Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

Supporting Information

Recent advances of catalytic hairpin assembly and its application in bioimaging and biomedicine

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Table S1. Newly developed CHA strategies

Newly developed CHA strategies	Target	Detection method	Linear range	LOD	Ref
bCHA with three-way junctions	DNA	Colorimetry	10-600 pM	7.7 pM	1
bCHA with three-way junctions	EGFR mutant DNA	Colorimetry	870 aM-87 pM	7.7 fM	2
bCHA with three-way junctions	DNA	Colorimetry	0.01-5 pM	0.1 pM	3
bCHA with three-way junctions	DNA	Colorimetry	0.02-1 pM	0.01 pM	4
bCHA with three-way junctions	has-miR-143-3p, has-miR-18b-5p, has-miR-424-5p, has-miR-93-5p	Colorimetry	0-100 pM	5 pM	5
bCHA with three-way junctions	Cancer cells	Colorimetry	25-10^7 cells	10 cells	6
bCHA with three-way junctions	DNA	Fluorescent	100 pM-400 nM	10 pM	7
bCHA with three-way junctions	Bisphenol A	Fluorescent	10 fM-10 nM	5 fM	8
bCHA with three-way junctions	DNA, adenosine triphosphate	Fluorescent	1 pM-100 nM	4.59×10^{-13} M	9
bCHA with three-way junctions	Hg^{2+}	Fluorescent	10 pM-100 nM	4.5 pM	10
bCHA with three-way junctions	Hg^{2+}	Fluorescent	50-1200 pM	25 pM	11
bCHA with three-way junctions	Exosomal microRNA-181	Electrochemistry	10 fM-100 nM	7.94 fM	12
bCHA with four-way junctions	Nucleic acid biomarkers	Colorimetry	100 aM-10 pM	100 aM	13
bCHA with four-way junctions	DNA	Fluorescent	Not shown	Not shown	14
bCHA with four-way junctions	Chloramphenicol (CAP), kanamycin (Kana)	Microchip electrophoresis	0.001-40 ng/mL	0.52 pg/mL (CAP), 0.41 pg/mL (Kana)	15
bCHA with dendrimer-like DNA nanostructures	DNA	Colorimetry	50-700 pM	9 pM	16
C-CHA	MicroRNA	Colorimetry	100 fM-10 nM	36.2 fM	17
C-CHA	MicroRNA	ECL/ Electrochemistry	100 aM -100 pM	28.75 aM	18

C-CHA	MicroRNA	Fluorescent	Not shown	Not shown	19
C-CHA	Cytokines	Fluorescent	1 pM-50 nM	0.6 pM	20
C-CHA	DNA, microRNA	Fluorescent	1 pM -10 nM	0.67 pM	21
SRCHA	DNA, small molecule	Colorimetry	10-500 pM	5 pM	22
SRCHA	DNA	Fluorescent	1-60 nM	58 pM	23
SRCHA	Cd^{2+}	Fluorescent	10 pM -100 mM	5 pM	24
SRCHA	Aflatoxin B1	Fluorescent	0.30-15 ng/mL	0.13 ng/mL	25
SRCHA	ATP	Fluorescent	20-1000 nM	7 nM	26
LCHA	MicroRNA	Fluorescent	Not shown	2.0 pM	27
LCHA	MiRNA-21	Fluorescent	100 pM- 10 nM	58.1 pM	28
LCHA	MicroRNAs	Fluorescent	Not shown	Not shown	29
LCHA	Messenger RNA	Fluorescent	Not shown	Not shown	30
LCHA	Exosomal microRNA	Electrochemistry	10 aM -100 pM	7.2 aM	31
DCHA	MicroRNAs	ECL	50 aM-1 nM（miRNA-21），20 aM-1 nM（miRNA-155）	14.8 aM（miRNA-21），5.3 aM（miRNA-155）	32
DCHA	MiRNA-21 and MUC1	ECL	20 aM-50 pM（miRNA-21），1 fg/mL-10 ng/mL（MUC1）	11 aM（miRNA-21），0.40 fg/mL（MUC1）	33

