1. The enrichment process of biocathode

A two-chambered MFC with cation exchange membrane (Qianqiu Co. Ltd., China) as separator between the anode and cathode was used for biocathode culture (Figure S1). Both anode and cathode chambers were cylinder rooms of the same sizes (4 cm long by 3 cm in diameter). Carbon fiber brushes (2.5 cm long and 3 cm in diameter) were used as anode and cathode electrode.

Both anode and cathode was inoculated with municipal wastewater. The anode medium contained glucose (1.0 g/L), NH₄Cl (0.1 g/L), phosphate buffered nutrient solution (KCl 0.13 g/L, NaH₂PO₄·2H₂O 3.32 g/L and Na₂HPO₄·12H₂O 10.32 g/L), vitamins, and trace minerals. The cathode medium contained NaHCO₃ (1.0 g/L), NH₄Cl (0.3 g/L), phosphate buffered solution (KCl 0.13 g/L, NaH₂PO₄·2H₂O 3.32 g/L and Na₂HPO₄·12H₂O 10.32 g/L), vitamins, and trace minerals.
g/L and Na$_2$HPO$_4$·12H$_2$O 10.32 g/L), vitamins and trace minerals. The anode was sealed with rubber plug to keep anaerobic condition except for the refreshment of medium. The cathode was sparging with a pump continuously. Both chambers were refilled with inoculation medium everyday until a stable voltage was achieved. All tests were conducted at a constant temperature of 30 ºC and triple repeated. To avoid the growth of algae and photosynthetic bacteria, the MFC reactors were wrapped with aluminum foil.”

![Fig. S1 Digital photography of biocathode microbial fuel cell (MFC)](image1)

**Fig. S1** Digital photography of biocathode microbial fuel cell (MFC)

![Photo-anode Biocathode](image2)

**Fig. S2** Digital photography of biocathode coupled photoelectrochemical cell (Bio-PEC)