Electronic Supplementary Information (ESI)

Nanomechanical actuation driven by light-induced DNA fuel

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Experimental details

Preparation of Sample: We have utilized i-motif DNA sequence as a probe molecule in order to functionalize a microcantilever surface. Here, i-motif DNA sequence is given as SH-5'-CCCT AACC CTAA CCCT AACCC-3' (Integrated DNA Technology, CA, USA), and its complimentary DNA exhibits the sequence of 3'-G<u>T</u>GA TTGG GATT <u>T</u>GGA TTG<u>T</u>G-5' (Integrated DNA Technology, CA, USA), where underlined sequences indicate the mismatching sequences to i-motif DNA sequence. For light irradiation-induced chemical oscillation in the pH of a buffer solution, we have prepared malachite green carbinol base (MGCB)-dissolved solution in such a way that deionized water (with resistance of 2 M Ω) is mixed with cetyrimethylammonium bromide (CTAB; 50 mM) [Sigma-Aldrich, USA], NaCl (200 mM; otherwise specified), malachite green carbinol base (MGCB; 100 mM) [Sigma-Aldrich, USA], and dimethysulfoxide (DMSO).

Cantilever Bioassay: We have employed a commercially available V-shaped microcantilever with force constant of 0.12 N/m (Veeco Inc., CA, USA). The microcantilever surface was coated with gold thin layer for immobilization of probe DNA onto a cantilever surface due to gold-thiol conjugation. In particular, just after the gold coating, the immobilization was implemented by immersing the gold-coated cantilever into Tris EDTA buffer solution (pH 8) that contains thiol-modified i-motif DNA sequence (1 μ M). We mounted such a functionalized cantilever onto a liquid cell, which is filled with prepared MGCB solution. We have optically measured the bending deflection change for a microcantilever using Nanoscope V controller available from atomic force microscope (AFM) PicoForce (Veeco Inc., CA, USA). For light irradiation-driven chemical oscillation, we have utilized 6W UV lamp with wavelength of 312 nm (UVITEC, UK) such that UV light is turned onto a liquid cell for 2 hr.

Quantitative Analysis: We have considered Stoney's formula in order to calculate the surface stress change due to light irradiation-driven DNA conformational transitions. Stoney's formula provides the relationship between the surface stress change and the cantilever's bending deflection change such as¹

$$\Delta \tau = \frac{2}{3(1-\nu)} \frac{L_1^2}{\left[WtL_2 + (tb/4L_1)(L_1 - L_2)^2\right]} K\Delta z$$
(1)

where $\Delta \tau$ and Δz represent the surface stress change and bending deflection change, respectively, *K* and *v* indicate the force constant and Poisson's ratio of a microcantilever, respectively, and L_1 , L_2 , *W*, *t*, and *b* are the geometric parameters of a V-shaped microcantilever as shown in Figure S1. In order to quantify the light irradiation-driven actuation, we have introduced the work done by a cantilever bending motion due to light irradiation, which is defined as²

$$\Delta U = -\Delta \tau A \varepsilon_s + \frac{2K}{3(1-\nu)} \Delta z^2 \tag{2}$$

Here, ΔU is the work done due to a light irradiation-driven cantilever motion, ε_s is surface strain given as $\varepsilon_s = \kappa t = (2\Delta z/L_1^2)t$, where κ is a bending curvature, and A is the surface area of a microcantilever. It has to be noted that, in Eq. (2), the first term represents the contributions of the surface stress change to the work, while the second term indicates the strain energy due to cantilever bending deflection motion. Accordingly, the work done due to light irradiation-driven cantilever motion can be estimated as

$$\Delta U = \frac{2K\Delta z^2}{3(1-\nu)} \left[1 - 2\frac{(b/W)(-1+L_1/L_2)+2}{1+(b/4W)(-1+L_1/L_2)^2} \right]$$
(3)

Moreover, in order to quantitatively characterize the kinetics for DNA conformational transition, we have used Langmuir kinetic model such that the kinetics for the conformational change of i-motif DNA from *R*-form to *Q*-form (due to removal of UV light) is denoted as *k*. The Langmuir kinetic model provides the differential equation for the number of DNA molecules in *R*-form such as³

$$\frac{dN(t)}{dt} = -kN(t) \tag{4}$$

where N(t) is the number of molecules in the conformation of *R*-form at time *t*. Consequently, the number of molecules that undergo the conformational transition from R to Q is given by

$$N_{C}(t) = N_{0} \left[1 - \exp\left(-kt\right) \right]$$
⁽⁵⁾

Here, N_0 is the total number of DNA molecules in *R*-form at t = 0. Because the surface stress change due to DNA conformational changes obeys the Langmuir kinetics, the surface stress change is in the form of

$$\Delta \tau(t) = \Delta \tau_0 \left[1 - \exp(-kt) \right] \tag{6}$$

where $\Delta \tau_0$ is the surface stress changes when all DNA molecules undergo conformational transitions (i.e. $t \rightarrow \infty$). Because of linear relationship between the surface stress change and bending deflection change as shown in Eq. (1), the bending deflection change due to conformational change from *R*-form to *Q*-form is given as

$$\Delta z(t) = \Delta z_0 \left[1 - \exp(-kt) \right] \tag{7}$$

Here, Δz_0 is the steady-state value of the bending deflection change.

Table

	Experimental condition	Bending deflection change (nm)	Surface Stress change (mN/m)	Work done in one acutation cycle (× 10 ⁻¹⁵ J/cycle)	Kinetic rate for DNA conformational change due to removal of UV light (× 10 ⁻⁵ s ⁻¹)	Experimental Results
Effect of hybridization	Without complimentary DNA	71.67±3.64	83.03 ± 4.22	3.33	11.06 ± 1.19	Blue graph in Fig. <mark>2</mark> c
	With complimentary DNA (1 µM)	47.11±5.05	$54.59 \pm \\5.86$	1.44	9.36 ± 7.60	Red graph in Fig. <mark>2</mark> c
Effect of monovalent salt concentration	0 M NaCl	-1.19±3.39	-1.38 ± 3.92	N/A	N/A	Red graph in Fig. <mark>3</mark> c
	0.2 M NaCl	71.67±3.64	83.03 ± 4.22	3.33	11.06 ± 1.19	Blue graph in Fig. <mark>3</mark> c
	0.5 M NaCl	107.65 ± 4.32	124.73 ± 5.01	7.54	21.10 ± 9.66	Green graph in Fig. <mark>3</mark> c

Table S1. Summary of experimental results on the nanomechanical actuation of microcantilevers induced by light-driven DNA fuel



Figure S1. Schematic descriptions of V-shaped microcantilever with its geometric parameters



Fig. S2. Light-driven pH change of a MGCB solution: UV irradiation increases the pH due to change of MGCB into MG cation, whereas removal of UV light reversibly reduces the pH of a solution.



Figure S3. (a) light-driven oscillating pH changes of MGCB solution and (b) reversible cantilever's bending motion driven by UV light on-off switching.

References

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