

Colorimetric mercury detection with enhanced sensitivity using Magnetic-Au hybrid nanoparticles

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Synthesis of iron-oxide nanoclusters

Iron-oxide nanoclusters were synthesized using a synthetic hydrothermal method. $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (0.68 g), polyacrylic acid (0.293 g), NaOAc (3.681 g), and ethylene glycol (10 mL) were vigorously stirred for 1 hour in an oil bath at 160°C in a round-necked flask. The resulting mixture was placed in a Teflon vial and autoclaved at 200°C for 10 hours. After the reaction, the solution was centrifuged three times with acetone and separated.

Silica coating on Fe_3O_4

A solution containing the iron-oxide nanoclusters (6.01 mg), ethanol (444.42 mL), distilled water (8.88 mL), and aqueous ammonia (28~30%, 15.12 mL) were placed in a beaker and stirred. While stirring the solution, tetraethylorthosilicate (15 μL) in 5 mL ethanol were added dropwise at a flow rate of 8 mL/h. After the reaction was completed, (3-aminopropyl)trimethoxysilane (25 μL) in 1.67 mL ethanol were added and stirred for 1 hour to achieve surface amination.

Synthesis of the Au seed solution

A solution containing NaOH (0.16 g), tetrakis(hydroxymethyl)phosphonium chloride (96 μL), and 848 mL distilled water were stirred for 15 min. Then, mixture containing 0.192 g $\text{HAuCl}_4 \cdot x\text{H}_2\text{O}$ (0.68 mM) and 19.2 mL of distilled water was rapidly added to the vigorously stirred solution. The resulting mixture was stirred for 30 min. It was then stored at 4°C for three days.

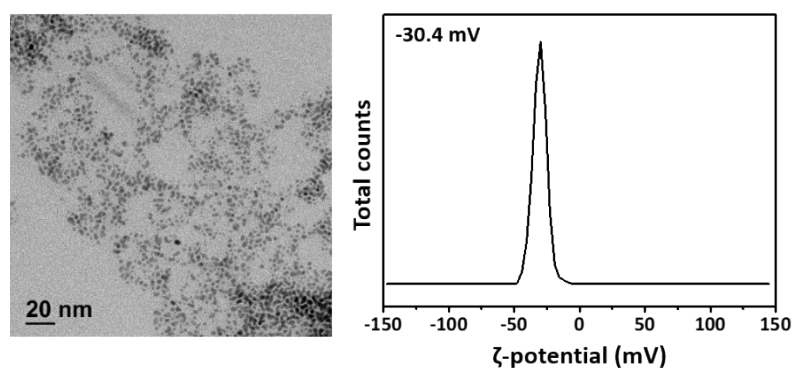


Figure S1. Transmission electron micrographs (TEM) and zeta-potential results of the Au seed.

Preparation of the gold growth solution

HAuCl₄·xH₂O (0.6 g) was added to 20 mL distilled water and stored overnight at 4°C in the dark. To this solution, 1 g of K₂CO₃ and 1.58 L of distilled water were mixed, stored overnight in the dark at room temperature, and then stored in a refrigerator (4°C).

Formation of Mag-Au hybrid nanoparticles

After mixing 1.6 L of the growth solution (0.2 mM) and 20 mL of bis(p-sulfonatophenyl)phenylphosphine dihydrate dipotassium salt (22.6 μM) with 1 mg of Fe₃O₄@SiO₂ attached to the Au seed, 32 mL NH₂OH·HCl (363.2 μM) was added while stirring. Subsequently, the mixture was stirred for three days. Finally, the separation was performed using a centrifuge.

