

Electronic supplementary information

Reusable interface-decoupled organic photoelectrochemical transistor biosensing for acetylcholinesterase activity and its inhibitor detection

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Experimental Section

Reagents and Materials. All solvents and reagents were commercially available and used without further purification, unless for special needs. Acetylcholinesterase (AChE), choline oxidase (ChOx), soluble starch, amyloglucosidase (AG), and zinc chloride (ZnCl_2) were purchased from Macklin Biochemical Company (Shanghai, China). Acetylcholine chloride (ACh), potassium iodide (KI), chlorpyrifos (CPF), glyphosate (GLY), urea, oxytetracycline base (OTC-base), 2,4-dichlorophenoxyacetic acid (2,4-D), ascorbic acid (AA), and thioacetamide were obtained from Aladdin Reagent Company (Shanghai, China). Horseradish peroxidase (HRP), alkaline phosphatase (ALP), glucose oxidase (GOx), and biological PBS buffer (10 mM, pH 7.2-7.6) were sourced from Sangon Biotech Company (Shanghai, China). Tetracycline hydrochloride (TC-HCl) and lysozyme (LYZ) were acquired from Energy Chemical Company (Shanghai, China). Leucine aminopeptidase (LAP) was procured from Yuanye Bio-Technology Company (Shanghai, China). Indium (III) chloride (InCl_3) was purchased from Adamas Reagent Company (Shanghai, China).

Instruments. Photoelectrochemical (PEC) and organic photoelectrochemical transistor (OPECT) detection were performed using a PEC/OECT detector (Nandaguang, Nanjing, China). Scanning electron microscopy (SEM) images were acquired using an S-4800 microscope (Hitachi, Japan). Transmission electron microscopy (TEM) images were obtained with a Thermo Scientific Talos F200i instrument (Thermo Fisher Scientific, Australia). X-ray diffraction (XRD) patterns were recorded on an Ultima IV diffractometer (Rigaku, Japan) with copper $K\alpha$ radiation over the 2θ range of 25° to 70° . A Thermo Scientific K-Alpha X-ray photoelectron spectrometer (Thermo Electron, USA) was used to collect X-ray photoelectron spectroscopy (XPS) spectra. UV-vis absorbance spectroscopy and UV-Vis diffuse reflectance spectroscopy (UV-vis DRS) were recorded on a Cary 60 UV-vis spectrophotometer (Agilent Technologies, USA) and a UV-3600i Plus UV-Vis-NIR spectrophotometer (Shimadzu, Japan), respectively. Luminous power was measured using a PLS-MW2000 Luminous power meter (Perfectlight Technology, Beijing, China).

Synthesis of ZnIn_2S_4 . The synthesis of ZnIn_2S_4 was performed following a modified procedure.¹ Briefly, ZnCl_2 (0.0682 g, 0.5 mmol), InCl_3 (0.2212 g, 1 mmol), and $\text{C}_2\text{H}_5\text{NS}$ (0.1503 g, 2 mmol) were dissolved in 45 mL of ultrapure water and stirred at 25°C for 1 h. The solution was transferred to a Teflon autoclave, and several FTO substrates (25×8 mm) were placed vertically within the autoclave. Then the autoclave was sealed and maintained at 160°C for 4 h. After cooling, the samples were rinsed thoroughly with ultrapure water and ethanol, dried at room temperature, and finally annealed at 200°C for 2 h.

Device Testing.

PEC and OPECT measurements were performed in PBS containing 10 mM AA. A gate with an effective area of 19.63 mm^2 and a white light source (5 W) with 420 nm filter were employed for signal excitation. PEC characterization utilized a standard three-electrode system comprising an Ag/AgCl reference electrode, Pt wire counter electrode, and sample/FTO working electrode, with responses recorded at 0 V relative Ag/AgCl. Transistor characterization involved acquiring I_D - V_D output curves through V_G sweeps from 0 to 0.6 V at fixed $V_D = 0.1$ V, and determining the I_D -time and I_D - V_G curves.

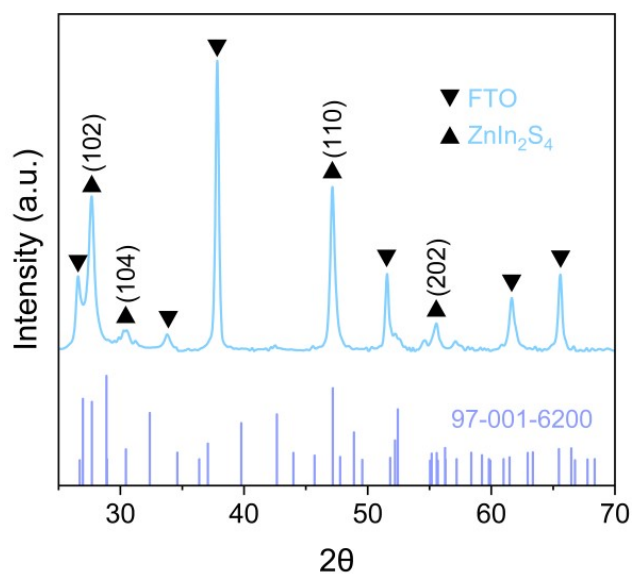


Fig. S1 XRD patterns of ZnIn_2S_4 .

