Supporting Information

Oxidative Capacity Storage of Transient Singlet Oxygen from Photosensitization with a Redox Mediator for Improved Chemiluminescent Sensing

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Table S1. DNA or RNA sequences used in this work

Name	Sequences (5'→3')
BRCA1	GAG CAT ACA TAG GGT TTC TCT TGG TTT
CS-BRCA1	AAA CCA AGA GAA ACC CTA TGT ATG CTC
BRCA2	AAA GGG CTT CTG ATT
CS-BRCA2	AAT CAG AAG CCC TTT
p53	TTC CTC TGT GCG CCG GTC TCT CCT
CS-p53	AGG AGA GAC CGG CGC ACA GAG GAA
One base mismatch-BRCA1	GAG CAT ACA TAG GCT TTC TCT TGG TTT
Three base mismatch-BRCA1	GAG CTT ACA TAG GCT TTC TCT TCG TTT
One base mismatch-p53	TTC CTC TGT GCG CCA GTC TCT CCT
Three base mismatch-p53	TTC CTC TGT ACG CCA GTA TCT CCT
Radom sequence DNA	ACA CAC ACA CAC ACA CAC ACA CAC ACA CAC ACA CAC ACA CAC ACA CAC AC

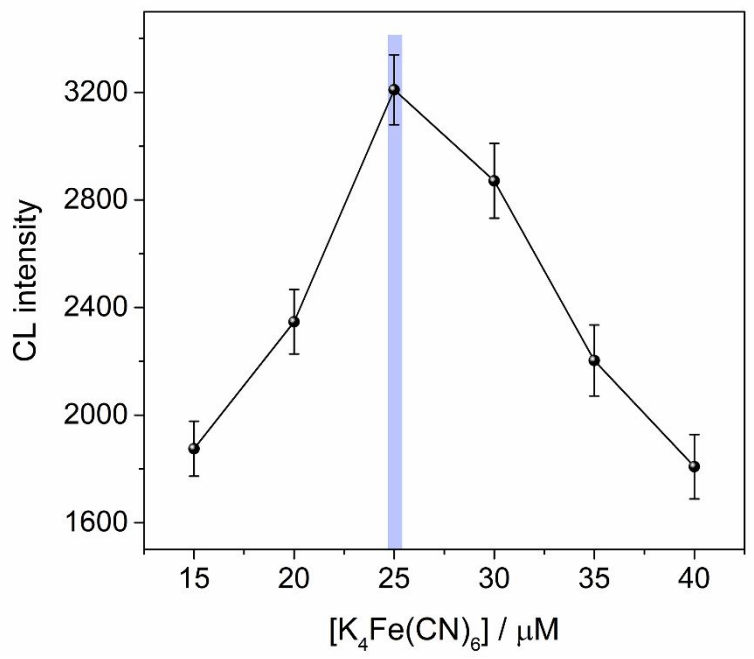

Figure S1. Optimization of the ferrocyanide concentration for amplification of the luminol CL.

