

## Supporting information

Identification and technical accessibility of the carbon self-assembly concept hidden  
in catalytic carbon nanotube evolution

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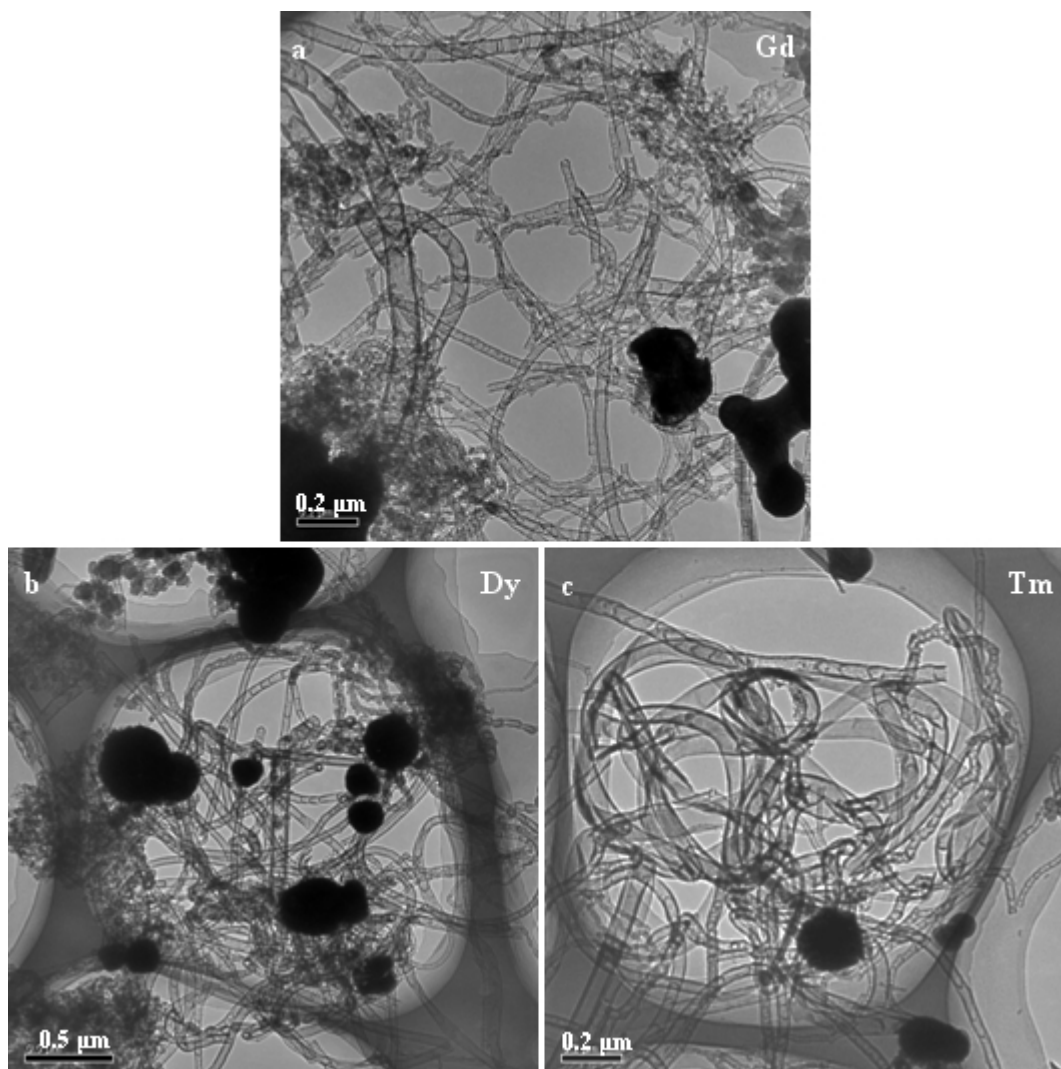


Figure S1. TEM images of the CNTs obtained from the catalyses of (a) Gd, (b) Dy, and (c) Tm.

