

## Supporting information

Table S1 DNA sequences used in this paper.

Name	Sequences (from 5' to 3')	Length (n.t.)
1	CAAGCCCAATAGGAAC CCATGTACAAACAGTT	32
2	AATGCCCCGTAACAGT GCCCGTATCTCCCTCA	32
3	TGCCTTGACTGCCTAT TTCGGAACAGGGATAG	32
4	GAGCCGCCCCACCACC GGAACCGCGACGGAAA	32
5	AACCAGAGACCCTCAG AACCGCCAGGGGTCAG	32
6	TTATTCATAGGGAAGG TAAATATT CATTCAAGT	32
7	CATAACCCGAGGCATA GTAAGAGC TTTTAAAG	32
8	ATTGAGGGTAAAGGTG AATTATCAATCACCGG	32
9	AAAAGTAATATCTTAC CGAAGCCCTTCCAGAG	32
10	GCAATAGCGCAGATAG CCGAACAATTCAACCG	32
11	CCTAATTTACGCTAAC GAGCGTCTAATCAATA	32
12	TCTTACCAGCCAGTTA CAAAATAAATGAAATA	32
13	ATCGGCTGCGAGCATG TAGAAACCTATCATAT	32
14	CTAATTTATCTTTCCT TATCATTATCCTGAA	32
15	GCGTTATAGAAAAAGC CTGTTTAG AAGGCCGG	32
16	GCTCATTTTCGCATTA AATTTTTG AGCTTAGA	32
17	AATTACTACAAATTCT TACCAGTAATCCCATC	32
18	TTAAGACGTTGAAAAC ATAGCGATAACAGTAC	32
19	TAGAATCCCTGAGAAG GTCAATAGGAATCAT	32
20	CTTTTACACAGATGAA TATACAGTAAACAATT	32
21	TTTAACGTTTCGGGAGA AACAAATAATTTCCCT	32
22	CGACAATAAGTATTA GACTTTACAATACCGA	32
23	GGATTTAGCGTATTAA ATCCTTTGTTTTCAGG	32
24	ACGAACCAAAACATCG CCATTAATA TGGTGGTT	32
25	GAACGTGGCGAGAAAG GAAGGGAA CAACTAT	32
26	TAGCCCTACCAGCAGA AGATAAAAACATTTGA	32
27	CGGCCTTGCTGGTAAT ATCCAGAACGAACTGA	32
28	CTCAGAGCCACCACCC TCATTTTCCTATTATT	32
29	CTGAAACAGGTAATAA GTTTTAACCCCTCAGA	32
30	AGTGTAAGGAAAGTA TTAAGAGGCCGCCACC	32

