Electronic Supplementary Material (ESI) for Nanoscale
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Supplementary information

3-order-of-magnitude density control of single-walled carbon nanotube networks by maximizing catalyst activation and dosing carbon supply

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Fig. S1. Comparisons of surface morphology of catalyst nanoparticles formed after fast-heating and normal heating processes. The morphology of catalyst nanoparticles was measured by atomic force microscopy (AFM; Asylum Research MFP-3D) with a Si cantilever having a spring constant of 50 N/m and a frequency of 275 kHz (Budget Sensors). In the **(a)** fast-heating process, a large number of small nanoparticles with a size of 1 – 2 nm and only a few large nanoparticles with a size of >3 nm were formed. In comparison, catalysts pretreated at 900 °C in **(b)** Ar (200 sccm) atmosphere at 760 Torr or **(c)** Ar (200 sccm) and H_2 (40 sccm) atmosphere at 760 Torr showed many nanoparticles with a size of >3 nm. The corresponding line profiles of these AFM images are also plotted.

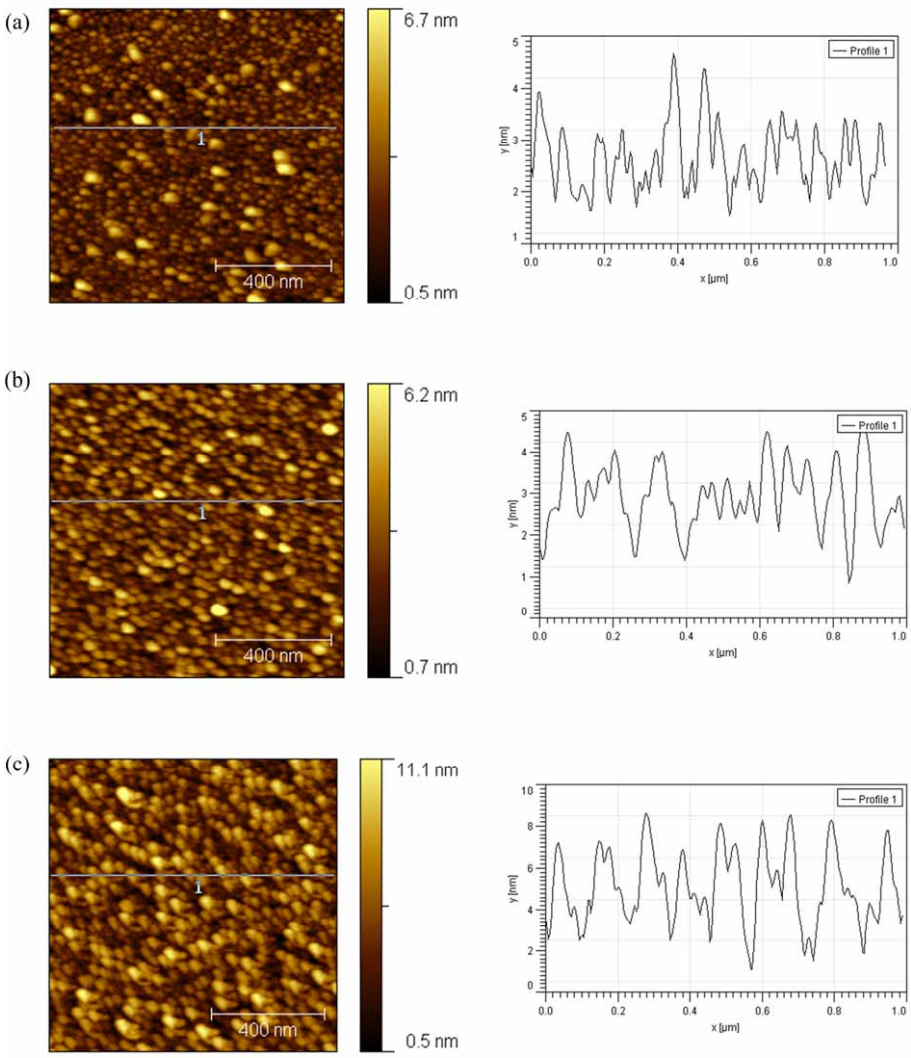

Fig. S2. Chemical states of catalyst nanoparticles formed after fast-heating and reduction. The chemical states of catalyst nanoparticles formed after fast-heating and reduction was measured by x-ray photoelectron spectroscopy (XPS; Specs SAGE 150) with Mg Kα excitation at 1253.6 eV. Fe 2p and Al 2p spectra are clearly shown below. *Mattevi et al.* indicated that as compared to the Fe/SiO_2 catalyst system, a strong interaction between the Fe catalyst and the Al_2O_3 supporting layer could cause a shift of the Fe 2p peak from the metallic state (at binding energy of 707.3 eV) to higher binding energy [see *Mattevi et al., J. Phys. Chem. C 112, 12207-12213 (2008)*]. This strong interaction helped restrict Fe mobility and led to a much higher density of Fe catalyst nanoparticles. Indeed, the Fe 2p peak in our samples was seen at binding energy of 710.3 eV (of course the slight oxidation of these Fe nanoparticles was not excluded as the measurements were done *ex situ* after the samples were exposed to atmosphere). A higher carbon nanotube nucleation density was therefore expected using the Fe/Al_2O_3 catalyst system.

