Electronic Supplementary Information (ESI):

Graphene-Sponges as High-Performance Low-Cost Anodes for Microbial Fuel Cells

Xing Xie,\textsuperscript{a,b} Guihua Yu,\textsuperscript{c} Nian Liu,\textsuperscript{d} Zhenan Bao,\textsuperscript{c} Craig S. Criddle\textsuperscript{a}\textsuperscript{*} and Yi Cui\textsuperscript{b,e}\textsuperscript{*}

\textsuperscript{a} Department of Civil and Environmental Engineering, Stanford University, Stanford, California 94305, USA. E-mail: ccriddle@stanford.edu; Tel: +1-650-723-9032

\textsuperscript{b} Department of Materials Science and Engineering, Stanford University, Stanford, California 94305, USA. E-mail: yicui@stanford.edu; Fax: +1-650-725-4034; Tel: +1-650-723-4613

\textsuperscript{c} Department of Chemical Engineering, Stanford University, Stanford, California 94305, USA.

\textsuperscript{d} Department of Chemistry, Stanford University, Stanford, California 94305, USA.

\textsuperscript{e} Stanford Institute for Materials and Energy Sciences, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, California 94305, USA.
Experimental Details:

Electrode fabrication. Graphene nanopowders (0.1% by weight, SkySpring Nanomaterials, Inc., USA) were dispersed in water with 1% sodium cholate (Sigma-Aldrich Co. LLC., USA) by sonicating for 2 hours. To prepare graphene-sponge (G-S) composite, artificial sponges made from polyurethane (McMaster-Carr, USA) were cut into desired shapes, dipped into graphene ink, then removed and dried at ~90 ºC. The dipping-and-drying process was repeated twice to increase graphene loading. The G-S-SS electrode was prepared by sandwiching two pieces of G-S composite (1 cm × 1 cm × 0.2 cm) with a stainless steel (SS) mesh (1 cm × 1 cm × 0.05 cm, 20-mesh) in between. A conductive carbon paste (Ted Pella, Inc., USA) was applied to provide the connection between the G-S composite and the SS current collector. The G-S electrode without SS current collector was 1 cm × 1 cm × 0.4 cm, having the same volume of G-S composite as the G-S-SS electrode does. A plain SS mesh functioned as a control sample. All the electrodes were connected to the external circuit through titanium (Ti) wires.

MFC construction and operation. A traditional H-shaped two-chamber MFC was built as described previously. Three different anodes were placed in the same anodic chamber. The cathode was carbon cloth (2 cm × 5 cm, Fuel Cell Earth LLC, USA) with a platinum (Pt) catalyst layer (0.5 mg cm⁻² 10wt. % Pt on XC-72). The MFC was inoculated with the anolyte of a previous MFC originally seeded with wastewater from the Palo Alto Regional Water Quality Control Plant. The anolyte started with glucose media (1.0 g L⁻¹). Additional glucose was added when the operating voltage dropped below 0.05 V. The voltage across a 475 Ω external resistor was recorded. All experiments were conducted at room temperature (~20 ºC).
Characterization. All the SEM images were taken by a field emission SEM (XL30 Sirion, FEI, USA). The bio-samples for SEM were pretreated by a fixing and critical point drying process. The electrochemical characterization was carried out using a potentiostat equipped with electrochemical impedance spectroscopy (EIS) board (VMP3, Bio-Logic SAS, France). For all electrochemical tests, a three-electrode system was applied: the working electrode was the target electrode (SS, G-S, or G-S-SS); the counter electrode was Pt; and the reference electrode was a double junction Ag\|AgCl\|KCl (3.5M) electrode. The electrolyte was the MFC anolyte as described previously. EIS was conducted at the OCV in the frequency range of $10^5$-0.1 Hz with a 10 mV peak-to-peak sinusoidal potential perturbation. To determine the maximum current density, linear staircase voltammetries were applied by increase the anode potential from -0.5 to 0.2 V vs. RE by 25 mV each time and recording the current after 3 minutes for equilibrium. The power outputs were calculated from the loading and the recorded voltage. The current densities and power densities were normalized to the projected surface area (1 cm$^2$) or the volume (0.4 cm$^3$) of the electrodes.

Supplementary Figures:

**Fig. S1** G-S composite before and after flushing with water (~100 mL per second) for 10 minutes.

![Fig. S1](image1)

**Fig. S2** A G-S-SS composite electrode: two pieces of G-S composite (1 cm × 1 cm × 0.2 cm) with a stainless steel (SS) mesh (1 cm × 1 cm × 0.05 cm, 20-mesh) in between. A Ti wire is applied to circuit connection.

![Fig. S2](image2)
Fig. S3 Nyquist curves of the electrochemical impedance spectroscopy (EIS) tests for different electrodes after colonization.
Fig. S4 SEM images of the colonized G-S-SS anode.