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31	GCCACCACTCTTTTCA TAATCAAACCGTCACC	32
32	GTTTGCCACCTCAGAG CCGCCACCGATACAGG	32
33	GACTTGAGAGACAAA GGGCGACAAGTTACCA	32
34	AGCGCCAACCATTTGG GAATTAGATTATTAGC	32
35	GAAGGAAAATAAGAGC AAGAAACAACAGCCAT	32
36	GCCCAATACCGAGGAA ACGCAATAGGTTTACC	32
37	ATTATTTAACCCAGCT ACAATTTTCAAGAACG	32
38	TATTTTGCTCCCAATC CAAATAAGTGAGTTAA	32
39	GGTATTAAGAACAAGA AAATAATTAAGCCA	32
40	TAAGTCCTACCAAGTA CCGCACTCTTAGTTGC	32
41	ACGCTCAAATAAGAA TAAACACCGTGAATTT	32
42	AGGCGTTACAGTAGGG CTTAATTGACAATAGA	32
43	ATCAAAATCGTCGCTA TTAATTAACGGATTCTG	32
44	CTGTAAATCATAGGTC GAGAGACGATAAATA	32
45	CCTGATTGAAAGAAAT TGCGTAGACCCGAACG	32
46	ACAGAAATCTTTGAAT ACCAAGTTCCTTGCTT	32
47	TTATTAATGCCGTCAA TAGATAATCAGAGGTG	32
48	AGATTAGATTTAAAAG TTTGAGTACACGTAAA	32
49	AGGCGGTCATTAGTCT TTAATGCGCAATATTA	32
50	GAATGGCTAGTATTA CACCGCCTCAACTAAT	32
51	CCGCCAGCCATTGCAA CAGGAAAAATATTTTT	32
52	CCCTCAGAACCGCCAC CCTCAGAACTGAGACT	32
53	CCTCAAGAATACATGG CTTTTGATAGAACCAC	32
54	TAAGCGTCGAAGGATT GGATTAGTACCGCCA	32
55	CACCAGAGTTCGGTCA TAGCCCCCGCCAGCAA	32
56	TCGGCATTCCGCCGCC AGCATTGACGTTCCAG	32
57	AATCACCAAATAGAAA ATTCATATATAACGGA	32
58	TCACAATCGTAGCACC ATTACCATCGTTTTCA	32
59	ATACCCAAGATAACCC ACAAGAATAAACGATT	32
60	ATCAGAGAAAGAACTG GCATGATTTTTATTTG	32
61	TTTTGTTTAAGCCTTA AATCAAGAATCGAGAA	32
62	AGGTTTTGAACGTCAA AAATGAAAGCGCTAAT	32
63	CAAGCAAGACGCGCCT GTTTATCAAGAATCGC	32
64	AATGCAGACCGTTTTT ATTTTCATCTTGCGGG	32
65	CATATTTAGAAATACC GACCGTGTTACCTTTT	32
66	AATGGTTTACAACGCC AACATGTAGTTCAGCT	32
67	TAACCTCCATATGTGA GTGAATAAACAAAATC	32
68	AAATCAATGGCTTAGG TTGGGTTACTAAATTT	32
69	GCGCAGAGATATCAAA ATTATTTGACATTATC	32
70	AACCTACCGCGAATTA TTCATTTCCAGTACAT	32
71	ATTTTGCCTTTTAGG AGCACTAAGCAACAGT	32
72	CTAAAATAGAACAAAG AAACCACCAGGGTTAG	32
73	GCCACGCTATACGTGG CACAGACAACGCTCAT	32
74	GCGTAAGAGAGAGCCA CAGCAAAAAGGTTAT	32

