

Supporting Information

Suppressed DNA Base Pair Stacking Assembly of Gold
Nanoparticles in an Alcoholic Solvent for Enhanced Ochratoxin
A Detection in Baijiu

Zhiyu He,^a Qianyuan Chen,^a Shansen Ding,^a Guoqing Wang,^{*,a,b} Tohru Takarada,^c

Mizuo Maeda^c

^a*College of Food Science and Engineering, Ocean University of China, 5 Yushan
Road, Qingdao 266003, China*

^b*Laboratory for Marine Drugs and Bioproducts, Pilot National Laboratory for
Marine Science and Technology (Qingdao), Qingdao 266237, China*

^c*RIKEN Cluster for Pioneering Research, 2-1 Hirosawa, Wako, Saitama 351-0198,
Japan*

Email: gqwang@ouc.edu.cn

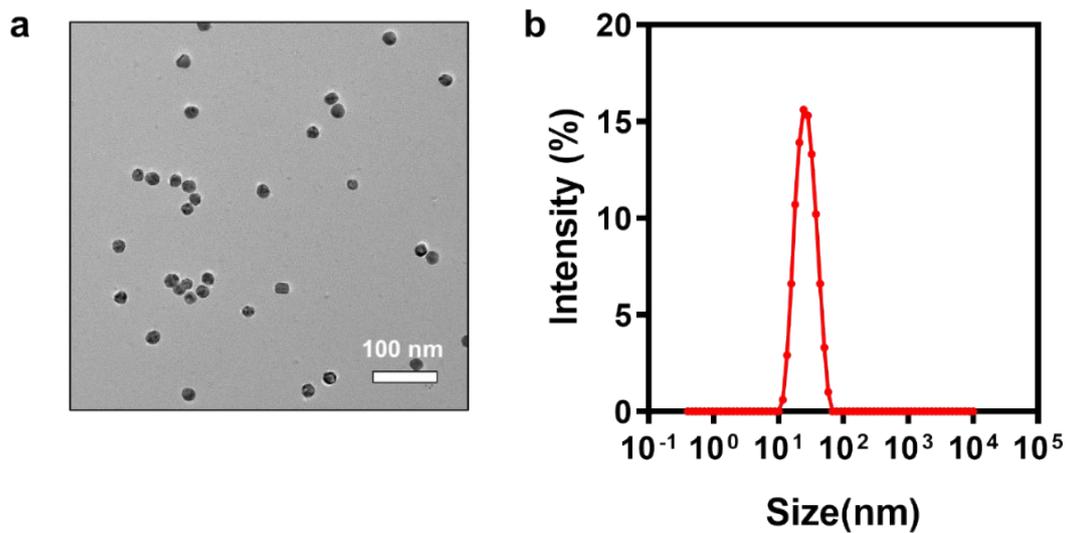


Figure S1 Characterizations of the AuNPs. (a) A typical TEM image of the AuNPs. (b) DLS analysis result of the AuNPs.

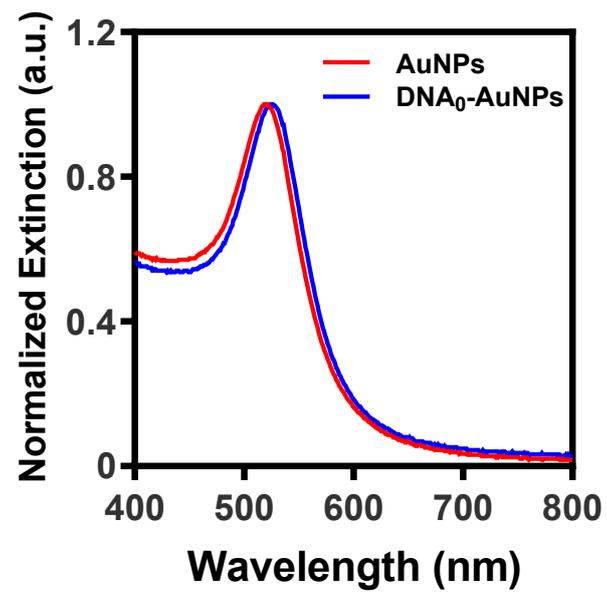


Figure S2 Spectra of the AuNPs before and after the functionalization with DNA₀.

