

**Supplementary Information for**  
**Liquid Crystal Self-templating Approach to Ultrastrong and**  
**Tough Biomimic Composites**

*Xiaozhen Hu, Zhen Xu, Zheng Liu, and Chao Gao\**

MOE Key Laboratory of Macromolecular Synthesis and Functionalization,  
Department of Polymer Science and Engineering, Zhejiang University, 38 Zheda  
Road, Hangzhou 310027, P. R. China.

\*Corresponding author. E-mail: [chaogao@zju.edu.cn](mailto:chaogao@zju.edu.cn)

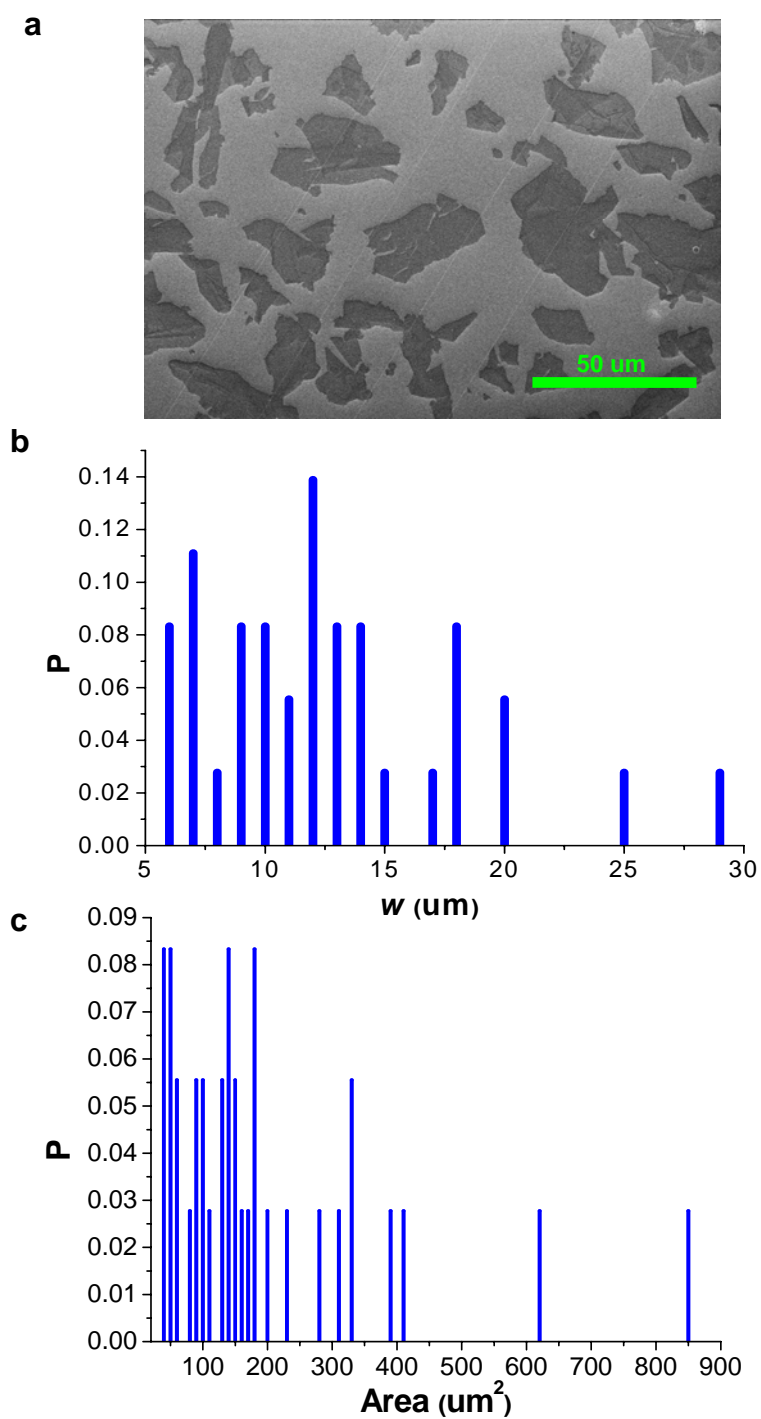
## Supplementary Methods

**Materials.** Graphite powder (~500  $\mu\text{m}$ ) was obtained from Qingdao Henglide Graphite Co., Ltd. Concentrated  $\text{H}_2\text{SO}_4$  (98%),  $\text{KMnO}_4$ ,  $\text{P}_2\text{O}_5$ ,  $\text{H}_2\text{O}_2$  (30%) solution,  $\text{K}_2\text{S}_2\text{O}_8$ ,  $\text{CaCl}_2$ , glutaraldehyde (25%), acetic acid,  $\text{KOH}$ , and hydrazine monohydrate were purchased from Sinopharm Chemical Reagent Co., Ltd. and used as received.  $\text{Vc}$ , L-tryptophan, and HI acid (~40%) were purchased from Aladdin and used as received. Potassium methylate solution in methanol ( $\text{CH}_3\text{OK}$ , 25wt%) from Fluka were used as received. Dioxane were distilled before use. Hyperbranched polyglycerol (HPG) was synthesized by ring-opening anionic polymerization with potassium methylate as initiator according to the previous protocol<sup>53</sup>.

