

## **Ti<sub>3</sub>C<sub>2</sub> MXene as an excellent anode material for high performance microbial fuel cells**

Da Liu,<sup>a†</sup> Ruiwen Wang,<sup>a†</sup> Wen Chang,<sup>a</sup> Lu Zhang,<sup>a</sup> Benqi Peng,<sup>a</sup> Huidong Li,<sup>a</sup> Shaoqin Liu<sup>ab</sup> Mei Yan<sup>\*ab</sup> and Chongshen Guo<sup>\*ab</sup>

*School of Life Science and Technology, Harbin Institute of Technology,*

*Harbin 150001, China. \*E-mail: [yanmei@hit.edu.cn](mailto:yanmei@hit.edu.cn),*

*[chongshenquo@hit.edu.cn](mailto:chongshenquo@hit.edu.cn)*

*Key Laboratory of Micro-systems and Micro-structures Manufacturing,*

*Ministry of Education, Harbin Institute of Technology, Harbin 150001,*

*China.*

† Da Liu and Ruiwen Wang contributed equally to this work.

## **Microbial community analysis**

After 500 h steady and repeatable voltage output, anodes were cut into pieces and taken for DNA extraction. DNA extraction was conducted using Power Soil DNA Isolation Kit (MoBio Laboratories, Inc., Carlsbad, CA) following the manufacturer's instructions. DNA concentration was confirmed by a spectrophotometer (NanoDrop 2000c, Thermo, USA). High-throughput microbial community analysis was conducted on MiSeq platforms. Raw sequence data to NCBI Sequence Read Archive (SRA) was uploaded with accession number PRJNA428311. Universal primers 515F (5'-GTGCCAGCMGCCGCGGTAA-3'), and 907R (5'-CCGTCAATTCCTTTGAGTTT-3') was used for PCR amplifying V4 and V5 regions of the bacterial 16S rRNA gene. PCR product was mixed and purified with Qiagen Gel Extraction Kit (Qiagen, Germany). Sequencing libraries were generated using TruSeq DNA PCR-Free Sample Preparation Kit (Illumina, USA). Individual samples were barcoded in one run of an Illumina HiSeq platform (2500, Illumina, CA) that generated 250 bp paired-end sequencing reads. OTUs were generated by sequences (analyzed by Uparse software) with  $\geq 97\%$  similarity. Phylogenetic relationship was constructed by phylogenetically assigning sequences obtained to the phylum, order, class, family and genes level using the MOTHUR program with distance level of 0.03 and confidence threshold of 97% for the phylogenetic classification. Relative abundance of a certain sample was calculated by dividing its total sequences to the total sequences.

---

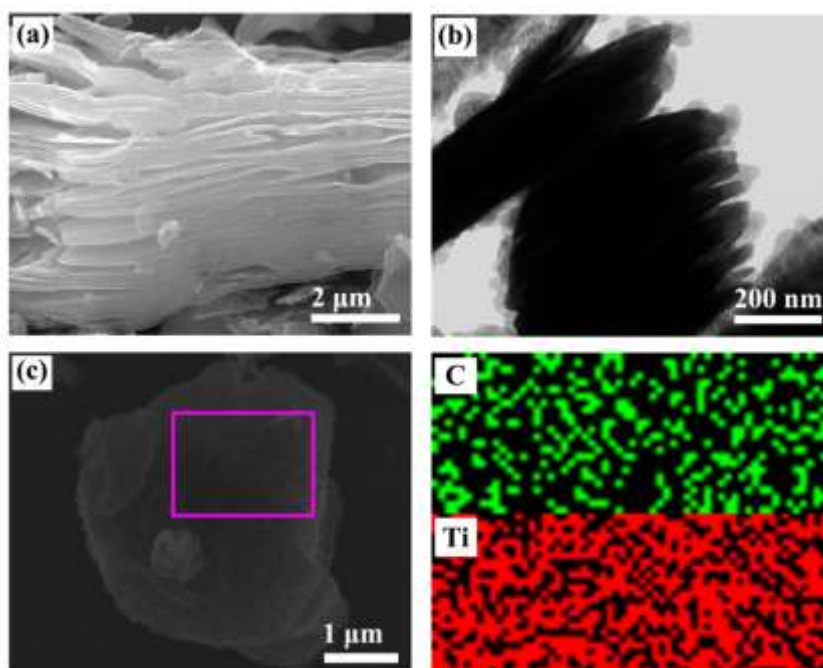


Fig. S1 (a)-(b) SEM and TEM images of multilayers  $\text{Ti}_3\text{C}_2$  MXene. (c) SEM image and corresponding elemental mapping of C and Ti

