

## Supporting Information

### **Carbon nanoparticles of Chinese ink-wrapped natural loofah sponge: a low-cost three-dimensional electrode for high-performance microbial energy harvesting**

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**Table S1 Stock suspension of 5 brands of CI used in this study**

Chinese ink <sup>o</sup>	Suppliers <sup>o</sup>
Zhujiang Ink <sup>o</sup>	Jinjian office supplies factory, Guangzhou, China <sup>o</sup>
Shuchang ink <sup>o</sup>	Shaoyang renfa ink factory, Hunan, China <sup>o</sup>
Red apple ink <sup>o</sup>	Linyi a pen stationery Co., Ltd, Shandong, China <sup>o</sup>
Song ink <sup>o</sup>	Sangzhi stationery factory, Hunan, China <sup>o</sup>
Xiangzi ink <sup>o</sup>	Changsha Dongfeng Ink Co., Ltd, Hunan, China <sup>o</sup>

**Table S2 Performances of various 3D electrodes with different skeletons and brands of CI**

Electrodes	Chinese ink	EIS ( $\Omega$ )		Conductivity (S/m)	j (mA/cm <sup>3</sup> )
		R <sub><math>\Omega</math></sub>	R <sub>ct</sub>		
CF	No	4.2	132.2	220.0 $\pm$ 6.0	4.0 $\pm$ 0.6
LSC	No	4.1	10.8	15.8 $\pm$ 1.0	18.6 $\pm$ 0.6
CI-SP	Zhujiang Ink	4.9	45.2	3.1 $\pm$ 0.3	5.6 $\pm$ 0.6
CI-LS-5	Zhujiang Ink	4.1	13.1	9.5 $\pm$ 0.8	16.3 $\pm$ 0.6
CI-LS-5	Shuchang Ink	4.2	42.8	4.3 $\pm$ 0.5	10.1 $\pm$ 0.4
CI-LS-5	Song Ink	4.0	61.2	1.8 $\pm$ 0.4	5.2 $\pm$ 0.3
CI-LS-5	Red apple Ink	3.9	102.8	0.8 $\pm$ 0.2	2.8 $\pm$ 0.4
CI-LS-5	Xiangzi Ink	4.0	127.5	0.3 $\pm$ 0.1	0.5 $\pm$ 0.1

**Table S3 Capital expenditure of various 3D electrodes**

Cost <sup>a</sup>	LSC <sup>b</sup>	CI-LS-5 <sup>c</sup>	Carbon Felt <sup>d</sup>	Graphene-Sponge <sup>e</sup>
Raw materials	--	\$ 1300		
Energy Consumption	\$ 4400	--		
Total	\$ 4400	\$ 1300	\$ 4200	\$ 2000

<sup>a</sup>: the cost was calculated for 1 m<sup>3</sup>.

<sup>b</sup>:The price of LSC is estimated as follows: The main cost of the LSC is assigned to the electricity for heat treatment. Our local electricity price is 0.11 US\$ for 1000 W/h and the power of the furnace used in this study is 4 kW. We can once maximally treat 200 pieces (1.0 × 1.0 × 0.5 cm). So for 1 hour heat treatment, a piece of LSC costs 0.0022 US\$, and the cost of LSC can be roughly calculated to be 4,400 UD \$/m<sup>3</sup>.

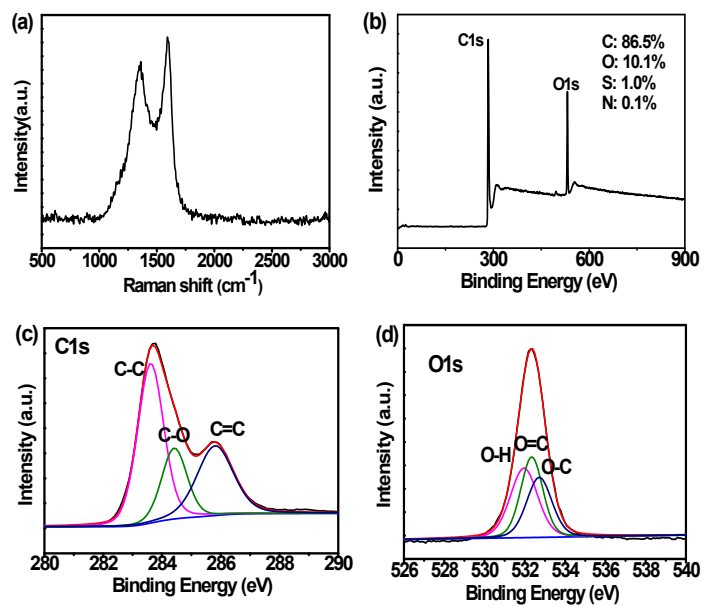
<sup>c</sup>:The price of CI-LS-5 is estimated as follows: The main cost of the CI-LS-5 is assigned to the CI. For instance, the price of Zhujiang ink is ca. 1.3 \$/L. To make one piece of CI-LS-5 (1.0 × 1.0 × 0.5 cm), 0.5 mL ink is consumed. The cost of CI-LS-5 can be roughly calculated to be 1,300 UD \$/m<sup>3</sup>.

