Supporting Information for

Electrochemical cloth-based DNA sensors (ECDSs): A new class of electrochemical gene sensors

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Chemicals and materials

The oligonucleotide sequences (**Table S1**), TE buffer (containing 10 mM Tris-HCl, and 1 mM ethrylene diamine tetracetic acid (EDTA), pH 8.0), 10× phosphate-buffered saline (PBS) (containing 0.1 M NaH_2PO_4, 0.1 M Na_2HPO_4, and 0.1 M KCl, pH 7.4), mercaptoacetic acid (MPA) (>90%), bovine serum albumin blocking buffer (BSA-BB), AluI restriction enzyme, 10× Buffer Tango (with BSA), Triton X-100 (>98%), and native PAGE preparation kit were acquired from Sangon Co., Ltd. (Shanghai, China). Multi-walled carbon nanotubes (MWCNTs) (>97%, main range of diameter: 10-30 nm, length: >2 μm) were obtained from Shenzhen Nanotech Port Co. Ltd. (Shenzhen, China). Ferrocenecarboxylic acid (FcA) (>98%) was acquired from Tokyo Chemical Industry Co., Ltd. (Shanghai, China). Cadmium chloride hemi(pentahydrate) ($CdCl_2 \cdot 2.5H_2O$) was purchased from J&K Scientific Ltd. (Beijing, China). Sodium borohydride ($NaBH_4$) (98%), Telluriu (99.99%), poly(diallyldimethylammonium chloride) (PDDA) solution (20%), N-(3-Dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride (EDC) (98%), N-Hydroxysuccinimide (NHS) (98%), glutaraldehyde (GA) (50%), chitosan (CS) (≥95%, deacetylation), and potassium ferricyanide (>98%) were acquired from Shanghai Aladdin Bio-Chem Technology Co., Ltd. (Shanghai, China). Potassium chloride was purchased from Sigma-Aldrich (St. Louis, MO, USA). NaOH (≥96%), HCl (36-38%), H_2SO_4 (95-98%) and HNO_3 (65-68%) were bought by Guangzhou Chemical Reagent Factory (Guangzhou, China). The bacterial strains (*Listeria monocytogenes* (*L. monocytogenes*) CMCC54007, *Escherichia coli* (*E. coli*) O157:H7 GW1.0202 and *Listeria ivanovii* (*L. ivanovii*) ATCC19119) were obtained from Guangzhou Institute of Microbiology (Guangzhou, China). *Salmonella*

typhimurium (*S. typhimurium*) ATCC14028 was purchased from Shanghai Luwei Technology Co., Ltd (Shanghai, China). The TIAMamp Bacterial Genomic DNA Extraction Kit, and proteinase K (20 mg/mL) were purchased from Tiangen Biotech Co., Ltd. (Beijing, China). Whole milk was purchased from an local supermarket. White plain weave cloth (100% cotton) was bought from Guangzhou Haiyin Cloth Confluence Co., Ltd. (Guangzhou, China). Conductive carbon ink (CNB-7, <60 Ω per square) was purchased from Jiangsu Kenuan Thermal Energy Science and Technology Co., Ltd. (Xuyi, China). Wax crayons (Detong Co., Ltd., Guangzhou, China) and smoothing utensil were acquired from a local grocery store.

Apparatus

Pt electrode (PT213) and Ag/AgCl electrode (R0305) were produced from Beijing Instrument Electric Technology Co., Ltd. (Beijing, China). An electrochemical workstation (CHI660E, Shanghai Chenhua Instrument Co., Ltd., (Shanghai, China). A microcomputer temperature-controlled heating board (YH-946B) was produced by Guangzhou Yihua Electronic Equipments Co., Ltd. (Guangzhou, China). A centrifuger (H1650) was purchased from Hunan Cence Instrument Co., Ltd. (Hunan, China). The A200 Gradient Thermal cycler was acquired from Hangzhou LongGene Scientific Instruments Co., Ltd. (Hangzhou, China). A constant temperature and humidity chamber (HWS-508) was acquired from Beijing Heng Nuo Li Xing Science and Technology Co., Ltd. (Beijing, China). A oven (DHG-9035A) was bought from Shanghai Tensuc Experimental Instrument Manufacturing Co., Ltd. (Shanghai, China). A ultrasonic cleaner (KQ-100DA, 100 W, 40 KHz) was provided by Kunshan

Ultrasonic Instruments Co. Ltd. (Kunshan, China). The gel imaging system (JY026) and electrophoresis apparatus (JY-SCZ2+) were produced from Beijing Junyi Electrophoresis Co., Ltd. (Beijing, China). The pH meter (PB-10) and analytical balance (BSA2245-CW) were purchased from Sartorius Scientific Instruments (Beijing) Co., Ltd. (Beijing, China).

Preparation of target DNA (TD)

The *Listeria monocytogene* strains stored in glycerol at -80 ° were revived for 30 min in a 37 °C water bath. Then, 0.5 mL of bacterial solution was inoculated into Luria-Bertani broth (20 mL deionized (DI) water, 0.06 g beef extract, 0.1 g peptone, 0.1 g NaCl, pH 7.0~7.2) and shaken for 12 h at 150 rpm in a 37 °C culture oscillator. Next, the cultured bacteria were collected by centrifuging for 20 min at 5000 rpm at room temperature.

Genomic DNA was extracted from the cultured bacteria by the extraction kit. The primers were designed to target 75-bp amplicons of the *hlyA* gene. PCR was performed in 25 μL reaction mixture with 1× PCR buffer, 0.3 mM dNTPs, 1 μM each primer, 0.05 units/μL Taq polymerase, and 2 μL extracted DNA. During the PCR, DNA was initially denatured at 95 °C for 5 min, followed by 35 cycles of amplification (95 °C for 30 s, 56 °C for 30 s, 72 °C for 30 s) and finally extended for 4 min at 72 °C. The double-stranded amplification products were observed in agarose gel electrophoresis (2.0%). The single-stranded target DNA (TD) was obtained by denaturing double-stranded amplicons (89 ng/μL) at 95 °C for 5 min and then rapidly cooling at -20 °C over 1 h.