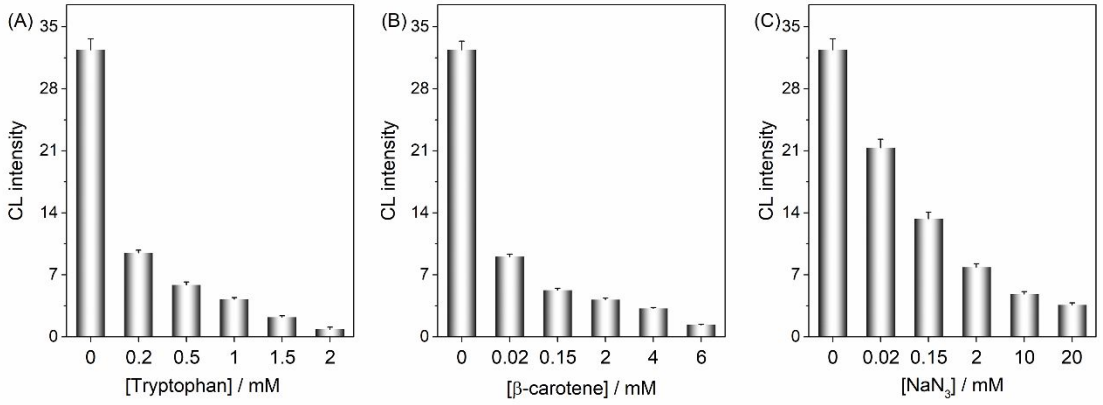

Figure S2. The concentration-dependent scavenging of singlet oxygen by tryptophan, β-carotene, and NaN_3.

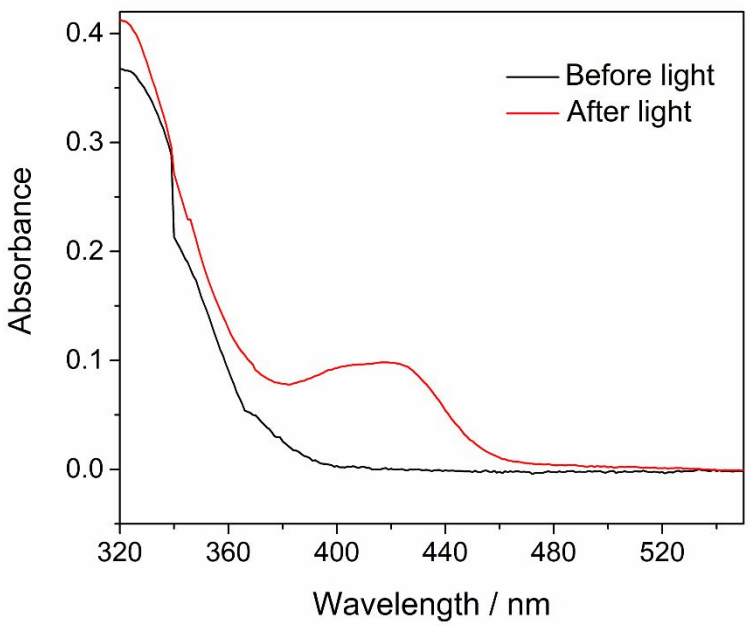

Figure S3. Absorption profiles of PB + $K_4Fe(CN)_6$ before and after green LED irradiation.

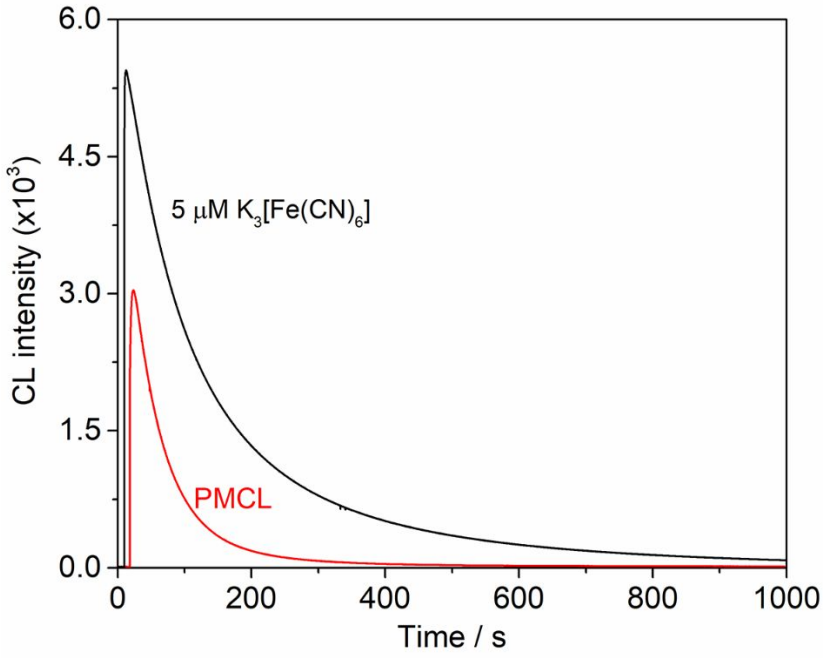

Figure S4. Time courses of the CL profiles from the luminol-singlet oxygen-$K_4Fe(CN)_6$ reaction and the luminol- $K_3Fe(CN)_6$ reaction.

