Supporting Information

C_{60}/Collapsed Carbon Nanotube Hybrids - A Variant of Peapods

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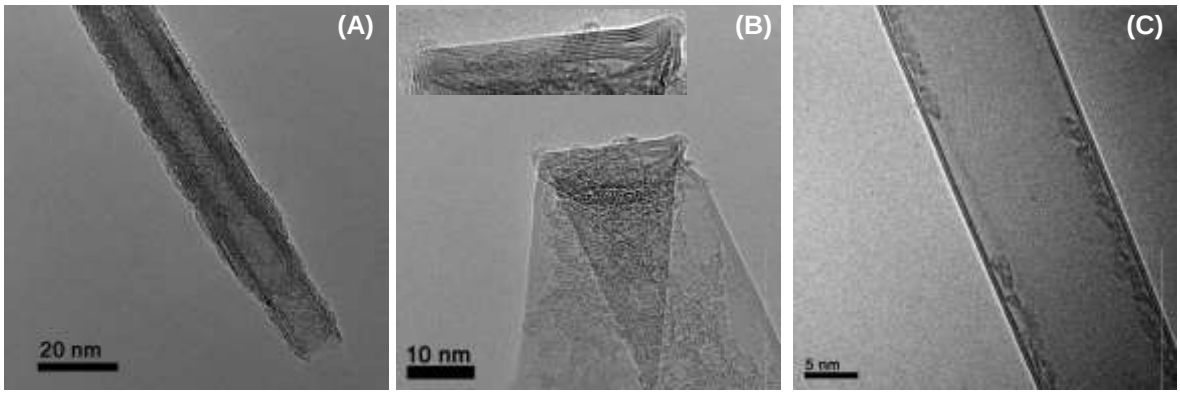

Figure S1. TEM image of (A) a typical MWCNT after its cap has been removed by thermal oxidation (B) A four-walled CCNT which is bent at the middle where it is possible to see its cross section consisting of eight graphene layers (inset). The teardrop shape of the duct is clear in the image. The width of the collapsed tube (21 nm) is the same as the bend region, which implies that the tube is completely collapsed, (C) a double-walled CCNT with a width of 14 nm, much above the theoretically calculated critical diameter for a double walled tube.[1] The image contrast at the edge duct regions is due to solvent contamination during the extraction process.