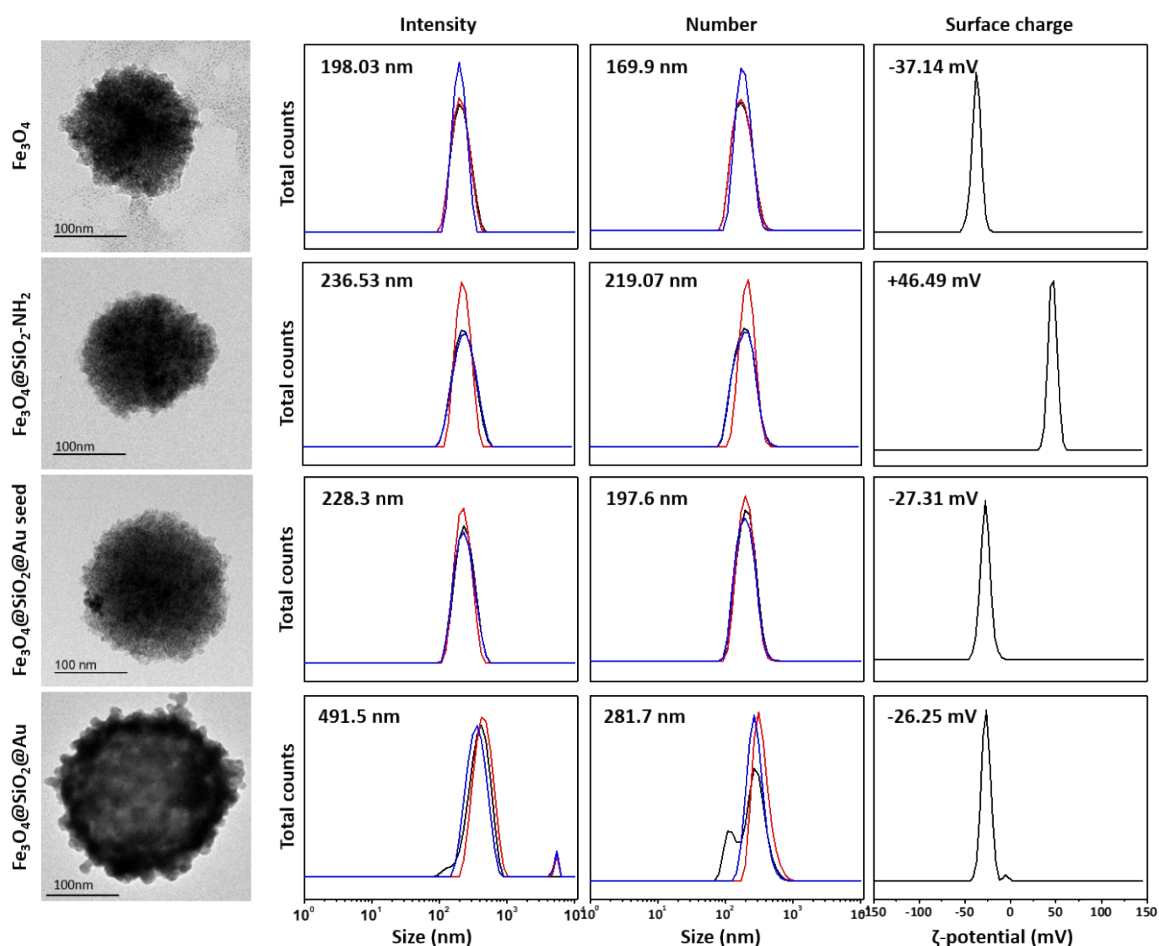


Figure S2. TEM images, hydrodynamic size, and surface charge of Fe₃O₄, Fe₃O₄@SiO₂-NH₂, Fe₃O₄@SiO₂@Au seeds, and Fe₃O₄@SiO₂@Au nanoparticles.

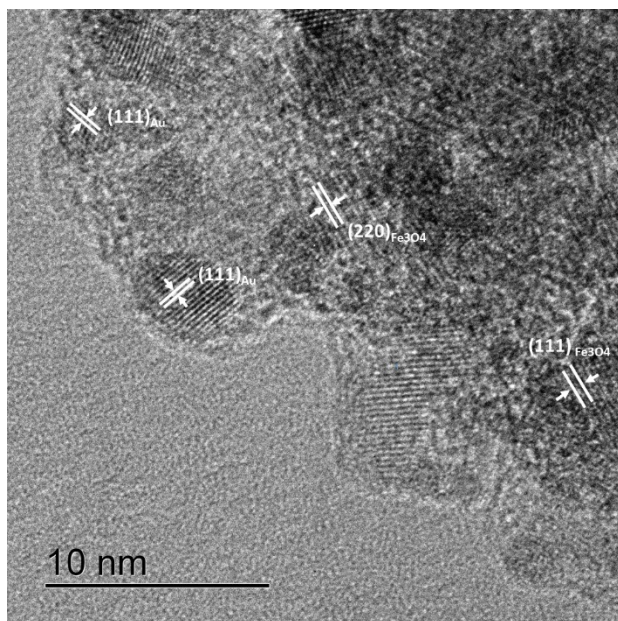


Figure S3. High-resolution TEM image of Mag-Au.