Preparation of CdTe QDs/MWCNTs nanocomposite (CdTe-MWCNTs)

First, mercaptoacetic acid (MPA)-capped CdTe QDs (MPA-CdTe QDs) were prepared using a procedure described elsewhere with slight modifications.[1] Briefly, 46 mg tellurium powder and 92 mg $NaBH_4$ were added into 20 mL DI water, and the solution was reacted under oxygen-free conditions in a 60 °C water bath until it turned mauve. Then, 140 μL MPA were added into 60 mL of 1.34 mM $CdCl_2$ solution, and 1 M NaOH solution was trickled into the system until the pH value was 11.2. The obtained mixture was transferred into a three-mouth flask, stirred and purged with nitrogen for 30 min. Last, the mauve NaHTe supernatant was rapidly injected into this mixture solution. The resulting mixture was heated and refluxed under nitrogen flow for 4 h at 100 °C to obtain the desired QDs. The MPA-CdTe QDs solution was then washed three times with alcohol for purification.

Second, the MWCNTs were pretreated with acid and poly(diallyldimethylammonium chloride) (PDDA) according to our previous method with a minor modification.[2] First, 1 g of MWCNTs was introduced into 40 mL mixture of HNO_3 (65-68%) and H_2SO_4 (95-98%) (v/v, 1:3), and the mixture was continuously ultrasonicated for 8 h at room temperature. Then, the solid-liquid mixture was centrifuged and washed many times with DI water until the pH got neutral. The acid-treated MWCNTs were further filtered, followed by drying in an 80 °C oven for 24 h. Next, 5 mg as-prepared MWCNTs were dispersed into 1 mL NaCl (0.5 M)-containing PDDA (0.25 wt%) solution, and ultrasonicated over 1 h. After removal of the supernatant, PDDA-modified MWCNTs (PDDA-MWCNTs) were obtained, washed three times, and redispersed in water.

Finally, the MPA-CdTe QDs solution and PDDA-MWCNTs dispersion were used to prepare the CdTe QDs/MWCNTs nanocomposite (CdTe-MWCNTs). In brief, 2 mL

MPA-CdTe QDs solution (3 mg/mL) was added to 2 mL PDDA-MWCNTs dispersion (1 mg/mL). After a brief ultrasonication, the reaction mixture was stirred for 30 min, and then centrifuged and redispersed in water. This procedure led to the formation of a homogeneous CdTe-MWCNTs nanocomposite solution.

Digestion of genomic DNA

First, genomic DNA was extracted from the *L. monocytogene* cultures, and its concentration was measured to be 18.5 ng/μL. Then, the *AluI* restriction enzyme digestion was performed to obtain the target fragment sequences from the genomic DNA samples. Different concentrations of genomic DNA samples (0, 0.05, 0.1, 0.2, 1, 2, and 10 ng/μL) were digested for 60 min at 37 ^{o}C in 20 μL reaction mixture (containing 2× Buffer Tango) with corresponding units of *AluI* restriction enzyme (0.001 unit for 1 ng genomic DNA). Next, to inactivate the enzyme and obtain the single-stranded target fragments, the enzyme digestion mixture was heated for 10 min at 90 ^{o}C, and then it was rapidly cooled to -20 ^{o}C and maintained for more than 60 min. Finally, the genomic DNA digestion products were detected by the ECDSs under the optimum conditions.

Preparation of milk samples spiked with *L. monocytogenes*

After the bacteria were cultured, the concentration of collected bacteria was confirmed using the optical absorbance at 600 nm, where the absorbance of 0.125 was equivalent to about 2.5×10^{7} CFU/mL. For milk samples used for PCR amplification, 10-fold serial dilutions ranging from 2.5×10^{7} to 2.5×10 CFU/mL were performed in 1× PBS. Then, different

concentrations of bacteria (100 μL) were inoculated into milk (900 μL) to prepare contaminated milk samples (0-2.5×10^6 CFU/mL). For the negative-control milk sample without any bacteria, 1× PBS buffer solution (100 μL) was used to replace the bacterial strain. Next, to reduce the effect of protein and lipid components in milk, each contaminated milk sample was mixed with 100 μL Triton X-100 (0.1%) and 10 μL proteinase K, and the mixture was maintained at 37 ^{o}C for 60 min. Finally, the genomic DNA samples were extracted from these pretreated milk samples, and were used for PCR amplification. For milk samples used for enzyme digestion of genomic DNA, similar procedures were used for preparation of milk samples with different concentrations of *L. monocytogenes* (0, 2.5×10^4, 5×10^4, 2.5×10^5, 5×10^5, and 2.5×10^6 CFU/mL), pretreatment of milk samples, and extraction of genomic DNA. Next, the extracted genomic DNA samples (5 μL) were used for enzyme digestion, during which the reaction mixture (20 μL) containing 2× buffer Tango and 0.05 units/μL AluI restriction enzyme was kept at 37 ^{o}C for 60 min.