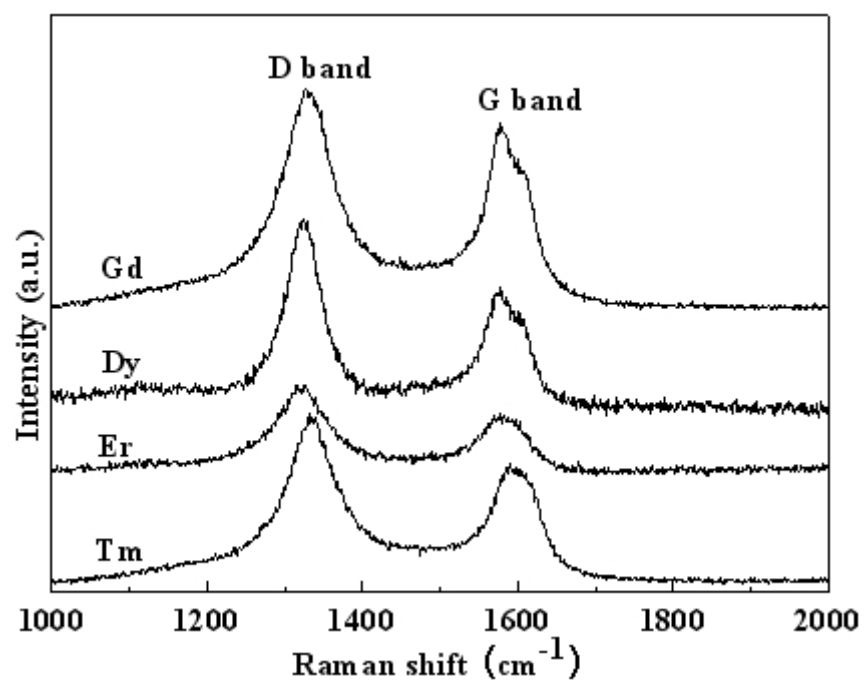


Figure S2 Raman spectra of the samples obtained from the catalyses of Gd, Dy, Er and Tm.

