Supporting Information

Layer-by-Layer Construction of In Situ Formed Polypyrrole and Bacterial Cells as Capacitive Bioanode for Paper-based Microbial Fuel Cell

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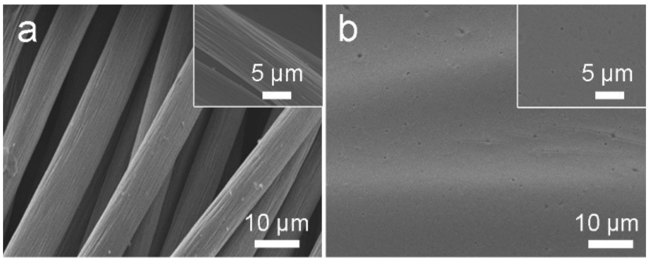

Fig. S1. SEM images of CC (a) and CC/PPy (b) electrodes.

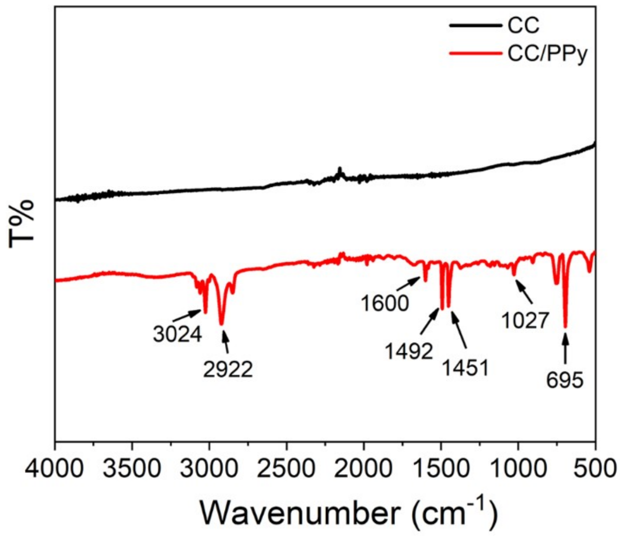

Fig. S2. FT-IR spectrum of CC and CC/PPy electrodes.

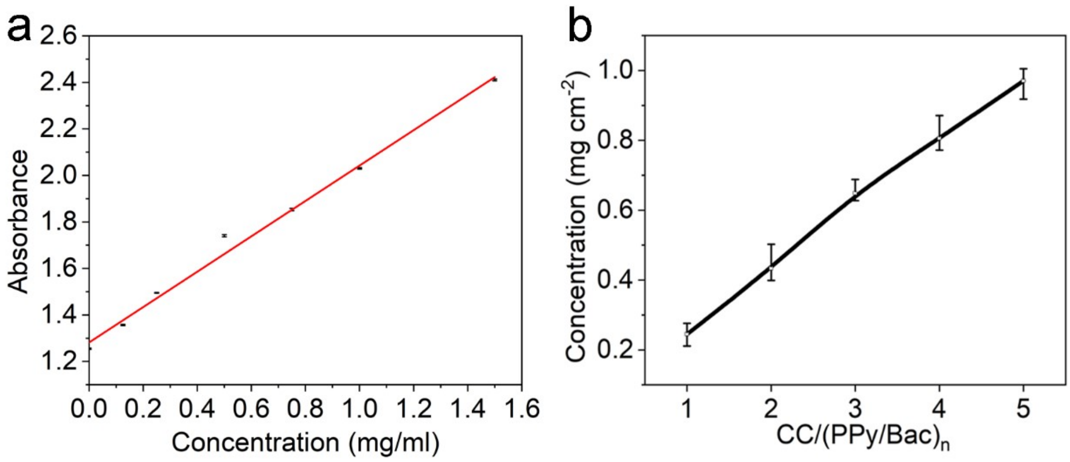

Fig. S3. a) Standard curve of the BCA protein assay (y = 0.7605 (±0.026) * x + 1.282 (± 0.017), R^2 = 0.9942). b) Protein amounts of CC/$(PPy/Bac)_n$ electrodes as a function of bilayer number.

