Electronic Supporting Information

Protein Recognition on Single Graphene Oxide Surface Fixed on Solid Support

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1. Average height determination from AFM topographies

We determined the average height of the piece of GO seen in Fig. 3 by undertaking a histogram analysis of the corresponding AFM topography rather than its sectional view. The results are shown in Fig. S1. We performed the histogram analyses within the rectangular area indicated by the dotted line in the topographies in Fig. S1 using Nanoscope Analysis (Bruker), the software supplied with Bruker Dimension FastScan AFM.

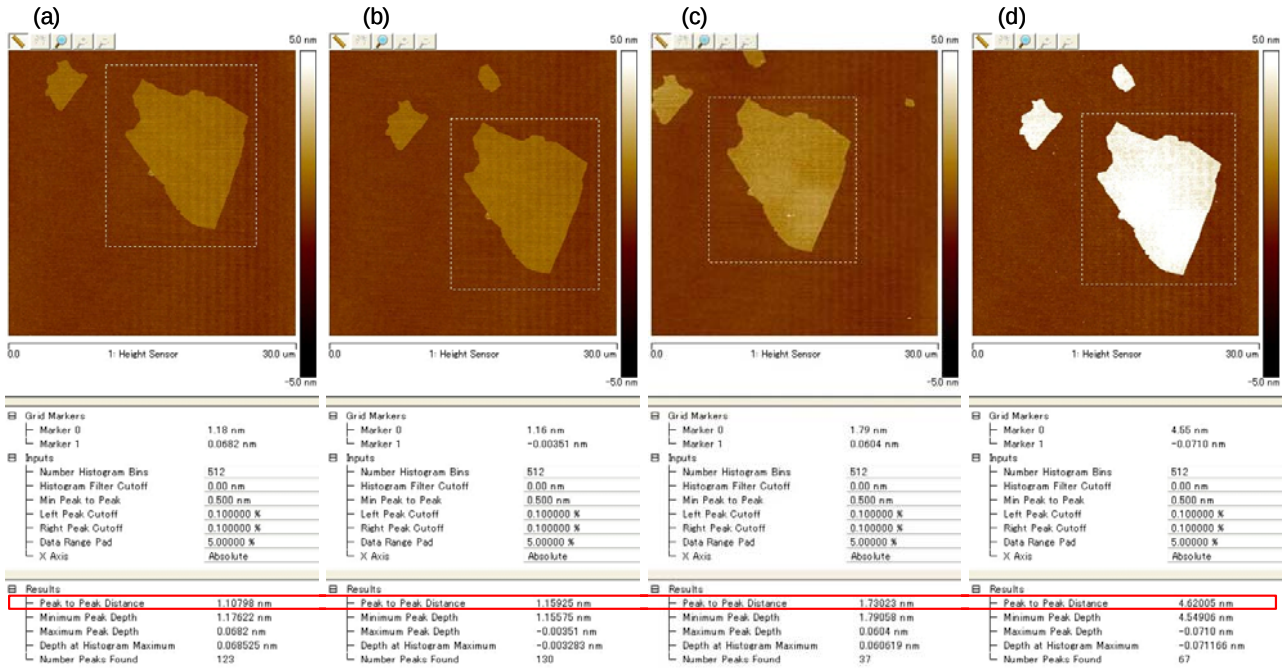

Fig. S1. Histogram analysis results for the GO piece seen in Fig. 3. The average heights of the GO piece are indicated in the red box.

2. Control experiments

We examined a similar reaction to that reported in the main text using 10 mM Tris-HCl buffer instead of phosphate buffer with the same concentration of thrombin (100 unit/mL). The chemical modification and thrombin detection were observed step-by-step using an LSM and an AFM in the same manner as described in the main text. The results are shown in Fig. S2 (LSM) and Fig. S3 (AFM).

