Supplementary Information for

Top-down fabrication of small carbon nanotubes

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Supplementary Figures: Fig. S1-S8

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1. Supplementary Figures

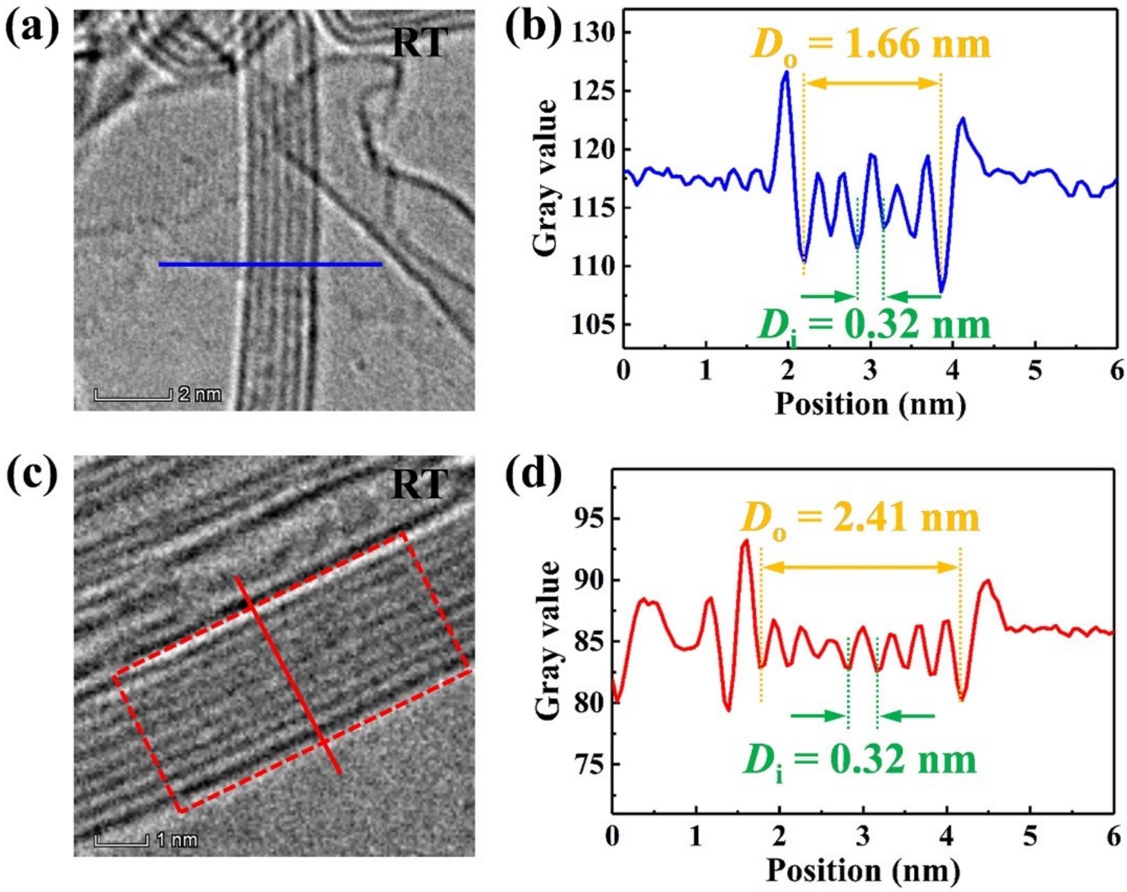

Fig. S1 Smallest triple-walled and four-walled carbon nanotubes obtained by the self-contracting process via irradiation/annealing. HRTEM images of the CNTs acquired at room temperature and the corresponding image intensity profiles for triple-(a-b) and four-walled CNTs (c-d) respectively.