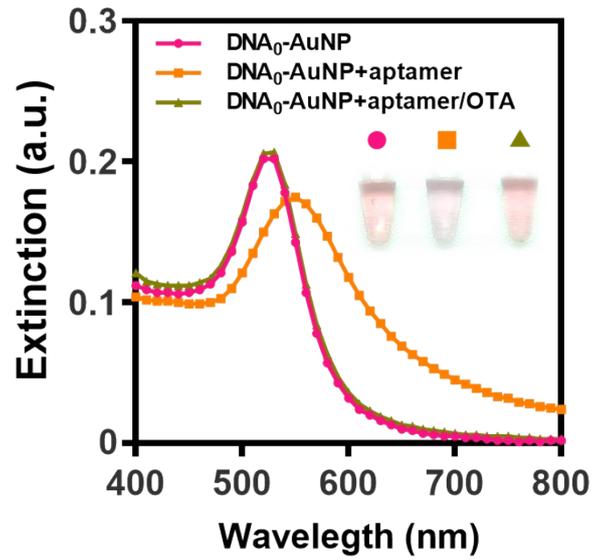


Figure S3 Spectra of DNA₀-AuNPs before and after mixing with aptamer or aptamer/OTA complex. Inserted in the graph is photograph of DNA₀-AuNPs solutions. The final concentration of DNA₀-AuNP was approximately 0.97 nM. The final concentrations of NaCl and aptamer were 800 mM and 62.5 nM, respectively. The concentration of OTA premixed with aptamer was 3.33 μ M. The reaction time was 6 min.

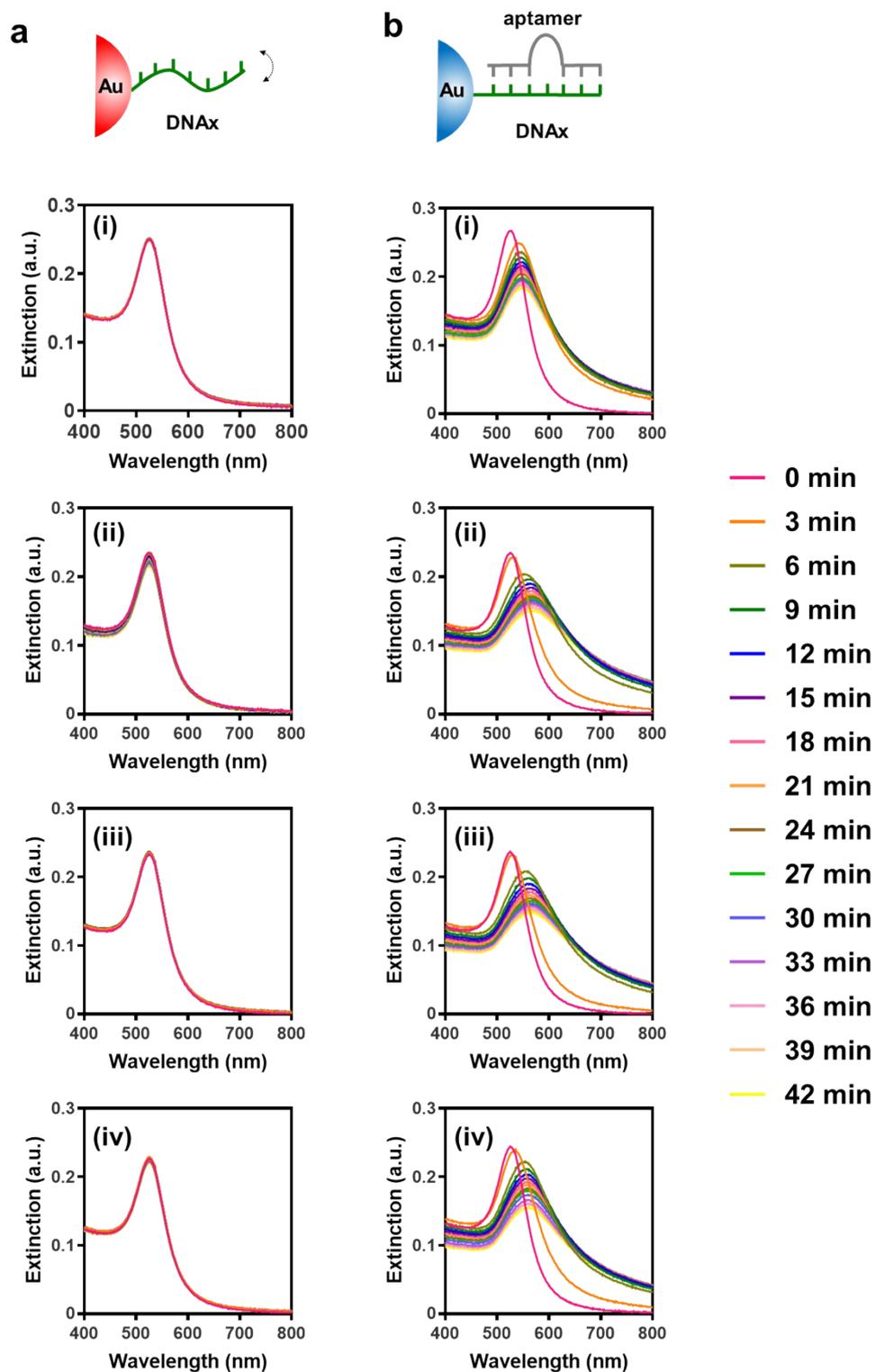


Figure S4 Time course of the assembly of the AuNPs functionalized with different DNA sequences, including DNA₀, DNA₁, DNA₂, and DNA₃. (a) Spectra of DNA_x-AuNPs (0.97 nM) recorded in 42 min. (b) Spectra of DNA_x-AuNPs (0.97 nM) in the presence of aptamer (62.5 nM) recorded in 42 min. From top to bottom: (i) DNA₀-AuNPs, (ii) DNA₁-AuNPs, (iii) DNA₂-AuNPs, and (iv) DNA₃-AuNPs. The final concentrations of PB, MgCl₂, and NaCl in the solution were 10 mM, 1 mM, and 800 mM, respectively.