Table S1. The sequences of oligonucleotides used in this work

Name	**Sequences (5' to 3')**
NH_2-DNA1	TAAGTCTCCGAGGTT ATTCAGAGGCTCCAA-$(CH_2)_6$-NH_2
NH_2-DNA2	AACCTCGGAGACTTATTGGAGCCTCTGAAT-$(CH_2)_6$-NH_2
CP	NH_2-$(CH_2)_6$-CTATCCTTCAAAGCCGTAATTTACGGAGGTT CCGCAAAAGATACGTGCGAAC
One-base mismatched CP (C1)	NH_2-$(CH_2)_6$-CTATCCTTCAAAGCCGTAATTTACGGAG**A**TT CCGCAAAAGATACGTGCGAAC
Two-base mismatched CP (C2)	NH_2-$(CH_2)_6$-CTATCCTTCAAAGCCGTAATTTACGGAG**AC**T CCGCAAAAGATACGTGCGAAC
HP (for DL-HCR)	TAAGTCTCCGAGGTTGTTCGCACGTGAAGTTCAAATCA TCGACGGCAACCTCGGAGACTTATTGGAGCCTCTGAAT
One-base mismatched HP (H1)	TAAGTCTCCGAGGTTGTTCGCACGTGAAGTTCAAATCA TC**A**ACGGCAACCTCGGAGACTTATTGGAGCCTCTGAAT
Two-base mismatched HP (H2)	TAAGTCTCCGAGGTTGTTCGCACGTGAAGTTCAAATCA TC**AT**CGGCAACCTCGGAGACTTATTGGAGCCTCTGAAT
HP_D (for Single-HCR)	GTTCGCACGTGAAGTTCAAATCATCGACGGCAACCTCG GAGACTTATTGGAGCCTCTGAAT
Forward/reverse primers (for *L. monocytogenes*)	TCCTTCAAAGCCGTAAT/TAAGTCTCCGAGGTTGC
Forward/reverse primers	TCAGCAAAAGAGAATCCGTGA/TGACGATGAAGTTAAT

(for *S. typhimurium*)	GATGGTATAAA
Forward/reverse primers (for *E. coli* O157:H7)	CACTCGAGGCGTTTTT/TGGAACTCCTGGCTGATTAAGT
Forward/reverse primers (for *L. ivanovii*)	GGTACGCCCGGATTTTTCTTAT/ACTAGGGTGAAGGCTG CATT
TD (amplicons of *L. monocytogenes hlyA* gene) (75-bp)	TAAGTCTCCGAGGTTGCCGTCGATGATTTGAACTTCATC TTTTGCGGAACCTCCGTAAATTACGGCTTTGAAGGA
Amplicons of *E. coli* O157:H7 (175-bp) (for evaluation of selectivity)[(a)]	CACTCGAGGCGTTTTTCGTTATGTATAAATAAGGAGCA CACCATGCAATATGCCATTG**C<u>A</u>G<u>G</u>GTGG<u>C</u>CT<u>G</u>T<u>T</u>GCT G<u>G</u>CT<u>G</u>CCCT<u>T</u>CC<u>GAA</u>TC<u>T</u>TTA<u>CTT</u>GA<u>GCG</u>A<u>A</u>T<u>C</u>AGG GCATTTAA<u>ACG</u>TGAC<u>TGA</u>C<u>GGA</u>**AACGCCTTATCGACA TACTTAATCAGCCAGGAGTTCCAAAAA
Amplicons of *S. typhimurium* (260-bp) (for evaluation of selectivity)[(a)]	TCAGCAAAAGAGAATCCGTGAATTTGTACTCGATTTTA CATTGTTGTCGATACTCAGTAATTATTGATATATAATTC AGCAAATTTGACAAA**T<u>T</u>C<u>GT</u>T<u>TC</u>A<u>GA</u>ATGAC<u>C</u>TTG<u>C</u>T T<u>T</u>TA<u>T</u>G<u>TGAACTTCAT</u>TCA<u>TT</u>TTTC<u>AA</u>A<u>C</u>CGTC<u>T</u>C<u>A</u>T ATAAT<u>G</u>T<u>TTT</u>C<u>A</u>T<u>G</u>C<u>A</u>**AATTTTCTTGTCATACACATTG AAAACATATAACAAAAATATGTTCACGACATATCCATT TAATATTTTATACCATCATTAACTTCATCGTCA
Amplicons of *L. ivanovii* (215-bp) (for evaluation	GGTACGCCCGGATTTTTCTTATCAAAATTAGCCCCTTGT TTTAAAATATCTCG**<u>TAA</u>A<u>TCT</u>CTTAAATCT<u>CC</u>A<u>TC</u>A<u>AT</u>**

of selectivity)[a]	**T<u>ATTT</u>CT<u>ACTTCATCTTTTGCT</u><u>GAACC</u>A<u>CC</u>A<u>TA</u>G<u>AT</u>C**
	<u>AC</u>C<u>GCTTT</u>A<u>AAT</u>G<u>A</u>AGAATTTTGAATGATATTCTCTAA
	TTCAGTATCACCTTTAACTGATTTACCTTTAAATGCAGC
	ATCGAATGCAGCCTTCACCCTAGT
Target fragment	**AG↓CT**AATTCATTGTCTTTTAAGAAGTTTGTTGTATAGG
sequences obtained by	CAATGGGAACTCCTGGTGTTTCCCGGTTAAAAGTAGCA
the *AluI* restriction	CCTTTTTTCAAAATATCTCG<u>TAAGTCTCCGAGGTTGCCG</u>
enzyme digestion of *L.*	<u>TCGATGATTTGAACTTCATCTTTTGCGGAACCTCCGTAA</u>
monocytogenes genomic	<u>ATTACGGCTTTGAAGGA</u>AGAATTTTTGATGATATTTGTC
DNA[b]	AGTTCTACATCACCTGAGACAGATTTCCCACTTACGGC
	AGCGTCAAAAGC**AG↑CT**

Note: Superscript (a)-based on the search from the NCBI (National Center for Biotechnology Information) genebank, the sequences written in bold (75 bp) are found to have a relative higher similarity to that of the TD (underlined for the similar sequences), and for amplicons of *E. coli* O157:H7, *S. typhimurium* and *L. ivanovii* gene fragments, their similarity is 38.7%, 48% and 69.3%, respectively. Superscript (b)-the enzyme digestion site is marked in bold, and the resulting sequence length is 243 bp (underlined for the 75-bp TD fragment).