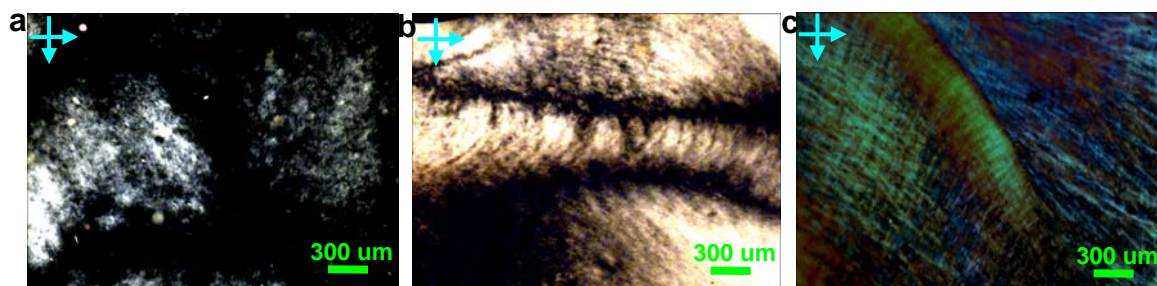
**Preparation of GGO.** GGO was synthesized by oxidation of graphite (500  $\mu\text{m}$ ) according to ref 30.

**Preparation of RGG-HPG fibers.** RGG-HPG fibers were obtained by reducing GGO-HPG fibers. As a typical example, the GGO-HPG fibers were immersed into the aqueous solution of  $\text{Vc}$  (5.8  $\text{mg mL}^{-1}$ ) and  $\text{KOH}$  (1.8  $\text{mg mL}^{-1}$ ) and kept at 90  $^\circ\text{C}$  for 12 h. After cooling to room temperature, the fibers were dried at 80 $^\circ\text{C}$  under vacuum for 12 h.

