

Electronic supplementary information (ESI)

Aerobic microbial electrochemical technology based on the coexistence and interactions of aerobes and exoelectrogens for synergistic pollutant removal from wastewater

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Preparation of GB/NPC

Graphite brushes (GBs) were immersed firstly in a 50% (w/w) concentrated sulfuric acid for five minutes to improve their hydrophilicity and then washed with distilled water for three times before further use. Polyaniline (PANi) was grown onto the GB by a chemical oxidative polymerization process. The polymerization was carried out at a temperature range of 0 to 5 °C, based on the protocol published elsewhere.¹ Briefly, separate solutions of 0.06 M aniline and 0.015 M ammonium persulfate was prepared by dissolving 0.559 g and 0.342 g, respectively in 100 mL 2 M HCl solution. Both solutions were then mixed vigorously, and the GB electrode was immersed into this mixture immediately. After oxidative polymerization for about 12 h, the composite GB/PANi electrode was obtained. After washing with distilled water for three times, the GB/PANi was de-doped by using 0.5 M ammonia solution and re-doped using 0.1 M phytic acid solution to form phytic acid doped GB/PANi composite. Finally, the phytic acid doped GB/PANi was placed in a high-temperature oven preheated to 900 °C at a heating rate of 5 °C min⁻¹ under the nitrogen atmosphere. After annealing for 1 h at the final temperature and cooling down to below 200 °C, N, P co-doped carbon modified GB composite electrode (GB/NPC) was obtained.

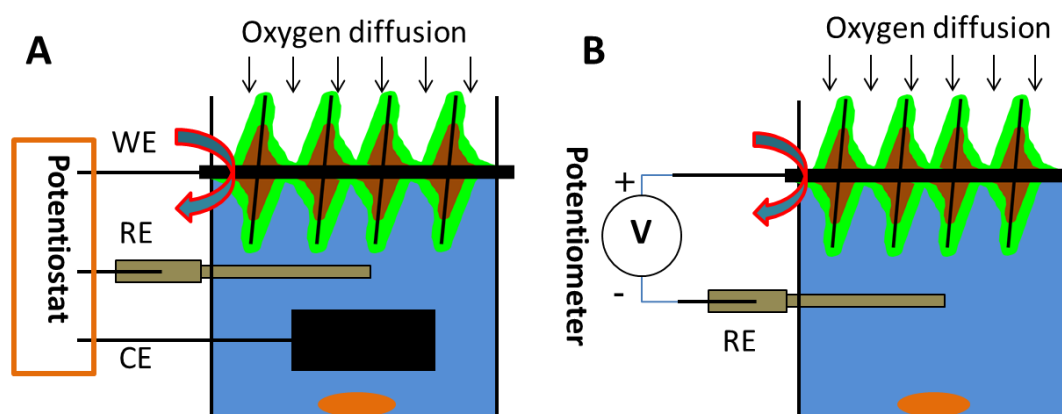


Fig. S1 Schematic diagrams of setups and steps for starting up and running the ABRB, which include (A) potentiostat controlled aerobic selection and accumulation of exoelectrogens and assembly of aerobic biofilms simultaneous and (B) operation mode of the ABRBs under aerobic condition. Notes: **WE**, working electrode, rotating brush electrode; **RE**: reference electrode, Ag/AgCl; **CE**: counter electrode, graphite plate.

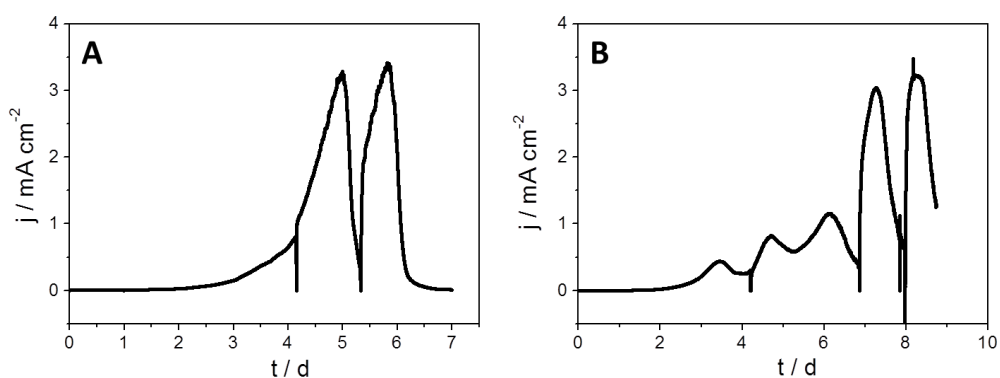


Fig. S2 Bioelectrocatalytic current generation profiles of the rotating (A) GB and GB/NPC (B) bioreactors under anaerobic conditions in chronoamperometry operation. A constant potential of +0.2 V vs. Ag/AgCl was applied at the working electrodes. Real wastewater with 10 mM acetate (total COD $740 \pm 85 \text{ mg L}^{-1}$) was used during open circuit operation process.

