

Fig S1. pH changes during experimental periods for all 14 MEC reactors, and inflow solution. The effluent pH varied slightly among different reactors but very stable over time for individual reactors.



Fig. S2. Current production during the experimental period for individual reactors. All 14 reactors produced current although some degrees of variations were observed among different reactors. Current production was also fairly stable over the experimental periods for each reactor.





(B) CH₄ production



(C) CO₂ production



Fig S3. Stability of gas production over experimental time: (A) H_2 ; (B) CH_4 ; and (C) CO_2 . The variation of the yield of gases (H_2 , CH_4 or CO_2) over time was measured by coefficient variations (CVs) of the production of each gas for each reactor. The CV at each time point was calculated based on the average and standard deviations of gas yield over a 15-day window. By shifting the window to next subsequent time points,

the dynamic changes of CVs in gas production over time can be visualized as shown in this Figure. Little changes of the CVs in gas yields were observed for most (H_2) or all (CH₄, CO₂) of the reactors after initial 10 days, suggesting that the functional performance for each reactor in terms of H_2 production was relatively stable for these reactors after initial 10 days. However, the H_2 yields varied substantially among different reactors whereas the CO₂ yields were very consistent among different reactors. The CH₄ yields also varied considerably among different reactors.



Fig. S4. Distribution of the functional groups which were most important in separating different bioreactor communities as shown in Fig. 2. The most important genes/populations were identified by SIMPER (similarity percentage) analysis (in PAST software). Only the functional genes belonging to top 10% contributors (i.e., 90% cumulative cutoff value) were shown in this figure. The number of genes/populations of each functional group was listed in the parentheses and total 59 genes/populations were included in the pie diagram.







Figure S5. Abundance changes of functional gene groups (A), phylogenetic groups (B), and key metal-reducing bacteria and several other dominant groups based on cytochrome genes (C). The overall functional and phylogenetic diversity patterns of these MEC reactor communities were substantially different at the levels of major functional categories (A) or phyla/classes (B) although the reactors were inoculated with the same inoculums and operated under identical conditions. *Geobacter, Shewanella, Desulfovibrio* and *Anaeromyxobacter* are typical metal-reducing bacteria. *Pseudomonas* and *Rhodobacter* are commonly found in MEC reactors. They could play important roles in substrate oxidation, electron transfer and hydrogen production. As shown in this Figure (C), their abundance varied substantially among different reactors.



Fig. S6. Canonical correspondence analysis (CCA) showing the relationships between community functional structure and environmental variables. Significant relationships (p < 0.1) were observed between community structure and all environmental variables as whole or individual gases.

Table S1. Average β - diversity of the MEC reactor communities based on individual functional gene categories

| Functional gene categories | Sorensen Dissimilarity | Bray Curtis Dissimilarity |
|-----------------------------|------------------------|------------------------------|
| Antibiotic resistance | 0.37± 0.09 | 0.46±0.11 |
| Carbon cycling | 0.51±0.13 | 0.60±0.12 |
| Energy process | 0.47±0.12 | 0.53 ± 0.13 |
| Metal resistance | 0.44±0.11 | 0.50±0.11 |
| Nitrogen | 0.50±0.12 | 0.57±0.13 |
| Organic remediation | 0.49±0.11 | 0.57±0.11 |
| Phosphorus | 0.43±0.12 | 0.54±0.13 |
| Sulphur | 0.47±0.12 | 0.54±0.12 |
| Phylogenetic markers (gyrB) | 0.49±0.13 | 0.59±0.13 |
| Average | 0.47±0.11 | 0.54±0.12 |

