Supporting Information

Development of the selective ligand for G-G mismatches of CGG repeat RNA inducing the RNA structural conversion from the G-quadruplex into the hairpin-like structure

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1. Supplementary date

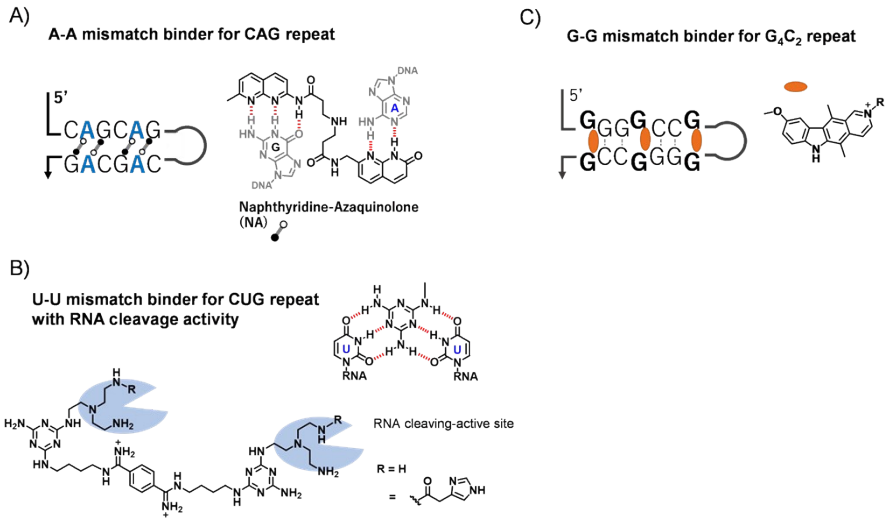


Fig. S1 Previously developed binders for the base mismatch sites of the repeat sequences. A) A-A mismatch binder for CAG repeat reported by Nakatani's group (ref. 9). B) U-U mismatch binder for CUG repeat reported by Zimmerman's group (ref. 13). C) G-G mismatch binder for G_4C_2 repeat reported by Disney's group (ref. 17).

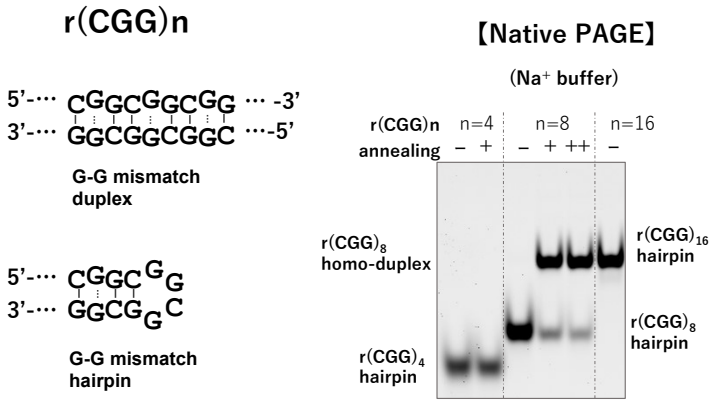
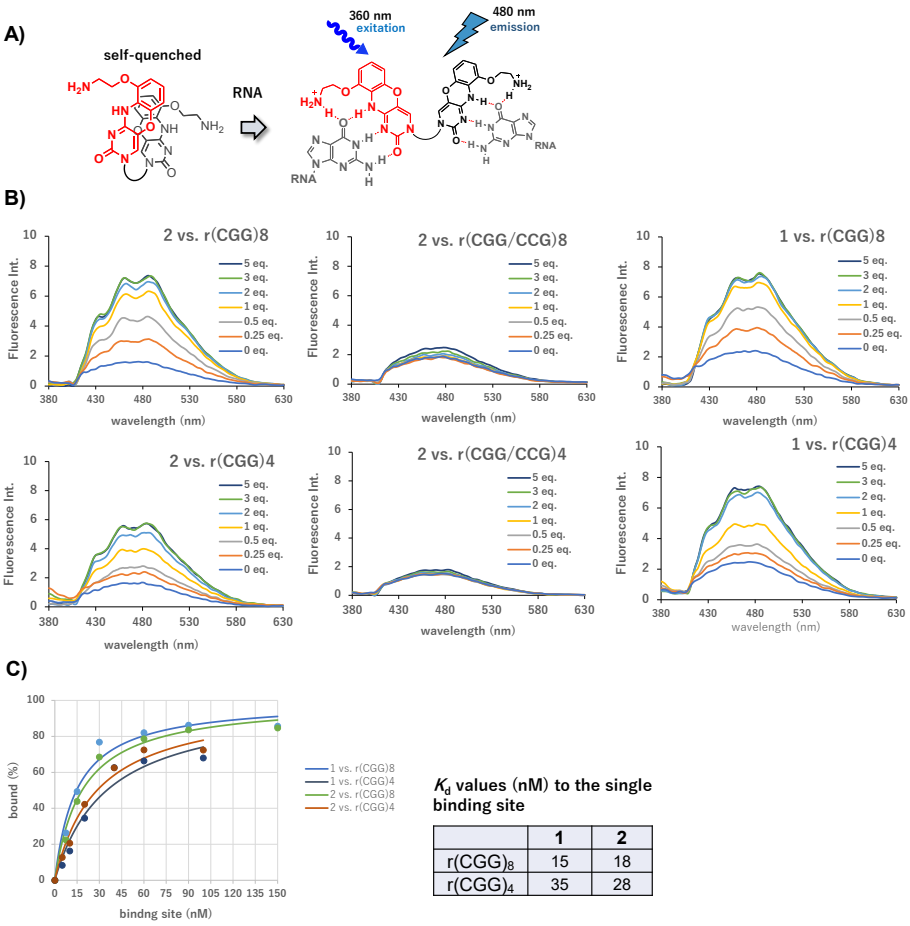