<sup>d</sup>: Purchased from JinGu Carbon Material CO., LTD. (Liaoning, China).

<sup>e</sup>: Calculated by Xie et al. (*Energy Environ. Sci.* **2012**, 5, 6862–6866).

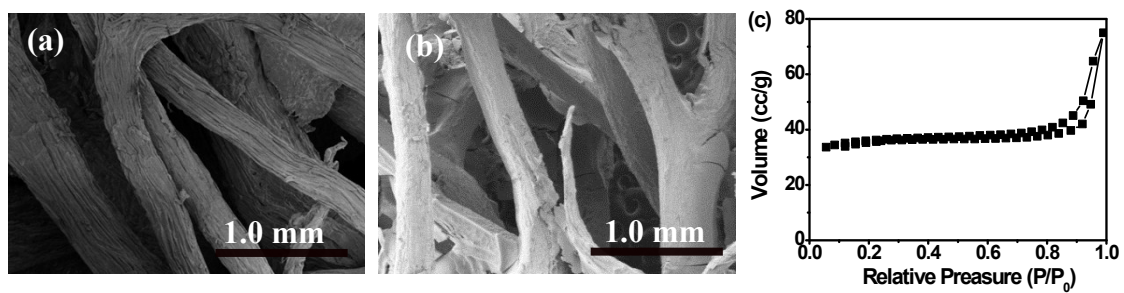
Note: the unit price used here is based on lab scale, which should be higher than unit price for industrial use.

**Figure S1**



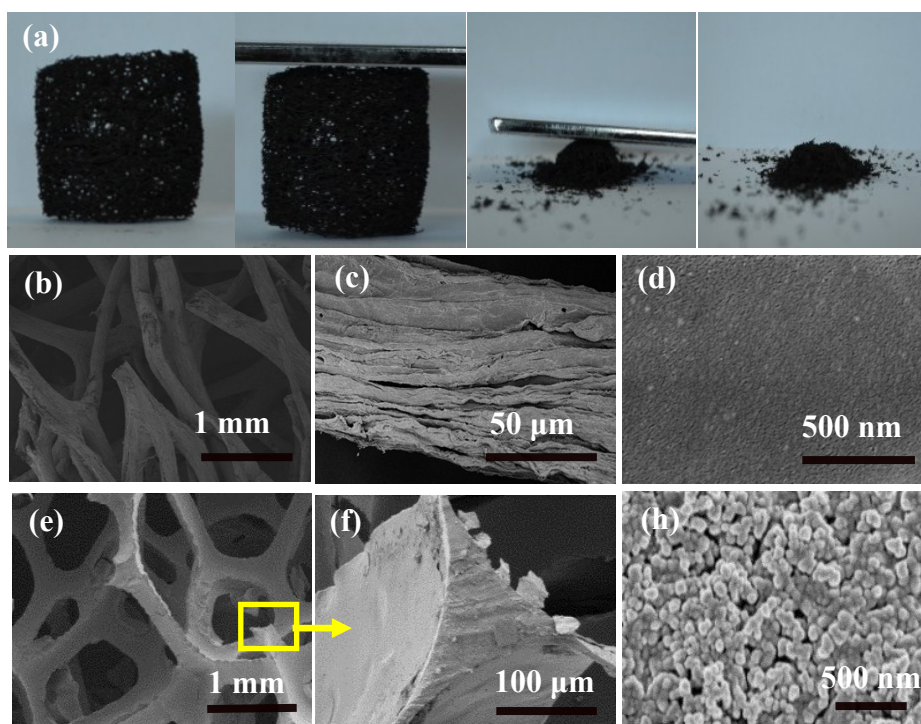
**Figure S1** Chemical characterization of the traditional CI. (a) Raman spectrum of the traditional CI; (b) Full-scan XPS of the CI; (c) high resolution C 1s XPS scan of the CI; (d) high resolution O 1s XPS scan of the CI.

