Supplementary Information (SI)

Can bicarbonate replace phosphate to improve the sustainability of bioelectrochemical systems for H₂ production?

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| Buffer type | Phosphate | Bicarbonate | рН | Conductivity |
|-------------|-----------|-------------|------|--------------|
| | (%) | (%) | | (S/m) |
| P/C@100% | 100 | 0 | 7.04 | 1.13 |
| P/C@20% | 20 | 80 | 8.40 | 1.30 |
| P/C@0.8% | 0.8 | 99.2 | 8.45 | 1.74 |
| P/C@0% | 0 | 100 | 8.47 | 1.80 |

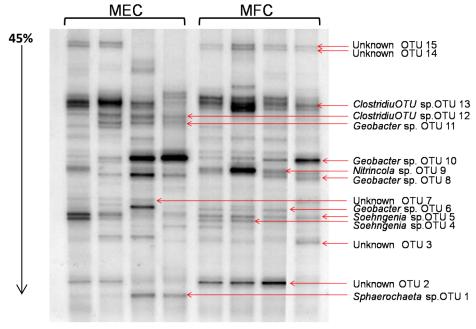
Table S1. pH and conductivity of different electrolyte buffer systems

| Table S2. The BLAST results of the distinct DGGE band DNA in the bioanode of |
|---|
| MECs and MFCs operated with different buffers |

| Sample | Species Closest gene sequence | | Identities | |
|--------|------------------------------------|--|-------------------|--|
| OTU-1 | Spirochaetaceae sp. | <i>Spirochaetaceae</i> bacterium S37_12_1 LK391556.1 | 151/151 (100%) | |
| OTU-2 | Geoalkalibacter ferrihydriticus | Uncultured bacterium C4 AB630334.1 | 147/148(9 9%) | |
| OTU-3 | _ | Uncultured bacterium TB003- 114 AB196087.1 | 131/132 (99%) | |
| OTU-4 | <i>Soehngenia</i> sp. | <i>Soehngenia</i> sp. B312138 HQ133002.1 | 120/122 (98%) | |
| OTU-5 | Soehngeniasp. | <i>Soehngenia</i> sp. B312138 HQ133002.1 | 127/127 (100%) | |
| OTU-6 | Geobacter sp. | Uncultured <i>Geobacter</i> sp. MEC25- 11 HM124838.1 | 152/152(1 00%) | |
| OTU-7 | Bacteroidetes sp. | Uncultured <i>Bacteroidetes</i> bacterium C1016S KF193877.1 | 147/148 (99%) | |
| OTU-8 | Geobacter sp. | Uncultured bacterium MFC4P_296 JF309187.1 | 152/152 (100%) | |
| OTU-9 | <i>Nitrincola</i> sp. | <i>Nitrincola</i> sp. LAR05R9 JX945779.1 | 157/158 (99%) | |
| OTU-10 | Geobacter sp. | Uncultured bacterium MEC_Bicarb_Ac-041 GQ152932.1 | 162/162 (100%) | |
| OTU-11 | Geobacter sp. | Uncultured bacterium MECB5- C04 KF171498.1 | 144/145 (99%) | |
| OTU-12 | Clostridium sp. | Uncultured <i>Clostridium</i> sp. 8 JX548536.1 | 147/147 (100%) | |
| OTU-13 | Clostridium sp. | Uncultured <i>Clostridium</i> sp. 8 JX548536.1 | 143/144 (99%) | |
| OTU14 | _ | Acholeplasmamorum strain 72- 043 NR_042959.1 | 112/120 (93%) | |
| OTU15 | _ | Acholeplasmamorum strain 72- 043 NR_042959.1 | 112/120 (93%) | |

| Sample | Species | Closest gene sequence | Identities | |
|---------|-----------------|---|-------------------|--|
| OTU-S1 | Clostridium sp. | Uncultured <i>Clostridium</i> sp. isolate DGGE gel band 8 JX548536.1 | 146/146(1 00%) | |
| OTU-S2 | Arcobacter sp. | Uncultured bacterium clone SanDiego_a6357 KF799750.1 | 138/138(1 00%) | |
| OTU-S3 | Pseudomonas sp. | Pseudomonas sp. GRPAa2 GU939693.1 | 144/148(9 7%) | |
| OTU-S4 | Geobacter sp. | Uncultured bacterium clone MFC4P_296 JF309187.1 | 151/151(1 00%) | |
| OTU-S5 | Geobacter sp. | Geobacter sp. GSS01 KJ620987.1 | 151/151(1 00%) | |
| OTU-S6 | Geobacter sp. | Geobacter sp. GSS01 KJ620987.1 | 148/152(9 7%) | |
| OTU-S7 | Geobacter sp. | Geobacter sp. GSS01 KJ620987.1 | 150/153(9 8%) | |
| OTU-S8 | Geobacter sp. | Geobacter sp. GSS01 KJ620987.1 | 149/149(1 00%) | |
| OTU-S9 | Geobacter sp. | Uncultured bacterium clone MFC4P_296 JF309187.1 | 149/149(1 00%) | |
| OTU-S10 | Geobacter sp. | Uncultured bacterium clone MFC4P_296 JF309187.1 | 149/149(1 00%) | |

Table S3. The BLAST results of the distinct DGGE band DNA in the bioanodeofMECs with different applied voltages (0.5 V~1.0 V) and different time slot



P/C@ 0% 0.8% 20% 100% 0% 0.8% 20% 100%

Fig. S1 The DGGE profile showing the microbial diversity and population in the bioanodes of MECs, together with their precursor MFCs. All MECs and MFCs were operated with electrolyte buffers at different P/C ratios (with a total concentration of 0.1 M). The formamide gradient in DGGE gel is from 45% to 60%.

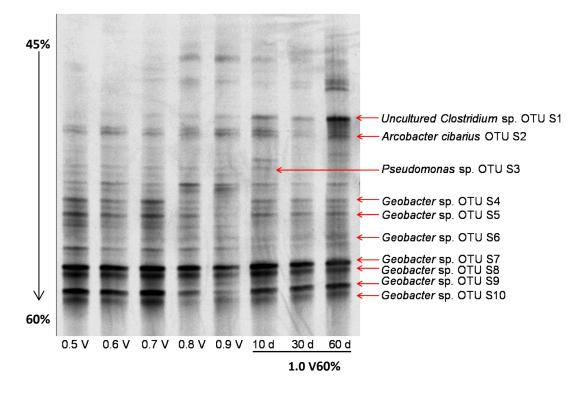


Fig.S2 The DGGE profile showing the microbial diversity and population in the bioanodes of MECs operated under different applied voltages (0.5 V~1.0V) and different time slot (1.0V: 10 d, 30 d, 60 d). All MECs were operated withthe electrolyte buffers at P/C@20% (total concentration: 0.1 M). The formamide gradient in DGGE gel is from 45% to 60%.