

Supporting Information

DNA analysis based on the toehold-mediated strand displacement on graphene oxide

Takaaki Miyahata,^a Yusuke Kitamura,^{*a,b} Akika Futamura,^a Hirotaka Matsuura,^a Kazuto Hatakeyama,^a Michio Koinuma,^{a,b} Yasumichi Matsumoto,^{a,b} and Toshihiro Ihara^{*a,b}

^aDepartment of Applied Chemistry and Biochemistry, Kumamoto University, 2-39-1 Kurokami, Chuo-ku, Kumamoto 860-8555, Japan

^bCREST, Japan Science and Technology Agency, 7 Gobancho, Chiyoda-ku, Tokyo 102-0076, Japan

Oligonucleotides. ODNs were synthesized by a conventional phosphoramidite method on a universal CPG (controlled pore glass) column (Glen Research). Cleavage from the CPG support and deprotection was carried out by the incubation in methylamine (40 %) for 8 h at 55 °C. The aqueous methylamine was evaporated under reduced pressure. From these crude mixtures, the resulting ODNs were purified with conventional two-step procedure using RP-HPLC (linear gradient: 0.1 M triethylammonium acetate (TEAA), pH7.0/acetonitrile) and identified with MALDI-TOF/MS.

Fluorescence measurement. Fluorescence titration experiments were carried out at 25 °C by the addition of increasing amount of GO (0 – 9.6 µg/mL) to the 50 nM probe (or probe/capture duplex) in the 5 mM sodium phosphate buffer solution (pH 7.0) containing 500 mM NaCl, and 2 mM MgCl₂. The fluorescence spectra were collected following the addition of GO and 3 min incubation. The fluorescence recovery experiments were conducted in the same buffer solution as described above. To the solution of 50 nM probe or probe/capture duplex, the GO was added to the point where FAM was completely quenched (the final concentrations of GO were 6.0 µg/mL and 9.6 µg/mL, respectively). Fluorescence spectra were measured at 5 °C after the addition of 100 nM of several targets (Table S1) accompanying 10 min incubation. The emission intensities (517 nm) obtained in the presence of several targets were shown in Fig. S1.

Table S1. Sequences of several targets.

Targets	Sequences*
fmDNA	5' CAGACCGGGGACACA 3'
misDNA1A	5' CAGA A CGGGGACACA 3'
misDNA1G	5' CAGA G CGGGGACACA 3'
misDNA1T	5' CAGA T CGGGGACACA 3'
misDNA2C	5' CAG C CCGGGGACACA 3'
misDNA2G	5' CAG G CCGGGGACACA 3'
misDNA2T	5' CAG T CCGGGGACACA 3'
misDNA3C	5' CAC A CCGGGGACACA 3'
misDNA3A	5' CAA A CCGGGGACACA 3'
misDNA3T	5' CAT A CCGGGGACACA 3'

* Bold letters denote bases that are not complementary to the probe.

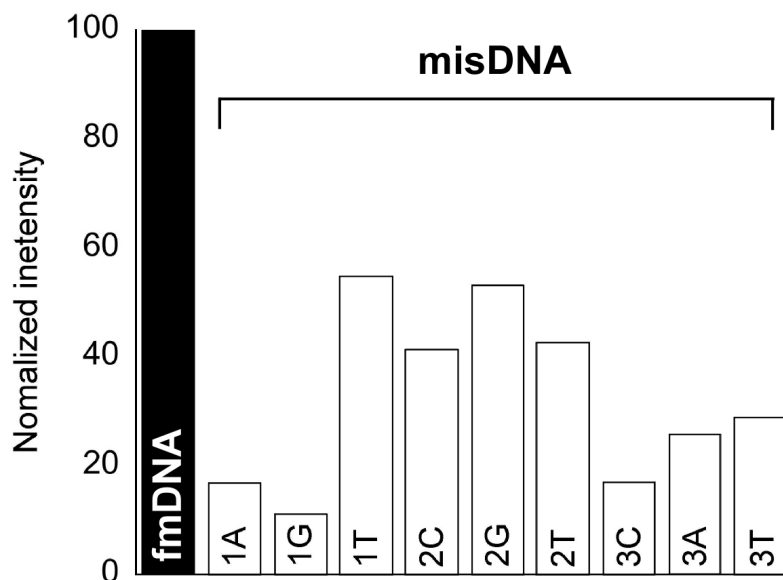


Fig. S1. Normalized emission intensities of the solution containing probe/capture DNA duplex and GO in the presence of several targets.