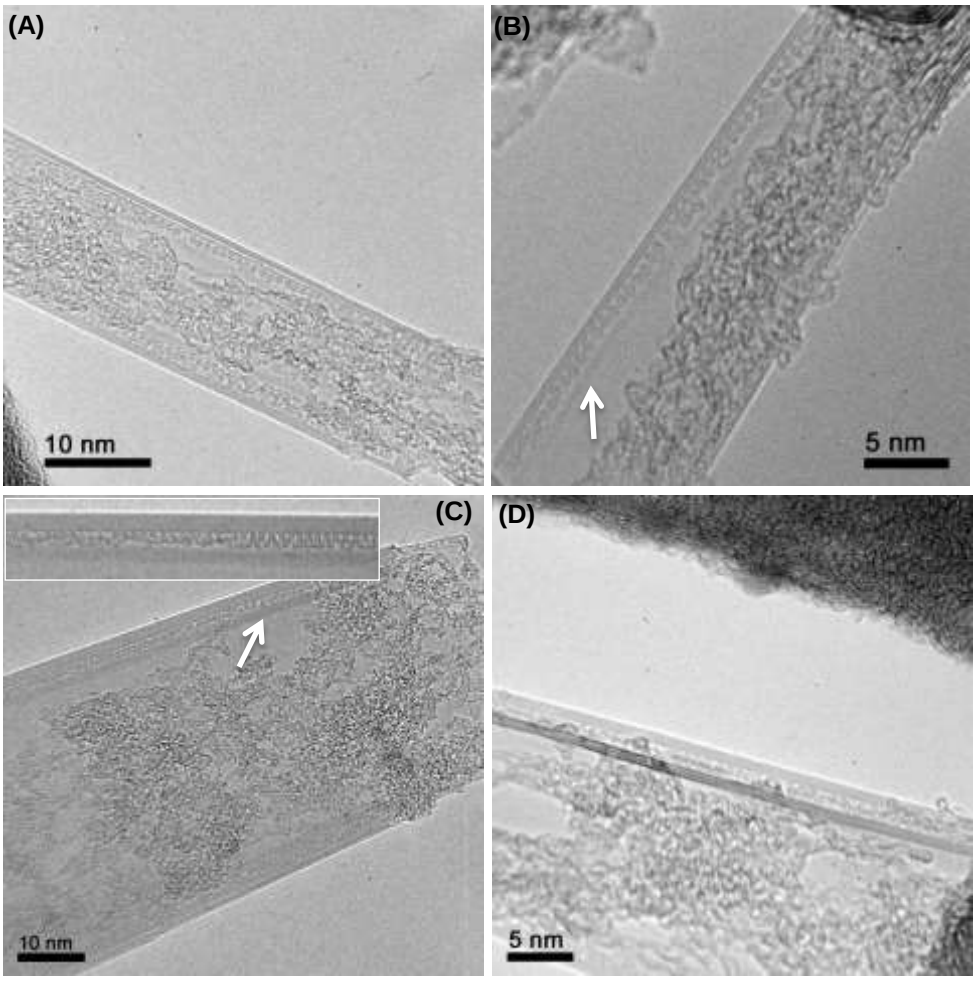

Figure S2. Low magnification TEM image of C_{60}/CCNTs synthesized by solution process; (A) and (C) show the structural damage at the end of the C_{60}/CCNTs where due to cap removal the structures suffers from dangling bonds. The C_{60}/CCNTs in image (B) and (C) have small angle with the image plane, causing an image contrast parallel to the CCNT walls (close to the C_{60} molecules, indicated by arrows in the image). This is due to the graphene layers surrounding the C_{60} molecules which now have higher interaction with the electron beam. The inset in figure (C) is higher magnification image of the edge of the C_{60}/CCNT indicating overlap of C_{60} molecules (due to tilting) as well as both linear and staggered C_{60} configuration along the duct. (D) TEM

image of a duct of twisted C_{60}/CCNT where (due to twisting) the graphene layers at both side of the C_{60} molecules are parallel to electron beam and cause strong image contrast.

