

Supplementary Information

**A Multiscale Hydrothermal Carbon Layer Modified Carbon Fiber for
Composite Fabrication**

Xianfeng Xi^{a, b}, Yousi Chen^a, Jie Wang^{a, b}, Yaoyao Lia^{a, b}, Xiangdong Shao^c, Liu
He^{a *}, Qing Huang^{a *}, Xueliang Pei^{a *}

^a Ningbo institute of Material Technology and Engineering, Chinese Academy of
Sciences, Ningbo Zhejiang 315201, China

^b University of Chinese Academy of Sciences, 19 A Yuquan Rd., Shijingshan District,
Beijing 100049, China

^c Zhejiang Zhongtian Fluorine Silicone Material Co., Ltd, 20 North Huaying Rd.,
Kecheng District, Quzhou 324004, China

* To whom correspondence should be addressed: heliu@nimte.ac.cn (Liu He*);
huangqing@nimte.ac.cn (Qing Huang*) peixueliang@nimte.ac.cn (Xueliang Pei*)

1. The experiment equipment and phenomena observed by eye directly

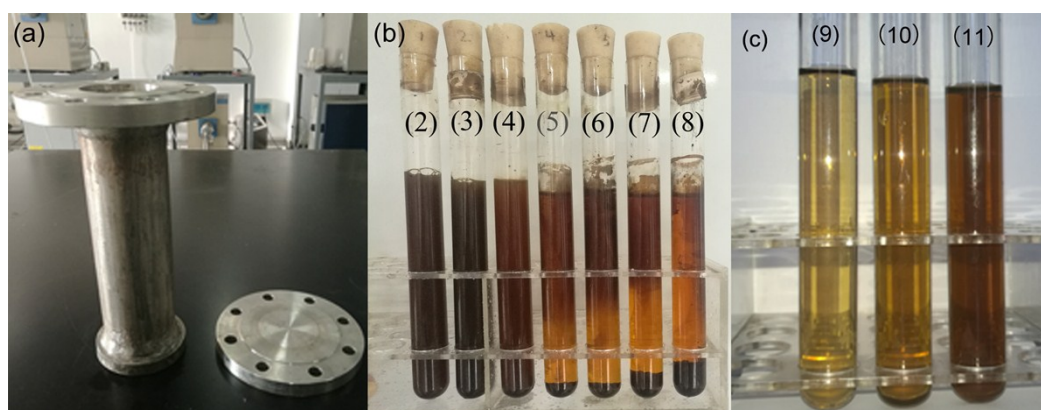


Fig. S1. Hydrothermal treatment equipments for the MHTCL experiment.

The samples in the paper were Hydrothermal treated in the sealed test tubes as same as tubes in image (b), which were sealed in stainless autoclave (a).

The test tubes are 25 cm long with a diameter of 2 cm. The stainless autoclave is 40 cm long with an inner diameter of 7.8 cm. The seven test tubes were treated at one time. Image (b) shows the residual solutions after hydrothermal treatment with the CFs: The glucose solution in the glass tube (2-8) is: (2) 2 gL⁻¹, (3) 5 gL⁻¹, (4) 10 gL⁻¹, (5) 15 gL⁻¹, (6) 20 gL⁻¹, (7) 30 gL⁻¹, (8) 40 gL⁻¹, respectively. The image (b) was taken after standing 24 hours. Image (c) shows the low-concentration glucose solutions after hydrothermal treated without CFs, whose glucose concentration is: (9) 2 gL⁻¹, (10) 5 gL⁻¹, (11) 10 gL⁻¹, respectively.

2. Colourful MHTCL CFs

After hydrothermal treatment, the CFs became colourful, as shown in Fig. S2. It appeared golden in the photograph, but a part of the CFs in the bundles is blue in fact.

It is a convenient way to estimate whether the MHTCL has covered on the CFs by

observing the change of the CFs colours. The sample CF2 is a little colourful; sample CF3 is more colourful; CF5, CF6, CF7 and CF8 are obviously colourful, as shown in the photograph.



