

Supporting Information

Aptazymes-induced cascade amplification integrated with volumetric bar-chart chip for highly sensitive detection of aflatoxin B1 and adenosine triphosphate

Yuanyue Han,^a Xinli Liu,^b Qiao Zhao,^a Yanfeng Gao,^b Dongtao Zhou,^b Wenxiu Long,^a Yuzhen Wang^{*a} and Yujun Song^{*b}

^a Key Laboratory of Flexible Electronics & Institute of Advanced Materials, Jiangsu National Synergistic Innovation Center for Advanced Materials, Nanjing Tech University, Nanjing 211816, China

^b College of Engineering and Applied Sciences, State Key Laboratory of Analytical Chemistry for Life Science, Nanjing University, Nanjing 210023, China

* Corresponding authors:

Yuzhen Wang, E-mail: iamyzwang@njtech.edu.cn

Yujun Song, E-mail: ysong@nju.edu.cn

Experimental Section

Materials and chemicals: Aflatoxin B1 (AFB1), aflatoxins B2 (AFB2), aflatoxins G1 (AFG1), aflatoxins G2 (AFG2), 2-methyl silicon oil and 1H, 1H, 2H, 2H-perfluorooctyltrichlorosilane were obtained from Macklin Biotechnical Co., Ltd. (Shanghai, China). Adenosine triphosphate (ATP), cytidine triphosphate (CTP), guanosine triphosphate (GTP), uridine triphosphate (UTP), $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, sodium acetate, H_2PtCl_6 , sodium citrate, tetraethylorthosilicate (TEOS), 3-(aminopropyl) trimethoxysilane (APTES), polyethylene glycol (PEG) and dopamine hydrochloride were purchased from Aladdin Chemical Co., Ltd. (Shanghai, China). The AFB1 ELISA kit was purchased from Meimian Biotechnology Co., Ltd. (Shanghai, China). Bovine serum albumin (BSA) was obtained from Sangon Biotechnology Co., Ltd. (Shanghai, China). 4-mercaptophenylboric acid (4-MPBA) was purchased from Energy Chemical. (Shanghai, China). Hydrogen peroxide (H_2O_2) solution (30 wt. % in H_2O) and glutaraldehyde (25% v/v) were obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). Amorphous diamond-coated drill bits with a diameter of 1.0 mm and a CNC 4303 engraving and milling machine were purchased from Jing Yan Instruments Technology Co., Ltd. (Shenzhen, China). All device graphics were designed using Auto CAD software and then printed by Ji Xian Guang Dian Co., Ltd. (Shenzhen, China) into transparency photomasks with a resolution of 10 μm . SPR220-7 photoresist and AZ400K developer were purchased from Suzhou Research Materials Microtech Co., Ltd. (Suzhou, China).

