Antibacterial sensitive Wearable Biosensor Enabled by Engineered-Metal-Boride-Based Organic Electrochemical Transistor and Hydrogel Microneedle

Ru Zhang[a,*], Md Sohel Rana[a], Lin Huang[b,c,*], Kun Qian[a,*]

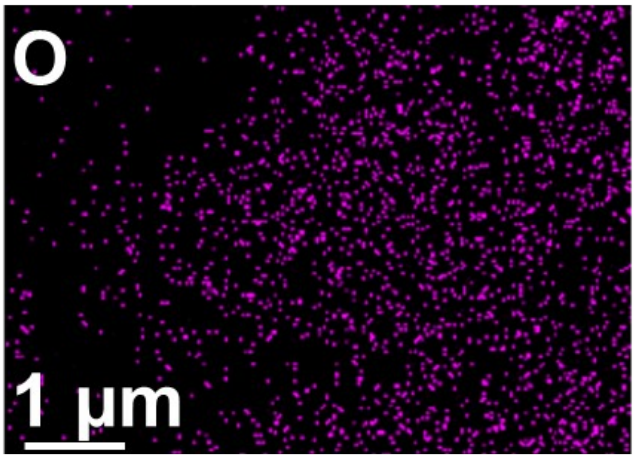
[a] School of Biomedical Engineering, Shanghai Jiao Tong University, Shanghai, 200030, China;

[b] Department of Clinical Laboratory Medicine, Shanghai Chest Hospital, Shanghai Jiao Tong University School of Medicine, Shanghai 200030, China;

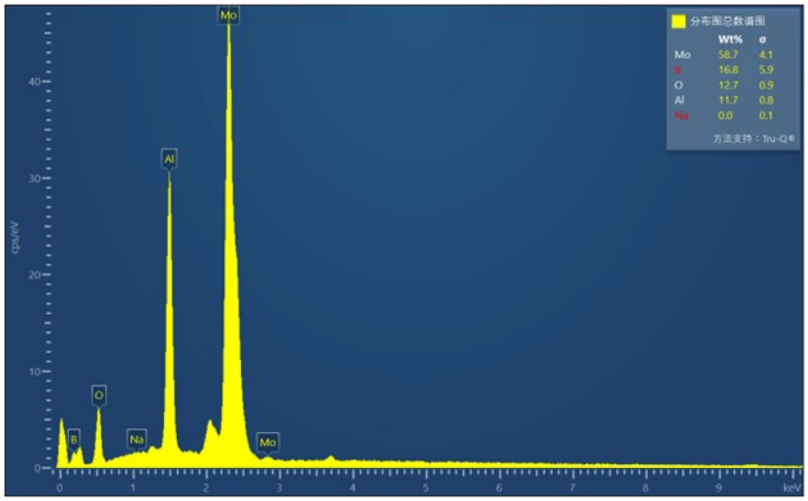
[c] Shanghai Institute of Thoracic Oncology, Shanghai Chest Hospital, Shanghai Jiao Tong University School of Medicine, Shanghai, 200030, China.

*Corresponding authors: ruzhang@sjtu.edu.cn; linhuang@shsmu.edu.cn; qian@sjtu.edu.cn