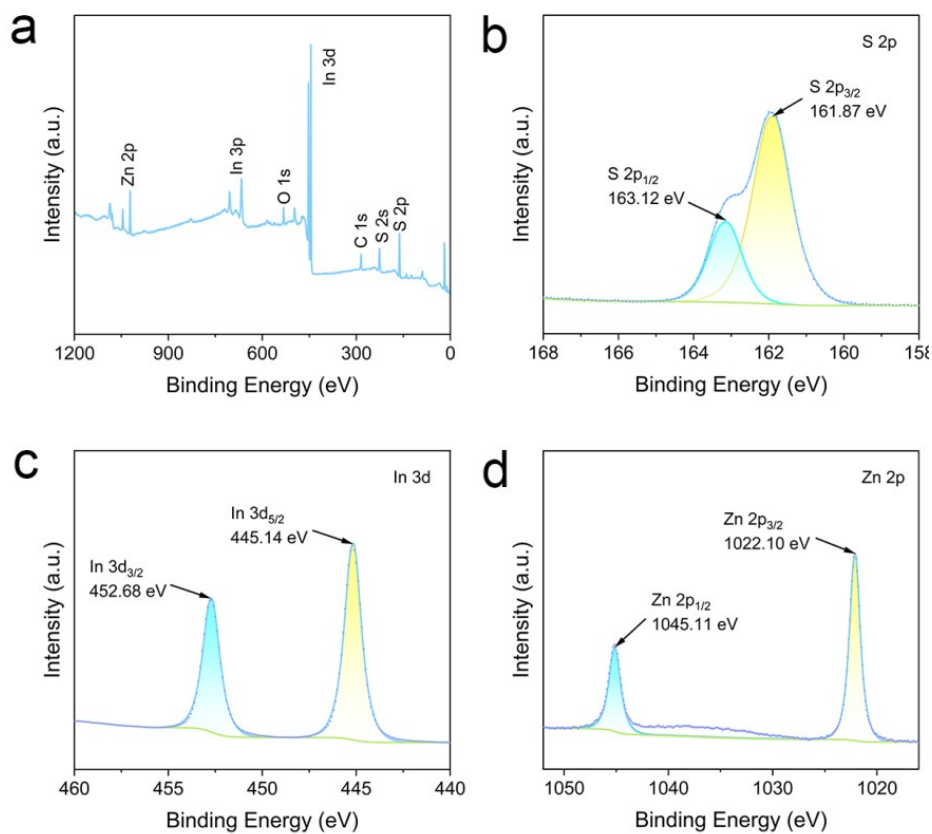


Fig. S2 XPS spectra of (a) survey-scan, (b) S 2p, (c) In 3d, and (d) Zn 2p for ZnIn_2S_4 .

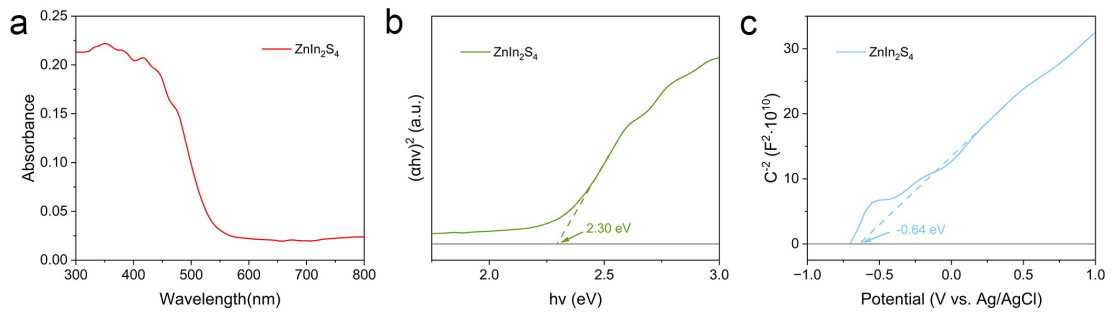


Fig. S3 (a) UV-vis DRS of ZnIn₂S₄. (b) Plots of $(\alpha hv)^2$ against photon energy ($h\nu$), and (c) Mott-Schottky plots of ZnIn₂S₄.