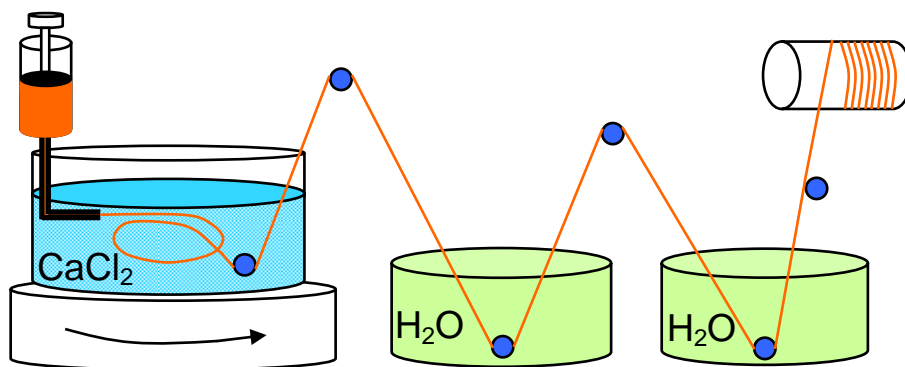
## Supplementary Figures



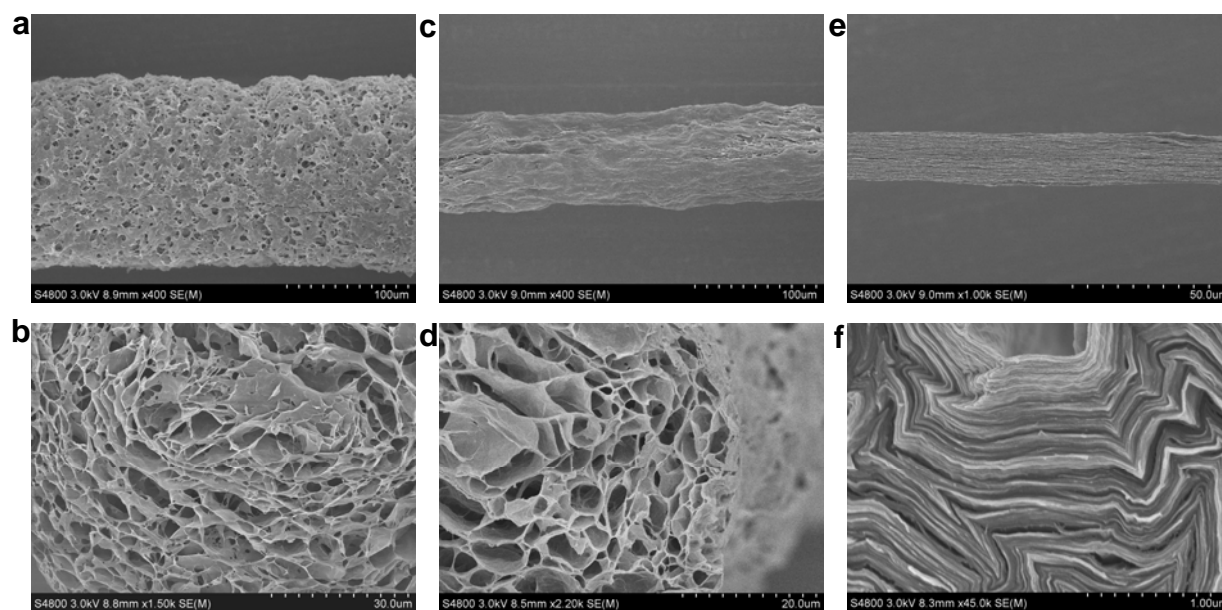
**Supplementary Figure S1 | Size and distribution of the as-prepared GGO sheets.** (a) SEM image of GGO sheets deposited on silicon. (b,c) The width and area distribution (P) of GGO sheets counted from their SEM image shown in a.



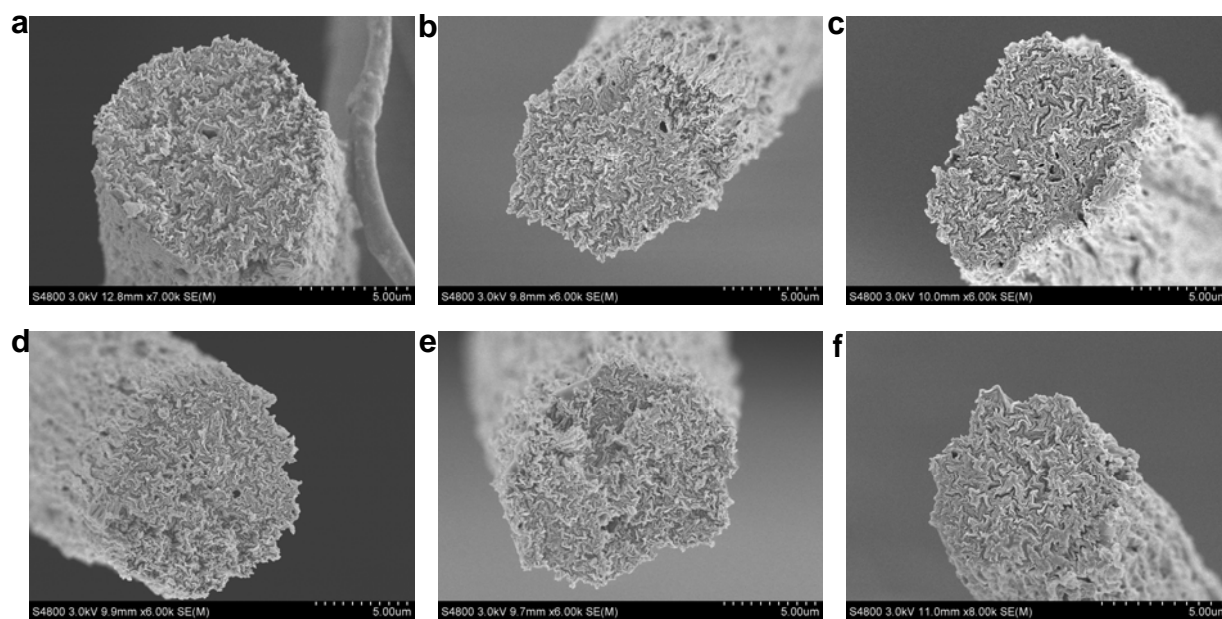
**Supplementary Figure S2 | POM images of GGO aqueous LCs loaded in the planar cells. (a)  $0.5 \text{ mg mL}^{-1}$ , (b)  $1 \text{ mg mL}^{-1}$ , and (c)  $4 \text{ mg mL}^{-1}$ .**



