Electronic Supplementary Information to

Facile and sensitive chemiluminescence detection of H₂O₂ and glucose by a gravity/capillary flow and cloth-based lowcost platform

Huijie Li, Dan Wang, Cuiling Liu, Rui Liu, Chunsun Zhang*

MOE Key Laboratory of Laser Life Science & Institute of Laser Life Science, College of Biophotonics, South China Normal University, Guangzhou 510631, China

* The contact information of the corresponding authors is:

Chunsun Zhang, PhD, Professor

MOE Key Laboratory of Laser Life Science & Institute of Laser Life Science,

College of Biophotonics, South China Normal University,

No. 55, Zhongshan Avenue West, Tianhe District,

Guangzhou, 510631,

P.R. China

Tel: +86-20-85217070-8501, Fax: +86-20-85216052

E-mail: zhangcs@scnu.edu.cn; zhangcs_scnu@126.com

Material	Quantity used per device	Assay cost per device (\$)
Cotton cloth (\$2.7/m ²)	15.8 cm ²	0.0043
Solid wax (\$0.02/g)	0.09 g	0.0018
HRP (\$0.47/mg)	10.5 µg	0.0049
Luminol (\$11.51/g)	6.2 μg	0.000071
PIP (\$3.17/g)	1.54 μg	0.0000049
GOD (\$0.00061/U)	3 U	0.00183
Total		0.011 (for H_2O_2 assay) or 0.013
		(for glucose assay)

Table S1. Materials cost estimation for a single H_2O_2 or glucose assay

Cloth material ^a	Weight of	Weight per	Threads per inch of	Thickness
	cloth	meter of thread	width	(µm)
	(mg/cm ²)	(mg)		
Cotton cloth	~10.12	~16.85	~77	~120
Linen-silk cloth	~14.80	~18.00	~71	~200
Linen cloth	~20.25	~70.69	~41	~260
Corduroy cloth	~30.47	~31.58	\sim 74 (crosswise), and	~450
			~43 (vertical)	

Table S2. Related parameters of white cloths with different components

Note: ^a these cloth materials were purchased from Guangzhou Haiyin Cloth Confluence Co. Ltd. (Guangzhou, China); Cotton cloth-white plain weave cotton cloth; Linen-silk cloth-white linen-silk blended cloth; Linen cloth-white linen cloth; and Corduroy cloth-white corduroy weave cotton cloth.

[Added] ^a (mM)	[Detected] ^b (mM)	RSD ^c (%)	Recovery ^d (%)
1	0.9946	3.97	99.46
2	1.9535	2.96	97.68
3	3.1157	5.58	103.86
4	4.1077	5.81	102.69
5	4.834	3.23	96.68

Table S3. Detection of H₂O₂ in spiked milk samples.

^a [Added] means the values that were added into the real sample. ^b [Detected] means the amount of H₂O₂ obtained according to the standard curve equation from five parallel detections. ^c The relative standard deviation (RSD) is calculated from five independent experiments. ^d Recovery means the [Detected]/[Added] ratio. Before the determination, the spiked samples were appropriately diluted with PBS.

Device types	CL reaction systems	Main apparatus used	LDR (mM)	LOD	References
				(µM)	
Large-size system	Luminol/H ₂ O ₂ /ferricy	Spiral flow cell, Peristaltic pump, four three-way	0.0022-0.45	1.8	[1]
	anide	solenoid valves, Perspex joint point, electronic			
		circuit, and photodiode			
Large-size system	Co(II)/H ₂ O ₂ /OH ⁻	Flow CL cell, and Biophysics CL analyzer with	0.005-1	2.6	[2]
		PMT detector			
Large-size system	Luminol/H ₂ O ₂ /AuNFs	Flow injection CL system consisting of a	0.03-3	10	[3]
		peristaltic pump, a CL detector and a PMT			
Large-size system	Co ₃ O ₄ -cored CDs/	N/A	0.01-1	10	[4]
	H_2O_2				
Large-size system	Luminol/H ₂ O ₂ /ferricy	Test tube, measuring cell of an Aminco Chem	0.02-10	20	[5]
	anide	Glow Photometer, and pulse integrator			

Table S4. Comparison of the presented cloth-based CL and other CL platforms for the H₂O₂ determination.