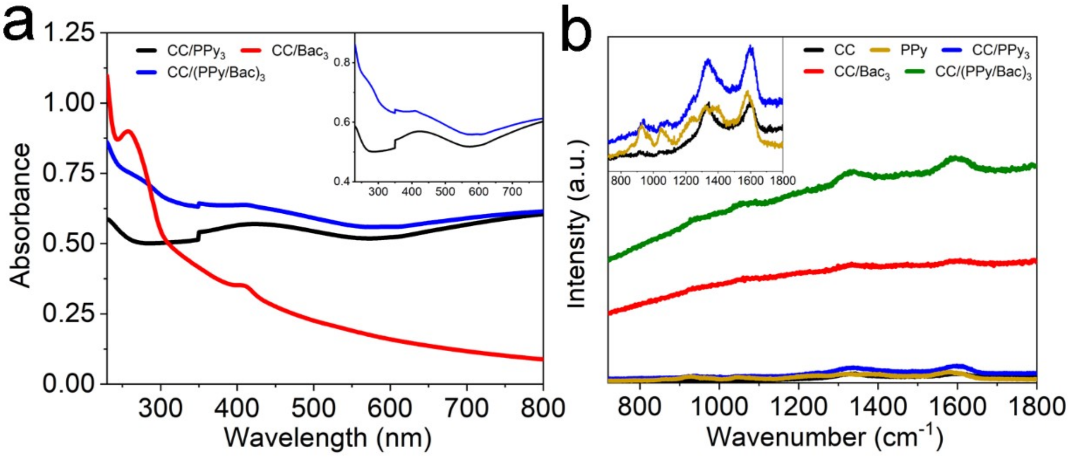

Fig. S4. UV-Vis spectra (a) and Raman (b) spectra of different electrodes. The samples on the electrodes were scraped and re-disperse in the solution for the UV-vis test, while the electrodes were directly used for Raman test.

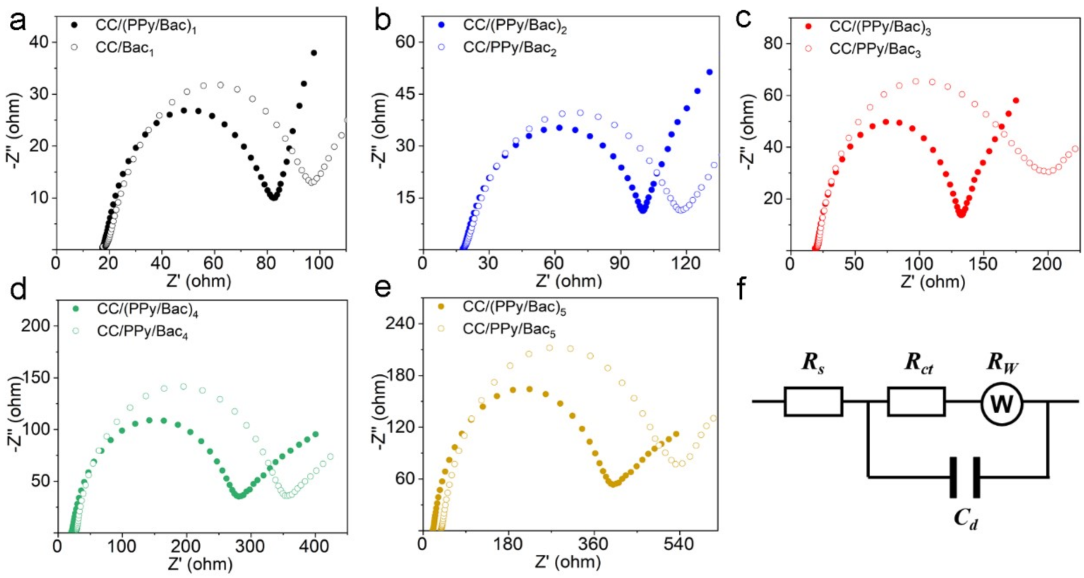

Fig. S5. (a-e) The Nyqusit curves of CC/(PPy/Bac)$_n$ electrodes and their corresponding control electrodes. (f) The equivalent circuit diagram of the Nyquist curves. The CC/Bac electrode was formed by the spin coating of bacterial layer onto the CC surface. The CC/PPy/Bac$_n$ electrodes were fabricated by the spin coating of different bacterial layers onto the surface of CC electrode with PPy film.

