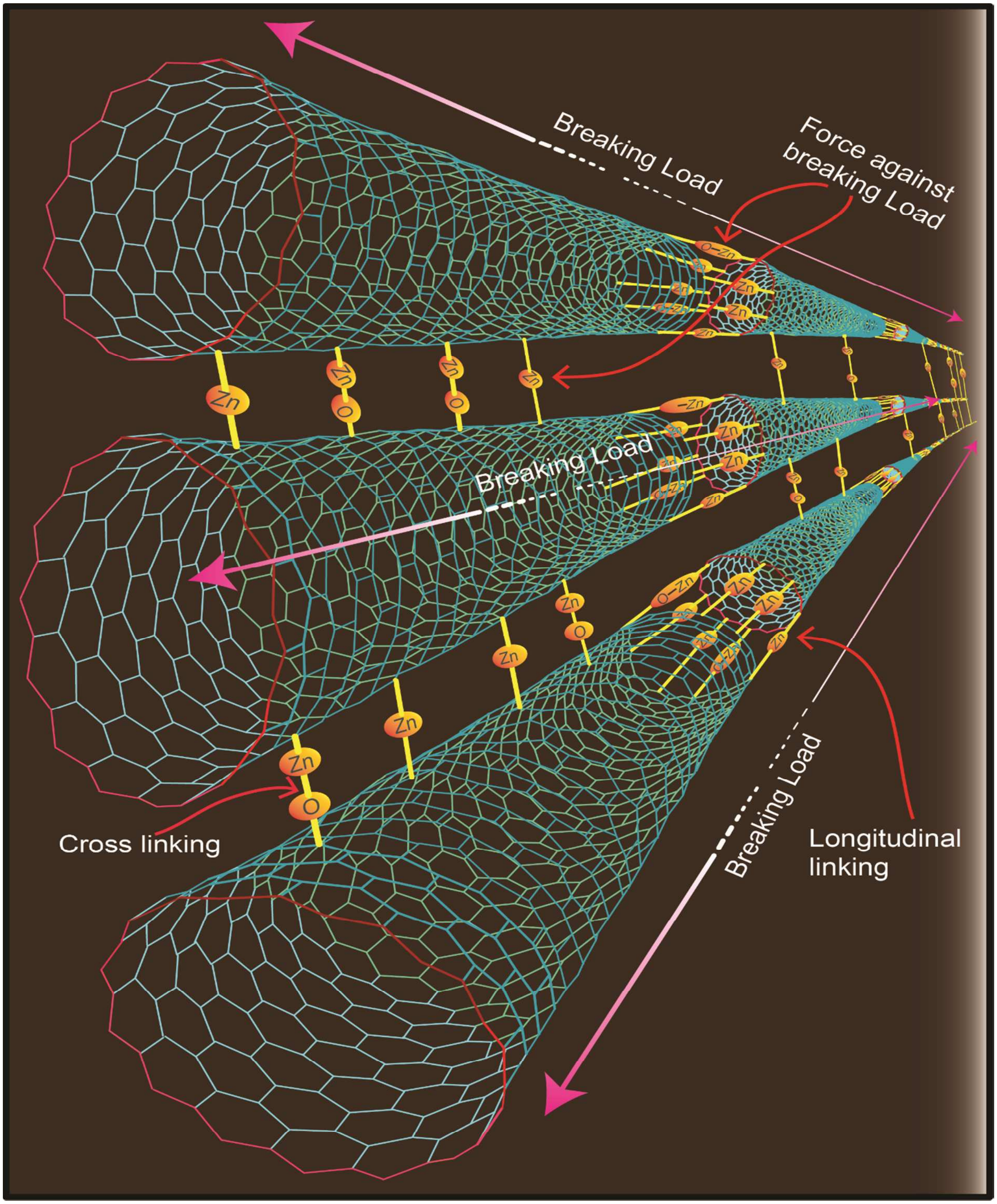

Figure S2. Inner architecture of aligned CNT fiber and its relation to mechanical breaking load.

