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PNA-Assembled Graphene Oxide for Sensitive and Selective Detection of DNA

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Abstract

DNA detection based on peptide nucleic acid (PNA)-DNA hybridization is emerging as an important method in the area of DNA microarrays and biosensors because PNA shows remarkable hybridization properties. In this work, we provide a novel, simple, sensitive, and selective strategy based on a PNA-graphene oxide (GO) assembled biosensor for fluorescence turn-on detection of DNA, in which the nano nova-GO was used as a scaffold for PNA and a quencher for the fluorophore. The PNA-GO assembled biosensor is capable of distinguishing sequence specificity including complementary, one-base mismatched and non-complementary targets. Moreover, the results show that the biosensor is able to detect target DNA down to hundreds of picomolar. This sensing platform has been demonstrated to be highly sensitive and specific, and we expect that it will find great applications in the field of biomedicine and disease diagnostics.

Introduction

Graphene is a novel single-atom-thick and two-dimensional carbon material with extraordinary electronic, mechanical and thermal properties.^{1,2} Despite its short history, the discovery of graphene has aroused diamond fever in the world scientific community. Specifically, graphene oxide (GO), an oxidized version of graphene, which displays good water-solubility and flexible modification due to the massive suspended hydroxyl and carboxyl groups present on the surface³ has also attracted great interest in biological and biomedical application areas.

In view of GO's properties of long-range nanoscale energy-transfer and adsorption with nucleobases or aromatic compounds noncovalently via π -stacking interaction,⁴⁻⁶ a number of sensors have been developed to detect nucleic acids,⁷⁻¹⁰ metal ions,¹¹⁻¹³ enzyme activity,^{14,15} and medicines.¹⁶ A typical ssDNA-GO assembled biosensor for DNA detection was developed by Lu and co-workers.⁷ The fluorescence was quenched upon physical adsorption of dye-labeled ssDNA probe on GO, whereas the fluorescence was recovered while the probe was hybridized with complementary target DNA and desorbed from the GO surface. Meanwhile, a new method of allowing multicolor DNA analysis based on GO was reported by He and co-workers.¹⁷ The multicolor fluorescence was quenched simultaneously while the large planar surface of GO adsorbed multiple DNA probes labeled with different dyes, allowing the rapid, sensitive and selective detection of multiple DNA targets in the same solution.

Almost all these sensors based on GO platform were projected using ssDNA as

a probe. Peptide nucleic acid (PNA) is an analogue of DNA in which the negatively charged sugar-phosphate backbone is replaced with an uncharged pseudo-peptide chain, thus resulting in a net neutral mimic.^{18,19} Owing to the special structural framework, it has been reported to possess more remarkable hybridization properties than DNA, such as faster, higher stability and higher sequence selectivity.¹⁹⁻²² PNA has thus been used as a probe molecule for detection of nucleic acids using microarrays and biosensors. Specific detection of DNA or RNA based on PNA-DNA/PNA-RNA hybridization was realized by conventional fluorescence-based techniques,²³ surface plasmon resonance (SPR),^{24,25} and silicon nanowire-based biosensor.^{26,27} However, these techniques require fabricated chips for sensing, which are expensive and tedious.

Very recently, the interactions of DNA and synthetic DNA mimics (PNA, PCNA) with GO has been studied by Vaijayanti Anil Kumar and co-workers.²⁸ In that paper, they demonstrate that a modified DNA backbone such as D-PCNA which is chiral and uncharged is better as a probe for DNA detection than natural DNA or PNA. Furthermore, DNA detection by using fluorescent dye-labeled PNA as a probe on GO sheet is not realized because PNA/DNA duplex can not be desorbed efficiently from the GO surface at room temperature. In this work, we present successful DNA detection with the platform of GO based on PNA-DNA hybridization. GO was synthesized using a simple traditional method, after which the dye-labeled PNA was physically adsorbed on GO, resulting in fluorescence quenching. The fluorescence signal was restored again while specific target DNA was hybridized with PNA under

the optimal reaction conditions and PNA was released from the GO sheets. The detection of DNA was then realized by the PNA-GO assembled biosensor. The analytical method only involves mixing GO with PNA, subsequently DNA, thus the principle of this PNA-GO assembled biosensor is very simple compared to other biosensor-based protocols. What is more, the biosensor shows good sequence specificity to target DNA and high sensitivity up to hundreds of picomolar. The method could be simple, fast, specific and sensitive for detection of DNA by monitoring the change of fluorescence signal.

