Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Self-Standing Aptamers by Artificial Defect-Rich Matrix

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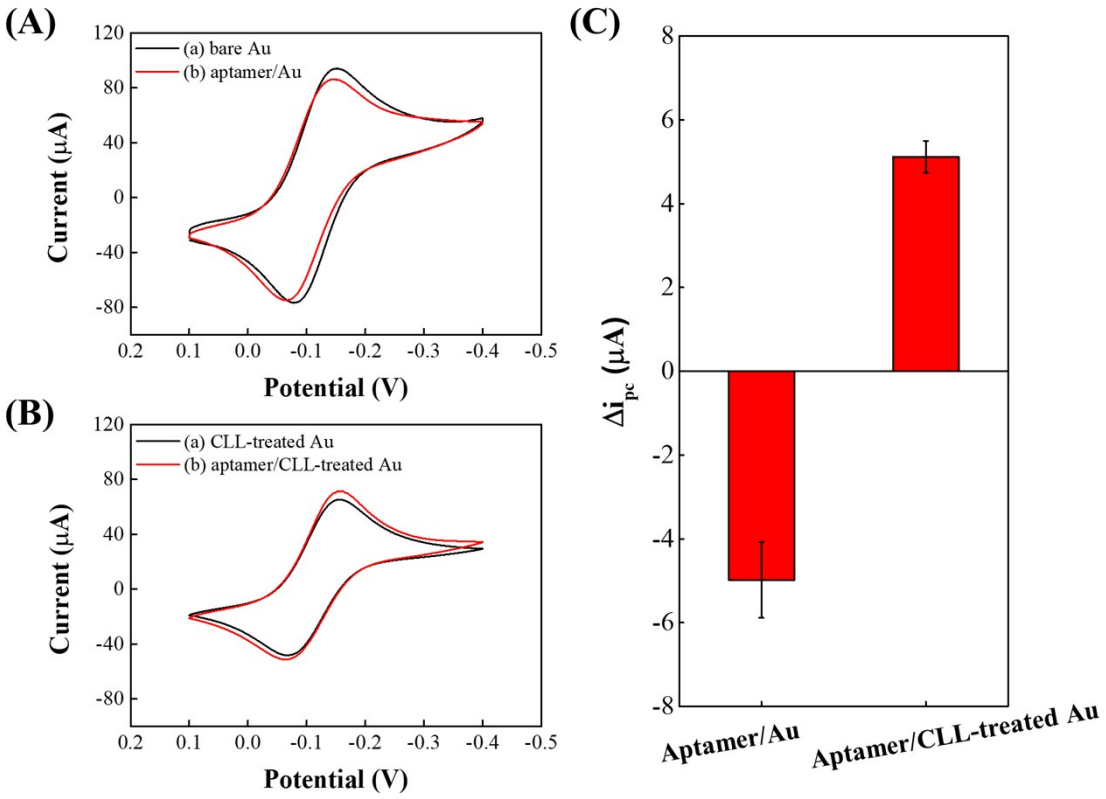

Figure S1. Cyclic voltammograms of 1 mM $[Ru(NH_3)_6]^{3+}$ on different surfaces. (A) Bare Au (curve a) and aptamer-modified Au (curve b). (B) CLL-treated Au (curve a) and aptamer-modified CLL-treated Au (curve b). These measurements were performed in 25 mM TRIS buffer at pH 7.4 with a scan rate of 100 mV/s. (C) The 1 mM $[Ru(NH_3)_6]^{3+}$ CV response change upon aptamer-modification of bare Au (A) and CLL-treated Au (B) surfaces. (N = 3) The error bars indicate the standard deviation.