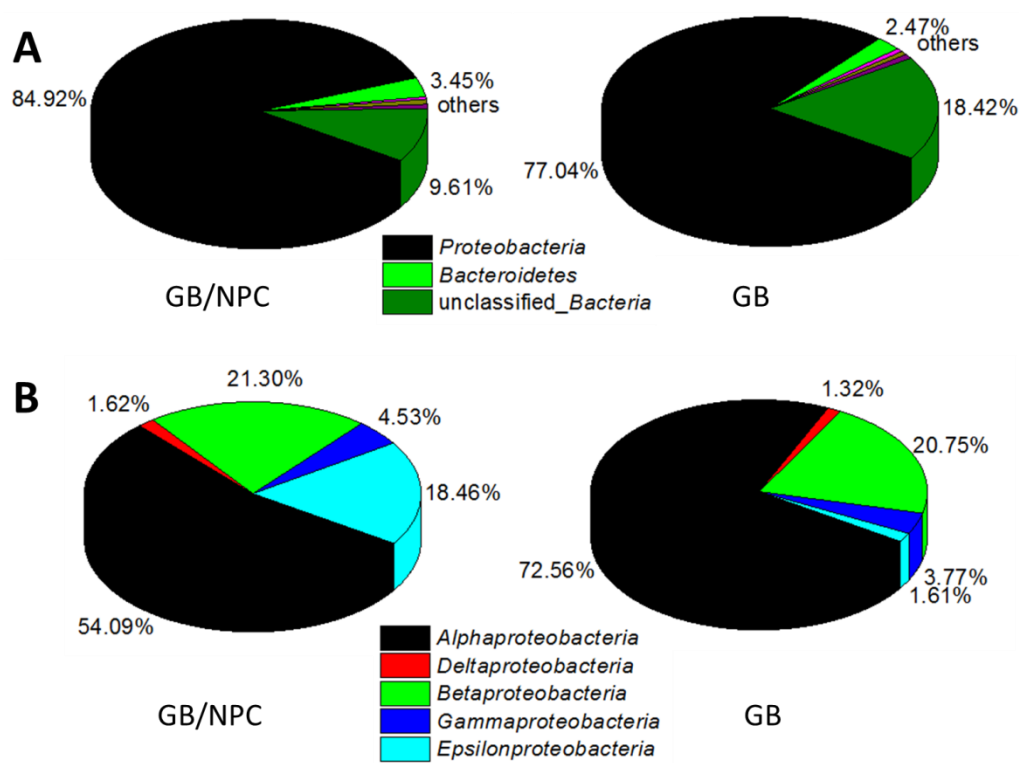


Fig. S3 Microbial community composition (A) at the phylum level in the GB/NPC and the GB. (B) The composition of the *Proteobacteria* at class level in the GB/NPC and the GB.

Table S1 Microbial community composition of biofilms in the GB/NPC and GB at different taxonomic levels

Bacteria	GB/NPC (%)	GB (%)
<u>Genus</u>		
<i>Neomegalonema</i>	40.39 ± 4.80	51.90 ± 3.74
<i>Hyphomonas</i>	2.38 ± 0.42	1.23 ± 0.23
<i>Thauera</i>	2.29 ± 0.20	9.20 ± 1.35
<i>Aquimonas</i>	0.64 ± 0.11	0.92 ± 0.22
<i>Geobacter</i>	0.033 ± 0.047	0.0029 ± 0.0015
<i>Arcobacter</i>	15.16 ± 4.03	1.21 ± 1.14
<u>Family</u>		
<i>Methylobacteriaceae</i>	40.40 ± 4.80	51.91 ± 3.73
<i>Hyphomonadaceae</i>	2.42 ± 0.43	1.26 ± 0.23
<i>Rhodocyclaceae</i> (unclassified_ <i>Rhodocyclaceae</i>)	13.94 ± 1.47 (11.41 ± 1.32)	12.40 ± 1.19 (3.06 ± 0.97)
<i>Xanthomonadaceae</i>	1.13 ± 0.19	1.33 ± 0.27
<i>Geobacteraceae</i>	0.032 ± 0.047	0.004 ± 0.0035
<i>Campylobacteraceae</i>	15.17 ± 4.03	1.21 ± 1.14
<u>Order</u>		
<i>Rhizobiales</i>	40.90 ± 4.82	52.34 ± 3.70
<i>Caulobacterales</i>	2.60 ± 0.45	1.40 ± 0.25
<i>Rhodocyclales</i>	13.94 ± 1.47	12.40 ± 1.19
<i>Xanthomonadales</i>	1.14 ± 0.19	1.35 ± 0.27
<i>Desulfuromonadales</i>	0.06 ± 0.060	0.02 ± 0.0072
<i>Campylobacteriales</i>	15.20 ± 4.04	1.21 ± 1.14
<u>Class</u>		
<i>Alphaproteobacteria</i>	44.58 ± 4.84	54.65 ± 3.40
<i>Betaproteobacteria</i>	17.55 ± 1.58	15.63 ± 1.59
<i>Gammaproteobacteria</i>	3.74 ± 0.50	2.84 ± 0.53
<i>Deltaproteobacteria</i>	1.33 ± 0.47	1.00 ± 0.26
<i>Epsilonproteobacteria</i>	15.21 ± 4.04	1.21 ± 1.14
<u>Phylum</u>		
<i>Proteobacteria</i>	83.55 ± 1.86	76.17 ± 1.48
unclassified_ <i>Bacteria</i>	9.45 ± 1.49	18.21 ± 1.26

1. He, S.; Hu, X.; Chen, S.; Hu, H.; Hanif, M.; Hou, H., Needle-like polyaniline nanowires on graphite nanofibers: hierarchical micro/nano-architecture for high performance supercapacitors. *Journal of Materials Chemistry* **2012**, 22, (11), 5114-5120.