Supplementary Figures for

Self-Assembly of Graphene on Carbon Nanotube Surfaces

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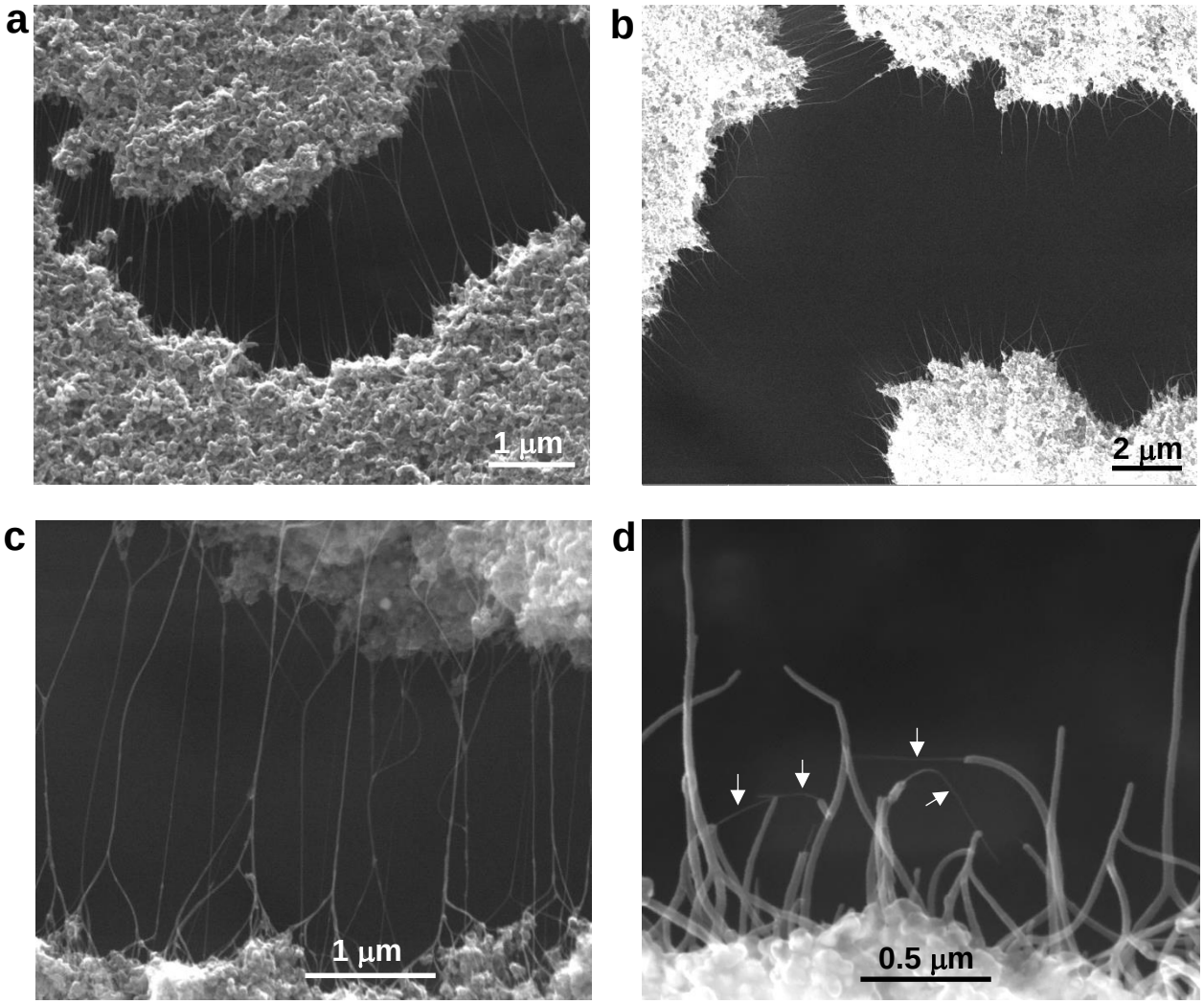

Figure S1. SEM images of SWCNT paper before and after the graphene growth. **a** and **b**, SWCNT paper before graphene growth. There exist many micron scale cracks in the paper. Pure individual and bundled SWCNTs either bridge the cracks or extrude from the fractured surface of the cracks. **C** and **d**, SWCNT paper after graphene growth. The SWCNT paper was well preserved during CVD and post growth handling steps. The white arrows in **d** mark the SWCNTs that were pulled out after fracture of the thickened tubes. The growth times for the tubes in **c** and **d** are 5 min and 10 min, respectively.