Table S1 Calculated melting temperature (T_m) of duplexes formed between aptamer and the coded DNA by UNAFold Web Server.^a

Code	T_m (°C)
DNA ₀	89.3
DNA ₁	68.3
DNA ₂	65.0
DNA ₃	60.3

^aThe simulations for the DNAs were performed under the condition of 800 mM of NaCl. The concentration of each DNA strand was 10 μ M.

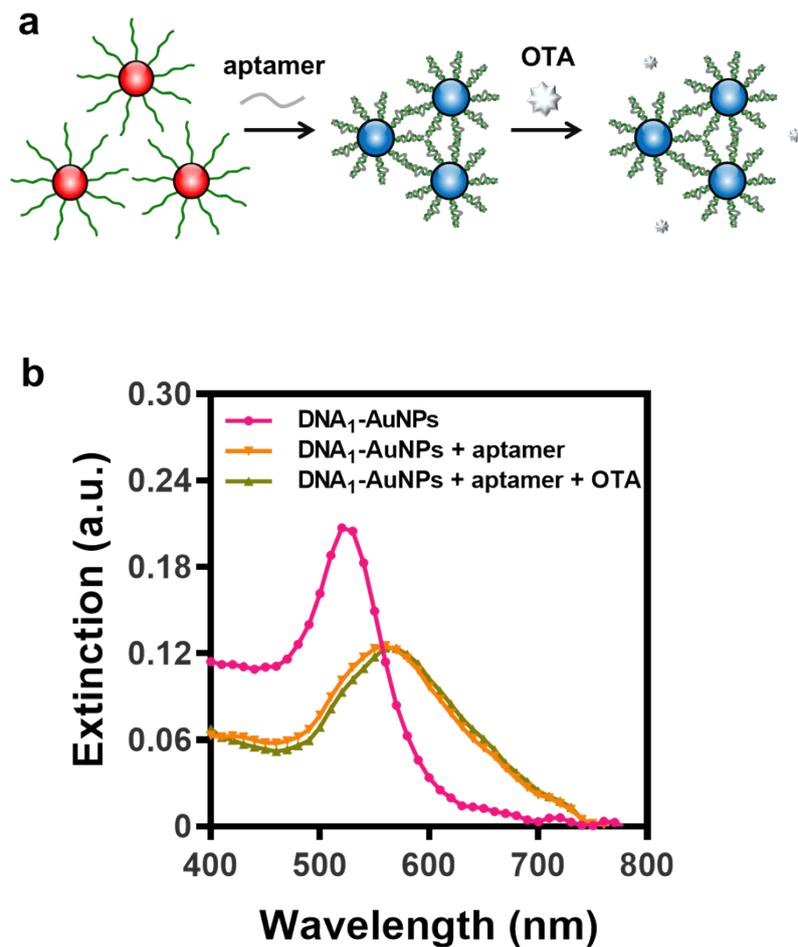


Figure S5 Unsuccessful detection of OTA based on the use of DNA₁-AuNPs caused by competitive binding of hybridized aptamer. (a) Schematic illustration for the OTA detection. (b) Spectra of DNA₁-AuNPs for discrimination of OTA. The final concentration of DNA₁-AuNP was 3 nM. The final concentrations of PB, MgCl₂, NaCl, and aptamer were 10 mM, 1 mM, 800 mM, and 62.5 nM, respectively. The final concentration of OTA was 20 μM.