Table S2. Materials cost estimation for a single sensing device

Material	Quantity used per device	Cost per device ($)
Cotton cloth ($3.70/m^2)	0.001 m^2	0.0037
Solid wax ($0.02/g)	0.04 g	0.0008
Conductive carbon ink ($0.087/g)	0.17 g	0.01479
Conductive double-sided tape ($0.05/m)	0.02 m	0.001
CdTe-MWCNTs ($0.005/mL)	0.002 mL	0.00001
CS($0.0003/mL)	0.002 mL	0.0000006
GA ($0.0001/mL)	0.002 mL	0.0000002
CP-DNA ($0.004828/μL)	5 μL	0.0241
BSA-BB ($0.0989/mL)	0.002 mL	0.0002
Total		0.045

Table S3. Comparison between the proposed ECDSs and other electrochemical sensors for gene detection of *L. monocytogenes*

Electrode	Modified materials	Method	Detection limit (fM)	Reference
Au electrode	NA	Cyclic voltammetry	46000000	**3**
Graphite electrode	Carboxymethylated dextran film	Cyclic voltammetry	200000	**4**
Au electrode	NA	Chronoamperometry	200000	**5**
Au electrode	NA	Chronoamperometry	600	**6**
Carbon ionic liquid electrode	Au nanoparticles/reduced graphene	Differential pulse voltammetry	290	**7**
Au electrode	NA	Electrochemical impedance spectroscopy	234	**8**
Carbon ionic liquid electrode	Partially reduced graphene/Au nanoparticles	Differential pulse voltammetry	31.7	**9**
Aluminum Interdigitated	Au nanoparticles	Differential pulse	10	**10**

electrode		voltammetry			
Carbon ink screen-printed cloth-based electrode	Glutaraldehyde/chitosan/CdTe-MWCNTs	Differential pulse voltammetry	8.74		**This work**

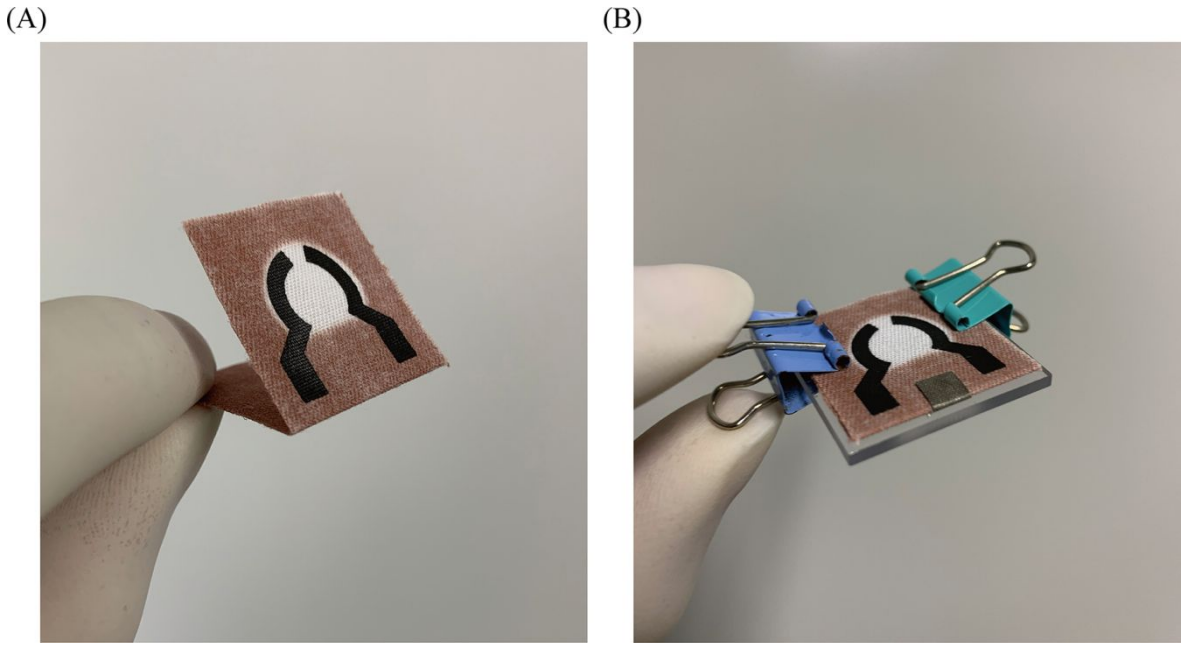

Figure S1. Photo of a typical 3D cloth-based DNA sensor. (A) The screen-printed side was folded to face outwards. (B) The sensor was fixed on a plastic support.

