

# **Controllable defluorination of fluorinated graphene and weakening of strong covalent C-F bonding under the action of nucleophilic dipolar solvent**

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## Synthesis of the spongy graphene oxide

GO was prepared by the modification of Hummers's method from flake graphite (average particle diameter of 4  $\mu\text{m}$ , 99.95% purity, Qingdao Tianhe Graphite Co. Ltd., Qingdao, China). 5 g of graphite and 3.75 g of  $\text{NaNO}_3$  (A.R.) were placed in a flask. Then, 375 mL of  $\text{H}_2\text{SO}_4$ (A.R.) was added with stirring in an ice-water bath, and 22.5 g of  $\text{KMnO}_4$ (A.R.) were slowly added over about 1 h. Stirring was continued for 2 h in the ice-water bath. After the mixture was stirred vigorously for 5 days at room temperature, 700 mL of 5 wt %  $\text{H}_2\text{SO}_4$  aqueous solution was added over about 1 h with stirring, and the temperature was kept at 98  $^\circ\text{C}$ . The resultant mixture was further stirred for 2 h at 98  $^\circ\text{C}$ . The temperature was reduced to 60  $^\circ\text{C}$ , 15 mL of  $\text{H}_2\text{O}_2$  (30 wt % aqueous solution) was added, and the mixture was 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 following procedure cycle 2 times: centrifugation, removal of the supernatant liquid, addition of 2 L of a mixed aqueous solution of 3 wt %  $\text{H}_2\text{SO}_4$ /0.5 wt %  $\text{H}_2\text{O}_2$  to the bottom solid, and dispersing the solid using vigorous stirring and bath ultrasonication for 30 min at a power of 140 W. Then a similar procedure was repeated: two times using 3 wt %  $\text{HCl}$  aqueous solution (2 L) and one time using  $\text{H}_2\text{O}$  (2 L). The final resultant water solution was dialyzed for two weeks to further remove the remaining  $\text{HCl}$  acid and other impurity. After centrifugation, water in the resultant solide was removed by freeze drying for 48 h.

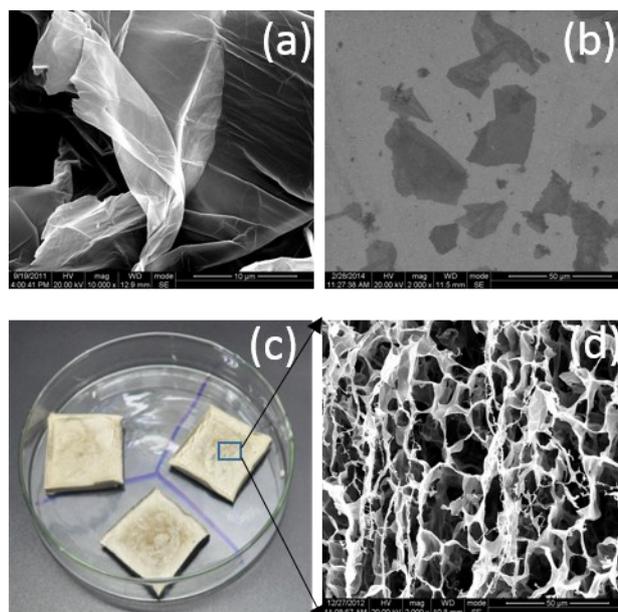
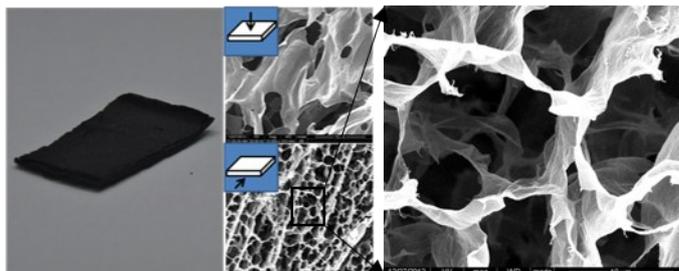
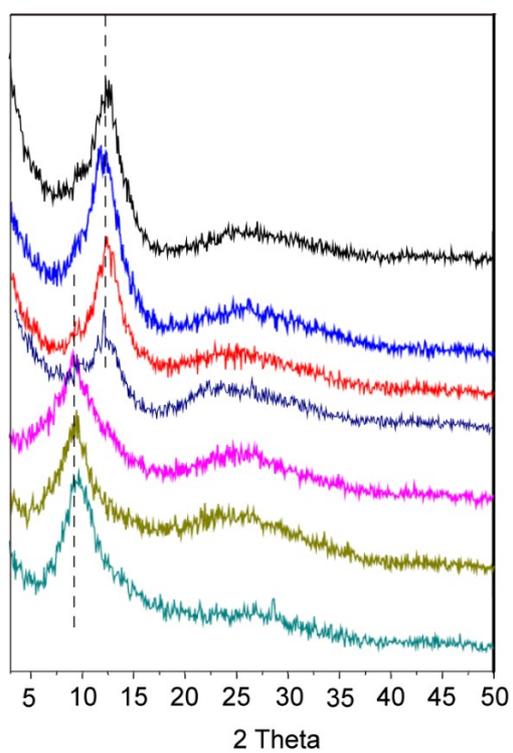