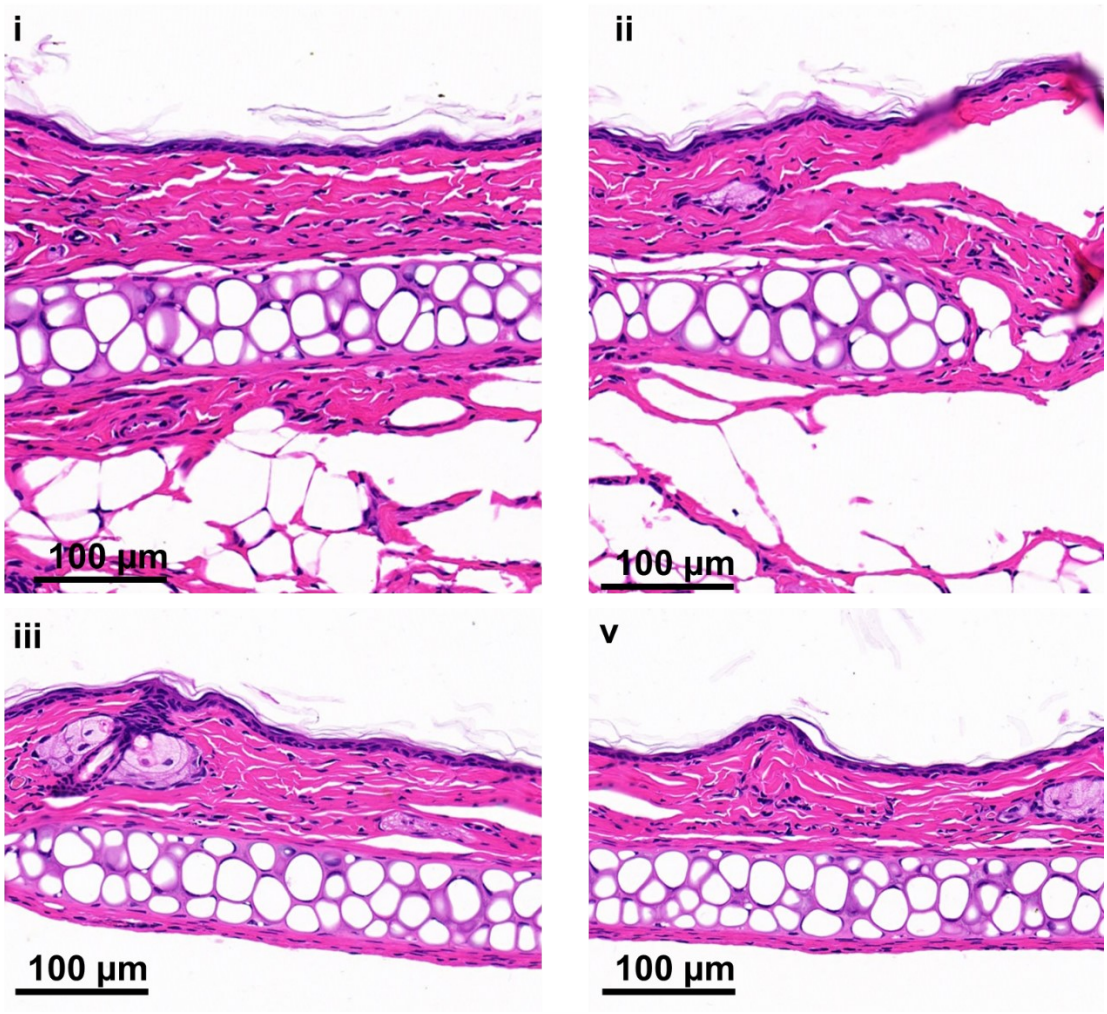
Keywords: hydrogel microneedle, antibacterial, wearing biosensor, two-dimensional-materials, organic electrochemical transistor



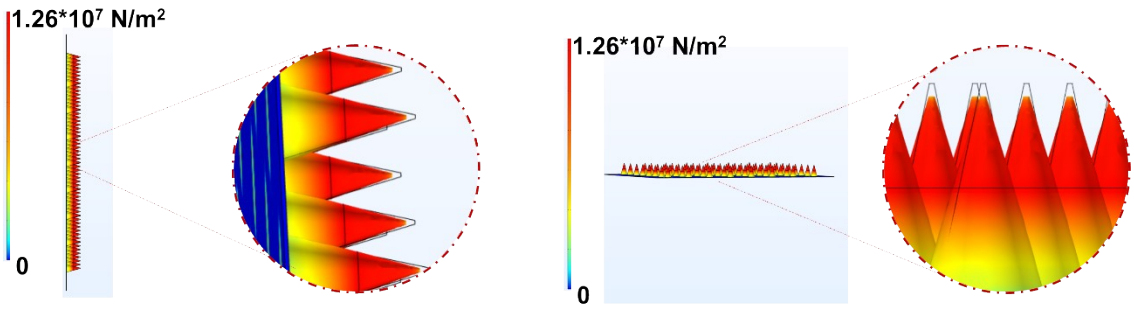
S1. Mapping analysis of O.