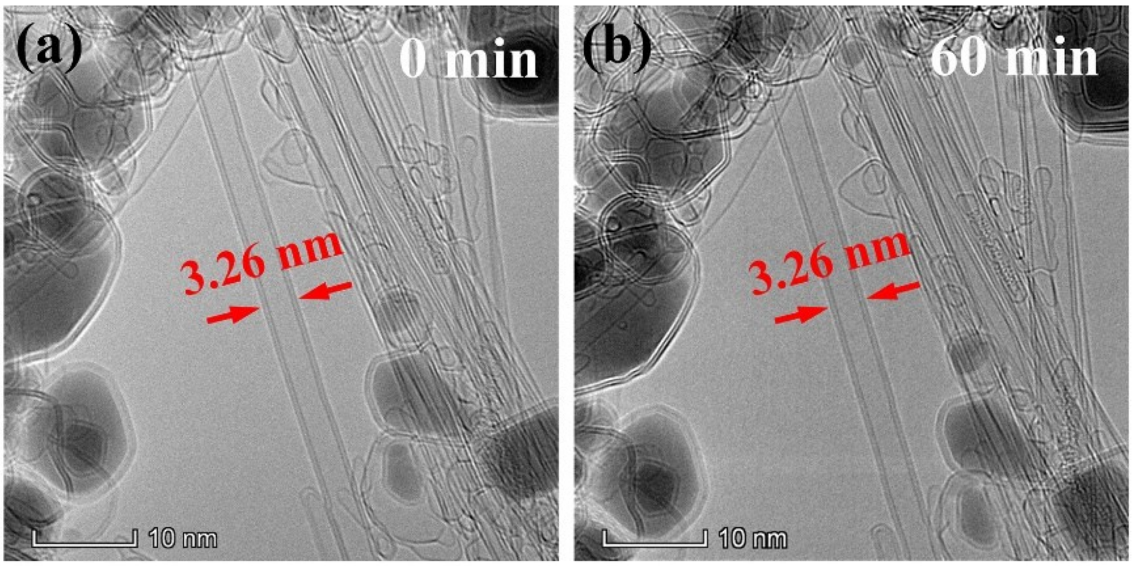

Fig. S2 DWNTs annealed at 1200 ºC without electron irradiation for 1 hour. (a) HRTEM image of initial DWNTs. (b) HRTEM image of DWNTs after one hour annealing. Without electron irradiation, the sole high temperature annealing does not bring any visible structural change to the DWNTs, which confirm the key role of the irradiation that indeed dominates the DWNT shrinking process.

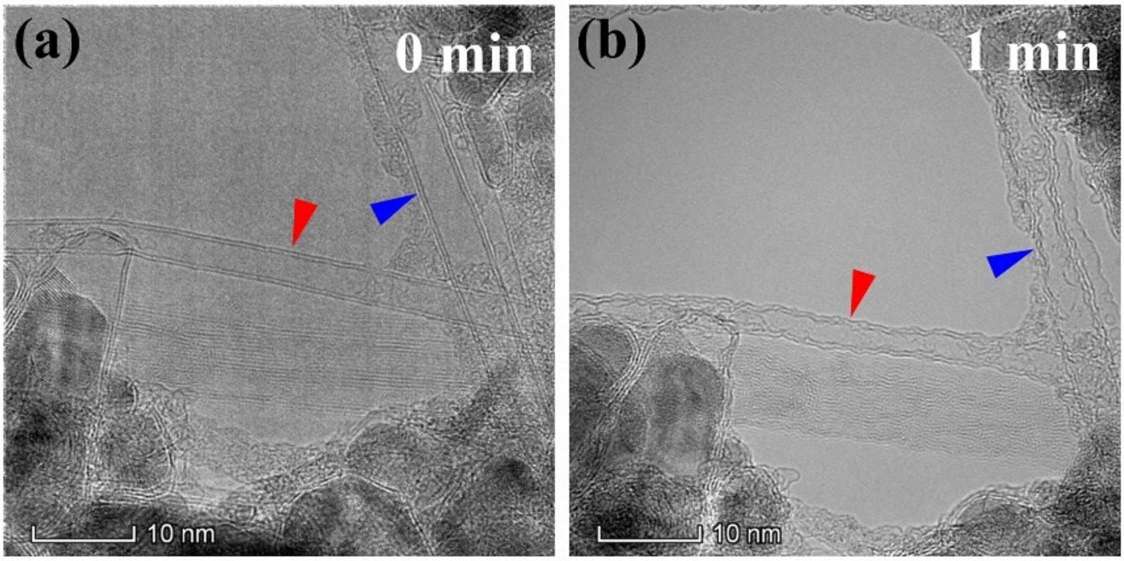

Fig. S3 DWNTs irradiated under 20 A/cm^2 at room temperature. HRTEM images of the DWNTs before (a) and after (b) 1 min's irradiation. The room-temperature irradiation lead the DWNTs to be disordered and amorphized, thus highlighting the importance of the concurrent annealing for maintaining the tube perfection.