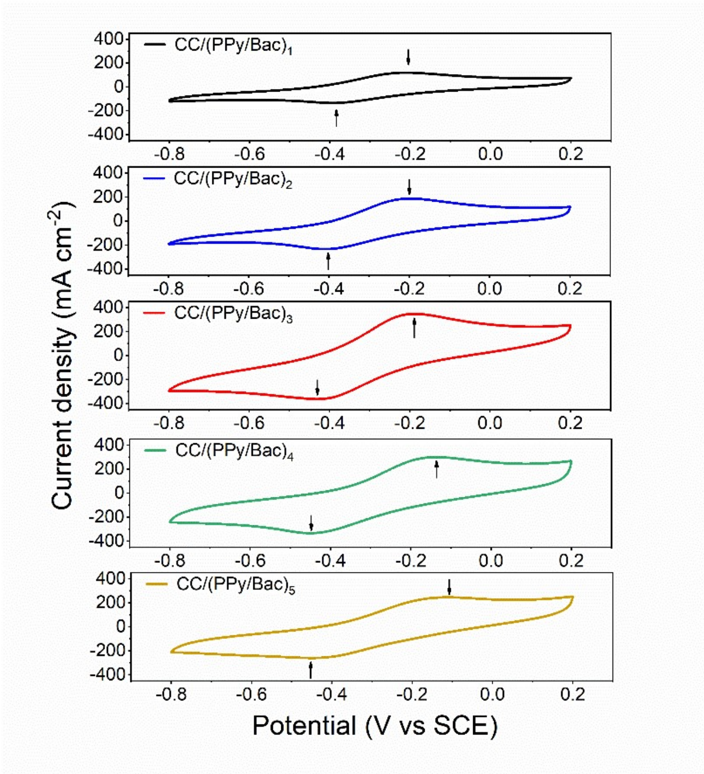

Fig. S6. CV curves of the CC/(PPy/Bac)$_n$ anodes (n = 1-5). The bigger redox potential difference indicates slower charge transfer.

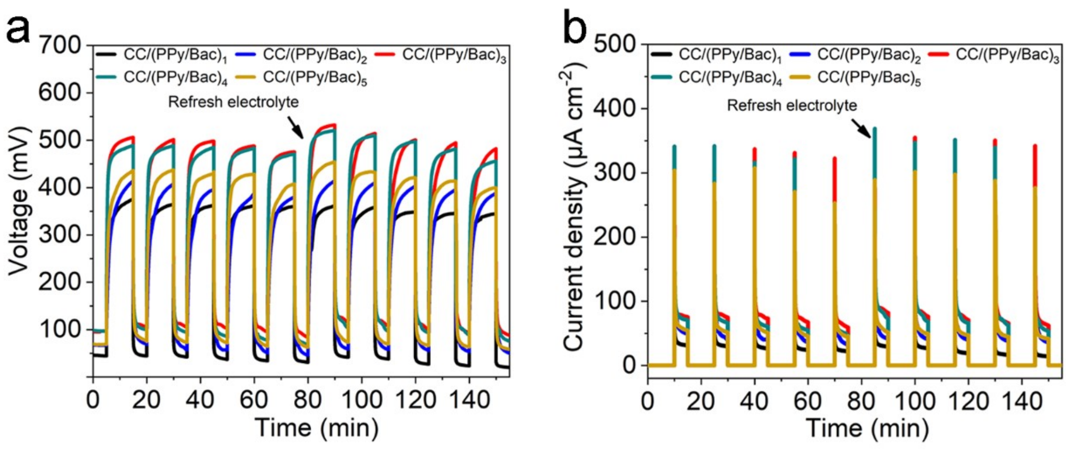

Fig. S7. Voltage (a) and current density (b) curves of SC-PBMFCs with different CC/(PPy/Bac)$_n$ anodes during ten of charge-discharge cycles (charging/discharging time, 10/5 mins for each cycle), when a load of 470 Ω was applied.

