S1. Binding Affinity of CNT

We have considered a negative control experiment in order to check whether there occurs an undesirable physisorption onto carbon nanotube (CNT) surface. Here, CNTs were incubated in thrombin-free TE buffer, and thrombin-dissolved PBS buffer (with thrombin concentration of 10 nM), respectively, followed by rinsing. Fig. S3 clearly elucidates that the incubation of CNT followed by rinsing does not include any possibility in physisorption. This indicates that aptamer-conjugated carbon anotubes (ACNT)-based bioassay does not exhibit any unfavorable physisorption due to interaction between CNT and biomolecules.

S.2. Binding Affinity of Carboxylated CNT

In order to validate the possibility of undesirable physisorption due to carboxylation of CNT, we have taken into account a negative control experiment in such a way that
carboxylated CNTs were incubated in various solutions (e.g. thrombin-free TE buffer solution, and thrombin-dissolved PBS buffer solution) followed by rinsing. Here, the details of the preparation of solutions were shown in Section S.1. Fig. S4 demonstrates that carboxylation of CNT does not lead to the unanticipated physisorption in our bioassay based on incubation followed by rinsing.

**S.3. Specific Binding Affinity of ACNT**

For verification of the ability of ACNT to selectively sense target molecules, we have devised a negative control experiment such that ACNTs were incubated in each PBS buffer solution containing non-target protein molecules such as human serum albumin (HSA, ≥97 %, Sigma-Aldrich, USA), fibrinogen (≥80 % of protein is clottable, Sigma-Aldrich, USA), and transferin (≥97 %, Sigma-Aldrich, USA), respectively. Here, in each PBS buffer solution, non-target protein molecules are dissolved at a concentration of 100 nM. These prepared buffer solution (containing non-target protein) with the amount of 100 μL was dropped onto ACNT patterned substrate and maintained for 2 hours. Subsequently, ACNT reacted with non-target proteins was rinsed with deionized water and dried in vacuum for 12 hours, and then imaged by tmAFM and KPFM.

**S.4. The Effect of Salt Concentration**

For studying the effect of salt concentration on binding affinity between thrombin and DNA aptamer, we have prepared solution in such a way that thrombin molecules are diluted with concentration of 100 pM in a buffer solution, which contain sodium chloride with concentration of 0.01 M, 0.1 M, and 0.2 M, respectively.
S.5. The Effect of pH

For understanding the effect of pH on the binding affinity between thrombin and ACNT, we have prepared solutions in such a way that thrombin was diluted in PBS buffer solution whose pH is given as 6.0, 7.0, 7.4 and 8.0, respectively. Based on such prepared solutions, we have conducted bioassay and then conducted AFM/KPFM-based imaging of ACNTs incubated with such prepared solution.
Fig. S1. Illustration of fabrication of horizontally aligned carbon nanotube by chemical vapor deposition (CVD) method.
Fig. S2. Fluorescence images of (a) pristin carbon nanotube, and (b) ACNT, respectively. Scale bar is 100 μm.
Fig. S3. Negative Control Experiments for CNT: (a) tmAFM images and (b) KPFM images for primeval CNT, and CNTs incubated in thrombin-dissolved PBS buffer, and TE buffer, respectively. (c) Average heights and (d) surface potentials for such prepared CNTs. Scale bar of all images is 100 nm.
Fig. S4. Negative Control Experiment for Carboxylated CNT: (a) tmAFM images and (b) KPFM images for carboxylated CNT, and carboxylated CNTs that were incubated in thrombin-dissolved PBS buffer, HSA-dissolved PBS buffer, and TE buffer, respectively. (c) Average height and (d) surface potentials for such prepared carboxylated CNTs. Scale bar is 100 nm.