

Article

Laser-Induced Graphene-Based Wearable Epidermal Ion-Selective Sensors for Noninvasive Multiplexed Sweat Analysis

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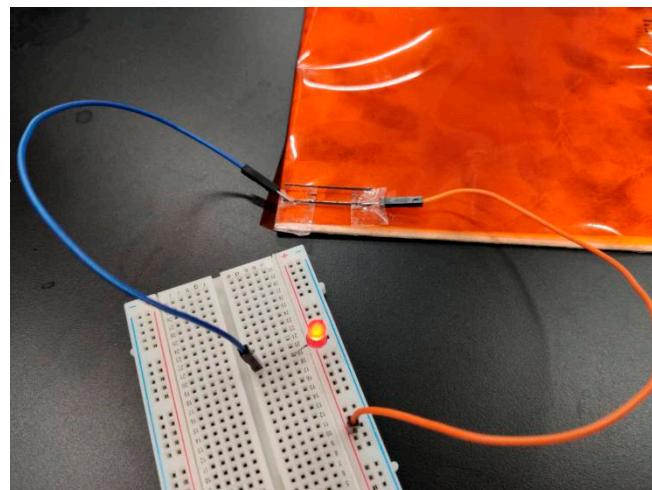


Figure S1. A LED bulb could be readily lighted through a LIG conductive wire, indicating good electrical conductivity of LIG.

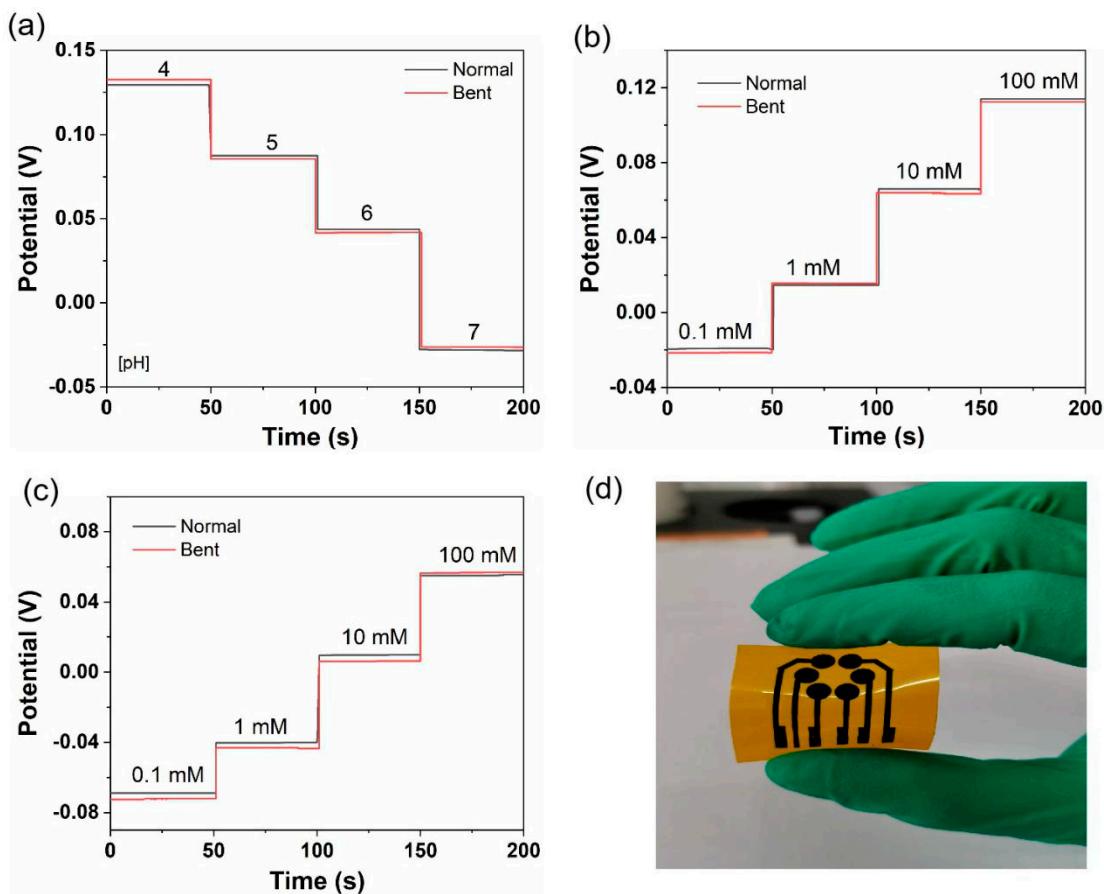


Figure S2. (a) pH, (b) Na^+ , and (c) K^+ sensing performance of flexible LIG-based sensors under normal and bent states.