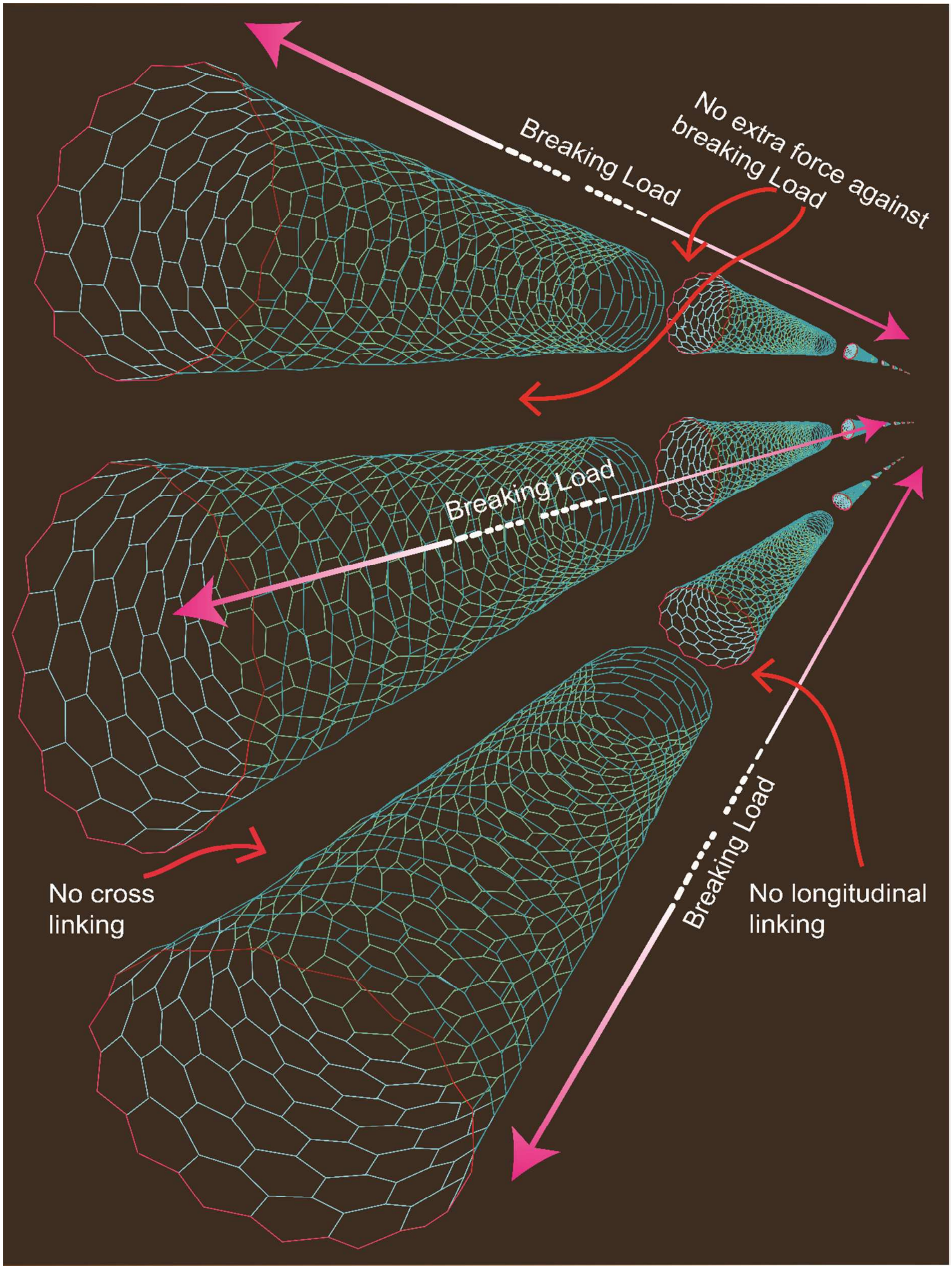

Figure S3. Bare CNT fiber inside architecture.

