

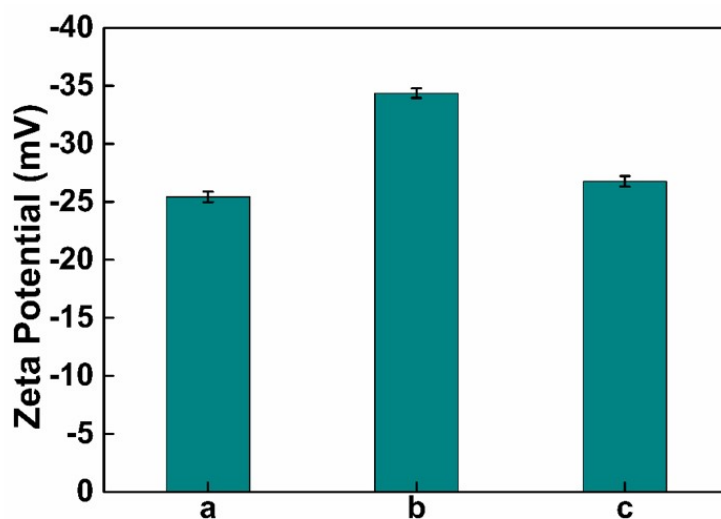
## **Electronic Supplementary Material**

# **Colorimetric determination of tetracyclines based on aptamer-mediated dual regulation of gold nanoparticle aggregation and in situ silver metallization**

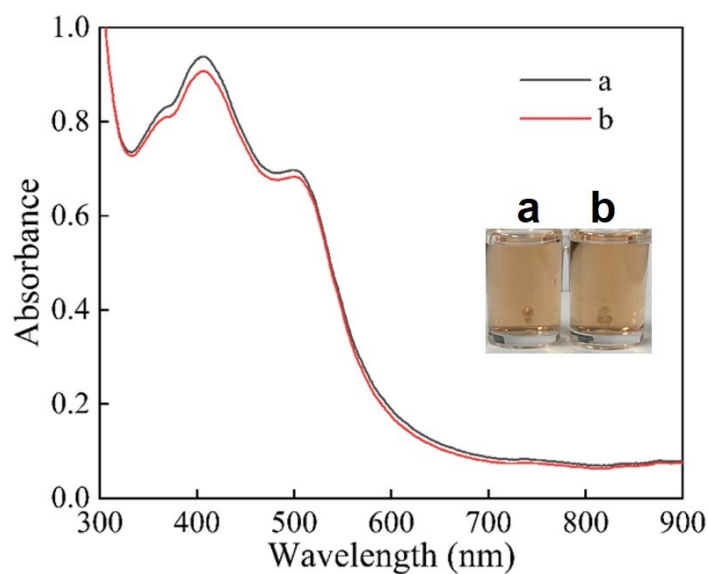
Dingmin Liu, Pengcheng Huang\*, Fang-Ying Wu\*

College of Chemistry, Nanchang University, Nanchang 330031, China

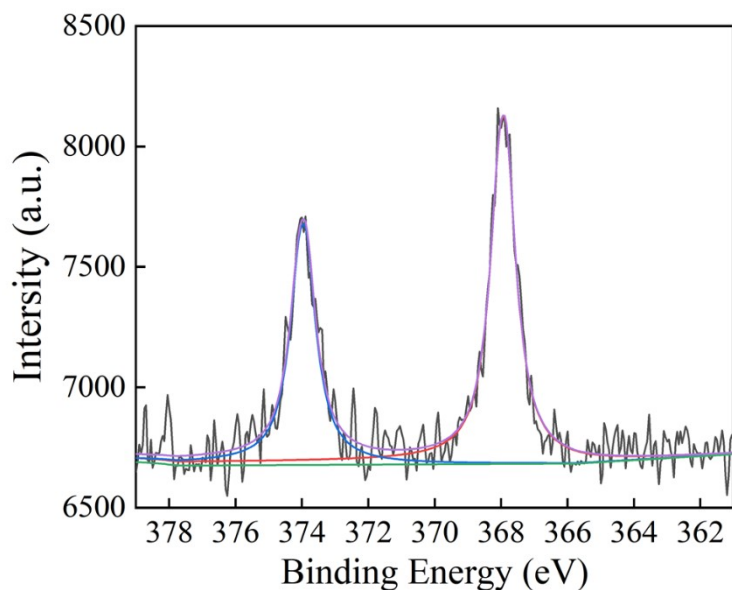
\*Corresponding Author: Pengcheng Huang, [pchuang@ncu.edu.cn](mailto:pchuang@ncu.edu.cn), Fang-Ying Wu, [fywu@ncu.edu.cn](mailto:fywu@ncu.edu.cn), Tel: + 86 79183969514, Fax: + 86 79183969514.