Table S2. CHA integrated with other techniques

Combination Method	Target	Detection method	Linear range	LOD	Ref.
CHA& DNAzyme	MicroRNA	Colorimetry	50 fM-50 nM	27 fM (Colorimetry) 500 fM (naked eye)	34
CHA& DNAzyme	MicroRNA-182	Fluorescent	10 fM-10 nM	6.8 fM	35
CHA& DNAzyme	Human papillomavirus -16 DNA	Fluorescent	10 pM-10 nM	5.7 pM	36
CHA& double-end DNAzyme	HIV-1 DNA	Fluorescent	1 pM-2 nM	1 pM	37
CHA& DNAzyme	Telomerase Activity	Fluorescent	Not shown	Not shown	38
CHA&3D-Bipedal DNA Walker& DNAzyme	Thrombin	Fluorescent	0.1 pM-5 nM	23 fM	39
CHA&DNA nanonet	MicroRNA	Electrochemistry	10 fM-1 nM	3.6083 fM	40
CHA& quadrivalent cruciform DNA nanostructure	Amyloid β Oligomers	Fluorescent	1 pM-100 nM	0.69 pM	41
CHA& DNA Nanotweezer	MicroRNAs	Colorimetry	100 fM-10 nM	30 fM	42
CHA& DNA Nanotweezer	MiRNA, uracil-DNA glycosylase	Fluorescent	0.1 pM-50 nM	8.7 fM	43
CHA& DNA Nanotweezer	MicroRNA	Electrochemistry	1 fM-100 pM	0.32 fM	44
CHA&HCR& DNAzyme	Carcinoembryonic antigen	Photoelectrochemistry	70 ag/mL-500 fg/mL	70 ag/mL (measured detection limit)	45
CHA&HCR& DNAzyme	MiRNA-21	Colorimetry	1.0-1000 pM	0.2 pM	46
CHA&HCR	Prostate-specific antigen	Colorimetry	1-100 pg/ mL	1 pg/mL	47
CHA&HCR	MiR-21	Fluorescent	0-1 nM	2 pM	48
CHA&HCR	MiRNA	Fluorescent	0.02-20 nM	6.9 pM	49
CHA&HCR	8-hydroxy-2'-deoxyguanosine	Electrochemistry	100 fM -10 nM	24.34 fM	50
CHA&HCR& alkaline phosphatase	MicroRNA	Photoelectrochemistry	1 fM-1 nM	0.41 fM	51

CHA&HCR	Single-Cell microRNA	Fluorescent	Not shown	60 copies	52
CHA&HCR& DNAzyme	Human telomerase RNA	Photoelectrochemistry	200 fM -20.0 nM	17.0 fM	53
LCHA&HCR	DENV gene	SERS	1 fM- 10 nM	0.49 fM	54
CHA&HCR& alkaline phosphatase	Nucleic acids	Photoelectrochemistry	0.1 fM -100 pM	0.052 fM	55
CHA&HCR& DNAzyme	Prostate specific antigen	Fluorescent	0.001-10 ng/mL	0.73 pg/mL	56
CHA&RCA	DNA	Fluorescent	1-100 nM	0.84 nM	57
CHA&RCA	MicroRNA	Electrochemistry	15 fM-250 pM	13.5 fM	58
CHA&RCA	Nucleic acids	Fluorescent	4.0-10 aM	1.2 aM	59
CHA&RCA& exonuclease III	Carcinoembryonic antigen	Electrochemistry	10 fg/mL-100 ng/mL	4.2 fg/mL	60
CHA& bipedal DNA walker& HCR& DNAzyme& exonuclease III	DNA	Electrochemistry	0.1 fM-100 pM	3.01 aM	61
CHA&T7 exonuclease	MicroRNA	Electrochemistry	1 fM-100 pM	200 aM	62
CHA& duplex-specific nuclease	MicroRNA	Electrochemistry	0-10 nM	25.1 aM	63
CHA&duplex-specific nuclease& DNAzyme	MicroRNA	Colorimetry	10 fM–1 nM	9.2 fM	64
CHA& Nt.BsmAI	HIV DNA	Fluorescent	10-500 pM	2.6 pM	65
CHA& Nt.BbvCI	P53 gene, cancer cells	Fluorescent	5 pM-2 nM (p53 gene), 1×10^2–1×10^4 cells/mL (cancer cells)	0.9 pM (p53 gene), 100 cells/mL (cancer cells)	66
CHA& CRISPR-Cas9	MicroRNA	Fluorescent	1 pM-10 nM	23.5 fM	67
CHA&CRISPR-Cas13a	Dengue virus	Electrochemistry	5 fM-50 nM	0.78 fM	68
CHA&CRISPR-Cas13a	MicroRNA-21	Electrochemistry	10 fM-1 nM	2.6 fM	69