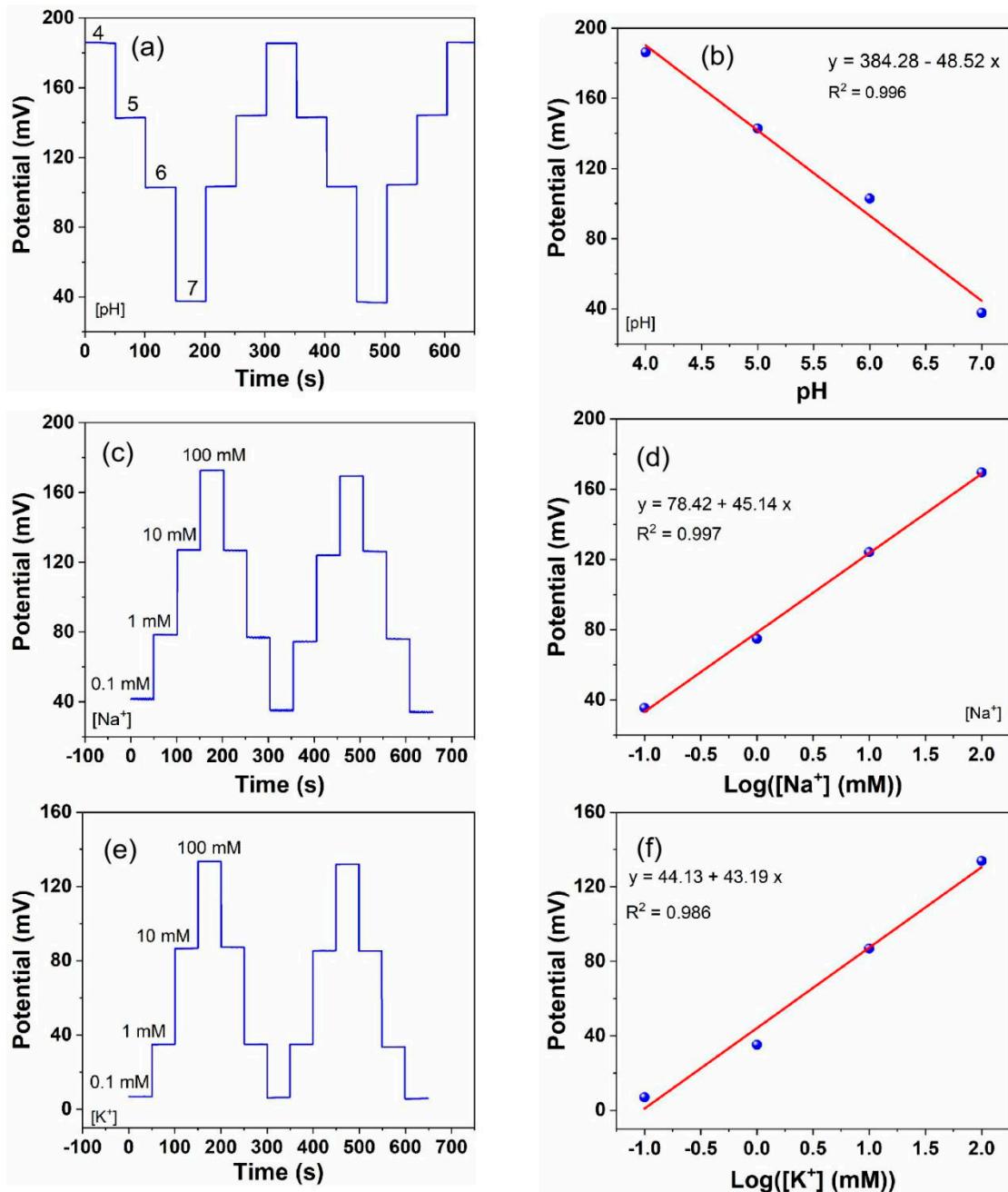
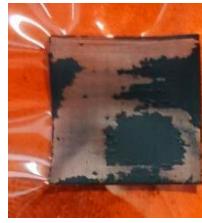
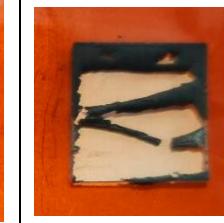


