A Spherical Nucleic Acids Platform Based on Self-Assembled DNA Biopolymer for High Performance Cancer Therapy

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Supporting Materials: figures and table

Entry	Sequence (5'-3')
Initiator Strand	TGCTGCTGCTGCTGCACGACG AAAAAA
M1	CGTCGTGCAGCAGCAGCAGCAACGGCTTGCTGCTGCTGCTGCTGC
M2	TGCTGCTGCTGCTGCACGACGGCAGCAGCAGCAGCAGCAGCCAGC
M3	<u>GGTGGTGGTGGTTGTGGTGGTGGTGG</u> TTTCGTCGTGCAGCAGCAGCAGCAGCA ACGGCTT
	GCTGCTGCTGCTGC
M3-FITC	<u>GGTGGTGGTGGTGGTGGTGGTGGTGG</u> TTTCGTCGTGCAGCAGCAGCAGCAGCAACGGCTT
	GCTGCTGCTGCTGC-FITC
M3-TMR	<u>GGTGGTGGTGGTTGTGGTGGTGGTGG</u> TTTCGTCGTGCAGCAGCAGCAGCAGCAGCAGCGGCTTG
	CTGCTGCTG CTGCTGC-TMR
Biotin-M3-FITC	Biotin-GGTGGTGGTGGTGGTGGTGGTGGTGGTGGTGGTGGTGGT
	GCTTGCTGCTGCTGCTGC -FITC
Biotin-M3-TMR	Biotin-GGTGGTGGTGGTGGTGGTGGTGGTGGTGGTGGTGGTGCAGCAGCAGCAGCAGCAACG
	GCTTCTGCTGCTGC -TMR
M4	<u>GGTGGTGGTGG</u> TTTT TGCTGCTGCTGCTGCACGACGGCAGCAGCAGCAGCAGCAAGC
	CGTTT <u>TGTGGTGGTGG TGG</u>
Sgc8 capped-Strand I-FITC	TGCTGCTGCTGCTGCACGACG TTTATCTAACTGCTGCGCCGCCGGGAAAATACTGTA
	CGGTTAGA-FITC
Sgc8 capped-Strand II-FITC	C <u>ATCTAACTGCTGCGCCGGCGGGAAAATACTGTACGGTTAGA</u> TTTACGGCTTGCTGCTGCT
	GCTGCTGC-FITC

Table S1. Oligonucleotides Used in This Work*

sequences are represented in the same color.

Size(nm)	Initiator(nM)	M1(nM)	M2(nM)
13.4 ± 1.2	0.0	0.0	0.0
19.0 ± 0.4	9.6 ± 1.2	8.7± 2.4	6.9±1.8
42.5 ± 0.8	18.9 ± 0.8	41.2±3.4	32.1± 1.5
112.3±2.4	40.8 ± 1.3	210.4± 4.7	189.3±2.5
130.4 ± 6.8	78.4 ± 3.4	312.8± 2.8	295.5± 2.7
110.4 ± 4.8	97.6 ± 4.2	368.7±2.3	373.5± 1.9

 Table S2. The concentrations of conjugated initiator and self-assembled M1, M2 for each size of AuNP-SNAs.



Figure S1. Absorption spectra of AuNPs and initiator strand before (black) and after (blue) conjugated.



Figure S2. Hydrodynamic size distributions of AuNP-SNAs characterized by DLS as a function of different concentrations of initiator strand conjugated on the AuNPs. (The concentrations of initiator strand from A to C, 0, 18.9 ± 0.8 and 78.4 ± 3.4 nM, respectively).



Figure S3. Topography AFM images and the corresponding height profiles of various sizes of AuNP-SNAs (A and B, the size are 13.4 ± 1.2 and 68.4 ± 4.8 nm, respectively). Scale bar: 400 nm.



Figure S4. EB-stained 4% agarose gel electrophoresis image of DNA biopolymer self-assembled on the surface of the AuNPs after treatment by 10 mM DTT. Lane 1: 25 bp Marker; Lane 2: Initiator conjugated-AuNPs; Lane 3: Initiator-AuNPs+M1+M2. (Concentrations: initiator strand is 80 nM; M1 and M2 are 400 nM)



Figure S5. EB-stained 4% agarose gel electrophoresis image of DNA biopolymer self-assembled as a function of different location of AS1411. (Lane 2-5) Initiator strand cascade of hybridization reaction by initiator strand M2 and M3. (Lane 6-9) Initiator strand cascade of hybridization reaction by initiator strand M2 and M4. (Lane 1: 50 bp Marker; Lane 2 and 6: (Initiator strand: M2:M3/M4=0.5:10:10); Lane 3 and 7: (Initiator strand: M2: M3/M4=1:10:10); Lane 4 and 8: Initiator strand: M2: M3/M4=2:10:10); Lane 5 and 9: (Initiator strand: M2: M3/M4=5:10:10).



Figure S6. Spectroscopic analysis of the interactions between hemin and different samples: initiator conjugated-MNPs (black curve), initiator conjugated-MNPs after HCR (red curve). [Hemin]= 1 μ M, [ABTS]= 1 mM, [H₂O₂]= 2 mM, The concentration of MNPs is 1 nM.



Figure S7. (A) Average hydrodynamic sizes of AS1411/MNP-SNAs characterized by DLS as a function of different incubation time in 3% FBS. (B) Fluorescent spectrum of TAMRA-labeled AS1411/MNP-SNAs as a function of different incubation time in 3% FBS (From top to bottom: 0, 0.5, 1.0, 2.0, 5.0 and 10.0 h). The concentration of MNPs is 5 nM.



Figure S8.Cytotoxicity assay of (black) SKOV3 cells (target cells) and (red) HBE135 cells (control cells) treated with various concentrations of AS1411/MNP-SNAs.



Figure S9. (A) 4% agarose gel electrophoresis image of DNA biopolymer before and after FITC-Sgc8 strand I capping. Lane 1: FITC-Sgc8 strand I; Lane 2: DNA biopolymer (Initiator strand: M1: M3=2:10:10); Lane 3: DNA biopolymer capped with FITC-Sgc8 strand I. (B) Fluorescent spectrum of Sgc8/MNP-SNAs before (black) and after (blue) FITC-Sgc8 strand I capping.



Figure S10. Fluorescence spectra of DOX solution (2 μ M) with increasing equivalences of Sgc8/MNP-SNAs (From top to bottom: 0, 0.5, 1.0, 2.0, 3.0, 4.0, 5.0 and 10 nM).



Figure S11. ESI Mass Spectra of SH-Initiator (A), Biotin-Initiator (B), M1(C), M2 (D) and M3 (E). Table F is the mass comparison between expected and observed value for A, B, C, D and E.