Supporting Information

A 3D origami electrochemical immunodevice based on Au@Pd alloy nanoparticles-paper electrode for the detection of carcinoembryonic antigen

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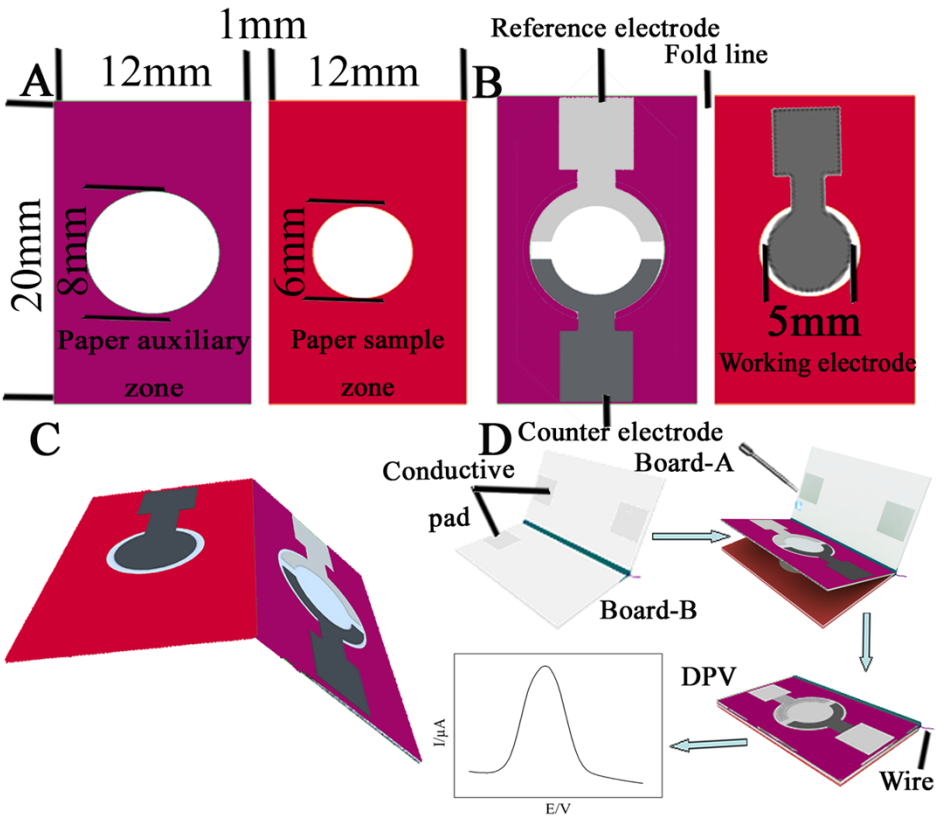
Fabrication and characterization of this 3D paper-based EC device

The preparation of this *3D paper-based EC device* was similarly to our previous work with modifications and detailed procedure was described below. Wax was used as the paper hydrophobization and insulation agent in this work to construct hydrophobic barrier on paper. The shape of hydrophobic barrier on origami device was designed using Adobe illustrator CS4 and the entire origami device could be produced in bulk on an A4 paper sheet by a commercially available solid-wax printer (FUJIXEROX Phaser 8560DN, Japan). After the curing process, the wax-penetrated paper sheet was ready for screen-printing of electrode. As shown in Scheme S1A, the entire paper-based device is 25.0 mm × 20.0 mm. The unprinted hydrophilic area, which constituted the reservoir of the paper electrochemical cell, contains two circular paper working zones (named circle-A: 6.0 mm in diameter on sample tab and circle-B: 8.0 mm in diameter on auxiliary pad) for screen-printing electrodes. The electrode array consisted of a screen-printed Ag/AgCl reference electrode and carbon counter electrode on the paper auxiliary zone (6 mm in diameter) and a screen-printed carbon working electrode on the paper working zone respectively. After folding, the three screen-printed electrodes (working electrode, reference electrode, and counter electrode) would be connected once the paper electrochemical cell was filled with solution.



Scheme S1 The schematic representation, size, shape, and detection of this 3D origami EC device. (A) One side of the paper-based EC device without the screen-printed electrodes; (B) The reverse side of (A) with the screen-printed electrodes; (C) The prepared paper-based EC device was folded as picture C; (D) After modification, the paper-based EC device was integrated with a transparent device-holder, and glucose was added to trigger the EC reaction.

