Supporting Information

Novel Method for the Fabrication of Flexible Film with Oriented Arrays of Graphene in Poly(vinylidene fluoride-co-hexafluoropropylene) with Low Dielectric Loss

Wangshu Tong[a], Yihe Zhang[*a], Li Yu[a], Xinglong Luan[a], Qi An[a], Qian Zhang[a], Fengzhu Lv[a], Paul K. Chu[b], Bo Shen[a] and Zhilei Zhang[a]

[a]National Laboratory of Mineral Materials, School of Materials Science and Technology, China University of Geosciences, Beijing 100083, China

[b]Department of Physics & Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong 999077, China

*E-mail: zyh@cugb.edu.cn. Phone: 86-10-82323433.

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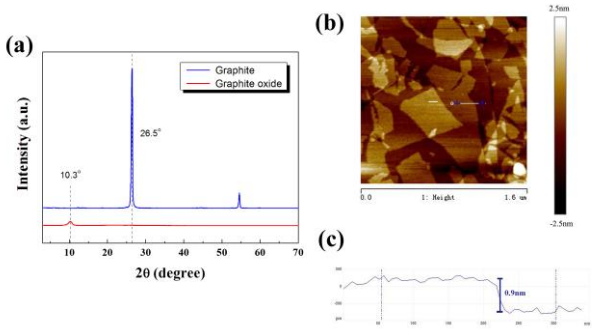
1. Fabrication of rGO/PVDF-HFP composite films with repeated spin-coating (SC) method

rGO/PVDF-HFP composite films were prepared by a spin-coating process. Typical procedures were as follows and are shown in Scheme 1: 1) the as-prepared GO/PVDF-HFP solution was spin-coated on a glass substrate with spin speed of 1400 rpm; 2) the glass substrate was kept in an oven at 80^{o}C for 1h to evaporate the solvent slowly and GO plates were fixed in PVDF-HFP; 3) GO/PVDF-HFP solution was spin-coated on the same glass substrate used in 1) and then a second layer of GO/PVDF-HFP on the first one was obtained; 4) steps 1 and 2 were repeated five times and a GO/PVDF-HFP film with six layers was obtained; and 5) the GO/PVDF-HFP film with six layers was kept in an oven at 80 ^{o}C for 3h and then at 220^{o}C for 1 h. Finally, rGO/PVDF-HFP composite films were obtained.

2. Fabrication of rGO/PVDF-HFP composite films with traditional drop-coating (TDC) method

The as-prepared GO/PVDF-HFP solution was drop cast on a glass plate and kept in an oven at 80 ^{o}C for 3 h to evaporate the solvent slowly to obtain the GO/PVDF-HFP composite films. Then the film was kept in an oven at 220^{o}C for 1 h and rGO/PVDF-HFP composite films (one layer) were obtained by TDC.

3. Morphology and structure of GO



Figuer S1. (a) WAXRD patterns of graphite and GO. (b) and (c) AFM of GO.

GO was synthesized from flake graphite by the Hummers method, and its structures were characterized by XRD (Figure S1a), AFM (Figure S1b and c), and raman spectra (Figure S2). The XRD pattern of GO only shows a peak at 2θ = 10.3^{o}, corresponding to the inter-planar spacing of 8.6Å. The XRD patterns of GO reveal the transformation of the interlayer spacing from 3.4 Å to 8.6 Å, indicating the incorporation of various functional groups in the sheet of

graphite. The expanded smooth XRD peak of the GO compared with the sharp peak of graphite indicates the poor crystallinity of the oxidized graphite.[1] AFM images show that the GO is a flake structure with about 0.5-1μm of the dimensions of GO diameter, and about 1nm in thickness (Figure S1c).

4. Raman spectra of GO

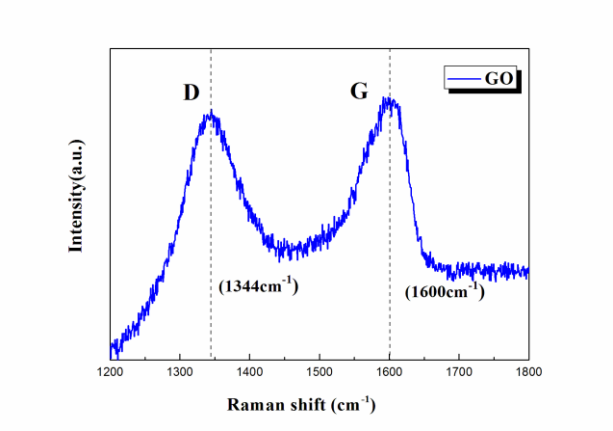


Figure S2. Raman spectra of GO.

The typical features of the raman spectra are the D band (about 1344 cm^{-1}) assigning to the disorder-induced D band from the multiple photo scattering of defects or amorphous carbon and the G band (about 1600 cm^{-1}) assigning to the stretching of conjugated double bonds corresponding to sp^2 hybridization.[2-3] The intensity ratio of D and G bands (I_D/I_G) is often taken to characterize the number of defects and impurities in GO.[4] The I_D/I_G=0.83 suggests that the GO was successfully oxidized with function groups defecting the ordered structure of sp^2 hybridization.

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