Table S1. Elemental analysis of Mag-Au using energy dispersive spectrometry (EDS)

Element	EDS mapping		
	Line Type	Wt%	Atomic%
Si	K series	2.45	11.64
Fe	K series	13.12	31.28
Au	L series	84.43	57.08

Comparison of Hg^{2+} detection capability between Fe_3O_4 , $\text{Fe}_3\text{O}_4@\text{SiO}_2$, and $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{Au}$ (Mag-Au)

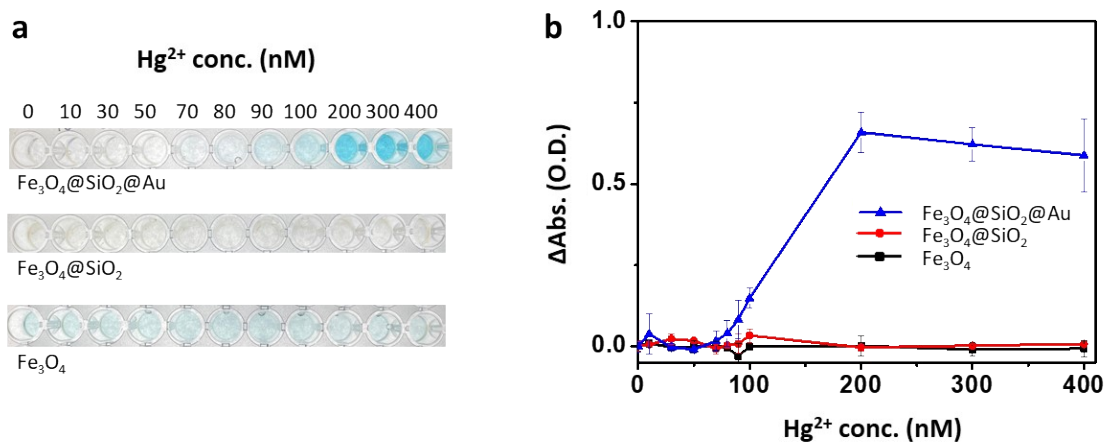


Figure S4. (a) Photograph of indicator solution reacted with varying concentrations of Hg^{2+} ions. The graph of Hg^{2+} ion concentration vs. absorption intensity (@ 652 nm) of the indicator solution.