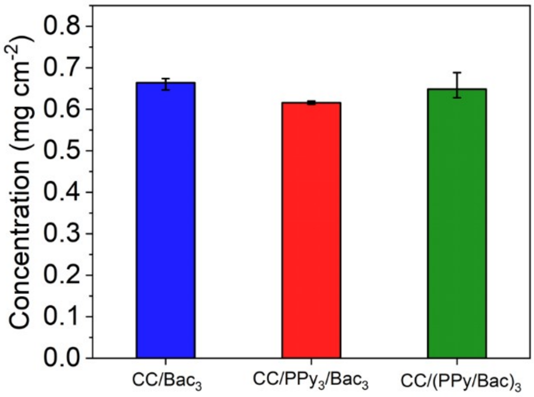

Fig. S8. Protein contents on the CC/Bac$_3$, CC/PPy/Bac$_3$ and CC/(PPy/Bac)$_3$ anodes.

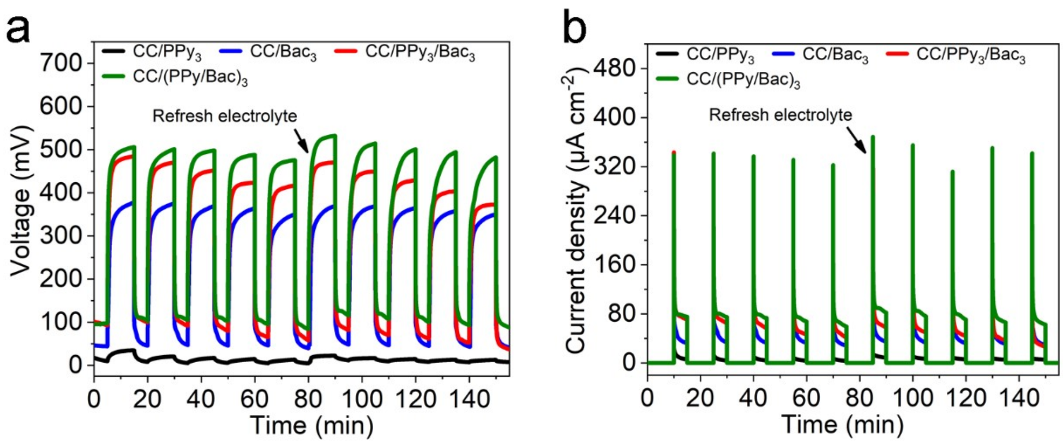

Fig. S9. Voltage (a) and current density (b) curves of SC-PBMFCs with CC/(PPy/Bac)$_3$ anode and control anodes during ten of charge-discharge cycles (charging/discharging time, 10/5 mins for each cycle), when a load of 470 Ω was applied.

