Property enhancement of graphene fiber by adding small loading of cellulose nanofiber

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Abstract Herein, a small amount of cellulose nanofiber (CNF) was introduced in graphene oxide (GO) spinning dope to improve the property of graphene fiber. It was found that the one-dimensional CNF could absorb on the GO sheets, and enhance the inter-layer interaction as well as the connection between GO sheets, resulting in an easy formation of lyotropic liquid crystalline structure and a better orientation of the resultant fiber. A large enhancement of tensile strength, modulus, and elongation of GO/CNF hybrid fiber has been achieved. After chemical reduction of GO, the fiber still maintains its original mechanical properties but with high conductivity and improved hydrophilicity. Our work further demonstrates the importance of a stable lyotropic liquid crystalline structure of GO dope in the spinning of graphene fiber and provides a facile way to produce graphene fiber with good performance.

Keywords Graphene fiber, Cellulose nanofiber, Composite fiber, Conductivity

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Introduction

Graphene, one of the most attractive materials of twenty-first century, with its unique structure, intrinsic mechanical strength, compelling heat, and electrical conductivity has received persistent attention.¹ Numerous researches have been carried out to translate the intrinsic attributes of individual graphene sheets into macroscopic assemblies for practical applications. Graphene paper, 3D graphene network, and graphene fiber were prepared in succession via various methods and techniques.²⁻⁶ Different from other carbon-based fiber such as traditional carbon fiber and carbon nanotube (CNT) fiber that all needs high temperature in the fiber processing either for post graphitization or in the CNT synthesis, graphene fiber emerges as an extremely attractive and promising carbon-based fiber of the next generation, which can be prepared via the facile and benign wet-spinning at room temperature and was originated from well-sourced graphite powder. This kind of carbon-based fiber could be applied as a supercapacitor, intelligent fiber, and a smart-fiber, via in situ or post functionalization. Usually, graphene fiber is obtained via fluidly wet-spinning from the concentrated graphene oxide (GO) aqueous suspensions followed by chemical reduction.⁵ Since GO sheets could be well dispersed in many solvents and easy to be functionalized, abundant enhancement method and modification could be proposed to increase the property of graphene fiber.⁷⁻¹³ Various attempts have been promoted mainly to further enhance the mechanical strength or functionalize the fiber.²⁻¹³ In our previous work we found that the defects, packing density in the fibers, orientation of graphene sheets, and interaction between graphene sheets could significantly affect the properties of the fiber.¹⁴ In a work reported by Gao, the stronger graphene composite fiber has been prepared by grafting PAN on GO sheets to enhance inter-layer adhesion¹⁵, while larger graphene sheets were utilized as building block to construct stronger π-π interaction of adjacent sheets in Tour’s group and Gao’s group;¹⁶,¹⁷ Also diamine cross-linker was introduced in Park’s group as coagulation bath to form ion bridges between the adjacent GO sheets.¹⁸ Other polymer such as hyperbranched polyglycerol (HPG), polyvinyl alcohol (PVA)¹⁹ were also introduced in graphene fiber, mainly to enhance the adhesion of the GO sheets. In
general, stronger sheet adhesion, better-alignment, less defect benefits a stronger fiber.

Since graphene fiber is prepared via fluidly wet-spinning from the concentrated GO aqueous suspensions and treated with reduction, the original state of GO dope will play an important role in determining the final property of graphene fiber. GO dope with stable lyotropic liquid crystalline structure was of key importance, which usually requires a high concentration or large sized GO sheets.14,20 Thus, it is highly desired to find an easy way for the formation of stable liquid crystalline structure in GO dope with less concentration or smaller GO sheets. Cellulose nanofiber (CNF) is one-dimensional nano-sized and usually utilized as nanobuilding block for nanocomposites. Moreover, it is highly water dispersible and super hydrophilic owing to the abundant hydroxyl groups. Its aqueous suspension shows liquid crystalline behavior and mechanical strong fiber could be wet-spun from the suspension.21–23 In this work, a small amount of CNF was introduced into the GO dope with a hope to obtain stable liquid crystalline structure, thus graphene fiber with better alignment and mechanical property could be obtained. The interaction between GO sheets and CNF was investigated by Fourier-transform infrared (FTIR). The liquid crystalline behavior of the spinning dope was characterized via Polarized Optical Microscopy, SEM, and rheological property. With the enhancement of liquid crystalline behavior of GO dope via adding CNF, a large enhancement of tensile strength, modulus, and elongation of GO-CN F hybrid fiber has been achieved. After chemical reduction, the reduced GO–CNF (r-GO-CN F) fiber still maintains the original mechanical properties of GO–CNF fibers, but with high conductivity and improved hydrophilicity.