Experimental section

Materials and reagents

The PNA probe utilized was synthesized by Bio-Synthesis, Inc. (Lewisville, Texas), the DNAs were purchased from Takara Biotechnology Co.Ltd. (Dalian, China), which were purified by HPLC. Each of PNA and DNAs was 22-mer in length. The sequence of the fluorescent dye-labeled PNA (P1) is N-AACCACACAACCTACTACCTCA-Lysine (FAM)-C and the sequence of the fluorescent dye-labeled ssDNA (P2) is the same as PNA's: 5'-AACCACACAACCTACTACCTCA-FAM -3'. The sequences of the DNAs employed for hybridization are 5'-TGAGGTAGTAGGTTGTGTGGTT-3' (complementary, T1), 5'-TGAGGTAGTAGGATGTGTGGTT-3' (one-base mismatched, T2), 5'-ATGCATGCATGCATGCATGCAA-3' (non-complementary, T3), respectively. Graphite powder (99.99995%, 325 mesh) was purchased from Alfa Aesar.

4-(2-Hydroxyethyl)-1-piperazineethanesulfonic acid (HEPES), KMnO_4 , NaCl and other chemicals were purchased from Generay Biotech Co. Ltd. (Shanghai, China). Ultrapure water obtained from a Millipore water purification system (18.2 M Ω resistivity, Milli-Q Direct 8) was used in all runs.

Instruments

The Fourier transform-infrared spectroscopy (FT-IR) spectrum of GO was collected with a Nicolet Nexus-6700 spectrometer (Thermo Scientific), and the Raman spectrum was recorded by a DXR Raman Microscope (Thermo Scientific). Subsequent fluorescence measurements were carried out on a Hitachi F-4600 spectrophotometer (Hitachi Co.Ltd., Japan) equipped with a Xenon lamp excitation source. The excitation was set at 495 nm and the emission was monitored at 520 nm.

Synthesis of Graphene Oxide (GO)

GO was synthesized by Hummer's method with some modifications.²⁹ Graphite powder (1.0 g) was put into 0□ concentrated H_2SO_4 (23 mL), followed by gradual addition of KMnO_4 (3.0 g) and kept at the reaction temperature below 20□ by ice bath. Afterwards, the mixture was stirred at 35□ for 2h, at which water (80 mL) was added slowly, and stirring was continued for 3h at room temperature. Additional water (125 mL) was added to end the reaction. Subsequently, adequate H_2O_2 (30%) were added drop by drop and the color of the mixture changed to bright yellow. The mixture was filtered and washed with 10% HCl solution (v/v 1:10) to remove metal ions, and then washed with lots of water repeatedly until the solution reached a neutral pH. The resulting solid was dried in air. Finally, the synthesized product was further purified

by dialysis for 1 week to remove the remaining metal species, and dispersed in water under sonication for 4 h to get the homogeneous GO suspension (200 $\mu\text{g/mL}$). The Raman spectrum and FT-IR spectrum were used for the characterization of as-prepared GO. (See ESI, Fig. S1-S2)

Fluorescent DNA assay

In a typical DNA assay, the working solution of P1 (5 μL , 10 μM) was diluted with 980 μL of HEPES buffer (25 mM, 100 mM NaCl, pH 7.4), followed by addition of 5 μL of GO aqueous solution (200 $\mu\text{g/mL}$) and reacted at room temperature for 1 min. After that, different concentrations of target (10 μL) were added to the above system, making the final volume 1 mL, and incubated at 45°C for 40 min. Finally, the fluorescence of the mixture was measured.

Results and discussion

GO-based fluorescent DNA assay

Scheme 1 depicts this new detection platform. First, the fluorescent dye-labeled PNA is physically absorbed onto GO sheet, accompanied with the quenching of the dye fluorescence. However, in the presence of target DNA, the adsorbed PNA can be desorbed from the GO surface by Watson-Crick specific hydrogen bonding with its complementary DNA, and then the initially quenched fluorescence can be recovered. Hence DNA is detected based on the fluorescence change before and after PNA-DNA hybridization in the solution of GO.

