Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supplementary Information

Enhancing the anode performance of microbial fuel cells in the treatment of oil-based drill sludge by adjusting the stirring rate and supplementing oil-based drill cuttings

Qi Feng [a], Longjun Xu [a, *], Chenglun Liu [a, b], Yu Hao[c], Zhengxin Yang [a], Teng Zhang [a], Yanling Liu [a], Huaren Su [a]

[a] State Key Laboratory of Coal Mine Disaster Dynamics and Control, Chongqing University, Chongqing, 400044, China

[b] College of Chemistry and Chemical Engineering, Chongqing University, Chongqing, 401331, China

[c] School of Resources and Security, Chongqing Vocational Institute of Engineering, Chongqing, 402260, China

* Corresponding author.

E-mail address: xulj@cqu.edu.cn (L. Xu).

Section 1: The details for microbial community analysis

The microbial community characteristics in these samples were assayed through high-throughput sequencing and determined using V3~V4 region of the 16S rRNA gene. For high-throughput sequencing, the genes were amplified by PCR with TransStart Fastpfu DNA Polymerase. The sequencing was performed on the Meijiyun platform and the analysis was carried out with the universal bacterial primers of 338F (5′-ACTCCTACGGGAGGCAGCAG-3′) and 806R (5′-GGACTACHVGGGT-WTCTAAT-3′). Clustering of sequence into operational taxonomic units (OTUs) was performed at 97% sequence identity. From each OTU selected the representative sequences, and annotated with species information using Ribosomal Database Project classifiers with the Silva database (SSU123Genus level heatmap was prepared using

Heatmap Illustrator (HEMI, version 1.0)).

Section 2: Data analysis

1. The current density (I_A) and power density (P_A) of the MFC were obtained with the following equation (1) and (2):

$$I_A = \frac{U}{R_{ext}A} \qquad (1)$$

$$P_A = I_A * U \qquad (2)$$

Where U is the measured voltage (V), *Rext* is the external resistance value (Ω) and A is the surface area of anode (m^2).

2. Exchange current density (i_0, A/cm^2) was calculated by the Tafel curve expressed as the Butler-Volmer:

$$Log\ i = log\ i_0 - \beta nF\eta/2.303\ RT \qquad (3)$$

Where i is the current density at which an electric current passes (A/m^2); i_0 is exchange current density (A/cm^2); β is the symmetry factor; F is the Faraday constant (96485 C/mol); η is served as the cathodic overpotential (V); R is as the ideal gas constant (8.31 J / (mol K); and T is the absolute temperature (K).

3. Coulomb efficiency (CE) is the relative contribution index of MFC power generation to organic matter removal, and the calculation equation is as follows (4):

$$CE(\%) = \frac{M_s \int_0^t \frac{U}{R} dt}{F \cdot b \cdot V_{anode} \cdot \Delta COD} \times 100\% \qquad (4)$$

Where Ms is molar mass of substrate, g/mol; F is Faraday constant, 96485 C/mol; b is number of electrons to be transferred by oxidized 1mol organic matter; Vanode is total effective volume, L, ΔCOD is variation of COD versus with time of 0~t, g/L.

Tab. S1 Main characteristics of anolyte and catholyte.

Parameters	OBDS	Landfill leachate	Aerobic sludge
Color	Dark black	Black brown	Light yellow
Odor	Stench of heavy oil	Unpleasant	Bad smell

Temperature (℃)	25 ± 1	25 ± 1	25 ± 1
Phase state	Viscous liquid	Opaque suspension	Transparent suspension
COD (mg/L)	21900 ± 210	2864 ± 2.32	3219 ± 23
NH_3-N (mg/L)	395±17.8	418 ± 4.7	105.4 ± 9.6
NO_3^-−N (mg/L)	32 ± 0.17	170 ± 2.4	210.4 ±10.3
NO_2^-−N (mg/L)	2 ± 0.0014	5.65 ± 0.14	7.3 ± 0.23
pH	7.2 ± 0.1	8.98 ± 0.1	6.3 ± 0.05
Conductivity (mS/cm^2)	36.3 ± 0.7	6.95 ± 0.4	17.3 ± 0.5

Tab. S2 EDS characteristic of OBDCs.

Element	Wt (%)	Atom (%)
C K	42.41	56.56
O K	38.36	38.37
Si K	2.15	1.23
Ba K	9.93	1.16
Fe K	3.85	1.1
Al K	1.86	0.93
K K	1.12	0.46
Fe K	0.32	0.19

Tab. S3 Anode chambers initial data of different OBDCs content.

