Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary Information

In Situ Mechanical Investigation of Carbon Nanotube-Graphene Junction in Three-Dimensional Carbon Nanostructures

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Section S1: TEM analysis of CNTs from CNTs/graphene nanostructure

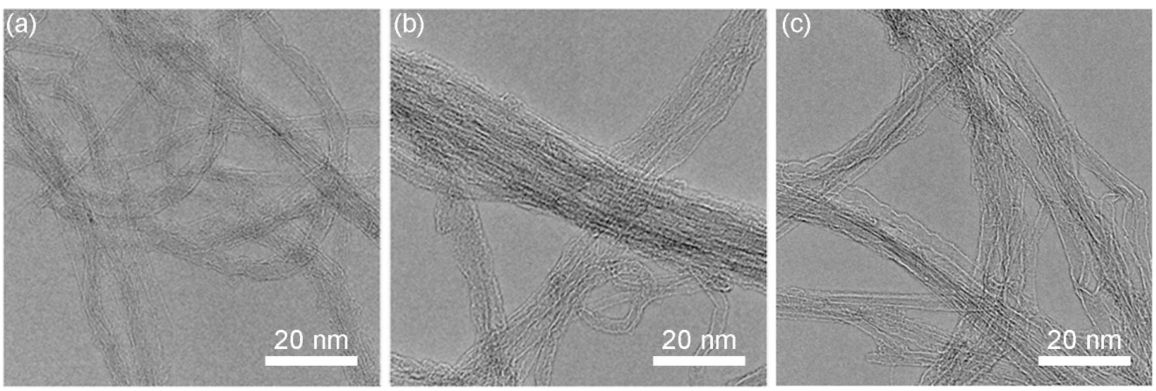

Fig. S1 HRTEM analysis of CNTs from CNTs/graphene nanostructure

Determination of average diameter of CNTs in the CNTs/graphene nanostructures is very important, because it will directly affect the estimation of real contact area and subsequent calculation of junction strength between CNTs and graphene. The TEM images in Fig. S1 show that single walled carbon nanotubes (SWNTs) with small diameter are highly bundled, making it difficult to identify them properly amidst their larger diameter counterparts that remains unbundle.[1] It should be noted that SWNTs are dominant in bundles, where few nanotubes with 2-3 walls can be observed as well. Such result is in agreement with the observation from the previous study.[2] Based on the diameter distribution analysis before, we assume that the average diameter of CNTs in bundle is 3 nm.[1]

Section S2: In-house nanomechanical tester

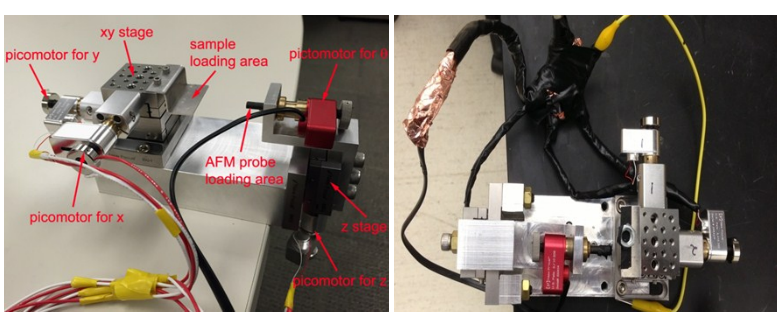

Fig. S2 Configuration of in-house nanomechanical tester

Section S3: Examination of cantilever

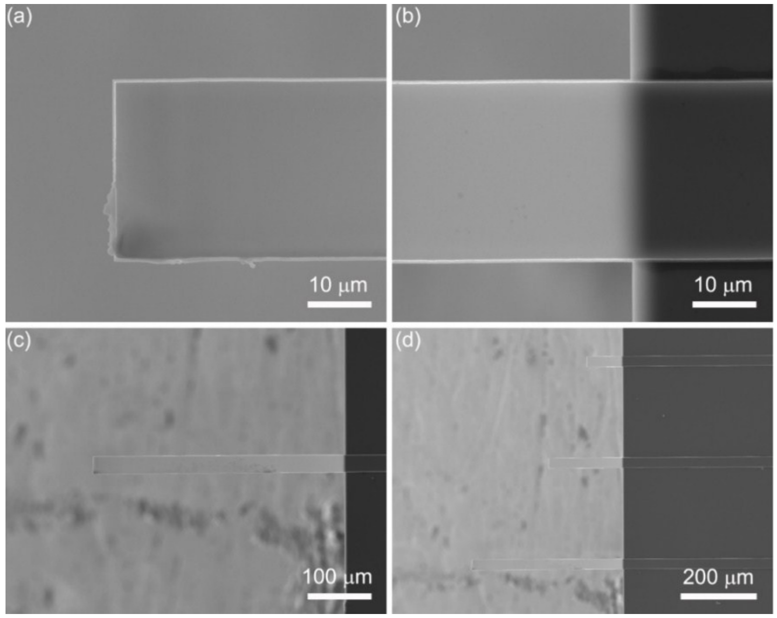

Fig. S3 SEM images of cantilever after debonding test. (a) Front of cantilever. (b) Bottom of cantilever. (c) Entire cantilever. (d) Three cantilevers sitting on one AFM probe.