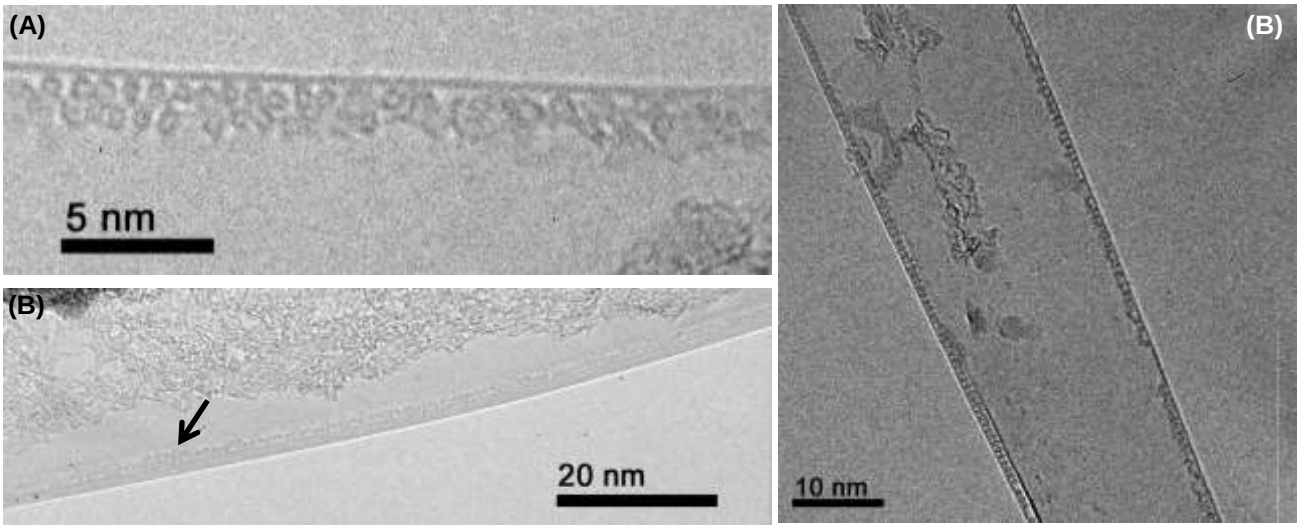

Figure S3. (A) TEM image of staggered C_{60} configuration along the duct of a single-walled CCNT, revealing that such a configuration is independent of number of walls. The C_{60} molecules on the right side of the image are fused to each other due to high energy electrons during imaging. (B) Lower magnification TEM image of C_{60}/CCNT, indicating that the duct of CCNT can be completely filled by C_{60} molecules all along its length. The C_{60} molecules are mainly in linear chain form however in some region they staggered configuration can also be observed (indicated by arrow in the image), which suggest that the C_{60} configuration in the duct is independent of number of walls. (C) single-walled CCNT filled with C_{60} on both edges except small gap between C_{60}s on right edge.

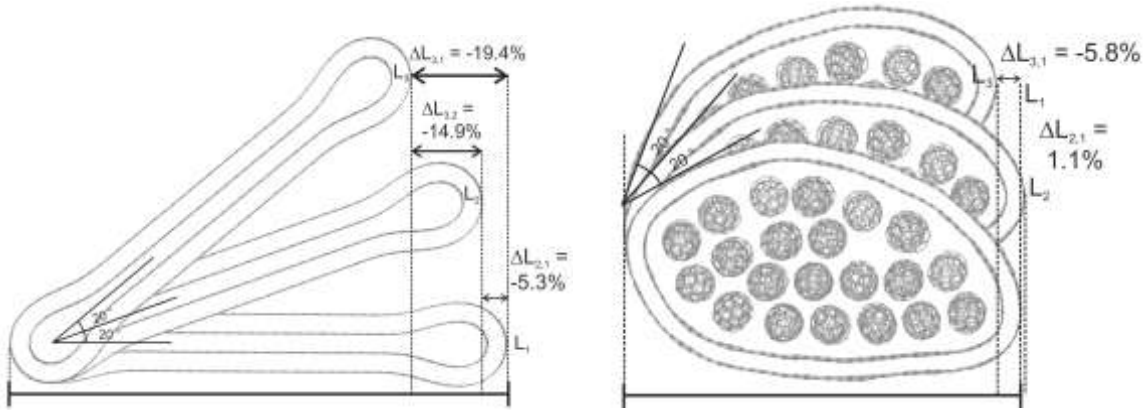

Figure S4. Simulated model of how the projected diameter of a flat CCNT (left), and a more than half-full C_{60}/CCNT changes by tilting the tube 20° around the tube axis.

