

Supplementary Information

Experimental details

Materials: MWNTs were purchased from Iljin Nanotec, Inc. and purified by acid treatment using the mixture of sulfuric acid and nitric acid (volume ratio of 3:1) and following heat treatment. A limited amount of carboxylic acid groups were generated on the surface of MWNTs during the acid treatment. The subsequent heat treatment was carried out at 400 °C for 40 min under atmospheric condition to remove amorphous carbon and residual acid solutions. Benzene and Dimethylformamide (DMF) were purchased from Merck. Monodisperse amine terminated Polystyrene (PS-NH₂) having the same M_w of 3,000 g/mol was purchased from Polymer Source, Inc.

Functionalization of MWNTs with end-functionalized polymers: MWNTs were dispersed in benzene through covalent or noncovalent functionalization with PS-NH₂. The noncovalent grafting through zwitterionic interaction was accomplished by simple solution mixing in benzene solution. The amide reaction between amine groups of PS-NH₂ and carboxylic acid of MWNTs were carried out by refluxing them in DMF at 140 °C for 120 hours. After the amide reaction, the functionalized CNTs (CNT-PS) were collected by vacuum filtration.

Fabrication of CNT scaffolds: The benzene solution containing the predetermined amounts of PS-NH₂ (0.5 mg/ml ~ 10 mg/ml) and MWNTs (0.1 mg/ml ~ 1 mg/ml), or CNT-PS (0.05 mg/ml) was deposited on the silicon wafer or indium-tin-oxide (ITO) glass under a humid air flow of 3 L/min (relative humidity of 80 %) at room temperature. The solution was left until all solvents were evaporated, yielding porous nanocomposite films. Cellular CNT film or mat was prepared by the subsequent calcination of PS matrix through pyrolysis conducted at 400 °C for 20 min.

Characterization of CNT Scaffolds: The morphology of a porous CNT film was investigated using a field emission scanning electron microscopy (FE-SEM, HITACHI S4800). Surface resistance was measured using four point probe (Mitsubishi Chemical Corporation, MCP-T360). Field emission properties were measured using diode structure in high vacuum chamber at the pressure of 10⁻⁷ Torr.^[21] Porous CNT film and ITO glass were used as cathode and anode, respectively. The space between cathode and anode was maintained to 300 μm and applied voltage to cathode was up to 850 V. The sample size for field emission was 1.1 cm².

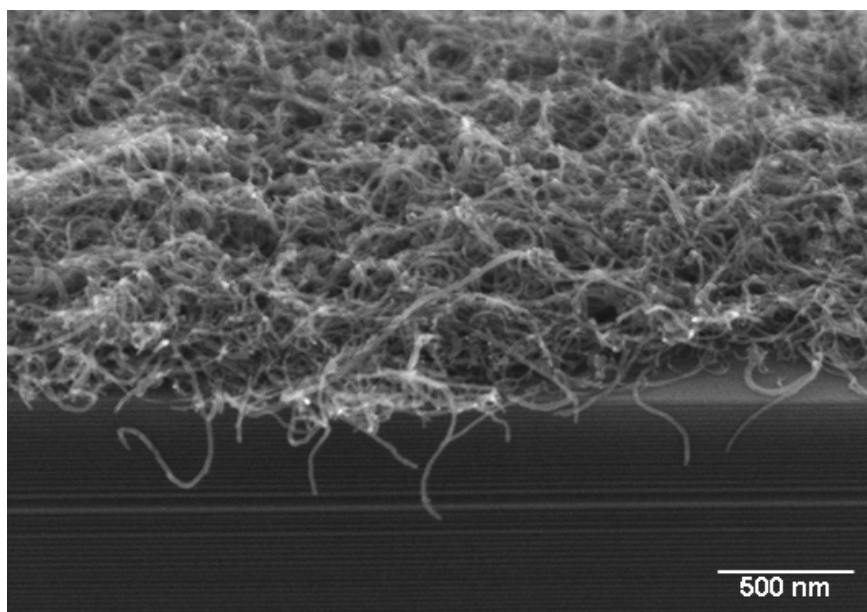


Fig. S1 60° tilted scanning electron microscopy (SEM) image of the fractured carbon nanotube (CNT) scaffold prepared by applying ‘dry N₂’ air over a nanocomposite film. The concentration of amine-terminated polystyrene (PS-NH₂) and multi-walled carbon nanotubes (MWNTs) in the precursor solution were 3 mg/ml and 1 mg/ml, respectively. Without applying humid air, the CNT scaffold does not construct robust three-dimensional architecture. It is a direct evidence demonstrating that the highly-entangled three-dimensional scaffolds of CNTs presented in Fig. 2 & 3 were driven by the nucleation and growth of the closely packed aqueous droplets.