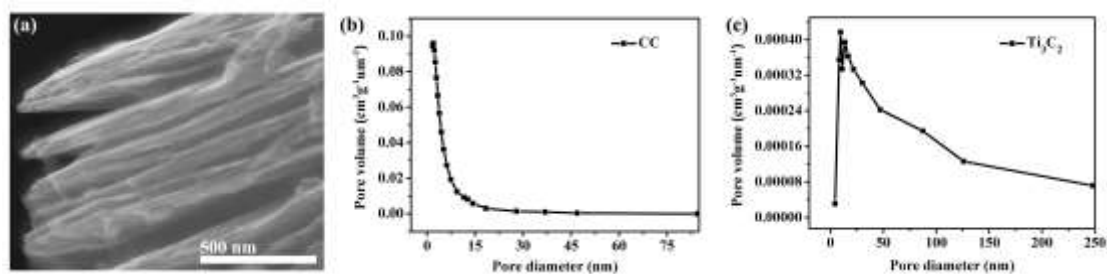


Fig. S2 (a) High-magnification SEM image of the  $\text{Ti}_3\text{C}_2$  MXene. (b)-(c) Pore size distribution curves of the bare carbon cloth and the  $\text{Ti}_3\text{C}_2$  MXene powder

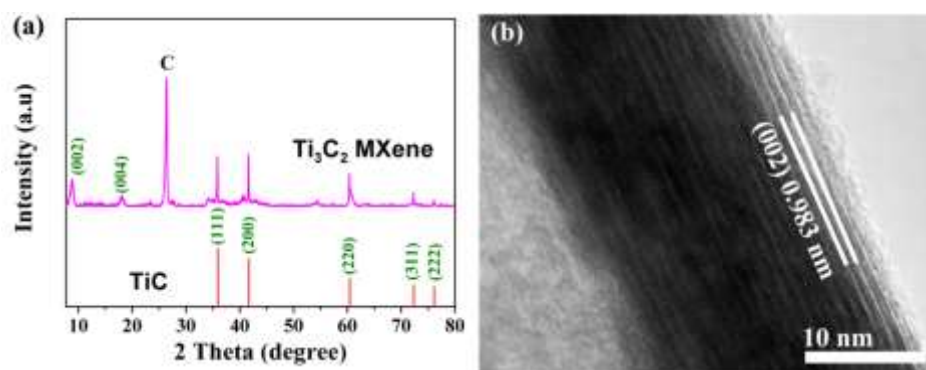


Fig. S3 (a) XRD patterns and (b) HRTEM image of  $\text{Ti}_3\text{C}_2$  MXene

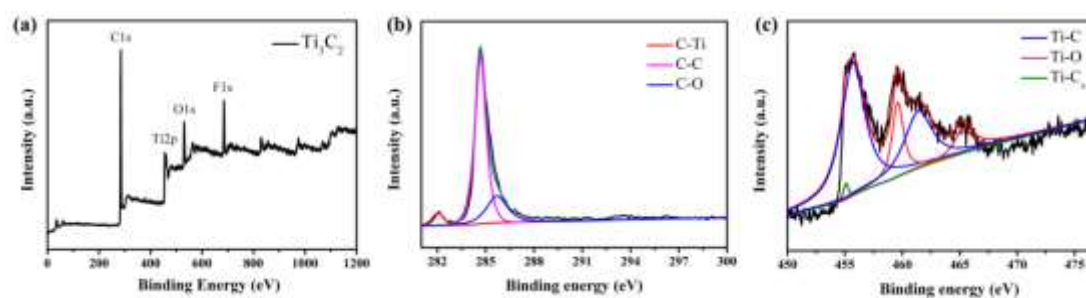


Fig. S4 (a) Full-range XPS spectra of  $\text{Ti}_3\text{C}_2$  MXene. (b) High-resolution of C 1s and (c) Ti 2p spectra

