

High-Performance Graphene Fibers Enabled by Hydration



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 Article Recommendations

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A small amount of water hydrates graphene oxide in organic solvent to promote the formation of a liquid crystal phase for robust graphene fiber.

Graphene fibers have attracted growing interest due to properties derived from individual graphene sheets such as their light weight, high mechanical strength, and high electrical conductivity. Thus, they are considered to be a promising electrode material for fiber electronics. Wet spinning of graphene oxide (GO) dispersion is currently the most used method to synthesize graphene fibers. In addition to using GO aqueous dispersions, it is important to develop GO dispersions based on organic solvents that are more capable of dispersing functional nanomaterials than aqueous media. In this issue of *ACS Central Science*, Kim and co-workers reported that the addition of a small amount of water to a GO-dispersed organic solvent could effectively enable hydration of GO sheets,¹ thus promoting the formation of a highly stable liquid crystalline GO phase and electrochemically exfoliated graphene (EG) (Figure 1). This method may provide a general and effective strategy to produce high-performance hybrid graphene fibers from GO organic dispersions.

Earlier, the GO dope was prepared in a typical wet-spinning process by dispersing GO sheets to form a stable solution and then injecting it into a coagulation bath to produce GO fibers. The graphene fibers were obtained after reduction of GO sheets by using reduction agents or thermal treatment. To endow the graphene fibers with enhanced mechanical strength and electrical conductivity, it was essential to realize a stable liquid crystalline phase in the GO spinning dope, so that the highly aligned arrangement of GO sheets could be effectively transferred to the graphene fibers.^{2–4} The formation of a liquid crystalline phase in the GO dispersion depended on the concentration of GO sheets and effective

aspect ratio (i.e., ratio of lateral size to thickness).⁴ More specifically, a higher aspect ratio meant that a lower critical concentration was required for the formation of liquid crystalline.⁵ It is well-known that hydrophilic GO sheets with abundant oxygen functional groups could be easily dispersed in water and exhibit a colloidal liquid crystalline phase. However, in the case of common polar organic solvents such as *N,N*-dimethylformamide (DMF) and *N*-methyl-2-pyrrolidone (NMP), it was difficult to form a highly stable GO liquid crystalline phase due to weak charge interactions and a mismatch of surface energy between solvent molecules and polar oxygen groups of GO.¹ The capability of GO sheets to form effective hydrogen bonds with solvents was considered the most important factor to realize the GO liquid crystalline phase.⁶

Unlike the previous strategies to modify GO sheets by increasing aspect the ratio and functional groups,^{6–8} Kim and co-workers discovered that adding only 2 wt % water to the GO/NMP dispersion could effectively enhance the solubility of the GO sheets.¹ The authors concluded that the hydrated surface of the GO sheets, after introducing water, could serve as an effective intermediate layer to achieve high colloidal stability in NMP. They carefully investigated the formation behaviors of GO liquid crystalline phase in the presence of water. It was observed that the sheets from the water-assisted GO/NMP dispersion showed a highly planar conformation without obvious wrinkles. Optical microscopy and scanning electron microscopy further demonstrated a stable dispersion state of the GO sheets in NMP/water mixture solvent. As expected, the as-spun GO fibers with a flat

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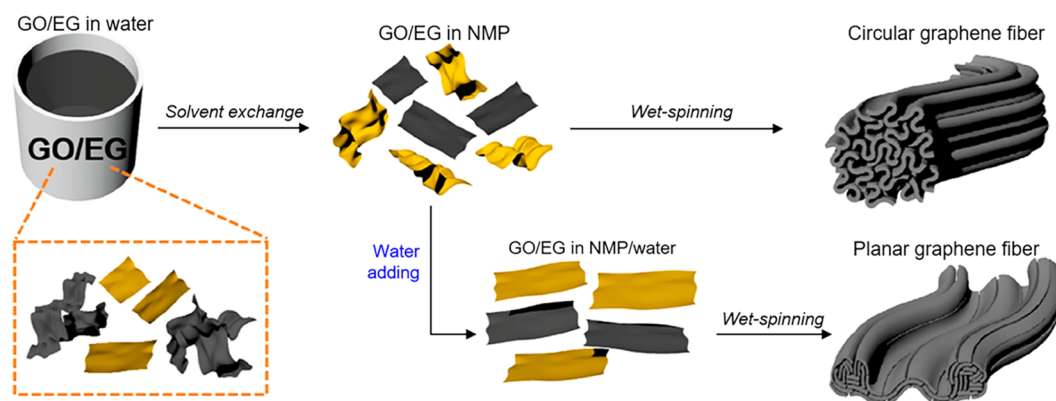


Figure 1. Schematic illustration of the effect of water addition on the conformation of GO/EG and cross section of resultant wet-spun graphene fibers. The addition of a small amount of water to the GO/EG NMP dispersion enabled a planar cross-sectional graphene fiber owing to hydration of GO sheets. In contrast, circular-shaped graphene fibers with wrinkled graphene sheets were wet-spun from GO/EG NMP dope without water addition. Reproduced with permission from ref 1. Copyright 2020 American Chemical Society.

shape can be consciously produced by adding over 2 wt % water. In contrast, the GO sheets in anhydrous NMP dispersion presented random wrinkles and obvious agglomeration, thus easily breaking as-spun GO fibers during the wet-spinning process.

The hydrated surface of the GO sheets, after introducing water, could serve as an effective intermediate layer to achieve high colloidal stability in NMP.

This interesting finding on the formation of GO liquid crystalline in organic solvent has greatly increased the potential of assembling GO with other functional nanomaterials to produce robust hybrid graphene fibers. To demonstrate this, Kim et al. introduced mildly oxidized EG flakes into water-assisted GO/DMF dispersion to form a mixed wet-spinning dope. The cross-sectional morphological evolution of the fiber was largely influenced by the solvent extraction rate in a coagulation bath such as ethyl acetate or acetone. Because of the slow NMP extraction caused by GO hydration, the anisotropic planar GO sheets could be spontaneously rearranged into a flat fiber, which favored the EG flakes with a highly oriented arrangement along the axial direction of graphene fiber. After systematic optimization, the resultant dumbbell-shaped hybrid graphene fiber exhibited both a high tensile strength of 597 MPa and an electrical conductivity of 527 S/cm, which makes it superior to the previously reported chemically reduced graphene fibers without metal additives.

The hybrid graphene fiber also exhibited high flexibility, and its structure was maintained well, even after bending it for 1000 cycles, which is highly desired for applications in fiber and textile electronics. For instance, this fiber can be

woven into a textile as a conductor and used as flexible fiber electrodes to fabricate fiber-shaped supercapacitors. From a viewpoint of future practical applications, the hybrid graphene fiber's mechanical and electrical properties can be further enhanced to meet the requirements of advanced electronic devices.⁹ We can expect to take advantage of this NMP-based GO spinning dope under mild operating conditions to introduce a wide range of other functional nanomaterials, such as carbon nanotubes, metal-nanowires, and polymers, into the graphene fiber for more functionalities and improved performance.

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Notes

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