During debonding test, the cantilever sustained a deflection up to ~165 μm. To see whether there is any crack on cantilever after debonding test, the cantilever was reexamined using SEM. Fig. S3 shows high-magnification SEM images of front and bottom of the cantilever. No crack was found.

Section S4: Pull-out CNT bundles with a stiffer cantilever

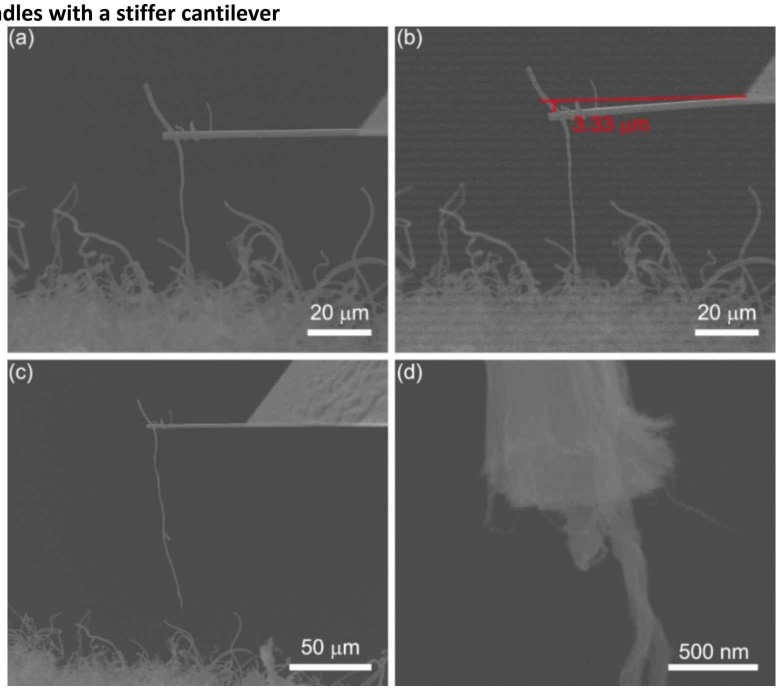

Fig. S4 SEM images of a CNT bundle debonding from graphene using a cantilever with spring constant of 10.6 N/m. (a) The CNT bundle was engaged with an AFM cantilever. (b) The last moment of CNT bundle debonding from graphene. (c) The pulled-out CNT bundle. (d) High-magnification SEM image of the CNT bundle root.

Section S5: Pull-out of a standing-along CNT bundles from graphene

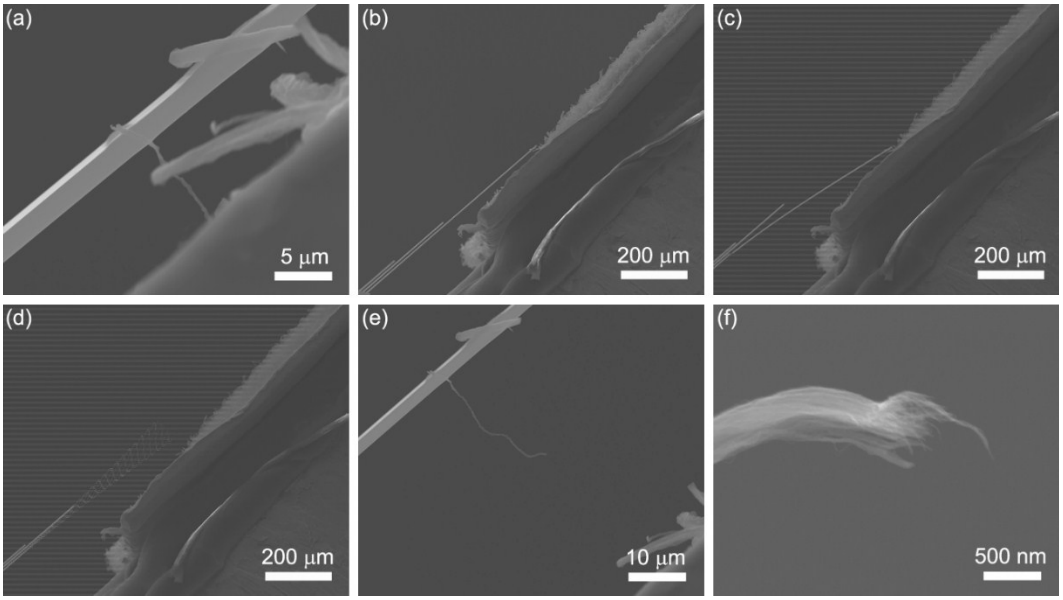

Fig. S5 Snapshots of a standing-alone CNT bundle debonding from graphene.

To see whether the entanglement at the bottom of CNT bundles where one is pulled out would bring error to the junction strength, a standing-along CNT bundle was pulled out from graphene. The measured junction strength is 2.24 GPa, which is in agreement with junction values listed in Table 1. Therefore, the entanglement does not affect the junction strength obviously. Fig. S5 shows the selected snapshots of the CNT bundle pulling out from graphene without any entanglement at the bottom.