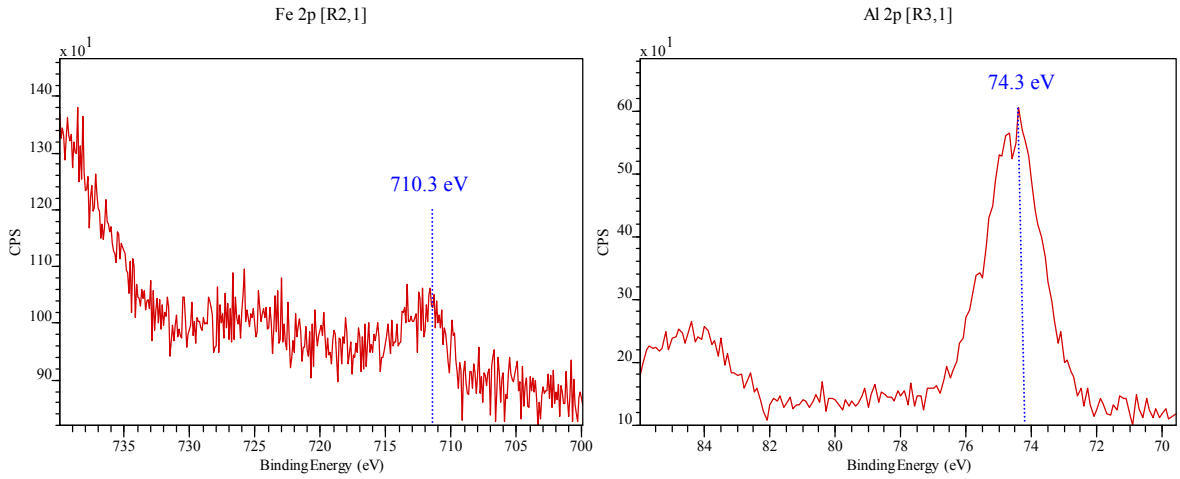
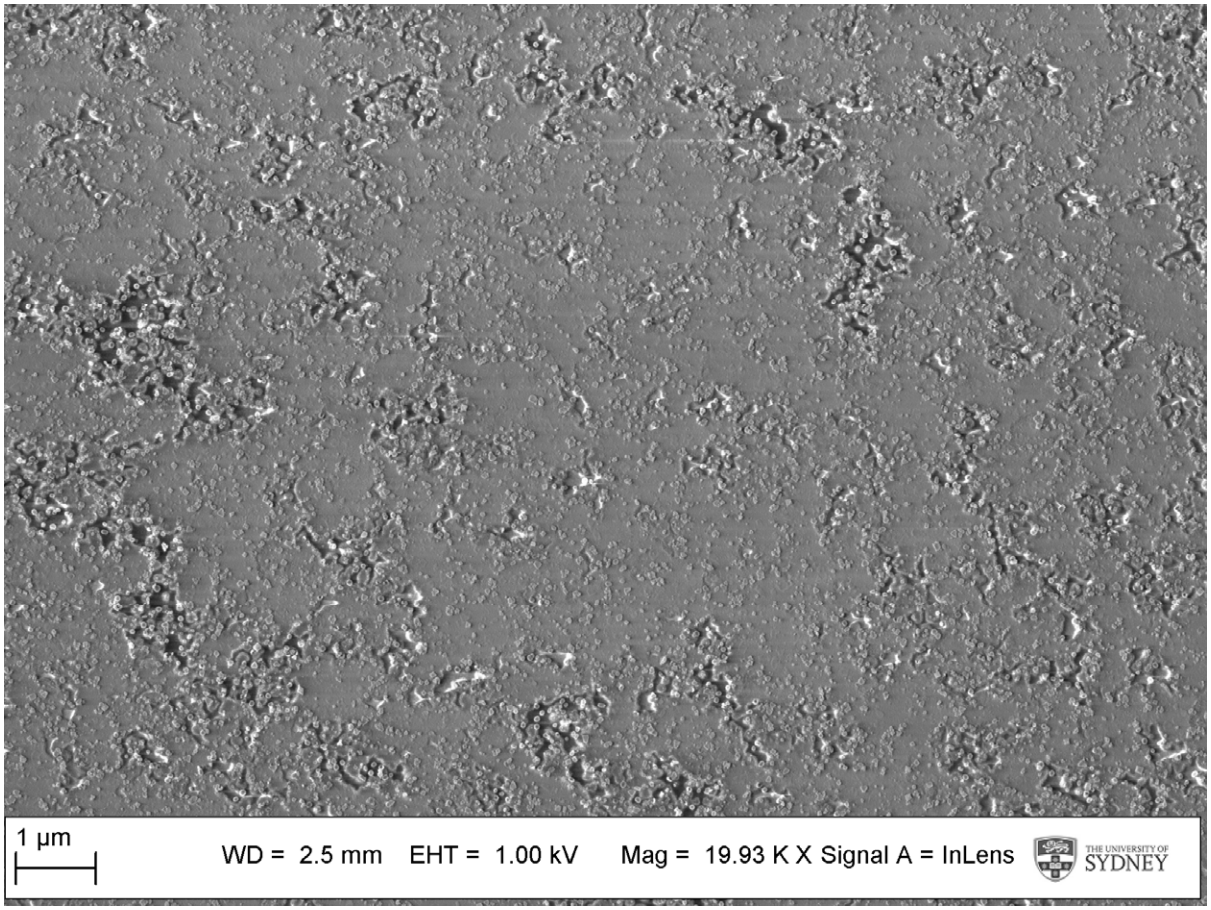
Fig. S3. Excessive acetylene used in the CCVD growth process results in amorphous carbon coating. If excessive acetylene was added, *e.g.*, by increasing the time of the pulses from 2 s to 15 s in the 3-pulse acetylene CCVD growth process (flow rate was kept at 60 sccm), we obtained amorphous carbon coatings instead of random SWCNT networks.