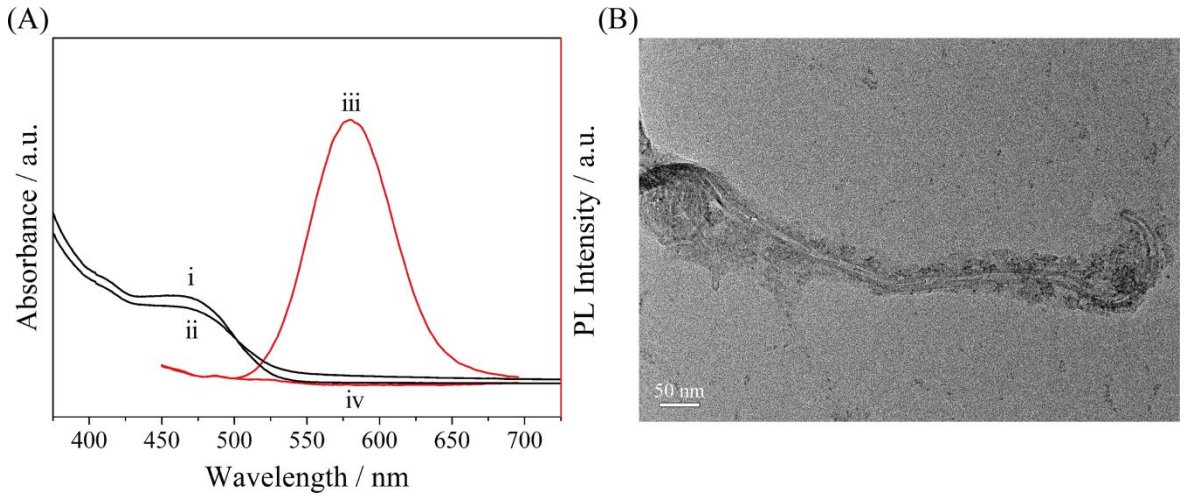

Figure S2. (A) UV-vis absorption and fluorescence (photoluminescence, PL) spectra of MPA-CdTe QDs (i, iii) and CdTe-MWCNTs (ii, iv). (B) TEM image of the CdTe-MWCNTs.

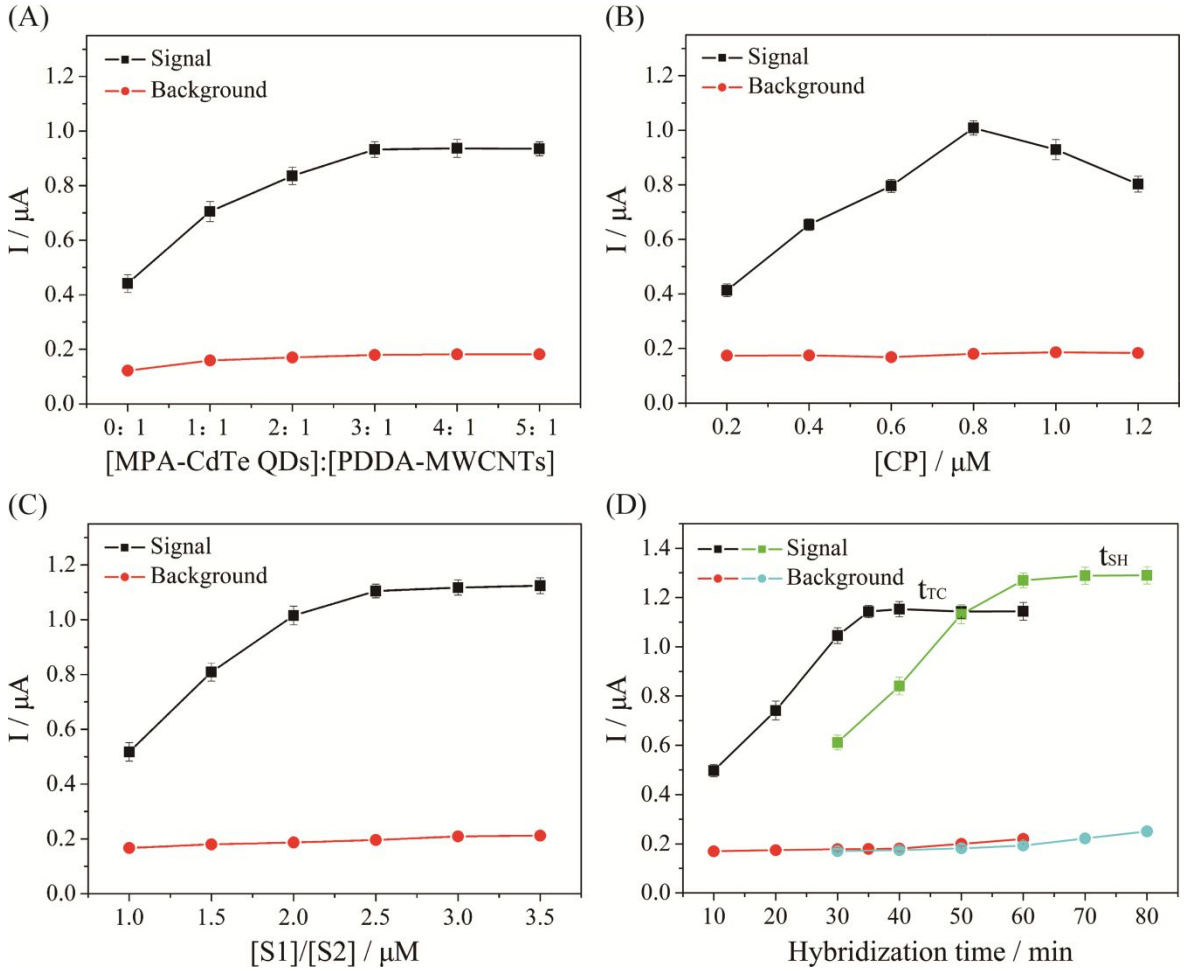

Figure S3. Dependences of DPV peak currents on mass ratios of MPA-CdTe QDs to PDDA-MWCNTs ([MPA-CdTe QDs]:[PDDA-MWCNTs]) (A), concentration of CP ([CP]) (B), concentration of S1/S2 ([S1]/[S2]) (C), and hybridization time of TD with CP (t_{TC}) and S1/S2 with HP) (t_{SH}) (D). In panel (A), [CP]-1 μM, [S1]/[S2]-2 μM, t_{TC}-30 min, t_{SH}-40 min and [TD]-50 pM. In panel (B), [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [S1]/[S2]-2 μM, t_{TC}-30 min, t_{SH}-40 min and [TD]-50 pM. In panel (C), [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, t_{TC}-30 min, t_{SH}-40 min and [TD]-50 pM. In panel (D), [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, [S1]/[S2]-2.5 μM, t_{SH}-40 min and [TD]-50 pM (for t_{TC} optimization); and [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, [S1]/[S2]-2.5 μM, t_{TC}-35 min and [TD]-50 pM (for t_{SH} optimization). The error bars represent the standard deviations of five independent

measurements.