Fig. S2 Native PAGE of r(CGG)n [n=4: 5 μM, n=8: 2 μM, n=16: 1 μM] in 10 mM HEPES-NaOH buffer pH 7.4 containing 100 mM NaCl. (12% acrylamide, acrylamide/bis=29/1, 0.25xTBE, 160 V for 2 h under 4 ºC, stained by SYBR®Gold.

[annealing conditions: 90 ºC to r.t. over 2 h (+), overnight (++) or to ice bath immediately (-)]



K_d values (nM) to the single binding site

	1	2
$r(CGG)_8$	15	18
$r(CGG)_4$	35	28

Fig. S3 A) The proposal mechanism of the light-up response of G-clamp-dimer for RNA binding. B) Fluorescence spectra of **2** or **1** (10 nM) in the presence of RNAs (0 to 50 nM) in 10 mM HEPES-NaOH buffer pH 7.4 containing 100 mM NaCl excited by 360 nm light at 25 °C. C) The K_d values to the single binding site was calculated by the plots of the 480 nm intensity based on the concentrations of the binding sites [r(CGG)$_8$: three binding sites, r(CCG)$_4$: two binding sites].

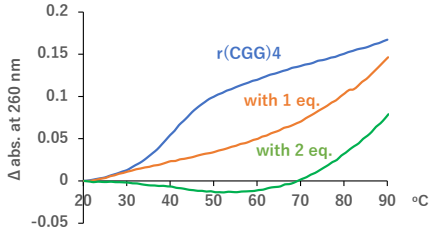


Fig. S4 Thermal melting of 5 μM r(CGG)$_4$ was recorded at 260 nm in the presence of **2** (0, 1 or 2 eq.) in 10 mM HEPES-NaOH buffer pH 7.4 containing 100 mM NaCl under +1 °C/min heating condition.

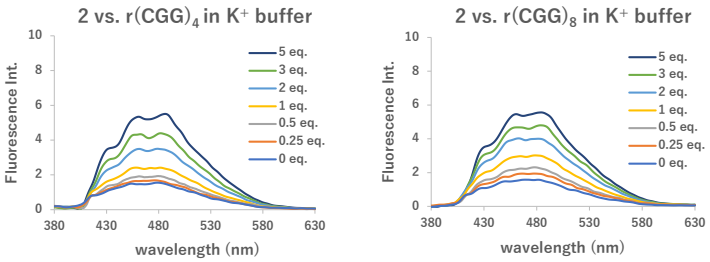


Fig. S5 Fluorescence spectra of **2** (10 nM) in the presence of RNAs (0 to 50 nM) in 10 mM HEPES buffer pH 7.4 containing 100 mM KCl excited by 360 nm light at 25 °C.

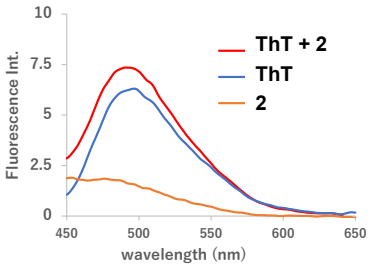


Fig. S6 Fluorescence spectra of **2** (0.2 μM), ThT (0.1 μM) and ThT + **2** in 80 % glycerol excited by 435 nm light at 25 °C.