Subsequently, experiments were performed to demonstrate the ability of the

PNA-GO assembled biosensor for detecting DNA. Fig. 1a shows the fluorescence emission spectra of P1 (PNA) in the presence of GO and complementary DNA (T1). The fluorescence spectrum of P1 in HEPES buffer without GO showed strong fluorescence emission intensity because of the presence of the fluorophore-FAM. Upon addition of GO, the fluorescence intensity of P1 decreased. The intensity decreased along with the increasing concentration of GO (data not shown). It was found that over 98% fluorescence was quenched instantly in the presence of 1.0 $\mu\text{g/mL}$ GO. In this work, 1.0 $\mu\text{g/mL}$ GO was selected for the following analysis. However, after T1 was added to the P1-GO complex and incubated under the optimal conditions, an obvious fluorescence-signal enhancement was observed, resulting from the formation and release of PNA/DNA duplex from the GO surface.

In contrast, we treated P2 (DNA) with the same GO concentration (1.0 $\mu\text{g/mL}$) and the same reaction buffer. The fluorescence quenching of P2 was found to be much less (~12%) (Fig. 1b). After T1 was added to the P2-GO complex, nearly no fluorescence change was found. Compared to PNA, DNA is not quenched completely using the same amount of GO. In such a case, the un-adsorbed DNA is free in the buffer solution. The target T1 is preferentially hybridized with DNA free in the buffer solution rather than DNA adsorbed on the surface of GO. As a result, the fluorescence intensity does not change. The results indicate that PNA has a stronger binding capability to GO than DNA does. On the other hand, it is well-understood that this high fluorescence quenching ability was contributed to the strong adsorption of P1/P2 on GO surface via π -stacking attraction and the electronic transference between P1/P2

and GO. However, the different quenching efficiency between P1 and P2 could be caused by their different backbone characters. P1 with its net neutral backbone interacts more competently with GO compared to P2 with the negatively charged sugar-phosphate backbone. That is to say, P2 would be repelled by the electrostatic repulsion between itself and the negatively charged GO, but P1's neutral backbone eliminates the electrostatic repulsion, resulting in faster and higher adsorption efficiency.

Effect of temperature on detection

It has been reported in recent literatures that PNA/DNA duplex may have additional hydrogen-bonding interactions through amides and π -stacking interactions with the surface of GO.^{28, 30, 31} Compared with dsDNA, PNA/DNA duplex is not desorbed efficiently from the GO surface at room temperature because of the absence of electrostatic repulsion which is caused by the negatively charged sugar-phosphate backbone with the negatively charged GO. This does not lead to a fluorescence restoration and an effective detection. However, we assume that the interactions would be damaged by increasing the temperature and PNA/DNA duplex can be released from the GO surface in view of high melting temperature (T_m) of PNA/DNA duplex. Therefore, high temperature is expected to facilitate adsorption. Although higher reaction temperature promotes desorption effectively and achieves higher fluorescence signal, high temperature would induce dissociation of PNA/DNA complex if the temperature is above T_m . On the other hand, PNA would be desorbed from the GO surface if the temperature is too high, making higher noise.³² Therefore,

choosing an appropriate reaction temperature is very important.

PNA/DNA duplex usually has a higher T_m than DNA/DNA duplex because PNA has a flexible uncharged peptide backbone. As reported,^{33,34} the predicted T_m of PNA/DNA duplex we used is 70.5°C. In this work, thermal dissociation of PNA/DNA duplex from the GO surface was conducted based on PNA/DNA duplex's good thermostability, and the fluorescence response was measured at different reaction temperatures (25-50°C) in the presence of 100 nM complementary DNA (T1). Fig. 2 shows temperature-dependent fluorescence intensity of PNA associated with GO in the absence and presence of T1. When T1 was added to the PNA-GO complex and reacted at room temperature, the desorption phenomenon measured was not considerably dramatic. That is in consistent with what was discussed in the literature mentioned above.²⁸ To further assist desorption, we tried to raise reaction temperature. The enhancement of fluorescence intensity could be observed in the temperature region from 25°C to 50°C in the case that T1 was added, suggesting that PNA/DNA duplex is effectively desorbed from the GO surface. Similarly, the fluorescence also increased when the control sample without T1 was treated in the same way. This is probably because PNA is desorbed from the GO surface along with the increased temperature. As shown, the fluorescence intensity in the presence of T1 tended to balance as the temperature over 45°C, whereas the fluorescence intensity in the absence of T1 continued increasing with further increasing temperature. It was found that the maximized signal-to-noise ratio was obtained at 45°C. Thus, the temperature of 45°C was used in the subsequent work.