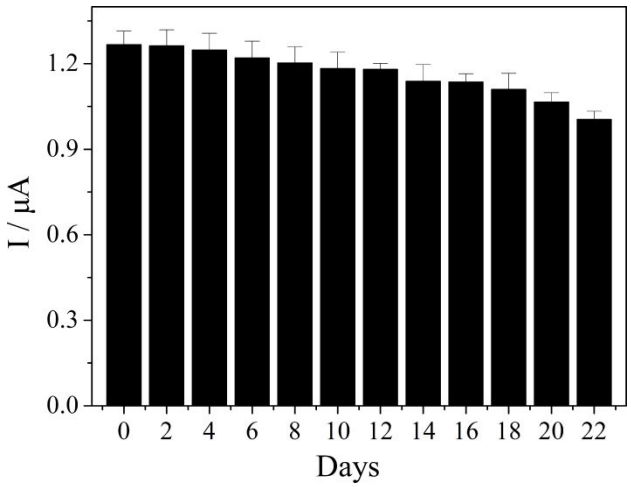

Figure S4. Storage stability of the proposed sensor. Experimental conditions: [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, [S1]/[S2]-2.5 μM, t_{TC}-35 min, t_{SH}-60 min, and [TD]-50 pM. The error bars represent the standard deviations of five independent measurements.

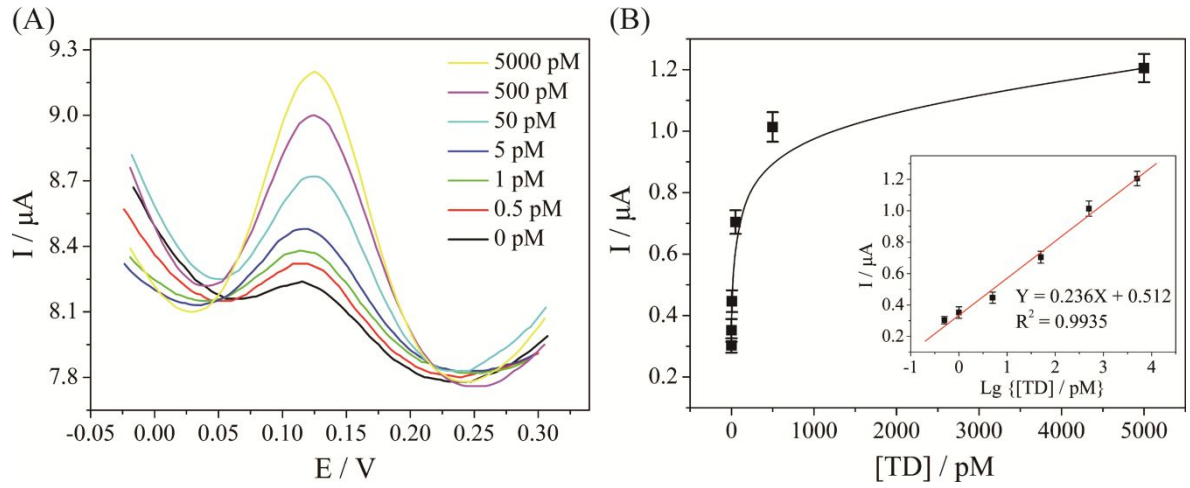

Figure S5. (A) DPV response curves of the sensor with traditional linear HCR at different TD concentrations (0, 0.5, 1, 5, 50, 500, and 5000 pM). (B) Plot of the DPV peak current versus the TD concentration (insert: the linear relationship between the peak current and the logarithm of TD concentration). Experimental conditions: [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, [S1]/[S2]-2.5 μM, t_{TC}-35 min, and t_{SH}-60 min. The error bars represent the standard deviations of five independent measurements.

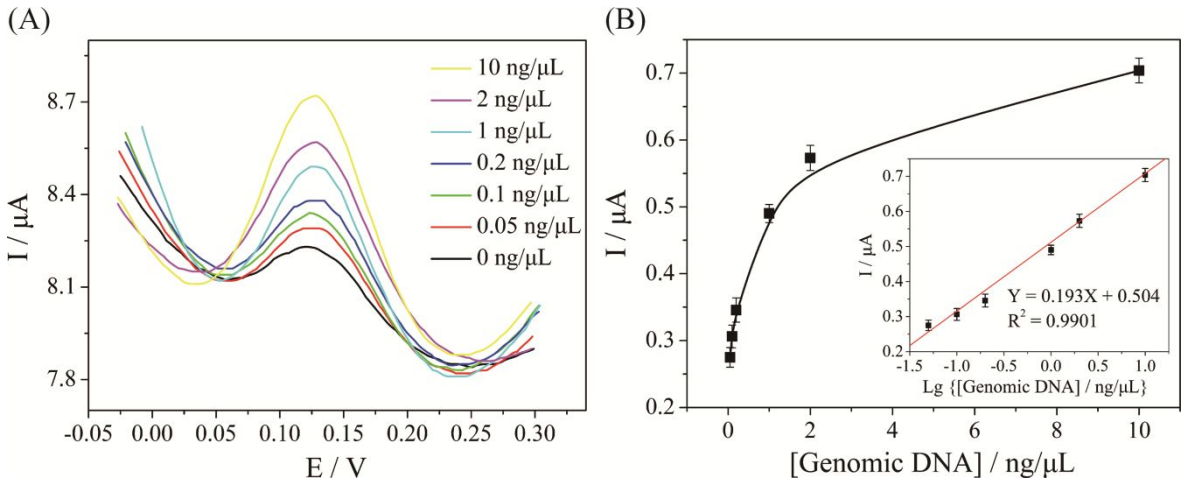

Figure S6. (A) DPV response curves at different concentrations of *L. monocytogenes* genomic DNA (0, 0.05, 0.1, 0.2, 1, 2, and 10 ng/μL). (B) Plot of the DPV peak current versus the genomic DNA concentration (insert: the linear relationship between the peak current and the logarithm of genomic DNA concentration). Experimental conditions: [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, [S1]/[S2]-2.5 μM, t_{TC}-35 min, and t_{SH}-60 min. The error bars represent the standard deviations of five independent measurements.