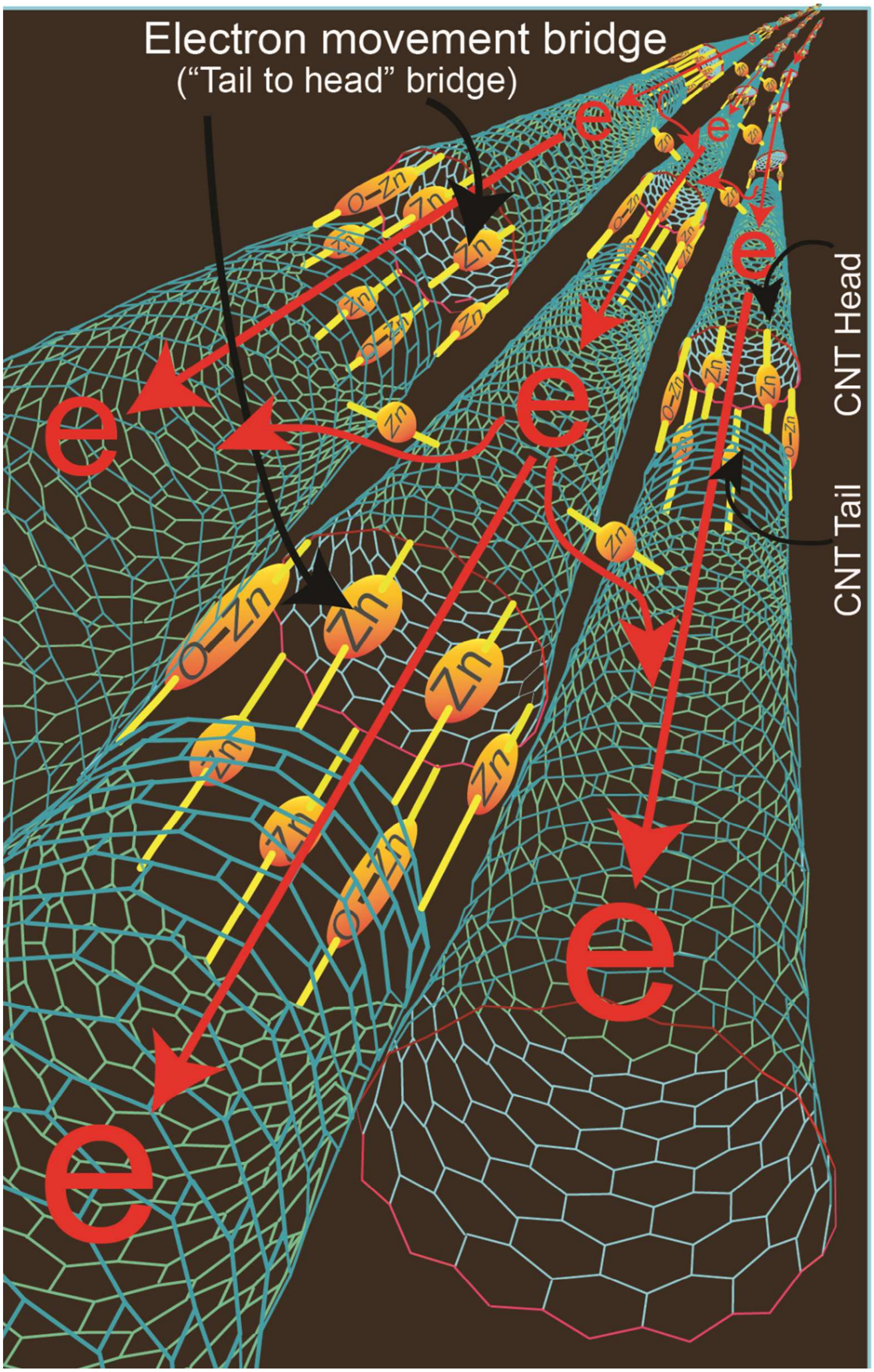

Figure S4. Inner architecture of aligned CNT fibers for improved electrical conductivity due to cross and longi-tudinal linking groups.

Reference

1. Bairamov, B. H.; Heinrich, A.; Irmer, G.; Toporov, V. V.; Ziegler, E. Raman Study of the Phonon Halfwidths and the Phonon-Plasmon Coupling in ZnO. *Physica Status Solidi B* **1983**, *119*, 227-234.
2. Son, D. I.; Kwon, B. W.; Park, D. H.; Seo, W. S.; Yi, Y.; Angadi, B.; Lee, C. L.; Choi, W. K. Emissive ZnO-graphene Quantum Dots for White-Light-Emitting Diodes. *Nat. Nanotechnol.* **2012**, *7*, 465-471.