Kinetic behaviors of PNA-GO complex

The kinetic behaviors of the P1-GO complex with T1 as well as GO quenching reaction to P1 were studied by monitoring the fluorescence emission spectra as a function of time. Fig. 3 shows the fluorescence quenching of P1 (50 nM) in the presence of GO (1.0 $\mu\text{g/ml}$) as a function of time (curve a). Due to PNA's strong binding capability to GO as discussed above, the quenching phenomenon was found to be rather fast, it reached equilibrium in a few seconds. However, the fluorescence restoration of P1-GO complex in the presence of T1 was relatively slow and needed almost 40 min (curve b).

Sensitivity of the method towards DNA detection

To investigate the sensitivity of this assay, DNA detection was carried out with the fixed concentrations of P1 (50 nM) and GO (1.0 $\mu\text{g/mL}$). Fig. 4a shows the fluorescence emission spectra of P1-GO complex upon incubation with varying T1 concentrations. The fluorescence intensities increased remarkably as the concentration of T1 increased over the range of 0-200 nM. Fig. 4b describes the linear relationship between the relative fluorescence intensity and the different concentrations of T1. A dynamic range from 1 nM to 200 nM for T1 was achieved with a good linear relationship range until 50 nM ($y=1.59x+63.82$, $R^2=0.9831$). The detection limit of 800 pM for DNA was estimated based on the $3\sigma/S$ calculation (σ is the standard deviation for the blank solution, and S is the slope of the calibration curve), which was much lower than the previously reported ssDNA-GO complex sensing for DNA detection with a LOD of 10 nM.⁷ The RSD (relative standard deviation) was

calculated to be 4.33%, indicating a satisfactory reproducibility for fluorescent detection of DNA by using the assay. These results suggest that the proposed method is suitable for accurate quantification of DNA with a high sensitivity.

Selectivity study

The specificity of the sensing platform described herein was evaluated by a couple of control experiments. Experiments were performed using PNA-GO complex in the presence of complementary target (T1), one-base mismatched target (T2) and non-complementary target (T3) each at a same concentration of 100 nM, respectively. Meanwhile, the sample treated with buffer solution was also prepared as a blank control for comparison. Fig. 5a shows fluorescence graphs of P1/GO complex in the presence of T1, T2 and T3 targets, respectively. As illustrated, the fluorescence response to T1 is significantly higher than T2 and T3. Fig. 5b shows relative fluorescence value in the presence of T1, T2 and T3, respectively, where F refers to the fluorescence intensity in the presence of target and F_0 refers to the fluorescence intensity of blank control. The proposed system perfectly discriminated T1, T2, and T3. The relative fluorescence (F/F_0) for T1 was 2.70, while it was 1.21 for T2 and 1.03 for T3. The above data indicate that the desorption efficiency of the perfect match (T1) is remarkable and the response to T2 or T3 is much less which is almost equal to the blank control. The high sequence specificity of the results clearly demonstrate that the PNA-assembled graphene oxide could be used as a sensitive and selective sensor for target nucleic acid detection, or even be applicable for SNPs detection.

Conclusions

In this paper, we have developed a PNA/GO-based platform for the sensitive and selective detection of DNA, which relies on the strong binding ability as well as the super quenching capacity between GO and PNA. So far, this has been the first report that used the neutral PNA as a fluorescent nanoprobe for recognition of ssDNA with the platform of GO. Owing to PNA's special framework, the PNA-GO assembled biosensor shows higher detection sensitivity than the DNA-GO assembled one. Therefore, the proposed method represents our first attempt at connecting the highly promising nanomaterial GO with PNA probe for the DNA detection, showing a new navigator for the detection of nucleic acids. Since PNA interacts with DNA duplex, further work will be directed to the detection of dsDNA by formation of PNA/DNA duplex complex.

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Figure captions:

Scheme 1 Scheme for fluorescent DNA detection based on the GO platform by using PNA as the probe. FAM is the carboxfluorescein.

