Supporting Information

Preparation of N-doped Graphene by Reduction Graphene Oxide with Mixed Microbial System and Its Hemocompatibility

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DISSCUSSION

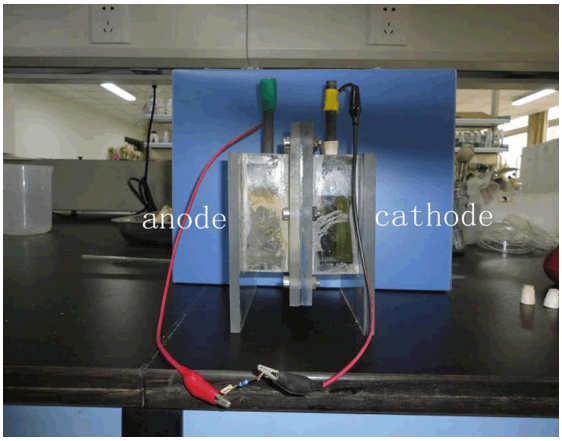

Figure S-1. the image of microbial fuel cells (*MFC*).

Microbial fuel cells (*MFC*) was assembled by our laboratory which can generate electricity steadily. The original strains were originated from anaerobic sludge in Nanjing Sewage Treatment Plant and inoculated into anode chamber after domestication, cultivation and separation.

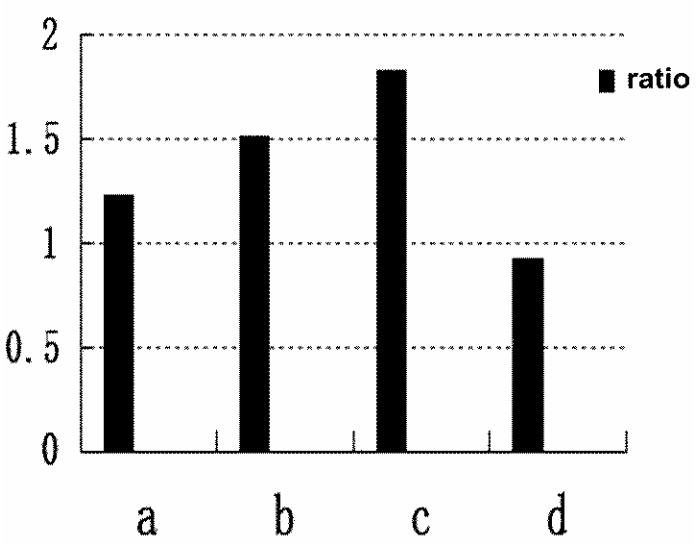

Figure S-2. the ratio of I_D/I_G :(a) *MG-1*, (b) *MG-2*, (c) *CG*, (d) *CGO*

The ratio analysis: The ratio was calculated according to the peak area of Raman spectra. The results show that *MG-1*and *MG-2* possess small ratio indicating lower defects/disorders in the graphitized structure.

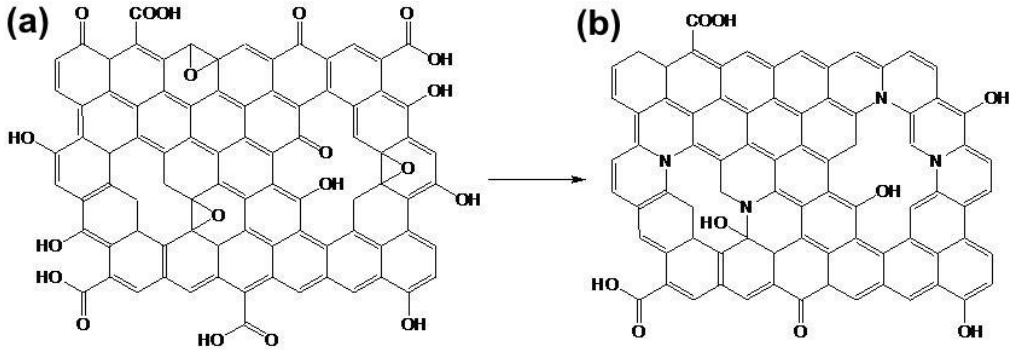

Figure S-3. simulation of the reaction equation:[1] (a) graphene oxide prepared by chemical oxidation method, (b) N-doped graphene prepared by reaction with mixed microorganisms

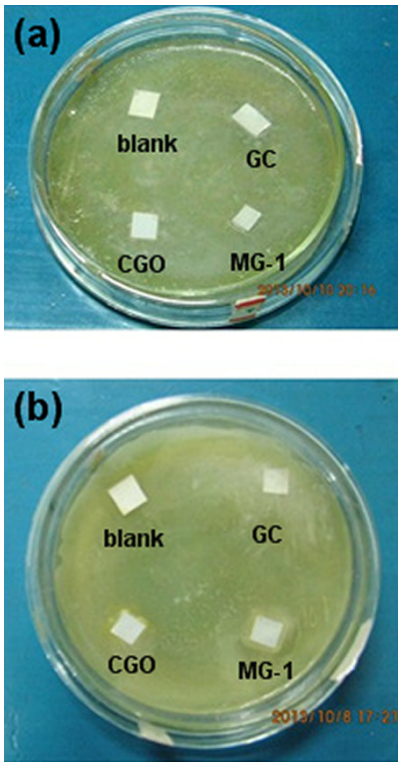

Figure S-4. antibacterial experiment of (a) *Escherichia coli* (b) *Staphylococcus aureus*: ***blank*** blank 1.2 um filter membrane, ***CG*** graphene prepared by chemical reduction method, ***CGO*** oxide graphene, ***MG-1*** contact with mixed microorganisms for 24 hours.