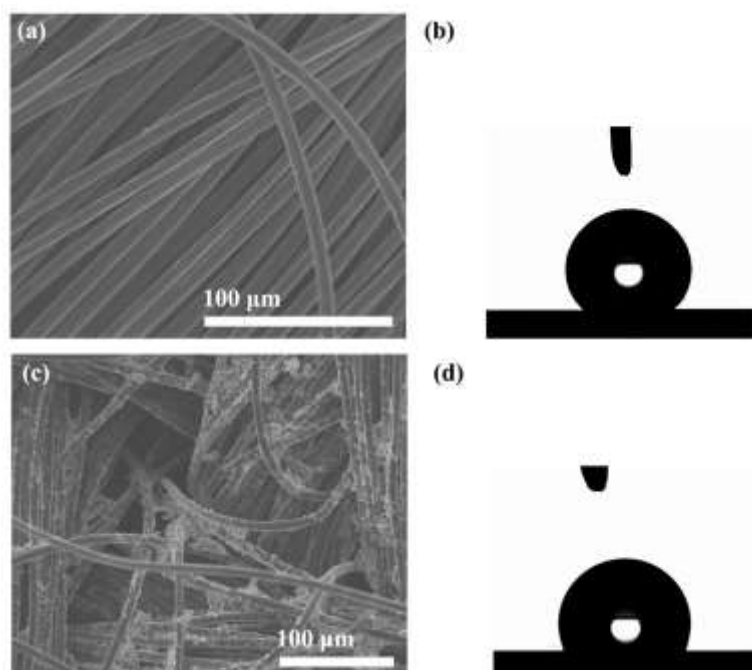


Fig. S5 (a)-(b) SEM image and contact angle ( $\theta = 140.0^\circ$ ) of carbon cloth. (c)-(d) SEM image and contact angle ( $\theta = 114.8^\circ$ ) of  $\text{Ti}_3\text{C}_2$  MXene coated

on the carbon cloth

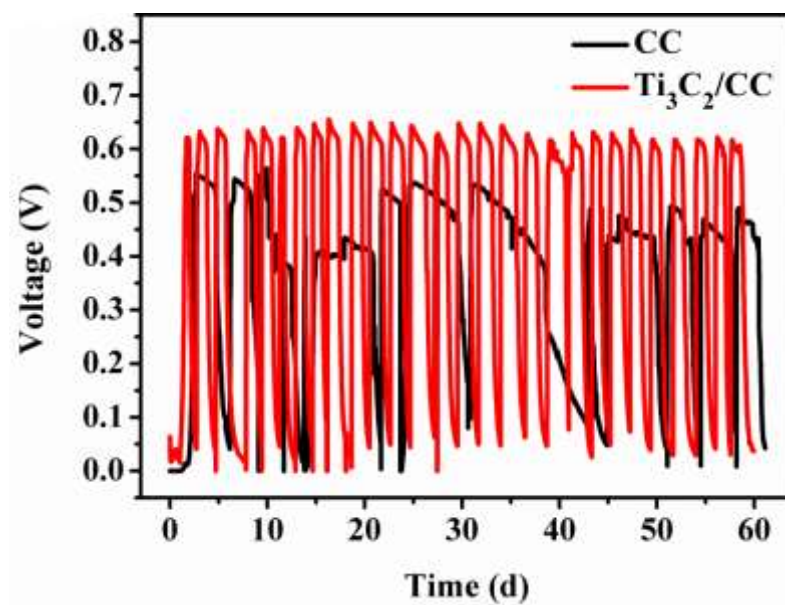


Fig. S6 The output voltage of different Ti<sub>3</sub>C<sub>2</sub>/CC (red)- and CC (black)-based MFCs with an external loading resistance of 1000  $\Omega$  in long-term operation