Elemental analysis of Mag-Au after reaction with Hg^{2+} ions

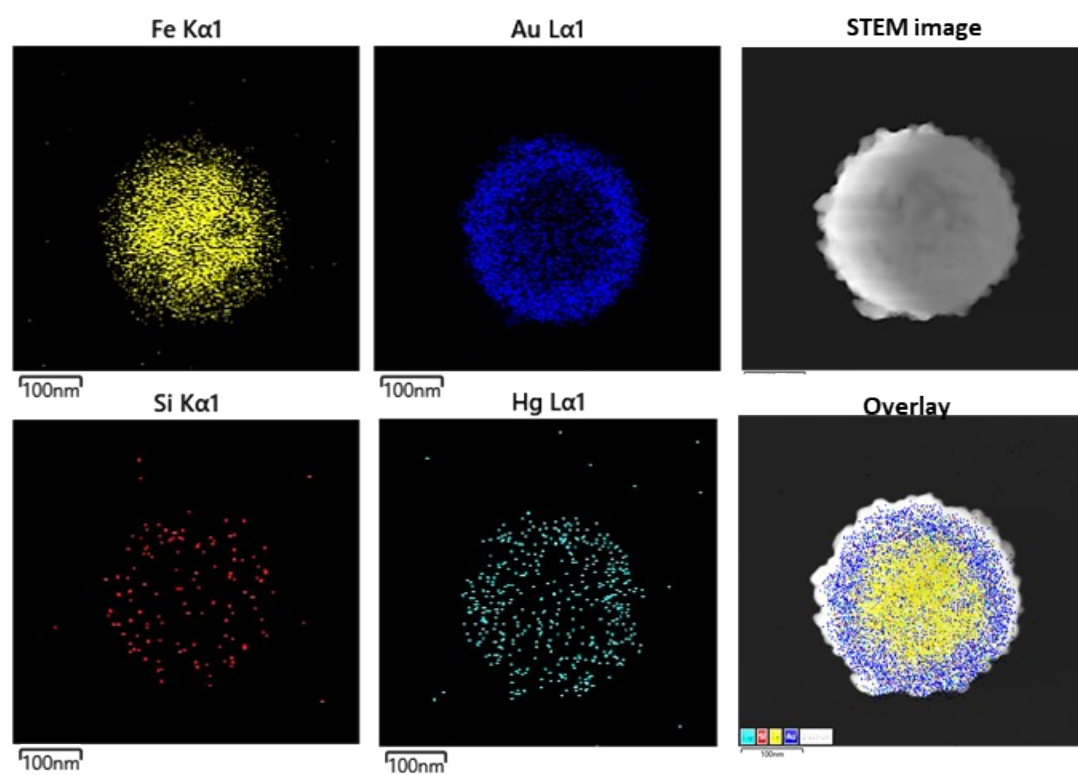


Figure S5. The color-mapped Scanning transmission electron microscopic images of Mag-Au after reaction with Hg^{2+} ions to observe the distribution of elements.

Composition analysis of Mag-Au before and after the reaction with Hg^{2+} using X-ray photoelectron spectra (XPS)

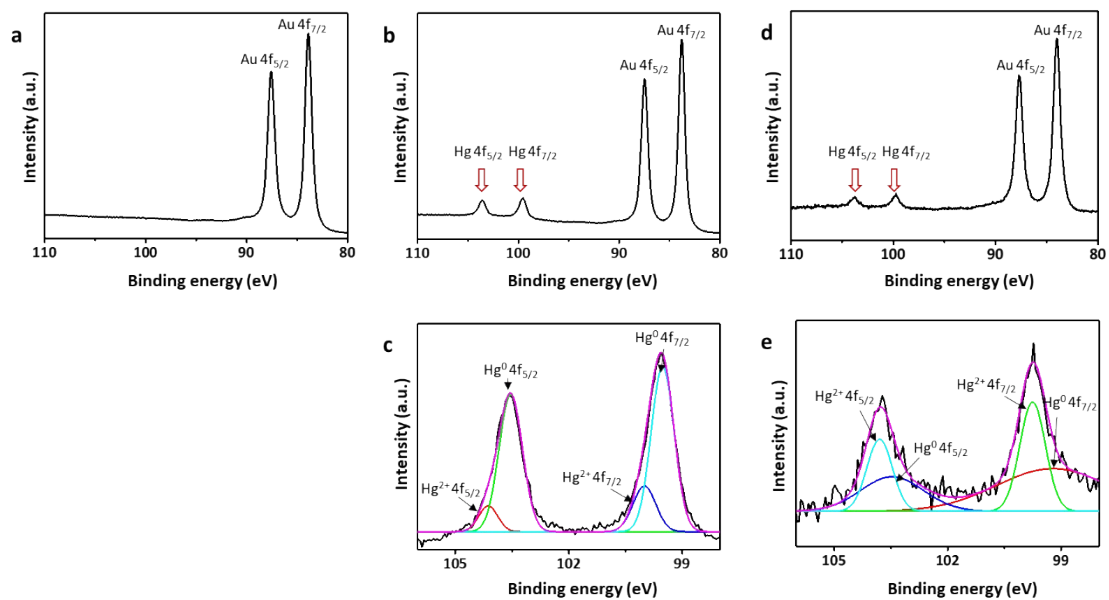


Figure S6. XPS of pristine Mag-Au (a), Mag-Au purified after reaction with Hg^{2+} ions (Mag-Au@Hg, b and c), and Mag-Au@Hg after the reaction with TMB and H_2O_2 (d and e).