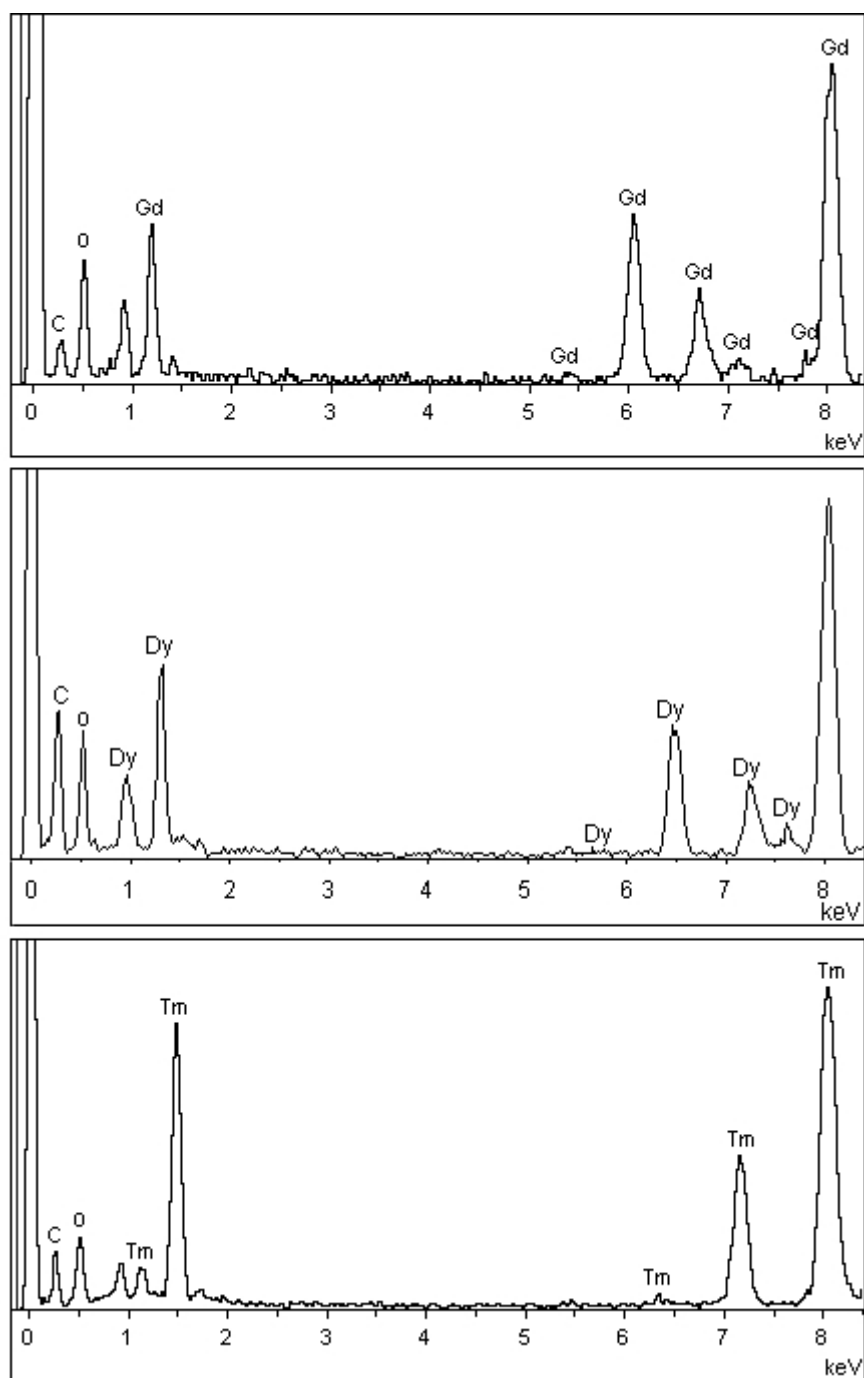


Figure S3. Typical energy dispersive X-ray spectrum (EDS) of the catalyst particles.