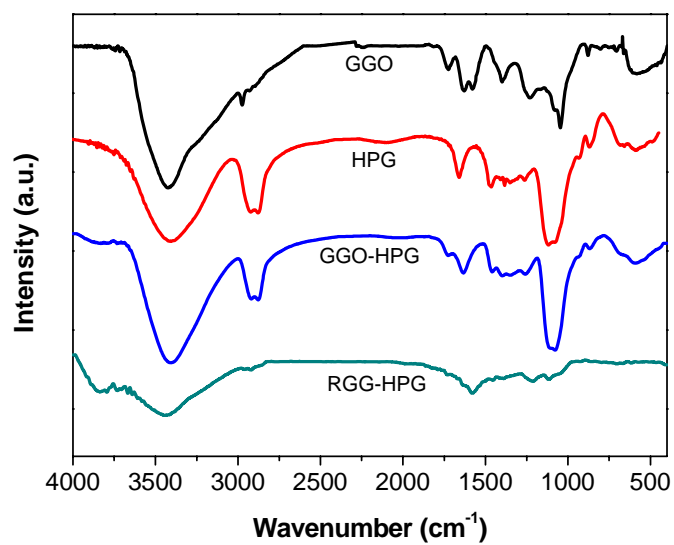
**Supplementary Figure S3 | Spinning apparatus for GGO-HPG fibres from GGO-HPG LCs in water.**



**Supplementary Figure S4 | SEM images of GGO-HPG fibers for observation of the drying procedure.** (a) The surface of a gel fiber as soon as spun into the coagulation bath. (b) The cross-section of gel fiber. (c,d) Morphology of gel fiber after coagulated for 3 minutes. (e,f) The structure of finally dried fiber.