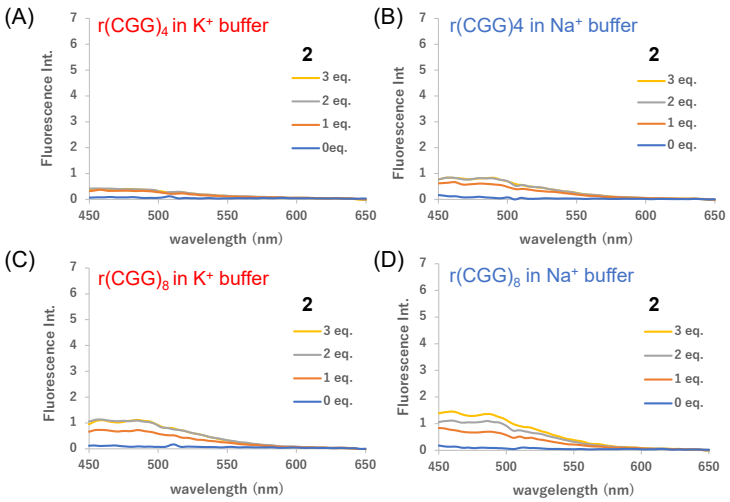


Fig. S7 Fluorescence spectra of **2** (0 to 0.3 µM) and 0.1 µM RNAs without ThT in 10 mM HEPES buffer pH7.4 containing 100 mM KCl or NaCl, excited by 435 nm light at 25 °C. A) $r(CGG)_4$ in K^+ buffer, B) $r(CGG)_4$ in Na^+ buffer, C) $r(CGG)_8$ in K^+ buffer, D) $r(CGG)_8$ in Na^+ buffer.

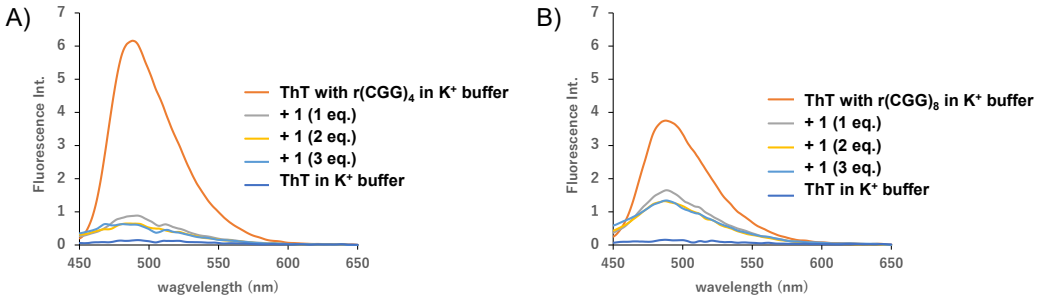


Fig. S8 Fluorescence spectra of thioflavine T (ThT, 0.1 µM) in the absence or presence of 0.1 µM $r(CGG)_4$ (A) or $r(CGG)_8$ (B) and **1** (0 to 3 eq.) in 10 mM HEPES buffer pH7.4 containing 100 mM KCl, excited by 435 nm light at 25 °C.

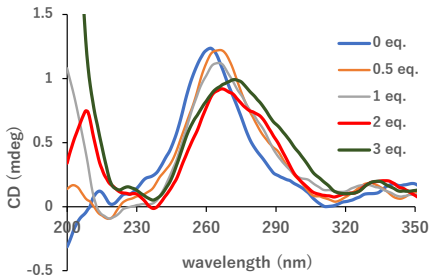


Fig. S9 CD spectrum of 5 µM $r(CGG)_4$ in the presence of **2** (0 to 3 eq.) in 10 mM HEPES-KOH pH 7.4 buffer containing 100 mM KCl at 25 °C.

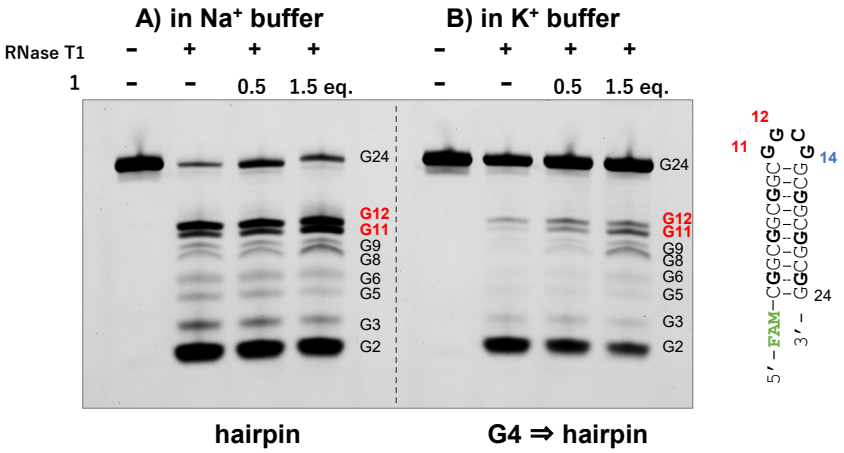
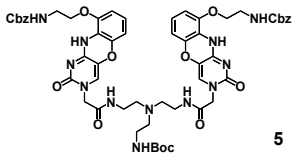