$$Conversion\ rate\ (\mathrm{K4Fe(CN)6}) = \frac{[\mathrm{K3Fe(CN)6}](\mathrm{photo-generated})}{\mathrm{irradation\ time(min)}}$$

The concentration of the generated ferricyanide was derived from the CL intensity of PMCL, based on the linear relationship between the CL intensity and the concentration of $K_4Fe(CN)_6$. The conversion rate was thus calculated according to the above equation (photo irradiation time of 30 min).

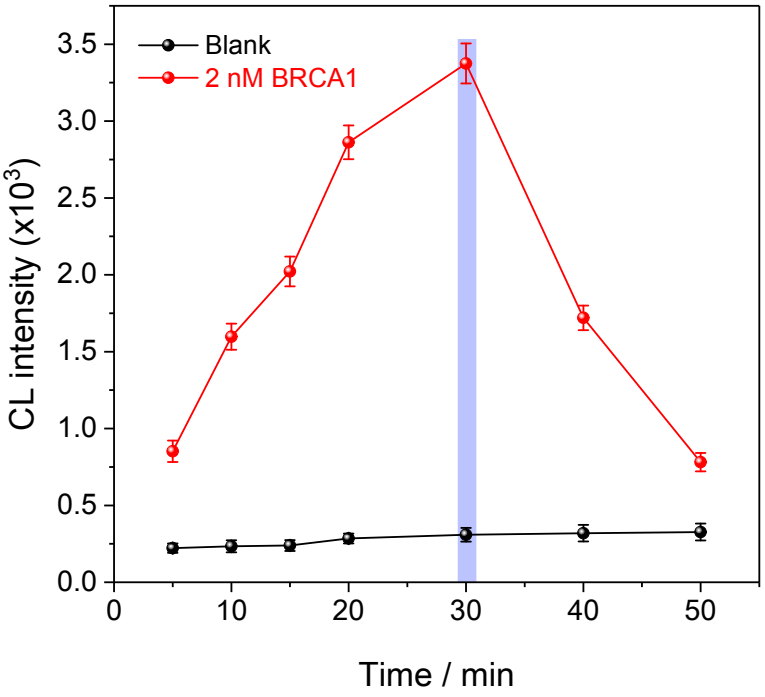

Figure S5. Optimization of the irradiation time for PMCL sensing.

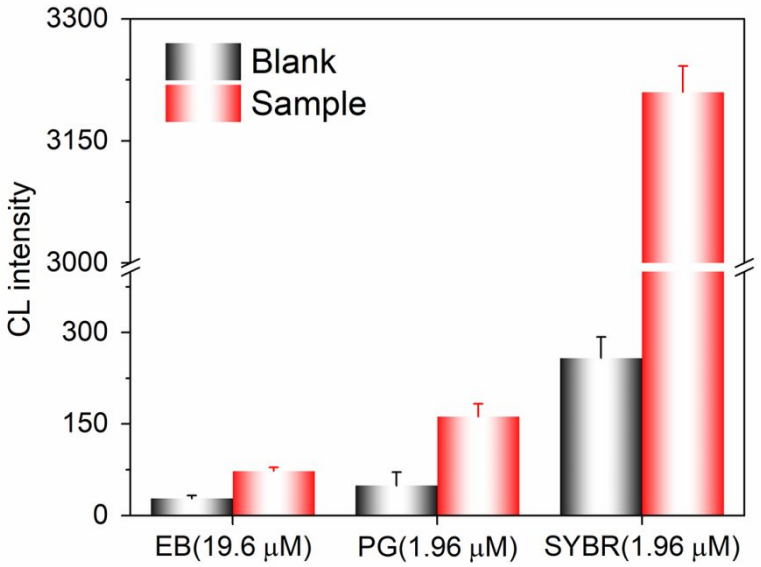

Figure S6. Comparison of the different dsDNA-staining dyes for PMCL. Experimental conditions: SG, 1.0 X; PG, 10 X ; EB, 19.6 μM; K4[Fe(CN)6], 25 μM; luminol, 1.0 mM; and luminol pH, 11.5.