**Experimental section**

**Materials**

Graphite powder (200 mesh) was purchased from Qingdao Black Dragon graphite Co., Ltd. Concentrated $\text{H}_2\text{SO}_4$ (98%), KMnO$_4$, $\text{H}_2\text{O}_2$ (30%), HCl, CH$_3$COOH, NaBr, NaClO (8% available chlorine), and NaOH were purchased from Kernel Chemical reagent plant (Chengdu, China), Chitosan (deacetylation degree of 95%) was purchased from Zhejiang Golden Shell Biological Technology Co., Ltd. Microfibrillated cellulose (Celish MFC KY100-S) was purchased from Daicel Chemical Industries, Ltd., Japan. 2,2,6,6-tetramethylpiperidin-1-oxyl radical (TEMPO) was purchased from Sigma–Aldrich. All reagents were used as received.

**Preparation of GO and CNF**

GO was prepared from natural graphite by a modified hummer’s method. The as-prepared GO was then centrifuged and washed with 1 M hydrochloric acid and deionized water until the supernatant turned to neutral. The clean, washed GO was then concentrated by repeated centrifugation. Nanofibriled cellulose (CNF) was prepared according to TEMPO-mediated oxidation method. The 2wt% of microfibriled cellulose was suspended in water containing TEMPO and NaBr. The pH was maintained at 10.5 by adding 1 M NaOH. The reaction was finished until no more decrease in pH was observed after 3–4 h. The resulting mixtures were then thoroughly washed with deionized water by filtration until the filtrated solution turned neutral and re-dispersed in water at a concentration of around 10 mg/ml with vigorous stirring.

**Wet-spinning of GO fibers**

A series of spinning solution was prepared by adding quantitative CNF aqueous suspension to concentrated GO gel of 46 mg/ml. The CNF suspension was diluted to a calculated concentration and then ultra-sonic treated for 30 mins using a water bath ultrasonic cleaner before use. The overall concentration of GO was kept at 3 wt%. Different loading of 2/100, 5/100, and 10/100 CNF with respect to GO were prepared and was denoted as GO-CN F-2, GO-CN F-5, and GO-CN F-10, respectively. GO dope without CNF was also prepared as reference sample. These spinning solutions were then vigorously stirred for 20 h to ensure a homogeneous distribution. GO spinning dope with or without CNF was fluidly wet-spin by a capillary with an inner diameter of 450 μm, The spinning dope was injected into the chitosan coagulation (1 wt% chitosan, 0.8 wt% acetic acid) under N$_2$ pressure. After soaking in the coagulation bath for 1 min, the fiber was then collected on the Teflon drum, and washed with deionized water three times. The fiber was then naturally dried for 24 h, further vacuum dried for 8 h at 60$^\circ$C, finally the GO-CN F fibers were obtained. Reduced GO (r-GO) fiber was obtained by immersing in hydroiodic acid for 15 mins at 80$^\circ$C, then washed with ethanol and deionized water.

**Characterization**

Polarized optical microscopy (POM) was carried out on Leica DM EP in transmission mode. Scanning electron microscopy (SEM) was performed on Inspelt F (FET) at 5 kV accelerate voltage. Rheology tests were conducted on HAAKE Mars III rheometer equipped with a cone-shaped spindle (1° tapper, 30 mm diameter) and gathered on steady shear mode varied from 0.01 to 10 s$^{-1}$, logarithmically increased and 24 points in all, and each points was kept for 1 min. Zeta potentials of the GO–CNF suspensions were carried out on a laser electro-photorisis zeta-potential analyzer (Zetasiser 3000HSA Malvern Instrument), X-ray diffraction (XRD) tests were carried out in X’Pert Pro XRD instrument to access the graphite intercalating space. Raman spectra were taken at Renishaw invia Raman Microscopy (RM 100) at a Ni-Ne laser of 532 nm. FTIR spectra was collected on Nicolet 6700 (Thermo Electron Corporation) on reflection mode. The Mechanical strength and stress–strain curve were collected from the monofilament tensile testing machine YG-001A with gauge length of 10 mm and a strain rate of 1 mm/min, and the cross-section area was calculated from the SEM images, the final results represented the average value for at least ten samples. Electrical behavior was examined with the standard four-probe method on FIUKE 45 dual display multimeter. Water contact angle tests were performed on OCA20 contact angle goniometer equipped with video capture (Dataphysics, Germany).