**Fig. S1**  $\zeta$ -potential analysis of different mixtures. (a) AuNPs (b) Apts+AuNPs (c) Apts+Mg(NO<sub>3</sub>)<sub>2</sub>+AuNPs+TET. Substance concentration: 0.0317  $\mu\text{mol}\cdot\text{L}^{-1}$  AuNPs, 0.08  $\mu\text{mol}\cdot\text{L}^{-1}$  Apts, 500  $\mu\text{mol}\cdot\text{L}^{-1}$  Mg(NO<sub>3</sub>)<sub>2</sub>, 4  $\mu\text{mol}\cdot\text{L}^{-1}$  TET



**Fig. S2** UV-vis absorption spectra of different mixtures and the corresponding photographs. (a) Random Apts+AuNPs+p-aminophenol+AgNO<sub>3</sub>. (b) Random Apts+Mg(NO<sub>3</sub>)<sub>2</sub>+AuNPs +TET+p-aminophenol+AgNO<sub>3</sub>. Substance concentration: 120  $\mu\text{mol}\cdot\text{L}^{-1}$  AgNO<sub>3</sub>, 120  $\mu\text{mol}\cdot\text{L}^{-1}$  p-aminophenol, 0.0317  $\mu\text{mol}\cdot\text{L}^{-1}$  AuNPs, 0.08  $\mu\text{mol}\cdot\text{L}^{-1}$  Apts, 4  $\mu\text{mol}\cdot\text{L}^{-1}$  TET, 500  $\mu\text{mol}\cdot\text{L}^{-1}$  Mg(NO<sub>3</sub>)<sub>2</sub>

