Supporting Information

Figure S1. SEM image of a CNT array from a side view.

Figure S2. High-resolution TEM image of a CNT.
**Figure S3.** Raman spectrum of a CNT array.

**Figure S4.** Electrocatalytic characterization by the cyclic voltammetry performed in 1 mM I\textsubscript{2}, 10 mM LiI, and 0.1 M LiClO\textsubscript{4} acetonitrile solution with a scan rate of 100 mV s\textsuperscript{-1} on a three-electrode setup. Two oxidation/reduction peaks clearly show a catalytic activity derived from CNTs for reduction of I\textsubscript{3}\textsuperscript{-}. The left and right peaks correspond to the oxidation/reduction of I/I\textsubscript{3} and I\textsubscript{2}/I\textsubscript{3}, respectively (Electrochim. Acta 2008, 53, 2890).
**Figure S5.** A typical tensile stress-stain curve of a perpendicularly aligned and penetrated CNT/resin composite film.

**Figure S6.** J-V curve of a dye-sensitized solar cell by using the platinum as counter electrode.
**Figure S7.** SEM images of aligned CNT/resin composite film derived from a pressed CNT array (improved for three times in CNT number density).

**Figure S8.** SEM images of aligned CNT/resin composite film after incorporating (a) polydiacetylene, (b) polyaniline, and (c) polypyrrole into pure CNT arrays.
**Figure S9.** Photo of a flexible dye-sensitized solar cell by using the perpendicularly aligned and penetrated CNT/polymer composite film as counter electrode.

**Figure S10.** A typical J-V curve of a flexible dye-sensitized solar cell by a perpendicularly aligned and penetrated CNT/resin composite film as a counter electrode measured under AM1.5 illumination.
**Figure S11.** Schematic illustration to the mechanical measurement.