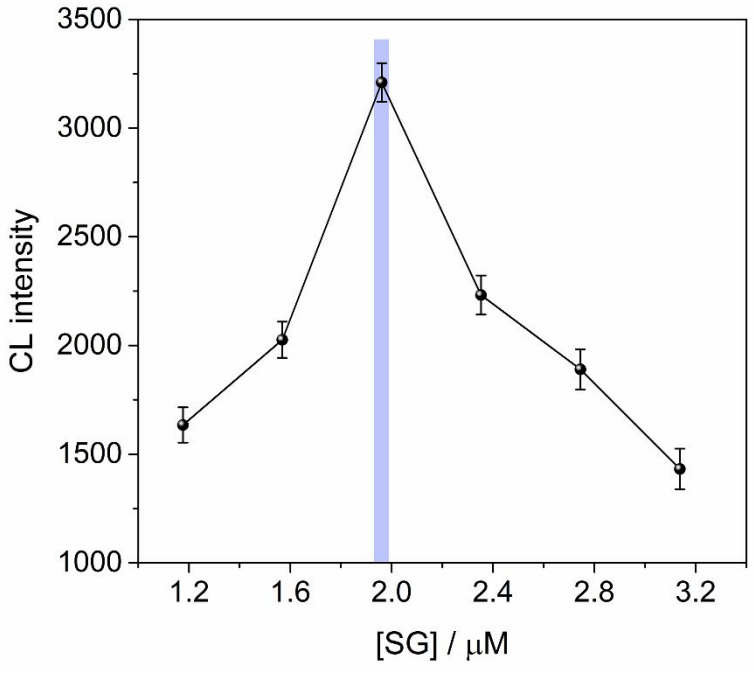

Figure S7. Optimization of the SG concentration for photosensitization.

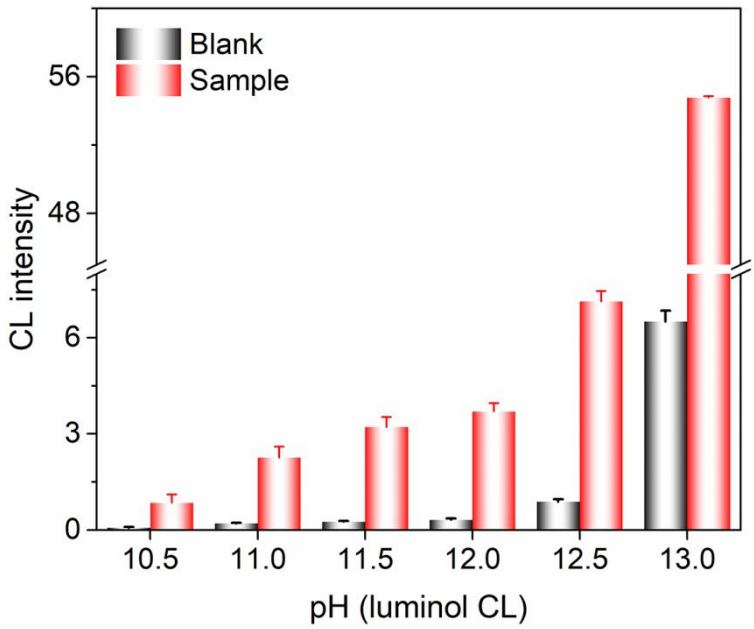

Figure S8. Investigation of pH on luminol CL reaction and PMCL performance. Experimental conditions: SG, 1.0 X; luminol, 1.0 mM.