Large-size system	Luminol/H ₂ O ₂ /HRP	Perkin-Elmer	fluorescence	spectrophotometer	N/A	52.2	[6]
		with PMT det	ector, and silica	fibre			
Large-size system	Luminol/H2O2/HRP	Perkin-Elmer	fluorescence	spectrophotometer	0.1-3	670	[7]
		with PMT det	ector, and silica	fibre			
Lab-on-chip	CPPO/Cyalume green	PDMS chip, s	syringe pump, sp	oring-loaded probes,	0.01-1	10	[8]
(microfluidic) system	dye/H ₂ O ₂ /DMAP	and solution	on-processed	thin-film organic			
		photodiodes					
Lab-on-chip	Luminol/H2O2/HRP	Single planar	EWOD glass ch	ip, photomultiplier,	0.01-100	10	[9]
(microfluidic) system		and amplifier	circuit				
Lab-on-chip	Luminol-H ₂ O ₂ -Co(II)	Silicon/glass	chip, two syrin	ge pumps, injector	0.1-1	100	[10]
(microfluidic) system		valve, fluid/e	electrical conne	ctors, photodiodes,			
		transconducta	nce amplifier, an	d voltmeter			
Lab-on-chip	CPPO/9,10-	PDMS chip,	two syringes, s	yringe pump, thin-	~1-1000	~1000	[11]
(microfluidic) system	diphenylanthracene	film organic p	hotodiodes, and	electrometer			
	dye /H ₂ O ₂ /DMAP						

Lab-on-chipLuminol/H2O2/HRPWax-patterned cloth, and CCD camera0.5-5460[12](microfluidic) systemLuminol/H2O2/HRPWax-patterned cloth, and CCD camera0.01-109.07This work(microfluidic) system

Note: CL: chemiluminescence; LDR: linear dynamic range; LOD: limit of detection; Luminol: 3-aminophthalhydrazide; ferricyanide: potassium hexacyanoferrate(III); PMT: photomultiplier tube; AuNFs-flowerlike gold nanostructures; CDs: carbon dots; N/A: not available; HRP: horseradish peroxidase; CPPO: bis(2-carbopentyloxy-3,5,6-trichlorophenyl) oxalate; Cyalume green dye: 9,10-bis(phenylethynyl)anthracene; DMAP: 4-dimethylaminopyridine; PDMS: poly(dimethyl-siloxane); EWOD: electrowetting-on-dielectrics; and CCD: charge-coupled device.

Device types	CL reaction systems	Main apparatus used	LDR (mM)	LOD	References
				(µM)	
Large-size	Luminol/H ₂ O ₂ ^a	Ten-port selection valve, peristaltic pump, optical flow-	0.01-1	4	[13]
system		cell, PMT, and multi-function interface card			
Large-size	Luminol/H ₂ O ₂ /HRP-	Peristaltic pump, six-way injection valve, U-shaped glass	0.01-1	5	[14]
system	AuNPs	flow cell, and Biophysics CL analyzer with PMT detector			
Large-size	Luminol/H ₂ O ₂ /ferricy	Loop-type microdialysis probe, flow-through	0-14	10	[15]
system	anide	microdialyzer, microdialysis syringe pump, variable-speed			
		peristaltic pump, eight-channel injector valve, and PMT			
Large-size	Luminol/H ₂ O ₂ /FRP	Flow cell, and photodiode with preamplifier	0.05-2	~10	[16]
system					
Large-size	Luminol/H ₂ O ₂ /ferricy	Ten-position valve, peristaltic pump, holding coil,	0.03-0.6	15	[17]
system	anide	photodiode, and Radiometer chart recorder,			

Table S5. Comparison of the presented cloth-based CL and other CL platforms for the glucose determination.

Large-size	Luminol/H ₂ O ₂ /HRP/	Flow cell, peristaltic pump, six-way injection valve,	0.05-5	44	[18]
system	GO	Biophysics CL analyzer with PMT detector, and			
		fluorescence spectrophotometer			
Large-size	Luminol/H2O2/HRP	Microdialysis syringe pump, microdialysis probe,	0-12	50	[19]
system		peristaltic pump, injection valve, U-shaped glass flow cell,			
		and Biophysics CL analyzer with PMT detector			
Large-size	Luminol/H2O2/HRP	CL cuvette, X-Y-t recorder, and PMT	0.2-2	120	[20]
system					
Lab-on-chip	Luminol/H2O2/HRP-	Microchip, external magnet, vacuum pump, microscope,	0.02-350	5.2	[21]
(microfluidic)	MNPs	and luminescence spectrometer with PMT detector,			
system					
Lab-on-chip	Luminol/H2O2/ferricy	PDMS/glass chip, syringe pump, multi-position selector	0.01-5	10	[22]
(microfluidic)	anide	valve, and PMT			
system					
Lab-on-chip	Luminol/H ₂ O ₂ /ferricy	Glass chip, peristaltic pump, and Biophysics CL analyzer	1.1-110	100	[23]