MFC	COD (mg/L)	NH_3-N (mg/L)	NO_3^-−N (mg/L)	NO_2^-−N (mg/L)
1 g/L	22015 ± 132	395 ± 17.8	29.4 ± 0.15	2.70±0.02
2 g/L	23100 ± 114	404 ± 20.1	32.5±0.21	2.92±0.04
4 g/L	26209 ± 168	417 ± 16.5	34.2±0.45	3.11±0.03
6 g/L	27320 ± 153	421 ± 11.9	37.5±0.62	3.15±0.07

---Control group data was same as OBDS from Tab.S1

Tab. S4 Fitting parameters value in Tafel curve.

MFC	βc (mV/dec)	βa (mV/dec)	Onset Potential (mV)	Exchange Voltage (mV)	Exchange Current (A/cm^2)
Control group	82.1	112.3	350	287	1.7×10^{-3}
100 rpm	103.6	54.1	380	310	2.1×10^{-3}
2 g/L	88.9	51.3	370	300	1.8×10^{-3}
100 rpm + 2 g/L	49.9	43.2	393	380	2.2×10^{-3}

Tab. S5 Proportion of elements in biofilms under the conditions of control group and 100 rpm +2 g/L.

Element	Samples			
	Control group		100 rpm +2 g/L	
	Wt (%)	Atom (%)	Wt (%)	Atom (%)
C K	28.83	40.85	40.15	52.04
N K	7.91	9.61	8.47	9.41
O K	34.72	36.93	31.12	30.28
Na K	2.30	1.71	1.38	0.93
Si K	0.98	0.59	1.38	0.77
P K	5.57	3.06	1.82	0.91
Cl K	5.90	2.83	4.85	2.13
Ca K	1.83	0.78	1.55	0.60
Fe K	11.97	3.65	7.93	2.21
Al K	/	/	0.49	0.28
S K	/	/	0.87	0.42

Tab. S6 Proportion of elements in the treated samples (control group and the group of 100 rpm + 2 g/L).

Element	Samples			
	Control group		100 rpm +2 g/L	
	Wt (%)	Atom (%)	Wt (%)	Atom (%)
C K	25.28	42.77	26.05	37.69
O K	34.04	43.23	41.56	45.14
Mg K	0.63	0.53	0.79	0.57
Al K	1.09	0.82	3.58	2.31
Si K	3.66	2.65	17.09	10.57
S K	6.22	3.94	0.76	0.41
Cl K	1.61	0.92	1.21	0.59
Ca K	2.98	1.51	2.78	1.20
Ba L	24.49	3.62	3.41	0.43
K K	/	/	1.64	0.73
Fe K	/	/	1.13	0.35

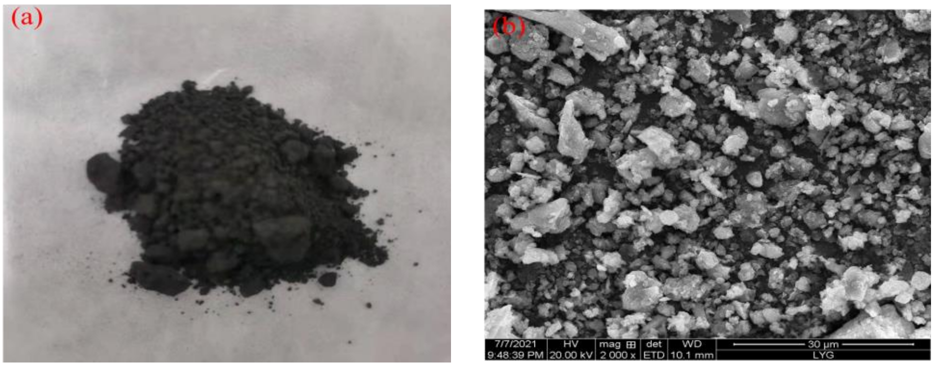


Fig. S1 The photo (a) and SEM (b) of OBDCs.

