

**A Sensitive Electrochemical Platform Integrated with 3D graphene
aerogel for Point-of-Care Testing for Tumor Markers**

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Biochip Fabrication & Preparation of GO

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Figure S1. Fabrication of the 3D GA microfluidic biochip.

The biochip consists of three layers including the ITO coated glass substrate, the microchannel layer and the upper PDMS cap. Porous 3D GA film is functionalized with PCA and modified with antibodies before the channel being sealed. The GA film is obtained via in-situ chemical reduction of graphene oxide with LA in the channel. a) Preparation procedures; b) Antibody Modification; c) Photograph of the microfluidic chip.

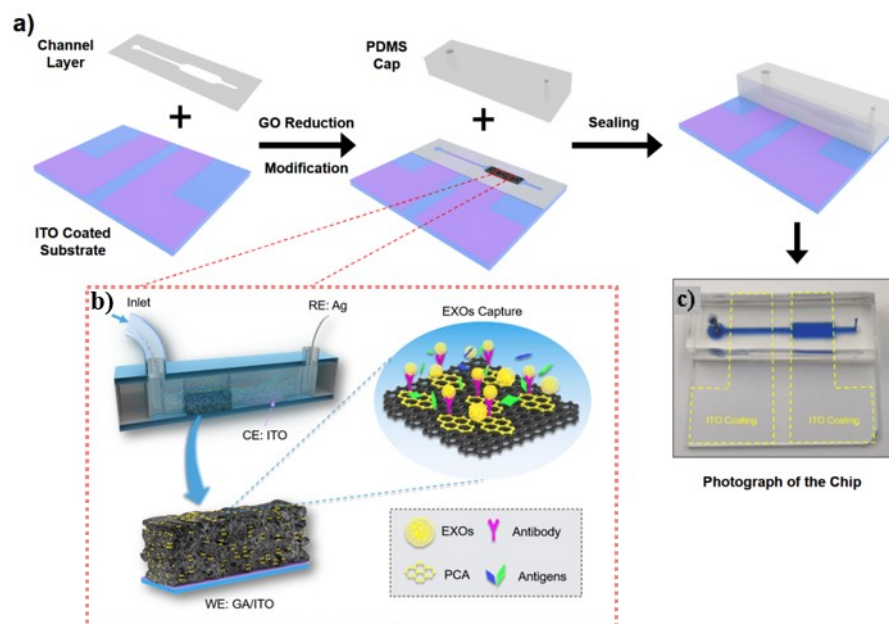


Figure S2. EIS detection platform.

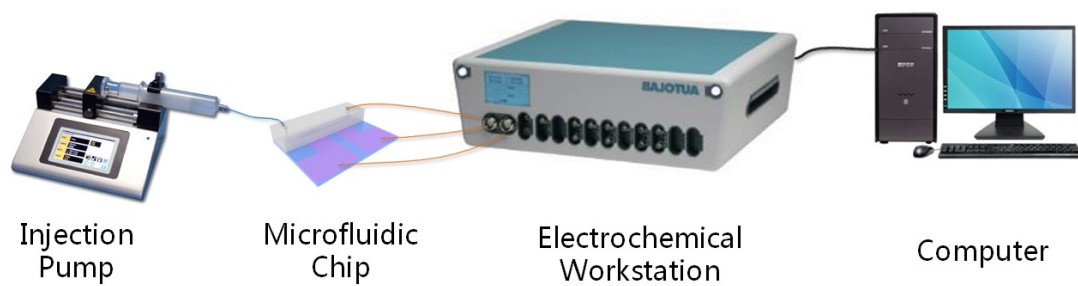
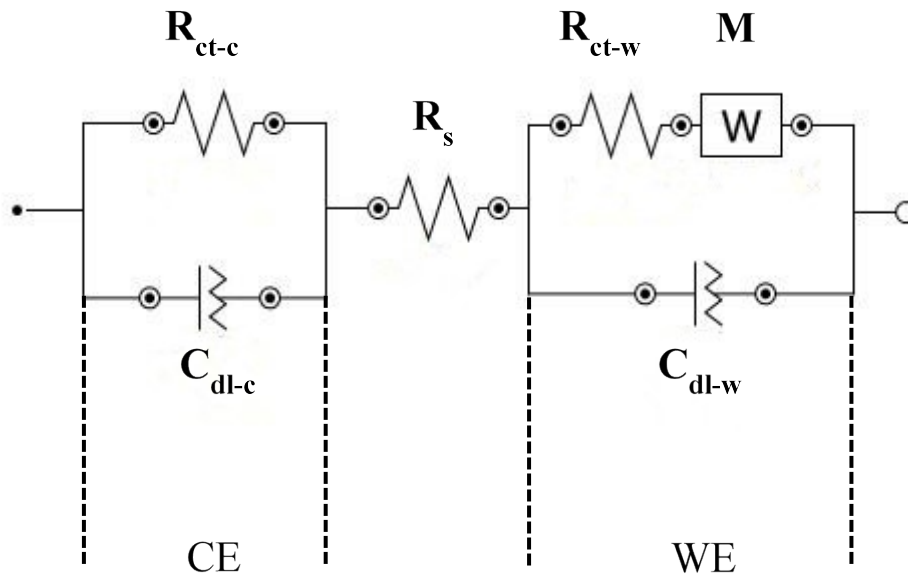