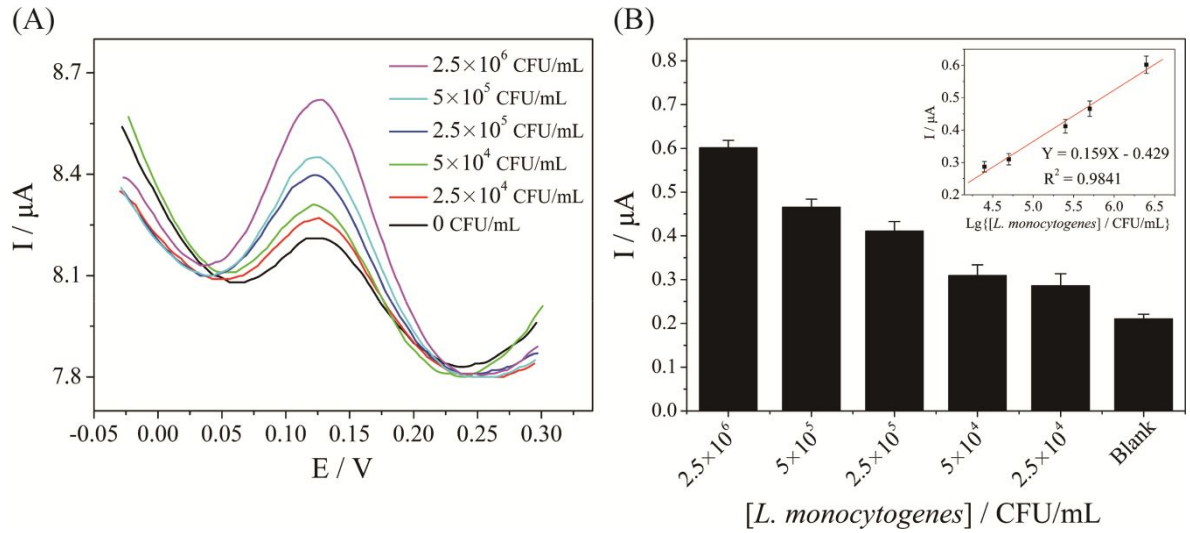

Figure S7. (A) DPV response curves of enzyme digestion products from genomic DNA samples at different concentrations of *L. monocytogenes* in milk (0, 2.5×10^4, 5×10^4, 2.5×10^5, 5×10^5, and 2.5×10^6 CFU/mL). (B) Plot of the DPV peak current versus the *L. monocytogenes* concentration (insert: the linear relationship between the peak current and the logarithm of *L. monocytogenes* concentration). Experimental conditions: [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, [S1]/[S2]-2.5 μM, t_{TC}-35 min, and t_{SH}-60 min. The error bars represent the standard deviations of five independent measurements.

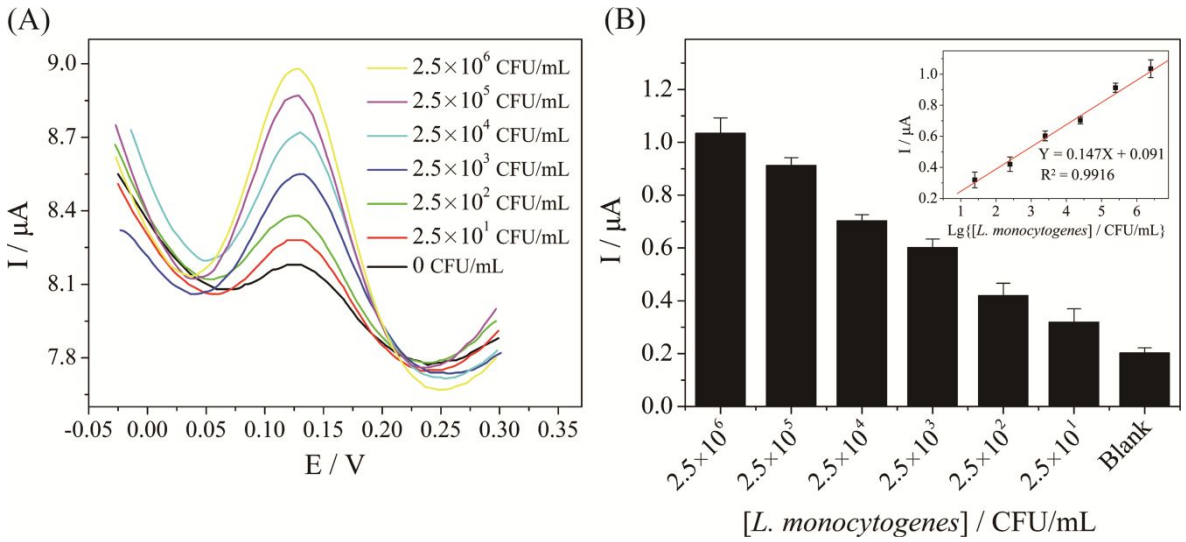

Figure S8. (A) DPV response curves of PCR products obtained from different concentrations of *L. monocytogenes* in milk samples (0, 2.5×10^1, 2.5×10^2, 2.5×10^3, 2.5×10^4, 2.5×10^5, and 2.5×10^6 CFU/mL). (B) Plot of the DPV peak current versus the *L. monocytogenes* concentration (insert: the linear relationship between the peak current and the logarithm of *L. monocytogenes* concentration). Experimental conditions: [MPA-CdTe QDs]:[PDDA-MWCNTs]-3:1, [CP]-0.8 μM, [S1]/[S2]-2.5 μM, t_{TC}-35 min, and t_{SH}-60 min. The error bars represent the standard deviations of five independent measurements.

