

**Metal Sulfide Nanoparticle-Based dual bar-Codes triggered
DNAzyme Cascade for Multiplex miRNA detection in a Single Assay**

Jiao Wang,^{a#} Liyuan Wen,^{b#} Ruyuan Cao,^a Xiaorong Gao,^a Xia Li,^a Ensheng Xu,^a

Qi Zhang,^a Shuling Xu,^a Caifeng Dai,^{c*} Qingwang Xue,^{a*}

^a *School of Chemistry and Chemical Engineering, Liaocheng University, Liaocheng, 252059, Shandong, China*

^b *Earthquake Monitoring Center station of Liaocheng, Earthquake Administration of Shandong Province*

^c *Center for Reproductive Medicine, Department of Obstetrics and Gynecology, Qilu Hospital of Shandong University, Ji'nan 250012, Shandong, P.R. China*

[†] *Jiao Wang and Liyuan Wen contributed equally to this work.*

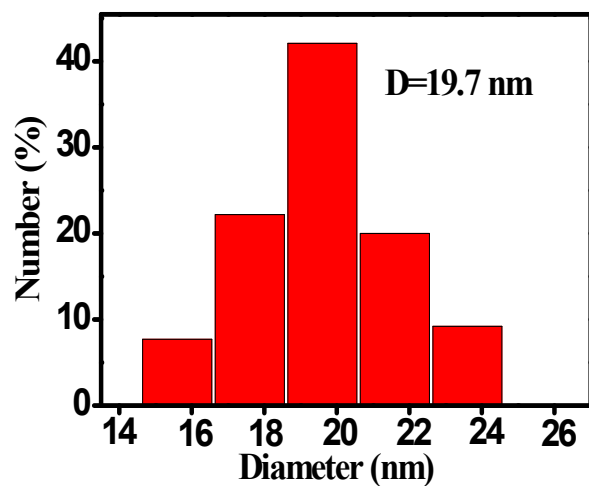
*Corresponding author:

Tel: +86-635-8239001; Fax: +86-635-8239001

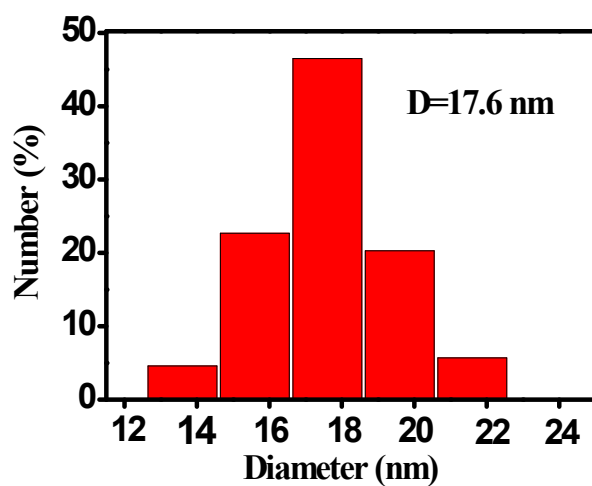
Email: xueqingwang1983@163.com; daicaifeng1982@163.com

Tabel S1 Sequence of oligonucleotides

Name	Sequence(5'-3')
hairpin 1 (H1)	5'- TCAACATCAGTCTGATAAGCTACCATCTGTTGATAGC TTATCAGACTGATCCAAGACACACGAC-3'-biotin
hairpin 2 (H2)	5'- CCATCTTTACCAGACAGTGTTACCTAGAGATGTTAAC ACTGTCTGGTAACCATAACTGTCCTG-3'-biotin
hairpin 3 (H3)	5'- TAAGCTATCAACAGATGGTAGCTTATCAGACTGATCC ATCTGTTGAGTA-3'-NH ₂
hairpin 4 (H4)	5'- GTGTTAACATCTCTAGGTAACACTGTCTGGTAACCTA GAGATGTGAT-3'-NH ₂
miRNA-21	5'-UAGCUUAUCAGACUGAUGUUGA-3'
miRNA-141	5'-UAACACUGUCUGGUAAAGAUGG-3'
miRNA-122	5'-UGGAGUGUGACAAUGGUGUUUG-3'
let-7d	5'-AGAGGUAGUAGGUUGCAUAGUU-3'
Zn ²⁺ -specific DNase (Zn- Enz)	5'- CATCTCTTCTCCGAGCCGGTTCGAAATAGTGAGTAAA AAAAAA-3'
Zn-Sub (MB1)	FAM-5'- CCACCACATTCAAATTCACCAACTATrAGGAAGAGAT GTTACGAGGCGGTGGTGG-3'-BHQ-1
Cu ²⁺ -specific DNase (Cu-Enz)	5'- GGTAAGCCTGGGCCTCTTTCTTTTAAAGAAAGAAGTT -3'
Cu-Sub (MB2)	Cy3-5'- CACCACCATACTTTTCAACTTCTTTCTTATACGGCTTA CCGCTCGGGGGTGGTG-3'-BHQ-2



(A)