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75	GGAAATACCTACATTT TGACGCTCACCTGAAA	32
76	TATCACCGTACTCAGG AGGTTTAGCGGGGTTT	32
77	TGCTCAGTCAGTCTCT GAATTTACCAGGAGGT	32
78	GGAAAGCGACCAGGCG GATAAGTGAATAGGTG	32
79	TGAGGCAGGCGTCAGA TGTAGCGTAGCAAGG	32
80	TGCCTTTAGTCAGACG ATTGGCCTGCCAGAAT	32
81	CCGAAACACACCACG GAATAAGTAAGACTCC	32
82	ACGCAAAGGTCACCAA GAAACCAATCAAGTT	32
83	TTATTACGGTCAGAGG GTAATTGAATAGCAGC	32
84	TGAACAAACAGTATGT AGCAAATAAAAGAA	32
85	CTTTACAGTTAGCGAA CCTCCCGACGTAGGAA	32
86	GAGGCGTTAGAGAATA ACATAAAAGAACACCC	32
87	TCATTACCCGACAATA AACACATATTTAGGC	32
88	CCAGACGAGCGCCCAA TAGCAAGCAAGAACGC	32
89	AGAGGCATAATTTTCAT CTTCTGACTATAACTA	32
90	TTTTAGTTTTTCGAGC CAGTAATAAATTCTGT	32
91	TATGTAAACCTTTTTT AATGGAAAAATTACCT	32
92	TTGAATTATGCTGATG CAAATCCACAAATATA	32
93	GAGCAAAAACCTTCTGA ATAATGGAAGAAGGAG	32
94	TGGATTATGAAGATGA TGAAACAAAATTTTCAT	32
95	CGGAATTATTGAAAGG AATTGAGGTGAAAAAT	32
96	ATCAACAGTCATCATA TTCCTGATTGATTGTT	32
97	CTAAAGCAAGATAGAA CCCTTCTGAATCGTCT	32
98	GCCAACAGTCACCTTG CTGAACCTGTTGGCAA	32
99	GAAATGGATTATTTAC TTGGCAGACATTCTG	32
112	CCGAAATCCGAAAATC TGTTTGAAGCCGGAA	32
113	CCAGCAGGGGCAAAAT CCCTTATAAAGCCGGC	32
114	GCATAAAGTTCCACAC AACATACGAAGCGCCA	32
115	GCTCACAATGTAAAGC CTGGGGTGGGTTTGCC	32
116	TTCGCCATTGCCGGAA ACCAGGCATTAATCA	32
117	GCTTCTGGTCAGGCTG CGCAACTGTGTTATCC	32
118	GTTAAAATTTTAACCA ATAGGAACCCGGCACC	32
119	AGACAGTCATTCAAAA GGGTGAGAAGCTATAT	32
120	AGGTAAAGAAATCACC ATCAATATAATATTTT	32
121	TTTCATTTGGTCAATA ACCTGTTTATATCGCG	32
122	TCGCAAATGGGGCGCG AGCTGAAATAATGTGT	32
123	TTTTAATTGCCCGAAA GACTTCAAAACACTAT	32
124	AAGAGGAACGAGCTTC AAAGCGAAGATACATT	32
125	GGAATTACTCGTTTAC CAGACGACAAAAGATT	32
126	GAATAAGGACGTAACA AAGCTGCTCTAAAACA	32
127	CCAAATCACTTGCCCT GACGAGAACGCCAAAA	32
128	CTCATCTTGAGGCAAA AGAATACAGTGAATTT	32
129	AAACGAAATGACCCCC AGCGATTATTCATTAC	32
130	CTTAAACATCAGCTTG CTTTCGAGCGTAACAC	32

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131	TCGGTTTAGCTTGATA CCGATAGTCCAACCTA	32
132	TGAGTTTCGTCACCAG TACAACTTAATTGTA	32
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135	GAGTTGCACGAGATAG GGTGAGTAAGGGAGC	32
136	GTGAGCTAGTTTCCTG TGTGAAATTTGGGAAG	32
137	TCATAGCTACTCACAT TAATTGCGCCCTGAGA	32
138	GGCGATCGCACTCCAG CCAGCTTTGCCATCAA	32
139	GAAGATCGGTGCGGGC CTCTTCGCAATCATGG	32
140	AAATAATTTTAAATTG TAAACGTTGATATTCA	32
141	GCAAATATCGCGTCTG GCCTTCCTGGCCTCAG	32
142	ACCGTTCTAAATGCAA TGCCTGAGAGGTGGCA	32
143	TATATTTTAGCTGATA AATTAATGTTGTATAA	32
144	TCAATTCTTTTAGTTT GACCATTACCAGACCG	32
145	CGAGTAGAACTAATAG TAGTAGCAAACCTCA	32
146	GAAGCAAAAAAGCGGA TTGCATCAGATAAAAA	32
147	TCAGAAGCCTCCAACA GGTCAGGATCTGCGAA	32
148	CCAAAATATAATGCAG ATACATAAACACCAGA	32
149	CATTCAACGCGAGAGG CTTTTGCATATTATAG	32
150	ACGAGTAGTGACAAGA ACCGGATATACCAAGC	32
151	AGTAATCTTAAATTGG GCTTGAGAGAATACCA	32
152	GCGAAACATGCCACTA CGAAGGCATGCGCCGA	32
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154	CAATGACACTCCAAAA GGAGCCTTACAACGCC	32
155	AAAAAAGGACAACCAT CGCCCACGCGGGTAAA	32
156	TGTAGCATTCCACAGA CAGCCCTCATCTCCAA	32
157	GTAAAGCACTAAATCG GAACCCTAGTTGTTCC	32
158	AGTTTGGAGCCCTTCA CCGCCTGGTTGCGCTC	32
159	AGCTGATTACAAGAGT CCACTATTGAGGTGCC	32
160	ACTGCCCGCCGAGCTC GAATTCGTTATTACGC	32
161	CCCGGGTACTTTCCAG TCGGGAAACGGGCAAC	32
162	CAGCTGGCGGACGACG ACAGTATCGTAGCCAG	32
163	GTTTGAGGGAAAGGGG GATGTGCTAGAGGATC	32
164	CTTTCATCCCCAAAAA AGGAAGACCGGAGAG	32
165	AGAAAAGCAACATTAA ATGTGAGCATCTGCCA	32
166	GGTAGCTAGGATAAAA ATTTTTAGTTAACATC	32
167	CAACGCAATTTTTGAG AGATCTACTGATAATC	32
168	CAATAAATACAGTTGA TTCCCAATTTAGAGAG	32
169	TCCATATACATACAGG CAAGGCAACTTTATTT	32
170	TACCTTTAAGGTCTTT ACCCTGACAAAGAAGT	32
171	CAAAAATCATTGCTCC TTTTGATAAGTTTCAT	32
172	TTTGCCAGATCAGTTG AGATTTAGTGGTTTAA	32
173	AAAGATTCAGGGGGTA ATAGTAAACCATAAAT	32
174	TTTCAACTATAGGCTG GCTGACCTTGTATCAT	32