Supplementary Figures and Tables

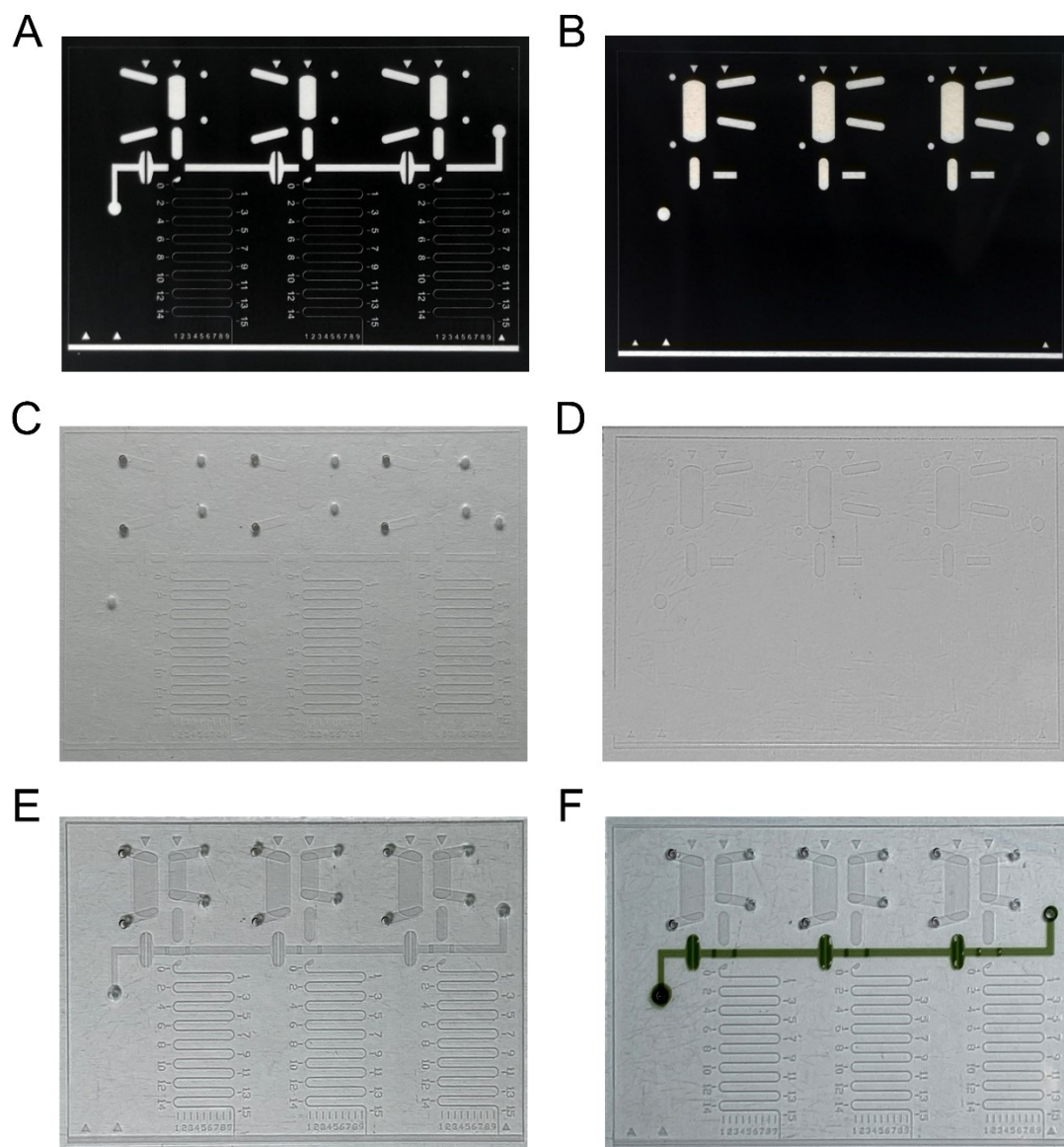


Fig. S1 Fabrication of TV-Chip. Masks used for the preparation of TV-Chip (A) top plate and (B) bottom plate. Photographs of TV-Chip (C) top plate and (D) bottom plate. Photographs of (E) a typical assembled TV-Chip and (F) a loaded TV-Chip with three independent analysis units in each chip.