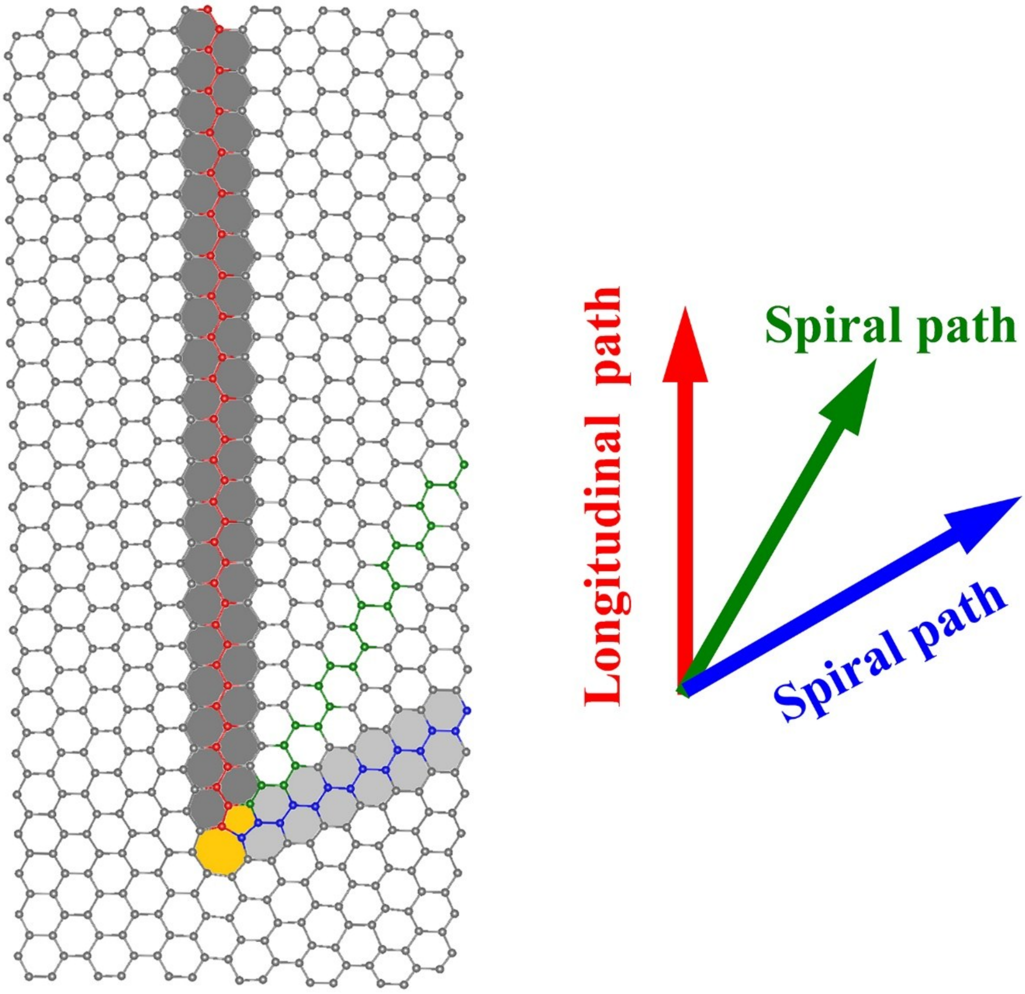

Fig. S4 Climb paths of 5|7 dislocation in an armchair CNT. An armchair nanotube theoretically allows both longitudinal and spiral kink motions. In a longitudinal path, the 5|7 dislocation climbs due to the sequential departure of the C_2 dimers marked as red circles along a zigzag chain. In the spiral paths, the 5|7 dislocation moves along another zigzag chain (in blue) or an armchair chain (green) as the result of the removal of the C_2 dimers in the tube lattice.