We also examined the detection of avidin using the present GO system modified with thrombin aptamer. In this control experiment, we used a 10 mM phosphate buffer solution of avidin (1mg/mL) instead of thrombin. The LSM observations showed no difference between fluorescence intensities at the GO surface before and after avidin addition (Fig. S4). This supports the idea that the modified GO responds specifically to thrombin. The surface of the GO piece in Fig. S4 was also observed with an AFM (Fig. S5). Unlike Fig. 3 and Fig. S3, no apparent increase in the GO height could be confirmed from the AFM topography after the avidin addition. This further supports the view that the modified GO does not respond to avidin. Note that part of the piece of GO has peeled off in Fig. S5, although a piece of GO fixed on a hydrophilic SiO_2 solid support was usually stable in the operations we undertook as shown in the other results (Fig. 3 and Fig. S3). Thus we did not use the histogram analyses to determine the average GO height in this case.

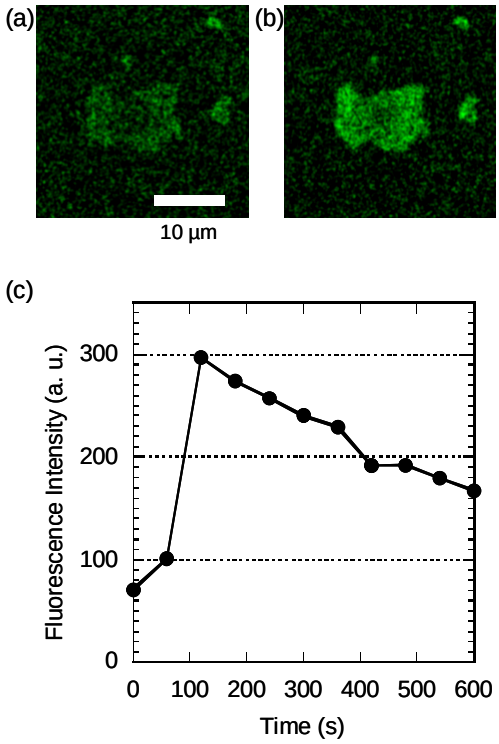

Fig. S2. Change in the fluorescence intensities of aptamer-modified single GO in Tris-HCl buffer. (a) LSM image (30 × 30 μm) of the specific GO piece before thrombin addition. (b) After thrombin addition. (c) A plot of fluorescence intensity vs. time after the thrombin addition. The fluorescence intensities are corrected by subtracting the average fluorescence intensity of the background from that of the GO area.