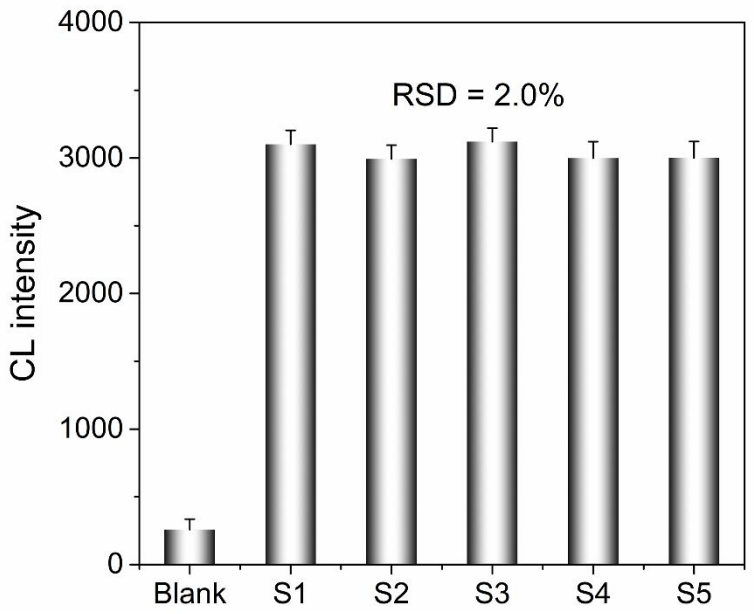

Figure S9. Reproducibility evaluation of the BRCA1 gene assay. Experimental conditions: BRCA1, 2.0 nM; CS-BRCA1, 5.0 nM; SG, 1.0×; $K_4Fe(CN)_6$, 25 μM; luminol, 1.0 mM.

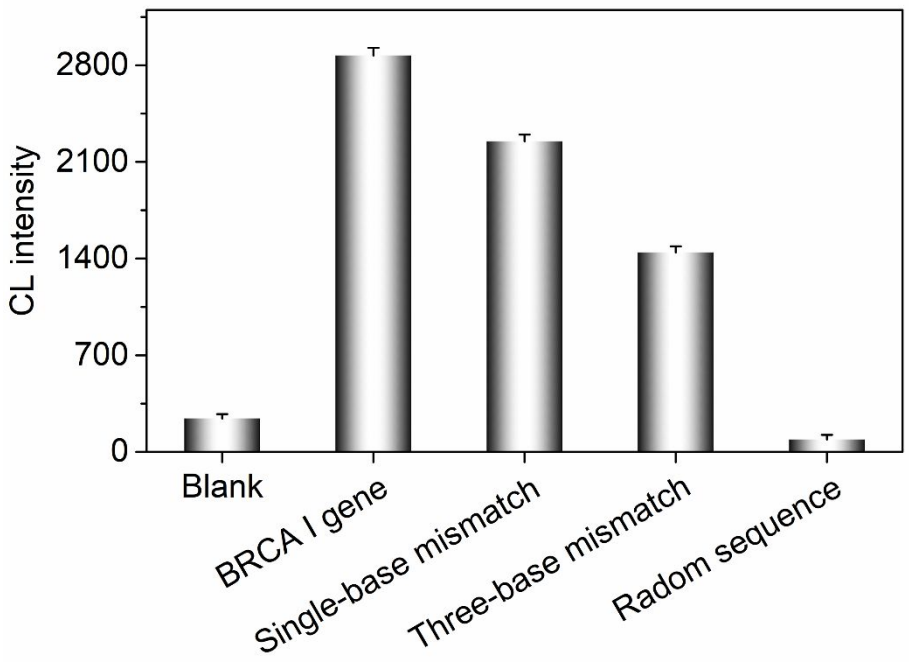

Figure S10. Specificity investigation of PMCL sensing of BRCA1 gene. Experimental conditions: BRCA1 gene, 2.0 nM; probe DNA, 5.0 nM; Other DNA, 2.0 nM; SG, 1.0 ×; $K_4[Fe(CN)_6]$, 25 μM; luminol, 1.0 mM.