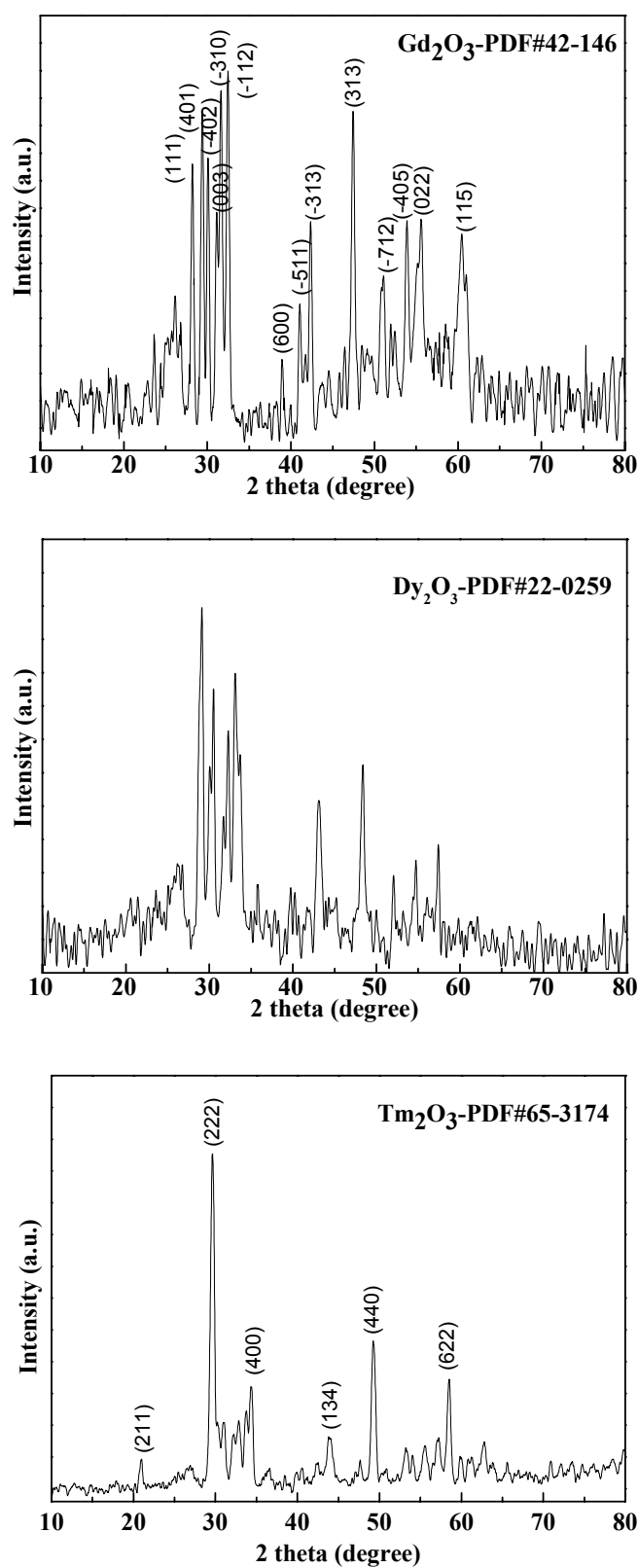


Figure S4. X-ray diffraction (XRD) patterns of the samples obtained the catalyses of Gd, Dy, Tm. Note: the detailed index of  $Dy_2O_3$  crystal is still lack in the published PDF cards (#22-0259).

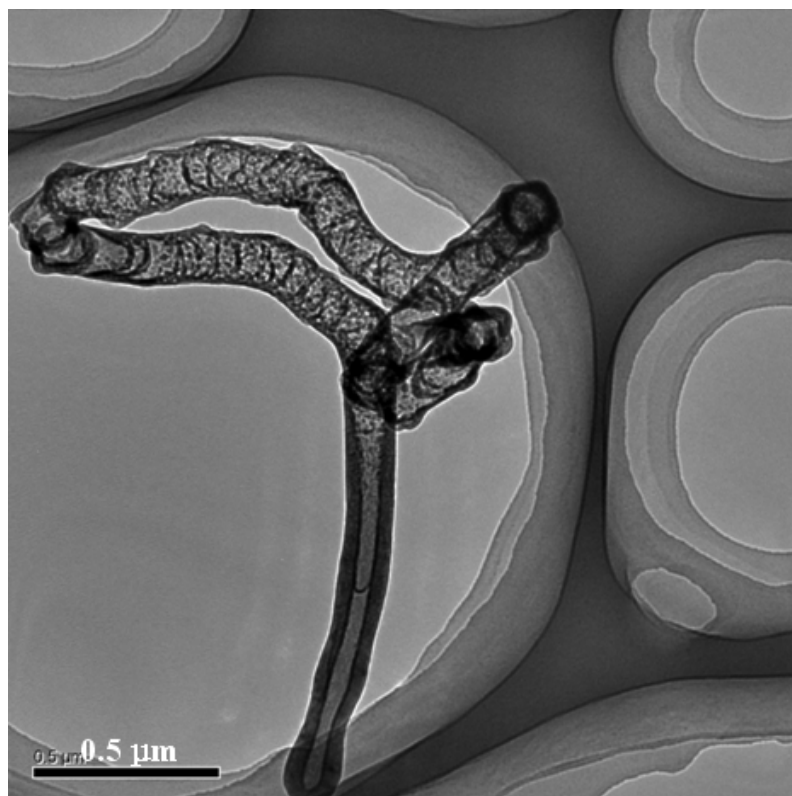


Figure S5. A typical tube with morphologically different moieties, reflecting stepwise evolution process: from nanowire assembled by small carbon nanoparticles to bamboo-like tubular structure and finally to well-developed hollow tube.

