MATERIALS SCIENCE

Reversible fusion and fission of graphene oxide–based fibers

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Stimuli-responsive fusion and fission are widely observed in both bio-organizations and artificial molecular assemblies. However, the design of a system with structure and property persistence during repeated fusion and fission remains challenging. We show reversible fusion and fission of wet-spun graphene oxide (GO) fibers, in which a number of macroscopic fibers can fuse into a thicker one and can also separate into original individual fibers under stimulation of solvents. The dynamic geometrical deformation of GO fiber shells, caused by solvent evaporation and infiltration, is the key to the reversible fusion-fission cycles. This principle is extended to implement flexible transitions between complex fiber assemblies and the inclusion or expulsion of guest compounds.

Fusion and fission behaviors have been extensively studied in biology, chemical engineering, and theoretical physics to understand cellular processes, develop morphological events of artificial assemblies, and create multimetallic compounds. Fusion and fission of lipid/surfactant/small organic molecules/polymer micelles and vesicles are usually triggered by either introducing salts, surfactants, ions, oxidants, and reductants or applying ultraviolet and visible light to change the interactions inside bilayer membranes, as well as by dissolving additives such as saccharides to alter the osmotic pressure between the aqueous interior of vesicles and the bulk solution (*1*–*6*). Similar behaviors of metal particles and clusters are also induced by heat or cluster deposition (*7*–*9*). Although progress has been made on fusion-fission of artificial vesicles and nanoparticles, reversible fusion and fission are still difficult to realize, mainly because of the irreversible physical or chemical changes at the interface between individual assemblies. The exploration of reversible and controllable fusion and fission would inspire the development of stimuli-responsive materials, which show promise in deriving dynamic transformable systems and structural materials with customized fibrous substructures. The recyclability of the assembled structures is also a beneficial attribute.

We propose a solvent-triggered topography-regulation strategy to implement reversible fusion and fission. Graphene oxide (GO) fiber was selected as a model because of the nature of GO sheets, including two-dimensional (2D) topology, abundant chemical moieties, superflexibility, and self-adhesion capability (*10*–*13*). After swelling (*14*), the wet-spun GO fiber features a shell (an outermost skin) that confines the movement of internal GO sheets and shows a solvent-triggered large volume change and elastic deformation capability. Upon stimuli of water and polar organic solvents, the topography of fiber shells reversibly switches between a wrinkled, tubular state and a spread, cylindrical state through swelling and deswelling, thus causing a transient fiber interface and leading to cyclic self-fusion and self-fission of an arbitrary number of GO fibers (Fig. 1A). In each cycle, the number, size, composition, structure, and properties of GO fibers are recovered after fission, exhibiting the precise reversibility of fusion and fission.

Continuous GO fibers were prepared following a wet-spinning protocol (fig. S1) (*15*, *16*). In Fig. 1B, we demonstrate the reversible fusion and fission involving up to 100 GO fibers (movie S1). For simplicity, the fused GO fiber assembled by n individual fibers is denoted as FuF-n, and the corresponding fissured, individual dried GO fiber is denoted as FiF-n. A typical fusion process requires three steps, including solvent swelling of fibers in a bundle, drawing the fiber bundle out of the solvent, and air drying. The fission steps comprise reswelling of the fused fiber in a solvent, splitting of swelled fiber bundles, and drying of the fissured fibers separately (see the supplementary materials).

In situ optical microscopy and ex situ scanning electron microscopy revealed the water-induced fusion and fission processes (Fig. 1, C to F; figs. S2 and S3; and movies S2 and S3). The swelling ratio, defined as the ratio of swelled fiber diameter to raw fiber diameter, is used to quantify the extent of swelling, which is influenced by solvent type, the chemical nature of the GO fibers, and the soaking time (fig. S4) (*17*). The equilibrium-swelled GO fibers randomly immersed in water exhibited a swelling ratio of 541% with a core shell structure, where the core (pore size of 2 to 5 μm after freeze drying; inner wall thickness of 28 ± 8 nm) was enclosed by a skin-like shell (80 ± 18 nm thick) of densely packed GO sheets that oriented along the circumference (Fig. 1, C1 and D1, and figs. S5 and S6). This different arrangement of GO sheets within the outer layer and inner fiber is a featured structure of wet-spun fibers that originates from the aligning effect during fiber extrusion and is enhanced by double-diffusive convection in the coagulation bath (*18*). The thickness of both the shell and inner wall depends on the swelling of fibers, which relates to the quantity of absorbed solvent. Driven by the surface tension of water, these swelled fibers collected together and deformed into a cylindrical group spontaneously as they were drawn out from the liquid. During air drying (0 to 40 min), the swelled fibers bonded together accompanied by adaptive crumpling of fiber shells, exhibiting a volume shrinkage of 98% (Fig. 1, C1 to C4 and D1 to D4). Meanwhile, the interlayer spacing decreased from >2.21 to 0.84 nm according to in situ x-ray diffraction characterizations (fig. S7A). Consequently (40 min), the resulted FuF-100 featured compact packing of GO platelets (interlayer spacing of 0.84 nm; density of 1.51 g cm^{-3}) with a tensile strength of 281 MPa (fig. S2, G to I). GO fibers also fused while not drying under tension despite the decrease in density and tensile strength compared with those drying under tension (fig. S8).