Fig. S2. The photograph of the MHTCL CFs (sample CF5) and unsized CFs in the sunshine

3. Topography of the sediments in the hydrothermal treatment

The sediments samples separated from the residual solutions of the hydrothermal treatment were studied by SEM. Fig. S3 shows the topography of the sediments.

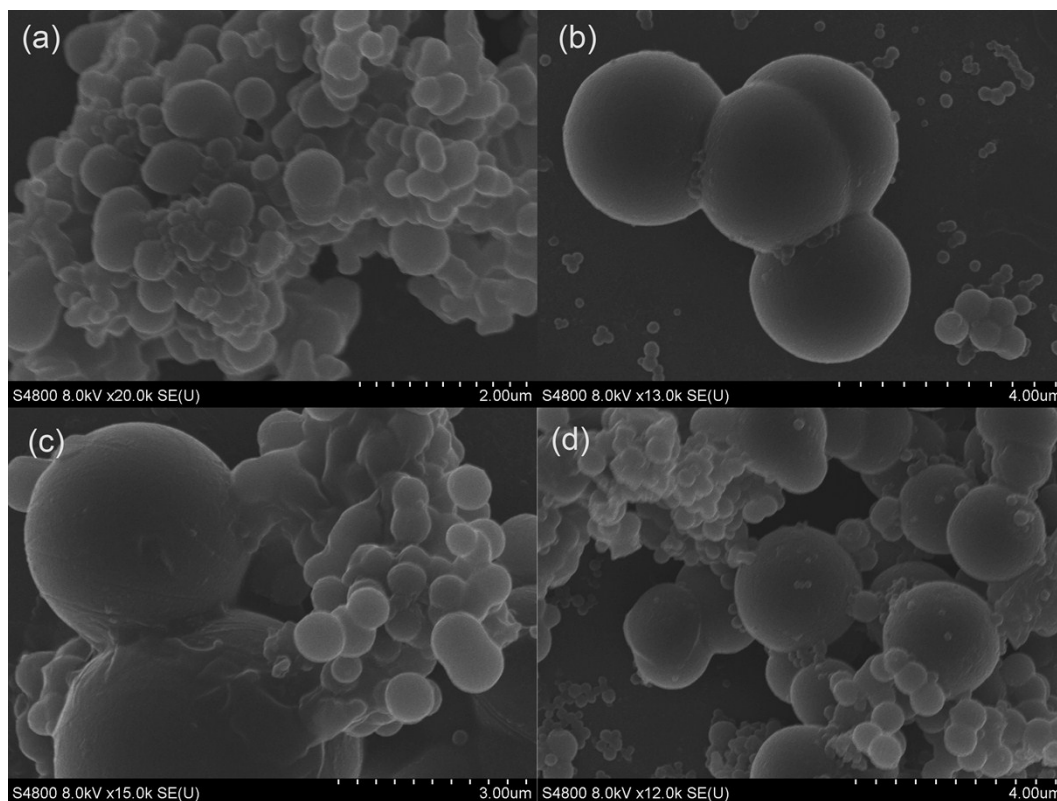


Fig. S3. The SEM images of the sediments in the hydrothermal treatment. Image (a-d) is sediments in tube (4), tube (5), tube (6) and tube (7) respectively.

4. The curve fitting of the XPS C1s spectra

The curve fitting of the CFs XPS C1s spectra performed using a Gaussian-Lorentzian peak shape after performing a Shirley background correction, as shown in Fig. S4. Deconvolution of the C 1s spectra gives three peaks [2]: that represent C=C and C-C groups (peak I, 284.6 eV), carbon present in phenolic, alcohol or ether groups (peak II, 286.1–286.3 eV), carbonyl or quinone groups (peak III, 287.3–287.6 eV). It is obvious that the peak II and peak III rose with increase of the glucose solution concentration. The organic groups increase with the glucose solution, coincided with the O/C ratio plot we discussed in the communication.

