Supporting Information

Unique Three-Dimensional Ordered Macroporous Dealloyed Gold-Silver Electrochemical Sensing Platform for Ultrasensitive Mercury (II) Monitoring

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Content

Chemicals

Au substrates were provided by Shanghai Institute of Micro-system and Information Technology (Shanghai, China). $HAuCl_4 \cdot 4H_2O$, $H_2PtCl_4 \cdot 4H_2O$ were purchased from Nanjing Chemical Reagent Co., Ltd (Nanjing, China). $AgNO_3$, diethyl pyrocarbonate (DEPC) and 6-mercapto-1-hexanol (MCH) were purchased from Baoman Bio-tech Co., Ltd. (Shanghai, China). Ethylene glycol (EG), $NaNO_3$, hexadecyltrimethylammonium bromide (CTAB) were obtained from Shanghai Lingfeng Chemical Reagent Co. LTD (Shanghai, China). Polyvinylpyrrolidone (PVP, Mw = 40000), $CuSO_4$, $ZnCl_2$, $CoCl_2$, $MgCl_2$, $CaCl_2$, $CdCl_2$ were purchased from Sinopharm Chemical Reagent Co., Ltd. Thi and the silica spheres with the diameter of 500 nm were obtained from Alfa Asear (Shanghai, China). 40% acrylamide mix solution, ammonium persulfate (APS), ethidium bromide, and 1,2-bis (dimethylamino)-ethane (TEMED) were obtained from Sangon Biotechnology Co. Ltd (Shanghai, China) The supporting electrolyte phosphate buffer saline (PBS, 0.1 M) with various pH values were prepared by mixing the stock solutions of KH_2PO_4 and K_2HPO_4. The supporting electrolyte of cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) was 0.1 M KCl containing 2.5 mM $[Fe(CN)_6]^{4-/3-}$ (1:1). The differential pulse voltammetry (DPV) was recorded in 0.1 M pH7.4 PBS. All chemicals were of analytical grade and used without further purification. All other chemicals, such as anhydrous ethanol (EtOH), acetone, H_2SO_4, HF, and HNO_3 were of analytical grade. All solutions were made up using ultrapure water of resistivity ~18.2 MΩ cm^{-1} (Millipore, SYZ-A).

The DNA sequences were synthesized from Shanghai Generay Engineering Technology and Services Co., Ltd. (Shanghai, China), showing as follows[1].

Capture DNA (cDNA)：5'- SH-$(A)_{20}$- GAT TTG TGA GCT GGT GAG-3'

Probe DNA (pDNA)：3'- CTA TTC TCT CGT CCT CTC GC-$(A)_{20}$-SH-5'

Instruments

CV, EIS and DPV measurements were carried out on a CHI 660D electrochemical workstation (Shanghai CH Instruments Co., China) using a traditional three-electrode system. A platinum foil and a saturated calomel electrode (SCE) were used as counter electrode and reference electrode, and the 3DOMD Ag-Au film modified electrode (Φ = 3 mm) was used as working electrode. The morphology of the 3DOMD Ag-Au film was verified by scanning electron microscopy (FESEM, HITACHI S-4800). The morphology, size, composition and structure of AgNPs, APNC were verified by FESEM, transmission electron microscopy (TEM, JEOL JEM-200CX), the energy-

dispersive spectrum (EDS) analysis and a TU-1901 Double beam UV/visible spectrophotometer (Beijing). Gel electrophoresis was performed using a DYCP-31 BN electrophoresis analyser (Liuyi Instrument Company, China) and imaging was carried out using a Bio-Rad imaging system (USA).

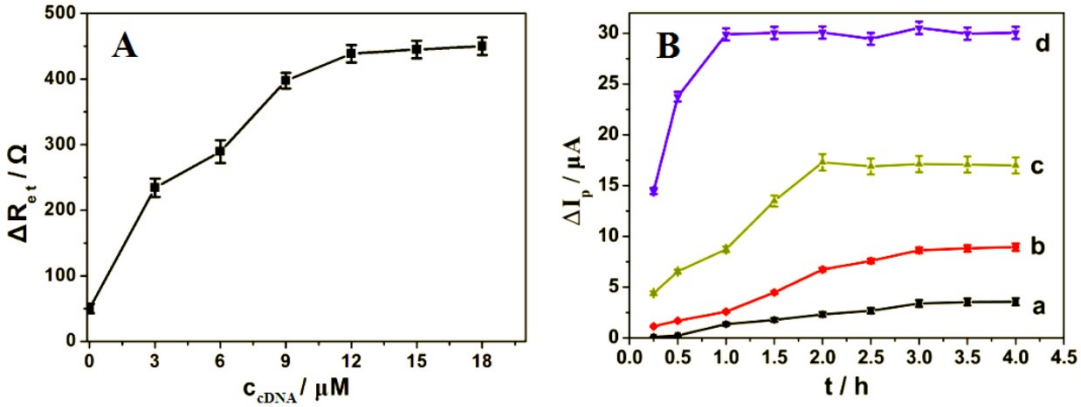

Figure S1. Optimization of the experimental variables. (A) Effect of cDNA concentration on the EIS responses of cDNA/3DOMD Au-Ag thin film electrode. (B) The effect of incubation time between the sensor and the different concentration of Hg^{2+} on the DPV peak current. a to d: 0.05 nM Hg^{2+}; 0.1 nM Hg^{2+}; 10 nM Hg^{2+}; 1000 nM Hg^{2+}. Error bar represents the standard deviation of three repetitive experiments.

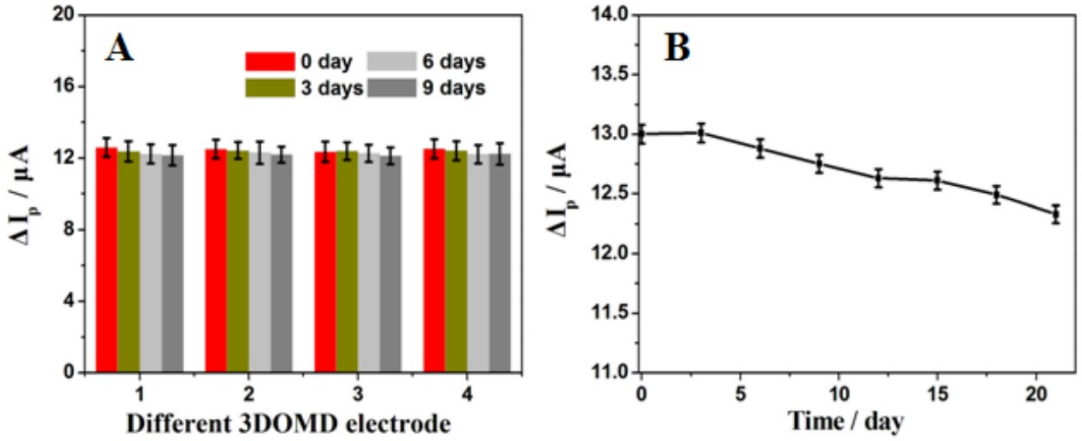

Figure S2. Stability and repeatability validation of Hg^{2+} electrochemical sensors. (A) The reproducibility in detecting Hg^{2+} solution once three days: 0 day, 3 days, 6 days and 9 days. (B) Stability of the Hg^{2+} biosensor in detecting 1 nM Hg^{2+} solution. Error bar represents the standard deviation of three repetitive experiments.

References

1. S. Wang, *Biomolecules*, 2021, **11**, 399.