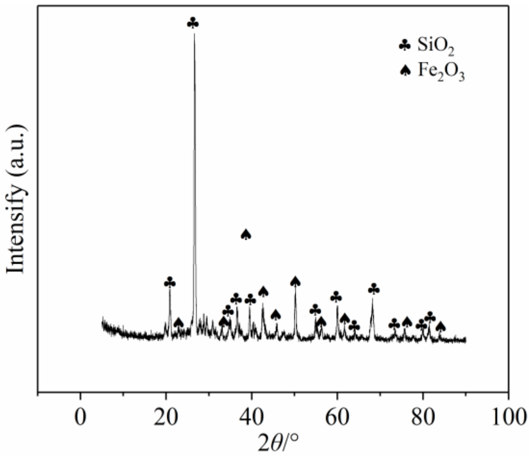


Fig. S2 XRD of pretreatment OBDCs.

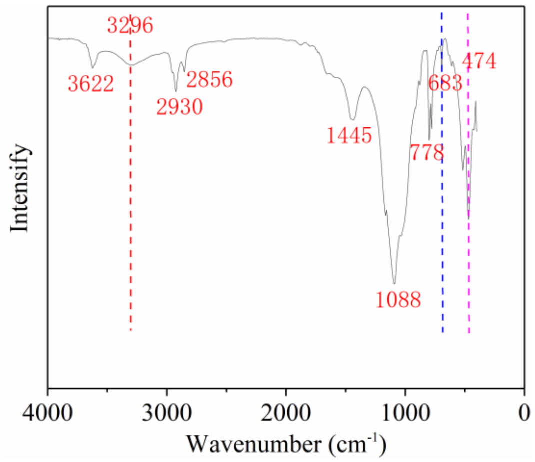


Fig. S3 FTIR of pretreatment OBDCs.

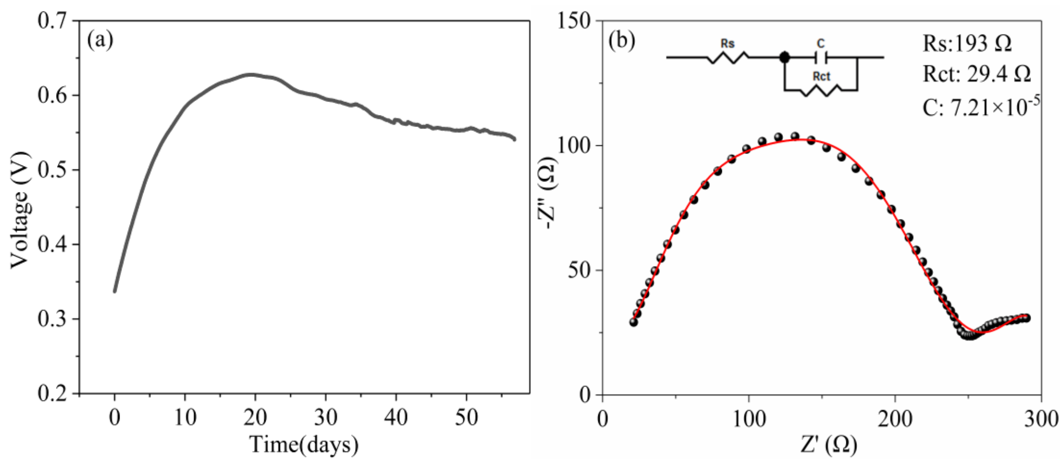
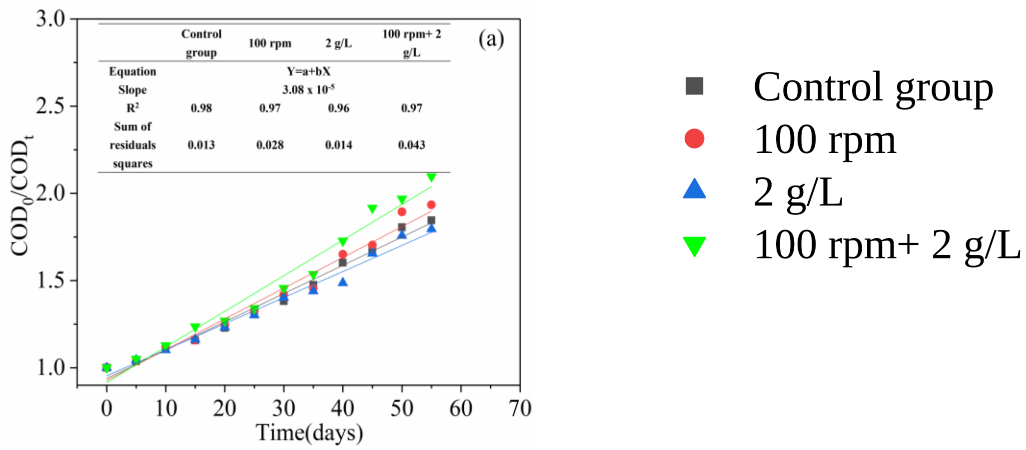


Fig. S4 (a) output voltage curves and (b) EIS under the optimized conditions.