Figure S3. Equivalent circuit of the three-electrode system in the micro-fluidic biochip.



Electrical element	Corresponding impedance expression
Solution resistance	R_s
Double layer capacitance	$1/j\omega C_{dl}$
Charge transfer resistance	R_{ct}
Restricted linear diffusion with reflecting boundary	$Z_M(\omega) = R_d \frac{\coth \sqrt{\tau_d j \omega}}{\sqrt{\tau_d j \omega}}$

Figure S4. Optimization of Antibody Concentration.

EIS responses of GA/ITO electrode modified with different concentrations (1 $\mu\text{g/mL}$, 5 $\mu\text{g/mL}$, 10 $\mu\text{g/mL}$) of antibodies.

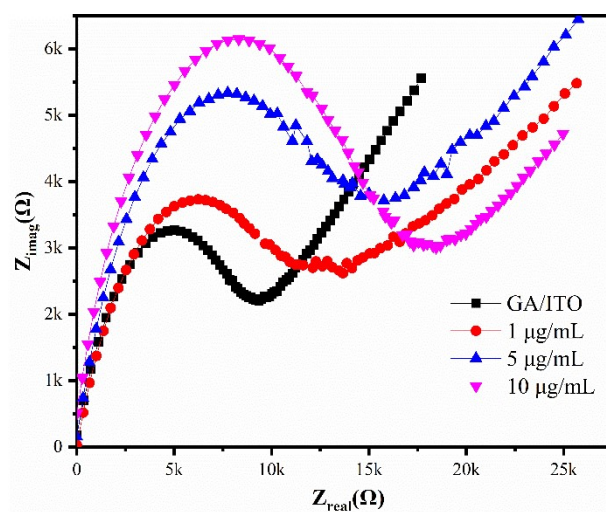


Figure S5. Selectivity of the biochip and the detection of CEA in diluted serum samples.

EIS responses of different analytes including AFP, AFP& CEA, AFP& IgG, AFP& Glucose, IgG, Glucose.

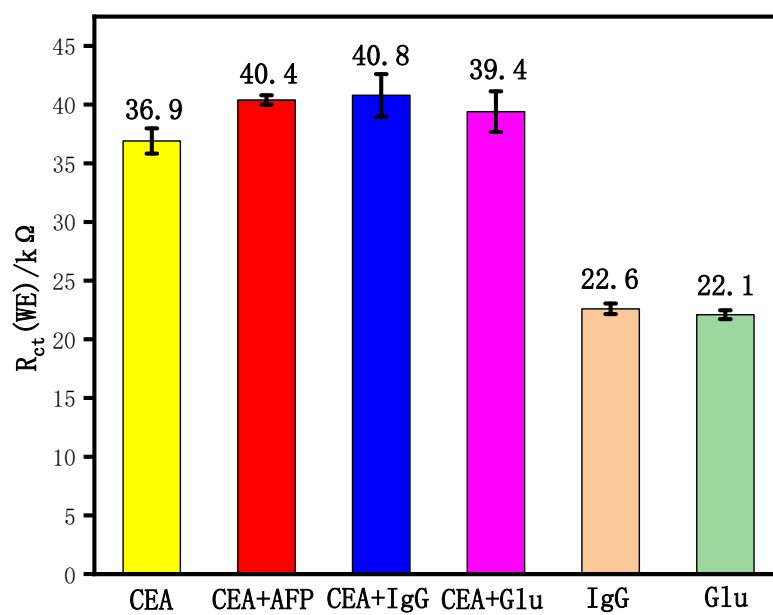
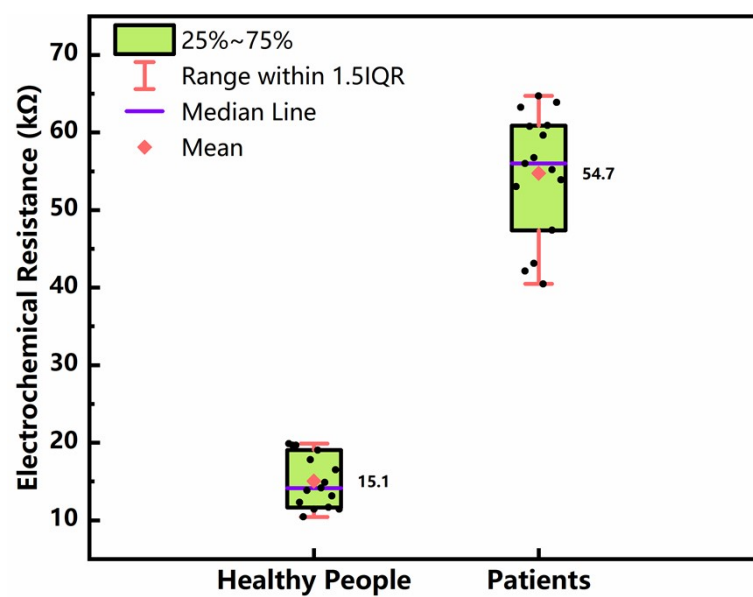