Table S1 Comparison of MFC performances with literature reports within five years

| Anode                                      | Microorganism               | Feed    | Configuration  | Maximum power density (mW m <sup>-2</sup> ) | Ref   |
|--------------------------------------------|-----------------------------|---------|----------------|---------------------------------------------|-------|
| N-CNTs/rGO                                 | <i>S. putrefaciens</i> CN32 | lactate | dual-chamber   | 1137                                        | [S1]  |
| Nitrogen-enriched graphitic carbon (NGC)   | <i>S. oneidensis</i> MR-1   | acetate | single-chamber | 750                                         | [S2]  |
| CNT-RTIL (room temperature ionic liquid)   | <i>Shewanella algae</i>     | lactate | dual-chamber   | 245.71                                      | [S3]  |
| PPy/NFs/PET                                | <i>Escherichia coli</i>     | glucose | dual-chamber   | 2420                                        | [S4]  |
| PANI/Carbon paper                          | <i>S. oneidensis</i> MR-1   | ---     | dual-chamber   | 693±36                                      | [S5]  |
| Magnéli-phase titanium suboxides (MM-TiSO) | mixed                       | acetate | ---            | 1541±18                                     | [S6]  |
| α-FeOOH                                    | mixed                       | acetate | single-chamber | 693±20                                      | [S7]  |
| Porous carbon                              | <i>E. coli</i>              | glucose | single-chamber | 1606                                        | [S8]  |
| rGO/MnO <sub>2</sub> /CF                   | mixed                       | acetate | dual-chamber   | 2605                                        | [S9]  |
| TiO <sub>2</sub> /rGO                      | <i>S. putrefaciens</i> CN32 | ---     | dual-chamber   | 3169                                        | [S10] |
| Graphene-containing foam (GCF)             | <i>S. putrefaciens</i>      | lactate | dual-chamber   | 786                                         | [S11] |
| CP/GNRs/PANI                               | <i>S. oneidensis</i>        | lactate | dual-chamber   | 856                                         | [S12] |
| PPy/GO                                     | <i>S. oneidensis</i> MR-1   | lactate | dual-chamber   | 1326                                        | [S13] |

Continued

|                                                       |                                |                |                          |             |                      |
|-------------------------------------------------------|--------------------------------|----------------|--------------------------|-------------|----------------------|
| PANI-ERGNO/C<br>C                                     | mixed                          | acetate        | dual-<br>chamber         | 1390        | [S14]                |
| Porous graphite                                       | <i>E. coli</i>                 | glucose        | single-<br>chamber       | 2600        | [S15]                |
| NiO/graphene                                          | <i>S. putrefaciens</i><br>CN32 | ---            | dual-<br>chamber         | 3632        | [S16]                |
| FeS <sub>2</sub> /rGO                                 | mixed                          | acetate        | dual-<br>chamber         | 3224        | [S17]                |
| 3D graphene/Pt<br>composites                          | <i>S. oneidensis</i><br>MR-1   | lactate        | dual-<br>chamber         | 1460        | [S18]                |
| Graphene/Au-m<br>odified carbon<br>paper<br>(CP/G/Au) | <i>S. oneidensis</i>           | lactate        | dual-<br>chamber         | 508         | [S19]                |
| Graphene-layer-<br>based graphite<br>plate (GL/GP)    | mixed                          | acetate        | single-<br>chamber       | 670±34      | [S20]                |
| PANI+G+CC                                             | mixed                          | acetate        | single-<br>chamber       | 884±96      | [S21]                |
| Graphene<br>microsheets                               | <i>E. coli</i>                 | glucose        | single-<br>chamber       | 2850        | [S22]                |
| G-CTAB-G                                              | mixed                          | acetate        | single-<br>chamber       | 731.3       | [S23]                |
| <b>Ti<sub>3</sub>C<sub>2</sub>/CC</b>                 | <b>mixed</b>                   | <b>acetate</b> | <b>dual-<br/>chamber</b> | <b>3740</b> | <b>This<br/>work</b> |