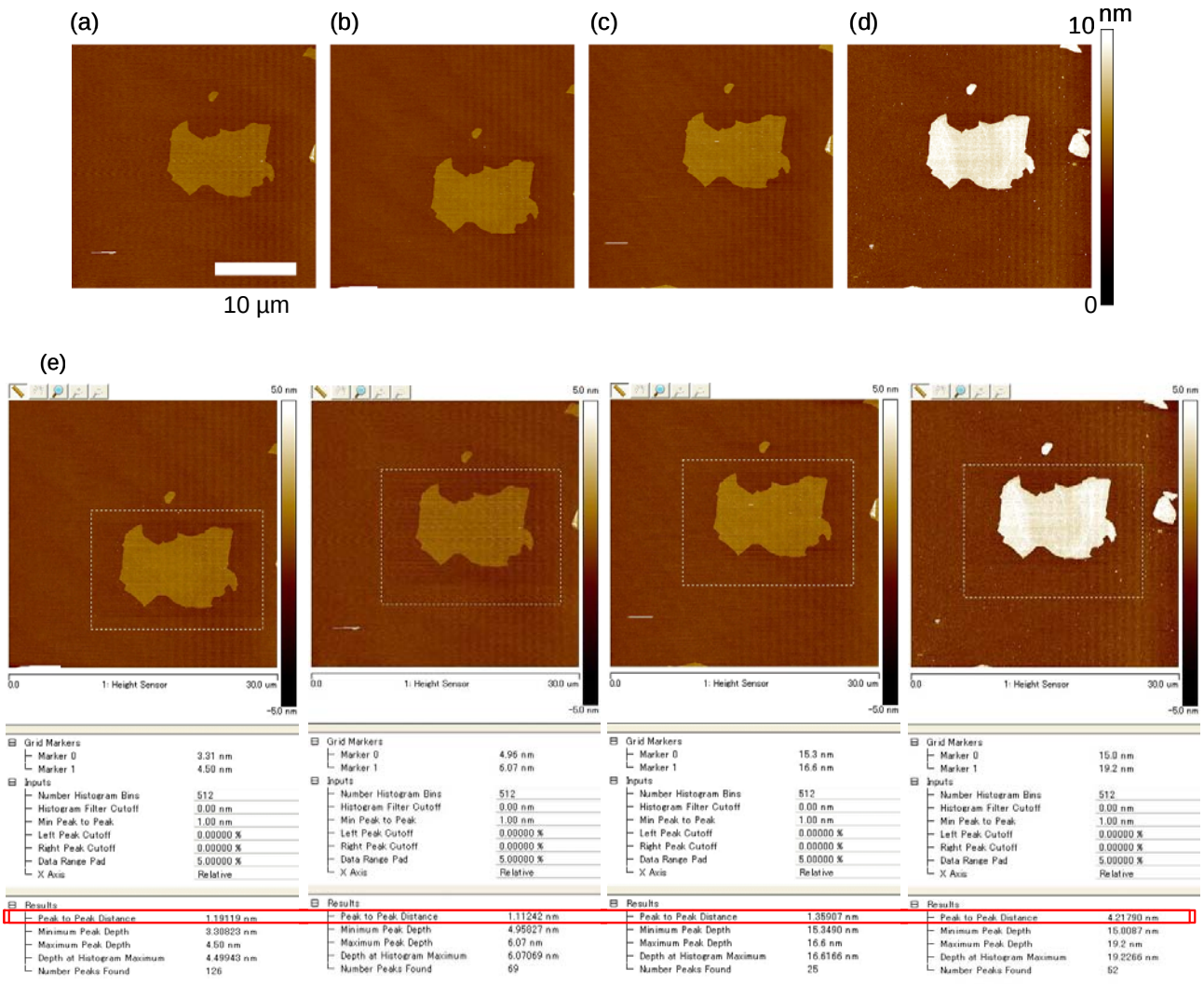

Fig. S3. AFM topographies of the GO piece in Fig. S2 observed (a) before chemical modification, (b) after modification of **1**, (c) after modification of **2**, and (d) after thrombin recognition. All the images were obtained under atmospheric conditions with an imaging area of 30 × 30 µm. (e) The average height of the GO piece was determined from a histogram analysis inside the rectangular area in each image. The initial average height of the GO piece is 1.19 nm, which is observed 1.11 and 1.36 nm respectively after the modification by **1** and **2**. After the thrombin recognition, the average height becomes 4.22 nm.

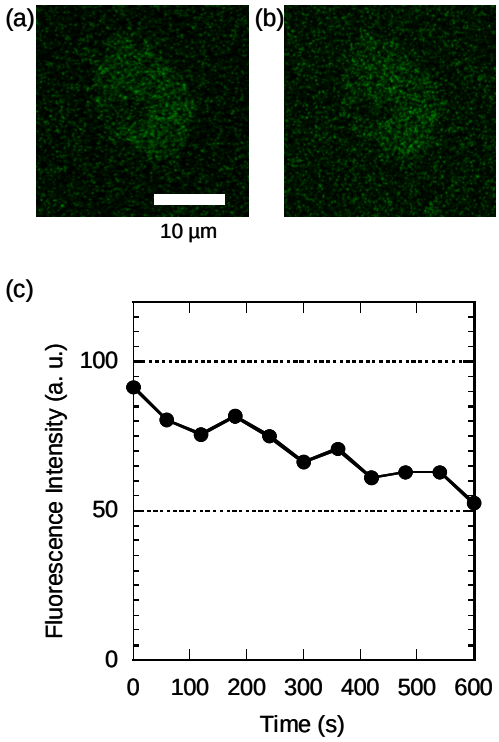

Fig. S4. Change in the fluorescence intensities of aptamer-modified single GO in PBS buffer. (a) LSM image (30 × 30 µm) of the specific GO piece before avidin addition. (b) After avidin addition. (c) A plot of fluorescence intensity vs. time after the avidin addition. The fluorescence intensities are corrected by subtracting the average fluorescence intensity of the background from that of the GO area.