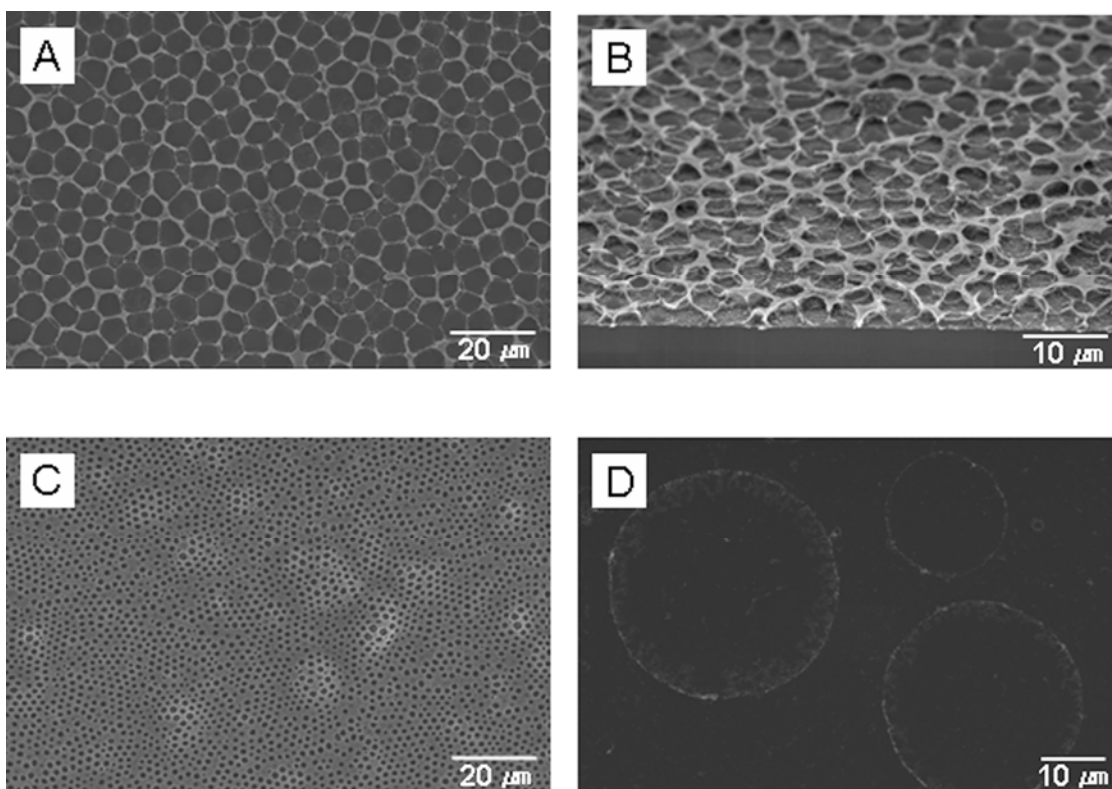


Fig. S2 Low magnified SEM images of cellular scaffolds with various concentrations of PS-NH₂ and CNTs. (A) Plane view of the monolayered cellular scaffold prepared from a precursor solution including 1 mg/ml of PS-NH₂ and 0.5 mg/ml of MWNTs. (B) 60° tilted image of a monolayered cellular scaffold. The precursor solution contained 3 mg/ml of PS-NH₂ and 1 mg/ml of MWNTs. (C) Plane image of porous CNT mat fabricated from a solution containing 10 mg/ml of PS-NH₂ and 5 mg/ml of MWNTs. As concentrations of polymer and CNTs are increased, pore sizes and their distributions are decreased. (D) Image of CNT ring scaffolds obtained from 1 mg/ml of covalently functionalized CNTs (PS-CNT) in benzene.