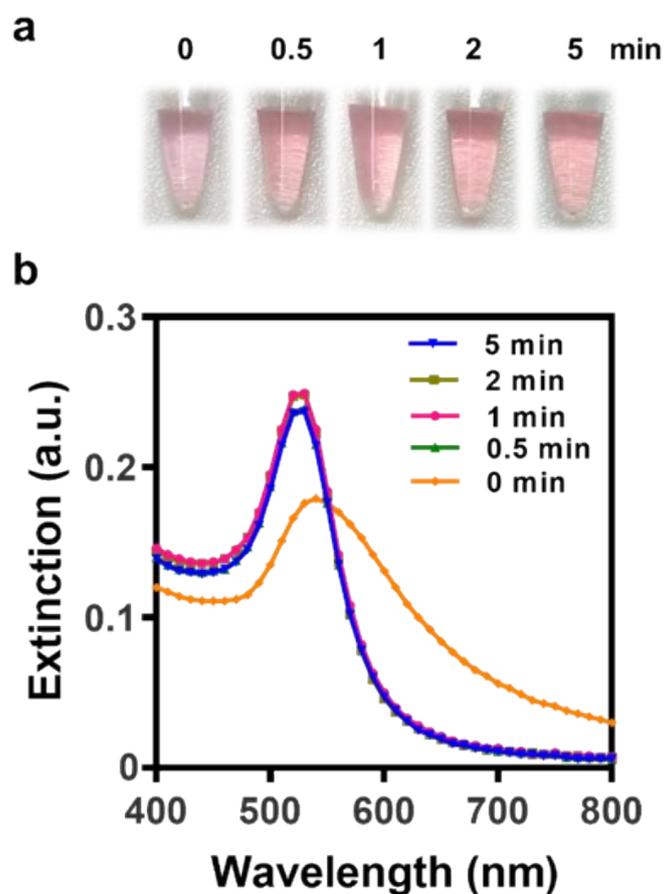


Figure S6 Optimization of the incubation time of OTA and aptamer. (a, b) Photograph (a) and spectra (b) of DNA₁-AuNPs after mixing with OTA and aptamer that were incubated for different times. The final concentration of DNA₁-AuNP was 0.97 nM. The final concentrations of PB, MgCl₂, NaCl, aptamer, and OTA were 10 mM, 1 mM, 800 mM, 62.5 nM, and 3.33 μ M, respectively. The results were recorded at 5 min after the addition of DNA₁-AuNPs.