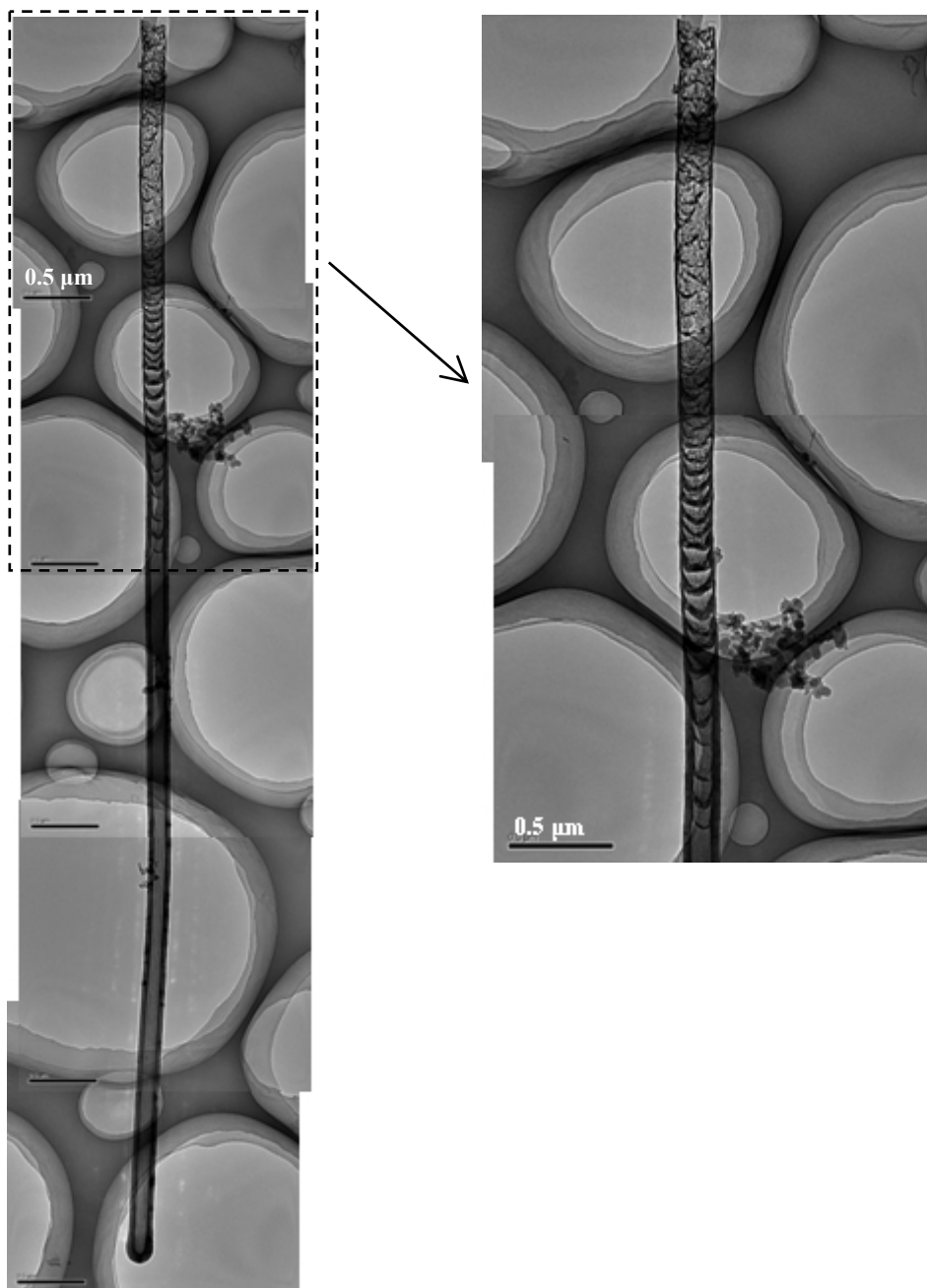


Figure S6. A tube intermediate, reflecting the self-assembly, coalescence and reorganization of carbon particles during tube evolution.