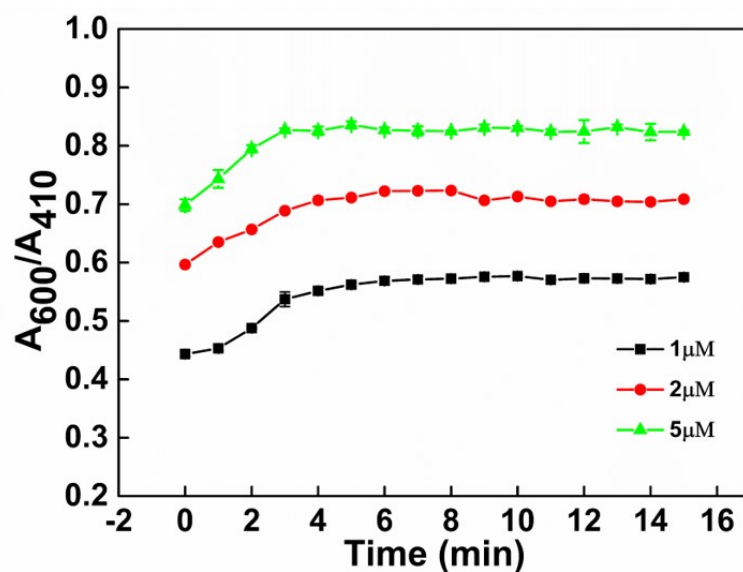


**Fig. S3** High resolution XPS spectrum for Ag3d

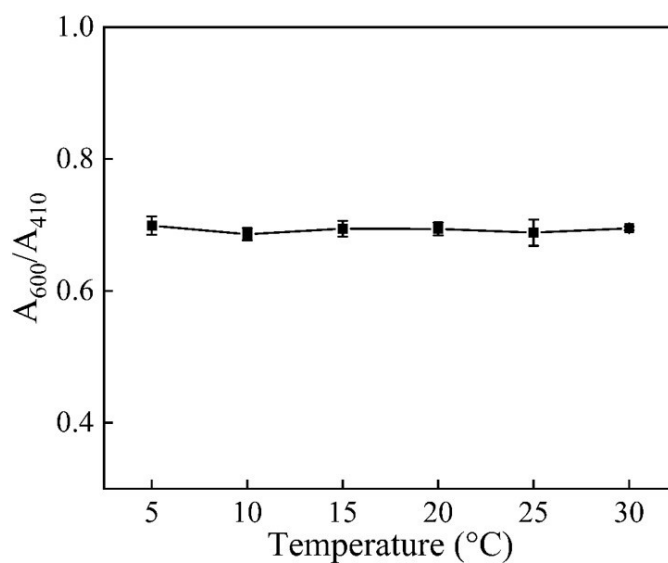
In 9 groups of experiments, by averaging the  $\Delta(A_{600}/A_{410})$  obtained for each concentration of each factor. It is not difficult to find that the optimal levels of  $Mg^{2+}$ , Apt, and  $Ag^+$  are  $500 \mu\text{mol}\cdot\text{L}^{-1}$ ,  $0.08 \mu\text{mol}\cdot\text{L}^{-1}$ , and  $120 \mu\text{mol}\cdot\text{L}^{-1}$ , respectively. At this concentration, we measured  $\Delta(A_{600}/A_{410}) = 0.60$ . This is better than the previous 9 sets of experiments. So  $Mg^{2+} = 500 \mu\text{mol}\cdot\text{L}^{-1}$ , Apt =  $0.08 \mu\text{mol}\cdot\text{L}^{-1}$ , Ag =  $120 \mu\text{mol}\cdot\text{L}^{-1}$  were determined.

**Table S1** Orthogonal experiment arrangement and results

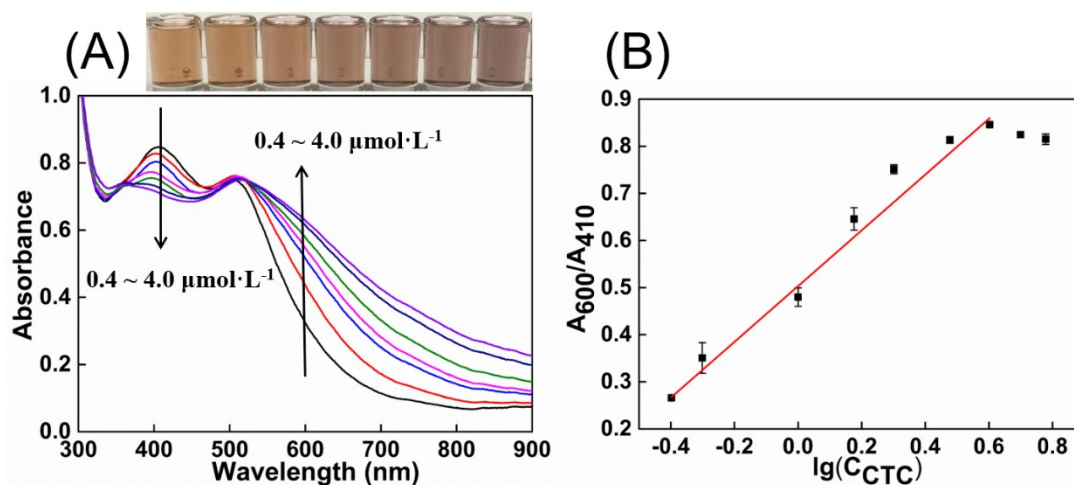
Experiment number	Factors			$\Delta(A_{600}/A_{410})$
	$Mg^{2+}$ ( $\mu\text{mol}\cdot\text{L}^{-1}$ )	Apt ( $\mu\text{mol}\cdot\text{L}^{-1}$ )	$Ag^+$ ( $\mu\text{mol}\cdot\text{L}^{-1}$ )	
1	400	0.08	80	0.56
2	400	0.10	100	0.47
3	400	0.12	120	0.39
4	500	0.08	100	0.52
5	500	0.10	120	0.50
6	500	0.12	80	0.43
7	600	0.08	120	0.39
8	600	0.10	100	0.38
9	600	0.12	80	0.47
Excellent level	500	0.08	120	0.60