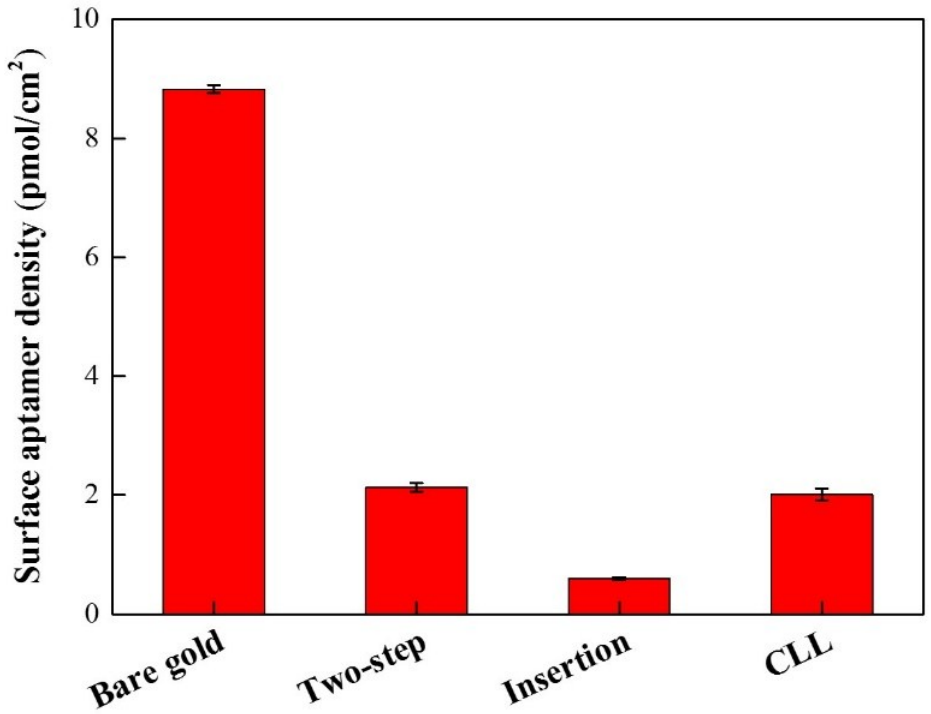

Figure S2. Surface aptamer density histograms from substrates fabricated by different processes. Each data point is obtained from the average fluorescence intensity of three samples (N = 3). The error bars indicate the standard deviation.

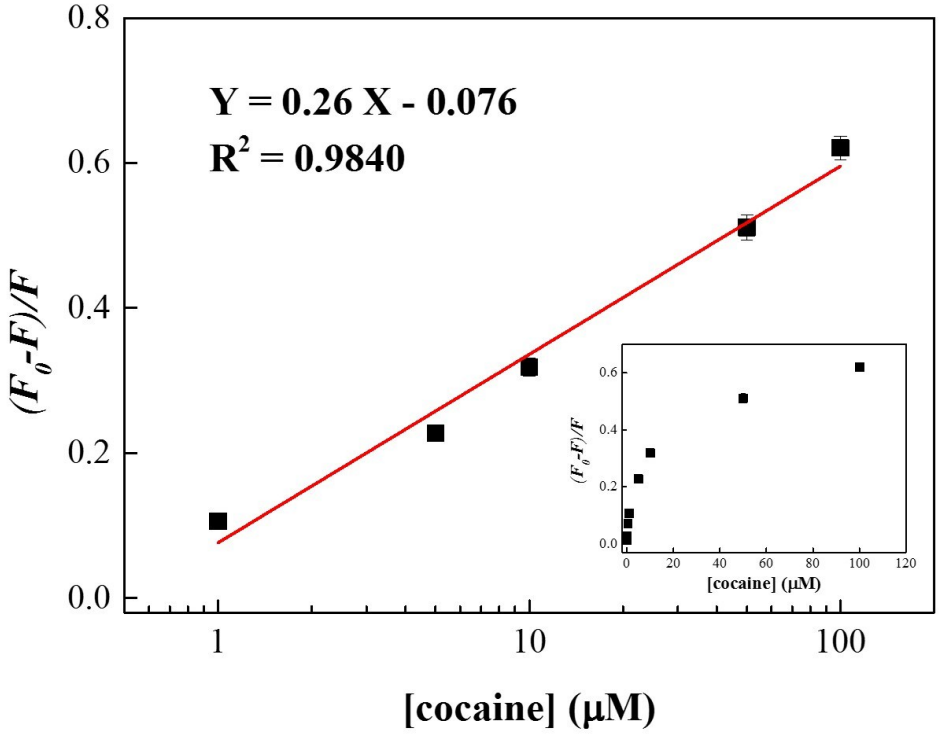

Figure S3. Linear correlation curve derived from $(F_0-F)/F$ against the logarithm of cocaine concentration and (inset) the cocaine concentration. Each data point is obtained from the average fluorescence signal of individual squares (N = 4). The error bars indicate the standard deviation.

Calculations of dissociation constants (K_d)

According to literature reports,[1-4] aptamer probe 3 can hybridize with cocaine with a molar ratio of 1:1. Therefore, the K_d can be defined as the formula given in equation (1):

$$K_d = \frac{[aptamer] \times [cocaine]}{[aptamer/cocaine]} \qquad (1)$$

where [aptamer], [cocaine], and [aptamer/cocaine] are the concentrations of unhybridized aptamer, unhybridized cocaine, and hybridized cocaine/aptamer on the gold surface, respectively. Based on 1:1 of aptamer/cocaine binding stoichiometry, when [aptamer] = [aptamer/cocaine], equation (1) is simplified to K_d = [cocaine]. In this case, when cocaine leads half the total of FAM-labeled aptamer to fluorescence quenching or half-maximal fluorescence image contrast change, K_d is equal to cocaine concentration.