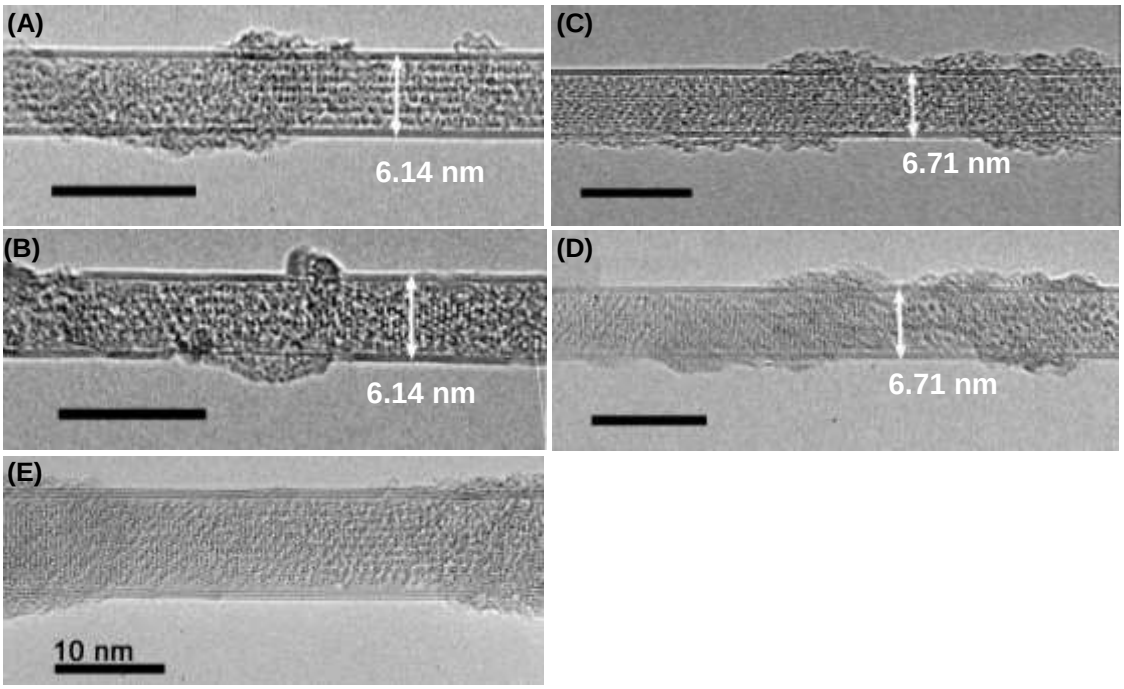

Figure S5. TEM images of a double-walled tube reinflated by C_{60} molecules; (A)&(C) shows the C_{60}/CCNT before, while (B)&(D) shows the same tubes after 20° and 10° tilting around the tube axis respectively. The diameter of the tube remain unchanged after tilting indicating that the tube is fully or close to fully reinflated by C_{60}. The scale bars in (A) to (D) are 10 nm. (E) TEM image of a three walled tube with 10 nm diameter; it is theoretically predicted that such a tube is unstable in tubular form.

Table S1. Theoretically and experimentally determined width ***L*** and inner height ***d*** of C_{60}-stagger-filled CCNT of wall number n.

CCNT/C_{60} n	Theoretical calculation		Experiment	
	L **(nm)**	***d*** **(nm)**	***L*** **(nm)**	***d*** **(nm)**
n=1	2.66	1.39	-	1.20
n=2	3.29	1.43	-	-
n=3	3.19	1.40	-	1.25
n=4	3.23	1.38	-	-

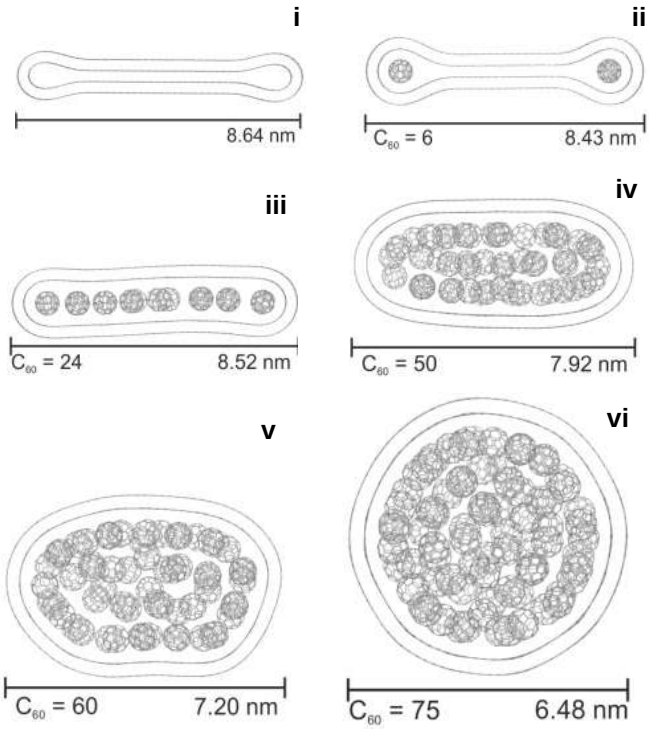

Figure S6. The starting configuration for the result presented in figure 3C.

1. Benedict, L. X.; Chopra, N. G.; Cohen, M. L.; Zettl, A.; Louie, S. G.; Crespi, V. H. *Chem Phys Lett* **1998,** 286, (5-6), 490-496.