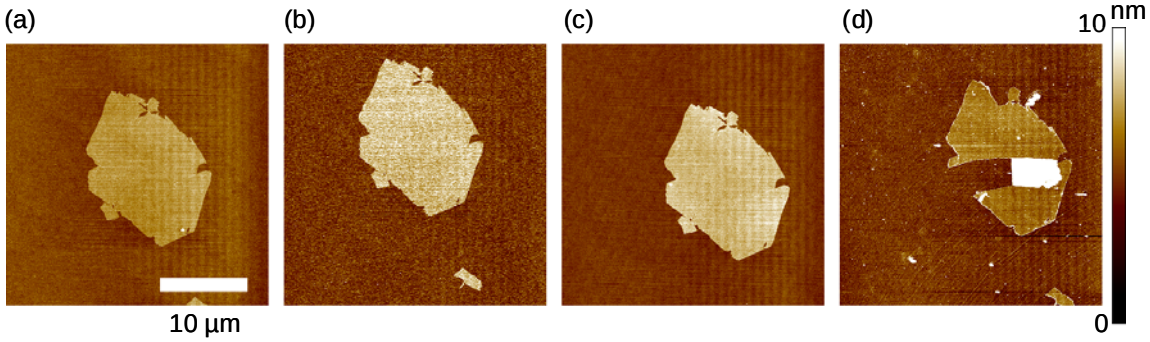

Fig. S5. AFM topographies of the GO piece shown in Fig. S4 observed (a) before chemical modification, (b) after modification of **1**, (c) after modification of **2**, and (d) after attempted avidin detection. All the images were obtained under atmospheric conditions with an imaging area of 30 × 30 µm.

3. Positions of GO pieces on coverslip mounted with a microchannel

We determined the size and shape of the GO pieces in Fig. 4 by changing the contrast and brightness of the fluorescence image as shown in Fig. S6 (a) in order to distinguish the border between GO pieces and the substrate. Fig. S6 (b) shows the result of the determination in which the GO pieces are indicated by white dotted lines. The orange square in Fig. S6 (b) corresponds to the same area in Fig. 4. The GO piece indicated by the white arrow in Fig. S6 (b) is the single piece positioned both inside and outside of the microchannel.

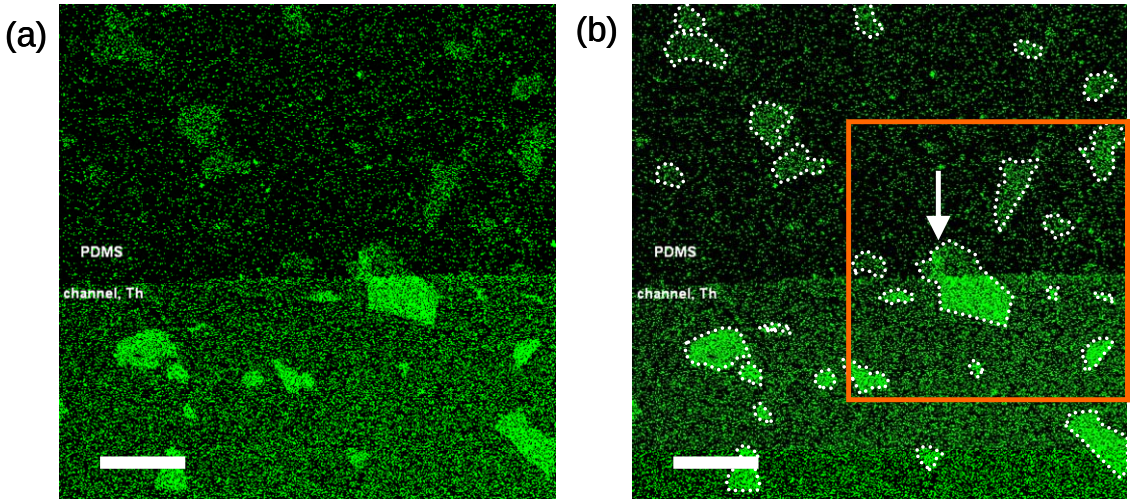

Fig. S6. (a) Fluorescence image in Fig. 4 with different contrast and brightness. Scale bar: 20 μm. (b) The same image as (a) with the white dotted lines that indicate the shape of GO pieces.