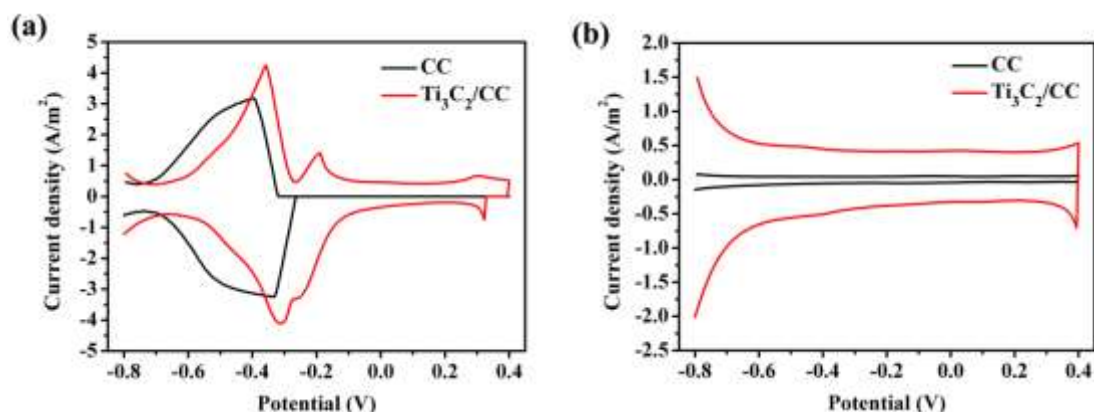


Fig. S7 DPV of (a) biofilm on  $\text{Ti}_3\text{C}_2/\text{CC}$  (red curve) and CC (black curve) anodes under turnover condition, (b)  $\text{Ti}_3\text{C}_2/\text{CC}$  (red curve) and CC (black curve) anodes without biofilm under turnover condition. Electrolyte: fresh anolyte (acetate  $2 \text{ g L}^{-1}$  in PBS with vitamin and trace element added), amplitude 60 mV, pulse width 200 ms, potential increment 6 mV, vs Ag/AgCl.

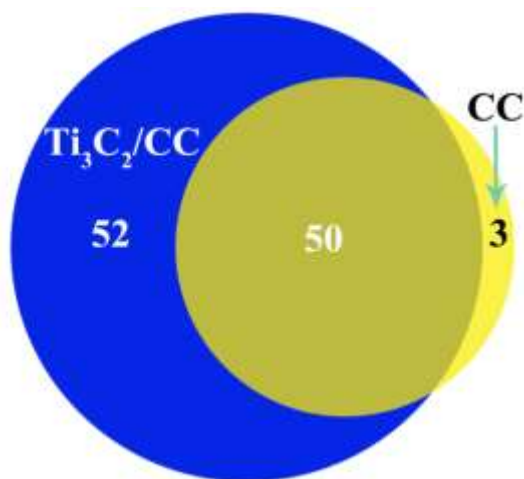


Fig. S8 The venn diagram of diversity species on  $\text{Ti}_3\text{C}_2/\text{CC}$  (blue colour, 52+50) and CC (yellow colour, 50+3) anodes.

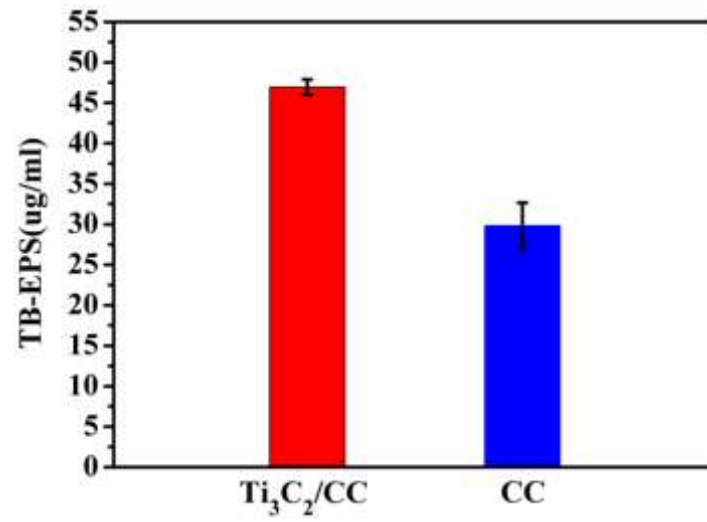


Fig. S9 The concentration of tightly bound extracellular polymeric substances (TB-EPS) in Ti<sub>3</sub>C<sub>2</sub>/CC and CC anode surface biofilm