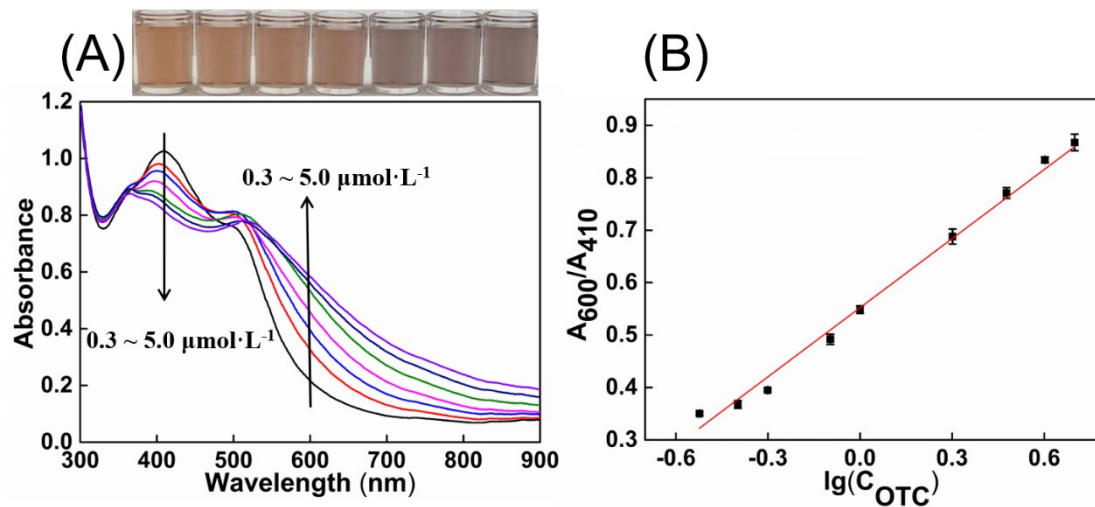
**Fig. S4** When the TET concentrations are  $1\mu\text{mol}\cdot\text{L}^{-1}$ ,  $2\mu\text{mol}\cdot\text{L}^{-1}$ ,  $5\mu\text{mol}\cdot\text{L}^{-1}$ , the values of  $A_{600}/A_{410}$  change with time. Substance concentration:  $120\mu\text{mol}\cdot\text{L}^{-1}$  p-aminophenol,  $120\mu\text{mol}\cdot\text{L}^{-1}$   $\text{AgNO}_3$ ,  $0.0317\mu\text{mol}\cdot\text{L}^{-1}$  AuNPs,  $0.08\mu\text{mol}\cdot\text{L}^{-1}$  Apts,  $500\mu\text{mol}\cdot\text{L}^{-1}$   $\text{Mg}(\text{NO}_3)_2$



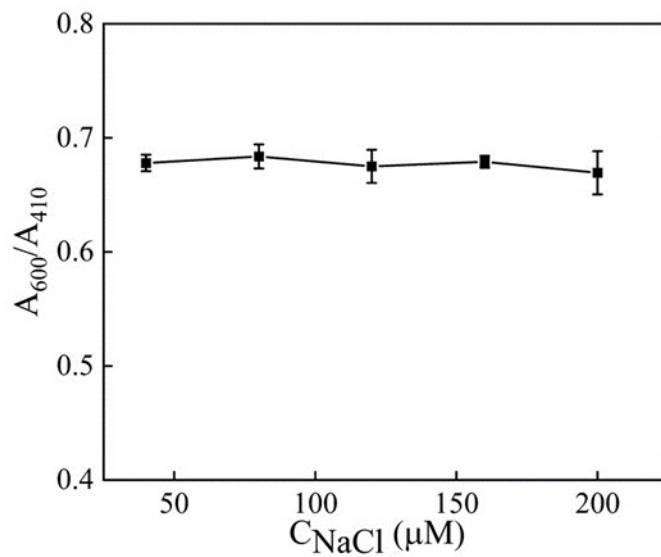
**Fig. S5** When the TET concentrations is  $2\mu\text{mol}\cdot\text{L}^{-1}$ , the values of  $A_{600}/A_{410}$  change with time. Substance temperature:  $120\mu\text{mol}\cdot\text{L}^{-1}$  p-aminophenol,  $120\mu\text{mol}\cdot\text{L}^{-1}$   $\text{AgNO}_3$ ,  $0.0317\mu\text{mol}\cdot\text{L}^{-1}$  AuNPs,  $0.08\mu\text{mol}\cdot\text{L}^{-1}$  Apts,  $500\mu\text{mol}\cdot\text{L}^{-1}$   $\text{Mg}(\text{NO}_3)_2$



**Fig. S6** (A) The UV-vis absorption spectra of the experiment at different concentrations of CTC ( $0.4, 0.8, 1, 2, 3, 4 \mu\text{mol}\cdot\text{L}^{-1}$ ) and the corresponding photographs. The upper illustration shows the photos of the corresponding concentrations. (B) Fitting curve of  $A_{600}/A_{410}$  and  $\lg(C_{\text{CTC}})$ . Error bars represent the standard deviation of three independent measurements. Substance concentration:  $120 \mu\text{mol}\cdot\text{L}^{-1}$  p-aminophenol,  $120 \mu\text{mol}\cdot\text{L}^{-1}$   $\text{AgNO}_3$ ,  $0.0317 \mu\text{mol}\cdot\text{L}^{-1}$  AuNPs,  $0.08 \mu\text{mol}\cdot\text{L}^{-1}$  Apts,  $500 \mu\text{mol}\cdot\text{L}^{-1}$   $\text{Mg}(\text{NO}_3)_2$