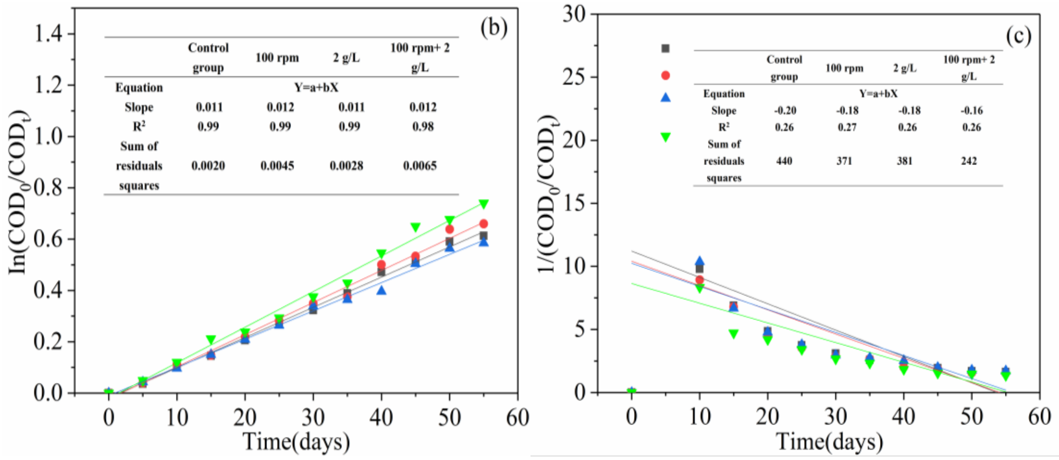


Fig. S5 (a) zero order kinetics fitting of COD removal in analyte, where COD_0 is the initial COD; (b) first order kinetics fitting of COD removal in anolyte; (c) second order kinetics fitting of COD removal in anolyte.