Comparison of Hg detection capability of Mag-Au in different buffer condition

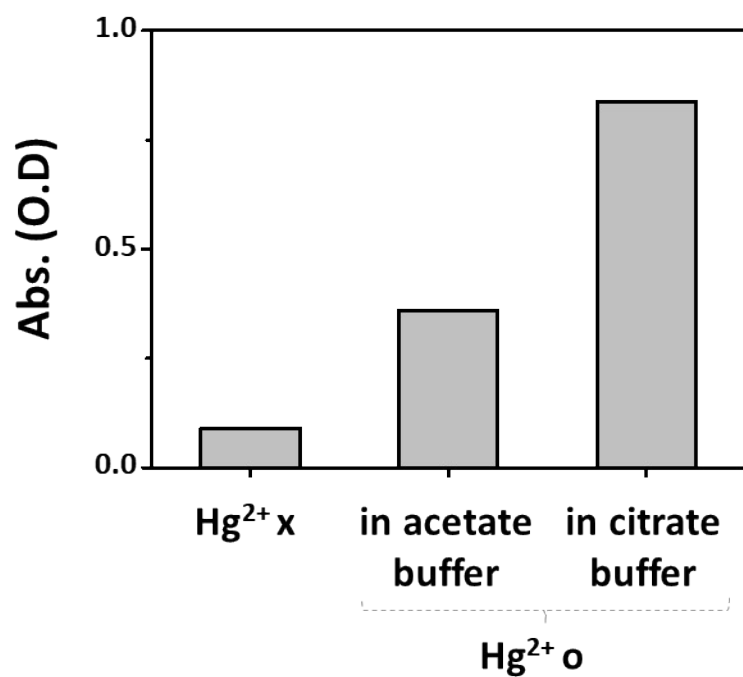


Figure S7. Absorbance comparison of Hg²⁺ detection solution in different buffers at the same pH.

Reactive oxygen species generation test using 1,3-diphenylisobenzofuran (DPBF)

The absorbance of a DPBF solution (2.5 μM , 5 μL dissolved in DMSO) containing Mag-Au NPs (300 $\mu\text{g}/\text{mL}$, 10 μL in 10 mM pH 5.4 citrate buffer) at 416 nm was monitored for 60 minutes. In the presence of reactive oxygen species (ROS), DPBF is converted to 1,3-dibenzoylbenzene (DBB), leading to a decrease in absorbance at 416 nm.

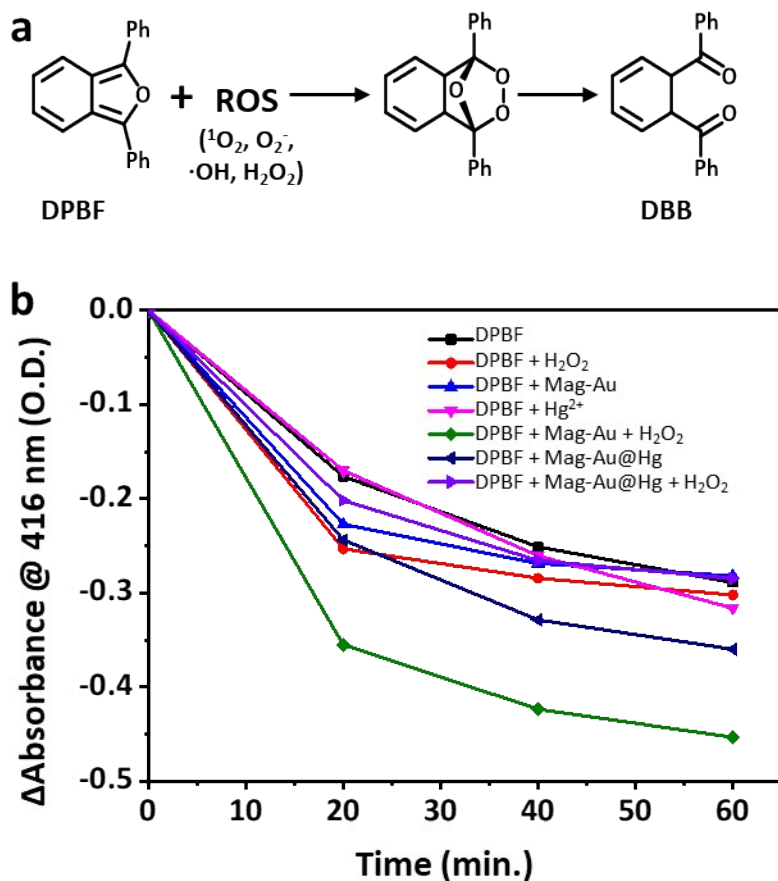


Figure S8. Chemical structure and fluorescent quenching scheme of the decomposition of 1,3-diphenylisobenzofuran (DPBF) to 1,2-dibenzoylbenzen (DBB) by ROS.