Fig. S10 PAGE analysis of RNase T1 assay. (A) Reactions in the Na^+ buffer, (B) Reactions in the K^+ buffer. The samples containing 5'-FAM-r$(CGG)_8$ (2 µM) and **1** (0, 0.5 or 1.5 eq.) in 10 mM HEPES buffer pH 7.4 containing 100 mM NaCl or KCl were treated with RNase T1 (2.5 U/µL) at 15 °C for 10 min. The digested samples were analysed on 15 % polyacrylamide gel containing 7.5 M urea and 18 % formamide.



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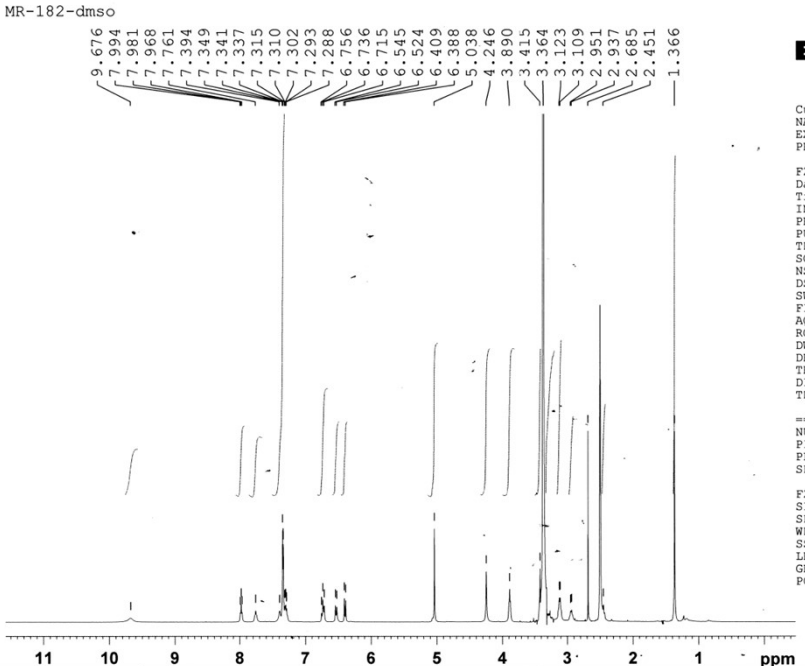

BRUKER

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TE                296.1 K
D1             1.00000000 sec
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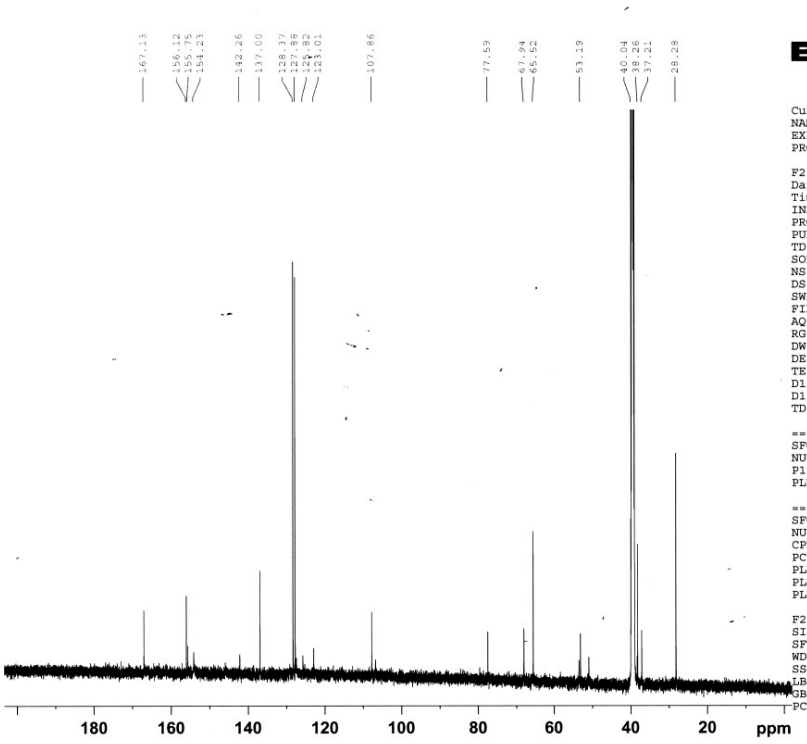