Figure S2



**Figure S2** SEM images of the LS before (a) and after (b) application of the ink coating at low magnification, and (c)  $N_2$  adsorption–desorption isotherm for CI-LS-5 electrode.

**Figure S3**



**Figure S3** (a) A set of pictures shows the irretrievable process of a compressed LSC; (b-d) SEM images of the LSC; (e-h) SEM images of the CI-SP.

Figure S4

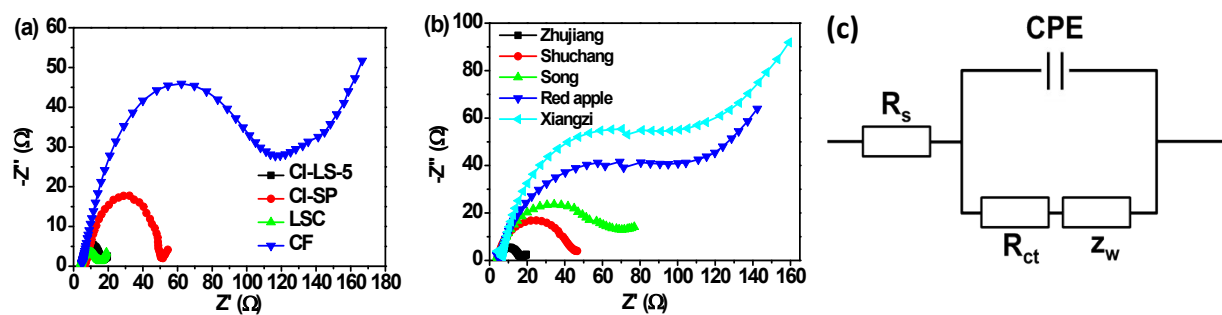


Figure S4 EIS tests for various 3D electrodes (a), CI-LS-5 electrodes with different brands of CI (b), and the equivalent circuit (c).

Figure S5

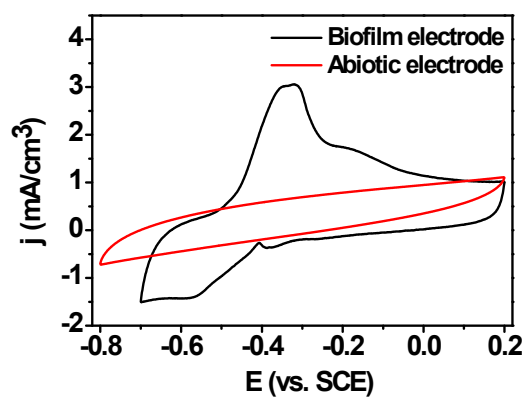
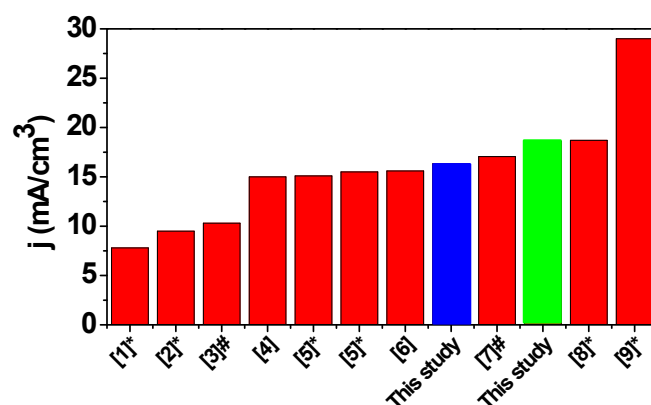


Figure S5 CV of electroactive biofilms grown on the CI-LS-5 electrode in the absence of acetate (Black line) and CV of CI-LS-5 electrode before biofilm growth (Red line) at scan rate of 5 mV/s.