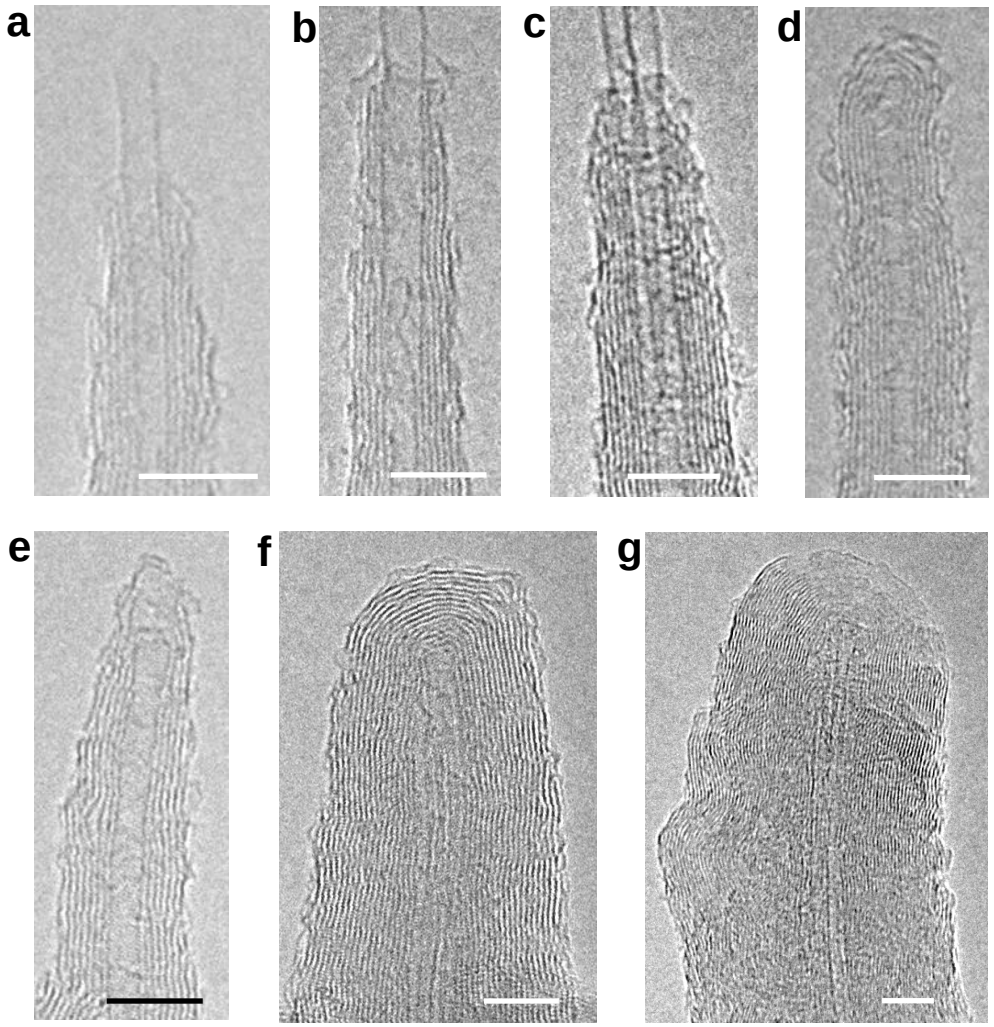

Figure S2. High-resolution TEM images of graphene grown on SWCNTs. **a**–**c**, Partially thickened nanotubes with cone-shaped morphology. **d**–**g**, Thickened nanotubes with different cap morphologies. The growth times for these thickened nanotubes are 4 min (**a, b**), 5 min (**c**–**e**), and10 min (**f, g**). The scale bars are 5 nm.