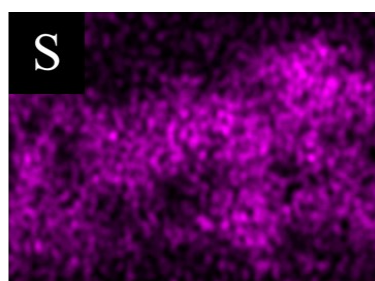


(B)

Fig. S1 A histogram of size distribution of metal sulfide nanoparticles; **(A)** CuS NPs, **(B)** ZnS NPs (n=50)



(A)



(C)

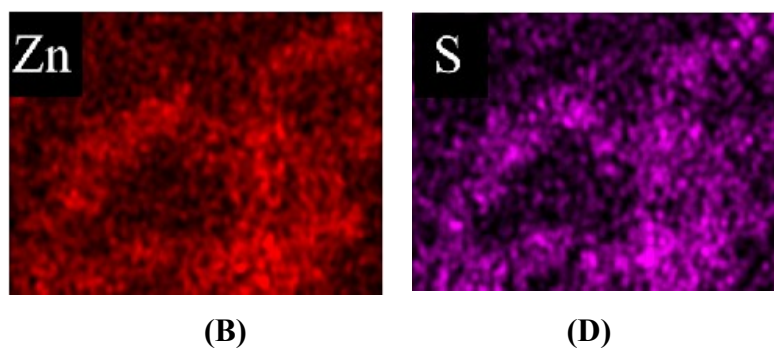
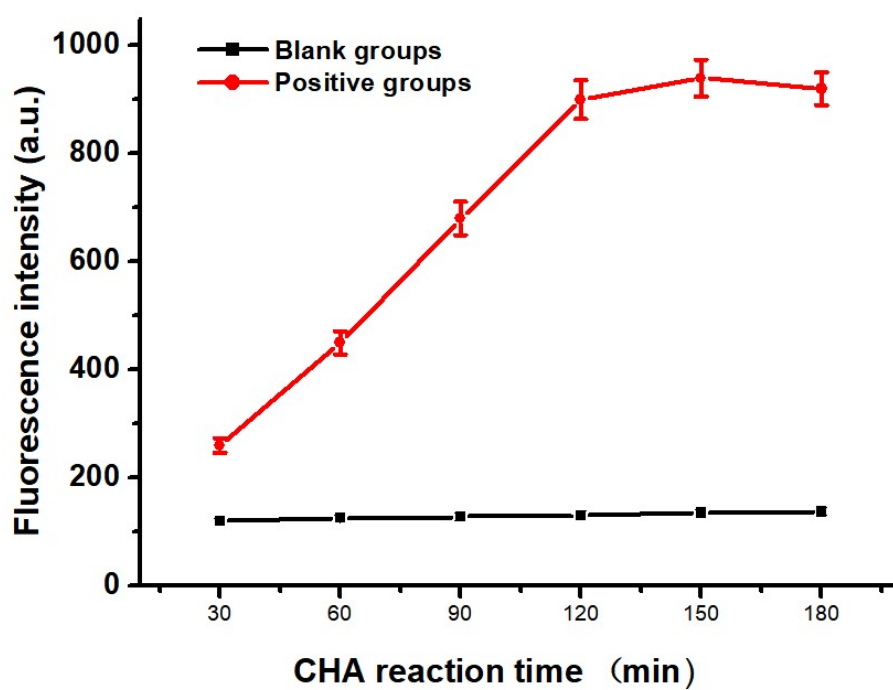
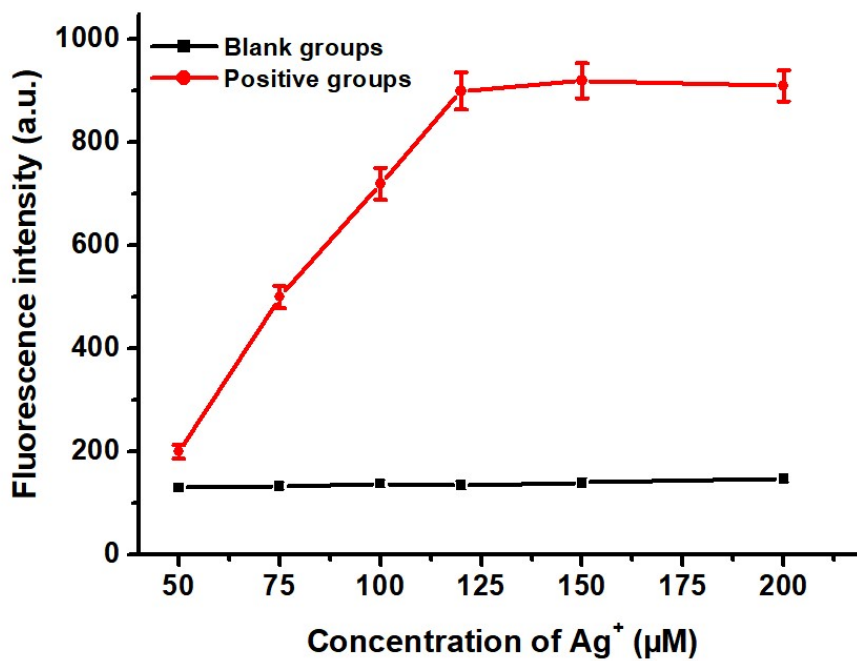