**Results and discussion**

SEM images of GO sheets and CNF are shown in Fig. 1a and b, respectively. The corresponding size distribution images show the diameter of GO sheets ranged from 0.84 to 7.1 μm and the
average diameter is 3.7 μm, which are small-sized GO sheets. And the thickness of GO was 1.0 nm, the corresponding aspect ratio was calculated to be 3700. The relatively small GO sheet size was generally higher with carboxyl and hydroxyl group and is more favored to be functionalized. The length of CNF ranged from 0.38 to 2.64 μm, the average length was 1.11 μm, the average diameter was 32 nm, and the aspect ratio was calculated to be 35. The CNF obtained by TEMPO-mediated oxidation is carboxyl and hydroxyl group rich, while GO sheets are also functionalized with many carboxyl and hydroxyl group after oxidation. Owing to the small size effect, the carboxyl and hydroxyl group of CNF could be fully exposed, thus a strong hydrogen bonding between GO and CNF is expected. FTIR was carried out to confirm the interaction between GO and CNF, this is shown in Fig. 1e. For GO, the adsorption peak at 1720 cm\(^{-1}\) is assigned to the C=O stretching vibration, similarly for CNF, the adsorption peak at 1723 cm\(^{-1}\) originated from the carbonyl group of the carboxyl group.24,25 However, for mixture of GO-CNF-10 sample, the carbonyl adsorption peak is red shifted to 1716 cm\(^{-1}\), on account of the hydrogen bond between GO and CNF, making CNF accessible to attach on GO sheets. 

Graphene fiber can be wet-spun from the liquid crystalline behavior of the GO, the pre-ordered GO sheets is the premise for the continuous wet-spinning of the fiber.1,20 CNF itself could form liquid crystalline for its high long-aspect ratio and high water dispersibility owing to the abundant carboxyl and hydroxyl group on CNF molecular chain.21,22 First of all, the liquid crystalline phase of hybrid spinning dope at different CNF loadings was examined under polarized optical microscopy observation. The vivid birefringence pattern is the typical characteristic of the liquid crystalline behavior. At the concentration of 10 mg/ml, the GO and GO aqueous suspension display the nematic liquid phase for the schlieren texture, a typical feature for nematic phase could be observed. For comparison, GO aqueous suspension was gradually diluted from 0.25 mg/ml and also observed under POM as shown in Fig. 2a. With the GO concentration decreased, the birefringence pattern became less vivid. At concentration of 0.25 mg/ml, the GO suspension exhibits an isotropic phase. The POM observation of GO–CNF at the CNF loading of 2/100, 5/100, and 10/10 are shown in Fig. 2b–d, respectively. As the concentration decreased the birefringence patterns also became faded. To better compare the liquid crystalline structure of GO and GO–CNF building block, the GO and GO–CNF hybrid suspensions at the same concentration were carefully studied, as marked in the rectangular area in Fig. 2. At the same concentration of 0.5 mg/ml, the GO suspension only partially displays birefringence in the tube and the birefringence is hard to observe. As the loading of CNF increased the patterns became more vivid and gradually spreads all over the tube, indicating an increased liquid crystalline behavior of GO suspension by adding CNF. The building block of GO–CNF exhibits liquid crystalline behavior in its aqueous suspension. A small loading of CNF was added in the GO aqueous suspension. The hydrogen bond between CNF and GO makes the CNF available to absorb on GO sheets. The freeze-dried samples could preserve the original arrangement of the GO sheets and CNF in the liquid state. One observes that the GO foam is less well organized compared with samples of GO-CNF-2 and GO-CNF-5, which displayed more ordered and uniformly organized as CNF loading increased. This phenomenon is in agreement with the POM observation results.
The shear-thinning behavior of the samples can be ascribed to the orientation under shear, which may be caused by the fact that GO sheets are better aligned to a more orderly aligned structure from a relatively disordered isotropic phase by adding CNF. This shear-thinning behavior and less viscous state suggested that the GO–CNF suspensions are highly spinnable under shear, which may give birth to a better oriented fiber and could provide instructions for the wet-spinning process.