Antibacterial analysis: As shown in Figure S-3, there is no inhibition zone for *Escherichia coli* but *MG-1* appears a small inhibition zone which is larger than that of *CGO* for *Staphylococcus aureus*. The possible reason was that the Gram-negative *Escherichia coli* with an outer membrane were more resistant to the damage caused by the sharp edges than the Gram-positive *Staphylococcus aureus* lacking the outer membrane.[2,3]

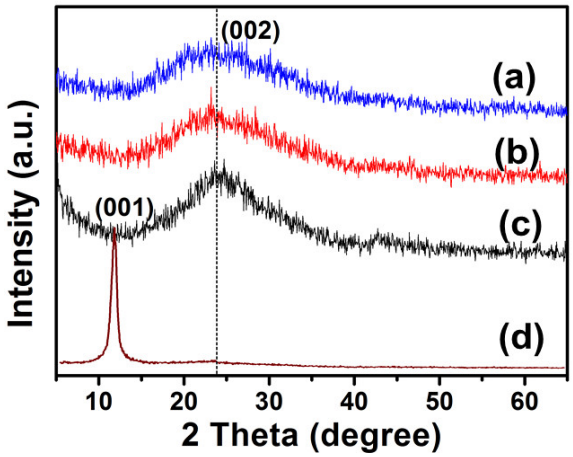

Figure S-5. X-ray diffraction patterns: (a)*MG-2* and (b) *MG-1* reaction with mixed microorganisms for 48 and 24 hours respectively, (c) *CG* graphene prepared by chemical reduction method and (d) *CGO* graphene oxide.

***XRD* Analysis.** *XRD* is a useful tool for analysis of structural distribution of atoms in space, especially crystal structure and phase purity. The *XRD* patterns were scanned and shown in Figure S-5. There is a sharp peak indexed to (001) at 2θ=13.3° for *CGO* (Figure S-5d) corresponding to an interlayer spacing of about 0.66nm because the majority of components is the sp3 domains from oxidized graphene.[4] However, the patterns of *MG-1*, *MG-2* and *CG* were

similar and only have a broadening peak indexed to (002) located at 2θ = 24.1° corresponding to an interlayer spacing of about 0.37nm due to heterogeneous nature comprised of both sp^2 domains from graphene and the sp^3 domains from graphene oxide and among them, more graphene domains. Generally, the diffraction peak of graphite located at 2θ = 26.3° and corresponds to an interlayer spacing of about 0.34 nm.[5] The interlayer spacing gradually decrease from *CGO*, N-doped graphene to graphite corresponding to 0.66, 0.37 and 0.34 nm respectively, which is in agreement with the experimental results from previous studies.

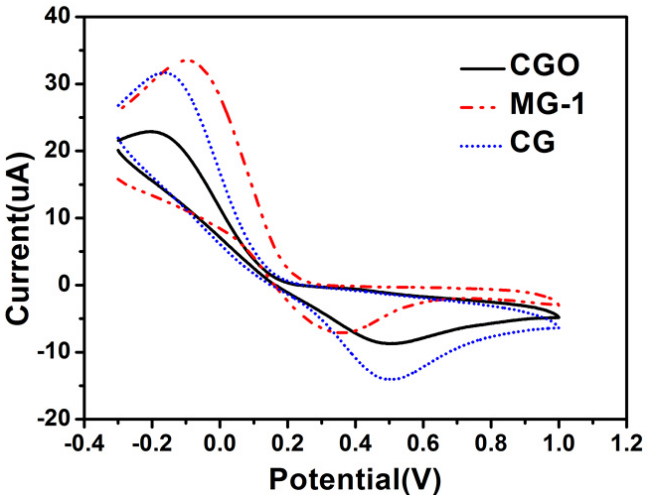

Figure S-6. Cyclic voltammograms in solution containing 5 mM of $[K_3(FeCN)_6]$, in 0.1 M KCl at scanning rate 5 mV/s : *CGO* graphene oxide, *CG* graphene prepared by chemical reduction method and *MG-1* contact with mixed microorganisms fro 24 hours.