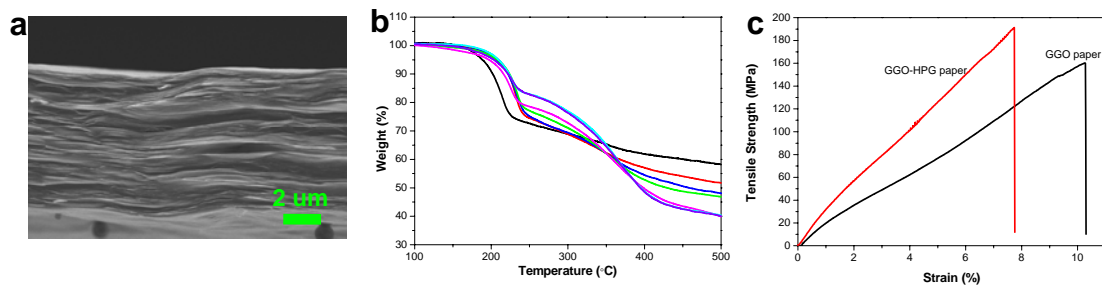


**Supplementary Figure S5 | SEM images of cross-section of GGO-HPG fibers.**

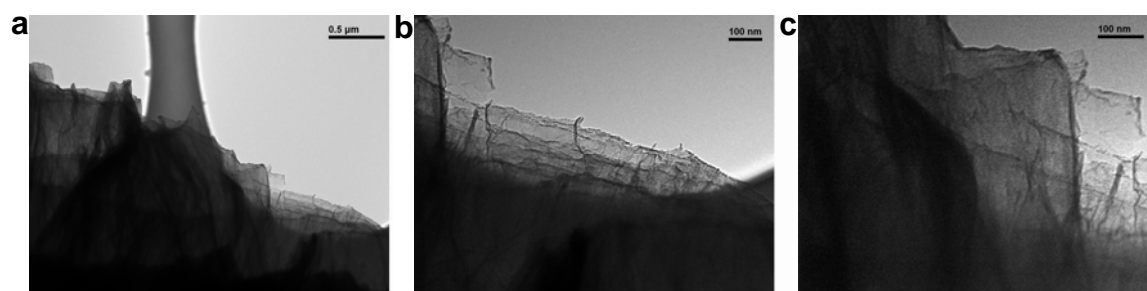


**Supplementary Figure S6 | FT-IR spectra of GGO, HPG, GGO-HPG, and RGG-HPG fibers.**

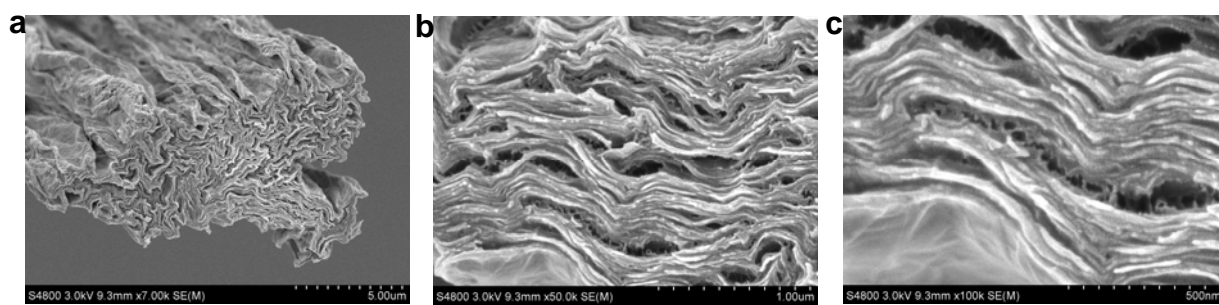




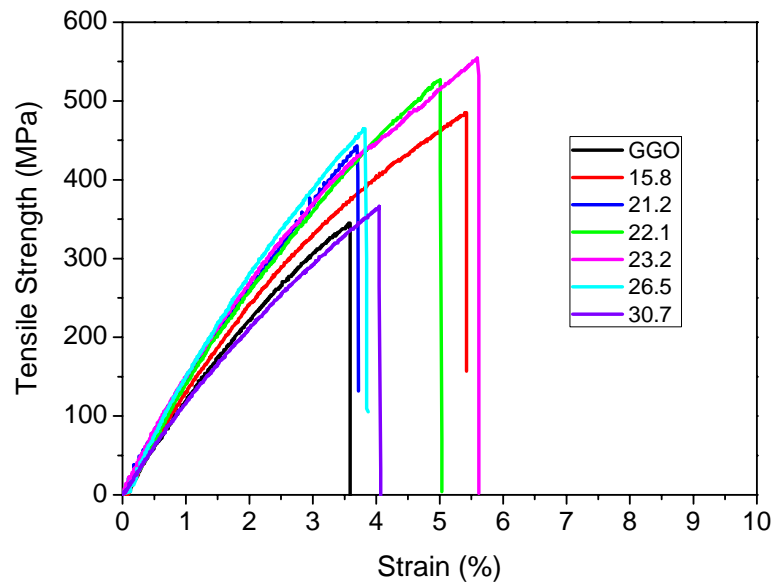
**Supplementary Figure S7 | Morphology and properties of GGO and GGO-HPG papers.** (a) SEM image of layered structure of GGO-HPG paper. (b) TGA curves of different GGO-HPG samples. (c) Typical stress-strain curves of GGO and GGO-HPG papers.



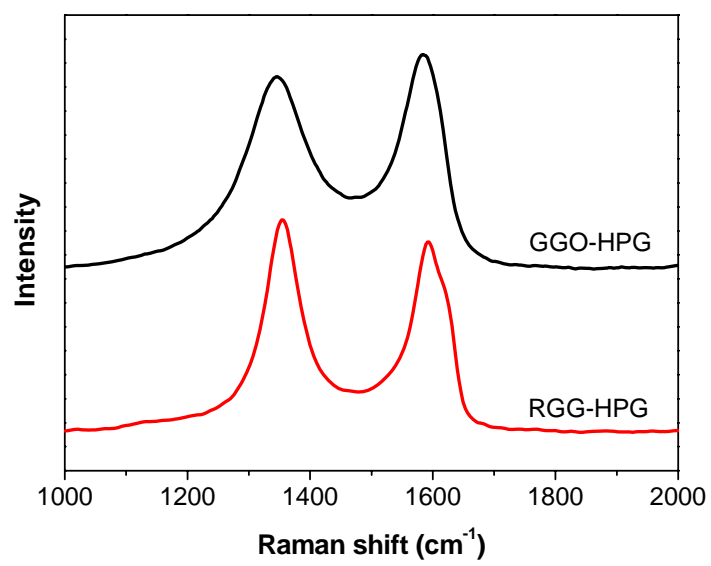
**Supplementary Figure S8 | TEM images of fracture section of GGO-HPG fibers at different magnification.**



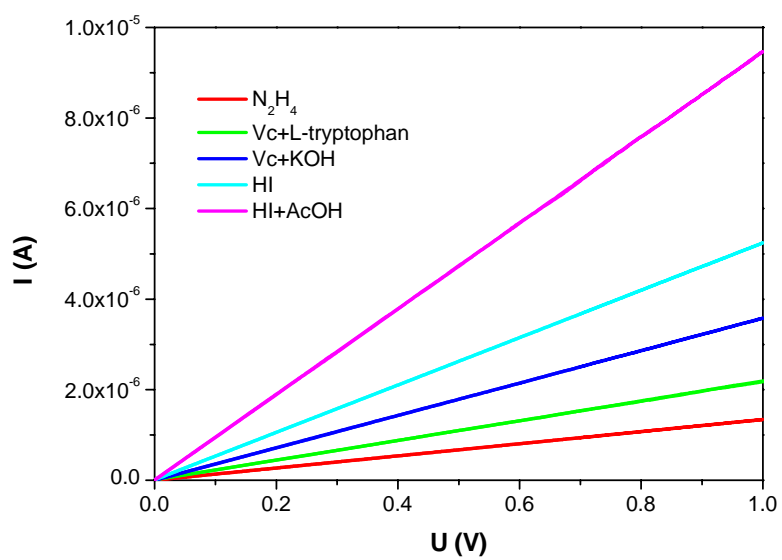
**Supplementary Figure S9 | SEM images of GGO-HPG-GA fibers at different magnification.**