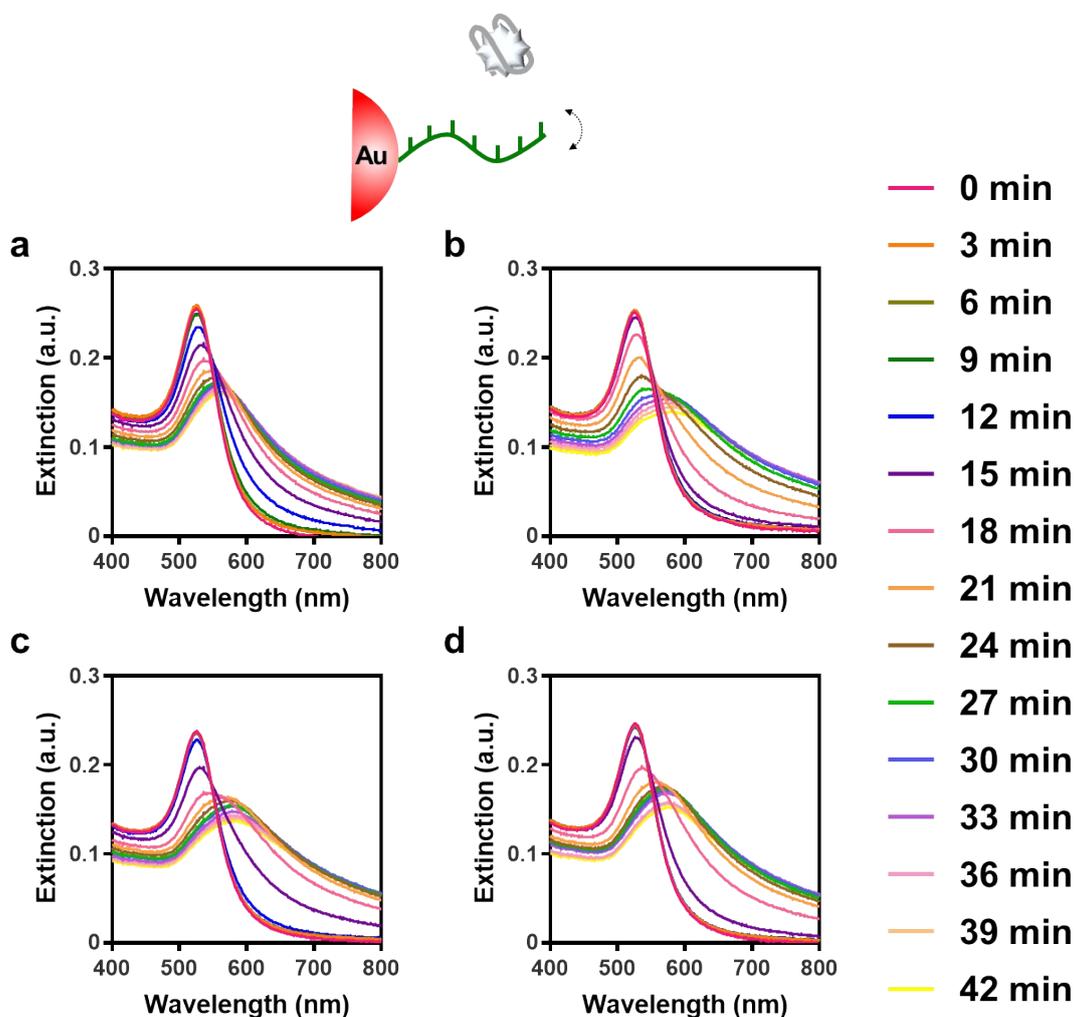


Figure S7 Time courses of the assembly of DNA₀-AuNPs, DNA₁-AuNPs, DNA₂-AuNPs, and DNA₃-AuNPs for detection of OTA. (a-d) Spectra of (a) DNA₀-AuNPs, (b) DNA₁-AuNPs, (c) DNA₂-AuNPs, and (d) DNA₃-AuNPs in the presence of aptamer/OTA complex recorded in 42 min. The final concentration of DNA-AuNPs was approximately 0.97 nM. The final concentrations of PB, MgCl₂, NaCl, aptamer, and OTA were 10 mM, 1 mM, 800 mM, 62.5 nM, and 3.33 μM, respectively. Schematic illustration on the top shows the proposed surface condition of the DNA₁-AuNPs when they are mixed with aptamer/OTA complex.

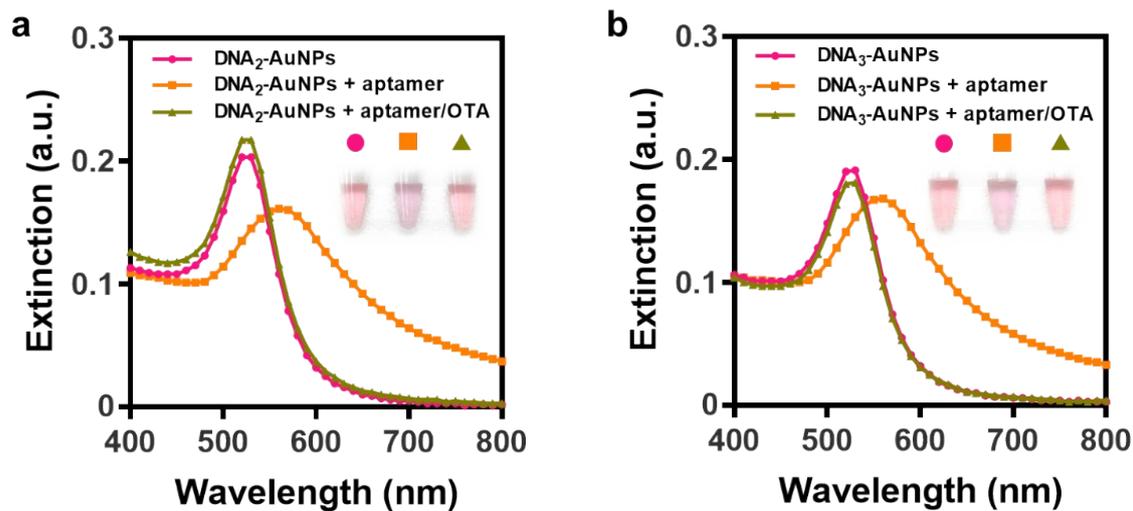


Figure S8 Detection of OTA using DNA₂-AuNPs and DNA₃-AuNPs. (a, b) Spectra of DNA₂-AuNPs (a) and DNA₃-AuNPs (b) before and after the addition of aptamer and aptamer/OTA complex. Insets are the corresponding photographs of the DNA-AuNPs. The final concentration of DNA-AuNPs was 0.97 nM. The final concentrations of PB, MgCl₂, NaCl, and aptamer were 10 mM, 1 mM, 800 mM, and 62.5 nM, respectively. The final concentration of OTA premixed with aptamer was 3.33 μM. The reaction time was 6 min.