Figure S1. SEM images of graphene oxide sheets (a and b); photographs of spongy graphene oxide (c) and its SEM image (d).

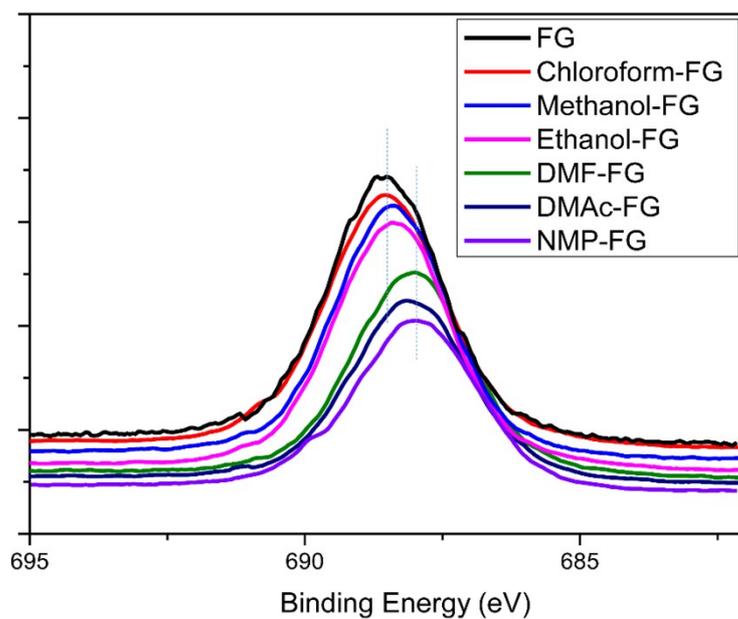
A 2 wt% GO suspension in water was prepared. The aqueous solution of GO was loaded into square aluminum molds. The specimens were then plunged into liquid nitrogen, then dried in a freeze drier for two days. Finally, the samples were further dried at 80  $^\circ\text{C}$  for two hours. The spongy graphene oxide GO was prepared, as shown in Figure S1



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



**Figure S3.** XRD spectra of FG samples dispersed by different solvents: FG, Chloroform-FG, Ethanol-FG, Methanol-FG, DMF-FG, DMAc-FG and NMP-FG (top to bottom).



**Figure S4.** XPS F 1s spectra of FG and samples treated by solvent.

The F 1s binding energy peak position of NMP-FG, DMF-FG and DMAc-FG shifted to a lower energy from 688.5 eV to 687.9 eV in comparison with that of pristine FG, chloroform-FG, ethanol-FG and methanol-FG, and the intensity of peaks decreases significantly, which indicate the reduction and weakening effect of DMF, DMAc, and NMP on FG.