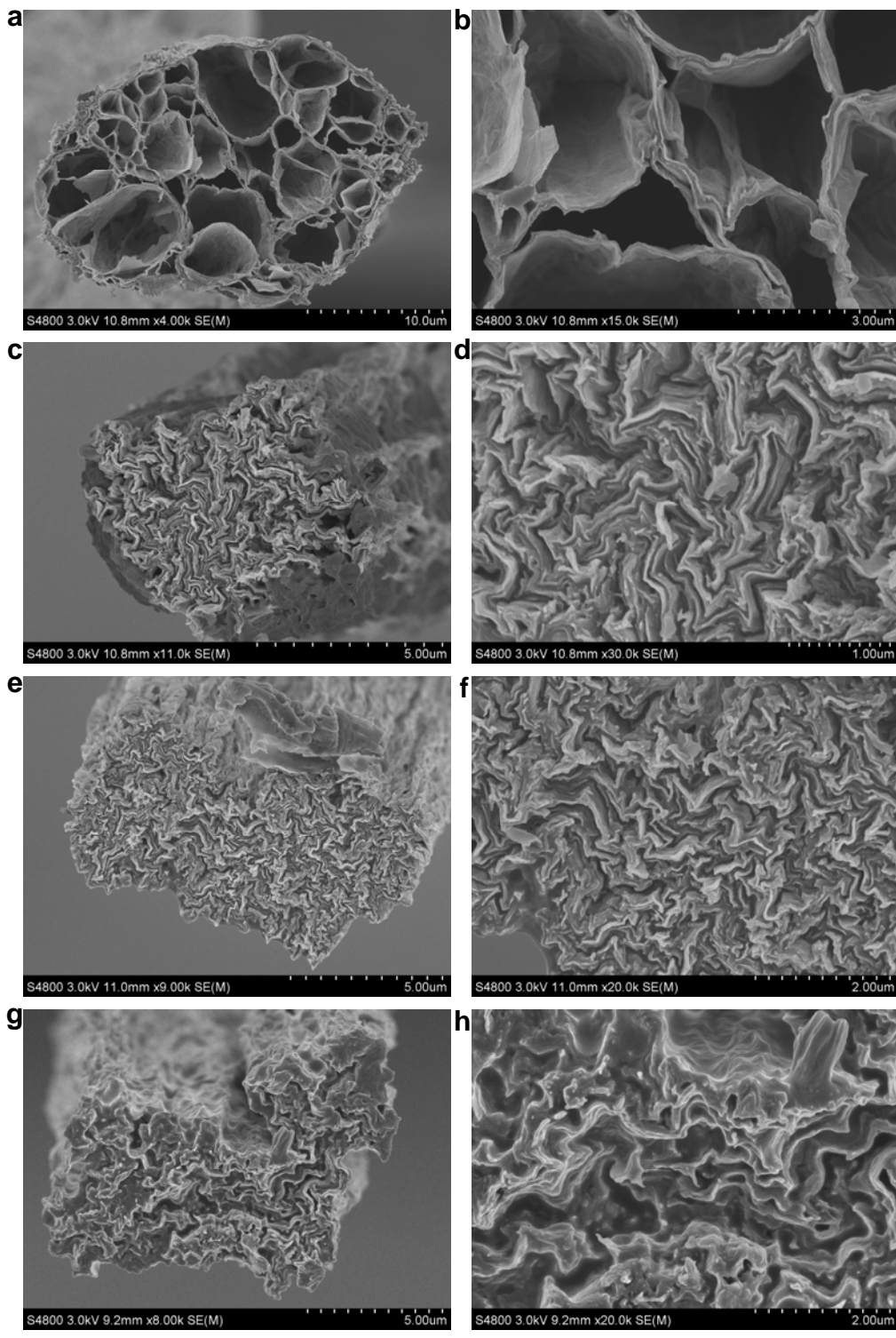
**Supplementary Figure S10 | Mechanical properties of GGO-HPG composites with different HPG contents.** Typical stress-strain curves of GGO-HPG fibers with different HPG contents.