Figure S6

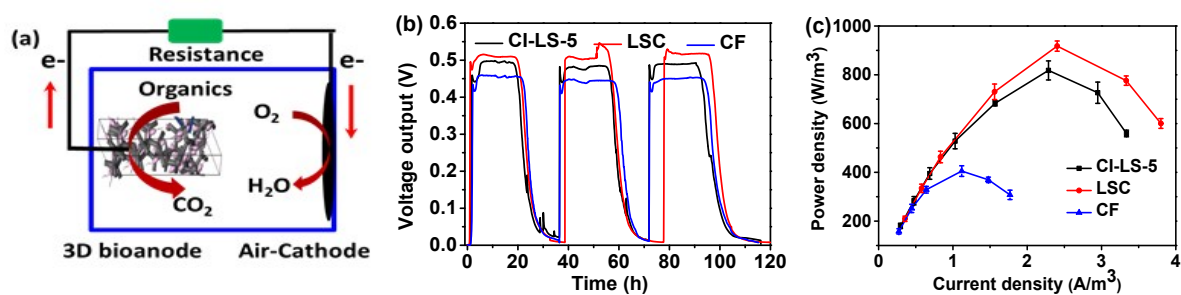


**Figure S6** Comparison of CI-LS-5 electrode performance with that of the current state-of-the-art electrodes (The number in brackets indicates the corresponding reference; the blue and green columns represent the CI-LS-5 and LSC electrodes, respectively. The # symbol indicates the use of graphene or carbon nanotubes for constructing 3D electrodes, and the \* symbol indicates the use of pyrolysis to fabricate 3D electrodes).

#### References

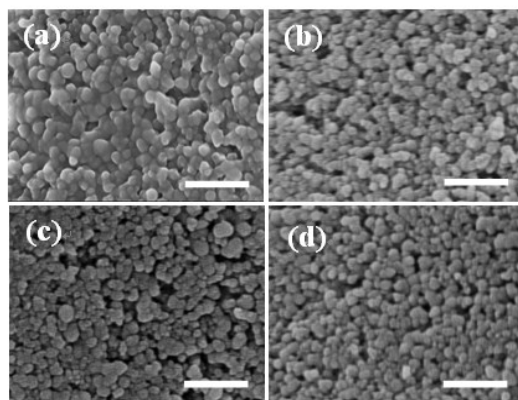
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Figure S7



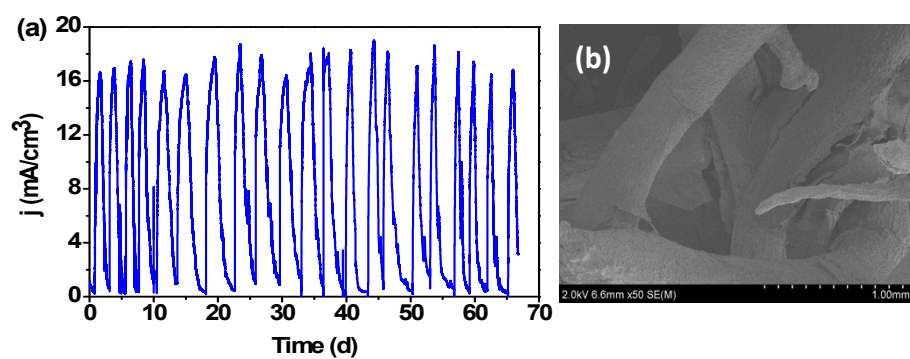
**Figure S7** The performance of the as-prepared 3D electrode was examined in a single-chamber microbial fuel cell with an air-cathode. (a) Schematic of the basic configuration of a single chamber air-cathode MFC, in which a wet-proofed carbon cloth loaded with Pt/C is used as an air cathode and a biofilm-attached 3D electrode is used as a bioanode; (b) voltage output curves of the MFCs equipped with different anodes at 1000 ohm; (c) power output curves of the MFCs equipped with different anodes.

**Figure S8**



**Figure S8** SEM images of four brands of CI used for coating LS (Scale bar: 1  $\mu\text{M}$ ): (a) Shuchang Ink; (b) Song Ink; (c) Red apple Ink; (d) Xiangzi Ink.

Figure S9



**Figure S9** (a) Bioelectrocatalytic current generation at the CI-LS-5 electrode versus time during long-term operation; (b) SEM image of the bioelectrode after 70 days of operation.