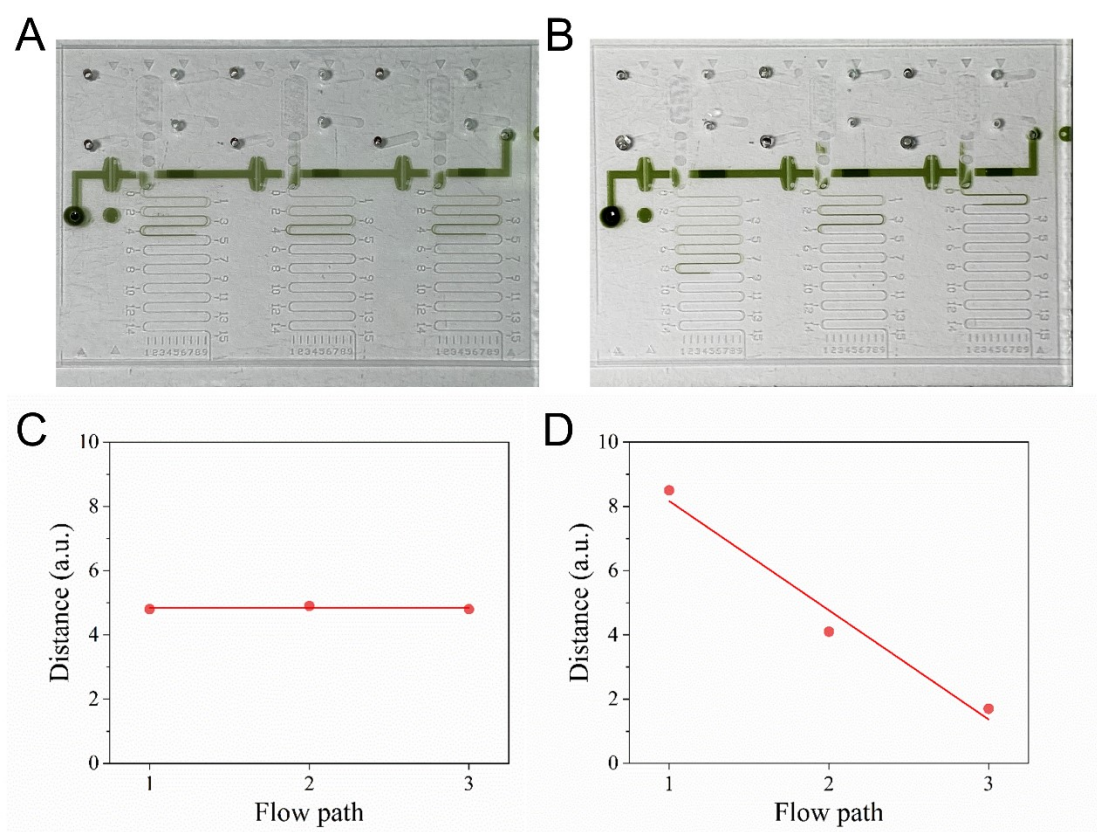


Fig. S2 The homogeneity and quantification of TV-Chip. Photograph (A) and corresponding functional diagram (C) loading PtNPs with the same concentration into three reaction wells of the TV-Chip ($10 \mu\text{g mL}^{-1}$). Photograph (B) and corresponding functional diagram (D) loading different concentrations of PtNPs into three reaction wells of the TV-Chip (from left to right, 15, 10 and $5 \mu\text{g mL}^{-1}$).