Fig. 1 (a) Fluorescence quenching of P1 (PNA, 50 nM) by GO and its restoration upon addition of T1. (b) Fluorescence quenching of P2 (DNA, 50 nM) by GO and its restoration upon addition of T1. All fluorescence spectra were carried out in HEPES buffer (25 mM, 100 mM NaCl, pH 7.4). The concentrations for GO and T1 were 1.0 $\mu\text{g/mL}$ and 200 nM, respectively.

Fig. 2 Fluorescence desorption of a mixture containing P1 (50 nM) and GO (1.0 $\mu\text{g/mL}$) in the presence (purplish red histogram) and absence (blue histogram) of T1 (100 nM) at different temperatures (25-50°C).

Fig. 3 (a) Fluorescence quenching of P1 (50 nM) by GO (1.0 $\mu\text{g/mL}$) as a function of time, and (b) fluorescence restoration of P1-GO complex in the presence of 100 nM T1 as a function of time.

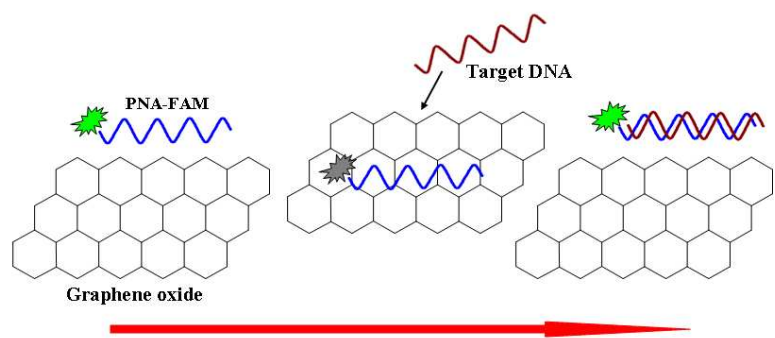
Fig. 4 (a) Fluorescence emission spectra of P1/GO complex in the presence of varying concentrations of T1 (a to k: 0, 1, 2.5, 5, 10, 25, 50, 75, 100, 150 and 200 nM, respectively).

(b) Calibration curve for the fluorescence emission spectra of P1 versus T1 concentrations.

Inset: linear responses to low concentrations of T1. Each data was measured for three independent experiments, and the standard deviation was plotted as the error bar.

Fig. 5 (a) Fluorescence emission spectra of P1/GO complex after incubation with T1

(complementary), T2 (one-base mismatched) and T3 (non-complementary), respectively. (b) The relative fluorescence response in the presence of T1, T2 and T3 targets, respectively. F and F_0 are the fluorescence intensity in the presence and absence of target.



