Electronic Supplementary Information (ESI):

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

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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 °C. 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 \times 1 cm \times 0.2 cm) with a stainless steel (SS) mesh (1 cm \times 1 cm \times 0.05 cm, 20-mesh) in between the G-S composite and the SS current collector. The G-S electrode without SS current collector was 1 cm \times 1 cm \times 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 ¹. 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)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 °C).

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 ². 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 AglAgCllKCl (3.5M) electrode. The electrolyte was the MFC anolyte as described previously ¹. EIS was conducted at the OCV in the frequency range of 10⁵-0.1 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 cm²) or the volume (0.4 cm³) of the electrodes.

- 1. X. Xie, M. Ye, L. Hu, N. Liu, J. R. McDonough, W. Chen, H. N. Alshareef, C. S. Criddle and Y. Cui, *Energy & Environmental Science*, 2012, 5265-5270.
- X. Xie, L. B. Hu, M. Pasta, G. F. Wells, D. S. Kong, C. S. Criddle and Y. Cui, *Nano Letters*, 2011, **11**, 291-296.

Supplementary Figures:



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



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

Electronic Supplementary Material (ESI) for Energy & Environmental Science This journal is o The Royal Society of Chemistry 2012



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.