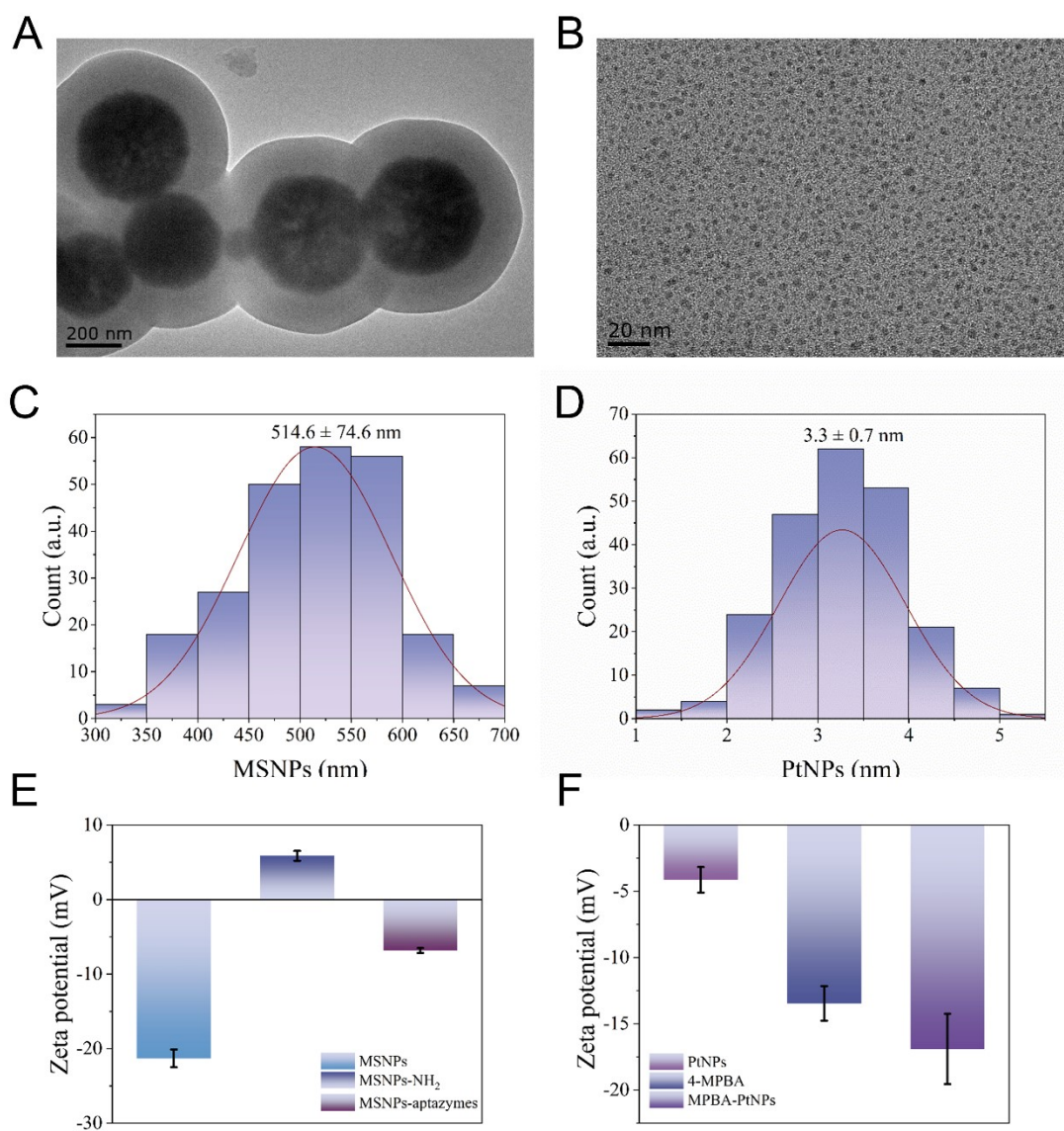


Fig. S3 Characterization of MSNPs and PtNPs. TEM images of (A) MSNPs and (B) PtNPs. Particle size distribution of (C) MSNPs and (D) PtNPs. (E) Zeta potentials of MSNPs, MSNPs-NH₂ and MSNPs-aptazymes. (F) Zeta potentials of PtNPs, 4-MPBA and MPt.

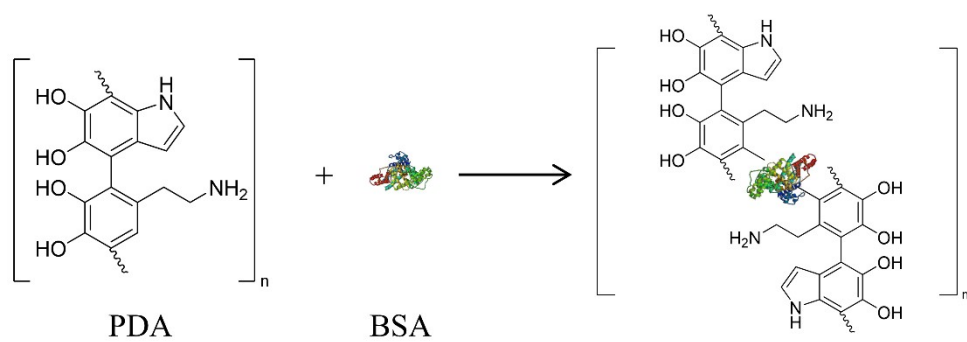


Fig. S4 Schematic representation of the PDA reaction with BSA.

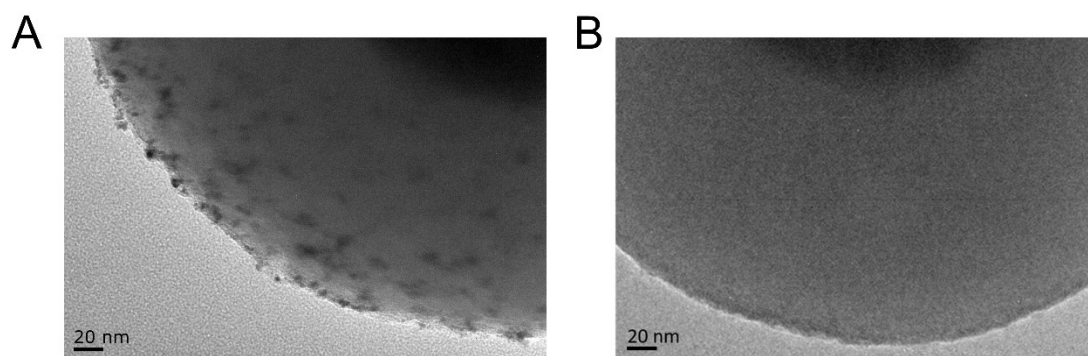


Fig. S5 TEM images of (A) PDA-deposited MSNP probe + MPt and (B) PDA-deposited MSNP probe + Pt, with scale bars corresponding to 20 nm.