Fission of FuF-100 started with a homogeneous swelling when it was resoaked in water, and small gaps emerged at the interfiber interfaces as the swelling persisted. Subsequently (150 s), a rapid gap propagation, along with volume expansion of the whole fiber assembly, led to the complete fission into 100 individual quasicylindrical fibers. The fissured fibers sustained in water for a long time rather than dissolving into pieces despite a swelling ratio of 538%, because of the protective effect of the outer shell. During the whole process, solvent infiltration caused expansion between GO sheets, with interlayer spacing restoring from 0.84 nm to >2.21 nm (fig. S7B). After drying separately, the interlayer spacing between GO sheets (0.84 nm), density (1.54 g cm^{-3}), and tensile strength (259 MPa) of FiFs-100 were close to those of FuF-100 (fig. S2). In situ optical microscopy and polarized optical microscopy observations on the cross section of two GO fibers during their fusion and fission further verified the above description (movie S3).

A GO fiber was labeled with fluorescent 1,1,2-triphenyl-2-(4-bromomethylphenyl) ethylene (TPE-Br) and silicon (Si) nanoparticles. Tracking on the fusion and fission of the labeled fiber and a pristine fiber under fluorescence microscope revealed no substance exchange across the

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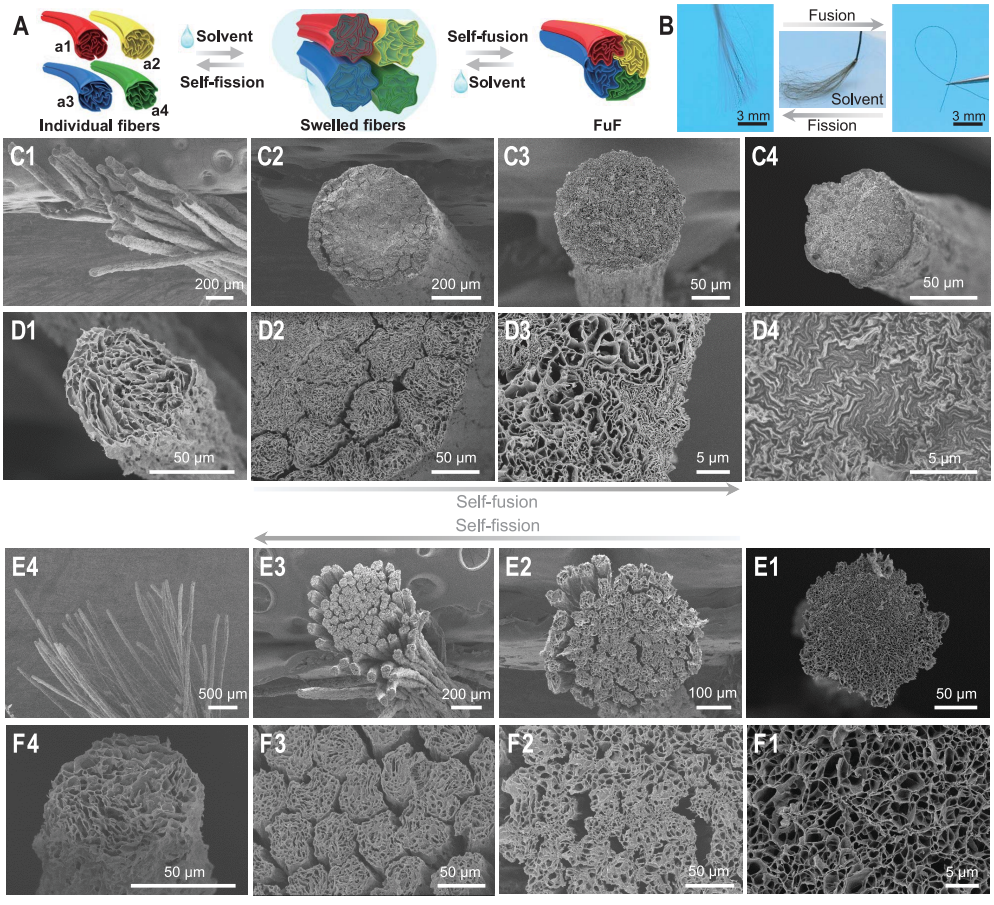