Section S6: Transfer of debonded CNT bundle onto Cu grid

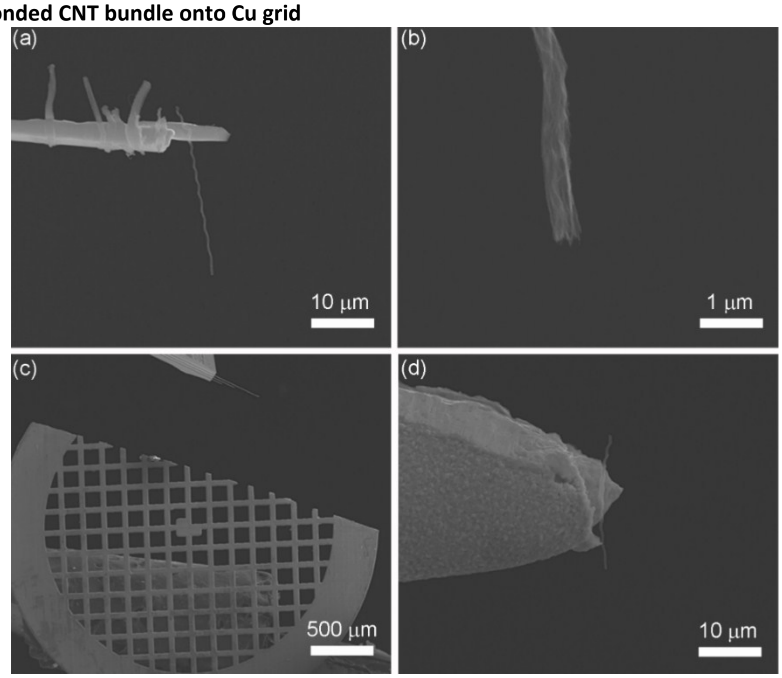

Fig. S6 The transfer of debonded CNT bundle onto Cu grid. (a) The pulled out CNT bundle. (b) The root of CNT bundle. (c) The sectioned copper grid. (d) The CNT bundle bonded onto sharp corner.

Fig. S6 shows the transfer of debonded CNT bundle onto Cu grid. In order to see whether there is residual of graphene connecting to the CNT bundle, we pulled out a CNT bundle shown in Figs. S6a and S6b and loaded it onto a sliced copper grid for observation in TEM. A copper grid was intentionally cut into two pieces. The pulled out CNT bundle was loaded onto the sharp conner of sliced copper grid as shown in in Figs. S6c and S6d. The carbon was deposited using EBID to bond the CNT bundle with the copper rod. Then, the AFM cantilever was moved away and the CNT bundle was left on the copper grid. The CNT bundle together with copper grid was inserted into TEM (JEOL 2100 F) to observe the residual graphene.

Section S7: Harvest graphene after removing CNT bundles

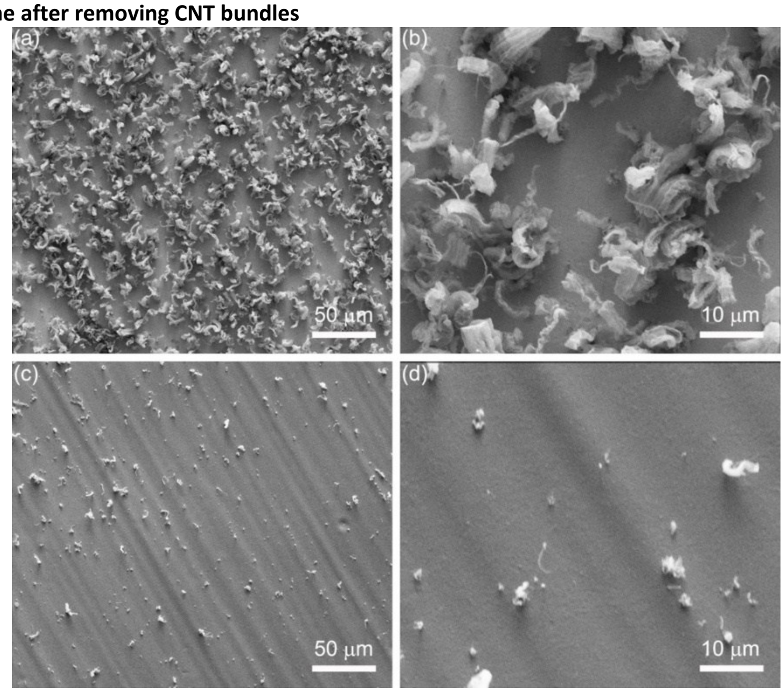

Fig. S7 SEM images of graphene/Cu after removing CNT forest from 3D CNTs/graphene nanostructures with scotch tape. (a and b) After first peel. (c and d) After second peel.

Captions of Videos

Video S1: The video for the CNT bundle debonding from graphene in Fig. 2.

Video S2: The video for the CNT bundle debonding from graphene, pushed back onto graphene, and pulled away from graphene in Fig. 4.

Video S3: A representative theoretical case of the CNT pull-test for strength prediction.