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175	CCAGGCGCTTAATCAT TGTGAATTACAGGTAG	32
176	CGCCTGATGGAAGTTT CCATTAACATAACCG	32
177	TTTCATGAAAATTGTGTCGAAATCTGTACAGA	32
178	ATATATTCTTTTTTCA CGTTGAAAATAGTTAG	32
179	AATAATAAGGTCGCTG AGGCTTGCAAAGACTT	32
180	CGTAACGATCTAAAGT TTTGTCGTGAATTGCG	32
181	ACCCAAATCAAGTTTT TTGGGGTCAAAGAACG	32
182	TGGACTCCCTTTTAC CAGTGAGACCTGTCGT	32
183	TGGTTTTTAACGTCAA AGGGCGAAGAACCATC	32
184	GCCAGCTGCCTGCAGG TCGACTCTGCAAGGCG	32
185	CTTGCATGCATTAATG AATCGGCCCGCCAGGG	32
186	ATTAAGTTCGCATCGT AACCGTGCAGTAACA	32
187	TAGATGGGGGGTAACG CCAGGGTTGTGCCAAG	32
188	ACCCGTCGTCATATGT ACCCCGGTAAAGGCTA	32
189	CATGTCAAGATTCTCC GTGGGAACCGTTGGTG	32
190	TCAGGTCACTTTTGCG GGAGAAGCAGAATTAG	32
191	CTGTAATATTGCCTGA GAGTCTGGAAAAGTAG	32
192	CAAAATTAAGTACGG TGTCTGGAAGAGGTCA	32
193	TGCAACTAAGCAATAA AGCCTCAGTTATGACC	32
194	TTTTTGCGCAGAAAAC GAGAATGAATGTTTAG	32
195	AAACAGTTGATGGCTT AGAGCTTATTTAAATA	32
196	ACTGGATAACGGAACA ACATTATTACCTTATG	32
197	ACGAACTAGCGTCCAA TACTGCGGAATGCTTT	32
198	CGATTTTAGAGGACAG ATGAACGGCGCGACCT	32
199	CTTTGAAAAGAACTGG CTCATTATTTAATAAA	32
200	GCTCCATGAGAGGCTTT GAGGACTAGGGAGTT	32
201	ACGGCTACTTACTTAG CCGGAACGCTGACCAA	32
202	AAAGGCCGAAAGGAAC AACTAAAGCTTTCCAG	32
203	GAGAATAGCTTTTTCG GGATCGTCGGGTAGCA	32
204	ACGTTAGTAAATGAAT TTTCTGTAAGCGGAGT	32
Loop 1	AACATCACTTGCCTGAGTAGAAGAACT	27
Loop 2	TGTAGCAATACTTCTTTGATTAGTAAT	27
Loop 3	AGTCTGTCCATCACGCAAATTAACCGT	27
Loop 4	ATAATCAGTGAGGCCACCGAGTAAAAG	27
Loop 5	ACGCCAGAATCCTGAGAAGTGTTTTT	26
Loop 6	TTAAAGGGATTTTAGACAGGAACGGT	26
Loop 7	AGAGCGGGAGCTAACAGGAGGCCGA	26
Loop 8	TATAACGTGCTTTCCTCGTTAGAATC	26
Loop 9	GTAATATGGTTGCTTTGACGAGCACG	26
Loop 10	GCGCTTAATGCGCCGCTACAGGGCGC	26
Capture 200-1	GCTCCATGAGAGGCTTTGAGGTAGTCTGTCAGGTCC	36
Capture 200-2	ACGCTCGGTCAGGATCTTCATGAGGACTAGGGAGTT	36
Capture 152-1	GCGAAACATGCCACTATGAGGTAGTCTGTCAGGTCC	36
Capture 152-2	ACGCTCGGTCAGGATCTTCACGAAGGCATGCGCCGA	36