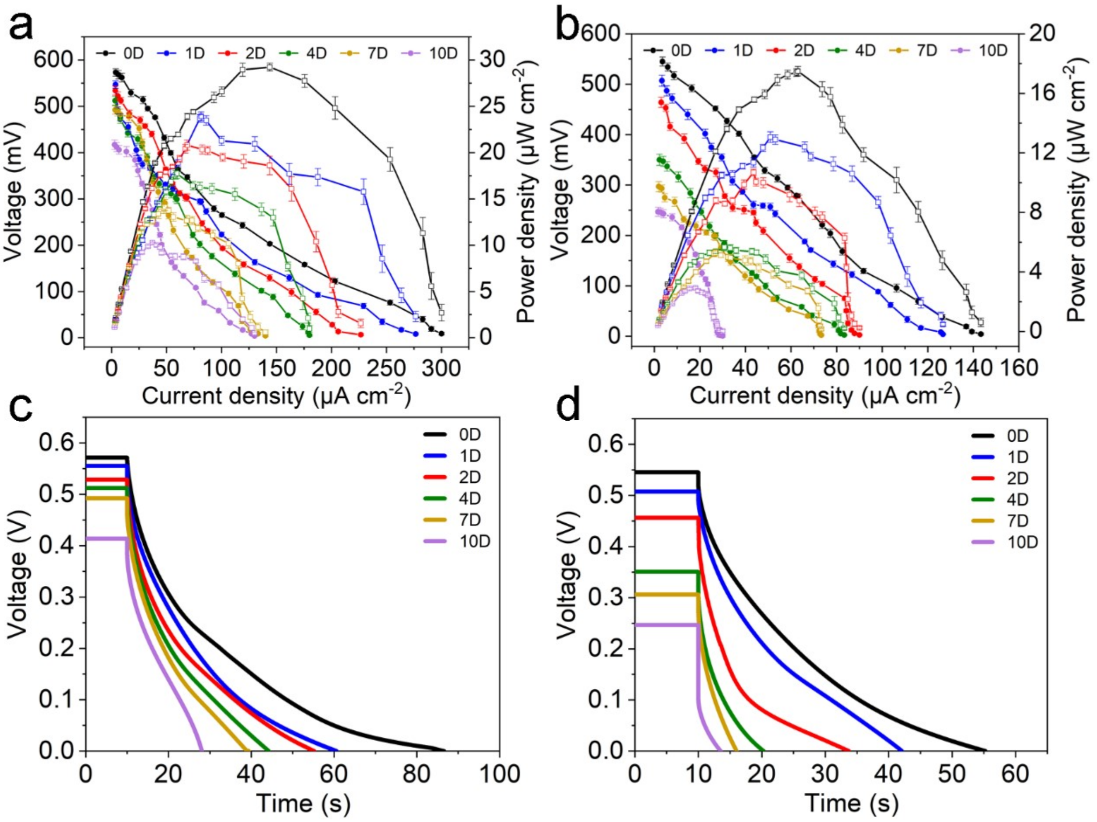

Fig. S10. The power density (a, b) and complete discharge (c, d) curves of SC-PBMFCs after the storage of CC/(PPy/Bac)$_3$ (a, c) and CC/PPy$_3$/Bac$_3$ (b, d) anodes for different times.

Table S1. A detailed comparison of PBMFCs performances with different anodes.

Elecrode Substartes	Anode Materials	Exoelectrogen Type	PMFC Type	Power Density	Ref.
Carbon Cloth	/	S.oneidensis	Cover type	5.5 $\mu W\ cm^{-2}$	1
Carbon Cloth	/	S.oneidensis	Cover type	10 $\mu W\ cm^{-2}$	2
Screen-printed Carbon	/	S.oneidensis MR-1	Origami type	9.3 $\mu W\ cm^{-2}$	3
Filter Paper	Graphite ink with AC	S.oneidensis MR-1	Origami type	0.14 $\mu W\ cm^{-2}$	4
Carbon Cloth	/	S.oneidensis MR-1	Origami type	1.7 $\mu W\ cm^{-2}$	5
Carbon Paper	activated carbon powder mixed with MWCNT	E. coli	Origami type	8.4 $\mu W\ cm^{-2}$	6
Activated Carbon Sheet	/	E. coli	Origami type	8.8 $\mu W\ cm^{-2}$	7

References

1. A. Fraiwan, S. Mukherjee, S. Sundermier, H. S. Lee and S. Choi, A paper-based microbial fuel cell: Instant battery for disposable diagnostic devices. *Biosens. Bioelectron.*, 2013, **49**, 410-414.

2. A. Fraiwan and S. Choi, Bacteria-powered battery on paper. *Phys. Chem. Chem. Phys.*, 2014, **16**, 26288-26293.

3. H. Lee and S. Choi, An origami paper-based bacteria-powered battery. *Nano Energy*, 2015, **15**, 549-557.

4. M. Mohammadifar, J. Zhang, I. Yazgan, O. Sadik and S. Choi, Power-on-paper: Origami-inspired fabrication of 3-D microbial fuel cells. *Renew. Energ.*, 2018, **118**,

695-700.
5. A. Fraiwan, L. Kwan and S. Choi, A disposable power source in resource-limited environments: A paper-based biobattery generating electricity from wastewater. *Biosens. Bioelectron.*, 2016, **85**, 190-197.
6. D. T. Nguyen and K. Taguchi, Optimization of electrodes in a disposable paper-based microbial fuel cell. *Ieej T. Electr. Electr.*, 2020, **15**, 311-312.
7. D. T. Nguyen and K. Taguchi, A disposable water-activated paper-based MFC using dry *E-coli* biofilm. *Biochem. Eng. J.*, 2019, **143**, 161-168.