Figure. S6. The EIS responses of real specimens from patients and healthy people.



S2: Supporting tables: Table S1~S2.

Table S1. Comparison with other reports on CEA/AFP detection

No.	Sensitive Material	Method	Test Range (ng/mL)		Detection Limit (pg/mL)		Ref
			AFP	CEA	AFP	CEA	
1	carboxy graphene nanosheets	Electrochemical Immunoassay	0.5 - 60		50.0	100.0	1
2	chitosan nanocomposites	Electrochemical Immunoassay	0.05 - 100		30.0	20.0	2
3	chitosan coated copper and cadmium hexacyanocobaltate nanocubes	Electrochemical Immunoassay	0.025 - 250		10.9	17.5	3
4	platinum porous nanoparticles hybrid with metal ions	Electrochemical Immunoassay	0.05 - 200		50.0	2.0	4
5	cadmium, lead and copper alginate nanobeads	Electrochemical Immunoassay	0.01 - 100		10.0	8.6	5
6	Fe Oxides	Fluoroimmunoassay	-		70.0	300.0	6
7	multicolor quantum dots	Immunochroma-Tographic Assay	-		300.0	200.0	7
8	gold nanoparticles and mesoporous silica	Electrochemical Immunoassay	0.5-100	0.5-50	100		8
9	multicolored Au NCs-MoS ₂	Fluoroimmunoassay	0.5-60	0.5 - 120	160	210	9
10	streptavidin-phycoobiliproteins	ELISA	0 - 50		250	280	10
11	gold nanoparticle and silver nanoparticles	Color Encoded Assay	0.5 - 100		500		11
12	Porous 3D graphene aerogel	Microfluidic Electrochemical Immunoassay	0.01 -10	0.01 - 50	7.9	6.2	This work

Table S2. Comparison with other reports on EXOs detection

No.	Sensitive Material	Method	Test Range (μL^{-1})	Detection Limit (μL^{-1})	Ref
1	Probe DNAs & $\text{Ru}(\text{NH}_3)_6^{3+}$	Electrochemical Immunoassay	-	70	12
2	Quantum dots	Electrochemical Immunoassay	$10^2 - 10^7$	100	13
3	Graphene oxide-DNA aptamer	Fluoroimmunoassay	$3.0 \times 10^4 - 6.0 \times 10^4$	2.1×10^4	14
4	Copper Nanoparticles	Fluoroimmunoassay	$7.5 \times 10^4 - 1.5 \times 10^7$	4.8×10^4	15
5	HER-2 antibody	Electrochemical Immunoassay	-	4.7×10^5	16
6	Surface marker mediated signal amplification	Electrochemical Immunoassay	$10^2 - 10^6$	200	17
7	Bivalent-cholesterol-labelled DNA anchor	Fluoroimmunoassay	$2.3 \times 10^3 - 2.3 \times 10^5$	2.2×10^3	18
8	Porous 3D graphene aerogel	Microfluidic Electrochemistry	$2.5 \times 10^1 - 2.5 \times 10^7$	9.8	This work

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