Fig. 1. Reversible fusion and fission of GO fibers. (**A**) Schematic of solvent-triggered precisely reversible self-fusion and self-fission of GO fibers, in which the individual fibers such as a1 to a4 are adaptively deformed by solvent swelling to form a thicker fiber FuF. The dried FuF is swelled in a solvent such as water to gradually fissure into original thinner fibers a1 to a4 without the exchange of GO sheets among them. (**B**) Photographs of reversible transition between ~100 GO fibers (left) and a single FuF-100 (right) through swelling-assisted (middle) fusion and fission. (**C** to **F**) Sequential scanning electron microscopy images of the water-induced fusion process (from left to right) of ~100 GO fibers and the reverse fission process (from right to left) of the FuF-100. (D) and (F) show corresponding enlarged local regions in (C) and (E), respectively. (C1) to (C4) and (D1) to (D4) correspond to 0, 25, 30, and 40 min in elapsed time, respectively. (E1) to (E4) and (F1) to (F4) correspond to 3, 75, 150, and 180 s in elapsed time, respectively. The time of fusion is set as zero when the GO fibers in a bundle are swelled in water just before being drawn out, as is the fission when the dried FuF-100 is resoaked in water.

fiber interface (Fig. 2, A to D; fig. S9; and movie S4). Energy-dispersive spectroscopy (EDS) analysis performed on a neat GO fiber and another composite Si/GO fiber (Fig. 2, E to L, and figs. S10 and S11) showed that fusion was finally accomplished by interlocking of the synergistically crumpled shells, and fission was induced by topographical recovery of shells with opposite normal vectors in geometry on both sides of the bonding interface (Fig. 2, K and L).

On the basis of the above characterizations, fusion and fission were found to be conducted by reversible crumpling and spreading of fiber shells while deswelling and reswelling (Fig. 3A and figs. S12 and S13). Here, the GO fiber shell is defined as the outermost dense layer of a saturated swelled GO fiber, characterized by macroscale cylindrical configuration, microscale ripples, and nanoscale closely organized GO platelets (fig. S13). The shell plays a major role in the reversible fusion-fission process because it is the boundary contacting with adjacent fibers, which provides interfiber bonding and debonding and protects inner fiber GO sheets from diffusion. A swelled GO fiber maintained a nearly constant perimeter during deswelling and reswelling courses, indicating that only elastic deformation occurred at the shell (Fig. 3B). The shell crumpled and spread because of wrinkling and unfolding of the ripple microstructures, with the curvature radius of the ripples varying in the range of ~10 nm to 83 μm (Fig. 3C and fig. S13, A1 to B4). At the same time, the inner fiber GO sheets deformed accordingly because they were interconnected in the swelled gel fiber. The reversible fusion-fission capability is only found in wet-spun GO fibers. Although dry-spun GO fibers can be fused, they failed to fissure, mainly because of the insufficiency of a dense and protective shell (fig. S14) (*19*). The topographical and volumetric evolution of fibers during the fusion process is driven by the surface tension of the solvent (*20*) and the Laplace pressure difference (P_c) (*21*). The attractive stress P_c facilitated further bonding of fiber shells through noncovalent interactions [π–π interaction and hydrogen bonding (*22*)], as well as subsequent wrinkling of GO sheets and densifying of the whole fiber bundle (fig. S15A). During the fusion process, the solvent-responsive fiber shell acts as an elastic barrier, preventing the sheet interdiffusion across the transient interface. Herein, the fusion degree, proposed to evaluate the fusion status (see the supplementary materials for the calculation), increased from 0% (unfused) to 100% (fused) upon swelling and traversed a hemifused region (swelling ratio from 165 to 358%) (fig. S16).

Conversely, fission was attributed to the cylindrical geometry–driven detachment between fiber shells (fig. S17A). As the FuF was soaked in good solvents for GO, the solvent infiltration weakened the adhesion strength (σ_{ad}*) between individual fibers (*23*). The tendency of cylindrical geometry reversion at the interface contributes to the interfacial detachability and compels the attached fibers to separate from each other. This repulsion is estimated by the net stress (denoted as fission stress σ_{fis}) of elastic tensile stress σ_{e1} and swelling pressure p_s along the inward-pointing normal direction of the curved microelement (see the supplementary materials). This hypothesis was further confirmed by finite element analysis, which showed that σ_{fis} was generated by the curved geometry of the shell when the swelling ratio of individual fibers surpassed a sufficiently high value (475 and 521% for separation of point 1 and 2, respectively) (fig. S17, B to E, and movie S5). Further swelling led to the rising σ_{fis}, which consequently reached a peak value equal to σ_{ad}* and triggered a sudden drop of σ_{fis} to 0 MPa. In fission experiments, the fission capability of FuF-2 was initiated when the average swelling ratio of individual fibers traversed a critical range from 310 to 419%, either by applying polarity-enhanced solvent or by reducing the pretreating temperature