Table S3. CHA-based biosensors for bioimaging and biomedicine

Application	Target	Method	Nanomaterials	Ref
Cell surface *in situ* imaging	ATP, target DNA, proton	bCHA	Not used	70
Cell surface imaging	Target cancer cell	LCHA&HCR	Not used	71
Intracellular imaging	MnSOD mRNA	CHA	Not used	72
Intracellular *in situ* imaging	Telomerase activity	CHA	Not used	73
In situ imaging in living CTCs	MicroRNA-21	CHA	Not used	74
Intracellular imaging	Endogenous metal ions	CHA& DNAzyme	Not used	75
Intracellular *in situ* imaging	MicroRNA-155	bCHA	Not used	76
Intracellular imaging	MicroRNA-155	bCHA	Not used	77
Intracellular imaging	MicroRNA-21	CHA&HCR	Not used	48
Intracellular imaging	Polynucleotide kinase	CHA&HCR&λ exonuclease	Not used	78
Intracellular imaging	Telomerase activity	CHA&RCA	Not used	79
Intracellular imaging	MicroRNA-21	CHA& DNAzyme	Not used	80
Intracellular imaging	MicroRNA-21	CHA& DNAzyme	Not used	81
Intracellular *in situ* imaging	MicroRNA-21	CHA&CRISPR–Cas9	Not used	67
Intracellular imaging	MiRNA-21	CHA&Glutathione	AuNPs	82
Intracellular imaging of base-excision repairing	Human apurinic/apyrimidinic endonuclease 1	DNA walker fueled by CHA	AuNPs	83
Intracellular imaging	MiRNA-21	3D DNA walker based on LCHA	AuNPs	84
Intracellular imaging	MiRNA-21	CHA	AuNPs	85
Intracellular imaging	MiRNA-21	CHA	AuNFs	86
Intracellular imaging	MiRNA-21, miRNA-155	CHA & 3D DNA walker	Janus nanoparticles	87
Intracellular SERS imaging	MicroRNA-1246	CHA	Au nanodumbbells (Au NDs) & Au NPs	88
Intracellular SERS imaging	MiR-125a-5p	CHA	Gold nanocage	89
Intracellular imaging	MiRNA-21	CHA&double-channel exciting single colour FL (DCESCF)	UCNPs& AuNPs	90
Intracellular imaging	MiRNA-21	CHA	UCNPs	91
Intracellular imaging	MiRNA-21	CHA&PC molecular switch	UCNPs	92
Intracellular imaging	MiRNA-21	CHA	GO	93
Intracellular imaging	Circ-Foxo3	CHA&HCR	GO	94
Monitoring Telomerase Activity in Living Cells	Telomerase activity	CHA&DNAzyme	MnO_2 nanosheets	38
Intracellular imaging	MiRNA-21	CHA	Near-infrared (NIR) light-propelled Janus-based nanoplatform	95
Intracellular imaging	Uracil DNA glycosylase	CHA&DNAzyme	MnO_2 nanosheets	96

Intracellular imaging	MiRNA-20a	CHA	Fe_3O_4@C nanoparticles	97
Intracellular imaging	MiR-21	LCHA	DNA nanowire	27
Intracellular imaging	MiR-21	Dispersion-to-localization of CHA	DNA nanowire	98
Intracellular imaging	Survivin and TK1	Crosslinking CHA	DNA nanotetrad	99
Intracellular imaging	MiR-21	LCHA	3D DNA cubic structure	29
Intracellular imaging	MnSOD mRNA	LCHA	DNA tetrahedron	30
Intracellular imaging	MiRNA-21, miRNA-155	CHA&DNAzyme	3D DNA scaffold	100
Intracellular imaging	MiR-155	CHA	Tetrahedral DNA frameworks	101
Intracellular and *in vivo* imaging	MiRNA-34a	Cascade CHA	Not used	19
Intracellular and *in vivo* imaging	Survivin mRNA	CHA	Y scaffold	102
Intracellular and *in vivo* imaging	MiR-21, let7a	CHA	DNA nanolantern	103
in vivo monitoring of drug-induced liver injury	MiR122	CHA&glutathione	UCNPs and gold nanorods	104
Imaging of microRNA and targeted drug delivery	Let-7a	CHA	DNA Nanohydrogels	105
Intracellular imaging and photothermal therapy	Survivin mRNA	CHA	Gold nanorods	106
Intracellular imaging and gene silencing	GalNAc-T mRNA, human early growth response-1	CHA& DNAzyme	Not used	107
In vivo magnetic resonance imaging (MRI), mRNAs silencing, photodynamic therapy	MiR-21	CHA&DNAzyme	Poly (D,L-lactic-co-glycolic acid) nanocarrier	108
Intracellular Imaging and Photodynamic−Photothermal Combination Therapy	Telomerase	CHA	Carbon Nanocage/Fe_3O_4	109

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