The GO–CNF composite fiber was wet-spinning as illustrated in Fig. 3. The morphology of the obtained fiber along with the orientation of CNFs can be observed in the images. The shear-thinning behavior of the samples can be ascribed to the orientation under shear, which may be caused by the fact that GO sheets are better aligned to a more orderly aligned structure from a relatively disordered isotropic phase by adding CNF. This shear-thinning behavior and less viscous state suggested that the GO–CNF suspensions are highly spinnable under shear, which may give birth to a better oriented fiber and could provide instructions for the wet-spinning process.
and vertical to the fiber direction was investigated via SEM (Fig. 3b and c). The cross-section morphology of the composite fiber shows curled and closely stacked GO sheets (Fig. 4a–c). The nanosized one-dimensional CNF could be clearly observed at the magnified cross-section images as shown in Fig. 4c, which is small dot-like as circled in red. GO sheets are closely and uniformly piled up on one another inheriting from the pre-aligned original spinning dope. For comparison, the as-spun GO fiber without CNF was also observed (Fig. 4d–f), compared with GO–CNF fibers it also displays layered structure similar to GO–CNF fibers, but it is less compactly stacked with more defects and voids as shown in Fig. 4f. The voids and defects of GO fiber may originate from the relatively low GO concentration, small size of GO sheets, and the fast solvent change in coagulation procedure. GO sheets are aligned along the flow direction, but on the radial direction it is stacked with several vectors, so there may be some defects and voids during stacking and wrinkling of the GO sheets in the wet-spinning or post-drying procedure. It was reported that a relatively low concentration GO generally leads to brittle fibers or ribbon-like fibers, especially for small GO sheets. But the GO–CNF composite fiber started from 3 wt.% GO aqueous suspension of small sheets and a small loading of CNF are more densely packed and regular circular shaped; again it is suggested that CNF may play a supporting and bridging role for GO fiber.

To better observe the morphology of GO fiber with and without CNF, the surface morphologies of GO fiber and GO–CNF fiber at varied CNF loadings at different magnifications are shown in Fig. 5. As demonstrated in Fig. 5, by comparison, much enhanced orientation of GO sheets by adding CNF can be clearly seen, better alignment of GO–CNF fiber could be observed. For GO–CNF-10 fiber (Fig. 5i and j), uniformly orientated GO sheets could be observed, at the magnified images (Fig. 5i) the CNF is orientated along the fiber axis which is different from the magnified surface section of GO fiber without CNF (Fig. 5a). Thus, it can be concluded that the CNF adsorbed on GO sheets well and the GO–CNF building
blocks are orientated along the flow direction during the wet-spinning procedure, resulting the orientation for both the GO sheets and CNF. The enhanced orientation of GO fiber after adding CNF could be proved by comparing the surface morphologies between GO and GO–CNF fibers as shown in Fig. 5. SEM images clearly reveal that the orientation increased and optimized after the incorporation of CNF. Though CNF cannot be easily observed form the fiber surface, GO sheets indeed show better orientation after adding CNF in GO–CNF fibers. The enhanced orientation degree is ascribed to the better aligned state of the original spinning dope. While at the loading of 10%, the alignment also shows a little drop which may be due to the local aggregation of the long-aspect ratio of CNF. The orientated structure originated from the pre-aligned spinning dope and the alignment was further facilitated by the elongational flow in the spinning procedure. After the solvent change in the coagulation bath and post-drying process, the GO sheets tend to wrinkle and the adjacent GO–CNF blocks become closely packed accompanying with the diameter shrinking. In the drying process, the fiber contracted both in diameter and length since the fiber was collected under wet state and restrained dried on the drum, the shrinkage in length direction was restricted and the aligned structure could be well preserved.

Mechanical strength is important for the practical application of the fiber. The mechanical properties of GO–CNF composite fibers were measured and the results are summarized in Table 1. Fig. 6 displays the typical stress–strain curves of the fibers at different CNF loadings. The elastic modulus and mechanical strength increased as the CNF loading increased, and for sample at a high loading of 10/100, the mechanical properties show a slight decrease, which may be ascribed to the partial agglomeration of CNF in the highly viscous GO gel.