Electrochemical Characteristics of N-doped Graphene. As shown in Figure S-6, the intensity of peaks of C*GO* was significantly lower than that of *MG-1* and *CG*, which was due to high surface charge caused by a great deal of oxygen-containing functional groups and then, ferricyanide ions were repelled at the electrode surface, which limited the electrochemical reactions.[6] To the contrary, there were higher peaks for MG-1 and *CG* indicating that oxygen-containing functional groups were reduced obviously. What's more, the peak current of *MG-1*

was slightly higher than that of *CG* and the peak separation potential is ΔEp = 0.43V which is much lower than that of *CG* and *CGO*, indicating that *MG-1* possessed a better electrochemical properties.

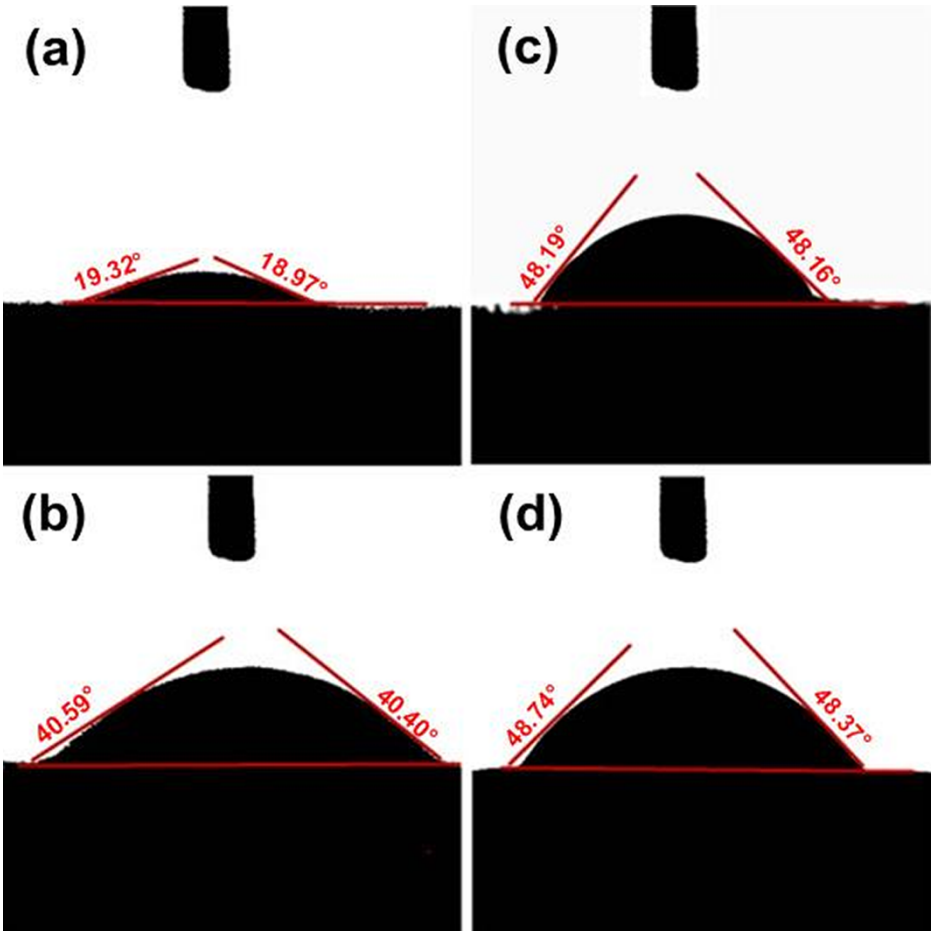

Figure S-7. water contact angle images after 4 seconds: (a) *MG-1*, contact with mixed microorganisms for 24 hours; (b) *CG*, graphene prepared by chemical reduction method; (c) blank control, 1.2 μm hydrophilic membrane; (d) *CGO*, oxide graphene.

Water Contact Angle Analysis: The contact angle of MG-1 is smallest in all four samples indicating that water-dispersity of as-prepared N-doped graphene obviously increases.[7]

METHODS

Experimental Method of Antibacterial Experiment: The three materials (*CG, CGO, MG-1*) of 0.01g were dispersed uniformly by ultrasonic dispersion for 2 hours and then vacuum filtration with 1.2 um filter membrane. The membrane covered with uniform material was clipped into rectangles with length of 5 μm. Before the antibacterial experiment, the rectangles were sterilized by ultraviolet irradiation for 20 min and finally the surface with material was affixed on agar plate before which *Escherichia coli* or *Staphylococcus aureus* was applied to the plate.

Experimental Method of Water Contact Angle: As the above method, the membrane covered with uniform material were prepared and flattened to complete the experiment of water contact angle.

Cyclic Voltammetry (*CV*) Experiments: *CV* experiments were conducted with platinum electrodes. Firstly, the platinum electrodes were polished to form a mirror surface with alumina slurry. Secondly, platinum electrodes were rinsed with deionized water and dried by nitrogen. Finally, the 10 μL suspension (sample of 10 mg, isopropyl alcohol of 5 mL and a drop of Nafion.) was dropped onto the electrodes and dried at room temperature using a 5 mM solution of potassium ferricyanide [$K_3(FeCN)_6$] containing 0.1 M KCl as electrolyte. Scanning rate is 5 mV/s and scanning scope is from -0.5 V to 1.0 V.

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