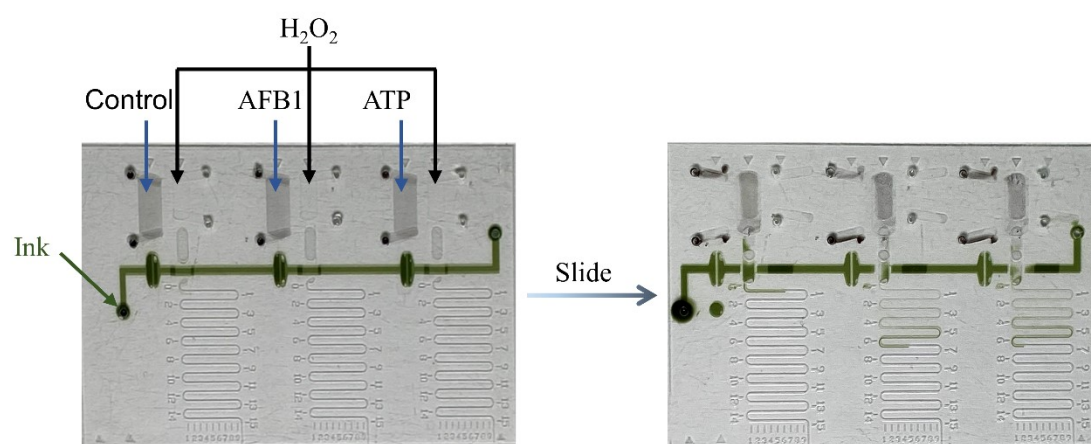


Fig. S6 Photographs of the TV-Chip used for AFB1 and ATP detection.