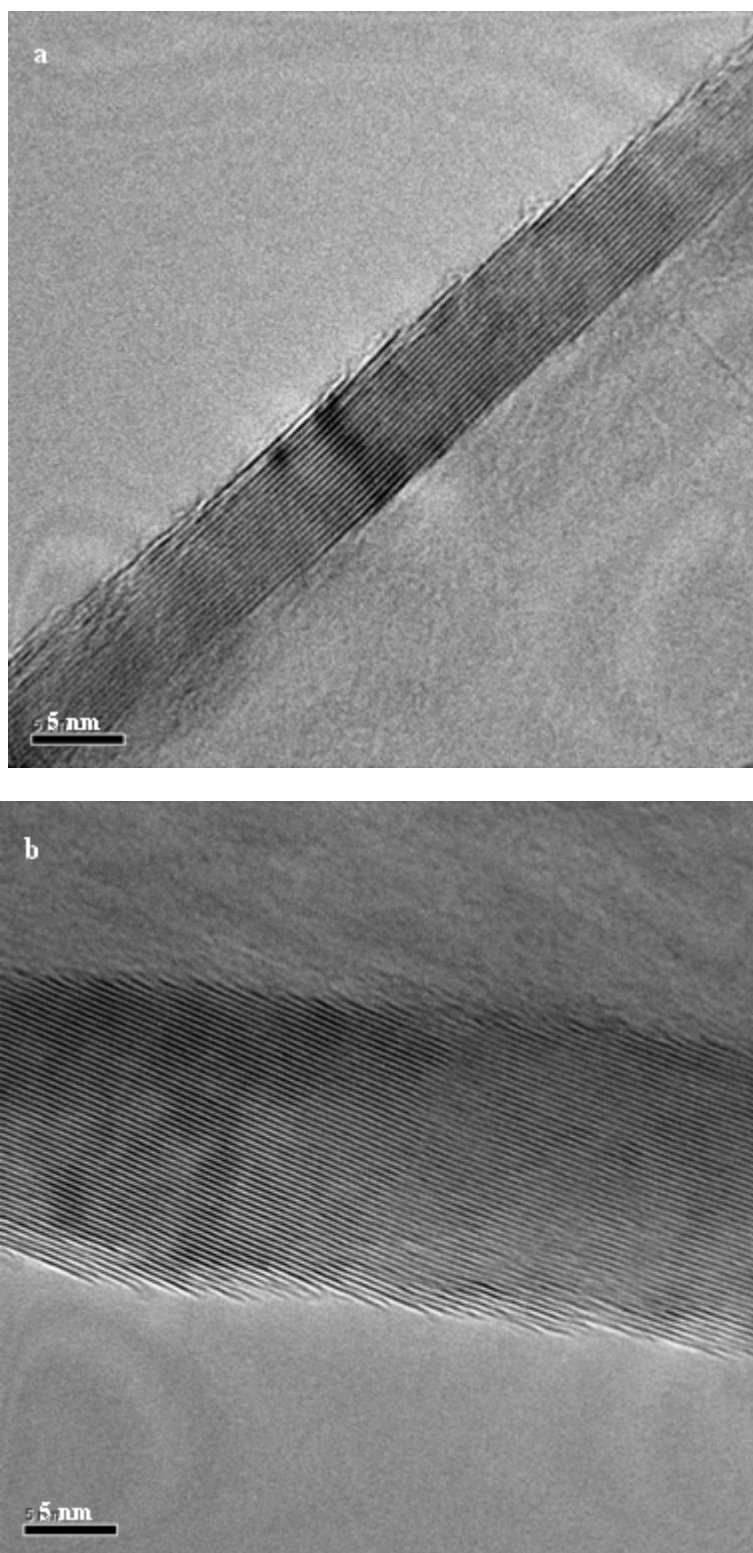


Figure S7. Representative high-resolution TEM images of the walls of the CNTs obtained from the non-catalysis transformation of AC. (a) Co-axial cylindrical graphitic structure. (b) fishbone-like structure.