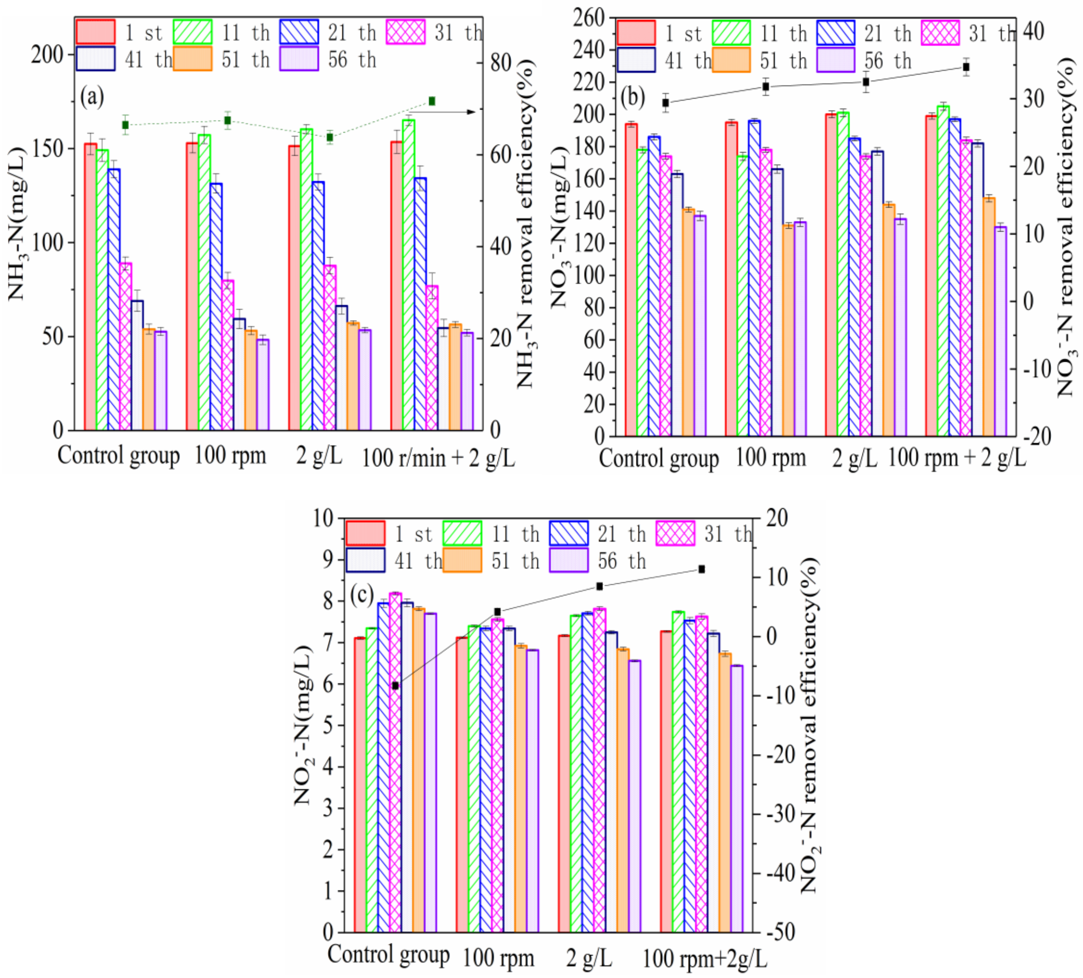


Fig. S6 Removal amounts of NH_3-N (a), NO_3^--N (b) and NO_2^--N (c)among the cathode chambers, respectively.

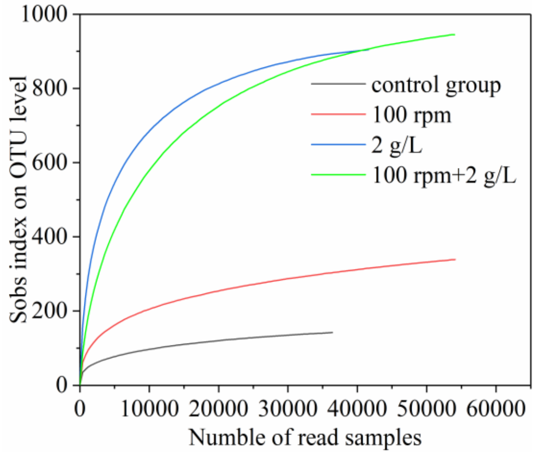


Fig. S7 Rarefaction curves for microbial communities.

Fig. S8 shows the analysis of the microbial community structure at the phylum level. Most of the bacteria belonging to *Proteobacteria* and *Bacteroides* involved in the process of important electricity-producing and organic compound-degrading that can perform long-distance electronic transmission. The abundance of *Actinobacteria* in the 100 rpm, 2 g/L and 100 rpm + 2 g/L groups was increased significantly compared with that in the control group, which suggested that the synergy of OBDCs and stirring rate enhanced the leading role of sulfur-reducing and nitrate-denitrifying bacteria. This might demonstrate the effects of simultaneous inorganic nitrogen and organic loads on the growth of autotrophic and heterotrophic bacteria. Meanwhile, other phyla, namely, *Chloroflexi* and *Firmicutes,* accumulated in the anode, which contributed to the electric acclimation, selection of oil-based microbes and anode microbial communities, and they were likely conducive to the removal of complex organic matter, such as anionic polyacrylamide [1].

