

## Electronic Supplementary Information

### **Synthesis and characterization of a new hierarchical reinforcement by chemically grafting graphene oxide onto carbon fiber**

Yibin Li <sup>a,\*</sup>, Qingyu Peng <sup>a</sup>, Xiaodong He <sup>a</sup>, PingAn Hu <sup>b</sup>, Chao Wang <sup>a</sup>, Yuanyuan Shang <sup>a</sup>, Rongguo Wang <sup>a</sup>, Weicheng Jiao <sup>a</sup>, Hongzhen Lv <sup>a</sup>

<sup>a</sup> Center for Composite Materials and Structures, Harbin Institute of Technology, Harbin 150080, P. R. China

<sup>b</sup> Research Centre for Micro/Nanotechnology, Harbin Institute of Technology, Harbin 150080, P. R. China

\*Corresponding email: [liyibin@hit.edu.cn](mailto:liyibin@hit.edu.cn)

## Materials

The natural graphite flakes is bought from Aldrich Inc.. Carbon fiber used is T300B-3000-40B (Toray Industries, Inc.). The amino is provided with Poly-(amidoamine) (PAMAM, Aldrich Inc.) dendrimers (Generation 0) with ethylenediamine core and amino surface groups, as shown in Fig.S1. The coupling agent used is N-[(dimethylamino)-1H-1, 2, 3-triazolo [4,5,6] pyridin-1-ylmethylene]-N-methylmethanaminium hexafluorophosphate N-oxide(HATU, GL Biochem Ltd) All other chemicals were purchased from Tianjin Bodi Organic Chemicals Co. Ltd.

## GO-CF hierachical reinforcement preparation

Graphite oxide was synthesized from natural graphite flakes using a modified Hummer's method. Graphite oxide was then fully exfoliated in DMF to produce suspension of graphene oxide (GO) sheet for grafting. The carbon fibers were desized in acetone at 60 °C for 48 h and then oxidized in concentrated nitric acid at 100 °C for 2 h. Subsequently, the carbon fibers were taken out and washed several times with de-ionized water until the pH value of the wash water was 7, followed by drying in a vacuum furnace.  $3 \times 10^{-5}$  mol PAMAM and a small amount of HATU were added into 30ml dry dimethylformamide (DMF) and dissolved with ultrasonic for several minutes. The carboxyl-functionalized carbon fibers were immersed into the DMF solution at room temperature for 4h in order to complete the amino-functionalization. After that, the amino-functionalized carbon fibers were washed with de-ionized water and dried under vacuum. Then, the amino-functionalized carbon fibers were immersed into the suspension of grapheme oxide and some HATU in 30ml DMF at room temperature for 4 h. The obtained fibers were rinsed in DMF.

## Characterization

### GO/CF reinforcement characterization

Imaging of GO sheets by AFM (DIMENSION icon, BRUKER, GER) was performed in contact

mode using BRUKER SNL-10 type silicon-tip on nitride lever cantilevers.

The surface morphologies of GO/CF reinforcement were observed by field emission scanning electron microscope (FESEM, model S-4300, Hitachi, Japan).

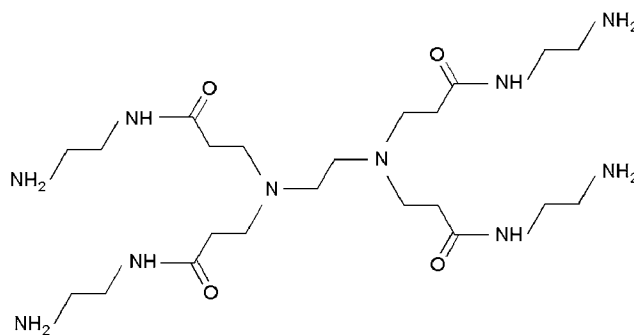
The nanoscopic-scale characterization of GO sheets was performed using a field emission transmission electron microscope (Tecnai G<sup>2</sup> F30, FEI, US),

The chemical reactions were confirmed by X-ray photoelectron spectroscopy (XPS, Model K-Alpha, Thermofisher Scientific Company, US) using a monochromated Al K $\alpha$  source and a pass energy of 50 eV at a base pressure of  $1 \times 10^{-8}$  mbar.

A Dynamic Contact Analyzer (DCAT21, GER) was chosen to evaluate the contact angles between test liquids and single fibers. The testing liquids are water and diiodomethane, whose surface energies are known. The dispersive and polar components can be easily determined by solving the following Eq.(1)

$$\gamma_l^T (1 + \cos \theta) = 2(\gamma_l^d \gamma_s^d)^{1/2} + 2(\gamma_l^p \gamma_s^p)^{1/2} \quad (1)$$

where  $\gamma_l^T$ ,  $\gamma_l^d$  and  $\gamma_l^p$  are the surface tension of immersion liquid, its dispersive and polar component, respectively. The  $\gamma_l^d$  of water is 22.1 mJ/m<sup>2</sup>. The  $\gamma_l^p$  of water is 50.7 mJ/m<sup>2</sup>. The  $\gamma_l^d$  of diiodomethane is 50.8 mJ/m<sup>2</sup>. The  $\gamma_l^p$  of diiodomethane is 0 mJ/m<sup>2</sup>.



**Figure S1** 0 generation PAMAM molecular structure.