Characterization of Au NPs seeded cellulose fibers

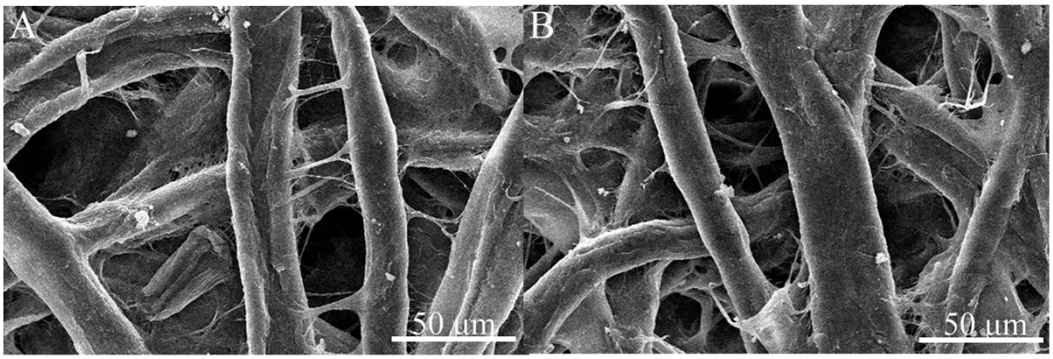


Fig. S1. SEM images of (A) bare PWE; (B) Au NPs seeded paper.

Optimization of detection conditions

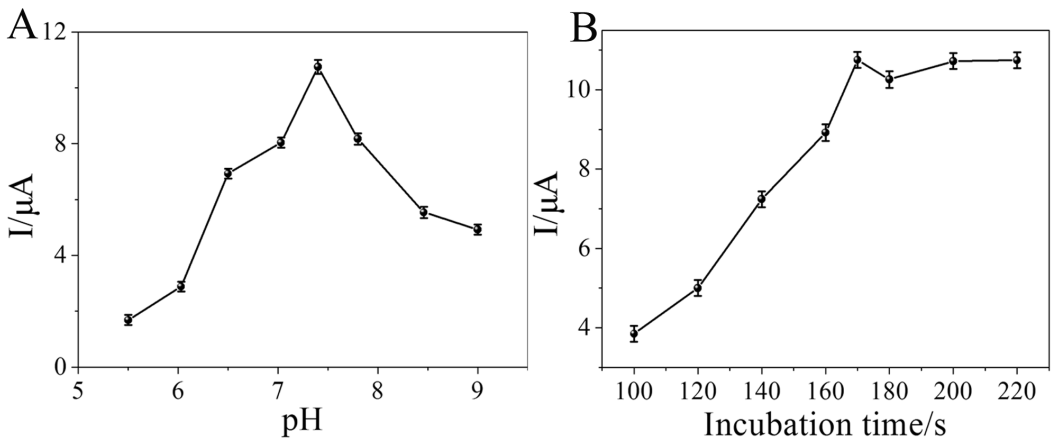


Fig. S2. Effect of (A) pH of detection solution, (B) incubation time on DPV peak current intensities in the presence of 1 ng·mL^{-1} CEA, where n=6 for each point.

Stability, selectivity and reproducibility of the immunodevice

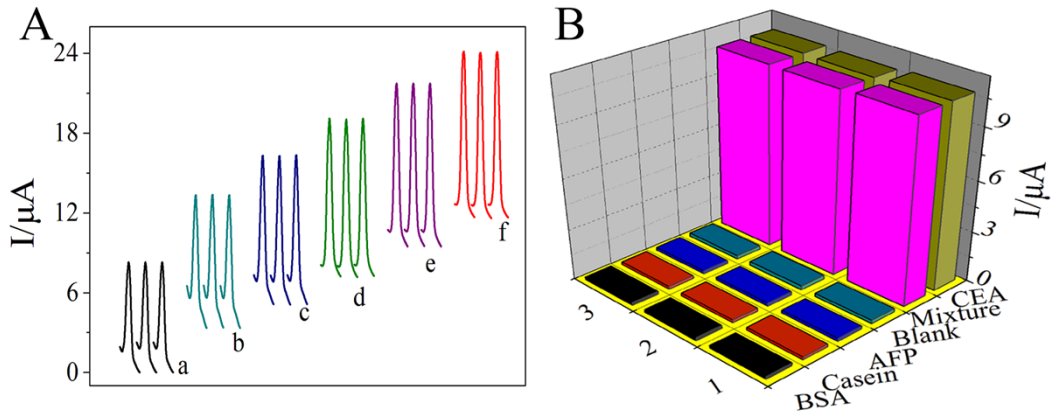


Fig. S3. (A) DPV peak current stability of proposed immunosensor to various concentrations (ng·mL^{-1}) of CEA (0.001, 0.01, 0.1, 1, 10, 100, from left to right). (B) The selectivity of the proposed EC immunosensor: BSA (100 ng·mL^{-1}), Casein (100 ng·mL^{-1}), AFP (100 ng·mL^{-1}), blank, CEA (1 ng·mL^{-1}), a mixture containing CEA (1 ng·mL^{-1}), BSA (100 ng·mL^{-1}), Casein (100 ng·mL^{-1}) and AFP (100 ng·mL^{-1}).

Table S1 The results of comparing with other immunoassay biosensing systems toward CEA.

Detection methods	Linear range ($ng \cdot mL^{-1}$)	Detection limit ($ng \cdot mL^{-1}$)	References
Flow injection chemiluminescence	5.0-100	2.7	1
Cyclic voltammetry	1.5-200	0.05	2
Electrochemical	0.01-100	0.01	3
Electrochemical	0.001-100	0.0004	Proposed method

References

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