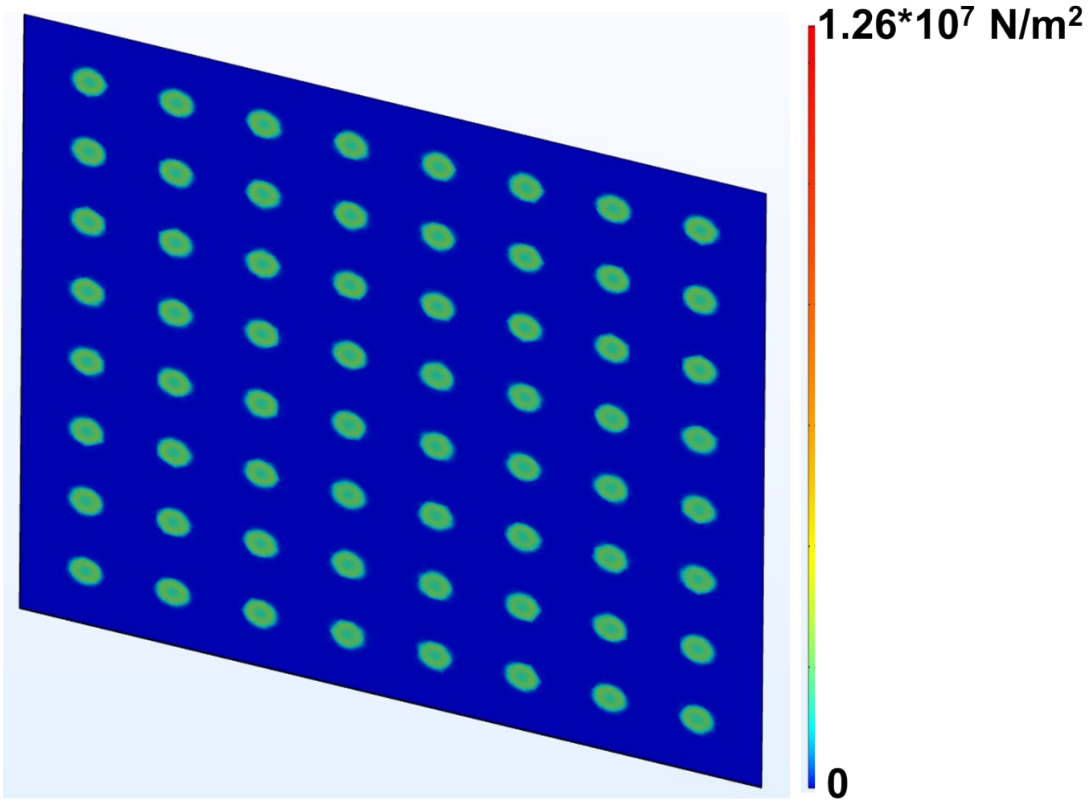
S2. Elemental distribution of MBene.



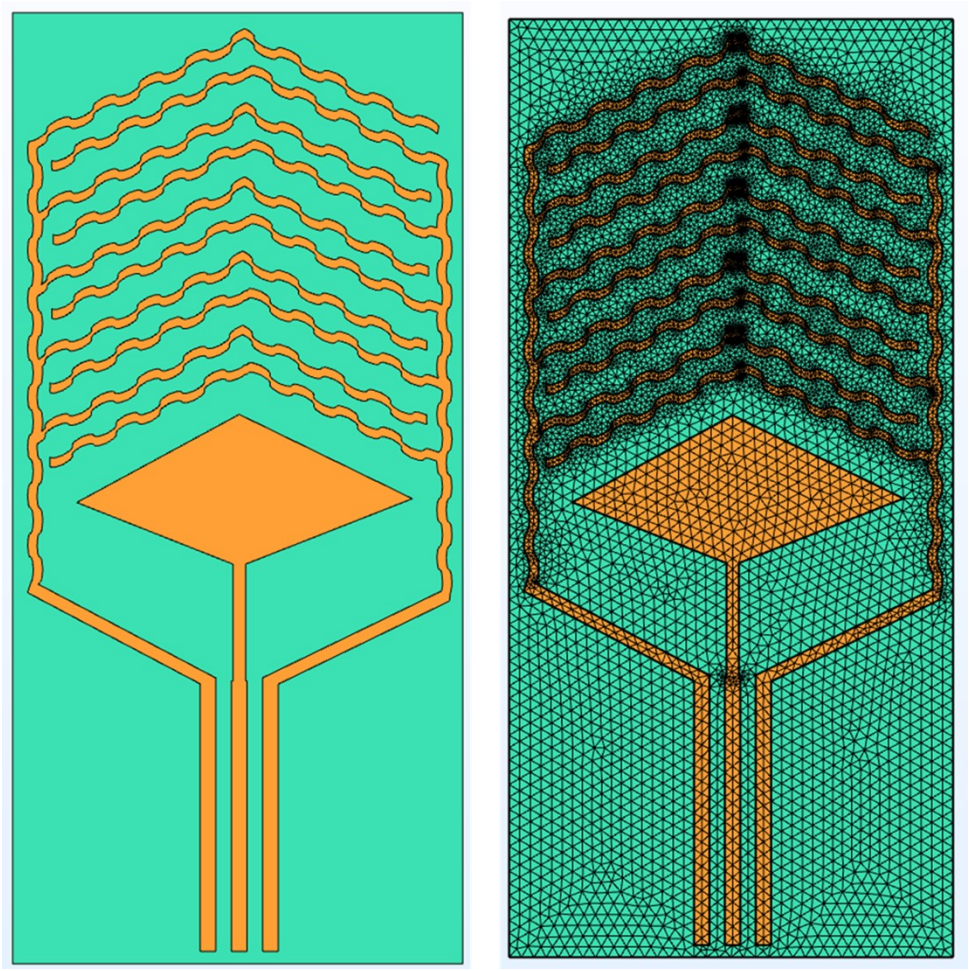
S3. Analysis of H&E staining of the skin at multiple sites punctured by hydrogel microneedles