Scheme 1

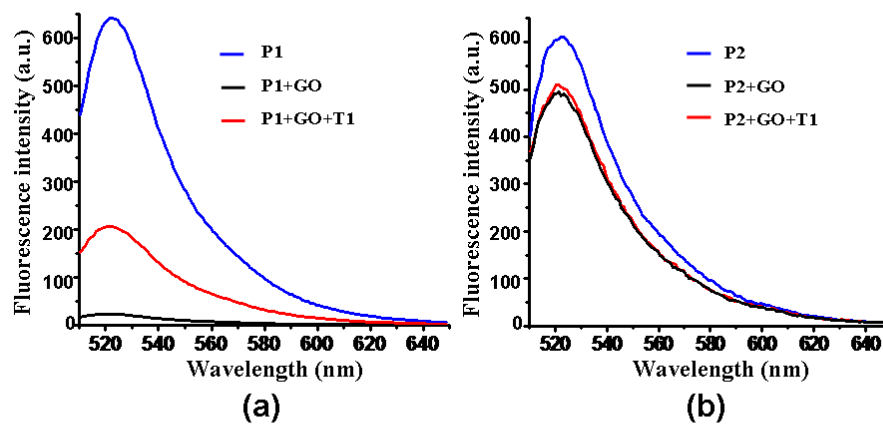


Figure 1

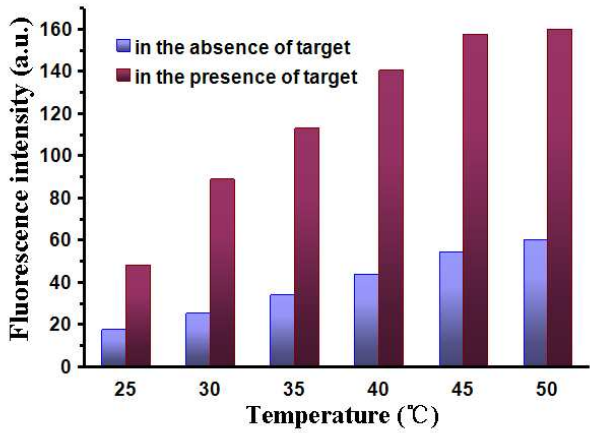


Figure 2

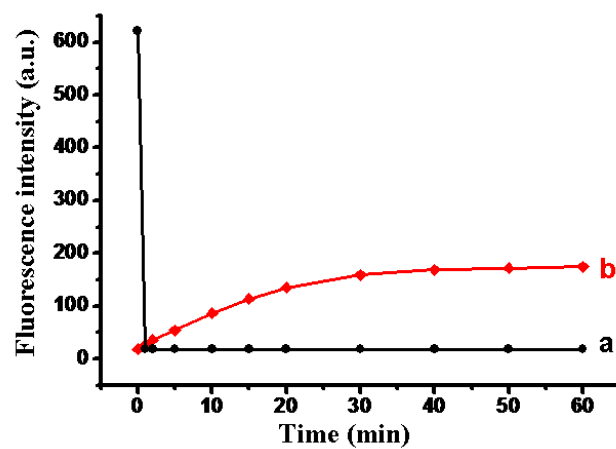


Figure 3

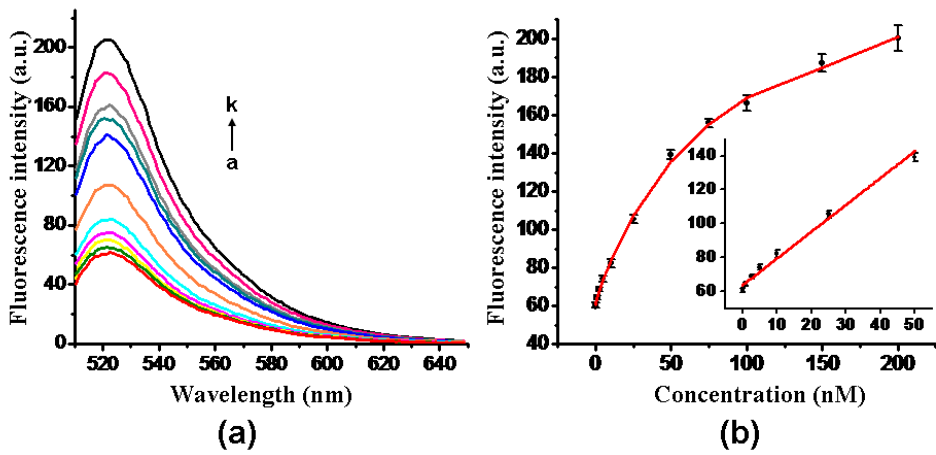


Figure 4

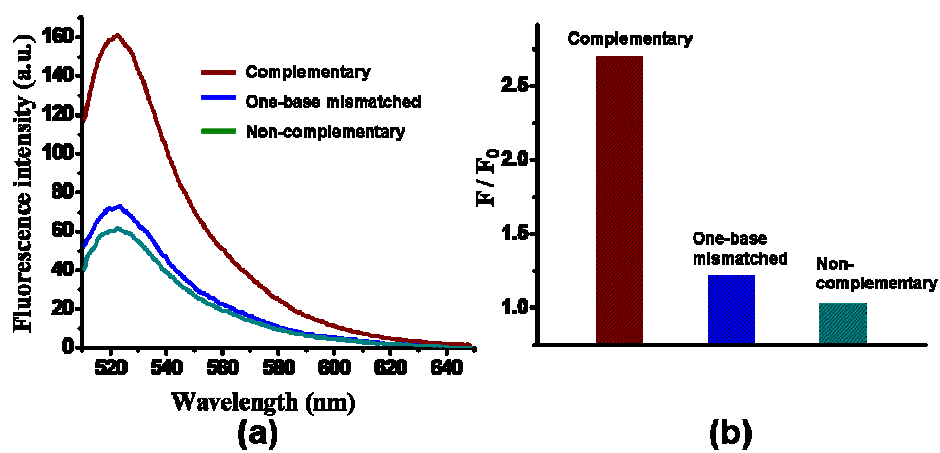


Figure 5

**PNA-Assembled Graphene Oxide for Sensitive and Selective
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