**Fig. S7** (A) The UV-vis absorption spectra of the experiment at different concentrations of OTC ( $0.3, 1, 2, 3, 4, 5 \mu\text{mol}\cdot\text{L}^{-1}$ ) and the corresponding photographs. The upper illustration shows the photos of the corresponding concentrations. (B) Fitting curve of  $A_{600}/A_{410}$  and  $\lg(C_{\text{OTC}})$ . Error bars represent the standard deviation of three independent measurements. Substance concentration:  $120 \mu\text{mol}\cdot\text{L}^{-1}$  p-aminophenol,  $120 \mu\text{mol}\cdot\text{L}^{-1}$   $\text{AgNO}_3$ ,  $0.0317 \mu\text{mol}\cdot\text{L}^{-1}$  AuNPs,  $0.08 \mu\text{mol}\cdot\text{L}^{-1}$  Apts,  $500 \mu\text{mol}\cdot\text{L}^{-1}$   $\text{Mg}(\text{NO}_3)_2$



**Fig. S8** Effect of salt concentration on TCs detection, NaCl+Apts+Mg(NO<sub>3</sub>)<sub>2</sub>+AuNPs +TET+p-aminophenol+AgNO<sub>3</sub>. Substance concentration: 120  $\mu\text{mol}\cdot\text{L}^{-1}$  AgNO<sub>3</sub>, 120  $\mu\text{mol}\cdot\text{L}^{-1}$  p-aminophenol, 0.0317  $\mu\text{mol}\cdot\text{L}^{-1}$  AuNPs, 0.08  $\mu\text{mol}\cdot\text{L}^{-1}$  Apts, 2  $\mu\text{mol}\cdot\text{L}^{-1}$  TET, 500  $\mu\text{mol}\cdot\text{L}^{-1}$  Mg(NO<sub>3</sub>)<sub>2</sub>

**Table S2** Comparison of different TET determination methods in analytical performances

<b>Probe</b>	<b>Procedure</b>	<b>Time/min</b>	<b>Linear range</b>	<b>LOD (nmol·L<sup>-1</sup>)</b>	<b>References</b>
AuNPs@Apt	AuNPs aggregation using NaCl	10	50 nM~3.0 μM	32.9	<sup>1</sup>
AuNPs@Apt	AuNPs aggregation using CTAB	30	10 nM~2 μM	122	<sup>2</sup>
AuNPs@Apt	AuNPs aggregation using NaCl	20	100 nM~5 μM	71	<sup>3</sup>
Enzyme	-	6	100 nM~10 μM	60	<sup>4</sup>
D-Trp-OMe@AuNCs	-	20	1.5μM ~30 μM	200	<sup>5</sup>
DNAzyme	-	17	11 nM~1 μM	3.1	<sup>6</sup>
AuNPs@Apt	AuNPs aggregation and silver metallization	5	400 nM~6 μM	40.88	This work

AuNPs= Gold nanoparticles, Apt= Aptamer, AuNCs= Gold cluster, D-Trp-OMe= D-tryptophane methyl ester. CTAB= hexadecyltrimethylammonium bromide.

## References

1. Y.-Y. Wu, P. Huang, F.-Y. Wu, *Food Chem.* , 2020 , **304**, 125377
2. L. He, Y. Luo, W. Zhi, Y. Wu, P. Zhou, *Aust. J. Chem.* , 2013, **66**, 485-490
3. M. Qi, C. Tu, Y. Dai, W. Wang, A. Wang, J. Chen, *Anal. Methods*, 2018, **10**, 3402-3407
4. M. Besharati, M.-A. Tabrizi, F. Molaabasi, R. Saber, M. Shamsipur, J. Hamed, S. Hosseinkhan., *Biotechnol. Appl. Biochem.*, 2020, DOI: 10.1002/bab.2078
5. Y. Song, J. Qiao, W. Liu, L. Qi, *Microchem. J.*, 2020. **157**, 104871
6. Y. Tang, X. Huang, X. Wang, C. Wang, H. Tao, Y. Wu, *Food Chem.*, 2022, **366**, 130560