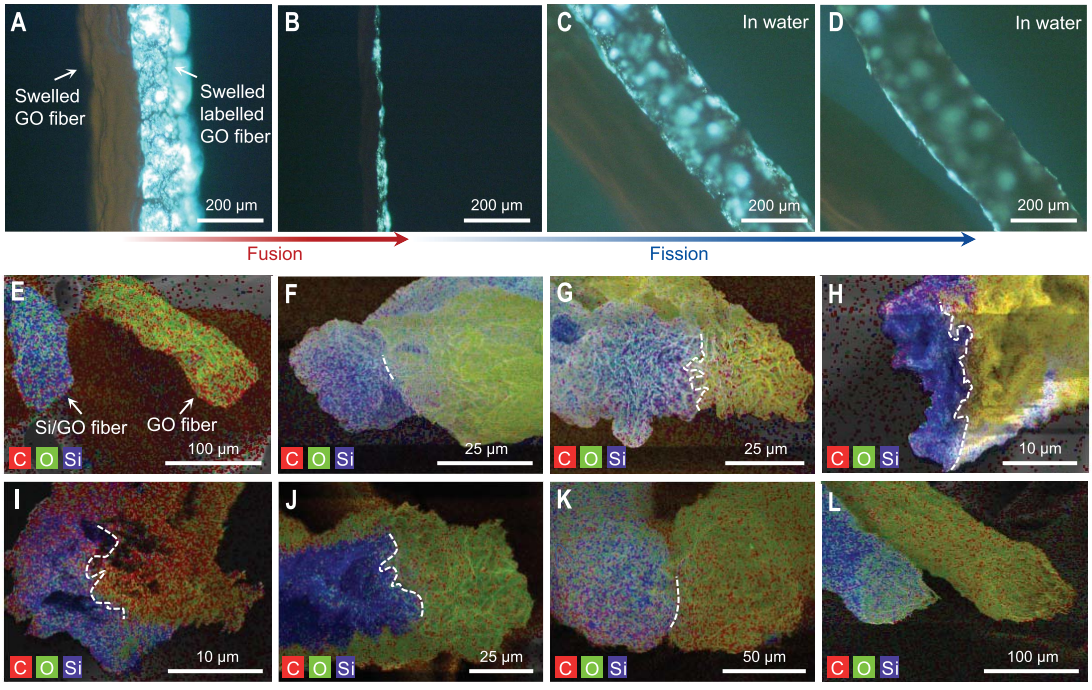

Fig. 2. In situ fluorescence observation and ex situ EDS analysis of a fusion-fission cycle. (**A** to **D**) Sequential fluorescent micrographs showing water-triggered fusion (A and B) and fission (B to D) of a neat GO fiber and a fluorescent TPE-Br–labeled GO fiber. A clear interface between the two fibers can be seen, demonstrating no exchange of GO sheets between them while fusing and fissuring. (**E** to **L**) Sequential overlapped elemental mapping images of carbon (red), oxygen (green), and silicon (blue) elements, respectively, showing the water-induced fusion (top) and fission (bottom) procedures involving a neat GO fiber and a Si/GO fiber. Dashed lines indicate the interface between the fibers, which suggests interlocking of the synergistically crumpled shells after fusion and the corresponding topographical recovery after fission.

