Supplementary Material (ESI) for Chemical Communications

Experimental

Vertically aligned carbon nanotubes (VACNTs) were synthesized on SiO_2/Si wafer by pyrolysis of iron(II) phthalocyanine (FePc) under Ar/H_2 at 800-1100 °C.[1] The thickness of Si substrate is about 0.4 mm. The VACNTs synthesized for this study have a typical tube length of about 6 micron and diameter of *ca.* 20-50 nm. For the controlled embedment of nantoubes into polymer films, a VACNT film grown on the SiO_2/Si substrate was held on a hot plate (Thermolyne) and heated up to a desired temperature. In a typical experiment, for example, the top of a VACNT array was fully covered with a thin film of polystyrene (~ 50 μm). Upon heating over the melting point and below the decomposition temperature of PS, the melted PS layer gradually infiltrated into the aligned nanotube "forest" (Figure S1). After a certain heating time, the polymer-embedded VACNTs were removed away from the heating stage and peeled off from the SiO_2/Si substrate by HF etching[1] to yield a free-standing film (See, Figure S2).

The selective deposition of metal (*e.g.* Au, Pt) nanoparticles onto polymer-free regions of the VACNTs embedded into a polymer film was carried out by the SEED method.[2] Briefly, for the deposition of Au nanoparticles, the VACNTs partially embedded into a PS film were supported with a copper foil, followed by immersing them into an aqueous solution of $HAuCl_4$ (3.8 mM) for 30 s. Pt cubic nanoparticles were formed at 3.8 mM K_2PtCl_4 and 10 mM $CuCl_2$ for 1 min, while Pt spherical nanoparticles were produced at 0.95 mM K_2PtCl_4 for 30 min (Figure S3).[2]

Iron sputter coating was carried out on a Denton Sputter coating system (Explorer 14) to directly deposit iron nanoparticles onto the tips of VACNT partially embedded into a

polymer matrix. Prior to the iron sputtering, the residual iron catalyst nanoparticles in the VACNT array were removed by immersing in an aqueous solution of HF (10%wt), water-plasma etching, and HCl treatment [3] (Figure S4). Scanning electron microscopic (SEM) images were recorded on a Hitachi S-4800 high-resolution SEM unit.

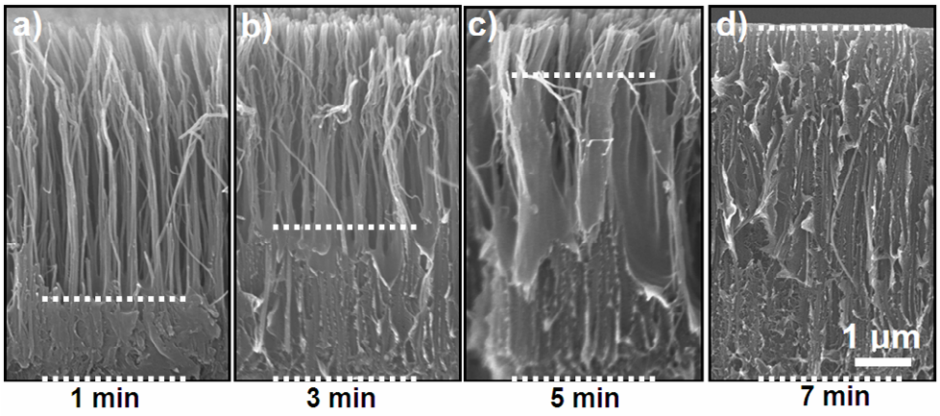


Figure S1. SEM images of VACNT arrays being embedded by PS with an increased heating time at 195°C.