At the CNF loading of 5%, the elastic modulus and fracture strength are promoted up to 8.6 GPa and 228 MPa compared to GO fiber of 6.3 GPa and 176 MPa. And the toughness of the fiber is also promoted from 2.53 to 3.48%. The alignment is one important aspect in the mechanical performance enhancement; the better alignment could also reduce the defects and facilitate the load transfer. In addition, the hydrogen bond between GO and CNF can also contribute to the mechanical performance of the GO–CNF fiber. Thus, it can be concluded that a simultaneous enhancement of strength and toughness of GO fiber can be achieved by adding CNF.

It was reported that the alignment of GO sheets, the interaction of the adjacent GO sheets, and the voids and defects in the fiber play a key important role in the mechanical performance of graphene or GO fibers. GO–CNF aqueous suspension shows orientated lyotropic liquid crystalline, the relative orientation degree of the as-prepared

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### Table 1 Mechanical Properties of GO and GO–CNF composite fibers

| Sample      | Tensile modulus (GPa) | Tensile stress (MPa) | Ultimate strain (%) |
|-------------|------------------------|----------------------|---------------------|
| GO          | 6.3 ± 0.5              | 176 ± 14             | 2.53 ± 0.33         |
| GO-NFC-2    | 7.1 ± 1.4              | 199 ± 17             | 2.89 ± 0.24         |
| GO-NFC-5    | 8.6 ± 0.9              | 228 ± 15             | 3.48 ± 0.32         |
| GO-NFC-10   | 8.1 ± 0.2              | 210 ± 5              | 3.00 ± 0.17         |

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![Figure 6](image6.png)  
**Figure 6** Typical stress–strain curve of GO–CNF composite fibers

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![Figure 7](image7.png)  
**Figure 7** a Polarized Raman spectra of GO-CNF-10 composite fibers in parallel and perpendicular to fiber axis and b orientation degree and tensile strength as a function of CNF loading
The fiber was further reduced to restore the electrical conductivity by immersing in hydroiodic acid aqueous solution (30 wt%) at 80°C for 15 mins. XRD was carried out to measure the reduction effectiveness. In Fig. 8a, XRD patterns of all the GO–CNF composite fibers show a d-spacing value of 8.4 Å. The r-GO-CNF fiber shows a wide 2θ degree at 25°, and the corresponding d-spacing value is ca. 3.55 Å, which implies that most of the oxygen groups were removed after reduction, and the conjugated network is partially restored. Raman spectra were also utilized to further characterize the reduction effectiveness as demonstrated in Fig. 8b. The intensity ratio of D band (1350 cm⁻¹) to G band (1580 cm⁻¹) is changed from...
The functional groups fully exposed. The reduced GO sheets were hydrophobic for the removal of oxygen functional group during reduction. After the incorporation of CNF, the water droplets’ contact angle decreased from 137° for r-GO fiber to 126°, as the CNF loading increased to 10/100 as shown in Fig. 10. The hydrophilicity of the r-GO fiber is promoted after CNF incorporation owing to the super hydrophilicity of the CNF. The improvement in hydrophilicity is beneficial for its application, for example, in electrochemistry filed. The hydrophobic attributes of graphene sheets will hinder the infiltration of the electrolyte, thus the inner graphene sheets are inaccessible to the electrolyte and the high specific area of graphene material could not be fully used. The highly conductive r-GO-CNF composite fiber with improved hydrophilicity has potential application in fiber-based flexible supercapacitor. Furthermore by an appropriate structure design such as porous fiber, r-GO-CNF fiber may lead to supercapacitor with much higher capacitance.34-36

The resulting mechanical strength for r-GO-CNF fiber was 236 MPa and had an electrical conductivity of 8300–11000 S/m. Compared with other carbon-based fibers such as wet-spun CNT fiber17 with a mechanical strength of 1.8 GPa, and directly spun twisted CNT yarns18,19 with a mechanical strength up to 1.0 GPa, traditional carbon fiber with a tensile strength of 6.0 GPa,20 the GO fiber or graphene fiber emerges as a new family of carbon-based fiber, its mechanical strength could be as high as 500 MPa17 using large GO sheets as building block and followed with ionic cross-linking up to now. Compared with carbon fiber or CNT fiber which has emerged for a long time, the graphene fibers still have a long distance both in mechanical strength and manufacture procedure, all need to be further promoted. The inferior mechanical strength of graphene fiber may be ascribed to that of the voids and defects in the GO sheets, and the conjugated network could only be partially restored after reduction. Numerous attempts have been proposed to enhance the mechanical properties of graphene or GO fiber,8,13,15-17 such as polymer blends, post drawing, or wet-drawing; larger size GO flakes have been used. Herein, the incorporation of CNF effectively enhances the mechanical properties of GO fiber from the small GO sheets. Compared with graphene fiber from large GO size or PAN-reinforced graphene fiber, the GO–CNF fiber was weaker. But this method does not require complex polymer grafting, post-drawing process, or large-sized GO sheets, only a small content of CNF is needed. The mechanical strength of the GO–CNF fiber could be further enhanced by post-drawing or ionic cross-linking. More importantly the small-sized CNF does not bring down the electrical conductivity appreciably. After reduction the fiber displays excellent electrical conductivity, good mechanical properties, and improved hydrophilicity.