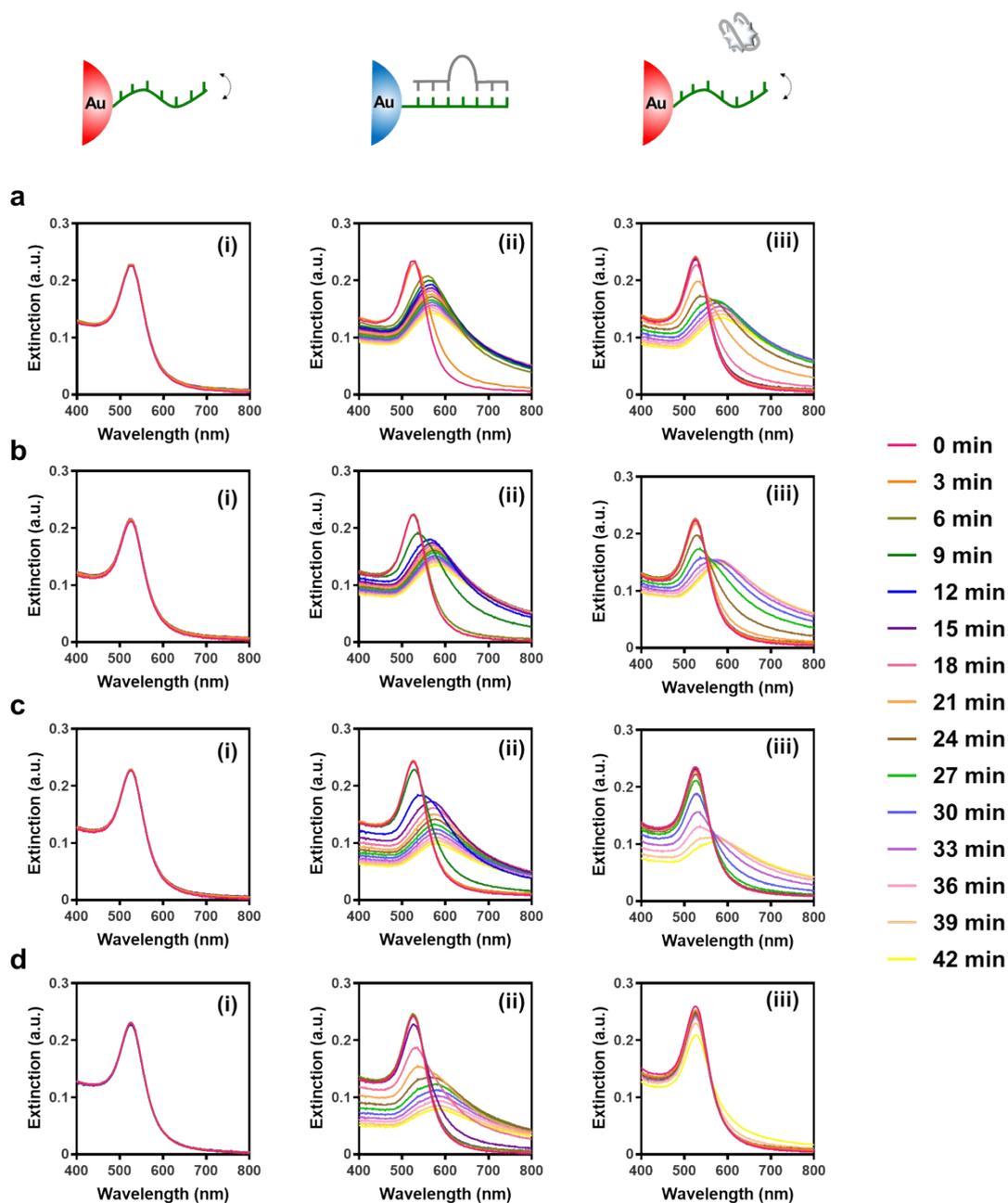


Figure S9 Kinetics of base pair stacking assembly of DNA₁-AuNPs in aqueous and ethanol solutions. (a-d) Spectra of DNA₁-AuNPs (0.97 nM) in aqueous solution (a), 2.5% ethanol (b), 5% ethanol (c), and 10% ethanol (d) without aptamer (i), with aptamer (62.5 nM) (ii), and with OTA (3.33 μM) premixed with aptamer (62.5 nM) (iii). The final concentrations of PB, MgCl₂, and NaCl were 10 mM, 1 mM, and 800 mM, respectively. Schematic illustration on the top shows the proposed surface condition of the DNA₁-AuNPs corresponding to each column of spectra.