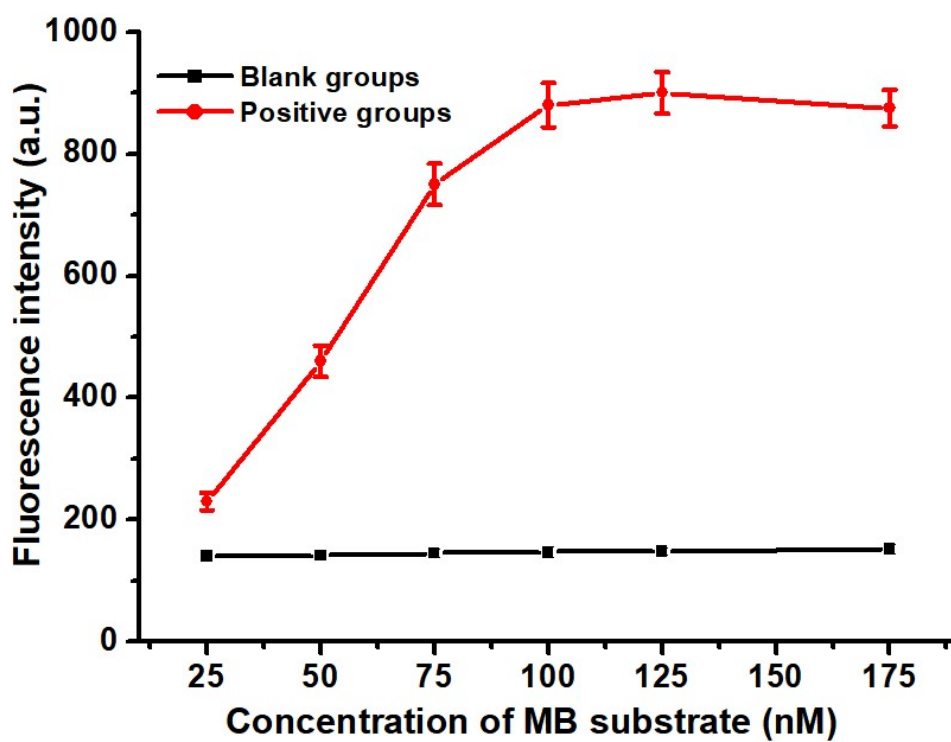
Fig. S2 the corresponding elemental mapping of (C) Cu, (D) Zn, and (E-F) S for SEM image of CuS and ZnS nanoparticles (Fig. 1), respectively.



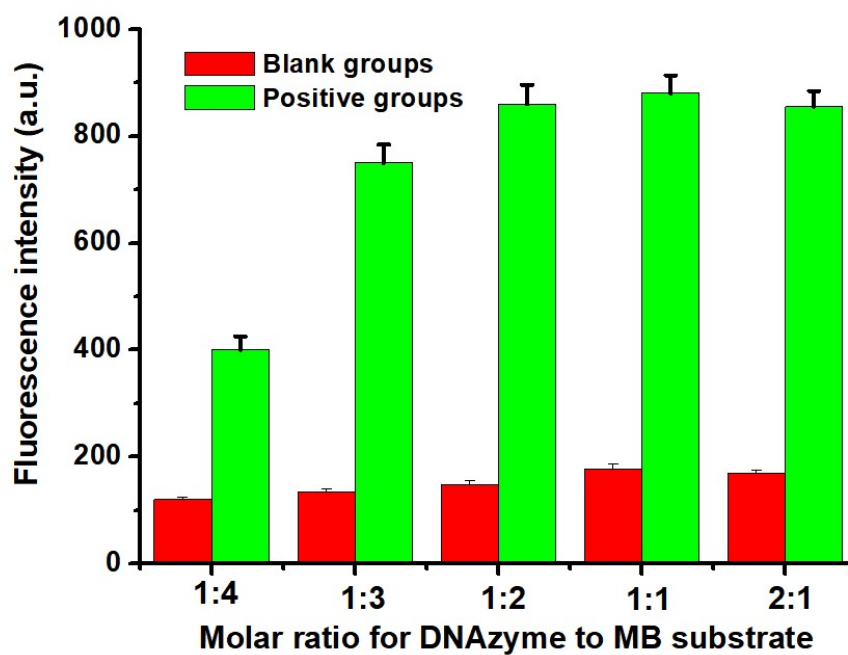
(A)



(B)



(C)



(D)

Fig. S3 Variance of the fluorescence intensity with (A) the CHA reaction time, (B) Ag^+ concentration, (C) MB substrate concentration, (D) molar ratio for the DNAzyme to MB substrate in the presence of 30 pM miRNA-141.