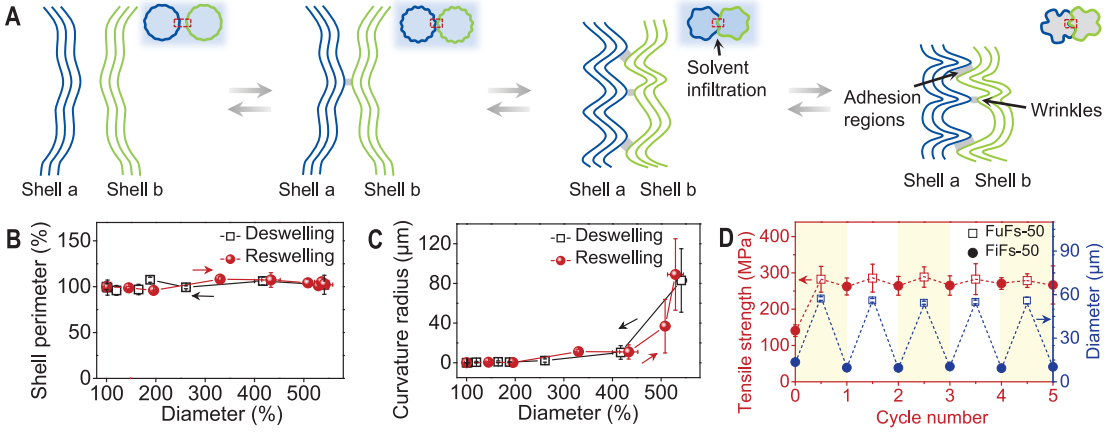

Fig. 3. Dynamic topographical deformation mechanism for reversible fusion and fission. (**A**) Schematic depicting the topographical crumpling of shells at the fiber interface that leads to self-fusion and reversible recovery that induces self-fission. (**B**) Shell perimeter of the water-swelled single GO fibers as a function of fiber diameter during the deswelling and reswelling courses. Corresponding values are plotted relative to that at the initial swelling state. (**C**) Average tip radius of curvature of a unit ripple in different fiber diameter during the deswelling and reswelling procedures of the water-swelled single GO fibers. The diameter of raw dry GO fiber in (B) and (C) is 52 μm. (**D**) Tensile strength (red) and corresponding diameter (blue) of FuF-50 and FiF-50 as a function of cycle number. The mechanical properties remained constant during several cycles.

on the FuF-2 (fig. S18A). Additionally, in contrast to GO fibers, GO belts featuring a distinct flat topography after swelling were able to be fused together but failed to fissure because of the absence of an arc conformation of GO sheets stored in wrinkles, which therefore lack a driving force σ_{fis} for self-fission (fig. S19).

The morphology and structure of both FiF-50 and FuF-50 were recovered throughout multiple fusion-fission cycles while exhibiting a relatively constant tensile strength of 281 and 259 MPa, respectively (Fig. 3D, fig. S20, and movie S6). After several cycles, the interlayer spacing of GO sheets in fibers remained unchanged at the same fusion or fission time (fig. S21). Additionally, there was no apparent decline in the tensile and compressive strength of FuFs when the fiber diameter increased from 23 to 78 μm (by increasing the number of individual GO fibers engaged in fusion from 10 to 100) (fig. S22). The values were stable at ~282 and 129 MPa, respectively. After thermal reduction at 1000°C, the FuF-100 exhibited a tensile strength of 597 MPa with a diameter of 58 μm. The consistency of the mechanical performances reflects that the stacking order within individual GO fibers remained intact. One advantage of thick FuFs is that firmly combined fibers are stronger than either the as-spun thick GO fibers or the unfused yarns with separated fibers (figs. S22B and S2I). Therefore, the affordable force on the thick FuFs is higher, which is better for structural materials that may exert mechanical superiority in engineering fields. Fluorescence tracking and EDS analysis on the labeled GO fibers further showed the cycles of reversible fusion and fission (figs. S23 and S24). Investigation on bundles composed of one Si/GO fiber and 99 neat GO fibers showed that the relative atomic content of Si in the Si/GO fiber remained constant whether