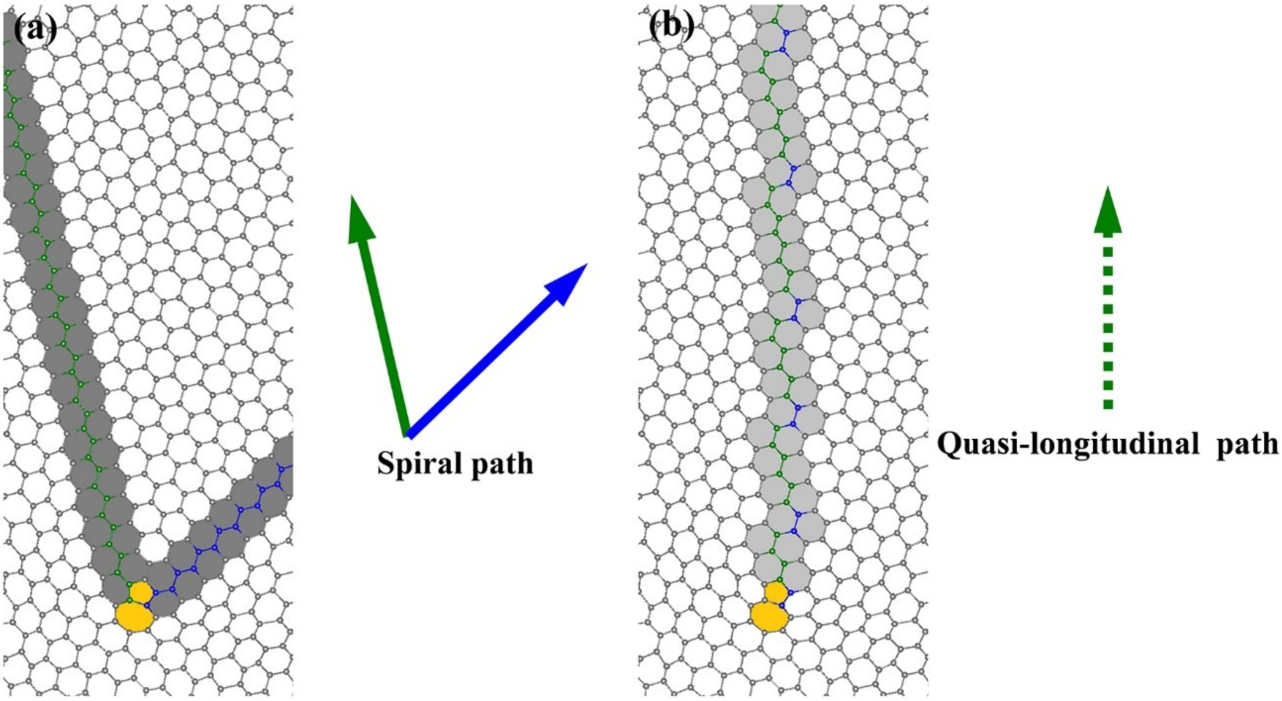

Fig. S5 Climb paths of 5|7 dislocation in a chiral CNT. (a) Two spiral paths. Due to the sequential removal of the C_2 dimers, the 5|7 dislocation can climb along two zigzag chains, marked in green and blue, resulting in the kink motion along the left-handed and right-handed spiral paths, respectively. (b) Quasi-longitudinal path. This is actually a combined path as the result of the mixture of left- and right-handed spiral paths at the atomic scale, leading the kink to move roughly along the tube axial direction.