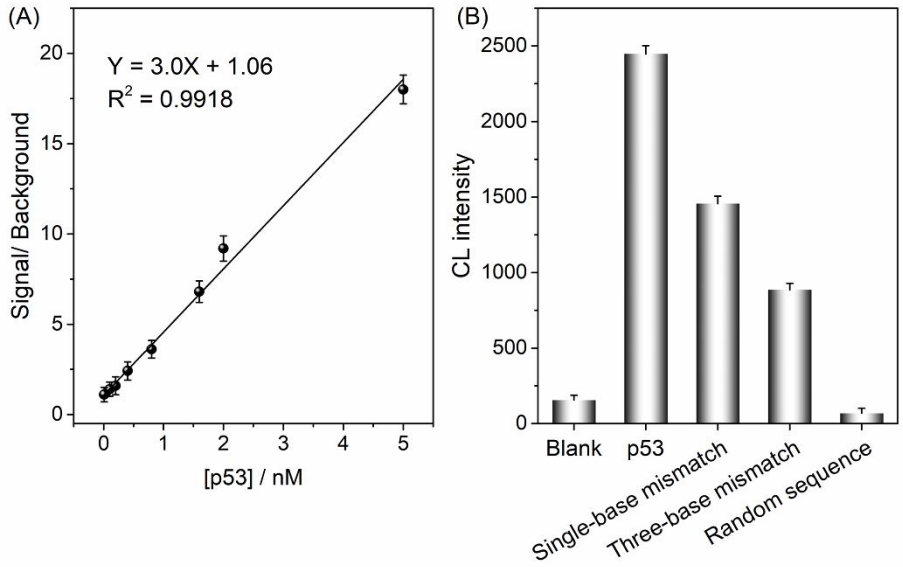

Figure S11. (A) The CL response of PMCL sensing system with the increase of p53 gene; and (B) Specificity investigation of PMCL sensing of p53 gene Experimental conditions: SG, 1.0 ×; $K_4[Fe(CN)_6]$, 25 μM; luminol, 1.0 mM; and luminol pH, 11.5.

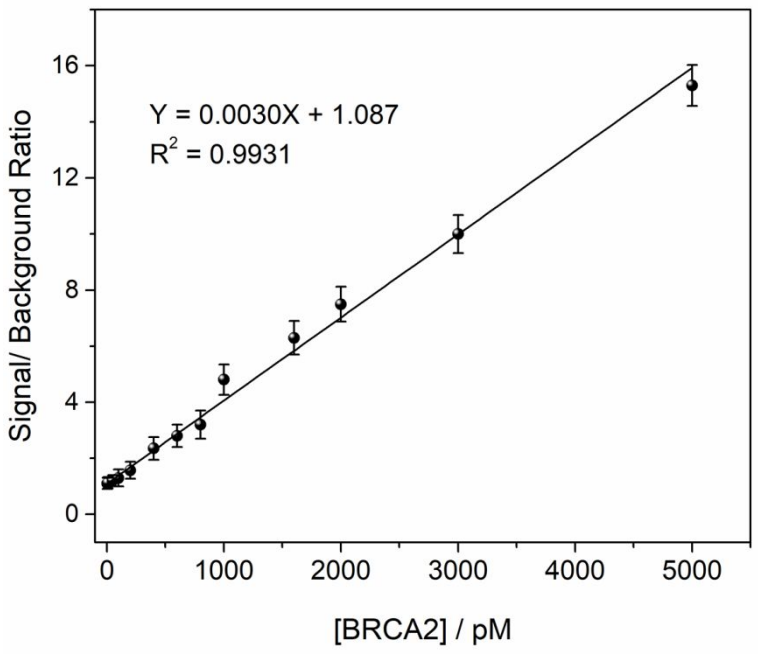

Figure S12. The CL response of PMCL sensing system with the increase of BRCAII gene. Experimental conditions: SG, 1.0 ×; $K_4[Fe(CN)_6]$, 25 μM; luminol, 1.0 mM; and luminol pH, 11.5.