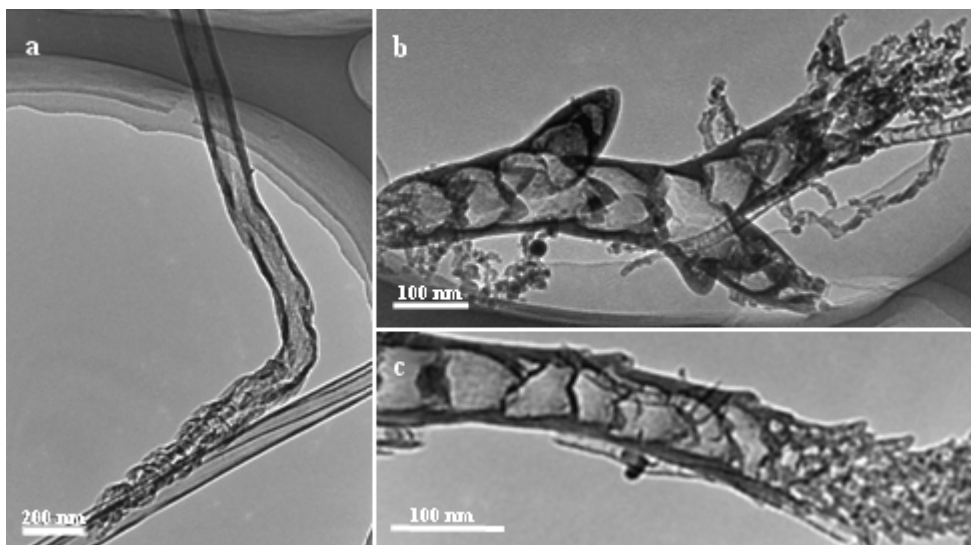


Figure S8. Tubular intermediate objects obtained from the detonation of the mixture of AC and TNP (without catalysts), displaying a close relationship between tube evolution and the self-assembly of carbon particles.

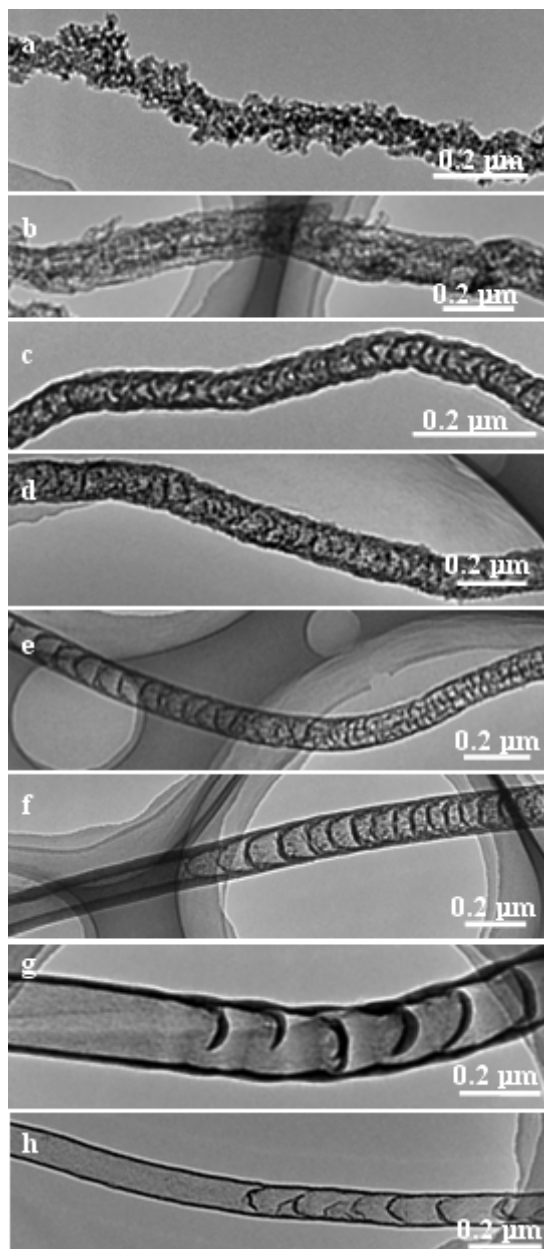


Figure S9. Tubular intermediate objects obtained from the first detonation of the mixture of AC and TNP (without catalysts). (a, b) Nanowires organized by carbon nanoparticles. (c, d) The nanoparticles on the wires become coalesced, leaving behind smooth outer surface and enlarged inner spaces. (e) Structural reorganization leads to a formation of bamboo-like tubule, in which inner spaces are significantly developed. (f-h) Further structural reorganization makes the arch parts smash up and joint to tube wall, resulting in fully hollow tubes.

