Supporting information

Materials

The as-prepared SWNTs (HiPCo) were purchased from Carbon Nanotechnologies, Inc.,（Houston, TX). All HPLC-purified DNA were purchased from Sangon Corp. Shanghai, China. The sequences of DNA are listed in Table S1. Different concentrations of FAM labeled probe DNA and target DNA were all prepared in phosphate buffer (pH = 7.4, containing 10 mM Na_2HPO_4, 2.0 mM KH_2PO_4, 2.5 mM Mg^{2+}, 140~300 mM NaCl). Exonuclease III from E. Coli were purchased from New England Biolabs (Ipswich, MA, USA) and used without further purification. N,N-dimethyl formamide (DMF) was purchased from Sigma-Aldrich. Milli-Q water (18.2 MΩ-cm) was used throughout the experiments.

General Procedure

Firstly, the as-prepared HiPCo SWNT (5mg) was sonicated in 5mL DMF for 5 h to give a homogeneous black solution. An aliquot of the SWNT suspension was added to a phosphate buffer containing the FAM labeled probe DNA (20nM to 500nM) and the mixed solution was set for ten minutes. The volume of SWNT in DMF is less than 5% in the whole mixed solution and so DMF has no influence on the following experiments. The fluorescence of DNA probe was completely quenched by the noncovalent assembled SWNT. And then, the complementary target with different concentrations was added to the SWNT-DNA solution and was incubated to hybridize for 3h at room temperature (or 37℃). The small fraction of quenched fluorescence of FAM-labeled probe was recovered because dsDNA has weaker affinity with SWNT than ssDNA probe. After that, 50unit Exonuclease III was added to the hybridized SWNT-DNA solution and incubated at 37℃ for 2h. The hybridized and enzymatic catalyzed SWNT-DNA solution was then centrifuged at 12000 rpm for 1h and the pellet comprising of impurities, aggregates, and bundles of nanotubes at the bottom of the centrifuge tube were discarded, and the supernatant was collected for the following fluorescence measurement.

To study the effect of SWNT amount on the quenching efficiency for ssDNA, dsDNA and FAM-TTT, oxidizing acid treated SWNT was used. SWNTs were treated by refluxing in 3:1 concentrated H_2SO_4:HNO_3 mixture for 24 h, then were filtered with a 220 nm millipore size membrane with the aid of a pump and thoroughly washed with water to obtain a neutral state. Finally, they were dried under vacuum at 60℃ overnight. The obtained SWNTs was highly hydrophilic and can be well dispersed in PBS buffer.

Detection method

Fluorescence of the DNA solution was measured on Cary Eclipse spectrofluorometer (Varian, USA) using a 100 μL quartz cuvette. Excitation was set at 490 nm and emission was monitored from 510~600 nm with the excitation slit and emission slit both at 10 nm.

Electrophoresis was performed with 18% PAGE in 1×TBE buffer (pH 8.3) at 80V for about 2 h. In order to avoid the interference of SWNT due to its big size and quenching property, gel electrophoresis was conducted on free antiP before and after reacted with antiT target DNA and Exo III. The concentrations of and antiT DNA were both 5uM and so the fluorescence is strong enough to visualize under the UV lights with no need to add extra fluorescence dye for imaging.

Figure S1

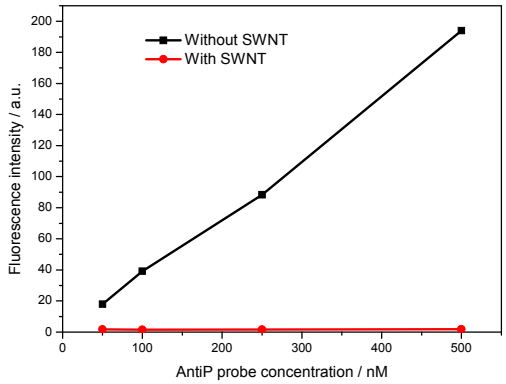

Figure S1 Fluorescence emission intensity of free antiP pobe and antiP with SWNT in the concentration rang of 50~500 nM in 0.20 M NaCl PBS. The quenching efficiency of SWNT for FAM labeled DNA is nearly 99%. The fluorescence was measured at λ_{ex}=490 nm, λ_{em}=520 nm, slit=10 nm, PMT voltage = 800 V.