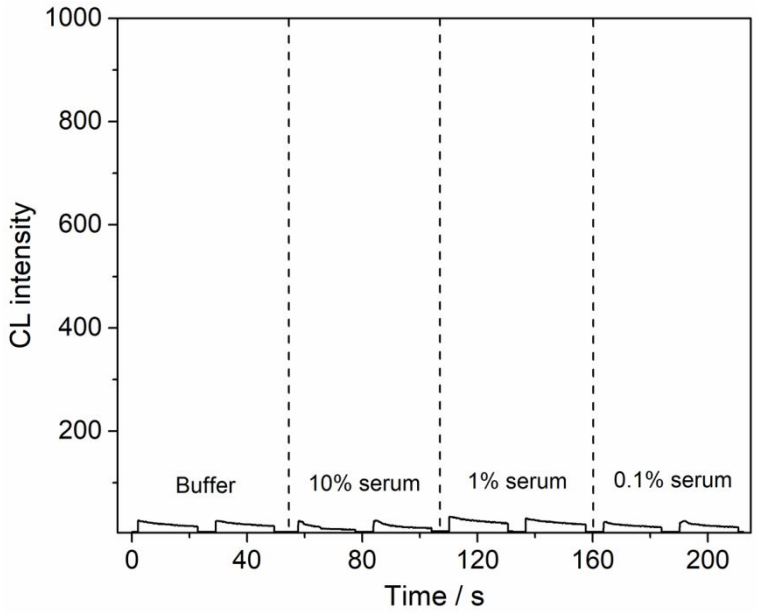

Figure S13. Potential interferences from endogenous photosensitizers on the PMCL performance.

Table S2 Comparison with other optical sensing methods for determination of DNA

strategy[a]	Detection Method	Amplification	Label/Tag	dynamic range	LOD	Ref
dsDNA-SG/ $K_4[Fe(CN)_6]$	CL	-	-	5.0 pM-5.0 nM	1.5 pM (0.45 fmol)	This work
dsDNA/SG	Fluorescence	-	-	400 pM-5 nM	150 pM	This work
Split DNAzyme	CL	-	-	100 pM-5 nM	33 pM	This work
Split DNAzyme	Colorimetry		-	10-300 nM	NM	[1]
Split DNAzyme	colorimetry		-	1-100 nM	NM	[2]
Caged DNAzyme	colorimetry		-	200 nM-4.3μM	NM	[3]
GQDs/dsDNA/ GO	fluorescence		-	6.7-46 nM	75 pM	[4]
MBs/HRP	CL/ CL imaging		HRP	0.3-300 pM	0.10 pM	[5]
CHA/DNAzyme	CL	CHA	-	0.25-25 nM	0.30 fmol	[6]

CHA/GO/AuNPs/ TMB	Colorimetry	CHA	-	0.1 -10 nM	57.4 pM	[7]
HCR/ AuNPs	Colorimetry	HCR	-	0.05-6 nM	50 pM	[8]
CHA/GO/FAM	fluorescence	CHA	FAM	0.4-5 nM	0.20 nM (0.1nmol)	[9]
CHA/ molecular beacon	fluorescence	CHA	FAM, DABCYL	1-50 nM	10 pM (5 fmol)	[10]
RCA/ molecular beacon	fluorescence	RCA	FAM. DABCYL	0.1-40 nM	1 pM	[11]
HCR/AuNPs	Colorimetry	HCR	AuNps	-	0.5 nM	[12]
RCA/DNAzyme	Colorimetry	RCA	-	1pM-5 nM	1 pM	[13]

[a]Abbreviations: LOD = limit of detection; NM = not measured; GO = graphene oxide; GQDs = graphene quantum dots; CHA = catalyzed hairpin assembly; FAM = fluorescein; MB = magnetic bead; HRP = Horseradish peroxidase。

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