Antibacterial activities and mechanisms of fluorinated graphene and guanidinemodified graphene

Xu Wang, ^aPengLu, ^a Yuan Li, ^aHuining Xiao*, ^a, Xiangyang Liu*, ^b

^a Department of Chemical Engineering, University of New Brunswick, Fredericton, NB
E3B 5A3, Canada.

^b State Key Laboratory of Polymer Materials Engineering, College of Polymer Science and Engineering, Sichuan University, Chengdu, Sichuan, 610065, P.R. China;

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Preparation of GO

GO was prepared by the modification of Hummers's method from flake graphite (average particle diameter of 4µm, 99.95% purity, Qingdao Tianhe Graphite Co. Ltd., Qingdao, China)^{1,2}.5 g of graphite and 3.75 g of NaNO3(A.R.) wereplaced in a flask. Then, 375 mL of H2SO4(A.R.) was added withstirring in an ice-water bath, and 22.5 g of KMnO4(A.R.) wereslowly added over about 1 h. Stirring was continued for 2 h in the ice-water bath. After the mixture was stirred vigorously for5 days at room temperature, 700 mL of 5 wt % H2SO4aqueoussolution was added over about 1 h with stirring, and the temperature was kept at 98 °C. The resultant mixture was further stirredfor 2 h at 98 °C. The temperature was reduced to 60 °C, 15 mLof H2O2(30 wt % aqueous solution) was added, and the mixture as stirred for 2 h at room temperature. To remove the ions of oxidant and other inorganic impurity the resultant mixture was purified by repeating the followingprocedure cycle 2 times: centrifugation, removal of the supernatant liquid, addition of 2 L of a mixed aqueous solution of 3 wt% H₂SO₄/0.5 wt % H₂O₂to the bottom solid, and dispersing thesolid using vigorous stirring and bath ultrasonication for 30 minat a power of 140 W. Then a similar procedure was repeated: two times using 3 wt % HCl aqueous solution (2 L) and one timeusing H2O (2 L). The final resultant water solution was dialyzed for two weeks to further remove theremaining HCl acid and other impurity. After centrifugation, water in the resultant solide was removed by freeze drying for 48 h.

Preparation of PHGH

Equimolar amounts of hexamethylenediamine (Sigma-Aldrich) and guanidine hydrochloride (Sigma-Aldrich) were mixed in a round-bottomed three-necked flask, which is equipped with a mechanical stirrer and vacuum system. The mixture reacted at 100 °C for 60 min, and then at 170 °C for a certain time. During the reaction, by-product ammonia is neutralized by bubbling through aqueous HCl. Thereafter the reaction continued on the condition of removing ammonia by vacuum system. At the end of reaction, the slightly yellow, viscous liquid solidifies upon cooling giving PHGH samples



Figure S1. SEM images of graphene oxide sheets (a and b); photographs of spongy graphene oxide (c) and it SEM image (d).



Figure S2. SEM images of reduced spongy graphene oxide (RSGO)



Figure S3.TGA (left) and DTA (right) linesof graphene derivatives: GO, reduced GO, FG, and PHGH-G



Figure S4. Antibacterial activity of PHGH-G against E. coli ATTCC-11229.