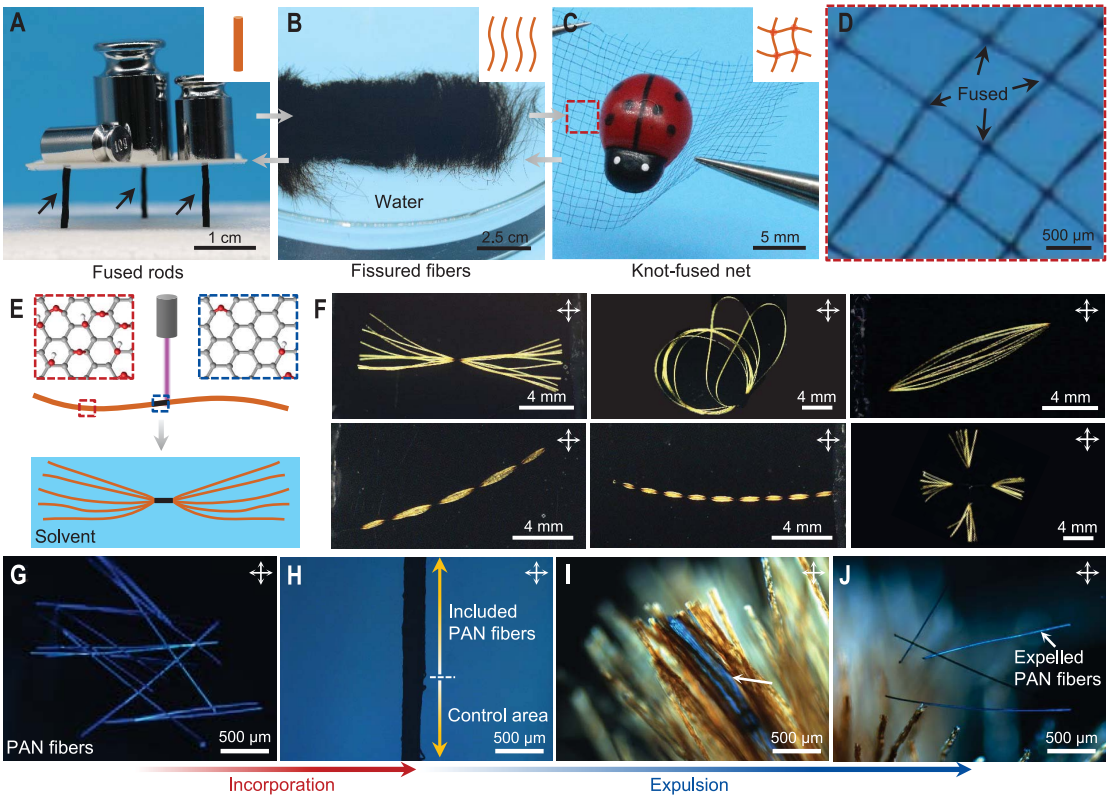

Fig. 4. Reversible fusion-fission promises controllable transformation. (**A** to **D**) Photographs of reversible transitions between a 3D, stiff fused GO rod (A) and a 2D, flexible knot–fused GO net (C and D) through fission (B) and refusion. (**E** and **F**) Illustration and polarized optical photographs showing the programmable 1D and 2D architectures of the locally laser-reduced FuF-10 under a water/isopropanol (8:2 v/v) mixture, including star, wristband, and multiblock wires with 3, 11, and 19 blocks and a dendrite with 40 arms. In the top of panel (E), the gray, red, and white spheres denote carbon, oxygen, and hydrogen atoms, respectively. (**G** to **J**) Polarized optical microscopy images showing the entrapment of polyacrylonitrile (PAN) staple fibers into a FuF by fusion (G and H) and the reversible expulsion by fission (H to J).

the Si-labeled fiber was within the fused fiber surrounded by neat GO fibers or at the fissured state (fig. S24, C to E; see the supplementary materials for the calculation).

Figure 4 demonstrates concepts for potential application of the fusion-fission behavior shown here. First, flexible transformation between diverse fiber–based assembled structures becomes possible. This would allow adaptive application of GO fiber–based systems in different scenarios with specific performance needs. For instance, the GO fiber assemblies were demonstrated to transform reversibly between a 3D stiff rod and a 2D flexible net through fission and refusion (Fig. 4, A to D; fig. S25A; and movie S7). In this case, as many as 13,500 fibers with micrometer-scale diameter and centimeter-scale length were fused into one 1.2-mm-thick rod that was rigid and strong enough to support 680 times its weight. Alternatively, transitions between a 1D fused GO fiber and various 1D and 2D sophisticated fiber assemblies were also conducted by localized fission and fusion (Fig. 4, E and F, and movie S8). The second application is that, through fusion and fission, a GO fiber bundle would be able to implement functions of including and expelling of guest objects, which may show promise in dynamic systems for controllable delivery. Various guest objects of different materials, sizes, and shapes, such as polyacrylonitrile chopped fibers, polystyrene microspheres, and glass beads in submillimeter scale, were absorbed into a FuF during fusion and then expelled during fission (Fig. 4, G to J; fig. S26; and movie S9). The third application is endowing ordinary fibers with reversible fusion and fission properties through GO coating. Conventional polymer, metal, and ceramic fibers were endowed with the reversible fusion-fission capability by simply coating a GO outer layer (figs. S27 and S28 and movie S10). This extension further expands the coverage of corresponding application fields.

Precisely reversible fusion and fission were induced here by the recoverable geometrical deformation of GO fibers. This enabled a dynamic feature in the fiber-assembled system so that transformation among structures and responsive actuation were realized. The concept was further expanded to conventional fibers through GO coatings while providing the potential for recyclability. The featured fusion-fission behavior constitutes a versatile strategy for the design of functional responsive materials.