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Capture 29-1	CTGAAACAGGTAATAATGAGGTAGTCTGTCAGGTCC	36
Capture 29-2	ACGCTCGGTCAGGATCTTCAGTTTTAACCCCTCAGA	36
Capture 77-1	TGCTCAGTCAGTCTCTTGAGGTAGTCTGTCAGGTCC	36
Capture 77-2	ACGCTCGGTCAGGATCTTCAGAATTTACCAGGAGGT	36
Capture 196-1	ACTGGATAACGGAACATGAGGTAGTCTGTCAGGTCC	36
Capture196-2	ACGCTCGGTCAGGATCTTCAACATTATTACCTTATG	36
Capture 148-1	AAATAATTTTAAATTGTGAGGTAGTCTGTCAGGTCC	36
Capture 148-2	ACGCTCGGTCAGGATCTTCATAAACGTTGATATTCA	36
Capture 33-1	ACGCTCAAATAAGAATGAGGTAGTCTGTCAGGTCC	36
Capture 33-2	ACGCTCGGTCAGGATCTTCATAAACACCGTGAATTT	36
Capture 81-1	AGAGGCATAATTTTCAT TGAGGTAGTCTGTCAGGTCC	36
Capture 81-2	ACGCTCGGTCAGGATCTTCACTTCTGACTATAACTA	36
Capture 188-1	ACCCGTCGTCATATGTTGAGGTAGTCTGTCAGGTCC	36
Capture 188-2	ACGCTCGGTCAGGATCTTCAACCCCGTAAAGGCTA	36
Capture 140-1	AAATAATTTTAAATTGTGAGGTAGTCTGTCAGGTCC	36
Capture 140-2	ACGCTCGGTCAGGATCTTCATAAACGTTGATATTCA	36
Capture 41-1	ACGCTCAAATAAGAATGAGGTAGTCTGTCAGGTCC	36
Capture 41-2	ACGCTCGGTCAGGATCTTCATAAACACCGTGAATTT	36
Capture 89-1	AGAGGCATAATTTTCATTGAGGTAGTCTGTCAGGTCC	36
Capture 89-2	ACGCTCGGTCAGGATCTTCA CTTCTGACTATAACTA	36
Capture 184-1	GCCAGCTGCCTGCAGGTGAGGTAGTCTGTCAGGTCC	36
Capture 184-2	ACGCTCGGTCAGGATCTTCATCGACTCTGCAAGGCG	36
Capture 136-1	GTGAGCTAGTTTCCTGTGAGGTAGTCTGTCAGGTCC	36
Capture 136-2	ACGCTCGGTCAGGATCTTCATGTGAAATTTGGGAAG	36
Capture 45-1	CCTGATTGAAAGAAATTGAGGTAGTCTGTCAGGTCC	36
Capture 45-2	ACGCTCGGTCAGGATCTTCATGCGTAGACCCGAACG	36
Capture 93-1	GAGCAAAAATTCTGATGAGGTAGTCTGTCAGGTCC	36
Capture 93-2	ACGCTCGGTCAGGATCTTCAATAATGGAAGAAGGAG	36
C2NP	ACTGGGCGAAACAAGTCTATTGACTATGAGC	31
Extended C2NP	GGA CCT GAC AGA CTA CCT CAT GAA GAT CCT GAC CGA GCG TACT GGG CGA AAC AAG TCT ATT GAC TAT GAG C	71