Table 2 Mechanical and electrical conductivity of r-GO-CNF composite fiber

| Sample      | Tensile modulus (GPa) | Tensile stress (MPa) | Ultimate strain (%) | Conductivity (S/m) |
|-------------|-----------------------|---------------------|---------------------|--------------------|
| r-GO        | 7.05 ± 0.39           | 193 ± 17            | 2.53 ± 0.30         | 13000              |
| r-GO-CNF-2  | 6.04 ± 0.34           | 202 ± 8             | 2.00 ± 0.52         | 11000              |
| r-GO-CNF-5  | 7.25 ± 0.21           | 236 ± 14            | 3.70 ± 0.31         | 9000               |
| r-GO-CNF-10 | 7.51 ± 0.47           | 201 ± 29            | 1.99 ± 0.24         | 8300               |

Figure 10 Water contact angle of r-GO-CNF fiber as a function of CNF loading

0.86 to 1.19 after reduction, which also implies an effective reduction.

SEM characterization was carried out to explore the morphology change of GO and GO–CNF after reduction as shown in Fig. 9. After reduction a reduction in diameter could be observed. For r-GO fiber (Fig. 9a and b), no significant change is observed from the cross-section morphology before and after reduction. But for GO–CNF composite fiber (Fig. 9c and d), the packing density of GO sheets decreases after the reduction, which may be correlated to the reduction procedure in which the fiber was immersed in hydroiodic acid (30 wt.%) in the reduction procedure, the super hydrophilic CNF might absorb water and swelling during the reduction thus some little voids were brought. The mechanical properties of the r-GO-CNF fibers are summarized in Table 2. The r-GO-CNF fiber still has a mechanical strength up to 236 MPa and flexibility of 3.7%.

After reduction, the mechanical properties are preserved and the electrical conductivity is simultaneously restored to a large extent as shown in Table 2. The GO–CNF composite fiber displays a high electrical conductivity of 8300 S/m even at a high loading of 10 wt% CNF. Different from the polymer-reinforced GO fiber such as using PAN, HPG, PVA as reinforcing materials, the polymers mainly act as glue and adhere the adjacent GO sheets which displayed nacre mimic structure that severely destructed the electrical conductivity. In our work, the mechanical strength promotion of GO–CNF fiber is mainly resulted from the better alignment of GO sheets as promoted by CNF. The linear shape and small-sized CNF does not entirely cover all the GO sheets. And the conductive path was only partially disrupted. And the GO–CNF composite fiber still shows highly conducted.

CNF is hydrophilic for its abundant carboxyl and hydroxyl groups. In addition, its nanosize, long-aspect ratio, makes
Conclusion

In conclusion, GO–CNF hybrid fibers have been successfully prepared by directly mixing GO gel with CNF aqueous suspension. The introduction of CNF facilitates the liquid crystalline behavior of the spinning dope; stable, highly spinnable, and well pre-aligned hybrid spinning are formed. GO–CNF fiber with better alignment, improved mechanical performance is obtained from the liquid crystalline of GO–CNF spinning dope. After reduction, the r-GO-CNf fiber was highly conductive and preserved the original mechanical performance of GO-CNf. Besides, the incorporation of CNF also endows r-GO-CNf fiber with improved hydrophilicity compared to the r-GO fiber. This work emphasizes the importance of the original liquid crystalline state in the fabrication of GO fiber, and provides an effective and facile method to reinforce GO fiber by enhancing the pre-alignment of the original spinning dope via adding small loading of CNF. Also multifunctional GO fibre could be obtained using CNF-absorbed GO sheets as new building blocks by further morphology control or functionalization.

Disclosure statement

No potential conflict of interest was reported by the authors.

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