As shown in Figure S1, the linear fitted equation of correlation is:

$$Y = 0.26\,X - 0.076 \qquad (2)$$

in which Y and X respectively represent the value of $(F_0\text{-}F)/F$ and the logarithm of cocaine concentration (μM). (Note: F_0 and F represent corresponding relative fluorescence intensities without and with targets, respectively.) The maximum value of $(F_0\text{-}F)/F$ (Y_{max}) is 0.62 ± 0.02, while the $(F_0\text{-}F)/F$ value of blank can be seen as zero due to $F = F_0$. Thus, $Y_{max/2}$ is estimated to be 0.31 ± 0.01, which indicates the half-maximal fluorescence image contrast change. According to equation (2), the calculated X is 0.90 ± 0.03, and thus the cocaine concentration is $10^{0.90}$ = 7.97 ± 0.57 μM. Due to K_d = [cocaine] is at which half-maximal fluorescence image contrast change occurs, the K_d is determined to be 7.97 ± 0.57 μM.

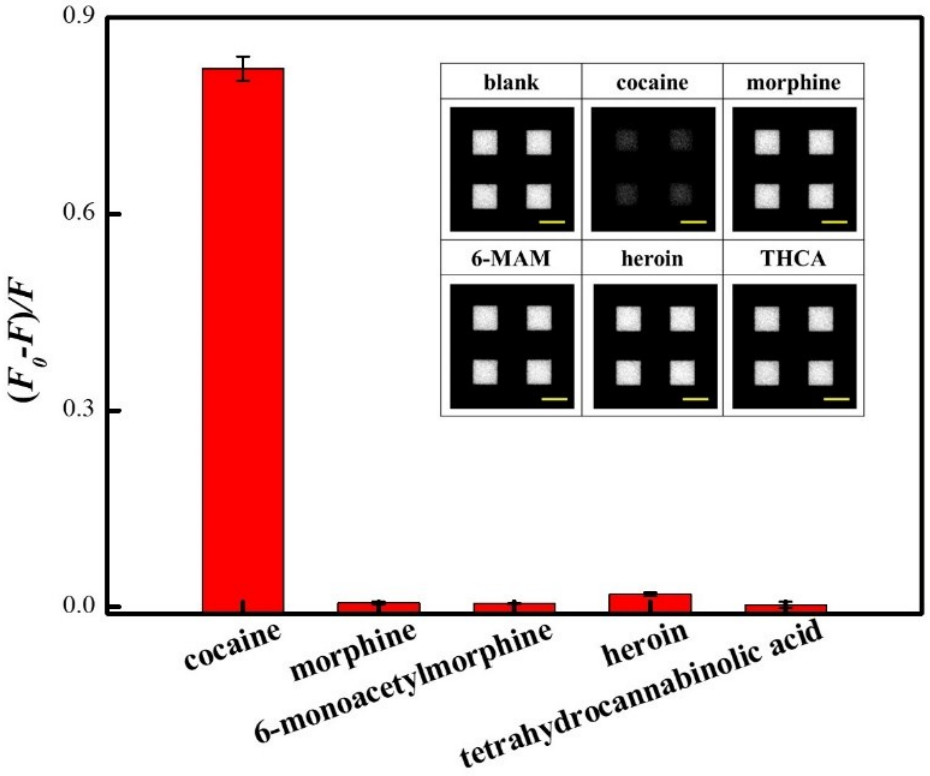

Figure S4. Selectivity examination of the CLL-fabricated aptamer-anchored platform toward cocaine. Each substrate was treated by 1 mM solutes. Each data point is obtained from the average fluorescence signal of individual squares (N = 4). The error bars indicate the standard deviation. The scale bars are 20 μm.