| Functional | Sorensen Dissimilarity | | | | Bray Curtis Dissimilarity | | | | | | | |
|-----------------------------------|------------------------|-------|--------|-------|---------------------------|-------|--------|-------|--------|-------|------|-------|
| gene | Adonis | | Anosim | | MRPP | | Adonis | | Anosim | | MRPP | |
| categories | F | р | R | р | δ | р | F | р | R | р | δ | р |
| Antibiotic resistance | 1.86 | 0.012 | 0.32 | 0.017 | 0.34 | 0.011 | 2.68 | 0.003 | 0.51 | 0.002 | 0.39 | 0.002 |
| Carbon cycling | 2.05 | 0.011 | 0.39 | 0.008 | 0.45 | 0.007 | 2.49 | 0.001 | 0.58 | 0.001 | 0.52 | 0.001 |
| Energy processing | 2.01 | 0.007 | 0.38 | 0.005 | 0.42 | 0.005 | 2.88 | 0.001 | 0.57 | 0.001 | 0.44 | 0.002 |
| Metal resistance | 1.89 | 0.01 | 0.32 | 0.021 | 0.4 | 0.014 | 2.56 | 0.001 | 0.53 | 0.001 | 0.44 | 0.001 |
| Nitrogen cycling | 2.09 | 0.003 | 0.4 | 0.003 | 0.44 | 0.009 | 2.66 | 0.001 | 0.54 | 0.001 | 0.49 | 0.002 |
| Organic remediation | 2.04 | 0.005 | 0.42 | 0.006 | 0.44 | 0.005 | 2.53 | 0.001 | 0.61 | 0.001 | 0.49 | 0.001 |
| Phosphorus utilization | 2.08 | 0.007 | 0.38 | 0.01 | 0.44 | 0.006 | 2.75 | 0.001 | 0.63 | 0.001 | 0.5 | 0.001 |
| Sulfur cycling | 2.55 | 0.004 | 0.42 | 0.003 | 0.37 | 0.001 | 2.96 | 0.001 | 0.64 | 0.001 | 0.45 | 0.001 |
| Phylogenetic markers (gyrB) | 2.15 | 0.003 | 0.4 | 0.005 | 0.41 | 0.009 | 2.85 | 0.001 | 0.6 | 0.001 | 0.46 | 0.001 |

Table S2. Significance tests of the differences of β -diversity among the four reactor groups based on individual functional gene categories^a.

^aSee the footnote in Table 1 for details. The degree freedom of Adonis test is (3,10).

Table S3. Estimated *p*-values of the goodness of fit to a neutral community model for different microbial communities in these reactors. The relative abundance of signal intensity $x10^5$ was used to fit the neutral community model. The metacommunity parameters are θ =367.578, m=0.0029. All communities are not fitted with the neutral community model as indicated by both Kolmogorov-Smirnov (KS) test and χ^2 test.

| Logal community | Group | J | KS | 3 | χ^2 -test | | |
|-----------------|-------|------------------------|----------|---------|----------------|---------|--|
| | | (local community size) | D | р | χ^2 | р | |
| #1 | С | 100291 | 0.979308 | < 0.001 | 844.3806 | < 0.001 | |
| #2 | В | 101066 | 0.991927 | < 0.001 | 20462.35 | < 0.001 | |
| #3 | В | 100630 | 0.989397 | < 0.001 | 5741.006 | < 0.001 | |
| #4 | В | 101245 | 0.992994 | < 0.001 | 23247.82 | < 0.001 | |
| #5 | В | 100796 | 0.989997 | < 0.001 | 9449.698 | < 0.001 | |
| #6 | В | 100509 | 0.982906 | < 0.001 | 3346.085 | < 0.001 | |
| #7 | В | 100690 | 0.987395 | < 0.001 | 6715.593 | < 0.001 | |
| #8 | С | 100634 | 0.989021 | < 0.001 | 5490.522 | < 0.001 | |
| #9 | А | 100554 | 0.984567 | < 0.001 | 4568.416 | < 0.001 | |
| #10 | С | 100662 | 0.988796 | < 0.001 | 6095.438 | < 0.001 | |
| #11 | С | 100387 | 0.983466 | < 0.001 | 1679.017 | < 0.001 | |
| #12 | D | 100873 | 0.993847 | < 0.001 | 7509.892 | < 0.001 | |
| #13 | С | 100853 | 0.990003 | < 0.001 | 10410.9 | < 0.001 | |
| #14 | С | 100389 | 0.981648 | < 0.001 | 2302.054 | < 0.001 | |