Figure S3. (a,b) pH, (c,d) Na⁺, and (e,f) K⁺ sensing performance of flexible LIG-based sensors tested on the custom-developed wearable multiplexed sensing system.

Table S1. The patterning of 2×2 cm squares on PI substrate was tested with different laser speed and power values.

Laser scribed graphene optimiza- tion		Max. Laser Power (12W)				
		50% (6W)	55% (6.6W)	60% (7.2W)	65% (7.8W)	70% (8.4W)
Laser speed (20 IPS)	10%					
	15%					

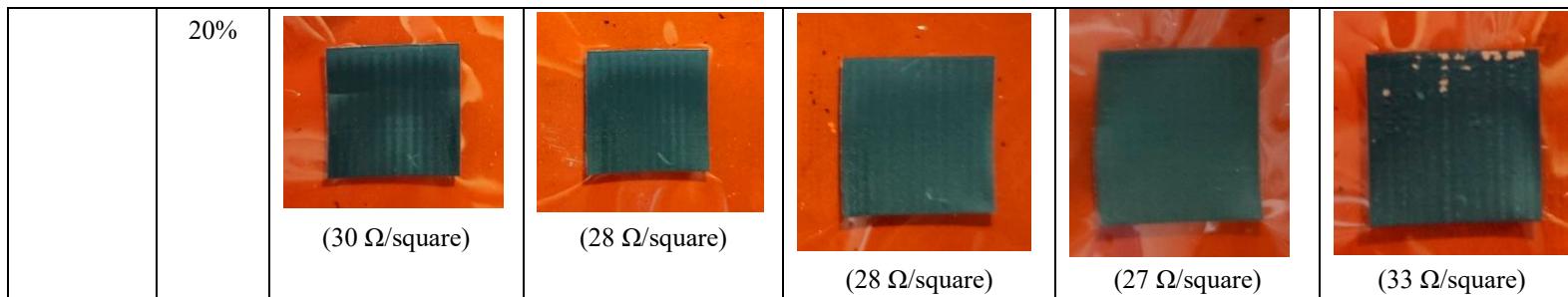


Table S2. Comparison of the sensing performances of the recently reported sweat sensors with this work.

Year/Reference	Detection method	Sensitivity	Linear range	Skin wearability	Integration level	Communication mode
2021 [1]	OCP	pH: 62.5 mV/decade	pH: 4-8	Yes	No	No
		Na ⁺ : 64.4 mV/decade	Na ⁺ : 10-160 mM			
		K ⁺ : 65.5 mV/decade	K ⁺ : 2-32 mM			
2018 [2]	OCP	pH: --	pH: 4-7	Yes	Yes	Bluetooth
		Na ⁺ : 45.8 mV/decade	Na ⁺ : 10-160 mM			
		K ⁺ : 35.9 mV/decade	K ⁺ : 2-32 mM			
2021 [3]	OCP	Na ⁺ : 50.3 mV/decade	Na ⁺ : 0.1-10 mM	NO	NO	NO