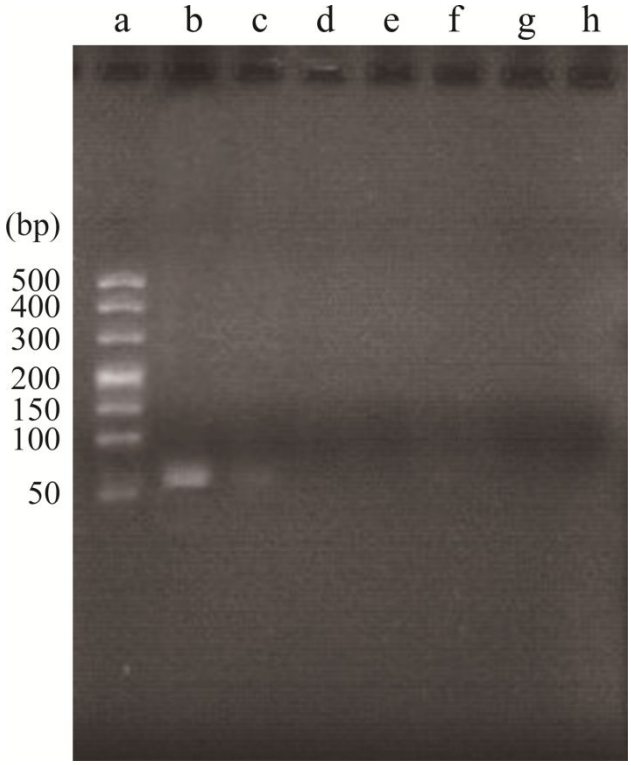

Figure S9. Agarose gel electrophoresis of PCR products amplified from genomic DNA samples extracted from different concentrations of *L. monocytogenes* in milk samples. Lane a-DNA ladder marker, and lanes b-h-2.5×10^6, 2.5×10^5, 2.5×10^4, 2.5×10^3, 2.5×10^2, 2.5×10^1 and 0 CFU/mL. Agarose gels (2%) were typically run at 90 V for 30 min in 1× TBE.

References

(1) Zhang, M.; Wan, F. W.; Wang, S. W.; Ge, S. G.; Yan, M.; Yu, J. H. Determination of L-proline based on anodic electrochemiluminescence of CdTe quantum dots. *J. Lumin.* **2012**, *132*, 938-943.

(2) Wang, D.; Liang Y.; Su, Y.; Shang, Q. P.; Zhang, C. S. Sensitivity enhancement of cloth-based closed bipolar electrochemiluminescence glucose sensor via electrode decoration with chitosan/multi-walled carbon nanotubes/graphene quantum dots-gold nanoparticles. Biosens. Bioelectron. 2019, 130, 55-64.

(3) Gao, H. W.; Qin P.; Lin, C.; Shang, Z. M.; Sun, W. Electrochemical DNA biosensor for the detection of *Listeria Monocytogenes* using toluidine blue as a hybridization indicator. *J. Iran. Chem. Soc.* **2010**, *7*, 119-127.

(4) Hajdukiewicz, J.; Boland, S.; Kavanagh, P.; Leech, D. An enzyme-amplified amperometric DNA hybridisation assay using DNA immobilised in a carboxymethylated dextran film anchored to a graphite surface. *Biosens. Bioelectron.* **2010**, *25*, 1037-1042.

(5) Kavanagh, P.; Leech, D. Redox polymer and probe DNA tethered to gold electrodes for enzyme-amplified amperometric detection of DNA hybridization. *Anal. Chem.* **2006**, *78*, 2710-2716.

(6) Hajdukiewicz, J.; Boland, S.; Kavanagh, P.; Nowicka, A.; Stojek, Z.; Leech, D. Enzyme-amplified amperometric detection of DNA using redox mediating films on gold microelectrodes. *Electroanalysis* **2009**, *21*, 342-350.

(7) Sun, W.; Qi, X. W.; Zhang, Y. Y.; Yang, H. R.; Gao, H. W.; Chen, Y.; Sun, Z. F. Electrochemical DNA biosensor for the detection of *Listeria monocytogenes* with dendritic nanogold and electrochemical reduced graphene modified carbon ionic liquid electrode. *Electrochim. Acta* **2012**, *85*, 145-151

(8) Kashish, Soni, D. K.; Mishra, S. K.; Prakash, R.; Dubey, S. K. Label-free impedimetric detection of *Listeria monocytogenes* based on poly-5-carboxy indole modified ssDNA probe. *J. Biotechnol.* **2015**, *200*, 70-76.

(9) Niu, X. L.; Zheng, W.; Yin, C. X.; Weng, W. J.; Li, G. J.; Sun, W.; Men, Y. L. Electrochemical DNA biosensor based on gold nanoparticles and partially reduced graphene oxide modified electrode for the detection of *Listeria monocytogenes* hly gene sequence. *J. Electroanal. Chem.* **2017**, *806*, 116-122.

(10) Rajapaksha, R. D. A. A.; Hashim, U.; Natasha, N. Z.; Afnan Uda, M. N.; Thivina, V.; Fernando, C. A. N. Gold nano-particle based Al Interdigitated electrode electrical biosensor for specific ssDNA target detection. In 2017 IEEE Regional Symposium on Micro and Nanoelectronics (RSM), Batu Ferringhi, Penang, Malaysia, August 23-25, pp **2017**, 191-194.