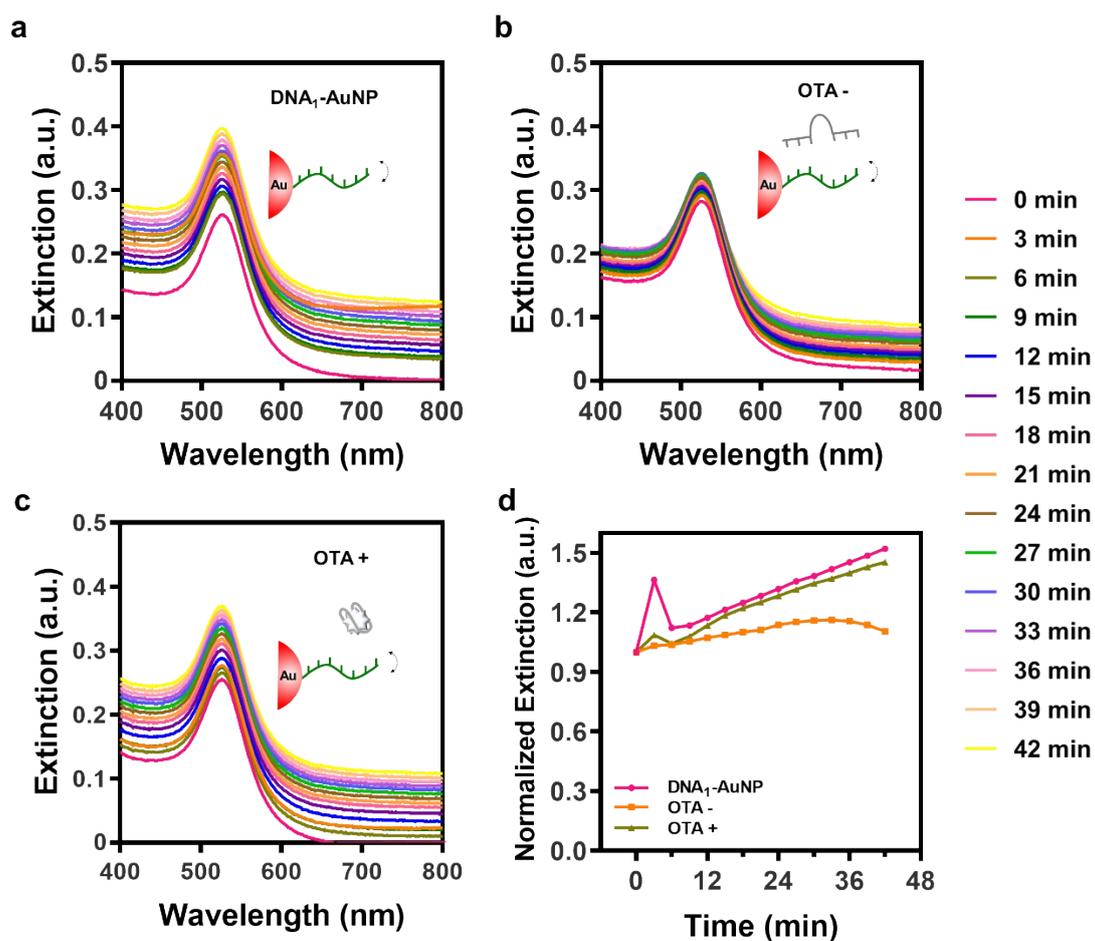


Figure S10 Kinetics of base pair stacking assembly of DNA₁-AuNPs in 20% ethanol solution. (a-c) Spectra of DNA₁-AuNP (0.97 nM) in 20% ethanol without aptamer (a), with aptamer (62.5 nM) (b), and with OTA (3.33 μM) premixed with aptamer (62.5 nM) (c). (d) Plots of the change in extinction of DNA₁-AuNPs at 525 nm recorded as a function of time. The final concentrations of PB, MgCl₂, and NaCl were 10 mM, 1 mM, and 800 mM, respectively.

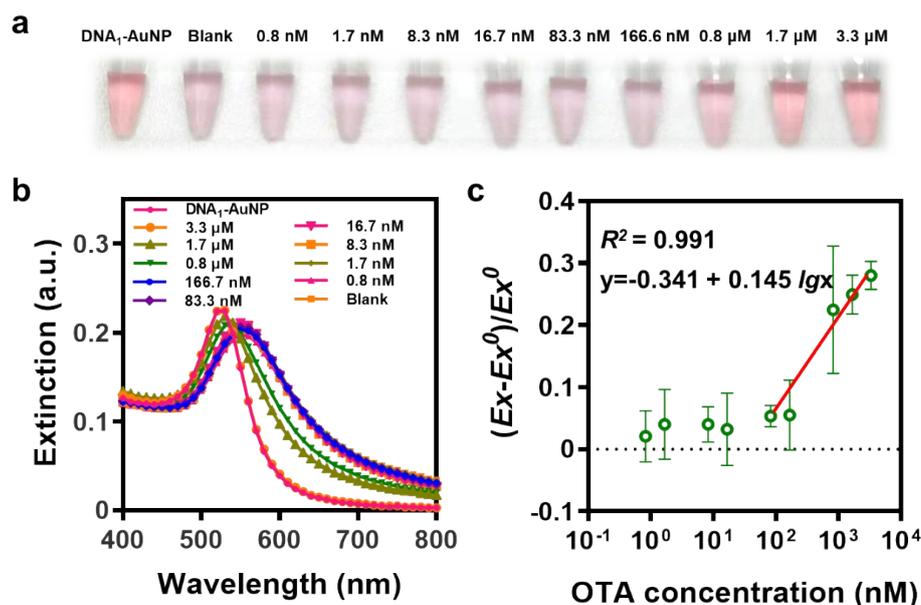


Figure S11 Test of the sensitivity of DNA₁-AuNPs toward OTA in aqueous solution. (a) Photograph of the sensing system in the presence of OTA at different concentrations. (b) Spectra of DNA₁-AuNPs (0.97 nM) with aptamer (62.5 nM) premixed with OTA in different concentrations. (c) Plots of the plasmonic responses $(Ex-Ex^0)/Ex^0$ of the sensing system as a function of OTA concentration. The final concentrations of PB, MgCl₂, and NaCl were 10 mM, 1 mM, and 800 mM, respectively. The reaction time was 6 min.