## References

- [S1] X. S. Wu, Y. Qiao, Z. Z. Shi, W. Tang, C. M. Li, *ACS Appl. Mater. Interfaces*, 2018, **10**, 11671-11677.
- [S2] S. J. You, M. Ma, W. Wang, D. P. Qi, X. D. Chen, J. H. Qu, N. Q. Ren, *Adv. Energy Mater.*, 2017, **7**, 1601364
- [S3] L. Mahrokh, H. Ghourchian, K. H. Nealson, *J. Mater. Chem. A.*, 2017, **5**, 7979-7991.
- [S4] Y. F. Tao, Q. Z. Liu, J. H. Chen, B. Wang, Y. D. Wang, K. Liu, M. F. Li, H. Q. Jiang, Z. T. Lu, D. Wang, *Environ. Sci. Technol.*, 2016, **50**, 7889-7895.
- [S5] R. B. Song, K. Yan, Z. Q. Lin, J. S. C. Loo, L. J. Pan, Q. C. Zhang, J. R. Zhang, J. J. Zhu, *J. Mater. Chem. A.*, 2016, **4**, 14555-14559.
- [S6] M. Ma, S. J. You, G. S. Liu, J. H. Qu, N. Q. Ren, *J. Mater. Chem. A.*, 2016, **4**, 18002-18007.
- [S7] X. H. Peng, H. B. Yu, X. Wang, N. S. J. Gao, L. J. Geng, L. N. Ai, *J. Power Sources*, 2013, **223**, 94-99.
- [S8] X. F. Chen, D. Cui, X. J. Wang, X. S. Wang, W. S. Li, *Biosens. Bioelectron.*, 2015, **69**, 135-141.
- [S9] C. Y. Zhang, P. Liang, X. F. Yang, Y. Jiang, Y. H. Bian, C. M. Chen, X. Y. Zhang, X. Huang, *Biosens. Bioelectron.*, 2016, **81**, 32-38.
- [S10] L. Zou, Y. Qiao, X. S. Wu, C. X. Ma, X. Li, C. M. Li, *J. Power Sources*, 2015, **276**, 208-214.
-

- [S11] L. Yang, S. Q. Wang, S. Q. Peng, H. M. Jiang, Y. M. Zhang, W. F. Deng, Y. M. Tan, M. Ma, Q. J. Xie, *Chem. Eur. J.*, 2015, **21**, 10634-10638.
- [S12] C. Zhao, P. P. Gai, C. H. Liu, X. Wang, H. Xu, J. R. Zhang, J. J. Zhu, *J. Mater. Chem. A.*, 2013, **1**, 12587-12594.
- [S13] Z. S. Lv, Y. F. Chen, H. C. Wei, F. S. Li, Y. Hu, C. H. Wei, C. H. Feng, *Electrochimica Acta*, 2013, **111**, 366-373.
- [S14] J. X. Hou, Z. L. Liu, P. Y. Zhang, *J. Power Sources*, 2013, **224**, 139-144.
- [S15] J. Xiong, M. H. Hu, X. P. Li, H. Y. Li, X. Li, X. Liu, G. Z. Cao, W. S. Li, *Biosens. Bioelectron.*, 2018, **109**, 116-122.
- [S16] X. S. Wu, Z. Z. Shi, L. Zou, C. M. Li, Y. Qiao, *J. Power. Sources*, 2018, **378**, 119-124.
- [S17] R. W. Wang, M. Yan, H. D. Li, L. Zhang, B. Q. Peng, J. Z. Sun, D. Liu, S. Q. Liu, *Adv. Mater.*, 2018, 1800618.
- [S18] S. Zhao, Y. Li, H. Yin, Z. Liu, E. Luan, Z. Feng, Z. Tang, S. Liu, *Sci. Adv.*, 2015, **1**, e1500372.
- [S19] C. E. Zhao, P. P. Gai, R. B. Song, J. R. Zhang, J. J. Zhu, *Anal. Methods*, 2015, **7**, 4640-4644.
- [S21] L. H. Huang, X. F. Li, Y. P. Ren, X. H. Wang, *Int. J. Hydrogen Energ.*, 2016, **41**, 11369-11379.
- [S22] A. T. Najafabadi, N. Ng, E. Gyenge, *Biosens. Bioelectron.*, 2016, **81**, 102-110.
- [S23] L. X. Xue, N. Yang, Y. P. Ren, X. F. Li, Y. G. Shi, Z. Z. Hua, X. H. Wang, *J.*
-

*Chem. Technol. Biotechnol.*, 2017, **92**, 157-162.