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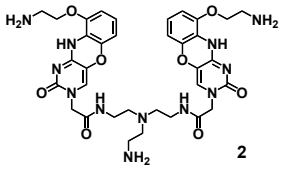

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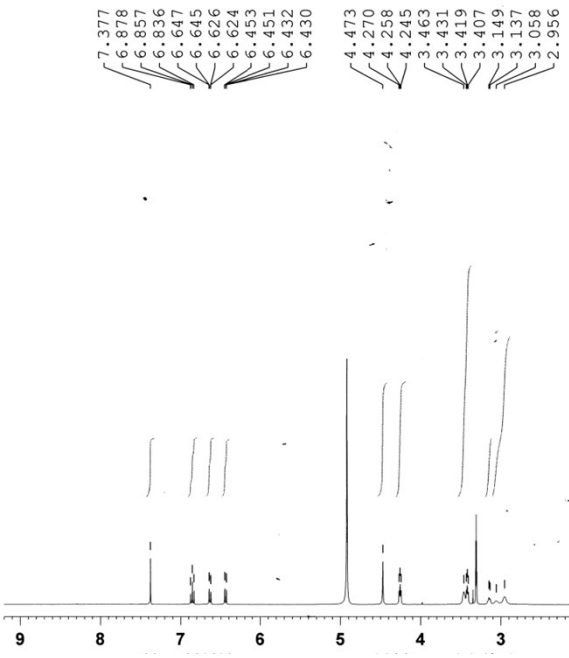
MR-202-HPLC



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FIDRES             0.125483 Hz
AQ                 3.9846387 sec
RG                      33.3
DW                   60.800 usec
DE                    6.50 usec
TE                   293.7 K
D1                1.00000000 sec
TD0                       1

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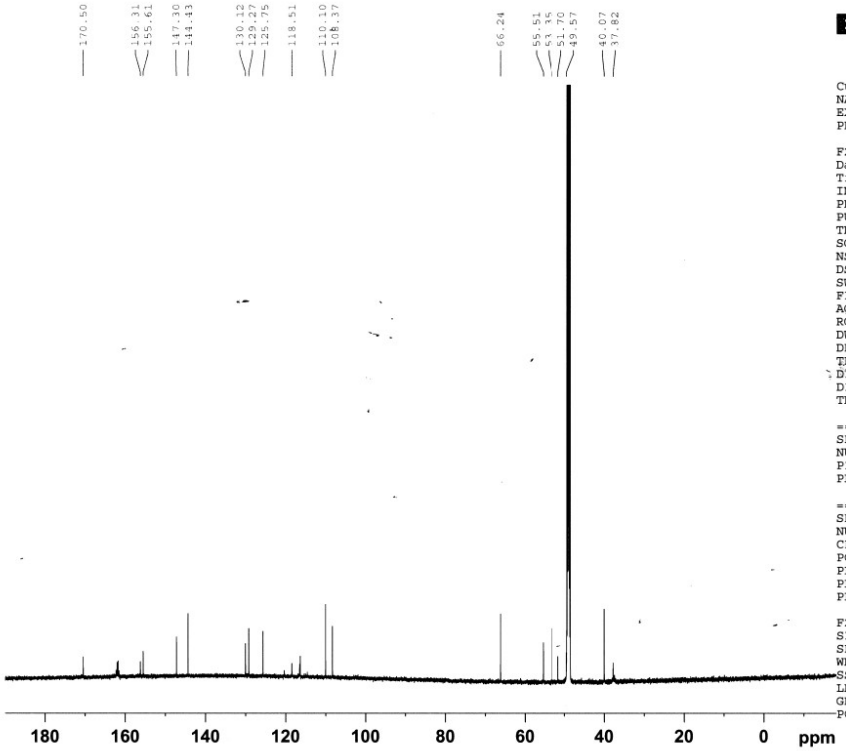
MR-202-C13



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SOLVENT                MeOD
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RG                    198.89
DW                   13.867 usec
DE                    18.00 usec
TE                   298.0 K
D1                2.00000000 sec
D11               0.03000000 sec
TD0                       1

======== CHANNEL f1 ========
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NUC1                    13C
P1                    12.00 usec
PLW1          83.50000000 W

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