		K^+ : 53.5 mV/decade	K^+ : 0.1-10 mM				
2018 [4]	OCP	pH: 71.44 mV/decade	pH: 4-8	Yes	Yes	Bluetooth	
				<input checked="" type="checkbox"/> Circuit			
					<input checked="" type="checkbox"/> Smartphone		
2014 [5]	OCP	Na^+ : 63.75 mV/decade	Na^+ : 0.1-100mM	Yes	Yes	Bluetooth	
				<input checked="" type="checkbox"/> Circuit			
					<input checked="" type="checkbox"/> PC		
2016 [6]	OCP	Na^+ : 64.2 mV/decade	Na^+ : 10-160mM	Yes	Yes	Bluetooth	
		K^+ : 61.3 mV/decade	K^+ : 1-32mM		<input checked="" type="checkbox"/> Circuit		
					<input checked="" type="checkbox"/> Smartphone		
2020 [7]	OCP	Na^+ : 56.4 mV/decade	Na^+ : 0.0001-10mM	Yes	Yes	Bluetooth	
		K^+ : 54.3 mV/decade	K^+ : 0.0001-10mM		<input checked="" type="checkbox"/> Circuit		
					<input checked="" type="checkbox"/> Laptop		
2021 [8]	OCP	pH: 79 mV/decade	pH: 3-8	NO	Yes	NO	
2021 [9]	OCP	Na^+ : 55.5 mV/decade	Na^+ : 1-1000mM	Yes	Yes	Bluetooth	

This work	OCP	pH: 51.5 mV/decade Na⁺: 45.4 mV/decade K⁺: 43.3 mV/decade	pH: 4-7 Na⁺: 0.1-100mM K⁺: 0.1-100mM	Yes	Yes	Bluetooth
					<input checked="" type="checkbox"/> Circuit	<input checked="" type="checkbox"/> Smartphone

References

- [1] B. Paul, S. Demuru, C. Lafaye, M. Saubade, D. Briand, Printed Iontophoretic-Integrated Wearable Microfluidic Sweat-Sensing Patch for On-Demand Point-Of-Care Sweat Analysis, *Adv. Mater. Technol.* 2021, 2000910.
- [2] L. Wang, L. Wang, Y. Zhang, J. Pan, S. Li, X. Sun, et al., Weaving Sensing Fibers into Electrochemical Fabric for Real-Time Health Monitoring, *Adv. Funct. Mater.* 2018, 28, 1804456.
- [3] F. Wang, Y. Liu, M. Zhang, F. Zhang, P. He, Home Detection Technique for Na^+ and K^+ in Urine Using a Self-Calibrated all-Solid-State Ion-Selective Electrode Array Based on Polystyrene-Au Ion-Sensing Nano-composites, *Anal. Chem.* 2021, 93, 8318-8325.
- [4] S.Y. Oh, S.Y. Hong, Y.R. Jeong, J. Yun, H. Park, S.W. Jin, et al., Skin-Attachable, Stretchable Electrochemical Sweat Sensor for Glucose and pH Detection, *ACS Appl. Mater. Interfaces* 2018, 10, 13729-13740.
- [5] A.J. Bandodkar, D. Molinnus, O. Mirza, T. Guinovart, J.R. Windmiller, G. Valdés-Ramírez, et al., Epidermal tattoo potentiometric sodium sensors with wireless signal transduction for continuous non-invasive sweat monitoring, *Biosens. Bioelectron.* 2014, 54, 603-609.
- [6] W. Gao, S. Emaminejad, H.Y.Y. Nyein, S. Challa, K. Chen, A. Peck, et al., Fully integrated wearable sensor arrays for multiplexed *in situ* perspiration analysis, *Nature* 2016, 529, 509-514.
- [7] P. Pirovano, M. Dorrian, A. Shinde, A. Donohoe, A.J. Brady, N.M. Moyna, et al., A wearable sensor for the detection of sodium and potassium in human sweat during exercise, *Talanta* 2020, 219, 121145.
- [8] V. Mazzaracchio, L. Fiore, S. Nappi, G. Marrocco, F. Arduini, Medium-distance affordable, flexible and wireless epidermal sensor for pH monitoring in sweat, *Talanta* 2021, 222, 121502.
- [9] H.-R. Lim, Y. Lee, K.A. Jones, Y.-T. Kwon, S. Kwon, M. Mahmood, et al., All-in-one, wireless, fully flexible sodium sensor system with integrated Au/CNT/Au nanocomposites, *Sens. Actuators, B* 2021, 331, 129416.