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SUPPLEMENTARY MATERIALS

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Materials and Methods
Supplementary Text
Figs. S1 to S28
References (*24*–*38*)
Movies S1 to S10

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Reversible fusion and fission of graphene oxide–based fibers

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Reversible fiber fusion and fission

Materials that can cycle between states are of interest for actuators, soft robotics, or recoverable membranes for separations. Chang *et al.* show that a collection of graphene oxide fibers can fuse into a single stronger fiber upon immersion in a solvent, extraction, and drying under tension (see the Perspective by Cruz-Silva and Elías). The geometrical deformation of the fibers during drying and swelling plays an important role in the reversible cycles, with a large volume change between the dried and swelled fibers. Moreover, fibers made from polymers, glass, metal, or silk can be given these abilities when coated with a micron-sized layer of graphene oxide.

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TEXTILES

Reversible fusion-fission fibers

Reversible assembly of graphene oxide fibers creates a pathway for practical applications

By **Rodolfo Cruz-Silva**[1] *and* **Ana Laura Elías**[2]

Materials scientists have long used highly coveted biomimetic techniques in the search for synthetic structural materials that resemble muscles and other natural fibers (*1*). Among these techniques, self-assembly processes inspired by cell fusion are gaining notoriety (*2*). Within biological systems, fission occurs when one entity can separate into two or more parts, and the opposite process, fusion, occurs when two or more parts merge into one object (*3*). These processes can be naturally triggered by stimuli present in the environment—such as light, temperature, or humidity—but are ultimately controlled by the organism's own metabolism. On page 614 of this issue, Chang *et al.* (*4*) demonstrate the assembly of wet-spun graphene oxide (GO) microfibers through a reversible solvent-triggered process, which is regulated by the individual fiber's chemistry and morphology, mimicking biological fusion.

The fusion process of Chang *et al.* results in hierarchical assemblies that involve thousands of individual GO fibers (see the figure). When the fiber assemblies are immersed in a suitable solvent, the process can be reversed, resembling fission. Notably, the transition between the individual fibers and the complex yarn can be repeated without damaging the primary fiber structure, ultimately preserving the GO flakes and their arrangement. The authors also used this technique to prepare crosslinked nets and functional yarns that can capture and release foreign particles. This fusion-fission behavior has not been found in other ceramic or polymeric fibers, and it grants a new functionality to GO fibers that can be used to manufacture highly complex architectures with many applications.

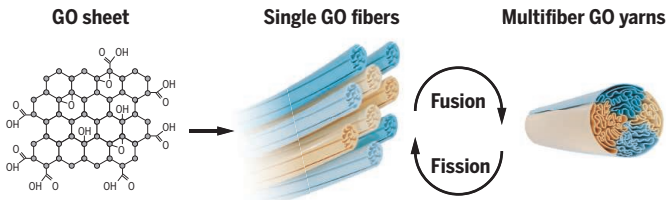
Humans have long relied on fibers to make clothes and various structural materials such as ropes and nets—from the primitive fibers used tens of thousands of years ago (*5*) to the modern functional fibers made using nanomaterials (*1*). In the 20th century, advanced fibers played a role in developing a myriad of technologies, including those used in aeronautics, electronics, and space exploration. Regardless of the era, fibers have been assembled hierarchically to form threads, yarns, ropes, and fabrics with different levels of complexity. As Chang *et al.* demonstrate, applying fusion and fission concepts previously studied in materials science through soft structures such as micelles and vacuoles is of paramount importance when making hierarchical fibers. Because modern fiber assembly techniques require complex machinery and a high-energy input, simplified fiber assembly processes are highly attractive, especially those with minimum energy requirements that can reduce the environmental footprint. Therefore, the development of a simplified and reversible fiber assembly process tackles one of the major challenges that human-made fibers have faced.

GO has attracted a lot of attention as a two-dimensional building block of complex architectures because of its water dispersibility, sub-nanometer-thin nature, synthesis scalability, and chemical reactivity. In the past decade, possibly inspired by the commercial success of carbon fibers, the wet-spinning technique has been used extensively for the integration of GO into fibers with micro- and nanometric diameters (*6*). Because GO fibers can be converted into electrically conductive fibers through reduction processes, they hold an enormous potential for multiple applications as sensors, electronic components, smart textiles, actuators, thermally conductive materials, and high-performance fibers, among others (*7*). Nevertheless, the industrial application of GO fibers has proven difficult. Developing processes to transform GO fibers into hierarchical assemblies like nonwoven fabrics, clothes, ropes, and nets (*8*), and possibly combining GO fibers with functional biological materials such as proteins (*9*), is necessary to achieve advanced functional nanocomposites useful for a broad range of applications.

