# Supporting Information

# High-throughput Detection of Multiple Contaminants Based on

# Portable Photo-electrochromic Sensor Chip

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# Content.

# 1. Materials and Apparatus

## 1.1 Experimental materials

Melamine ($C_3H_6N_6$, AR, ≥99.0%), Cyanuric acid ($C_3H_3N_3O_3$, AR, 99.0%) , Isopropanol ($C_3H_8O$, AR), Glutaraldehyde (GA, AR), Bovine Serum Albumin (BSA, AR)，Sodium Dihydrogen Phosphate ($NaH_2PO_4$, AR), Dibasic Sodium Phosphate ($Na_2HPO_4$, AR) and Chitosan (CS, AR, 99.0%) were purchased from Sinopharm Chemical Reagent Co., Ltd. (China), Titanium oxide, anatase ($TiO_2$, 99.8% metals basis, 25nm) purchased from Sinopharm Chemical Reagent Co., Ltd. (China). All of other chemicals are analytical grade, the ultrapure water was used throughout the experiment.

Ochratoxin A aptamer sequence: 5'-GAT CGG GTG TGG GTG GCG TAA AGG GAG CAT CGG ACA-3'

Lincomycin aptamer sequence: 5′-CGC GTG ATG TGG TCG ATG CGA TAC GGT GAG TCG CGC CAC GGC TAC ACA CGT CTC AGC GA-3′

Edifenphos aptamer sequence: 5'-C6-CGT ACG-GAA TTC GCT AGC TAA GGG ATT CCT GTA GAA GGA GCA GTC TGG ATC CGA GCT CCG-3'

All of aptamer was obtained from Sangon Biotech Co., Ltd (Shanghai, China).

## 1.2 Apparatus

The morphology and structure of samples image was obtained by a field emission scanning electron microscope (SEM, JSM-7800F, Japan). A D8 X-ray diffractometer equipped with (Bruker, Germany) high-intensity CuK α (λ = 1.5406) radiation was

used to record the X-ray diffraction (XRD) pattern. The electrochemical and photoelectrochemical (PEC) measurements were performed using a CHI 660E electrochemical workstation (Chen Hua Instruments Co., Shanghai, China) and a LED lamp (PL-LED100F, Beijing Precise Technology Co., Ltd.) in 0.1M phosphate buffer solution (PBS) at 0V. The laser etching is performed by Rui Zhiyi laser marking machine (rzy-bx-20b).

## 2 Experimental section

### 2.1 Synthesis of 3D-g-C$_3$N$_4$

To prepare 3D-g-C$_3$N$_4$ material, 1.3 g of melamine and 1.3 g of cyanuric acid were added into 100 mL of ultrapure water and stirred at room temperature for 12 h. The precursors were then centrifuged at 8000 rpm for 5 min, followed by filtering and drying in an oven at 75°C for 12 h. The dried precursors were finally ground and placed in a crucible and calcined in a muffle furnace at 430°C for 4 h. The final color of 3D-g-C$_3$N$_4$ material was yellow. The dried precursors were finally milled and placed in a crucible and calcined in a muffle furnace at 430°C for 4 h to obtain 3D-g-C$_3$N$_4$ material with a yellow color.

### 2.2 Synthesis of TiO$_2$/3D-g-C$_3$N$_4$

Preparation of TiO$_2$/3D-g-C$_3$N$_4$ composites was according previous works.[1] 1.3 g of melamine, 1.3 g of cyanuric acid and 0.4 g of titanium dioxide were added into 100 mL of ultrapure water and stirred at room temperature for 12 h for the self-assemble of TiO$_2$/g-C$_3$N$_4$ precursor by triple hydrogen bonding between melamine and cyanuric

acid. The precursors were then centrifuged at 8000 rpm for 5 min, followed by filtering and drying in an oven at 75°C for 12 h. Finally, the dried precursors were milled and placed in a crucible and calcined in a muffle furnace at 430°C for 4 h. $TiO_2$/3D-g-$C_3N_4$ composites with a light-yellow color were obtained.

To prepare g-$C_3N_4$ material,[2] 2 g of melamine was placed in a tube furnace and calcined at a rate of about 3°C/min to 550ºC for 4 h to obtain g-$C_3N_4$. g-$C_3N_4$ was ground into ultra-fine powder and this calcination process was repeated until obtaining bright yellow powder. After that, 2 mg of g-$C_3N_4$ was dispersed in 1 mL of isopropanol and sonicated for 2 h to obtain the suspension.

## 2.3 The detection process of actual sample

The preprocessing of the actual sample solution are as follows. 5 g of homogenized fish liver was placed into a 50 mL centrifuge tube. 10 mL methanol solution (80%) was added and fully shook evenly. Then this solution was centrifuged at 1800 rpm for 5 min, and the supernatant was taken into a new centrifuge tube. Next, the obtained supernatant was centrifuged twice for 10 min at 4000 rpm. Then, the supernatant was further filtered by organic microporous filter membrane. Afterwards, three standard solutions of high concentrations are added to the filtered solution and diluted to prepare three actual sample solutions. 20 µL of the obtained actual sample solution was dropped onto the aptamer-modified chip and incubated at 37°C for 45 min. After rinsed with ultrapure water, the chip was placed in PBS solution and illuminated by LED light for 15 minutes.