Comparison of the catalytic effect of Mag-Au under different reaction conditions

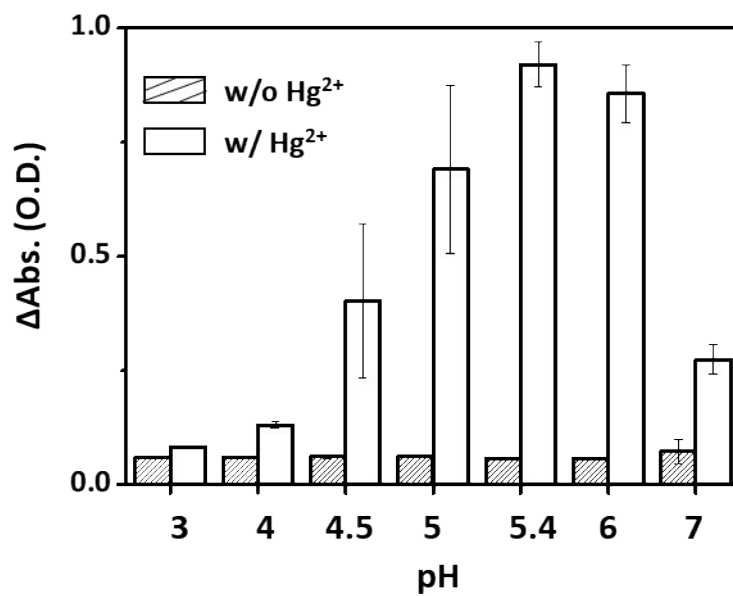


Figure S9. pH-dependent catalytic effect of Mag-Au.

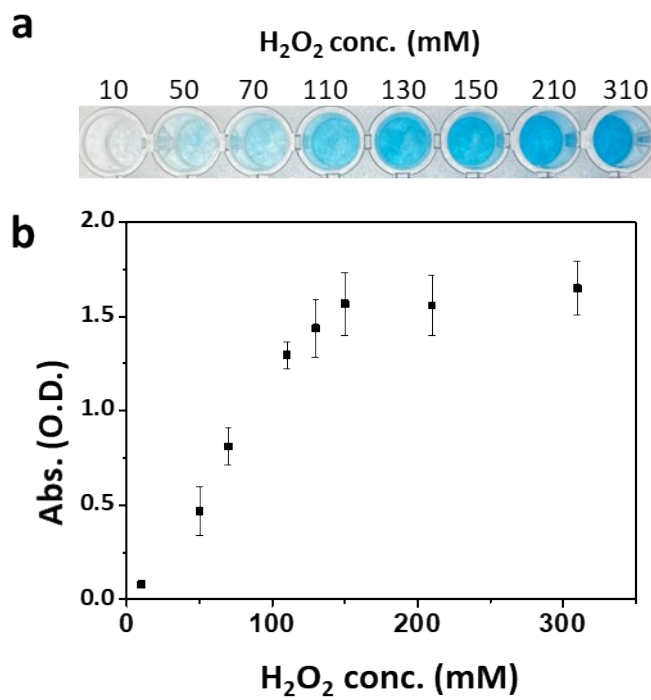


Figure S10. H₂O₂ concentration-dependent (a) color change and (b) absorption intensity.

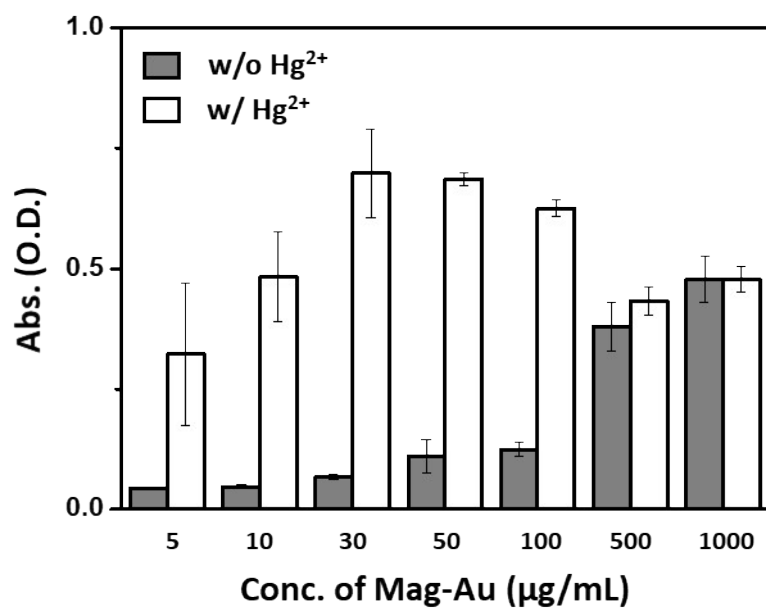


Figure S11. Concentration-dependent catalytic effects of Mag-Au.