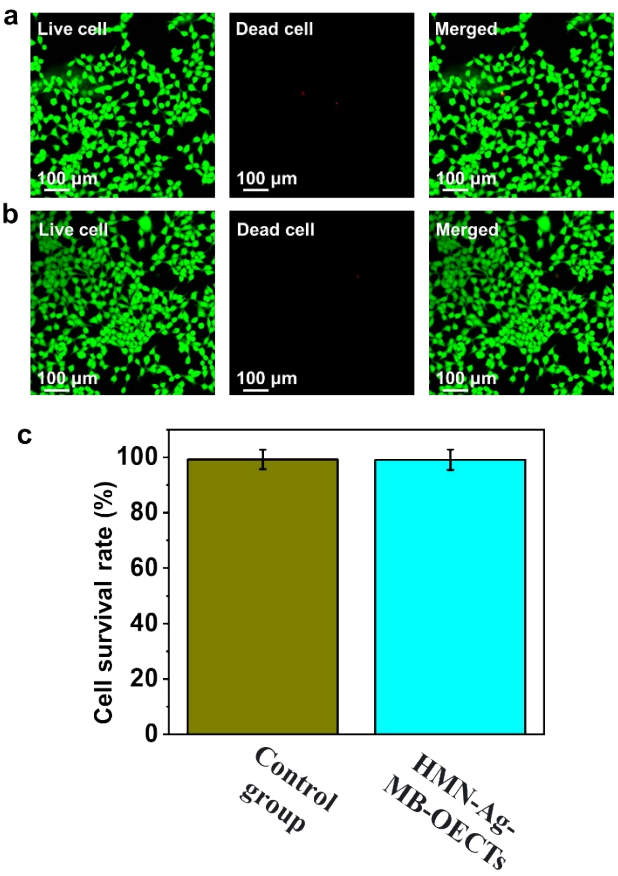
S4. COMSOL models other angles of the hydrogel microneedles.



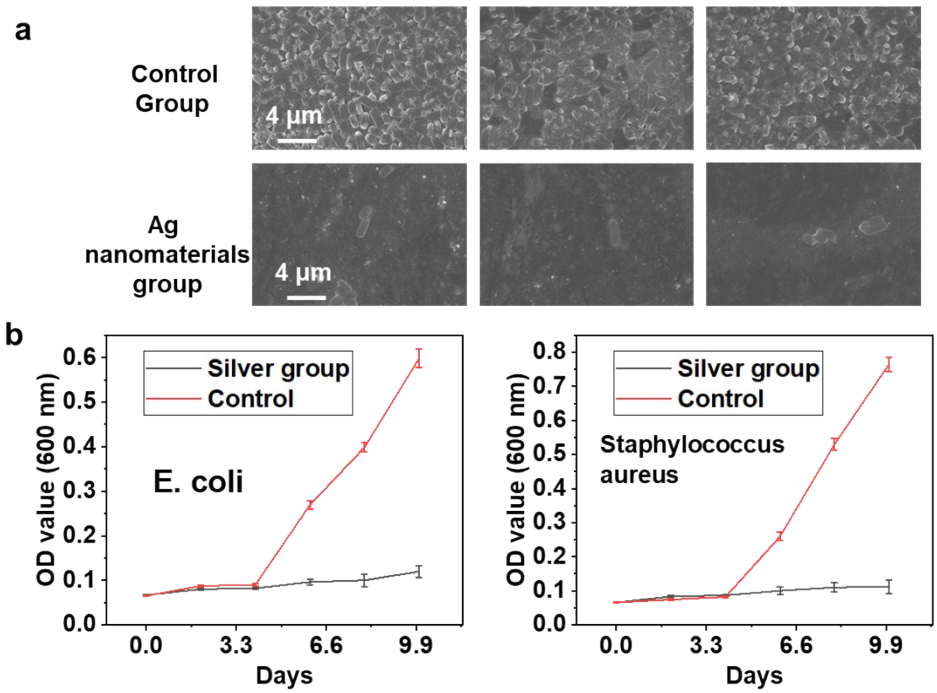
S5. Stress at the bottom of the hydrogel microneedle.