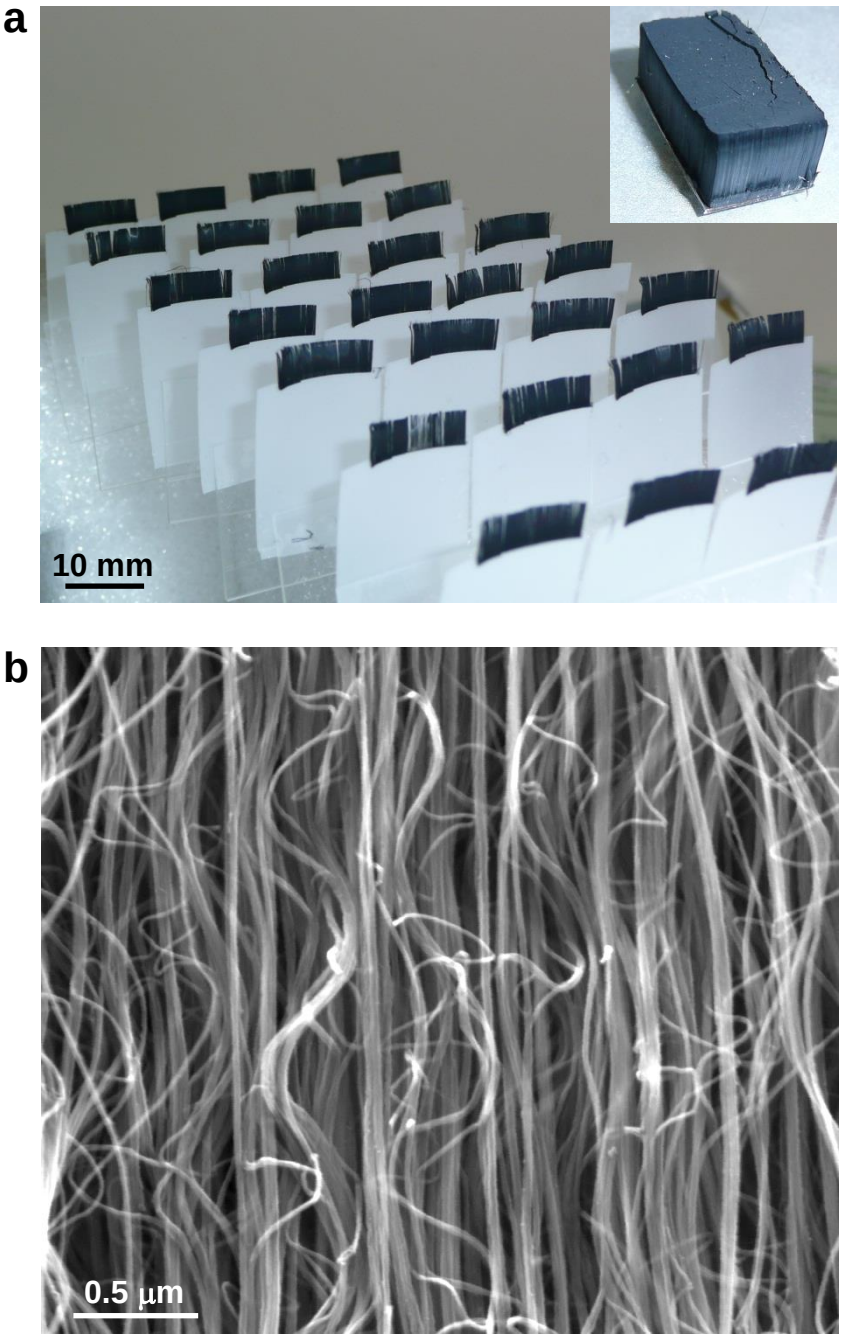

Figure S3. Aligned MWCNT sheets. a, Optical image of 6 mm tall vertically aligned nanotube sheets. The inset shows a 6 mm tall MWCNT forest. The nanotube sheets were drawn from a sidewall of the forest using adhesive tape. **b**, SEM image of the aligned MWCNT sheet.