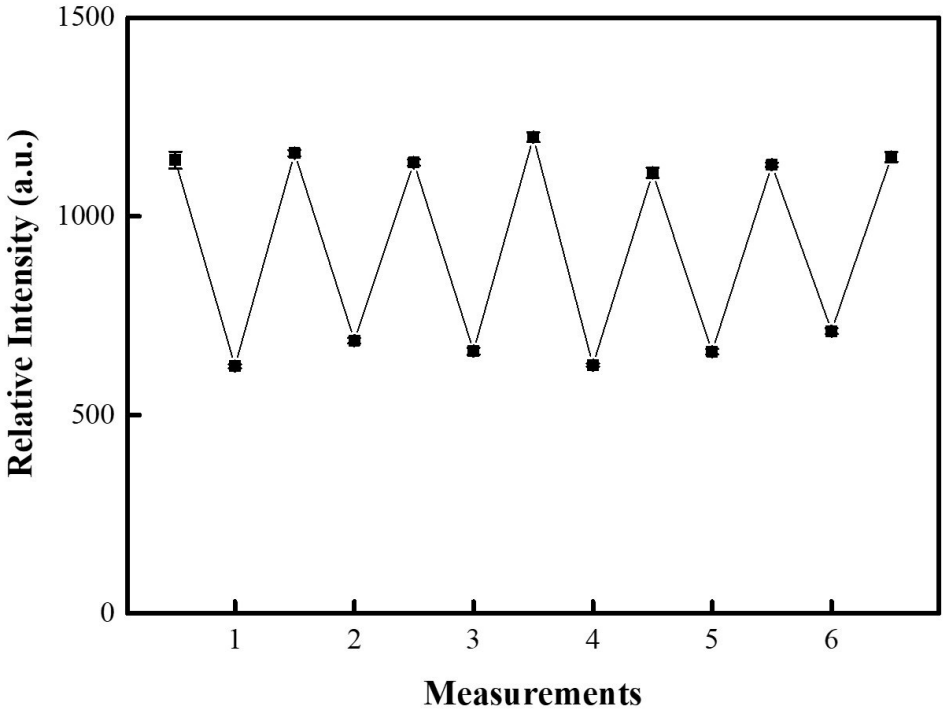

Figure S5. Reusability test of the CLL-fabricated aptamer-anchored platform for several measurement cycles of 100 μM cocaine. Each data point is obtained from the average fluorescence signal of individual squares (N = 4). The error bars indicate the standard deviation.

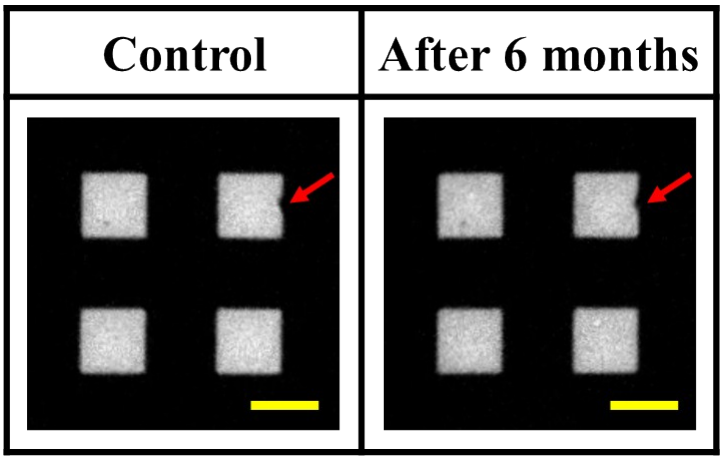

Figure S6. Comparison of the CLL-fabricated aptamer-anchored platform before and after 6-month of storage under 100% relative humidity at 4℃ in dark. The selected pattern defect site (red arrows) indicates that the data were collected from the same sample. The scale bars are 20 μm.

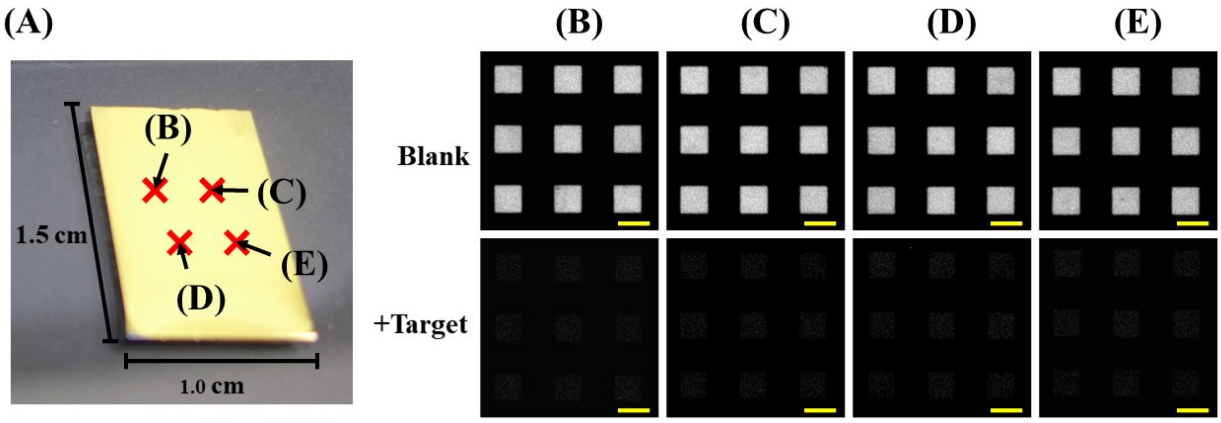

Figure S7. (A) Representing photo and (B-E) corresponding fluorescence images of different sampling spots obtained from the same CLL-fabricated aptamer-anchored substrate, and their responses toward the target. The scale bars are 20 μm.

REFERENCES

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