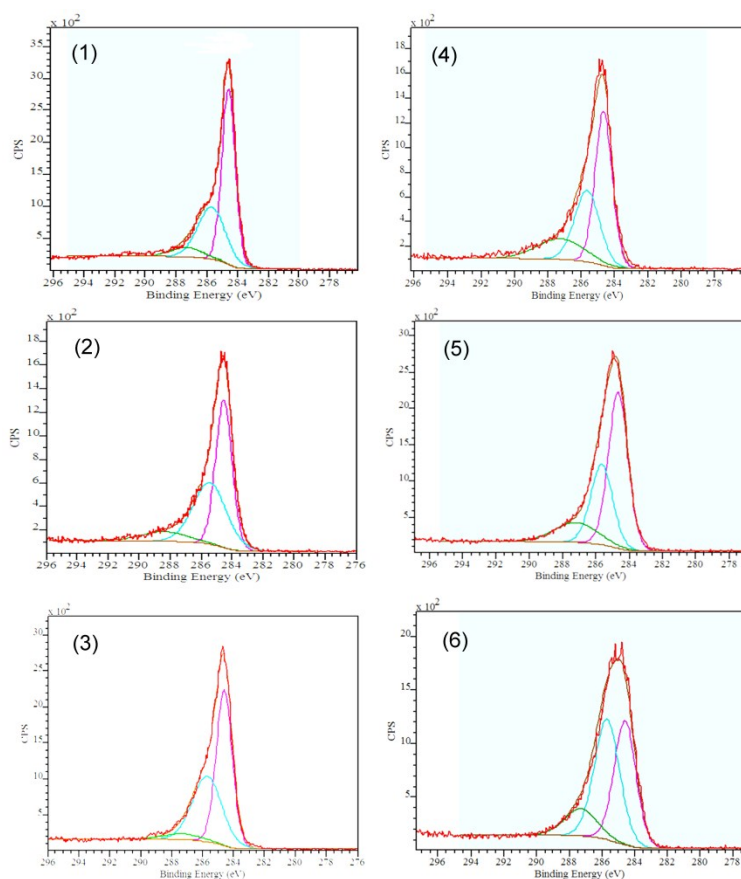


Fig. S4. The curve fits of the XPS C1s spectra were list: (1) unsized CFs, (2) CFs hydrothermal treated in 2 gL^{-1} glucose solution, (3) in 5 gL^{-1} glucose solution, (4) in 10 gL^{-1} glucose solution, (5) in 15 gL^{-1} glucose solution, (6) in 20 gL^{-1} glucose solution. The pink peaks at 284.6 eV correspond to the C-C and C=C; the blue peaks at $286.1\text{--}286.3 \text{ eV}$ correspond to the C-O; the green peaks at $287.3\text{--}287.6$ correspond to the C=O.

5. Contrast of the XPS C1s spectra

In this study, no compounds can decrease the bonding energy of the C1s, so we aligning the spectra at low bonding energy side of the C1s Peak to observe the shift of the C1s spectra.[3, 4]

