#### Supporting information for Chem. Commun.

# Electrically moving single-stranded DNA into and out of double-walled carbon nanotubes

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#### 1. Experimental details

DWNTs used here were prepared by an arc-discharge method with iron as catalyst. The raw DWNTs were purified by a combination of acid treatment and air oxidation before using. The purified DWNTs were dispersed by a brief supersonic treatment in ethanol. Then, droplets of this suspension were dripped and dried on Al substrates (0.5 cm  $\times$  7 cm). Single-stranded DNA (adenine, guanine, cytosine thymine) with the base number ranging from 5 to 100 were purchased from Nihon Gene Research Laboratories Inc. The synthesis of ssDNA@DWNTs was performed by applying an electric field equivalent to a 10 V positive DC voltage to the substrate on which ssDNA with concentration of 4 µmol/L was loaded, during which the ssDNA molecules were inserted into the cavity of DWNTs. The synthesis process is partially similar to the case of ssDNA encapsulated SWNTs using an Al-substrate bias method in a DNA electrolyte plasma.<sup>1</sup> The obtained sample on the aluminum substrate was dispersed in the DI water for 1h and then the resultant suspension was centrifuged at 40000 g for 1h to remove DWNTs to which ssDNA are attached outside. The sediment of ssDNA@DWNTs on the bottom of a sample container was collected and analyzed by field emission transmission electron microscopy (FE-TEM, Hitachi HF-2000) operated at 200 KV. In the case of releasing ssDNA from the inside of DWNTs, the above purified ssDNA@DWNT samples were placed again on the Al electrode substrate which was then immersed in the DI water. Then the negative DC bias -10 V was applied to the ssDNA@DWNT samples on the Al substrates for 10 min, 30 min, and 60 min, respectively. After that, the sonication process was repeated to remove DWNT samples from the Al substrates.

UV-Vis spectra were measured with a V-7200 spectrophotometer in a wavelength range of 250-500 nm. Quartz cells with volume of 5 mL were used to contain samples. In the case of measurements for released ssDNA, the DI water was used to dissolve ssDNA samples with a reference cell containing the 5mL DI water. In the case of ssDNA@DWNT sample, UV-vis spectra were taken by directly depositing various

DWNT samples on the surface of quartz cells, and also an empty quartz cell was used as a reference for the measurements.

In order to make field-effect transistor (FET) devices, DWNT samples including pristine DWNTs and ssDNA@DWNTs at various filling levels were firstly dispersed by supersonic treatment for over 9 hours in the *N*, *N*-dimethylformamide (DMF) solvent. Then the nanotube solution was spincoated onto an FET substrate, which consists of pairs of Au electrodes.<sup>2</sup> The individual DWNT bridging the two Au electrodes on the substrate was confirmed by atomic force microscopy (AFM; JSPM-5400). Electronic transport properties of DWNT-FETs are measured using a semiconductor parameter analyzer (Agilent 4155C) in vacuum. The source-drain current ( $I_{DS}$ ) is investigated as functions of gate bias ( $V_G$ ).

2. TEM images of G<sub>30</sub>, T<sub>30</sub> and A<sub>30</sub> ssDNA@DWNTs



Fig. S1. TEM images and their simulated images of ssDNA encapsulated in individual DWNT: (a)  $G_{30}$  ssDNA@DWNT, (b)  $T_{30}$  ssDNA@DWNT, (c)  $A_{30}$  ssDNA@DWNT (scale bar: 5 nm).

### 3. Absorbance spectra of G<sub>30</sub> ssDNA release from the inside of DWNTs



**Fig. S2** (a) Absorbance spectra of pristine DWNTs, the synthesized  $G_{30}$  ssDNA@DWNT, and three  $G_{30}$  ssDNA@DWNT samples after ssDNA release for 10 min, 30 min, and 60 min, respectively, in which the arrow shows that the peak intensity for  $G_{30}$  ssDNA in DWNTs gradually decreases with increasing the experimental time. (b) Absorbance spectra of released  $G_{30}$  ssDNA (252 nm) in water by applying DC bias for 10 min, 30 min, and 60 min, respectively, where the arrow indicates that the peak intensity for released  $G_{30}$  ssDNA (252 nm) gradually increases with increasing the experimental time.

# 4. Electrical transport properties of C<sub>30</sub> ssDNA@DWNTs



**Fig. S3**  $I_{DS}$ - $V_G$  curves measured with  $V_{DS} = 0.1$  V for FET devices fabricated with the synthesized C<sub>30</sub> ssDNA@DWNT by applying 10 V bias for (a) 5 min, (b) 10 min, (c) 15 min, and (d) 20 min, respectively.

## Reference

- 1. T. Okada, T. Kaneko, R. Hatakeyama and K. Tohji, Chem. Phys. Lett., 2006, 417, 288.
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