# 3. Results and discussion
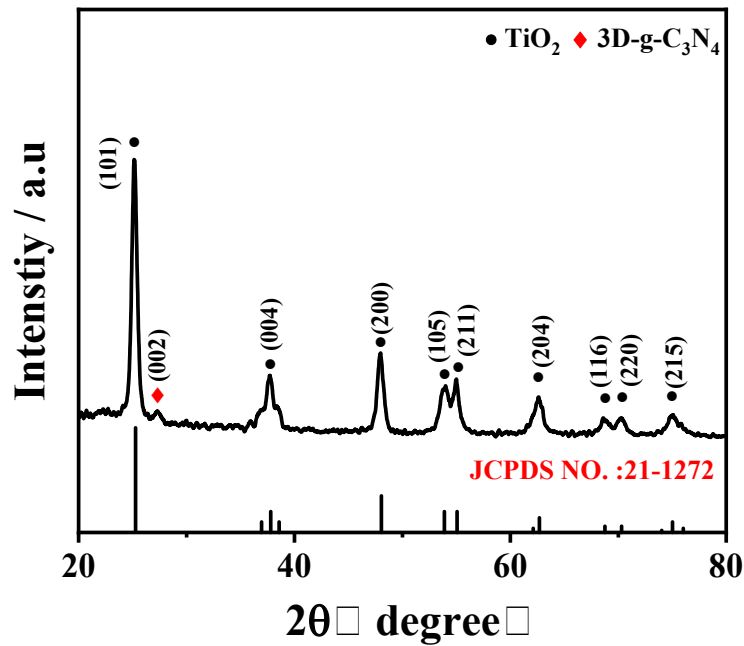
## 3.1 Characterization of materials



**Figure S1.** The XRD of the prepared $TiO_2$/3D-g-$C_3N_4$ composite.

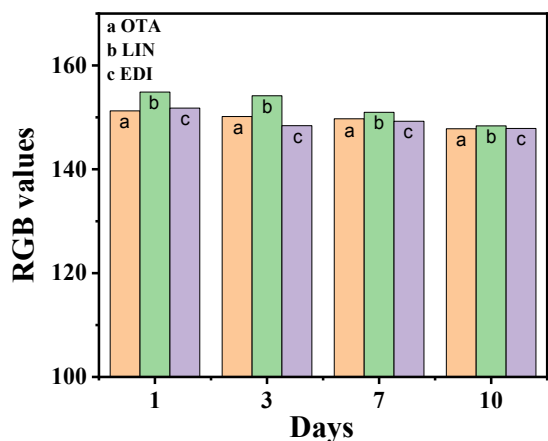## 3.2 Sensor stability and actual sample detection performance



**Figure S2.** The stability measurement of the sensor chips after storage.

**Table S1. Recovery tests of OTA, Lin and EDI in 10-fold diluted freshwater fish liver extract (n=3).**

| Spiked sample | Added (ng/mL) | Found (ng/mL) | Recovery (%) | RSD (%) |
|---|---|---|---|---|
| **OTA 1** | 1 | 1.01 | 101 | 2.83 |
| **OTA 2** | 10 | 10.3 | 103 | 2.21 |
| **OTA 3** | 30 | 31.7 | 105 | 3.07 |
| **Spiked sample** | **Added (mol/mL)** | **Found (mol/mL)** | **Recovery (%)** | **RSD (%)** |
| **LIN 1** | $1\times10^{-8}$ | $9.55\times10^{-8}$ | 95.5 | 2.96 |
| **LIN 2** | $1\times10^{-7}$ | $9.24\times10^{-8}$ | 92.4 | 2.24 |
| **LIN 3** | $2\times10^{-7}$ | $1.85\times10^{-7}$ | 92.5 | 3.61 |
| **Spiked sample** | **Added (ng/mL)** | **Found (ng/mL)** | **Recovery (%)** | **RSD (%)** |
| **EDI 1** | 1 | 1.04 | 104 | 3.96 |
| **EDI 2** | 10 | 10.8 | 108 | 2.17 |
| **EDI 3** | 50 | 48.7 | 97.4 | 2.53 |

### 3.3 Comparison of detection methods

**Table S2. Comparison with other detection methods.**

| Method | Liner range | Detection limit | Refs |
|---|---|---|---|
| PEC aptasensor with triple signal amplification (OTA detection) | 0.0005~0.5 ng /mL | 0.00021 ng /mL | [3] |
| aptamer–photonic crystal encoded suspension array (OTA detection) | 0.01~1 ng / mL | 0.25 pg / mL | [4] |
| chemiluminescence reaction system (LIN detection) | $1.8\times10^{-8}$~$2.2\times10^{-5}$mol/L | $5.6\times10^{-9}$ mol/L | [5] |
| liquid chromatography methods with charged aerosol detection (LIN detection) | $2.7\times10^{-7}$~$1.0\times10^{-3}$ mol/L | $8\times10^{-8}$ mol/L | [6] |
| Colorimetric method (EDI detection) | $1.5\times10^3$~ $7.7\times10^3$ng/L | $1.5\times10^3$ ng/L | [7] |
| Fluorescence spectroscopy (EDI detection) | $0.5\times10^3$~$6\times10^3$ ng/L | $0.13\times10^3$ ng/L | [8] |
| Photochromic visualization sensor (Simultaneous detection of OTA, LIN and EDI) | 1~100 ng/mL $10^{-11}$~$10^{-4}$mol/L 1~100 ng/L | 0.33 ng/mL $3.3\times10^{-12}$mol/L 0.33ng/L | This work |

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