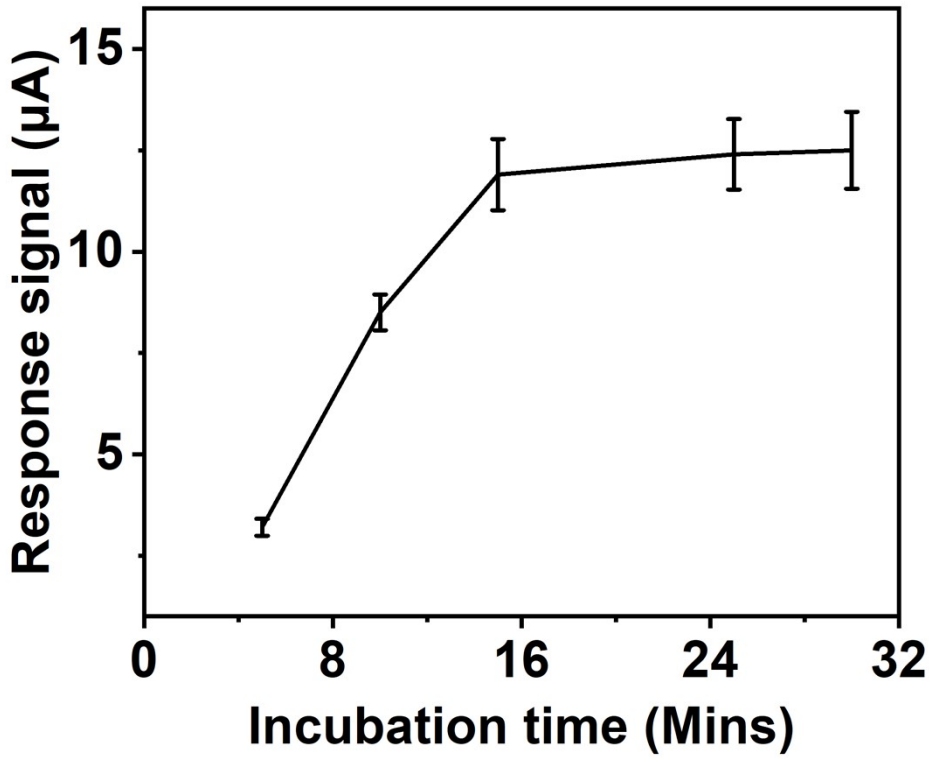
S6. Serpentine electrode's modeling structure.



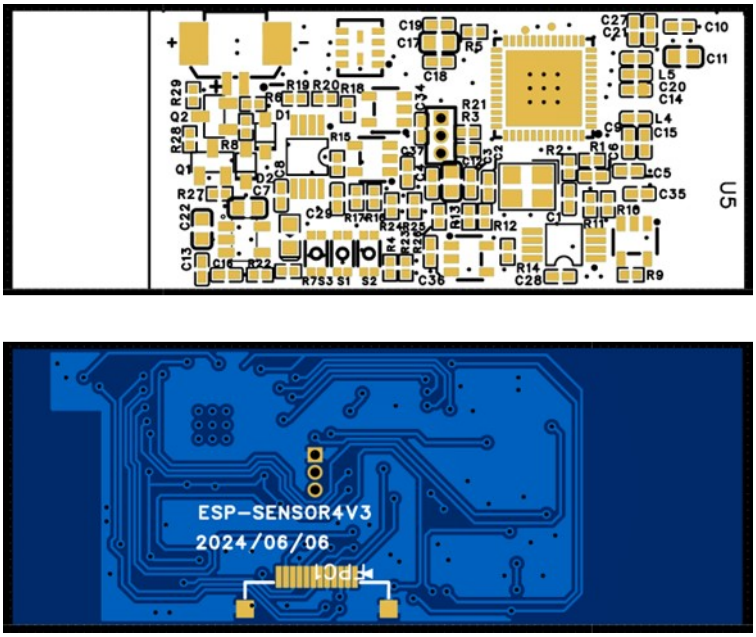
S7. a) Analysis of cellular fluorescence for the a) control group and b) the HMN-Ag-MB-OECTs. c) Corresponding analysis of the cell viability.