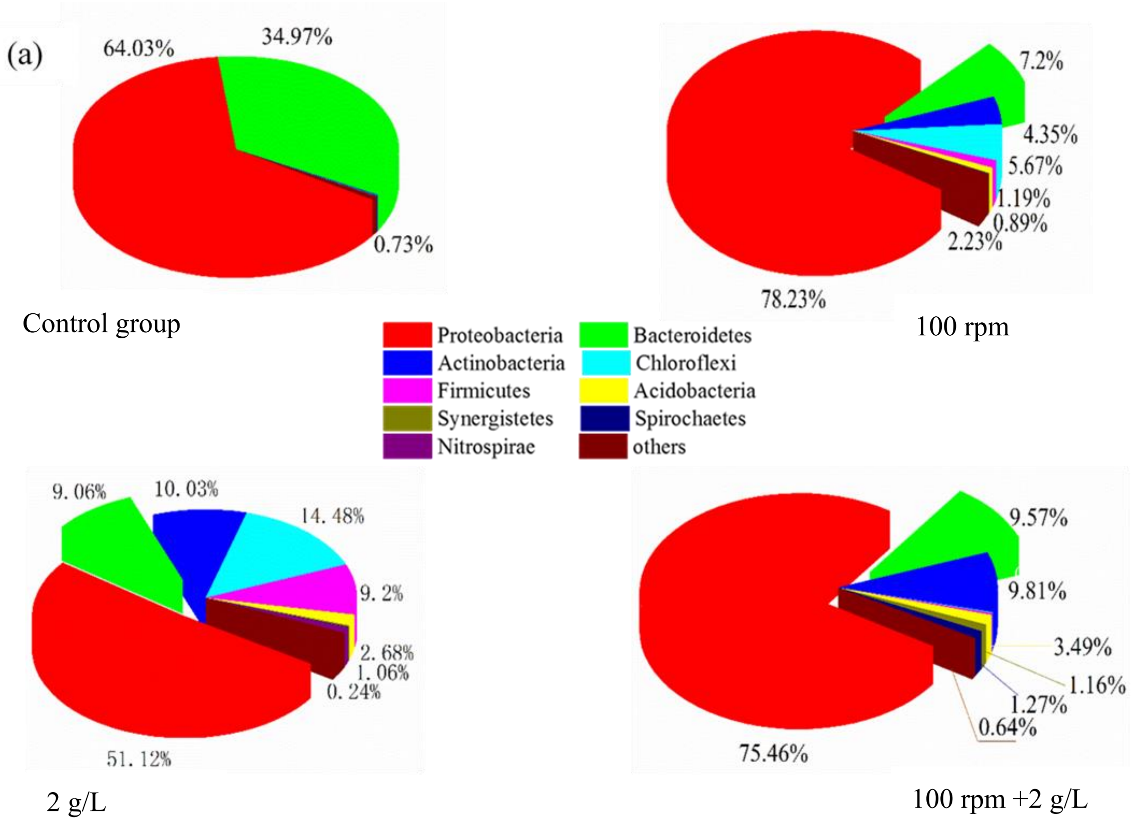


Fig. S8 Microbial communities on the anode biofilms at the phylum level.

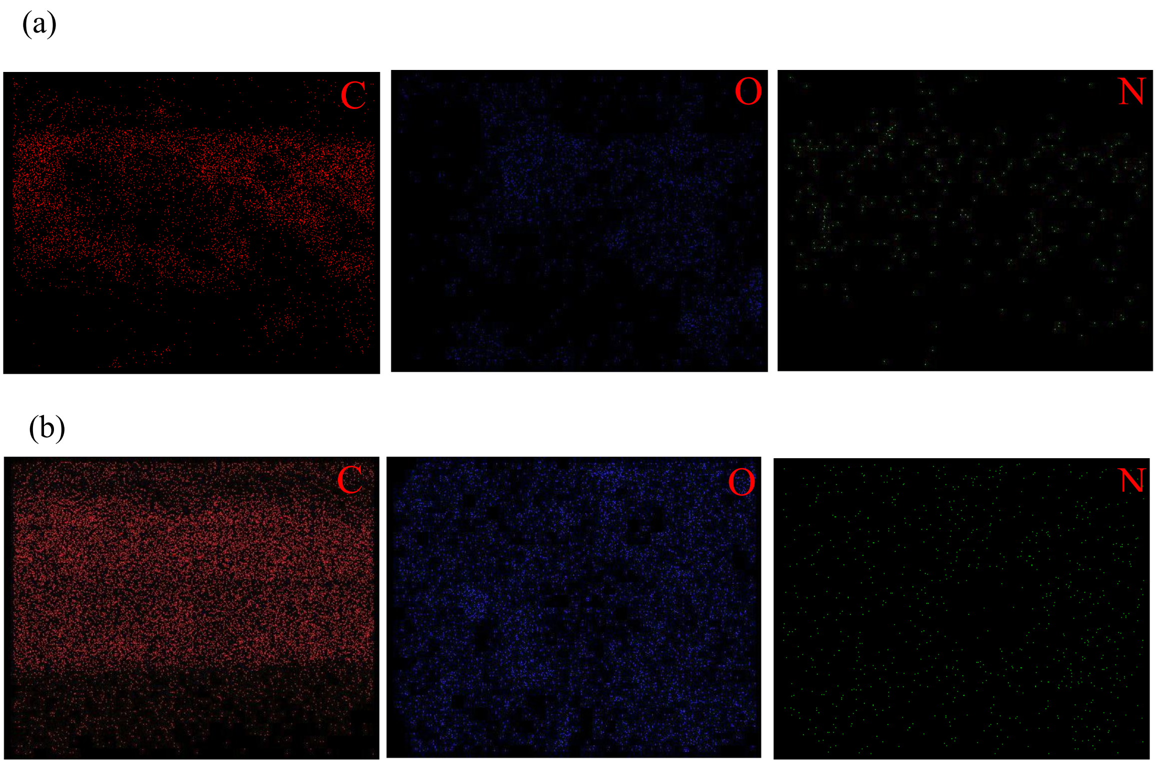


Fig. S9 Elemental mapping of biofilms: (a) control group, (b) 100 rpm + 2 g/L.