Figure S2 (A)

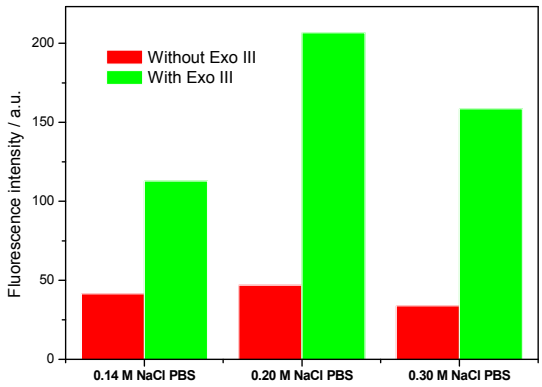

Figure S2 (B)

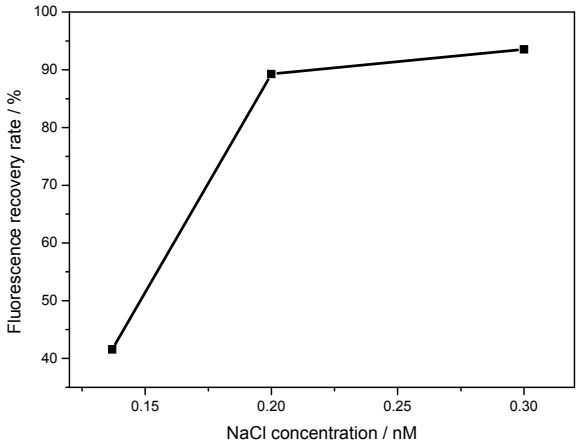

Figure S2 The influence of NaCl concentration (0.14 M, 0.20 M and 0.30 M) in PBS buffer: (A) The fluorescence

intensity of antiP probe hybridized with antiT target with and without Exo III. (B) The relationship between fluorescence recovery and the NaCl concentration in PBS buffer. The DNA hybridization is conducted at 37℃ for 3hours and the enzyme reaction is conducted at also 37℃ for 2 hours. The antiP probe concentration is 50 nM and the antiT target concentration is 1μM. The fluorescence recovery rate is defined as the ratio of the fluorescence intensity in the presence of 50 unit Exo III enzyme and target to the fluorescence of free antiP in the corresponding PBS buffer.

Figure S3

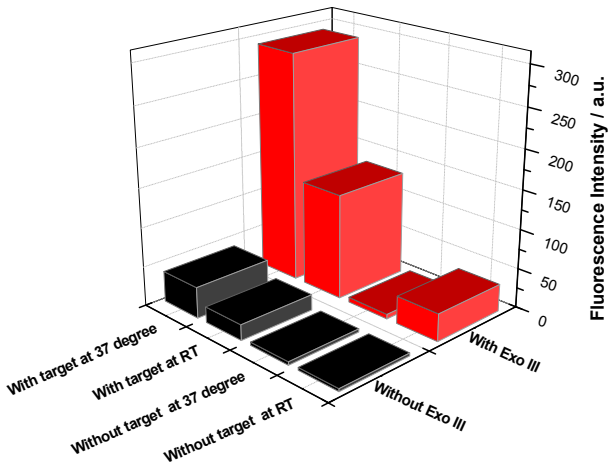

Figure S3 The influence of hybridization temperature. The DNA hybridization is conducted at 37℃ for 3hours and the enzyme reaction is conducted at also 37℃ for 2 hours. The antiP probe and antiT target concentration is 50 nM and 1 μM, respectively, and the Exo III amount is 50 unit.

Several important experimental conditions of DNA hybridization and enzyme catalyzed reaction were also optimized. The influence of NaCl concentration in PBS buffer was shown in Figure S2. Higher NaCl concentration resulted in higher DNA hybridization efficiency and higher fluorescence recovery rate due to the screen of negative charges of ssDNA by high ionic strength. Although the highest fluorescence recovery (93.52%) was obtained in the 0.30 M NaCl PBS, the fluorescence intensity in 0.20 M NaCl PBS is the highest after Exo III catalyzed amplification. The temperature of DNA hybridization and enzyme catalyzing also plays a prominent role in the signal amplification (Figure S3). The DNA hybridization and enzyme catalyzing efficiency at 37 ℃ is much higher than at room temperature. Although higher temperature will accelerate the hybridization rate of ssDNA, the optimum temperature of the enzyme Exo III is 37 ℃. So in the following experiments, to obtain the maximum signal amplification, the hybridization and enzyme catalyzing temperature were both set at 37 ℃.

Table S1 Synthesized oligonucleotides in the experiment

Name	Sequence (5'→3')
antiP	FAM-TTTTTGGAGCGCGCGGCATCGCGGG
antiT	CCCGCGATGCCGCGCGCTCCAATTT
antiT1mis	CCCGCGATGGCGCGCGCTCCAATTT
antiT3mis	CCCGGGATGGCGCGCGGTCCAATTT
TTTFAM	FAM-TTT
HIVP	TTTAGTCAGTGTGGAAAATCTCTAGC
HIVT	GCTAGAGATTTTCCACACTGACTTTTTT