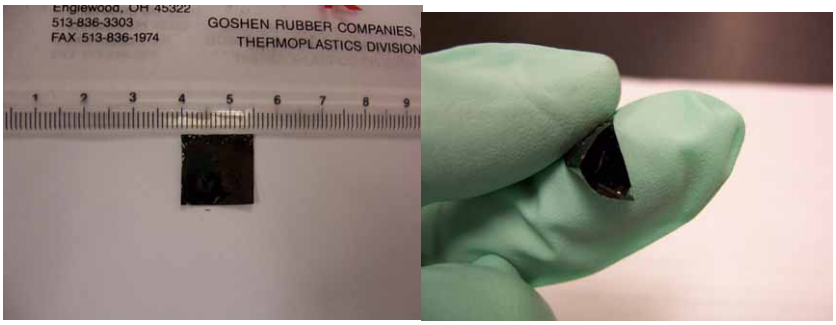


Figure S2. Photos showing a free standing flexible film of the PS-embeded VACNTs.

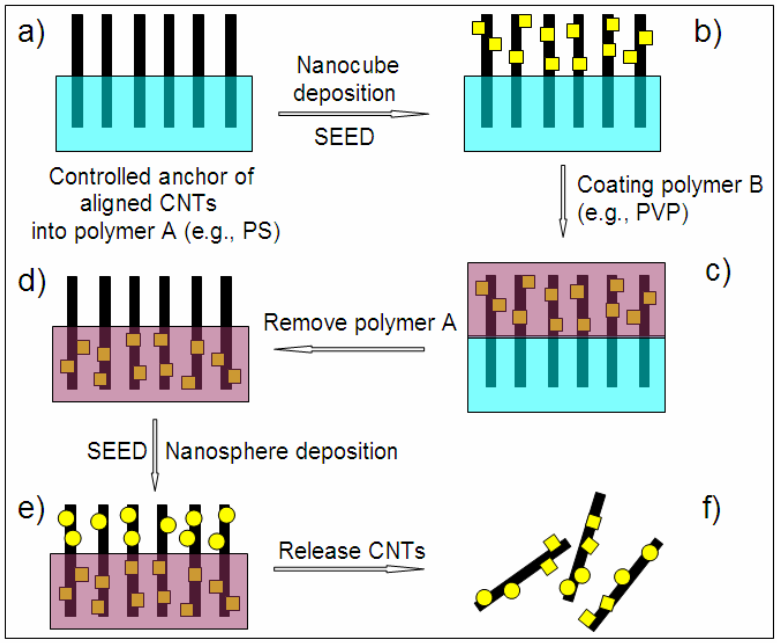
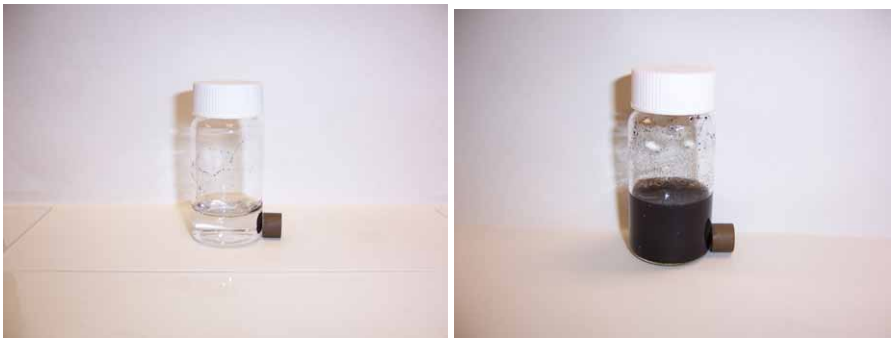


Figure S3. Schematic representation for the region-selective deposition of metal cubes and spheres onto the constituent nanotubes in VACNT arrays along the nanotube length.



(a) (b)

Figure S4. Photos of the HF/water-plasma/HCl-treated CNTs with (a) and without (b) sputter-coated iron after being dispersed in ethanol and applied with an external magnetic field. (a) standing 2 min; (b) standing 30 min.

(1) S. D. Huang, L. Dai, A. W. H. Mau, *J. Phys. Chem. B*, 1999, **103**, 4223-4227.

(2) (a) L. T. Qu, L. Dai, *J. Am. Chem. Soc.*, 2005, **127**, 10806-10807; (b) L. T. Qu, L. Dai, E. Osawa, *J. Am. Chem. Soc.*, 2006, **128**, 5523-5532.

(3) S. M. Huang, L. Dai, *J. Phys. Chem. B*, 2002, **106**, 3543-3545.