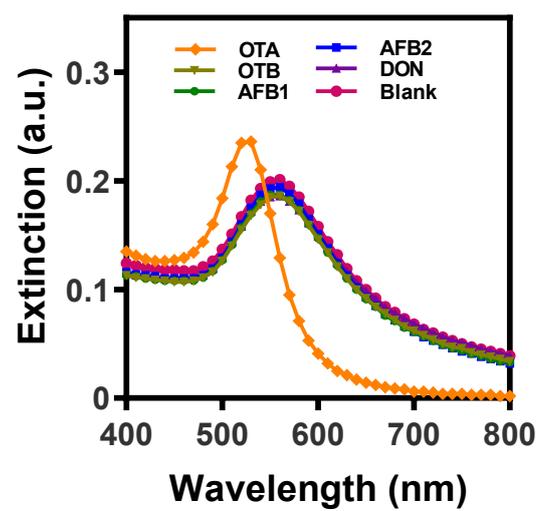


Figure S12 Spectra of DNA₁-AuNPs (0.97 nM) with different mycotoxins (3.33 μ M) premixed with the aptamer (62.5 nM). The final concentrations of PB, MgCl₂, and NaCl were 10 mM, 1 mM, and 800 mM, respectively. The reaction time was 6 min.

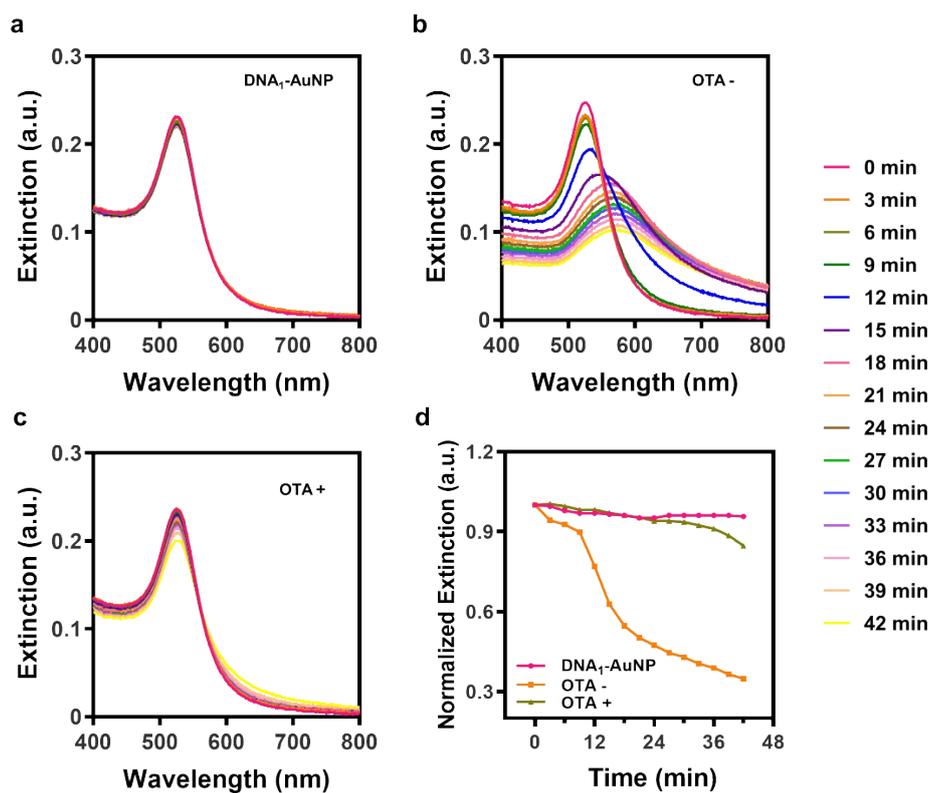


Figure S13 Kinetics of the base pair stacking assembly of DNA₁-AuNP for OTA detection in Baijiu. (a-c) Spectra of DNA₁-AuNP (0.97 nM) without aptamer (a), with aptamer (62.5 nM) (b), and with OTA (3.33 μM) premixed with aptamer (62.5 nM) (c) recorded in 42 min. (d) Plots of the change in the extinction of DNA₁-AuNPs at 525 nm recorded in 42 min. The final concentrations of PB, MgCl₂, and NaCl were 10 mM, 1 mM, and 800 mM, respectively.

Table S2 Comparison of this approach with reported colorimetric OTA assay based on AuNPs and aptamer.

Mechanism	LOD	Time required	Stability	Recoveries (%)	Reference
Electrostatic interaction among AuNPs	49 nM	25 min	poor	100~112.5	1
	5 nM	70 min	moderate	99.4~104.2	2
Induced growth of AuNRs	1 nM	>45 min	moderate	230.7	3
Induced etching of AuNRs	10 nM	82 min	good	101.0~108.0	4
Base pair stacking assembly of DNA-AuNPs	88 nM	<17 min	good	87.0~114.1	The present work

References

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- 2 Y. He, F. Tian, J. Zhou, Q. Zhao, R. Fu and B. Jiao, *J. Hazard. Mater.*, 2020, **388**, 121758.
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