

## DNA-SWNT Hybrid Hydrogel

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## Materials and equipments:

Oligonucleotides were purchased from Shanghai Sangon Biological Engineering Technology and Services Co.,Ltd., China. Single-walled carbon nanotubes (SWNTs) was purchased from Sigma Co.,Ltd. All other chemicals were of reagent grade or better. All experiments were performed using Millipore Milli-Q deionized water (15.6 MΩ cm resistivity).

Sequence A	5'-TACGCTGAATACCCCCAATCCCC-3'
Sequence B	5'-GTATTCAAGCGTACCCCCAATCCCC-3'
Sequence C	5'-GATGCG AGGCTATTCT-d(GT) <sub>20</sub> -3'
Sequence D	5'-AGAATAGCCTCGCATCCCCCTAACCCC-3'
Sequence E	5'-AGAATAGCCTCGCATCATGACTGCTCAGATCG-3'

UV/Vis spectra were recorded on a Varian Cary 100 spectrophotometer equipped with a programmable temperature-control unit. CD spectra were recorded on a JASCO-810 spectrometer. Rheology test experiments were performed on the AR2000ex rheometer manufactured by TA instrument company. The sonicator used was a UH-100A probe-type (Tianjin AutoScience Instrument Co., Ltd, China). All pH values mentioned were calibrated by a micro pH meter (FE 20 METTLER TOLEDO company).

## Gel electrophoresis experiment:

Native polyacrylamide gel electrophoresis (PAGE, 10%, Acr:Bis=29:1) of Fig. 1A was carried out in 1×TBE buffer (tris(hydroxymethyl) aminomethane (Tris, 89 mM), ethylenediaminetetraacetate (EDTA, 2 mM), and boric acid (89 mM), pH 8.0) and ran for 1 hour at 15 V/cm in the 4 °C refrigerator and native polyacrylamide gel electrophoresis (PAGE, 10%, Acr:Bis=29:1) of Fig. 1B carried out in 1× MES buffer (2-(N-morpholino) ethanesulfonic acid, 50 mM) and ran for 6 hours at 15 V/cm in the 4 °C refrigerator.

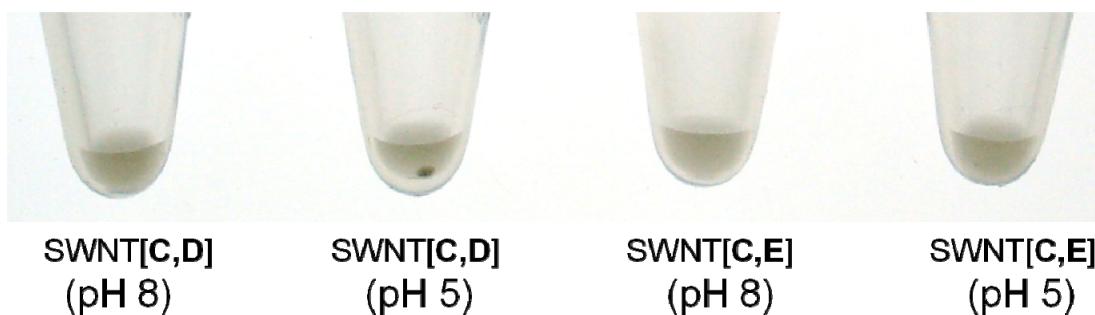
### **SWNT-DNA conjugate preparation:**

A 100  $\mu$ L solution containing about 0.1 mg single-walled carbon nanotubes, 0.025 mg DNA sequence C and 50 mM NaCl in 50 mM MES buffer (pH 8.0) was sonicated for 30 min at a power of about 4 W with a UH-100A sonicator. During sonication, the sample was maintained at 0 °C by an ice-water bath. The above solution containing SWNTs wrapped by sequence C (denoted as SWNT-C) was centrifuged at 16,000 g for 30 min to remove undissolved SWNTs as black precipitate. To the supernatant containing well-dispersed SWNT-C, oligonucleotide sequence D or sequence E was added in two times excess followed by an overnight incubation to produce SWNT-[C,D] or SWNT-[C,E] conjugates.

### **Free DNA removal:**

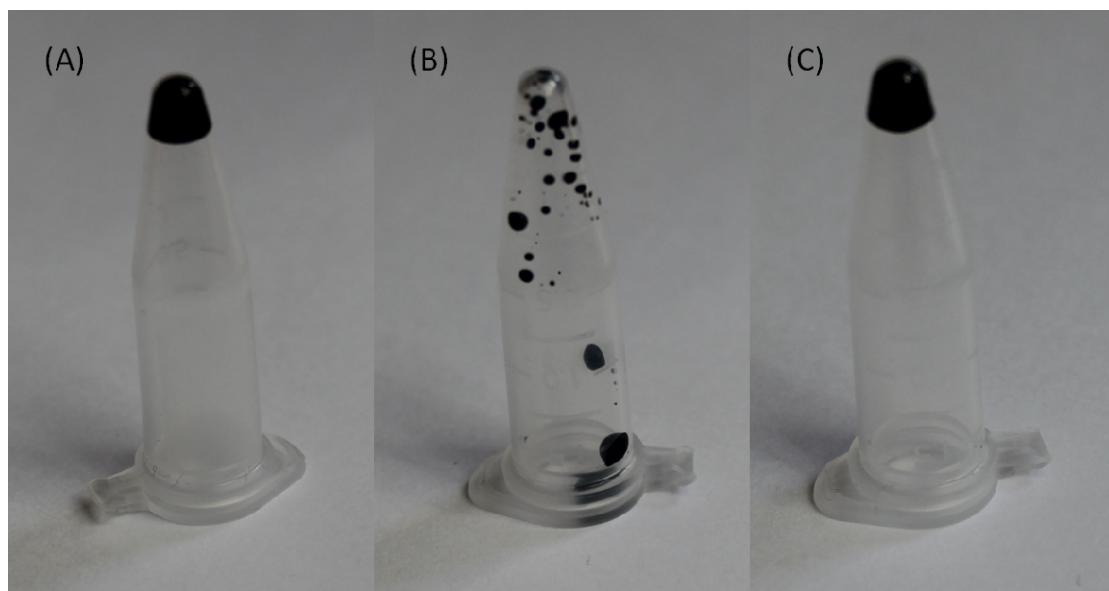
Briefly, up to 30 mM MgCl<sub>2</sub> was added to the DNA and SWNT mixture to facilitate the precipitation of the SWNT-DNA conjugates (SWNT-[C,D] or SWNT-[C,E]) by a centrifugation at 2,000 g for 1 min. The supernatant mainly containing free DNA molecules was removed. The precipitate corresponding to the SWNT-DNA conjugate was redispersed in 50 mM MES buffer containing 50 mM NaCl.

### **SWNT aggregation driven by i-motif formation:**



**Fig. S1** The pH of carbon nanotube solution in 50 mM MES containing 50 mM NaCl was adjusted from 8.0 to 5.0 by the addition of HCl. After incubation for several hours, the SWNT-[C,D] would aggregate under a 2,000 g centrifugation for one minute while the SWNT-[C,E] was in dispersed state after the same treatment.

### Gel to sol transition picture



**Fig. S2** The optical image of the gel-sol transition: (A) 40  $\mu\text{L}$  solution composed of linear unit at the concentration of 1 mM and crosslinking unit at the concentration of 1.5 mg/L was placed in a micro-centrifuge tube. When the environmental pH was 5.0, the system showed a hydrogel state. (B) By changing the environmental pH value to 8.0, the system was transformed to solution in less than a minute. (C) When the environmental pH was switched to 5, the system could be reversed to gel state.