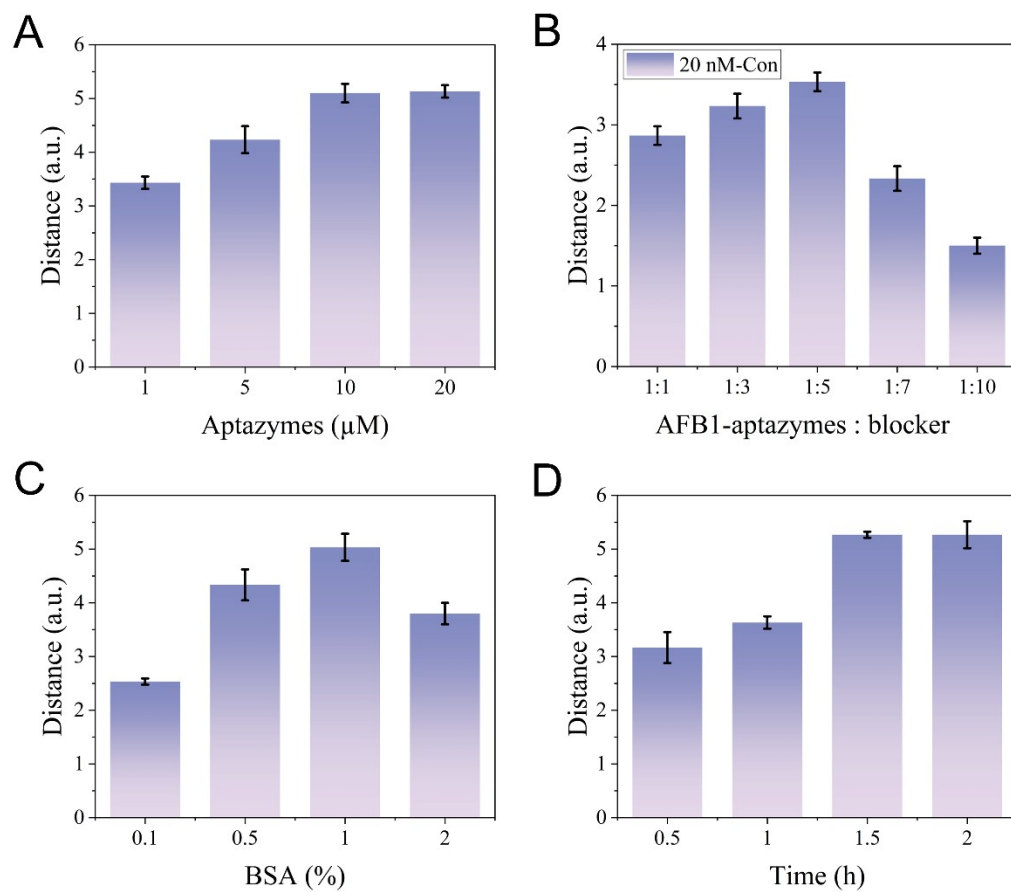


Fig. S7 Optimization of the experimental conditions: (A) the concentration of aptazymes, (B) the ratio of AFB1-aptazymes to blocker, (C) the concentration of BSA, (D) incubation time with the targets.

Table S1 Comparisons with other reported methods for AFB1 and ATP detection.

Target	Method	LOD (nM)	Reference
AFB1	Fluorescence based on aptamer induced assembly of nitrogen-doped carbon dots on gold nanoparticles	0.016	1
	Fluorescence based on aptamer/G-quadruplex DNAzyme probe	0.064	2
	Colorimetry based on gold nanoparticles	0.112	3
	Colorimetry based on an ingenious hairpin DNA probe integrated with exonuclease III-assisted signal amplification.	0.0001	4
	Colorimetry based on the Fe ₃ O ₄ /GO platform	16	5
	TV-Chip based on aptazymes-induced cascade signal amplification (ACSA)	0.000075	This work
ATP	Fluorescence based on a dual-function oligonucleotide	24.8	6
	Fluorescence based on CRISPR-Cas12a	400	7
	Fluorescence based on allosteric probe-conjugated strand displacement and CRISPR/Cpf1 trans-cleavage	0.0018	8
	Fluorescence based on gold nanorods coupled with enzyme assisted target recycling amplification	0.026	9
	Colorimetry based on the use of a magnesium (II)-dependent DNAzyme	0.0053	10
	Colorimetry based on Fe ₃ O ₄ nanozymes with aptamer-tuned catalysis	90	11
	TV-Chip based on aptazymes-induced cascade signal amplification (ACSA)	0.000818	This work

Supplementary References

1. B. Wang, Y. Chen, Y. Wu, B. Weng, Y. Liu, Z. Lu, C. M. Li and C. Yu, *Biosens. Bioelectron.*, 2016, 78, 23-30.
2. L. Wang, F. Zhu, M. Chen, Y. Zhu, J. Xiao, H. Yang and X. Chen, *Food Chem.*, 2019, 271, 581-587.
3. J. Qian, C. Ren, C. Wang, K. An, H. Cui, N. Hao and K. Wang, *Biosens. Bioelectron.*, 2020, 166, 112443.
4. J. Wu, L. Zeng, N. Li, C. Liu and J. Chen, *Food Chem.*, 2019, 298, 125034.
5. W. Zhu, L. Li, Z. Zhou, X. Yang, N. Hao, Y. Guo and K. Wang, *Food Chem.*, 2020, 319, 126544.
6. Y.-Y. Fan, X. Deng, M. Wang, J. Li and Z.-Q. Zhang, *Talanta*, 2020, 219, 121349.
7. L. Peng, J. Zhou, G. Liu, L. Yin, S. Ren, S. Man and L. Ma, *Sens. Actuators, B*, 2020, 320, 128164.
8. X. Wang, X. Chen, C. Chu, Y. Deng, M. Yang, Z. Ji, F. Xu, D. Huo, Y. Luo and C. Hou, *Sens. Actuators, B*, 2020, 323, 128653.
9. N. Xue, S. Wu, Z. Li and X. Miao, *Anal. Chim. Acta*, 2020, 1104, 117-124.
10. S. Zhu, X. Wang, C. Jing, Y. Yin and N. Zhou, *Microchim. Acta*, 2019, 186, 176.
11. S. Li, X. Zhao, X. Yu, Y. Wan, M. Yin, W. Zhang, B. Cao and H. Wang, *Anal. Chem.*, 2019, 91, 14737-14742.