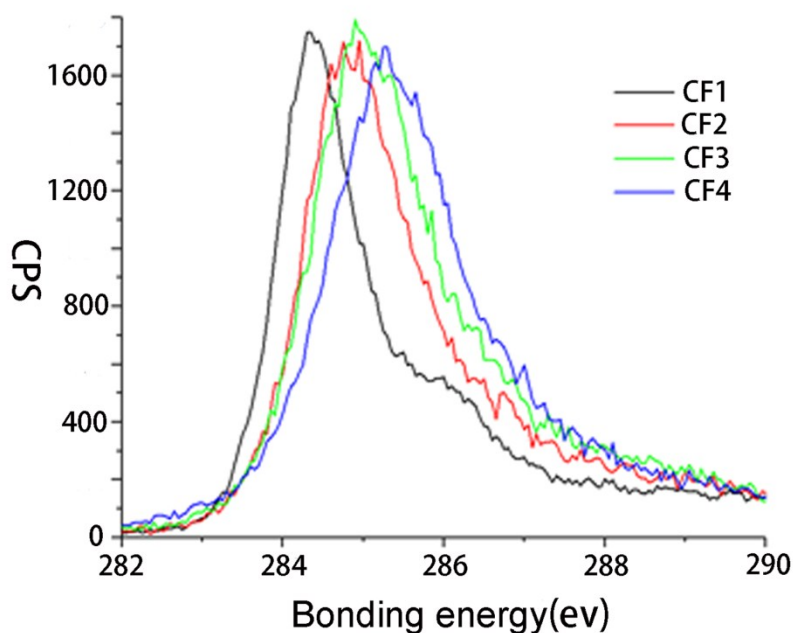


Fig. S5. The C1s spectra of the samples were contrast without calibration, the sample codes correspond with Table 1.

6. *The dewetting of the polyetherimide (PEI) on the CFs*

We mentioned the dewetting phenomena of the PEI particles on the CFs in the communication, which was illustrated by the SEM images as shown in Fig. S6. Fig. S6a showed the topography of the CFs sized by PEI emulsion, which was prepared according the literature[5]. Fig. S6b was that of the PEI sized CFs which undergone the hot-treated at 673K in the nitrogen atmosphere. The PEI Nano particles melted and dewetted in the hot-treated. It is obvious that the contact angle of the round PEI particles on the CFs is over 150 °after melted and cooled.

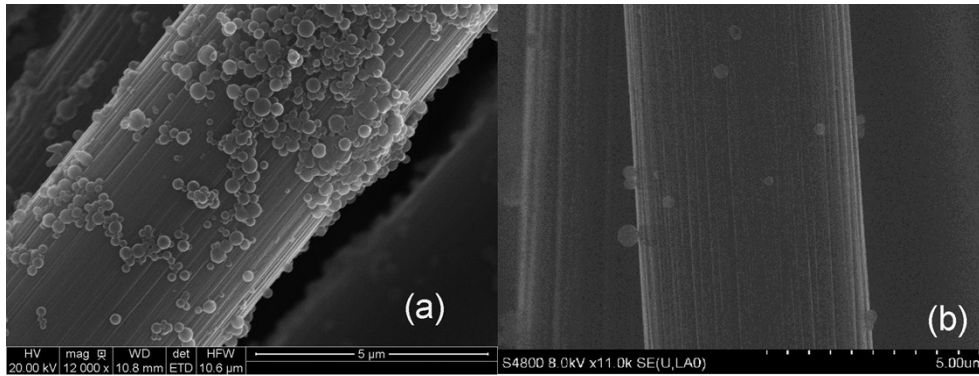


Fig. S6. The dewetting of the melted PEI particles on the CFs

Reference

- [1] M. Sevilla, A.B. Fuertes, The production of carbon materials by hydrothermal carbonization of cellulose, *Carbon* 47(9) (2009) 2281-2289.
- [2] Z.R. Yue, W. Jiang, L. Wang, S.D. Gardner, C.U. Pittman, Surface characterization of electrochemically oxidized carbon fibers, *Carbon* 37(11) (1999) 1785-1796.
- [3] P. Me´rel a, M. Tabbal b, M. Chaker a, S. Moisa a, J. Margot c, Direct evaluation of the sp³ content in diamond-like-carbon films by XPS., *Applied Surface Science* 136 (1998) 6.
- [4] G.P. Javier Di´az, † Salvador Ferrer, and Fabio Comin, Separation of the sp(3) and sp(2) components in the C1s photoemission spectra of amorphous carbon films .pdf, *Phys. Rev. B* 54 (1996) 8064.
- [5] I. Giraud, S. Franceschi-Messant, E. Perez, C. Lacabanne, E. Dantras, Preparation of aqueous dispersion of thermoplastic sizing agent for carbon fiber by emulsion/solvent evaporation, *Applied Surface Science* 266 (2013) 94-99.