(microfluidic)	anide	with PMT detector
system		
Lab-on-chip	Luminol/H2O2/HRP	PMMA chip, two syringe pumps, microdialysis probe, and 0.8-10 100 [24]
(microfluidic)		Biophysics CL analyzer with PMT detector
system		
Lab-on-chip	Luminol/H2O2/HRP	PDMS/glass chip, two microdialysis syringe pumps, two- 2-10 230 [25]
(microfluidic)		position actuator switching valve, silicon photodiode,
system		multifunctional transimpedance amplifier, and multimeter
Lab-on-chip	Rhodanine	Paper chip fabricated by stacking one layer of assembled 0.42-50 140 [26]
(microfluidic)	derivative/H ₂ O ₂	WCP#1s between two layers of water-impermeable
system		single-sided adhesive tape, ultraweak luminescence
		analyzer with PMT detector
Lab-on-chip	Luminol/H ₂ O ₂ /ferricy	Paper chip fabricated by the photolithographical ~0.3-1.2 ~300 [27]
(microfluidic)	anide	patterning or wax-printing technique
system		

Lab-on-chip	Luminol/H ₂ O ₂ /HRP	Wax-patterned cloth, and CCD camera	0.1-100	94.8	[28]
(microfluidic)					
system					
Lab-on-chip	Luminol/H ₂ O ₂	Wax-patterned cloth, and CCD camera	0.01-10	9.74	This work
(microfluidic)					
system					

Note: ^a: the catalyst is not available; CL: chemiluminescence; LDR: linear dynamic range; LOD: limit of detection; Luminol: 3-aminophthalhydrazide; PMT: photomultiplier tube; HRP-AuNPs: horseradish peroxidase (HRP) absorbed on the surface of gold nanoparticles; ferricyanide: potassium hexacyanoferrate(III); FRP: fungal peroxidase; GO: graphene oxide; HRP-MNPs: HRP immobilized on magnetic nanoparticles; PDMS: poly(dimethyl-siloxane); PMMA: poly(methyl methacrylate); WCP#1: whatman chromatography paper #1; and CCD: charge-coupled device.



Fig. S1. The influence of cloth colors on the CL intensity. Here, the similar cloths had five different colors: white, blue, red, pink, and brown. And, the experimental conditions were as follows: pH of test/substrate solutions-7.5/8.0, [Luminol]-1.0 mM, [HRP]-0.3 g L⁻¹, [PIP]-0.2 mM, [H₂O₂]-0, 0.05 and 0.5 mM, Angle of inclination-50°, and Volumes of test/substrate solutions-30 μ L/35 μ L. The error bars represent the standard deviations of five independent measurements.



Fig. S2. Photos of four kinds of white cloths with different components. (**A**) white plain weave cotton cloth ("Cotton cloth"); (**B**) white linen-silk blended cloth ("Linen-silk cloth"); (**C**) white linen cloth ("Linen cloth"); and (**D**) white corduroy weave cotton cloth ("Corduroy cloth").



Fig. S3. Effect of white cloths with different components on the CL intensity. Here, the used cloth materials included white plain weave cotton cloth ("Cotton cloth"), white linen-silk blended cloth ("Linen-silk cloth"), white linen cloth ("Linen cloth"), and white corduroy weave cotton cloth ("Corduroy cloth"). And, the experimental conditions were as follows: pH of test/substrate solutions-7.5/8.0, [Luminol]-1.0 mM, [HRP]-0.3 g L⁻¹, [PIP]-0.2 mM, [H₂O₂]-0, 0.05 and 0.5 mM, Angle of inclination-50°, and Volumes of test/substrate solutions-30 μ L/35 μ L. The error bars represent the standard deviations of five independent measurements.



Fig. S4. Proof-of-concept demonstration of simultaneous detection within two different CL units on a single cloth piece. (**A**) The typical CL images: (i) the diagram of the device; (ii) the "a" detection zone was loaded with the test solution without H_2O_2 , while the "b" detection zone was loaded with the H_2O_2 -containing test solution; (iii) the "a" detection zone was loaded with the H_2O_2 -containing test solution, while the "b" detection zone was loaded with the test solution without H_2O_2 ; and (iv) the "a" and "b" detection zones were loaded with the H_2O_2 -containing test solution, respectively. In panels (ii), (iii) and (iv), the experimental conditions were as follows: pH of test/substrate solutions-7.5/8.0, [Luminol]-1.0 mM, [HRP]-0.3 g L⁻¹, [PIP]-0.2 mM, [H_2O_2]-0.05 mM, Angle of inclination-50°, and Volumes of test/substrate solutions-30 µL/35 µL. (**B**) CL intensities within two detection zones according to the images in panels (ii), (iii) and (iv). The error bars represent the standard deviations of five independent measurements.