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Table S2.1 Cytotoxicity of folding buffer on K299 cells.

C* (mM)	12.5	6.25	3.125	1.25	0.625
Inhibition rate (%)	9.53±0.51	6.54±0.50	3.96±0.21	2.33±0.26	0.93±0.13

\*: Concentration of Mg<sup>2+</sup> in folding buffer.

Table S2.2 Cytotoxicity of RE on K299 cells.

C <sub>RE</sub> (nM)		4	2	1	0.5	0.1
Inhibition rate (%)	24 h	1.86±0.28	0.99±0.41	0.95±0.28	0.99±0.28	0.42±0.12
	48 h	1.36±0.39	0.99±0.34	1.57±0.37	2.13±0.31	1.49±0.33
	72 h	2.87±0.45	1.99±0.45	1.13±0.56	2.98±0.11	1.41±0.31

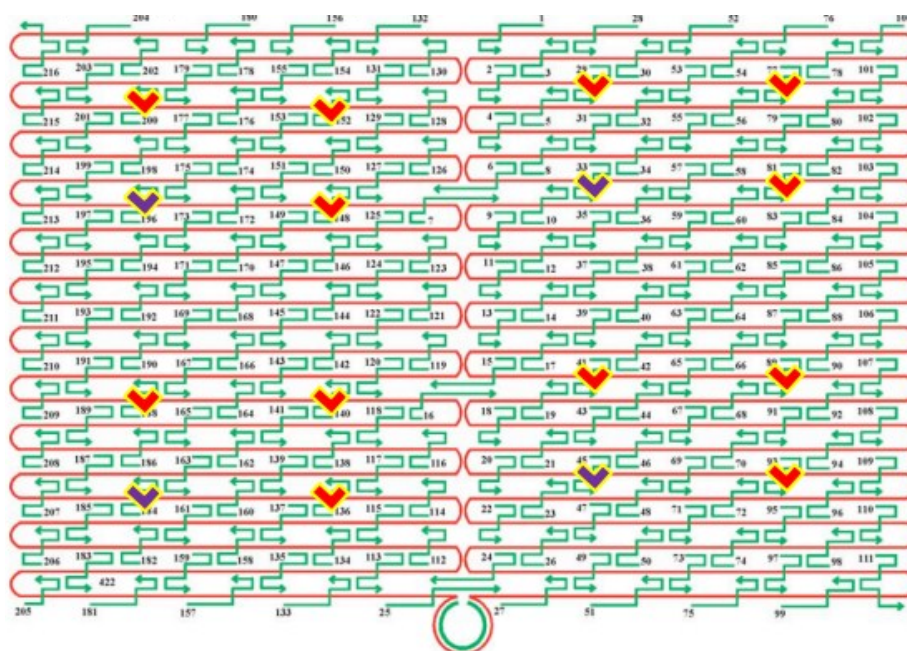


Fig. S1 Distribution of aptamers on DNA Origami nanostructure (For RE-4Apt, strand 196, 184, 33 and 45 were replaced with capture ssDNA. For RE-16Apt, strand 200, 196, 188, 184, 152, 148, 140, 136, 29, 33, 41, 45, 77, 81, 89 and 93 were replaced with capture ssDNA)<sup>1</sup>.

## Reference

1. Y. Ke, S. Lindsay, Y. Chang, Y. Liu and H. Yan, *Science*, 2008, **319**, 180-183.