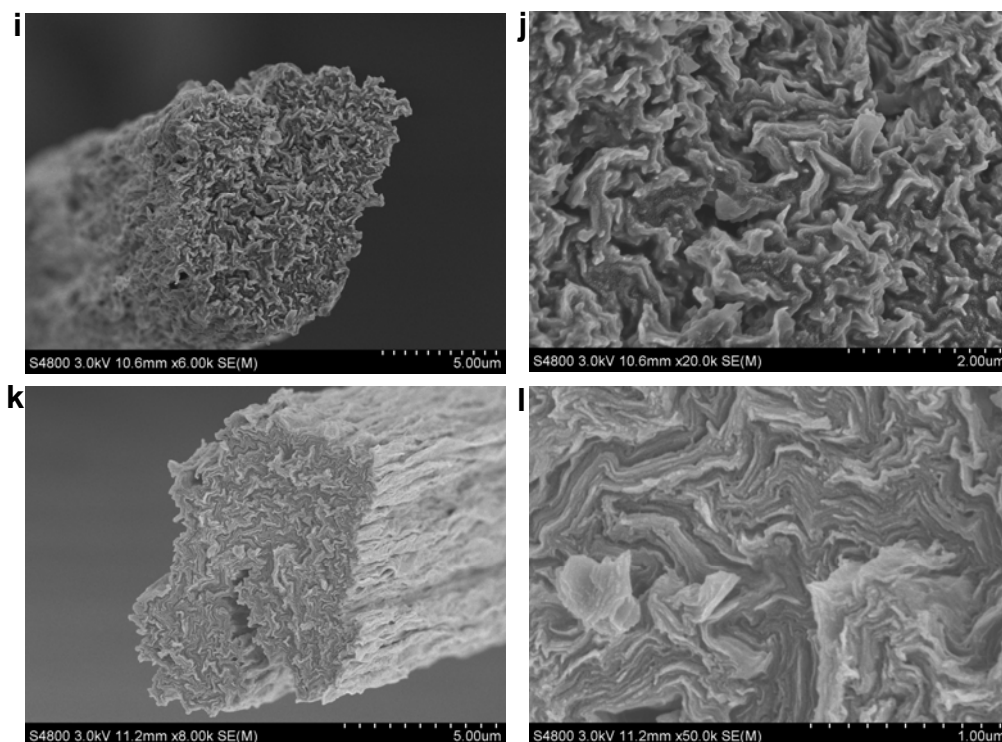


**Supplementary Figure S11 | Raman spectra of GGO-HPG, RGG-HPG fibres.**



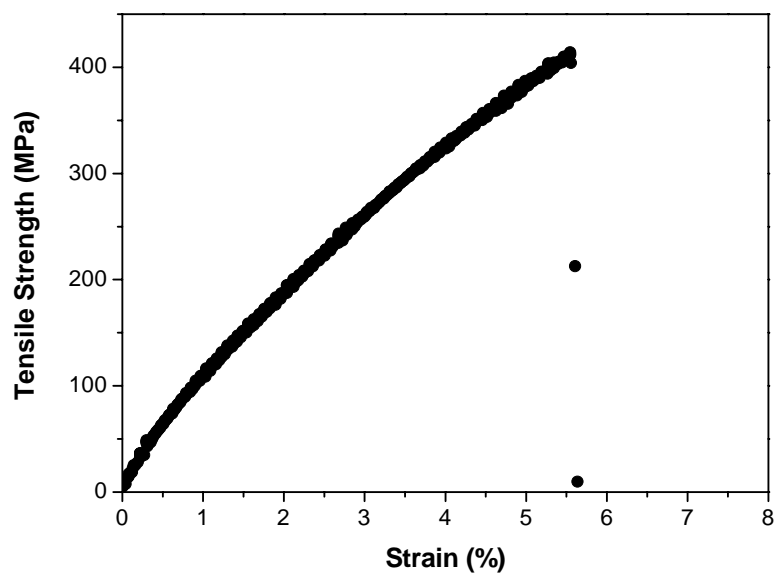
Supplementary Figure S12 | Typical I-V curves of RGG-HPG fibers.