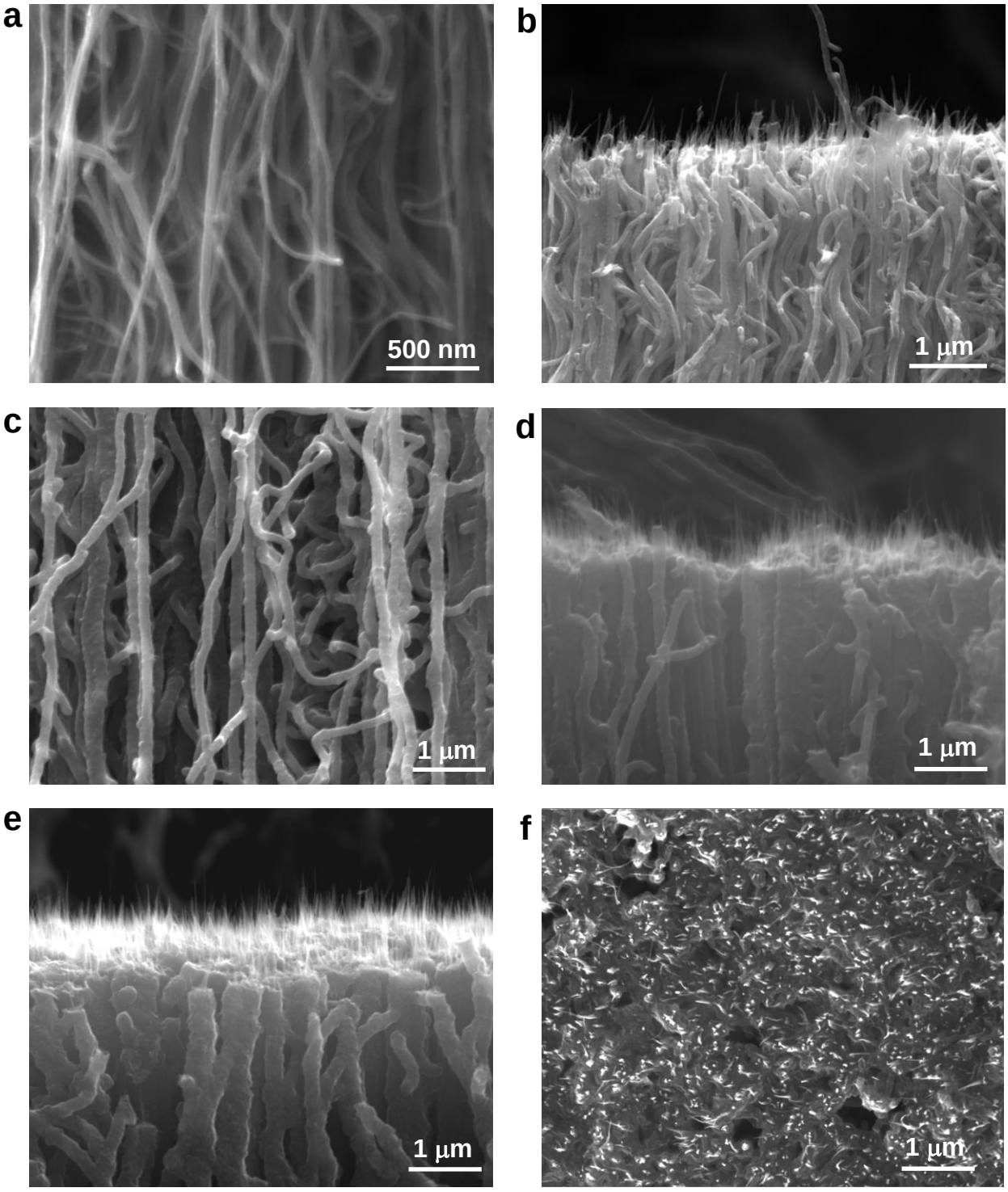

Figure S4. SEM images of MWCNT sheets after graphene growth. a, A sheet after 10 min of graphene growth. **b**, A fractured sheet after 30 min of graphene growth. **c**, A sheet after 60 min of graphene growth. **d**, A fractured sheet after 60 min of graphene growth. **e**, A fractured sheet after 120 min of graphene growth. **f**, Top view of the fractured sheet in **e**. The fractured sheets in **b**, **d**, **e** and **f** clearly show the pullout of the nanotubes. The diameters of the thickened tubes are ~20–30 nm, ~60–80 nm, ~100–150 nm, and ~200–250 nm after 10, 30, 60, and 120 min deposition, respectively. After 60–120 min deposition, solid nanotube sheets were formed by the coalescence of the thickened nanotubes.