Sensitivity. Sensitivities of both indirect and direct systems were assessed by titration curves for target DNA. The probe (or probe/capture DNA duplex) adsorbed GO was titrated with **fmDNA**. The measurement of fluorescence intensities (517 nm) were conducted in the same buffer solution as described above at 5 °C after the 90 min incubation. The titration curves were shown in Fig. S2. Each plot was obtained as an average value for three separated measurements.

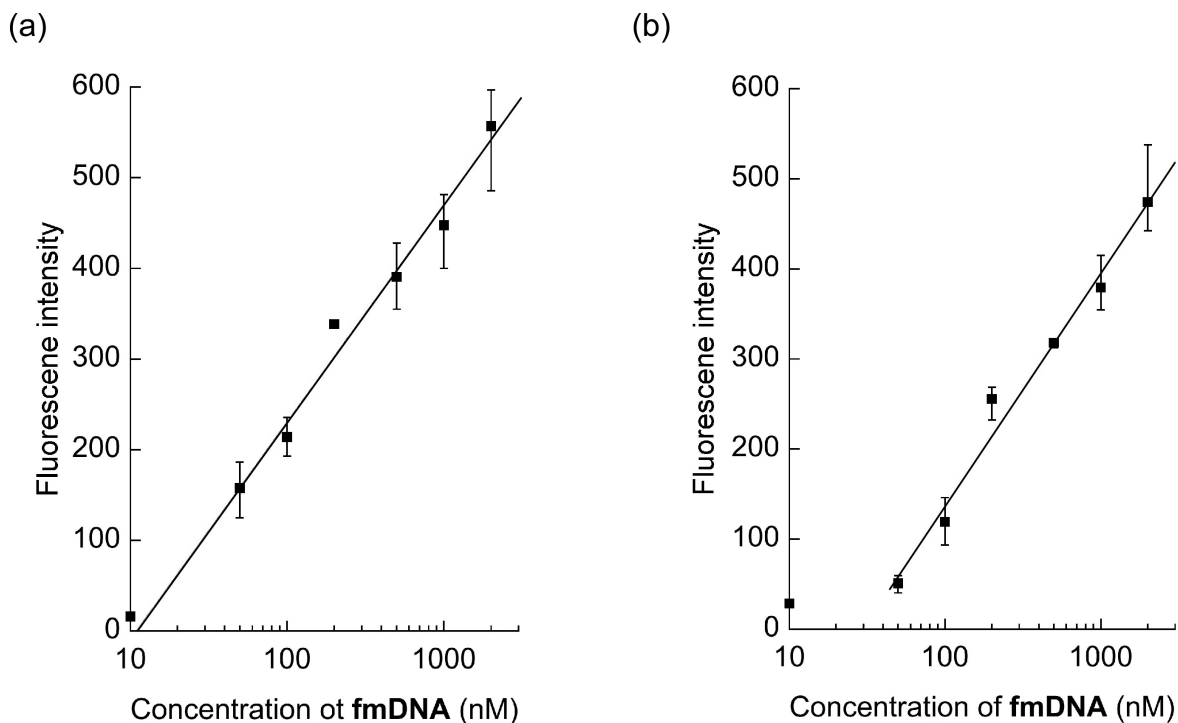


Fig. S2. Standard curves for the (a) indirect and (b) direct system.

AFM image. AFM images were obtained using Nanoscope V (Digital Instruments) in the tapping mode, where the samples were prepared by dipping the suspension on a mica substrate.

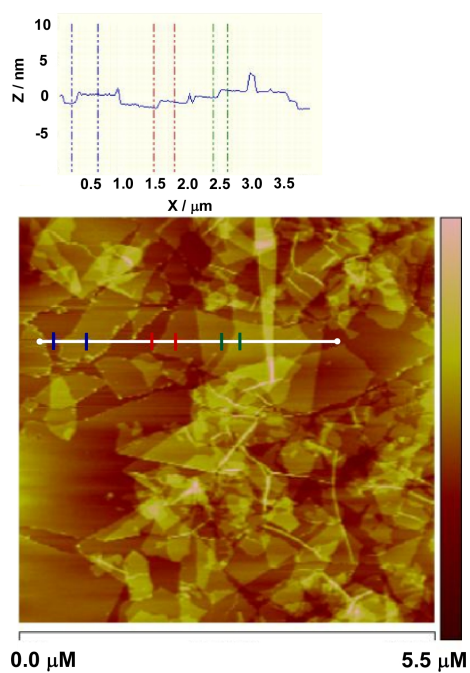


Fig. S3. AFM image and cross-section analysis of GO sheets deposited on a mica substrate.