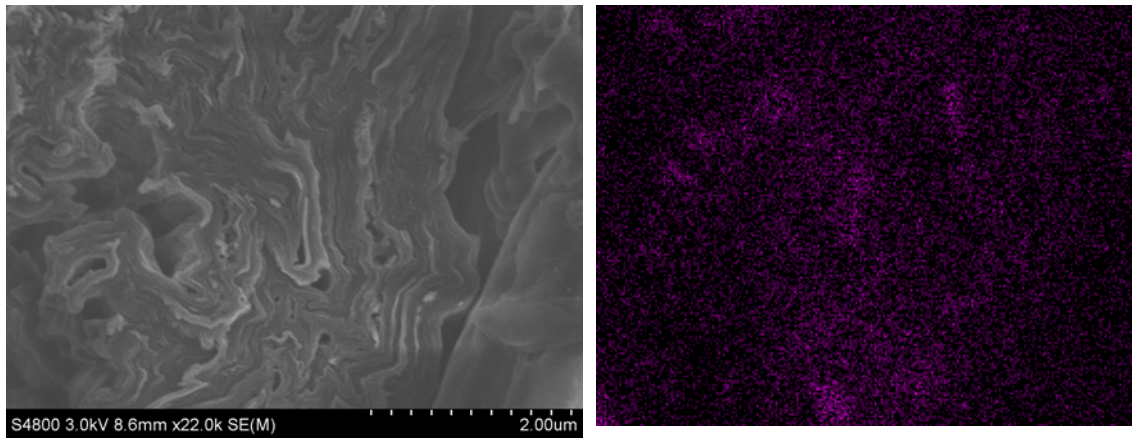


**Supplementary Figure S13 | SEM images of RGG-HPG fibers reduced by various chemicals and heat treatment.** (a,b) RGG-HPG fibers reduced by  $N_2H_4$ . (c,d) RGG-HPG fibers reduced by the previous Vc method. (e,f) RGG-HPG fibers reduced by our modified Vc method. (g,h) RGG-HPG fibers reduced by HI. (i,j) RGG-HPG fibers reduced by the vapor of HI and AcOH. (k,l) RGG-HPG fibers obtained by thermal reduction at 200 °C.

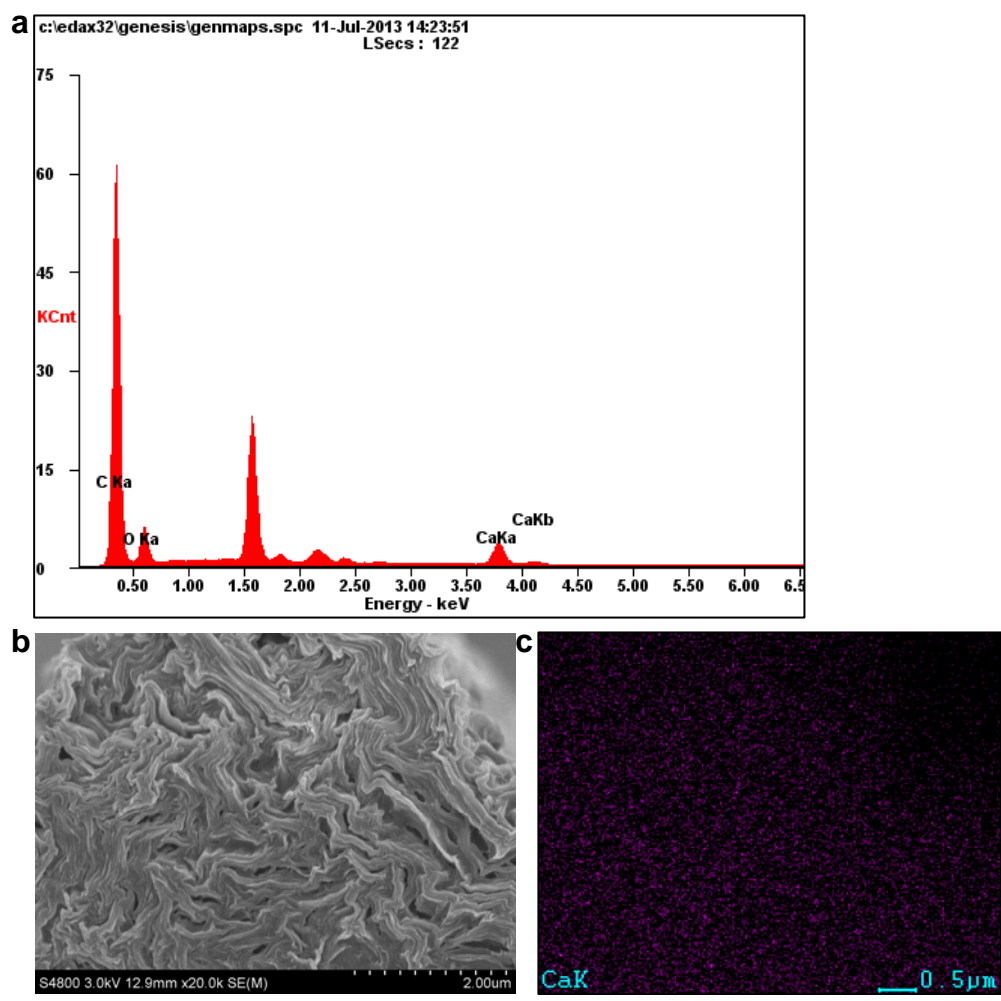




**Supplementary Figure S14 | Typical stress-strain curves of GGO-PVA fibers.**



**Supplementary Figure S15 | SEM image and Ag-mapping of GGO-HPG-Ag fibers.**



**Supplementary Figure S16** | (a) EDX spectrum of GGO-HPG fibers. SEM images of cross-section of GGO-HPG fibers (b) and their corresponding Ca-element mapping images (c).