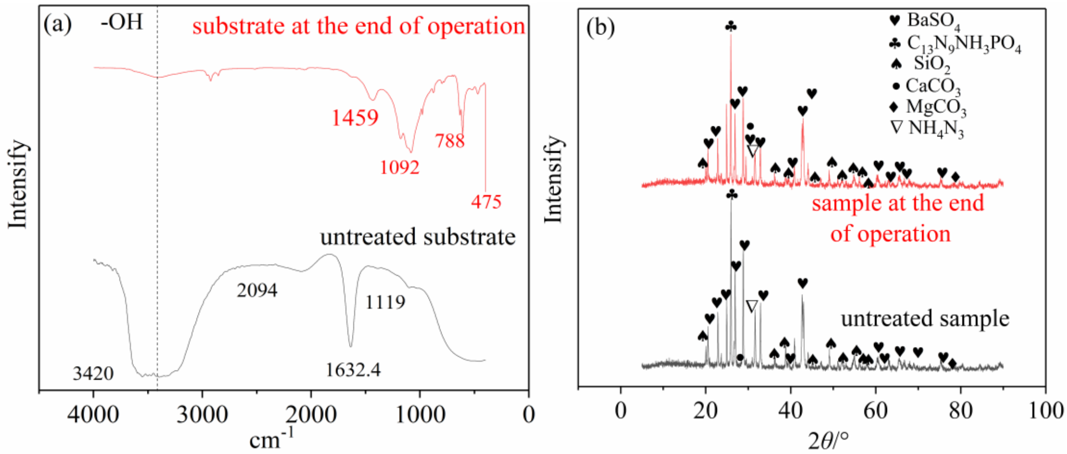
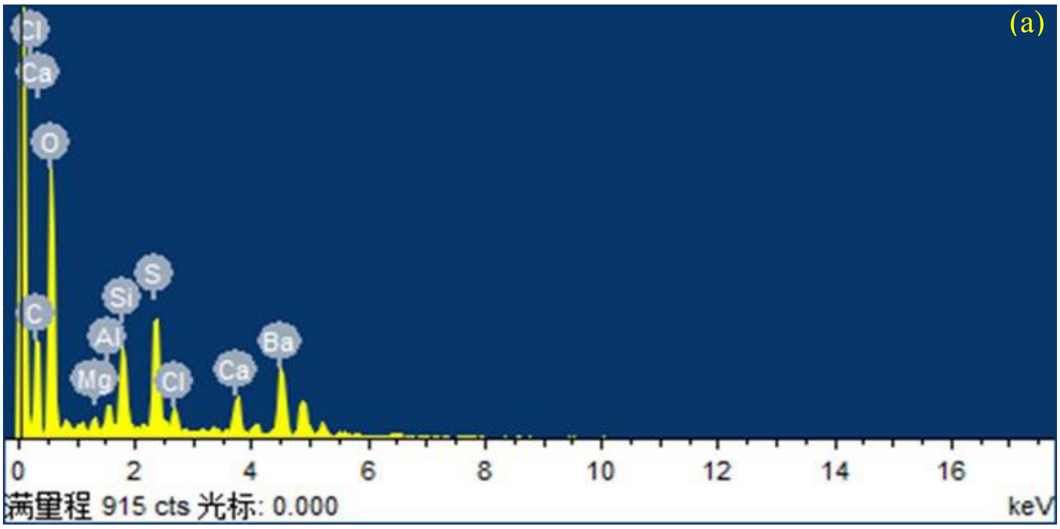


Fig. S10 Anode chamber substrates in the control group: (a) FTIR and (b) XRD.