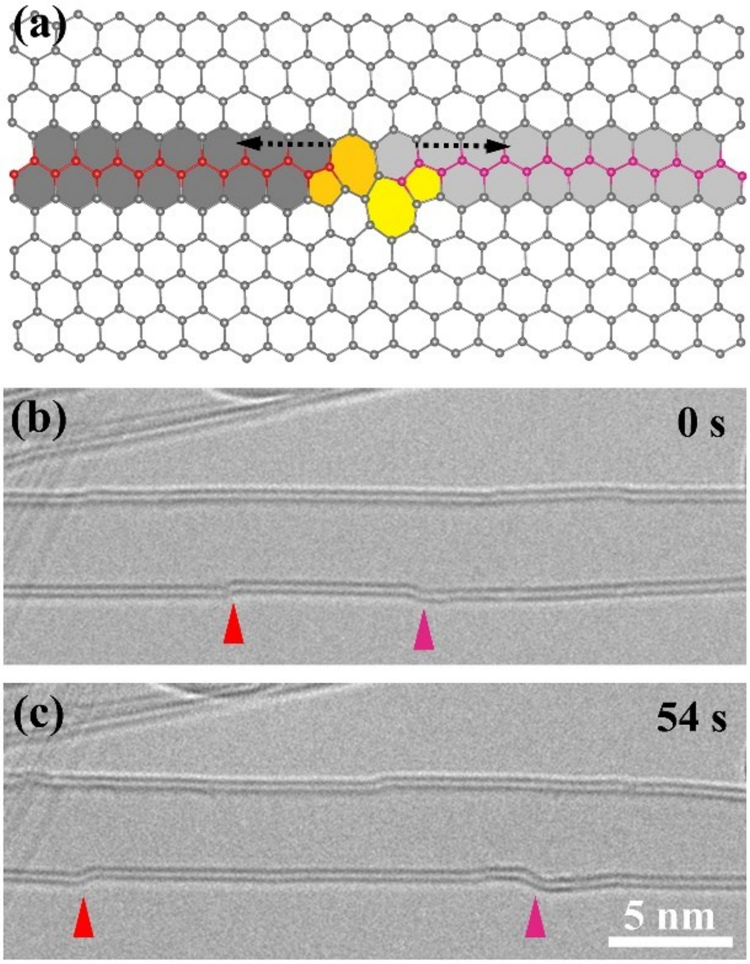

Fig. S6 A pair of head-to-head edge dislocations and their movements in the opposite directions simultaneously along the nanotube. (a) Schematic illustration of formation of two opposite 5|7 dislocations that can be generated from a di-vacancy through bond reconstructions, as described in reference 1. (b-c) HRTEM images showing the real process of two kinks moving in the opposite directions.

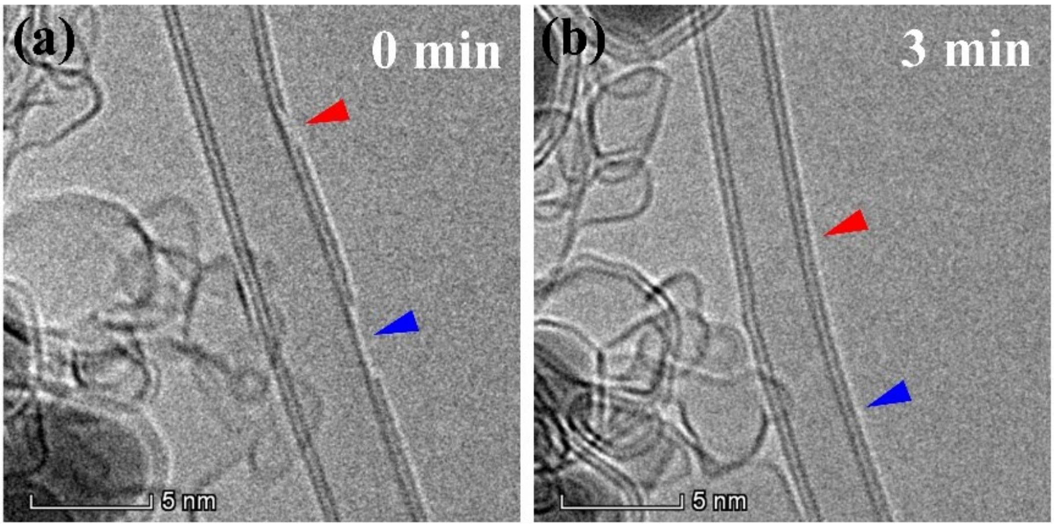

Fig. S7 Hole repairing in a DWNT during the self-contracting process. (a) Initial DWNT with holes in the tube walls, mark with red and blue arrows respectively. (b) After irradiation/annealing, the DWNT shrinks in the radial direction, companied with the mending of the holes that shrink in size and disappear completely when interacting with the propagating kinks, and the tube walls thus become continuous and straight.