Low-energy, stimulus-based assembly of GO-based architectures stands as an attractive field with many possible applications where reversibility is key. Textile and high-performance multifilament fibers account for the immediate applications of this technology. A potential use for the fibers may lie in the controlled release and capture of foreign materials such as particles and organic compounds. This requires developing a fundamental understanding of the role played by the individual GO sheets and their nanoscale assembly in the fusion and fission cycles. Even though biological fiber assemblies exhibit a much higher level of complexity and involve multiple components, the reversible assembly of GO fibers mimics nature and holds the refreshing potential to move the field forward. Specifically, GO fiber–based architectures featuring self-healing (*9*), self-sensing, and self-powered actuation should be vigorously pursued to finally bring GO fibers into practical applications. ■

Graphene oxide yarns

Graphene oxide (GO) fibers are assembled through wet-spinning techniques from GO sheets. When multiple GO fibers are immersed in a suitable solvent, they assemble into a hierarchical yarn. This assembly can be reversed, mimicking biological fusion-fission cycles.



GRAPHIC: N. DESAI/*SCIENCE* BASED ON CHANG *ET AL.* (*4*)

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The authors have a US patent (no. 9,284,193) for making graphene oxide films and fibers.

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Reversible fusion-fission fibers

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ELECTRONIC MATERIALS

Graphene oxide fibers fuse and come apart on demand

Simple solvent-based method forms strong yarns for electronics and smart materials

by ***Prachi Patel, special to C&EN***

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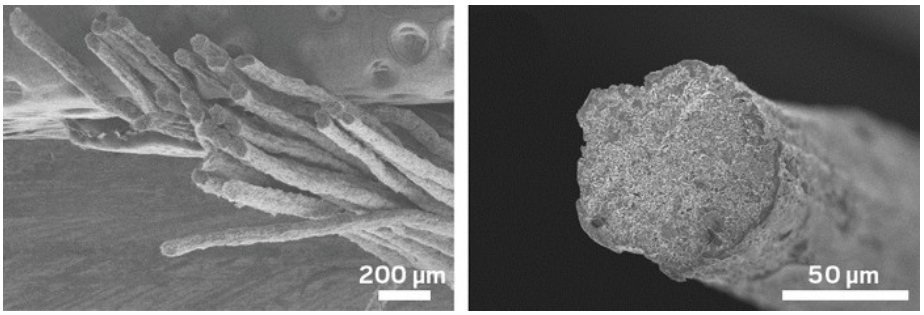

Credit: ***Science***

About 100 graphene oxide fibers are soaked in a solvent then dried, forming a thicker, stronger yarn. The yarn separates into the individual fibers when soaked again.

With a quick soak in a solvent bath, researchers can fuse hundreds of graphene oxide (GO) fibers into a thicker cable (*Science* 2021, DOI: **10.1126/science.abb6640**). Another solvent bath then splits the cable back into its original fibers.

The advance presents an easy, low-cost way to make strong, **graphene-based yarns** for **electronics** and smart materials that change shape when triggered. The yarn can also capture and release particles, which could be useful for pollutant-trapping fabrics and controllable delivery systems.

Chao Gao and Zheng Li of Zhejiang University, Yilun Liu of Xi'an Jiaotong University, and their colleagues soak a bundle of hundreds of fibers in water or another solvent for up to 10 min. The fibers, which are made of GO flakes, swell. Flakes in the outer layer of each fiber pack tightly, creating a skin around each fiber. When the researchers remove the bundle from the solvent, surface tension gathers the strands into a cylindrical cable, and the fibers bond together as they dry.

Immersing the yarn into a solvent reverses the process, splitting the yarn into its original strands. The skin around each fiber helps the fibers keep their structure. Such "precisely reversible" fusion has not been seen with other materials, Gao says.

As a demonstration, the team fused 13,500 GO fibers, each a micrometer wide, into a 1.2-mm-thick rod that was stiff and strong enough to support 680 times its weight. They also showed how the yarns could capture and release polymer fibers and microspheres via the fusion and separation process.

Polymeric, metallic, and ceramic fibers could also be reversibly fused and split by surrounding them with a GO outer layer, opening up a novel method for making composite fibers, says Ray Baughman, a chemist at the University of Texas at Dallas who was not a part of the study .

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“I find this simple but fascinating experiment remarkable,” says Mauricio Terrones, a chemist at Pennsylvania State University. One drawback is that GO fibers are not as stable as other materials, he says, but GO is a unique material “we are still unveiling its properties, structure and applications.”

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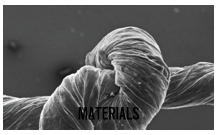

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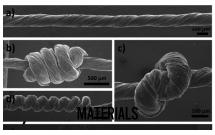

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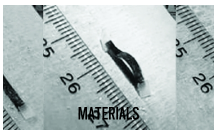

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