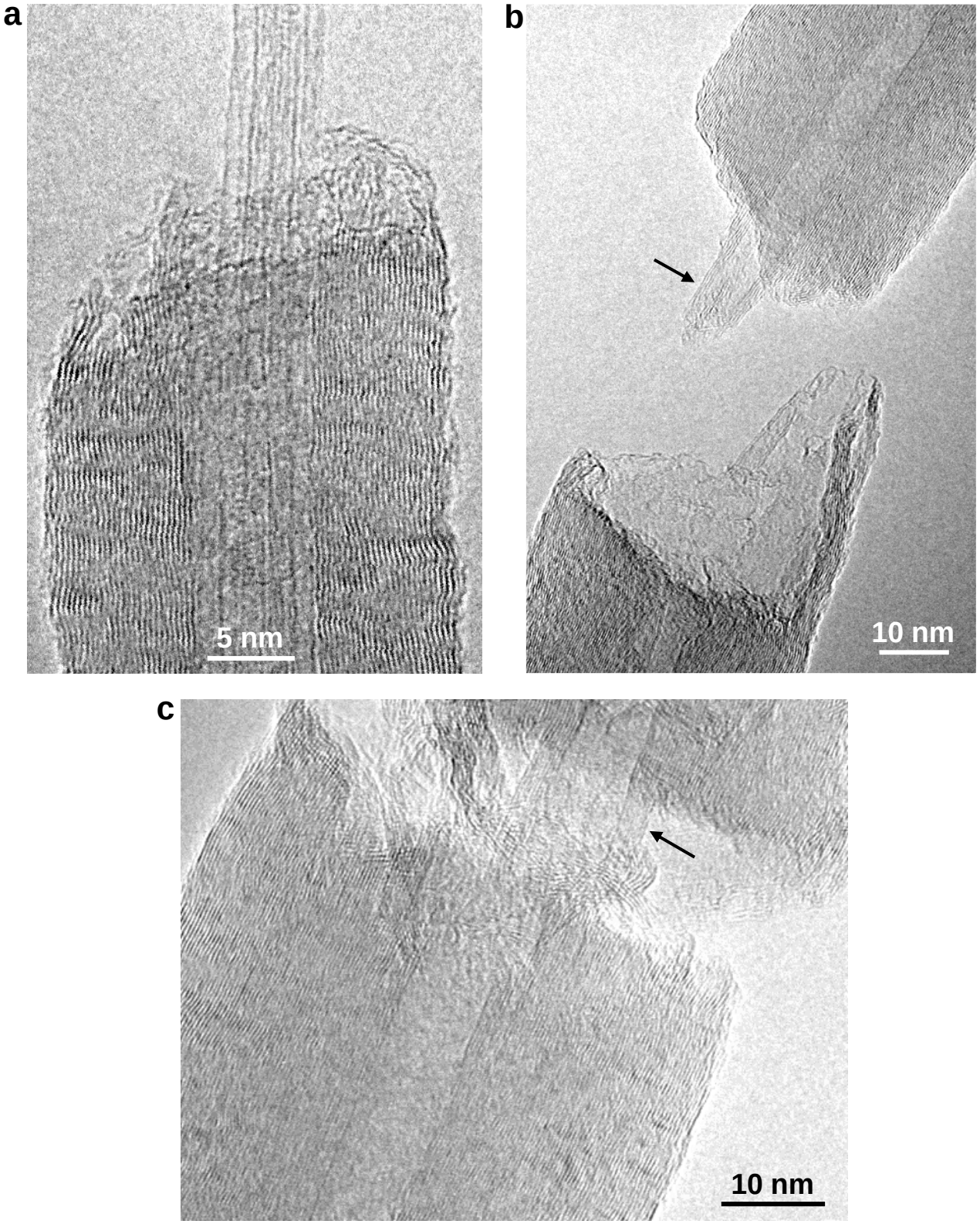

Figure S5. High-resolution TEM images of fractured nanotubes. a, A fractured nanotube showing the pullout of a bundle of primary SWCNTs. **b** and **c**, Fractured nanotubes showing the pullout of the primary MWCNTs (indicated by arrow heads).