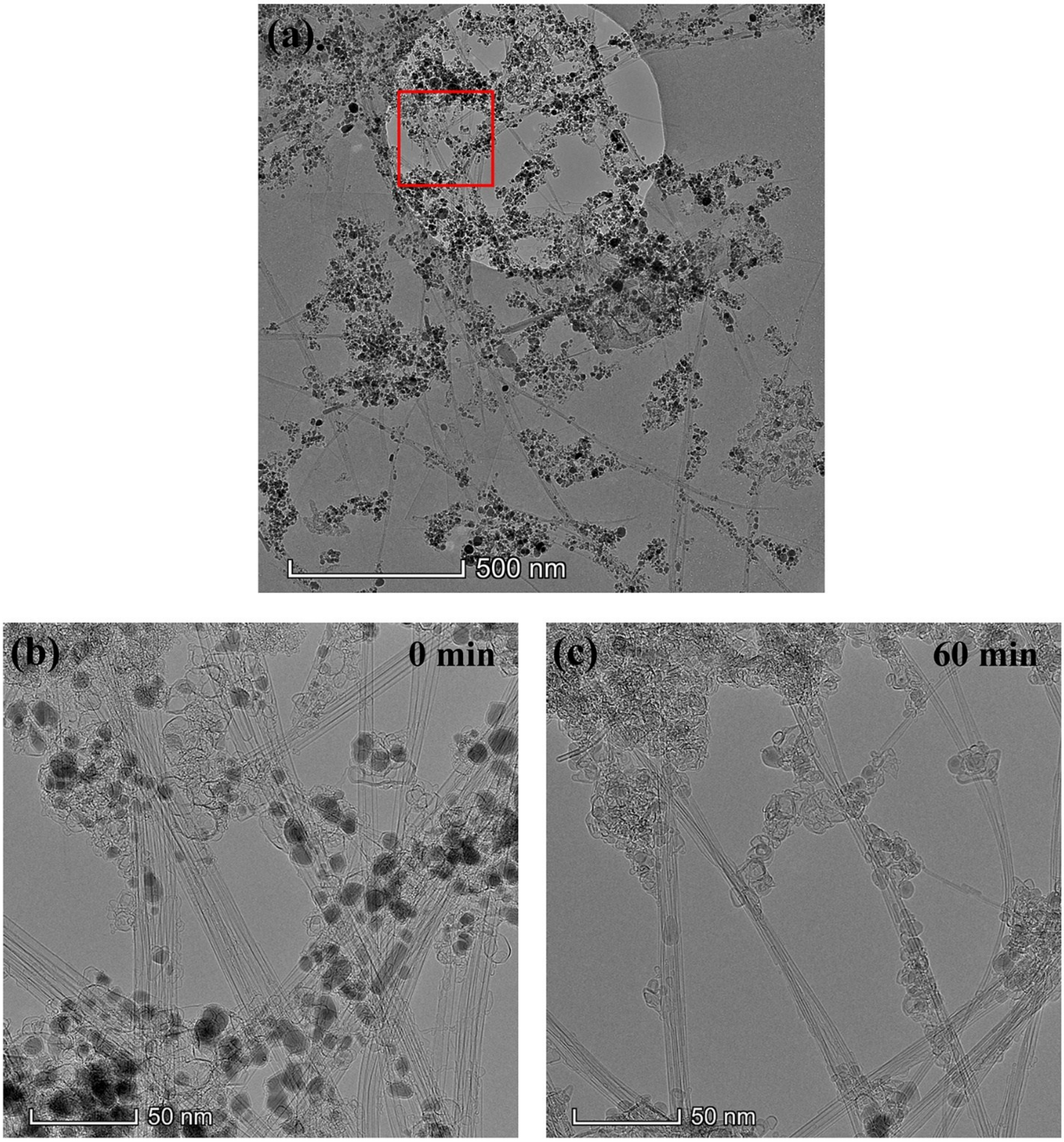

Fig. S8 Batch processing of a large number of DWNTs by irradiation/annealing. (a) TEM image of the prestine DWNTs lying on the SiN film of a heating chip (about one thousand in total that exposed to the irradiation of a broad e-beam). (b) The enlarged TEM image of the area marked with a red box in (a), showing the morphologies of the DWNTs before irradiation, (c) and their final morphologies after an hour of irradiation at 1200 ºC.

2. Supplementary Movies

Movie S1.mp4

An in-situ TEM movie showing the structural evolution of a DWNT that shrinks in a seamless and uniform fashion. The movie was recorded at a rate of 5 frames/s, and is played at 16× speed.

Movie S2.mp4

An in-situ TEM movie showing the breakdown of inner tube of a DWNT at its diameter limit. The inner tube at this stage becomes structurally unstable, and it then breaks and shrinks in length upon further irradiation at elevated temperature. The movie was recorded at a rate of 5 frames/s, and is played at 8× speed.

Movie S3.mp4

An in-situ TEM movie showing the longitudinal kink motion in a DWNT. The red arrows indicate the location of the kink that, however, disappears for a short while due to its changes in the advancing direction. The movie was recorded at a rate of 5 frames/s, and is played at 4× speed.

Movie S4.mp4

An in-situ TEM movie showing the spiral kink motion in a DWNT. The kink appears alternately on the two sidewalls, as marked with red and blue arrows respectively, indicating a spiral path along the tube surface. The movie was recorded at a rate of 5 frames/s, and is played at 8× speed.

Movie S5.mp4

An in-situ TEM movie showing the hole repairing process in a DWNT as the result of the interaction between the vacancy defects and the propagating kinks. The movie was recorded at a rate of 5 frames/s, and is played at 8× speed.

Reference

1. F. Ding, K. Jiao, M. Wu, B. I. Yakobson, *Phys. Rev. Lett.*, 2007, **98**, 075503.