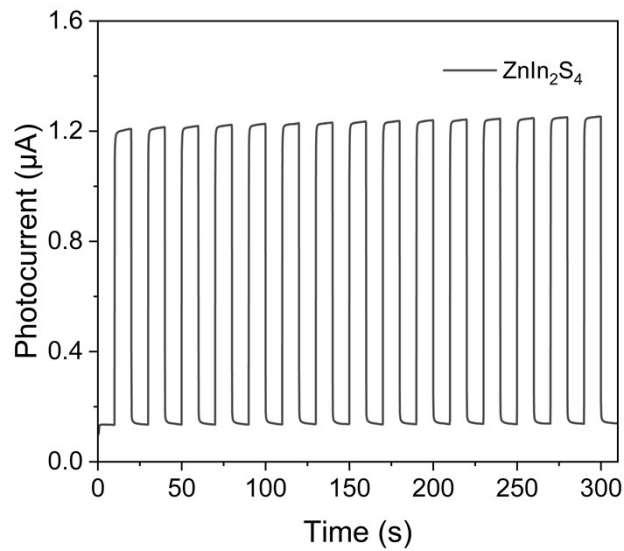


Fig. S4 Stability tests of ZnIn₂S₄ electrode.

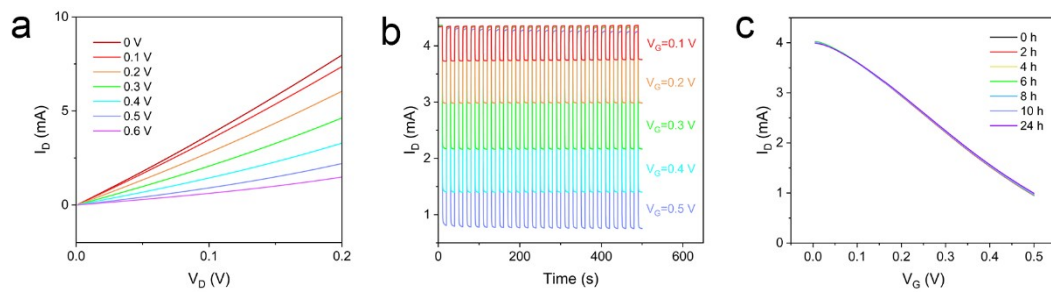


Fig. S5 (a) Output curves measured at different V_G . (b) Transient I_D responses at different V_G over 500 s. (c) Transfer curves measured at different time intervals.

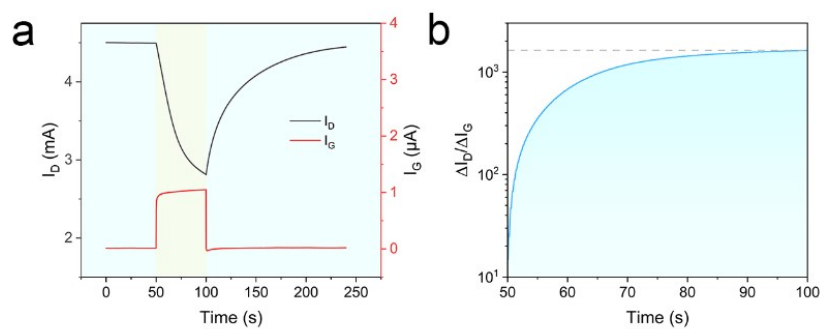


Fig. S6 (a) The transient I_D and I_G responses of $ZnIn_2S_4$ -gated OPECT with the light on/off, and (b) corresponding current gain. ($\Delta I_D/\Delta I_G$, where ΔI_D and ΔI_G are defined as the changed I_D and I_G signals upon light on/off).

Table S1 Evaluation of various biosensors for AChE analysis.

Method	Materials	Linear range ($U \cdot L^{-1}$)	LOD ($U \cdot L^{-1}$)	Reference
Electrochemical	PDA@dsPD	0-300	69	2
Colorimetric	μ PAD	100-1000	9	3
Fluorometric	DCM-N	200-1600	60	4
Fluorometric	CyN	0-8000	117	5
Fluorometric	Naph-3	0-1000	100	6
OPECT	I_2 -starch complex/ $ZnIn_2S_4$	20-200	15	This work

Table S2 Evaluation of various biosensors for CPF analysis.

Method	Materials	Linear range (ppb)	LOD (ppb)	Reference
Electrochemical	CuO/SA-g-PANI	350-42000	131	7
Electrochemical	CPSaptasensor	0-100	10	8
Fluorometric	dye@ALB	0-70000	200	9
Colorimetric	Ag-NanoZyme	35000-210000	11300	10
Colorimetric	γ -Fe ₂ O ₃ /t-SG	500-10 ⁶	170	11
OPECT	I ₂ -starch complex/ZnIn ₂ S ₄	30-180	16	This work

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