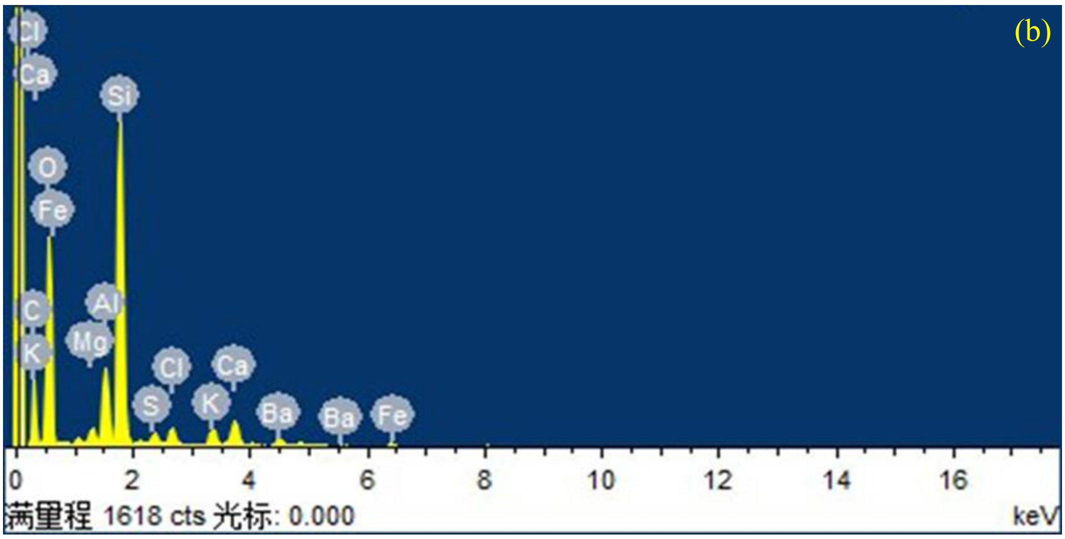


Fig. S11 EDS of the treated samples (a) control group; (b) 100 rpm + 2 g/L.

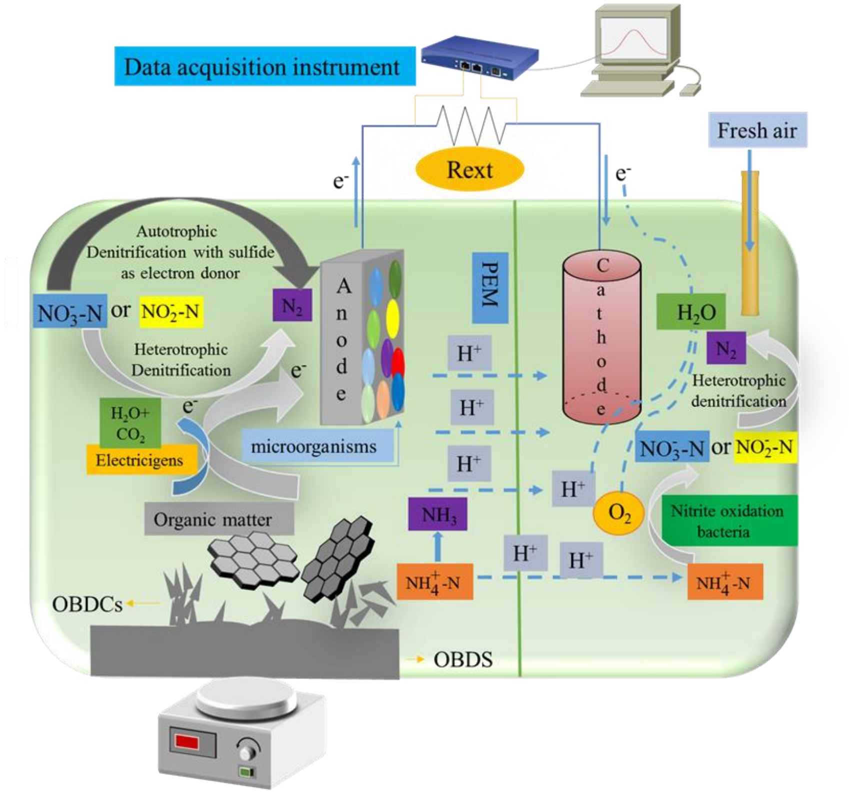


Fig. S12 Schematic diagram and possible mechanism of the simultaneous removal of complex organics and nitrogen from OBDS by an MFC.

References

[1] X. Zhang, A. Chen, D. Zhang, S. Kou, P. Lu, The treatment of flowback water in a sequencing batch reactor with aerobic granular sludge: Performance and microbial community structure, Chemosphere 211 (2018) 1065-1072.