Fig. S5. CL determination of H_2O_2 using the similar paper-based devices. The experimental conditions were as follows: pH of test/substrate solutions-7.5/8.0, [luminol]-1.0 mM, [HRP]-0.3 g L⁻¹, [PIP]-0.2 mM, [H₂O₂]-0, 0.05, 0.1, 0.5, 1, 5 and 10 mM, Angle of inclination-50°, and Volumes of test/substrate solutions-30 µL/35 µL. The insert showed that the CL intensities could be linearly proportional to the logarithms of H_2O_2 concentrations in the range of 0.05-10 mM. The error bar represents the standard deviation of five independent measurements.

References

- O. D. Leite, O. Fatibello, H. J. Vieira, F. R. P. Rocha and N. S. de Miranda Cury, *Anal. Lett.*, 2007, 40, 3148-3157.
- 2 F. Pan, P. K. Wei, M. L. Zhang and C. Lu, Anal. Methods, 2015, 7, 5667-5673.
- 3 W. Wang and H. Cui, J. Phys. Chem. C, 2008, 112, 10759-10766.
- J. Y. Zhou, J. J. Gu, C. X. Tian, D. C. Jiang, Y. Chen and K. Xi, *RSC Adv.*, 2016, 6, 39480-39483.
- 5 E. Warm and G. G. Laties, *Phytochemistry*, 1982, **21**, 827-831.
- 6 M. C. Ramos, M. C. Torijas and A. N. Díaz, Sens. Actuators B, 2001, 73, 71-75.
- 7 A. N. Díaz, M. C. R. Peinado and M. C. T. Minguez, Anal. Chim. Acta, 1998, 363, 221-227.
- X. H. Wang, O. Hofmann, R. Das, E. M. Barrett, A. J. deMello, J. C. deMello and D. D. C.
 Bradley, *Lab Chip*, 2007, 7, 58-63.
- 9 X. Y. Zeng, K. D. Zhang, J. Pan, G. P. Chen, A. Q. Liu, S. K. Fan and J. Zhou, *Lab Chip*, 2013, 13, 2714-2720.
- 10 A. M. Jorgensen, K. B. Mogensen, J. P. Kutter and O. Geschke, *Sens. Actuators B*, 2003, **90**, 15-21.
- O. Hofmann, P. Miller, P. Sullivan, T. S. Jones, J. C. deMello, D. D. C. Bradley and A. J. deMello, *Sens. Actuators B*, 2005, **106**, 878-884.
- 12 W. R. Guan, C. S. Zhang, F. F. Liu and M. Liu, Biosens. Bioelectron., 2015, 72, 114-120.
- 13 P. Panoutsou and A. Economou, *Talanta*, 2005, **67**, 603-609.
- 14 D. Lan, B. X. Li and Z. J. Zhang, Biosens. Bioelectron., 2008, 24, 934-938.

- 15 Q. Fang, X. T. Shi, Y. Q. Sun and Z. L. Fang, Anal. Chem., 1997, 69, 3570-3577.
- 16 D. Janasek and U. Spohn, Sens. Actuators B, 1997, 38-39, 291-294.
- 17 X. Z. Liu and E. H. Hansen, Anal. Chim. Acta, 1996, 326, 1-12.
- 18 M. J. Hao, N. Liu and Z. F. Ma, *Analyst*, 2013, **138**, 4393-4397.
- 19 B. X. Li, Z. J. Zhang and Y. Jin, Anal. Chim. Acta, 2001, 432, 95-100.
- 20 Y. X. Li, L. D. Zhu and G. Y. Zhu, Chem. Res. Chinese U., 2002, 18, 12-15.
- 21 Y. Zheng, S. L. Zhao and Y. M. Liu, Analyst, 2011, 136, 2890-2892.
- 22 Z. R. Xu and Z. L. Fang, Anal. Chim. Acta, 2004, 507, 129-135.
- 23 Y. Lv, Z. J. Zhang and F. N. Chen, Talanta, 2003, 59, 571-576.
- 24 W. Liu, Z. J. Zhang and H. S. Liu, Anal. Sci., 2005, 21, 413-416.
- 25 B. U. Moon, M. G. de Vries, B. H. C. Westerink and E. Verpoorte, *Sci. China Chem.*, 2012, 55, 515-523.
- 26 J. H. Yu, L. Ge, J. D. Huang, S. M. Wang and S. G. Ge, Lab Chip, 2011, 11, 1286-1291.
- 27 H. Jang and H. Noh, *Macromol. Res.*, 2015, 23, 493-495.
- 28 H. J. Li, C. L. Liu, D. Wang and C. S. Zhang, Biosens. Bioelectron., 2017, 91, 268-275.