Note: from the information shown above, bamboo-like tubule seems the intermediate objects during the evolution of fully hollow tubes. We have collected further

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evidences for it, which will be subsequently published.

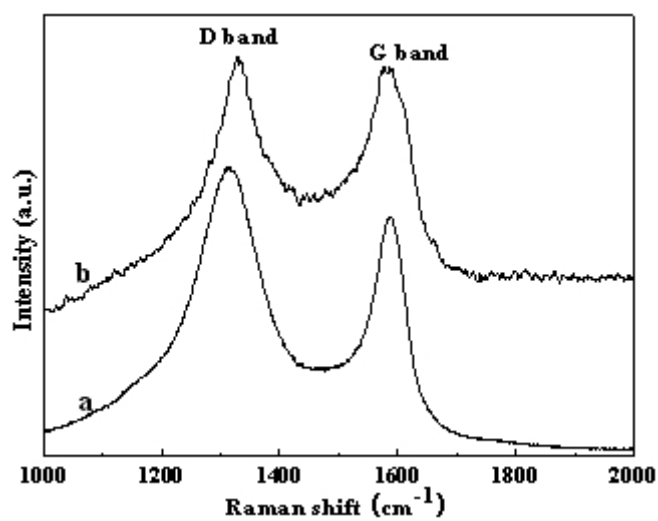


Figure S10. Raman spectra of the products obtained (a) from the first detonation reaction of the mixture of TNP and AC, and (b) from the additional annealing treatment of the first detonation products. The spectra display a significant increase of CNT content and CNT structure graphitization.

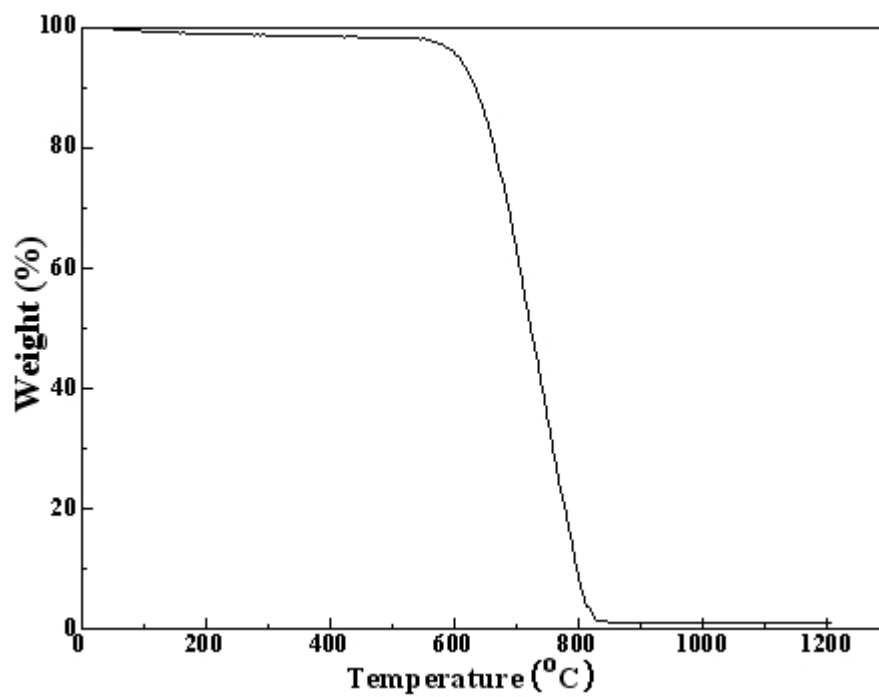


Figure S11. Thermogravimetric analysis of demineralized AC in air at a rate of 5 °C/min, showing that the content of total minerals is less than 1.2%.