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 μm, 99.95% purity, Qingdao Tianhe Graphite Co. Ltd., Qingdao, China). 5 g of graphite and 3.75 g of NaNO3 (A.R.) were placed in a flask. Then, 375 mL of H2SO4(A.R.) was added with stirring in an ice-water bath, and 22.5 g of KMnO4(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 % H2SO4 aqueous solution was added over about 1 h with stirring, and the temperature was kept at 98 °C. The resultant mixture was further stirred for 2 h at 98 °C. The temperature was reduced to 60 °C, 15 mL of H2O2 (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 % H2SO4/0.5 wt % H2O2 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 % HCl aqueous solution (2 L) and one time using H2O (2 L). The final resultant water solution was dialyzed for two weeks to further remove the remaining 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 it 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 °C for two hour. 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 disposed 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.