S8. a) SEM analysis images of Escherichia coli treated with and without silver nanomaterials; b) Analysis of the OD values of Escherichia coli and Staphylococcus aureus at different time points when treated with and without silver nanomaterials.



S9. Upon optimizing the incubation time of HMN-Ag-MB-OECTs, it was found that the device achieved sensitive and expeditious detection results when the incubation time was set at t = 15 mins.



S10. Back view of the flexible circuit board.

Table S1. Applications **of** PVA/PVP microneedle.

Number	Materials	Applications	References
1	PVA/PVP hydrogel microneedle patch	Extraction of interstitial skin fluid toward minimally invasive monitoring of blood glucose level	[1]
2	PVA/PVP dissolving microneedles	Transdermal drug delivery systems	[2]
3	PVA/PVP microneedle	enhanced transdermal delivery	[3]
4	PVA/PVP microneedle	Wearable device	This work

Table S2. Comparison of literature on cortisol testing.

Number	Materials	Strategies	Detection range (ng/mL)	Detection of limit (ng/mL)	References
1	PEDOT: PSS	OECTs	0.36 to 362.5	0.036	[4]
2	AuNPs	Electrochemical manner	0.073 to 0.36	/	[5]
3	L-cys/AuNPs/MXene	Amperometry	5-180	/	[6]
4	Au/PDMS	EIS	15-150	5	[7]
5	MBene/AgNWs/AgNPs	OECTs	0.001-1000	0.00026	This work

References

[1] N. Xu, M. Zhang, W. Xu, G. Ling, J. Yu, P. Zhang, Swellable PVA/PVP hydrogel microneedle patches for the extraction of interstitial skin fluid toward minimally invasive monitoring of blood glucose level, Analyst 147 (2022) 1478–1491. https://doi.org/10.1039/D1AN02288A.

[2] V. Ebrahiminejad, A. Malek-khatabi, Z. Faraji Rad, Influence of Low-Frequency Vibration and Skin Strain on Insertion Mechanics and Drug Diffusion of PVA/PVP Dissolving Microneedles, Adv. Mater. Technol. 9 (2024) 202301272. https://doi.org/10.1002/admt.202301272.

[3] R. Ghanma, Q.K. Anjani, Y.A. Naser, A.H. Bin Sabri, A.R.J. Hutton, L.K. Vora, A. Himawan, B. Greer, H.O. McCarthy, R.F. Donnelly, Risperidone-cyclodextrin complex reservoir combined with hydrogel-forming microneedle array patches for enhanced transdermal delivery, Eur. J. Pharm. Biopharm. 202 (2024) 114415. https://doi.org/10.1016/j.ejpb.2024.114415.

[4] S. Demuru, J. Kim, M. El Chazli, S. Bruce, M. Dupertuis, P.A. Binz, M. Saubade, C. Lafaye, D. Briand, Antibody-Coated Wearable Organic Electrochemical Transistors for Cortisol Detection in Human Sweat, ACS Sensors 7 (2022) 2721–2731. https://doi.org/10.1021/acssensors.2c01250.

[5] A.R. Naik, Y. Zhou, A.A. Dey, D.L.G. Arellano, U. Okoroanyanwu, E.B. Secor, M.C. Hersam, J. Morse, J.P. Rothstein, K.R. Carter, J.J. Watkins, Printed microfluidic sweat sensing platform for cortisol and glucose detection, Lab Chip 22 (2022) 156–169. https://doi.org/10.1039/d1lc00633a.

[6] T. Laochai, J. Yukird, N. Promphet, J. Qin, O. Chailapakul, N. Rodthongkum, Non-invasive electrochemical immunosensor for sweat cortisol based on L-cys/AuNPs/ MXene modified thread electrode, Biosens. Bioelectron. 203 (2022) 114039. https://doi.org/10.1016/j.bios.2022.114039.

[7] S. Reza Mahmoodi, P. Xie, D.P. Zachs, E.J. Peterson, R.S. Graham, C.R.W. Kaiser, H.H. Lim, M.G. Allen, M. Javanmard, Single-step label-free nanowell immunoassay accurately quantifies serum stress hormones within minutes, Sci. Adv. 7 (2021) abf4401. https://doi.org/10.1126/sciadv.abf4401.